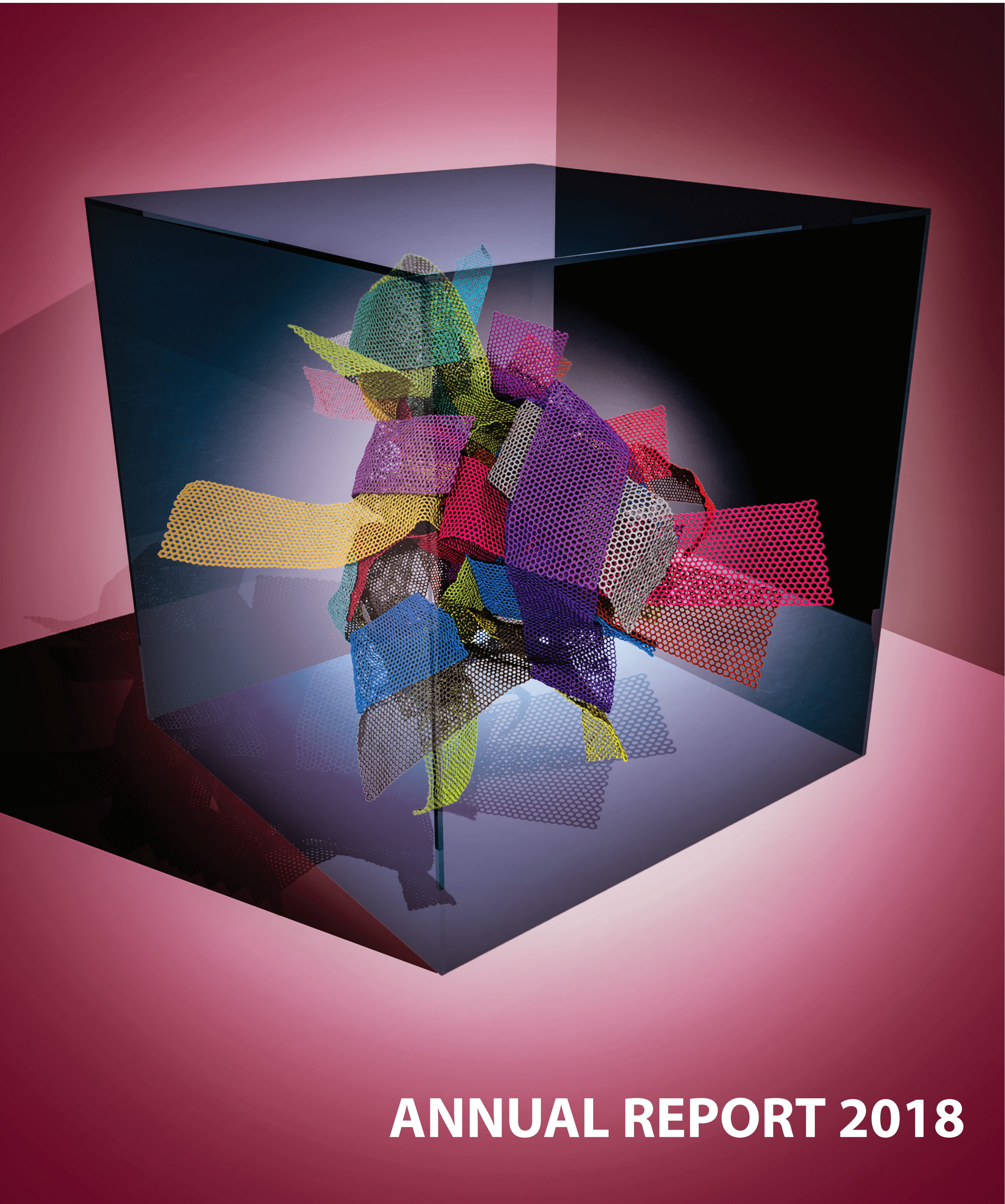
CHiMaD

Center for Hierarchical Materials Design



ANNUAL REPORT 2018

On the cover: A three-dimensional graphene melt composed of nanosheets, which characteristically exhibits a glass-transition behavior analogous to ordinary polymers [*Wenjie Xia*, Fernando Vargas-Lara, **Sinan Keten**, **Jack F. Douglas**]

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CHiMaD 2018

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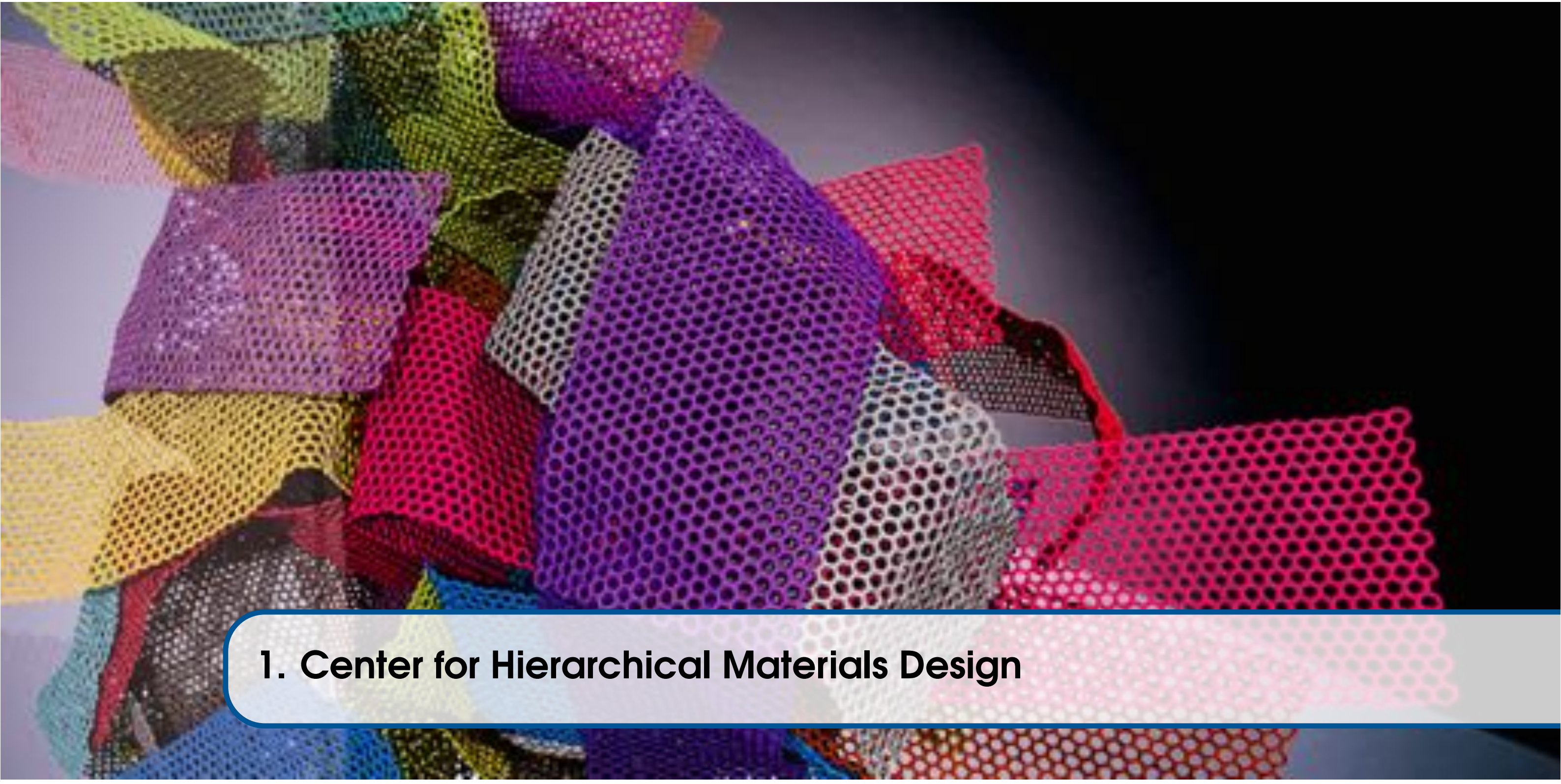
Center for Hierarchical Materials Design



1. Center for Hierarchical Materials Design

Foreword

The classical industrial materials development paradigm rests on a make-and-test strategy that results in unacceptably long times and increased costs before new materials can be inserted into applications. By contrast, materials design strategies offer the potential to radically transform U.S. manufacturing and cut costs dramatically by changing the way in which new products are deployed or inserted into production lines. The Center for Hierarchical Materials Design (CHiMaD) was founded to enable this transformation of U.S. manufacturing. Our mission is to demonstrate, advance, and disseminate the power of the materials design approach by providing opportunities to train scientists and engineers in materials design, by fostering the development of synergistic computational, experimental tools and, most importantly, the data that enable design, and by discovering new materials. CHiMaD has become a forum where multidisciplinary and multi-sector communities convene for in-depth discussions on all topics related to Materials Genome Initiative (MGI). A close partnership with the National Institute of Standards and Technology (NIST) has been central to the Center's success on all fronts.

We are pleased to present the 2018 annual report of the Center for Hierarchical Materials Design (CHiMaD), the National Institute for Standards and Technology (NIST) Center of Excellence in Advanced Materials. CHiMaD has become a focal point, both nationally and internationally, for the Materials Genome Initiative (MGI). We are committed to achieving the goals of the MGI through the development of broad research and outreach programs that involve participants from universities, national laboratories and companies. Bringing together researchers with expertise in computation, data science, and experiment to focus on the design of new materials is at the core of both the MGI and the CHiMaD. These collaborative efforts illustrate the power of incorporating experimental and computational databases in the materials design process.

CHiMaD is a Chicago-based consortium of Northwestern University (NU), University of Chicago (UC), Argonne National Laboratory (ANL), and small-business QuesTek Innovations, along with ASM Materials Education Foundation, a professional society. Over the past 5 years, CHiMaD has become a focal point, both nationally and internationally, for materials design. Building on its national and international visibility, CHiMaD has leveraged the NIST, Northwestern and University of Chicago investments to broaden our funding base and greatly enhance our materials design efforts. As a result, upwards of 180 researchers are now involved in CHiMaD, including more than 50 from NIST, in the design of innovative materials that range from Co-based superalloys to impact-mitigation materials and two-dimensional materials for heterostructures in applications. Our researchers have pioneered the translation of the materials design paradigm into the realm of soft materials and have created one of the most comprehensive polymer blend digital databases available, the Polymer Property Predictor and Database (PPPdB), providing many of the thermodynamic parameters needed for growing design efforts. More generally, we have partnered with NIST to create one of the largest materials databases available, the Materials Data Facility (MDF), with over 17TB of data and more than 260TB of discoverable data. Together, these databases, along with the Open Quantum Mechanical Database (OQMD) that we have used are being brought fully into CHiMaD, provide the data for emerging tools that rely on artificial intelligence to interpret sophisticated experiments and to design new materials; indeed, CHiMaD's work on developing machine learning tools for multiple aspects of the materials design paradigm has been foundational.

Last year, MDF developed and deployed a prototype service called MDF Connect with an aim to simplify the data ecosystem for researchers; to date over 200 authors from around the nation have uploaded more than 20 TB of data to MDF. An outgrowth of the Phase Field Methods workshops has been the establishment of community based benchmark problems for phase field codes, a second publication on the benchmark problems was published in 2018 and a third is in preparation. We are impacting materials design education throughout the country. For example, all the winners of the ASM undergraduate materials design competition for the past three years are ASM-CHiMaD materials design toolkit awardees. In 2018, our materials design training workshops expanded to include industry with the first successful demonstration through the pilot training conducted in March. In addition to these community building efforts, our materials design work continues to impact the community through the publication of high impact papers, technology transfer to industry, and high-profile talks.

Our national and international visibility and many design efforts are moving forward with great speed. We are making the promise of the Materials Genome Initiative a reality. We look forward to another exciting year.

Peter Voorhees, Juan de Pablo, Gregory Olson
CHiMaD Co-Directors

Mission and Vision

> *Accelerating materials discovery and commercialization by design and development of hierarchical methods and materials and enabling the complete integration of computation, experimentation and databases by building a strong community of current and future researchers*

Designing novel materials of specific properties for a particular application requires simultaneously utilizing physical theory, advanced computational methods and models, materials properties databases and complex calculations. This approach stands in contrast to the traditional trial-and-error method of materials discovery. CHiMaD aims to focus this approach on the creation of novel *hierarchical materials* which exploit distinct structural details at various scales, from the atomic on up, to obtain enhanced properties. The center's research focuses on both organic and inorganic advanced materials in fields as diverse as self-assembled biomaterials, smart materials for self-assembled circuit designs and, advanced metal alloys.

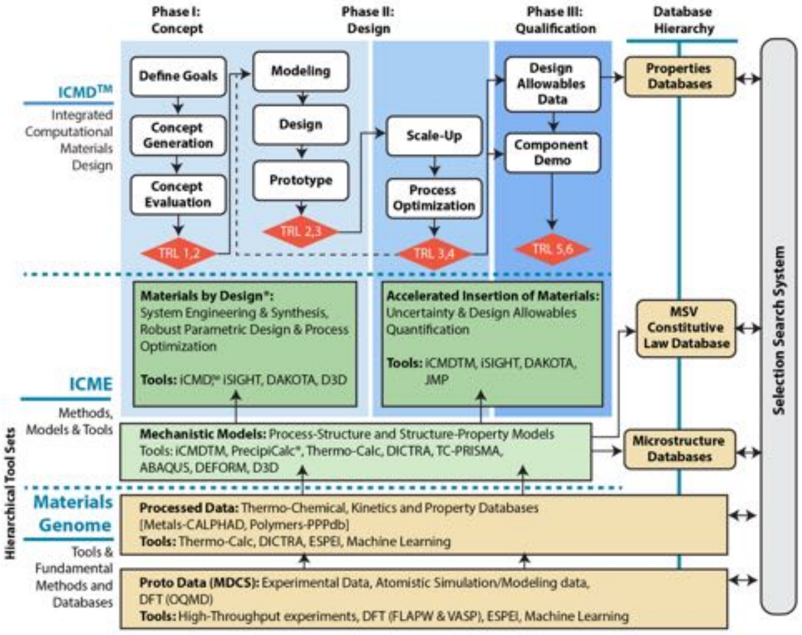


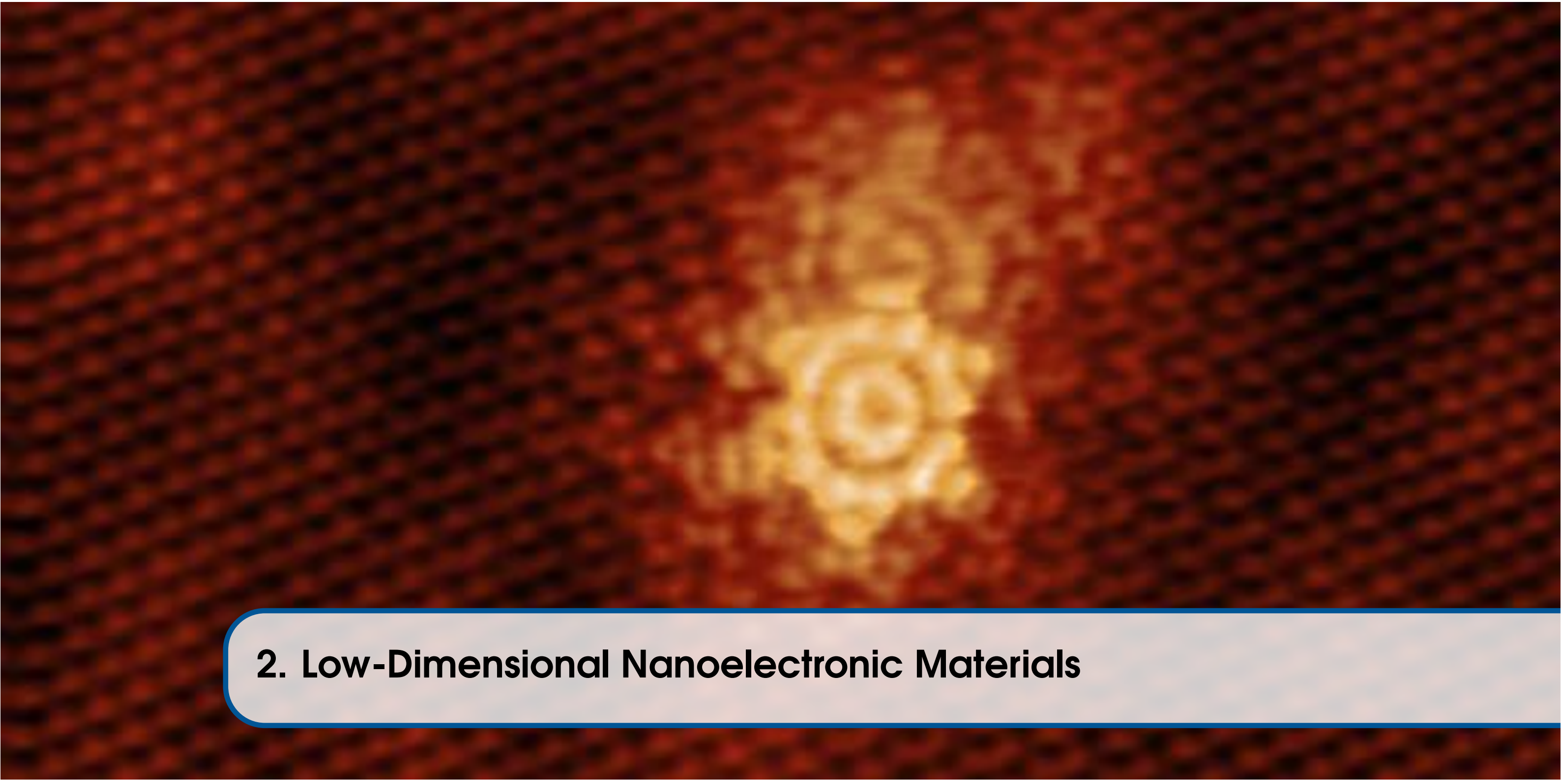
Figure 1.1: Hierarchical architecture of methods, tools (green), and databases (tan) for next-generation materials design and accelerated qualification.

Objectives

- Create a collaborative environment and concentration of scientific and technical capability to accelerate materials discovery and development
- Provide opportunities to transition new breakthroughs in advanced materials to industry
- Convene multidisciplinary and multi-sector communities for in-depth discussions
- Provide training opportunities for scientists and engineers in materials metrology
- Foster the development of integrated computation, modeling and data-driven tools
- Foster the discovery of new materials
- Establish opportunities for extended collaborations with NIST

II

CHiMaD Research



2. Low-Dimensional Nanoelectronic Materials

Mark Hersam (NU), Lincoln Lauhon (NU)

Albert Davydov (NIST), Francesca Tavazza (NIST), Benjamin Burton (NIST), Sergiy Krylyuk (NIST), Sugata Chowdhury (NIST), Kamal Choudhary (NIST)

Significance

The Low-Dimensional Nanoelectronic Materials use-case group seeks to understand and realize p-type and n-type doping in the low-dimensional limit. Since low-dimensional nanoelectronic materials have exceptionally high surface area to volume ratios, doping can be realized via two qualitatively different pathways: conventional substitutional doping and surface chemical functionalization. Furthermore, contacts can be designed to selectively inject carriers into the conduction or valence band, thus providing effective n-type and p-type behavior, respectively. Experimental methods in this use-case group include charge transport measurements, optical spectroscopy (e.g., absorbance, photoluminescence, and Raman spectroscopy), and high-resolution microscopy (e.g., scanning probe microscopy, electron microscopy, and atom probe tomography). These experimental methods are complemented by an extensive suite of computational techniques including multi-scale modeling, molecular dynamics, density functional theory, and finite element methods. The interplay between experiment and computation accelerates the understanding and design of doped low-dimensional nanoelectronic materials and their heterostructures.

2.1 Design Goals

The Low-Dimensional Nanoelectronic Materials Use-Case Group is pursuing the following design goals:

Substitutional Doping of Low-Dimensional Semiconductors. Substitutional doping is the most common means of controlling carrier concentration in bulk semiconductors. In the low-dimensional limit, substitutional doping is expected to play the same role, although the reduced degrees of freedom for charge transport suggest that this doping strategy will have greater deleterious effects on charge carrier mobility than in bulk materials. By combining unique characterization methods (e.g., atom probe tomography) with advanced computational methods (e.g., density functional theory), the design trade-offs between carrier concentration and mobility can be quantitatively understood and exploited to optimize materials properties for low-dimensional nanoelectronic devices.

Surface Chemical Functionalization Doping of Low-Dimensional Semiconductors. Since low-dimensional semiconductors have exceptionally high surface area to volume ratios, carrier concentration can also be controlled via surface chemical functionalization doping. The vast phase space for candidate surface chemical treatments necessitates an MGI-based approach to this design problem. Specifically, electron withdrawing and electron donating chemistries are being screened experimentally and computationally to realize controlled p-type and n-type doping, respectively. The results of this work are informing ongoing efforts to optimize spatially varying doping profiles that serve as the basis of many nanoelectronic devices including p-n junction diodes and transistors.

Developing Heterojunctions Consisting of Low-Dimensional Semiconductors. Since low-dimensional semiconductors have self-passivating surfaces, they can be directly integrated into van der Waals heterostructures without the epitaxial constraints of bulk semiconductors. Since this attribute implies a much greater number of heterojunction possibilities compared to bulk materials, efficient experimental and computational screening methods are required. Specific design goals include engineering of the band alignment and charge/energy transfer across heterojunction interfaces, which will allow optimization of nanoelectronic device metrics such as anti-ambipolarity, operating frequency, and optoelectronic response.

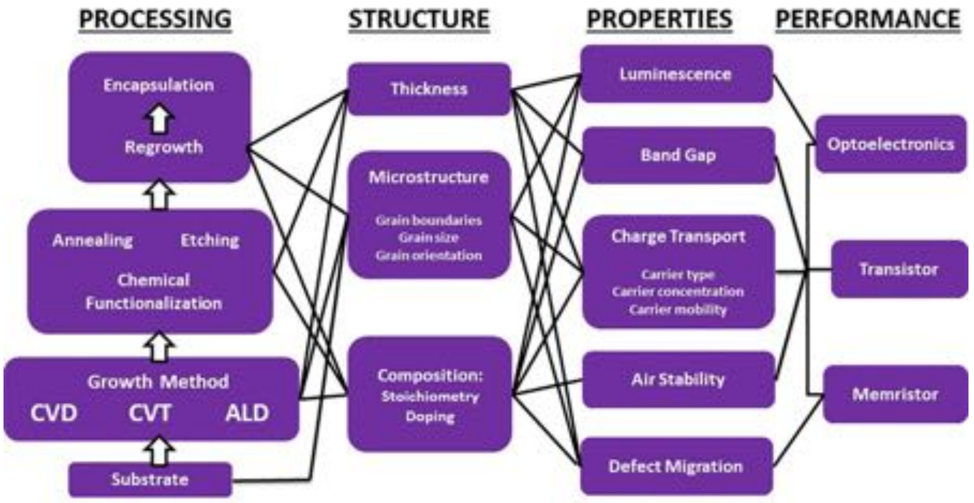


Figure 2.1: The system design chart for the low-dimensional nanoelectronic materials use-case group

2.2 Significant Accomplishments in 2018

Atomic Layer Deposition of Molybdenum Oxides with Tunable Stoichiometry Enables Controllable Doping of MoS_2 Following up on preliminary work reported in the previous year, **Lauhon** and **Hersam** in an inter-use-case group collaboration with **Marks** developed a new approach to doping MoS_2 using molybdenum oxides with tunable stoichiometry. Specifically, by using a low-oxidation state molybdenum precursor (Mo(NMe2)4) for atomic layer deposition, **Lauhon** grew a series of molybdenum oxides as dopants for the 2D semiconductor MoS_2. MoS_2 carrier density was probed with both Raman spectroscopy and field-effect transistor charge transport measurements (Figure 2.2) before and after deposition on MoS_2 grown by chemical vapor deposition by **Hersam**. Strong n-type accumulation and depletion were observed at the endpoints of the oxide series. Furthermore, dielectric deposition was shown to enhance mobility, and the dielectric could be removed by etching without decreasing the original mobility of the underlying 2D material, indicating that the low temperature ($80°C$) deposition process did not damage the 2D semiconductor. This work has been extended to in operando studies of field-effect transistors to explore directly the influence of the oxidants used in atomic layer deposition on doping of other transition metal dichalcogenides. The results of this study were recently published in Chemistry of Materials: M. J. Moody, A. Henning, T. Jurca, J. Y. Shang, H. Bergeron, I. Balla, J. N. Olding, E. A. Weiss, **M. C. Hersam**, T. L. Lohr, **T. J. Marks**, and **L. J. Lauhon**, "Atomic layer deposition of molybdenum oxides with tunable stoichiometry enables controllable doping of MoS_2," Chem. Mater., 30, 3628 (2018). DOI: 10.1021/acs.chemmater.8b03128.

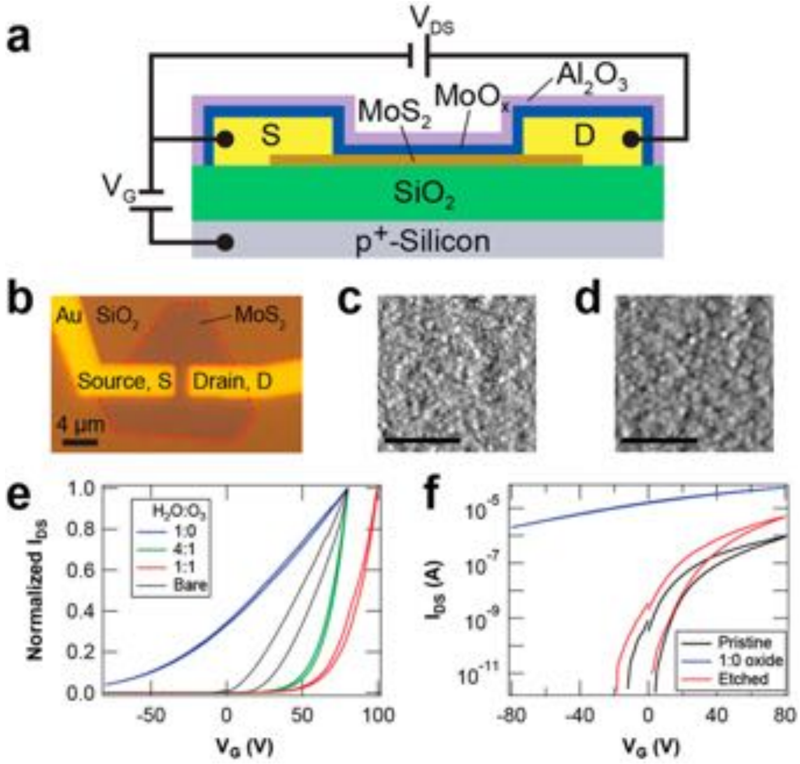


Figure 2.2: Deposition of molybdenum oxides with tunable stoichiometry on monolayer MoS_2 results in controlled doping. (a) Schematic of a doped field-effect transistor structure. (b) Optical image of a typical device. (c,d) Morphology via atomic force microscopy images of lithographically processed MoS_2 flakes (c) before (rms roughness 0.39 nm) and (d) after (rms roughness 0.3 nm) oxide deposition. (e) Field-effect transistor threshold voltage shifts after deposition of 10 nm $MoO_x/$10 nm Al_2O_3. (f) Reversibility of doping via wet etching. Z-scale is 2.5 nm and scale bar is 500 nm in panels c and d.

Transfer of Rotationally Commensurate 2D MoS_2 from Epitaxial Graphene In previous work, **Hersam** synthesized a 2D van der Waals heterostructure of rotationally commensurate monolayer MoS_2 on epitaxial graphene (EG) on SiC, resulting in MoS_2 with lower point defect density and grain boundaries orientations constrained to 30 and 60. In addition to its structural function in templating MoS_2 growth, the underlying EG dopes the MoS_2 via charge transfer. However, this interlayer coupling may be undesirable for many applications, thus requiring the selective transfer of the MoS_2 layer to arbitrary substrates. Over the past year, in an inter-use-case group collaboration with **Wolverton**, **Hersam** developed a method to selectively transfer monolayer MoS_2 from EG/SiC using a metal adhesion layer. First-principles opt-type van der Waals density functional theory calculations were used to design the technique, selecting for a metal with strong adhesion (large binding energy Eb) to MoS_2 and weak adhesion (small Eb) to EG. After computationally screening several candidate metals, Cu was chosen based on its calculated favorable interaction with the heterostructure materials: Eb(Cu/MoS_2) > Eb(Cu/EG) > Eb(MoS_2/EG). Following this computational prediction, a Cu metal layer was experimentally evaporated onto the MoS_2/EG heterostructure, after which thermal release tape (TRT) was placed on the Cu and mechanically peeled off, taking the Cu/MoS_2 and leaving behind the EG/SiC. The MoS_2 was then placed on its target substrate, and the TRT and Cu were removed. Demonstration of this technique with SiO_2/Si target substrates resulted in large-scale selective transfer of monolayer MoS_2 with preserved rotational alignment (Figure 2.3), recovery of photoluminescence previously quenched by EG, and high performance in field-effect transistors with mobilities up to 30 cm^2/Vs. Overall, this work leverages hierarchical design principles and computational screening to translate the advantages of 2D materials grown via van der Waals epitaxy to arbitrary substrates. The results of this study were recently published in Chemistry of Materials: J. Kang, I. Balla, X. Liu, H. Bergeron, S. Kim, **C. Wolverton**, and **M. C. Hersam**, "Selective transfer of rotationally commensurate MoS_2 from an epitaxially grown van der Waals heterostructure," Chem. Mater., 30, 8495 (2018). DOI: 10.1021/acs.chemmater.8b03128.

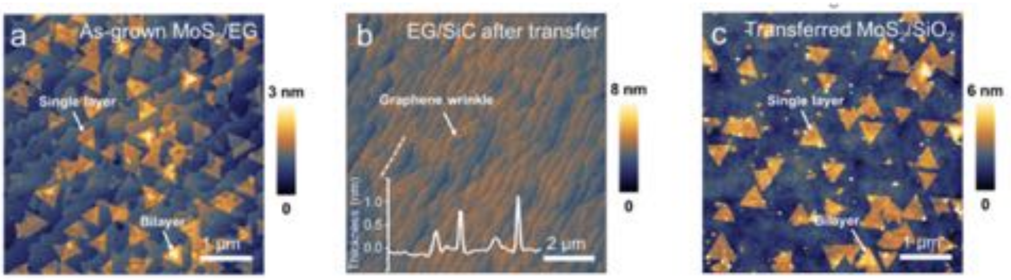


Figure 2.3: Atomic force microscopy height images of: (a) as-grown monolayer MoS_2 triangular domains on EG/SiC; (b) EG/SiC substrate after transfer, showing no MoS_2 domains remaining; (c) rotationally aligned transferred monolayer MoS_2 domains on an SiO_2/Si substrate

2.3 Research Accomplishments

Doping of $MoTe_2$ with Oxidizing Species The demonstration of doping by dielectric oxide deposition raises the question of whether the atomic layer deposition (ALD) precursors themselves can modify the carrier concentration in transition metal dichalcogenides (TMDs). Accordingly, p-type doping of $MoTe_2$ by exposure to H_2O, O_2, and O_3 was investigated by **Lauhon** using an in situ chip carrier that enabled direct in operando monitoring of the effects of oxidizing species on the behavior of transistors. Devices were fabricated with chemical vapor transport-grown $MoTe_2$ crystals provided by **Davydov** at NIST. Exposure to H_2O at $80°C$ increases hysteresis but does not permanently shift the voltage threshold (Figure 2.4). In contrast, exposure to increasingly oxidative species O_2 and O_3 at $80°C$ produces increasingly positive shifts in voltage threshold indicative of hole doping. Accordingly, with higher concentrations of atomic oxygen, it is possible to change the dominant carrier type from electrons to holes. Importantly, exposure to O_2 at $80°C$, which is the growth temperature of the MoO_x dielectric, does not induce doping, indicating that dielectric doping can be implemented independently of doping via controlled oxidation in these materials.

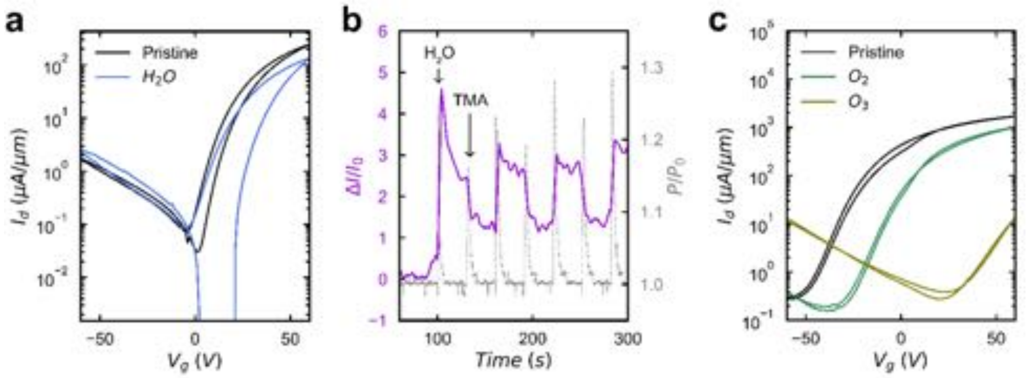


Figure 2.4: (a) Transistor transfer curves showing negligible threshold voltage shift but increased hysteresis when pristine $MoTe_2$ (black) is exposed to H_2O (blue) at $80°C$. The device was exposed to 68 cycles of 20 second pulses, each followed by a 5 second pump-out. (b) Time-dependent current in a $MoTe_2$ transistor during Al_2O_3 ALD growth at $80°C$. The device was biased into accumulation to increase the sensitivity to adsorbate induced changes. The pulse time for H_2O and TMA dosing was 0.015 seconds, with a 30 second pump-out in between each pulse. (c) Transistor transfer curve showing positive threshold voltage shift when pristine $MoTe_2$ is exposed to O_2 and O_3 at $80°C$. For O_2, the total dose was 68 cycles of 10 second pulses, each followed by a 3 second pump-out. For O_3, the total dose was 15 cycles of 10 second pulses, each followed by a 5 second pump-out

Laser Annealing of Atomic Layer Deposited SnS_2 **Lauhon** explored spatially selective processing schemes towards the patterning of p-n junctions in 2D materials. Specifically, atomic layer deposited (ALD) SnS_2 was laser annealed and analyzed by Raman spectroscopy and field-effect transistor (FET) measurements. Amorphous SnS_2 thin films were grown on SiO_2/Si or Si_3N_4/Si substrates at $120°C$ and encapsulated with AlO_x in situ. Laser annealing increases the intensity and decreases the width of the SnS_2 A1g Raman mode (Figure 2.5a-b), indicating that laser annealing promotes crystallization. Formation of SnO is observed at higher laser powers when annealing on SiO_2/Si substrates (Figure 2.5a); laser annealing of SnS_2 on Si_3N_4/Si suppresses oxidation (Figure 2.5b), highlighting the importance of the encapsulation to maintaining the desired stoichiometry. The annealed films are n-type semiconductors with field-effect mobilities that increase with increasing annealing power (Figure 2.5c-d). Surprisingly, the SnS_2 FETs on SiO_2/Si show significantly higher field-effect mobilities than those on Si_3N_4/Si, which suggests that an interfacial SnO layer may contribute to conduction and/or dope the SnS_2.

Charge Transfer in Ultrathin 2D MoS_2/Polymer Heterojunctions In previous work, **Hersam** reported the highest current density per thickness among photovoltaic technologies

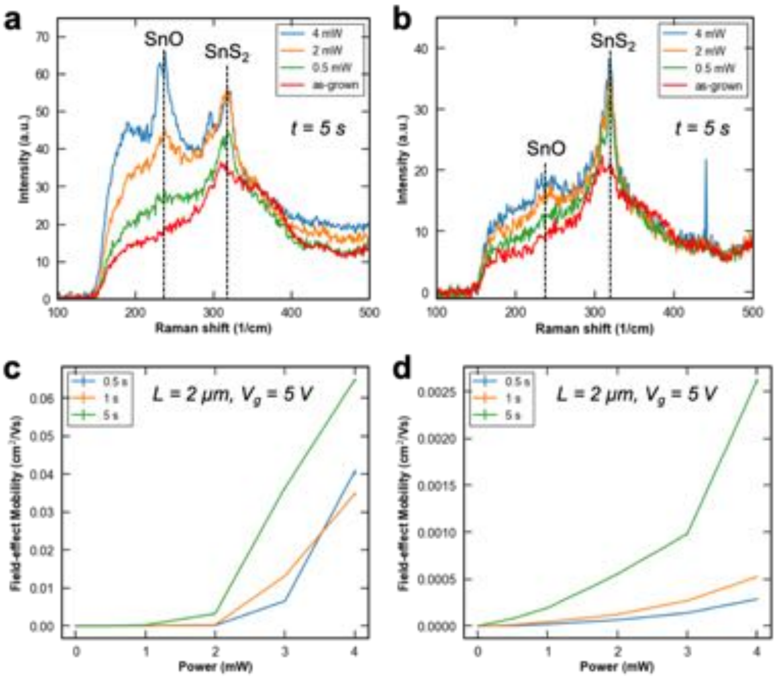


Figure 2.5: (a,b) Raman spectra versus annealing power of films grown on (a) SiO_2/Si and (b) Si_3N_4/Si. (c,d) Field-effect mobilities versus annealing power and time of films grown on (c) SiO_2/Si and (d) Si_3N_4/Si

using an ultrathin p-n heterojunction of monolayer MoS_2 and the polymer poly[[4,8-bis[(2-ethylhexyl)oxy] benzo[1,2-b:4,5-b']dithiophene-2,6-diyl] [3-fluoro-2-[(2-ethylhexyl) carbonyl] thieno[3,4-b]thiophenediyl]] (PTB7). To elucidate the mechanisms of charge transfer in these high-performance low-dimensional heterostructures, **Hersam** collaborated with Emily Weiss at Northwestern University to probe monolayer $MoS_2/PTB7$ van der Waals heterojunctions with transient absorption spectroscopy. This work revealed that charge transfer occurs on ultrafast time scales in $MoS_2/PTB7$ heterojunctions, with injection of electrons from the photoexcited PTB7 to the MoS_2 conduction band occurring in < 250 fs, and hole transfer from the photoexcited MoS_2 to PTB7 in 1-5 ps. The equilibrium between excitons and polaron pairs in PTB7 determines the charge separation yield, whereas the 3-4 ns lifetime of photogenerated carriers is limited by MoS_2 defects. Overall, this work elucidates the mechanisms of ultrafast charge carrier dynamics in $MoS_2/PTB7$ heterojunctions, which will inform ongoing efforts aimed at exploiting 2D semiconductor-polymer for photovoltaic and photodetector applications. This work was recently published in the Journal of Physical Chemistry Letters: C. Zhong, V. K. Sangwan, C. Wang, H. Bergeron, **M. C. Hersam**, and E. A. Weiss, "Mechanisms of ultrafast charge separation in a PTB7/monolayer MoS_2 van der Waals heterojunction," J. Phys. Chem. Lett., 9, 2484 (2018). DOI: 10.1021/acs.jpclett.8b00628.

Controlling the Semiconductor-to-Metal Phase Transition in 2D $MoTe_2$ $MoTe_2$ exists in two thermodynamically stable crystal forms: the semiconducting 2H phase at ambient conditions and the semimetallic 1T' phase at elevated temperatures. Reversibility of the 2H1T' phase transition can be controlled by temperature and other external stimuli, which makes this material attractive for advanced 2D electronics. Over the past year, Davydov prepared $MoTe_2$ single crystals by chemical vapor transport (CVT) that were subsequently studied by transmission electron microscopy, X-ray diffraction, X-ray photoelectron spectroscopy, secondary ion mass spectrometry, and Raman spectroscopy. The CVT growth conditions were optimized to stabilize the semiconducting 2H phase or the semimetallic 1T' phase as confirmed by field-effect transistor (FET) charge transport measurements that were performed by **Hersam**. The desired crystal structure was controlled by the growth temperature as well as by the post-growth annealing; namely, the 1T' phase was stabilized by quenching from $900°C$ processing temperature, while growth/annealing temperatures below $800°C$ resulted in the 2H phase. For the semiconducting 2H phase, the particular CVT transport agent dictated its intrinsic conductivity with iodine-assisted growth resulting in p-type behavior, while $TeCl_4$-assisted growth led to n-type behavior. The conductivity type was additionally tuned by scaling down the FET channel thickness. In particular, for the $TeCl_4$-assisted growth, the polarity switched from n-type for thick (> 50 nm) to p-type for thin (< 15 nm) layers, with ambipolar behavior observed for intermediate thicknesses. On the other hand, for the iodine-assisted growth, p-type polarity was independent of thickness in the range of 5-50 nm. The 2H-1T' phase transition in $MoTe_2$ was also observed by electric-field-induced reversible phase change during the formation of conducting filaments in 2H-$MoTe_2$ memristive devices. This work was the subject of an invited presentation at the Materials Research Society Fall Meeting in Boston on November 25, 2018: **A. V. Davydov**, **S. Krylyuk**, H. Zhang, L. A. Bendersky, B. Burton, A. Rani, I. Kalish, A. Bruma, B. Kalanyan, H. Hill, A. Hight Walker, M. Zaghloul, S. Oliver, P. Vora, F. Zhang, J. Appenzeller, H. Bergeron, M. Beck, and **M. C. Hersam**, "Control of conductivity type and semiconductor-to-metal phase transition in $MoTe_2$ single crystals."

Leveraging Computational and Experimental Data using Deep Transfer Learning In an inter-use-case group collaboration, **Tavazza** collaborated with **Agrawal** and **Wolverton** to use the JARVIS-DFT database to investigate deep transfer learning. The idea behind this work is to train machine leaning models on a large database (OQMD in this case) and then transfer that knowledge when learning from smaller databases (JARVIS-DFT in this case). This approach has significant implications for experimental databases since they are generally limited in size. This collaborative effort is currently being written up for publication: "Enhancing materials property prediction by leveraging computational and experimental data using deep transfer learning." Aspects of this work have also been included in the following publication in Scientific Reports: D. Jha, L. Ward, A. Paul, W.-K. Liao, **A. Choudhary**, **C. Wolverton**, and **A. Agrawal**, "ElemNet: Deep learning the chemistry of materials from only elemental composition," Scientific Reports, 8, 17593 (2018). DOI:10.1038/s41598-018-35934-y.

2.4 Impact Statement for CHiMaD Phase I

Using an MGI-based approach, the Low-Dimensional Nanoelectronic Materials Use-Case Group has advanced the understanding of p-type and n-type doping in the low-dimensional limit, thus enabling rational design of doping profiles for advanced electronic and optoelectronic applications. In particular, two of the design goals of this use-case group focus on alternative routes to doping of 2D materials, namely substitutional doping and doping via surface functionalization. Substitutional doping, which is the dominant paradigm for conventional 3D semiconductors, has been challenging to implement in 2D materials due in part to the challenge of measuring dopant concentration and distribution in extremely small volumes. The use-case group successfully addressed this challenge by developing the first application of atom probe tomography to analyze dopant locations in 2D materials. In parallel, the use-case group made major advances in doping via surface layers such as covalent organic adlayers on black phosphorus. The third design goal of this use-case group focuses on the realization of van der Waals heterojunctions between two-dimensional materials. Again, the use-case group made significant progress towards this goal, including the growth and transfer of rotationally commensurate MoS_2/graphene heterostructures. In addition, the role of stoichiometry on doping in MoS_2 has been elucidated, allowing control over electronic and optical properties. Overall, this use-case group has quantifiably impacted the field through high-impact papers in *Nature, Nature Chemistry, Nano Letters, ACS Nano, Advanced Functional Materials, and Chemistry of Materials* that have been highly cited including 4 ISI Highly Cited Papers (Top 1% in Citations).

2.5 Collaborations

2.5.1 CHiMaD Collaborations

Transfer of rotationally commensurate MoS_2 from epitaxial graphene
Chris ***Wolverton*** *(NU), Mark* ***Hersam*** *(NU)*
A metal layer mediated scheme to selectively transfer MoS_2 from epitaxial graphene was developed with computational guidance for the selection of the metal.

Controlled doping of MoS_2 via atomic layer deposition of molybdenum oxides
Tobin ***Marks*** *(NU), Lincoln* ***Lauhon*** *(NU), Mark* ***Hersam*** *(NU)*
Controlled doping of MoS_2 was achieved following the atomic layer deposition of molybdenum oxides with tunable stoichiometry.

2.5.2 NIST Collaborations

Characterization of doped and alloyed transition metal dichalcogenides
Albert ***Davydov*** *(NIST), Lincoln* ***Lauhon*** *(NU)*
Davydov group at NIST provides CVT-grown doped and alloyed transition metal dichalcogenides for characterization at Northwestern. The **Lauhon** group exfoliates these crystals as monolayers and few layers thin films to characterize how their electrical properties are impacted by alloying and charge transfer doping.

Charge transport and phase transition in $MoTe_2$ and $MoWTe_2$
Albert ***Davydov*** *(NIST), Mark* ***Hersam*** *(NU)*
Davydov is supplying bulk crystals of $MoTe_2$ and $MoWTe_2$ alloys that are being studied through field-effect transistor device fabrication and testing by **Hersam**.

Van der Waals epitaxy of MoS_2 on epitaxial graphene synthesized at atmospheric pressure
Randolph ***Elmquist*** *(NIST), Mark* ***Hersam*** *(NU)*
Elmquist is supplying epitaxial graphene, graphene nanoribbons, and graphene quantum dots on SiC for subsequent growth of MoS_2 heterostructures using CVD by **Hersam**.

Charge transport in MoS_2 and WS_2 grown by atomic layer deposition
James ***Maslar****(NIST), Mark* ***Hersam*** *(NU)*
Maslar is supplying 2D MoS_2, WS_2, and $MoWS_2$ alloy samples for charge transport measurements by **Hersam**.

2.5.3 External Collaborations

Charge transfer in ultrathin polymer-2D MoS_2 heterojunctions
Emily Weiss (NU), Mark ***Hersam*** *(NU)*
Charge transfer dynamics in ultrathin PTB7-MoS_2 p-n heterojunctions were studied using transient absorption spectroscopy.

Solution-processed nanoelectronic materials
Shashi Jasty, Jia Choi, and Adam Raw (Sigma-Aldrich), Mark ***Hersam*** *(NU)*
A series of solution-processed, printable 2D material inks have been formulated by **Hersam**, and are being distributed commercially by Sigma-Aldrich. In particular, 12 products are now available from Sigma-Aldrich including: Product Numbers 793663 (graphene inkjet ink), 796115 (graphene gravure ink), 798983 (graphene screen ink), 900695 (photonically annealable graphene ink), 900696 (graphene aqueous ink), 900710 (hexagonal boron nitride aqueous ink), 900724 (MoS_2 aqueous ink), 900960 (graphene spray coating ink), 901187 (MoS_2 inkjet ink), 901349 (hexagonal boron nitride blade coating ink), 9014140 (hexagonal boron nitride inkjet ink), 901857 (MoS_2 spray coating ink).

2.6 Technology Transfer

Patent
System and method for anti-ambipolar heterojunctions from solution-processed semiconductors
D. M. Jariwala, V. K. Sangwan, W. Xu, H. Kim, T. J. ***Marks****, and M. C.* ***Hersam***

Published on 03/27/18 Patent Number: 9,929,725 This invention reports the utilization of van der Waals heterostructures in anti-ambipolar transistors. Anti-ambipolarity is useful in several electronic applications including analog signal conditioning circuits and neuromorphic computing.

Product
Solution-Processed Nanoelectronic Materials
Shashi Jasty, Jia Choi, and Adam Raw (Sigma-Aldrich), Mark ***Hersam***

A series of solution-processed, printable 2D material inks have been formulated by **Hersam**, and are being distributed commercially by Sigma-Aldrich. In particular, 12 products are now available from Sigma-Aldrich including: Product Numbers 793663 (graphene inkjet ink), 796115 (graphene gravure ink), 798983 (graphene screen ink), 900695 (photonically annealable graphene ink), 900696 (graphene aqueous ink), 900710 (hexagonal boron nitride aqueous ink), 900724 (MoS_2 aqueous ink), 900960 (graphene spray coating ink), 901187 (MoS_2 inkjet ink), 901349 (hexagonal boron nitride blade coating ink), 9014140 (hexagonal boron nitride inkjet ink), 901857 (MoS2 spray coating ink).

Data
Datasets Uploaded to Materials Data Facility in 2018

1. Atomic layer deposition of molybdenum oxides with tunable stoichiometry enables controllable doping of MoS_2
   Moody, MJ; Henning, A; Jurca, T; Shang, JY; Bergeron, H; Balla, I; Olding, JN; Weiss, EA; ***Hersam, MC****; Lohr, TL;* ***Marks, TJ****;* ***Lauhon, LJ***
   DOI: 10.18126/M26351

2. Self-Aligned van der Waals Heterojunction Diodes and Transistors
   Sangwan, VK; Beck, ME; Henning, A; Luo, JJ; Bergeron, H; Kang, JM; Balla, I; Inbar, H; ***Lauhon****, LJ;* ***Hersam****, MC*
   DOI: 10.18126/M2FK9J
3. Multi-Terminal Memtransistors from Polycrystalline Monolayer Molybdenum Disulfide
   Sangwan, VK; Lee, HS; Bergeron, H; Balla, I; Beck, ME; Chen, KS; ***Hersam****, MC*
   DOI: 10.18126/M29W68
4. Mechanisms of Ultrafast Charge Separation in a PTB7/Monolayer MoS_2 van der Waals Heterojunction
   Zhong, CM; Sangwan, VK; Wang, C; Bergeron, H; ***Hersam****, MC; Weiss, EA*
   DOI: 10.18126/M21D12
5. Selective Transfer of Rotationally Commensurate MoS2 from an Epitaxially Grown van der Waals Heterostructure
   Kang, J.; Balla, I.; Liu, X.; Bergeron, H.; Kim, S.; ***Wolverton****, CM.;* ***Hersam****, MC*
   DOI:10.18126/M2WK9K

2.7 Publications and Presentations

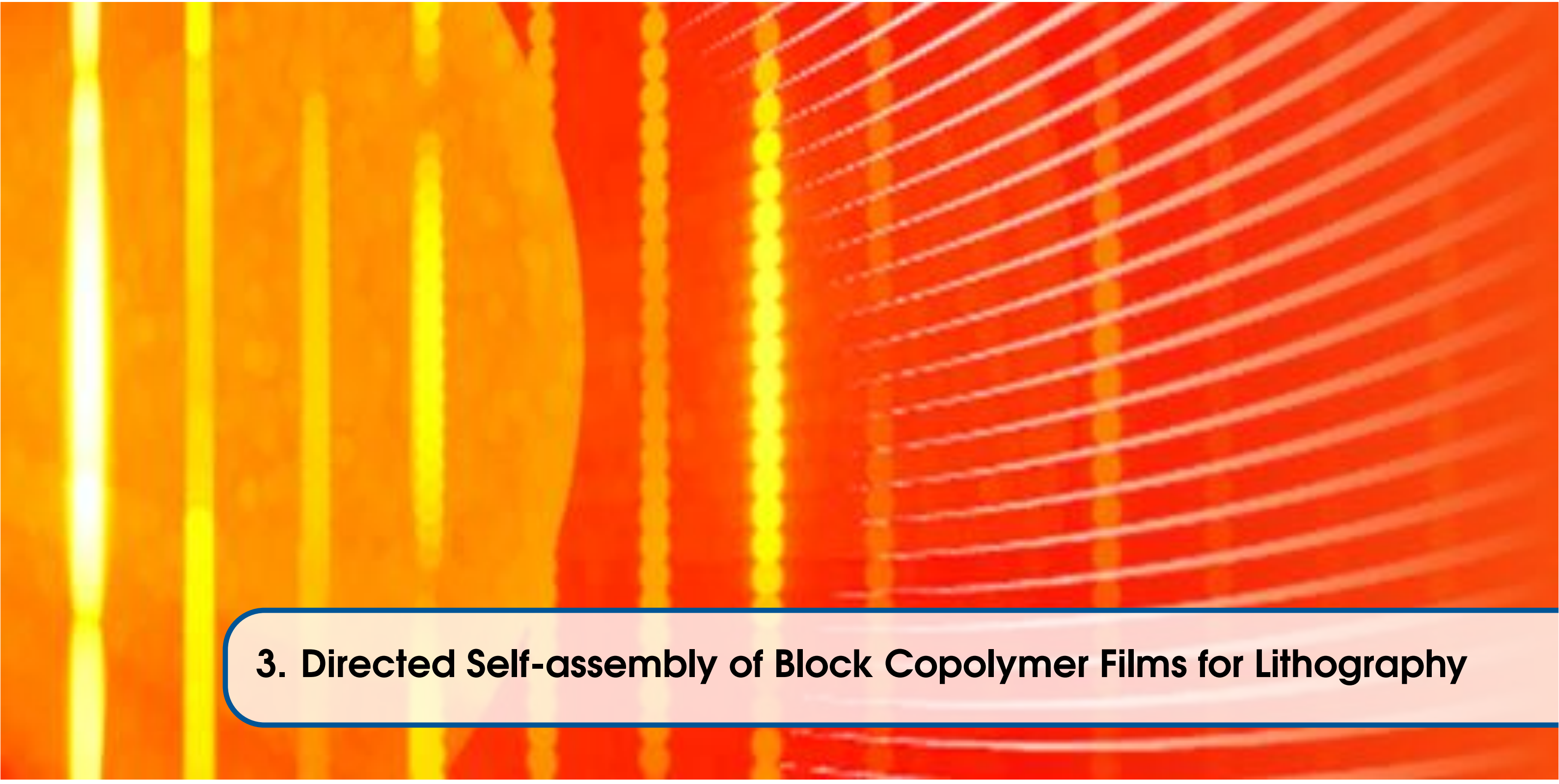
7 Number of CHiMaD-supported publications in 2018 by Low-Dimensional Nanoelectronic Materials use-case group. Please see chapter 27 for details. [27.1,27.3, 27.4, 27.5, 27.6, 27.7]

10 Number of CHiMaD-influenced NIST publications in 2018 by Low-Dimensional Nanoelectronic Materials use-case group. Please see chapter 27 for details. [27.a., 27.b., 27.c., 27.d., 27.e., 27.f., 27.g., 27.h., 27.i., 27.31]

40 Number of presentations on CHiMaD-supported research given in 2018 by Low-Dimensional Nanoelectronic Materials use-case group members, please see section 26.1 for the complete list.

2.8 CHiMaD Team

Low-Dimensional Nanoelectronic Materials Use-Case Group Researchers			
Name	**Position**	**Affiliation**	**Project (PI)**
Mark Hersam Lincoln Lauhon	Principal Investigator Principal Investigator	NU/MSE NU/MSE	
Jiangtan Yuan	Postdoc	NU/MSE	Low-dimensional semiconductor heterojunctions *(Hersam)*
Hadallia Bergeron	Graduate Student	NU/MSE	Low-dimensional semiconductor heterojunctions *(Hersam)*
Zhehao Zhu	Graduate Student	NU/MSE	Laser processing to control mobility and carrier concentration in transition metal dichalcogenides and monochalcogenides *(Lauhon)*
Ju Ying Shang	Graduate Student	NU/MSE	Doping and electrical characterization of transition metal dichalcogenides *(Lauhon/Davydov)*



3. Directed Self-assembly of Block Copolymer Films for Lithography

Paul Nealey (UC), Juan de Pablo (UC), Steven Sibener (UC), Heinrich Jaeger (UC)

Joseph Kline (NIST), Daniel Sunday (NIST)

Significance

Directed self-assembly (DSA) of block copolymers (BCPs) has the potential to revolutionize nanomanufacturing. BCPs self-assemble to form densely packed features with highly uniform dimensions and shapes in ordered arrays at the scale of 3 to 50 nm. Chemical prepatterns or templates may be defined using traditional lithographic materials and processes such as 193 immersion lithography at the scale of 80 nm. By directing the assembly of block copolymer films on the templates, the overall resolution of the lithographic process may be increased by three to four-fold or more and still meet the stringent requirements of manufacturing, including perfection, dimension control, and pattern placement accuracy. The interest and growth in research activity and expenditure is driven in the semiconductor industry by the prospect of manufacturing future generations of computer chips according to Moore's law using frequency division techniques. Even if billions are invested in new fabrication facilities based on extreme ultra violet lithography, for example, frequency division will still be required to meet the resolution requirements. For hard drives, block copolymer lithography is the only known technology to fabricate nanoimprint masters to manufacture bit patterned media at the required storage densities greater than 2 Tera dots/inch^2. This use-case collaborates with Intel, imec, the Semiconductor Research Corporation, CEA-Leti (Grenoble, France), Seagate and Mentor Graphics.

3.1 Design Goals

Directed Self-assembly of Block Copolymer Films for Lithography use-case group seeks to design block copolymer materials and assembly processes to enable manufacturing via DSA at length scales of 10 nm, scaling to 5 nm, and meet manufacturing constraints with respect to defects (0.01/cm^2), and line edge and width roughness (<1 nm). The structure of the assembled BCP depends on many variables describing material properties (molecular weight, block architecture, interaction parameter χ) and process parameters (temperature, solvent, assembly time, template chemistry and topography, interfacial energies, and surface energies); our design objectives can only be met by developing and validating predicative multi-scale models. Moreover, the immense parameter space requires evolutionary approaches to simultaneously understand and optimize numerous variables, in the context of analysis of experimental data and in the design of new materials and processes. To accomplish our design goals with respect to structure, our priority and main activity is to advance three-dimensional metrology tools and methods, and develop expertise in quantitatively comparing predictive models and experimental data. The focus of former mentioned activities are the independent variables (design criteria)- material properties, template properties, and processing parameters. Molecular simulation-based data analysis is the foundation for delineating complex structure-property-processing relationships and is accomplished by close collaboration with **Kline** (NIST) and **Sunday** (NIST). To accomplish our design goals with respect to design for minimization of defects, we are pioneers in understanding the dominant role of the dynamics and evolution of structure formation. Again, ultra-high information experiments involving high-speed AFM imaging are being analyzed in conjunction with predicative models that include dynamic information, leading to the determination of the key material and process criteria to meet manufacturing constraints.

3.2 Significant Accomplishments in 2018

Following are the significant accomplishments of the Directed Self-assembly of Block Copolymer Films for Lithography team in the past year:

- High-throughput synthetic strategies for high-X block copolymers with A-b-(B-r-C) architecture were developed to realize perpendicular lamellar without topcoat.
- To understand the mechanism of defect formation and control defectivity in DSA system, state-of-the-art defect inspection tools in 300-mm wafer fabrication facility has been used to reveal the dependence of defectivity on annealing time. This work was in collaboration with industry (IMEC).
- TEM tomography was exploited to probe the kinetics and structural evolution in both of the chemoepitaxy DSA with density multiplication and graphoepitaxial hole shrink process. In-depth knowledge was gained through the captured details of the three-dimensional shapes and dimensions of assembled BCP domains, fluctuations of domain interface through three-dimensions as well as molecular-level distribution and architecture. This work was in collaboration with industry (IMEC, TOK).
- Our understanding of metal - polymer interactions was expanded in order to use all inorganic chemical templates for DSA. These all inorganic templates allow for the possibility of DSA which is self-aligned with underlying active device layers, which thermodynamically guarantees placement error free patterning for industrial applications. This work was in collaboration with industry (IMEC, TOK).
- DSA of other soft materials, blue phase liquid crystal with self-organized 3D cubic symmetry, into perfect single crystal structure, which allows the in-depth analysis of the phase transition. Using in-situ Resonant Soft X-ray Scattering to investigated the martensitic transformation between two blue phases (BPI and BPII) to achieve a comprehensive understanding that the transformation must accompany the twinning to release residual lattice strain.

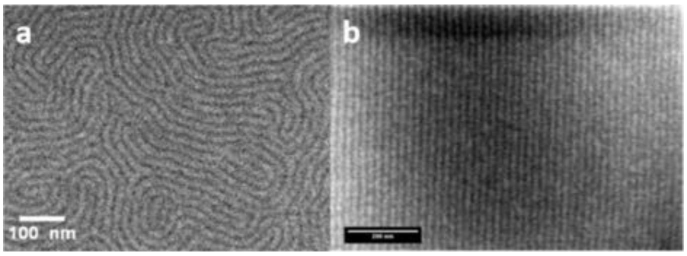


Figure 3.1: Top down SEM images of (a) perpendicular lamellar morphology (fingerprint) and (b) DSA of 10 nm half pitch demonstrated using LINE flow processes.

- Fundamental understanding of morphology development, including stitched to lamellar morphology transition, generated from predictive molecular models. Excellent agreement with real-time AFM experiments.

3.3 Research Accomplishments

New Block Copolymer Materials with High χ Value for DSA Applications In collaboration with Intel, **Nealey** and Rowan (UC) focused on the synthesis and processing of new materials and processes for 14 nm full pitch line and space patterns for semiconductor manufacturing, Figure 3.1. One of the most challenging materials properties to engineer is the interaction parameter, χ, and the related property, $\chi * N$, where N is a measure of the polymer molecular weight. The difficulty is that the parameters χ and $\chi * N$ are single valued for one particular block copolymer (BCP). If χ was the only material property to optimize for the BCP segregation strength and interfacial energy between blocks, it would be easy. Unfortunately, multiple template and processing parameters also depend on χ and χ*N, including at least 8 values of surface and interfacial energies. To address this challenge, **Nealey** and Rowan (UC) groups designed and synthesized a series of high χ block copolymers with A-b-(B-r-C) architecture using a robust thiol "click" chemistries. With such architecture, it is possible to decouple the segregation strength of the blocks, χ, from the surface energies of the blocks and the interfacial energies of the blocks in contact with the template. Polystyrene-b-poly(1,2-butadiene) was successfully functionalized with polar thiols, therefore, a 14-20 nm (full pitch) lamellae perpendicular orientation without topcoat was achieved. A wide variety of block copolymers with equal free energy can be synthesized. Applying the A-b-(B-r-C) architecture into block copolymers enables not only the equal surface energies, but also tunable χ. To understand the effect of thiol functionalities, χ N on the DSA related core features such as defectivity and etch selectivity is of immense significance for the future nanolithography applications.

Kinetics of Defect Annihilation in Chemo-epitaxy Directed Self-Assembly As a potential solution for next-generation lithography, directed self-assembly (DSA) of block copolymers (BCPs) is still restrained in high volume manufacturing partly due to its defectivity issue. Understanding the kinetics of defect annihilation is crucial to revealing the mechanism of defect formation and to further reducing defectivity in DSA. With the long-term partnership with IMEC, **Nealey** group has accomplished the most comprehensive study of defect annihilation in DSA so far using state-of-the-art defect inspection tools in 300-mm wafer fabrication facility, Figure 3.2. Statistical analysis of experimental data reveals the dependence of defectivity on annealing time, which can be well described by physical models. It was also revealed that defects with different spacing and configurations show different frequencies of occurrence. A simple yet powerful mathematical model is developed to predict the frequency of occurrence for a certain spacing or configuration, which agrees well with experimental results. Though these findings are based on PS-*b*-PMMA, it is

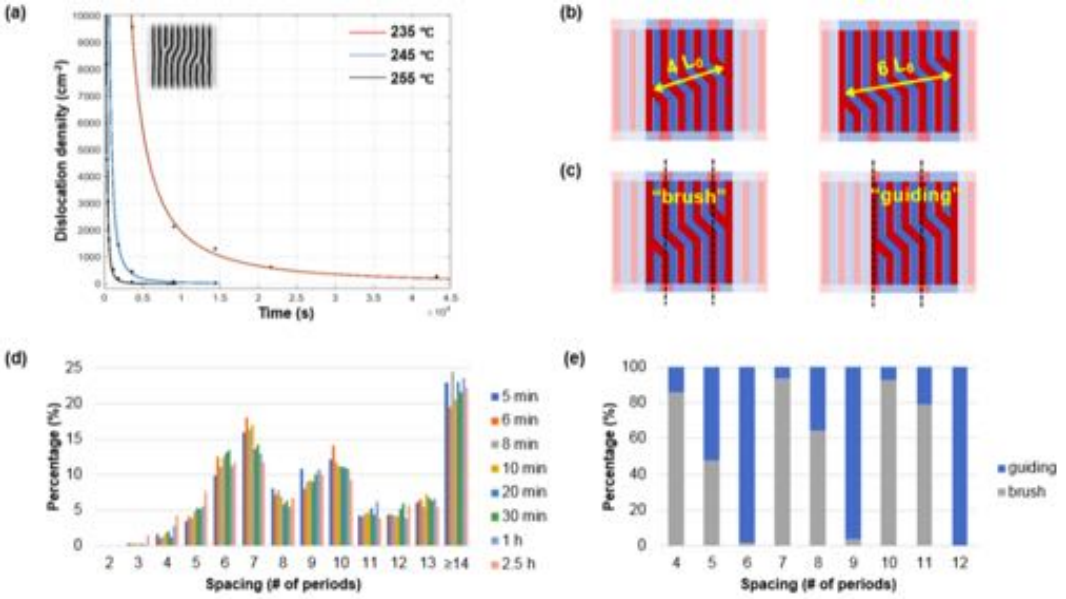


Figure 3.2: Dislocation density decreases faster under higher annealing temperatures, and the dependece on annealing time can be well described by power law models. (b) Disclocations can have different spacing, and each spacing can have (c) two different configureations. Configuration is defined as relative position of dislocation edges with respect to guiding stripes. (d) Distribution of spacing stays relatively constant over time. (e) Distribution of configurations is repeated every three periods.

anticipated that they could be applied to high-χ BCPs as well, and such fundamental understanding will open up new possibilities for further reducing defectivity in DSA. Building on these studies, the **de Pablo** group has developed a comprehensive understanding of defect annihilation based on theory and simulations.

Engineering the Kinetics of DSA of Block Copolymers Toward Fast and Defect-free Assembly Arrested annealing and TEM tomography were combined to probe the kinetics and structural evolution in the chemoepitaxy DSA of PS-*b*-PMMA with density multiplication. During the initial stages of annealing, BCP domains developed independently at first, with aligned structures at the template interface and randomly oriented domains at the top surface. As the grains coarsened, the assembly became cooperative throughout the film thickness, and a metastable stitch morphology was formed, representing a kinetic barrier. The stitch morphology had a three-dimensional structure consisting of both perpendicular and parallel lamellae. On the basis of the mechanistic information, the effect of key design parameters on the kinetics and evolution of structures in DSA were studied. Three types of structural evolutions were observed at different film thicknesses: (1) immediate alignment and fast assembly when thickness $< L_0$ (L_0 = BCP natural periodicity); (2) formation of stitch morphology for $1.25 - 1.45L_0$; (3) fingerprint formation when thickness $> 1.64L_0$. It was found that the DSA kinetics can be significantly improved by avoiding the formation of the metastable stitch morphology. Increasing template topography also enhanced the kinetics by increasing the PMMA guiding surface area. A combination of $0.75L_0$ BCP thickness and $0.50L_0$ template topography achieved perfect alignment over 100 times faster than the baseline process. This research, Figure 3.3, demonstrates that an improved understanding of the evolution of structures during DSA can significantly improve the

DSA process. The experimental observations are in excellent agreement with simulations by **de Pablo**.

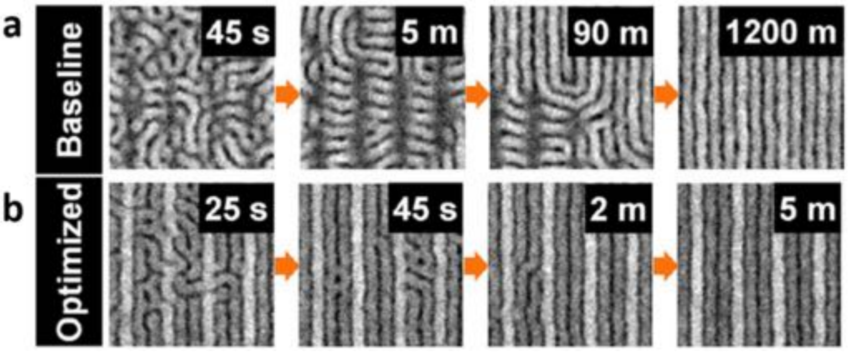


Figure 3.3: SEM images of (a) the kinetic evolution at 190°C; (b) DSA films combining $0.75L_0$ BCP thickness with $0.5L_0$ template topography. This optimized condition greatly enhanced the DSA kinetics, yielding complete alignment within 5 min of annealing at 190°C.

3D Characterization of Block Copolymer DSA in Graphoepitaxial Hole Shrink Process Using Scanning Transmission Electron Microscopy (STEM) Tomography Acquiring three-dimensional information becomes increasingly important for the development of block copolymer (BCP) directed self-assembly (DSA) lithography. Leveraging the nanofabrication capabilities at UC as well as the collaboration with IMEC (Belgium) and TOK (Japan), the use-case group investigated the 3D structures of BCP formed in templated holes with different surface chemistry and geometry. The 3D shapes of BCP in holes were reconstructed from STEM tomography data with high accuracy and high resolution without sample damage, Figure 3.4. The results reveal that strong PS-preferential sidewalls are necessary to create vertical DSA cylinders and that template size outside of process window could result in defective DSA results in three dimensions. The findings of the **Nealey** group were confirmed by simulation results using coarse-grained model done by **de Pablo** group. This work allowed for the development of a fundamental understanding of the key DSA factors for optimizing the graphoepitaxial hole shrink process and facilitated the implementation of DSA in semiconductor manufacturing.

Utilization of Metal-Polymer Interactions for Self-aligned Directed Self-assembly of Device Relevant Features The **Nealey** group has systematically studied the interactions between industry relevant metal films and PS-*b*-PMMA. It was discovered that both copper oxide, and gold were non-preferential for PS-*b*-PMMA and thus are promising materials for incorporation into an all inorganic chemical pattern. They showed that simple surface energy considerations do not adequately capture these polymer - metal interactions, but that some trends still appear to exist in terms of water contact angle vs relative preferentiality. Through close collaboration with IMEC (Belgium) and TOK (Japan), an all inorganic template has been created, and successfully directed the DSA of PS-*b*-PMMA with 4X density multiplication perfectly aligned with the underlying features, Figure 3.5. This allows DSA to be used in a self-aligned process in which the BCP thermodynamically snaps to the underlying layer. Self-aligned processes are required because today's feature sizes are beyond the resolution limit of the exposure tools.

In-situ Characterization of Liquid Crystal Martensitic Transformation by Resonant Soft X-ray Scattering Phase transformations between crystal structures of different lattice symmetries, which mediate crystal nucleation and growth, are of fundamental importance

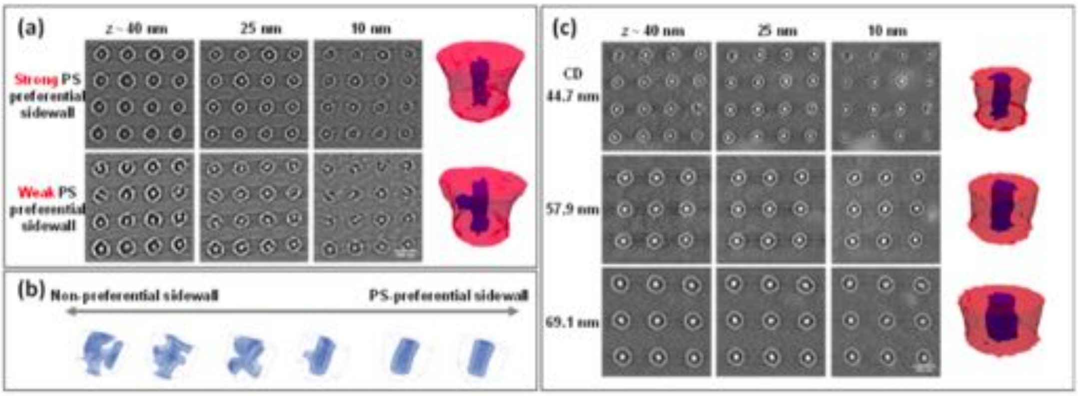


Figure 3.4: (a) STEM tomography results and the 3D rendering of DSA structures in templated holes with different surface chemistry. The grayscale images are xy slices (parallel to substrate) taken from the reconstruction volume at different sample heights, z. (b) Simulation results using coarse-grained model. Only the PMMA block is presented in blue color. (c) STEM tomography results and the 3D rendering of DSA cyclinders in templates with different critical dimensions, CD.

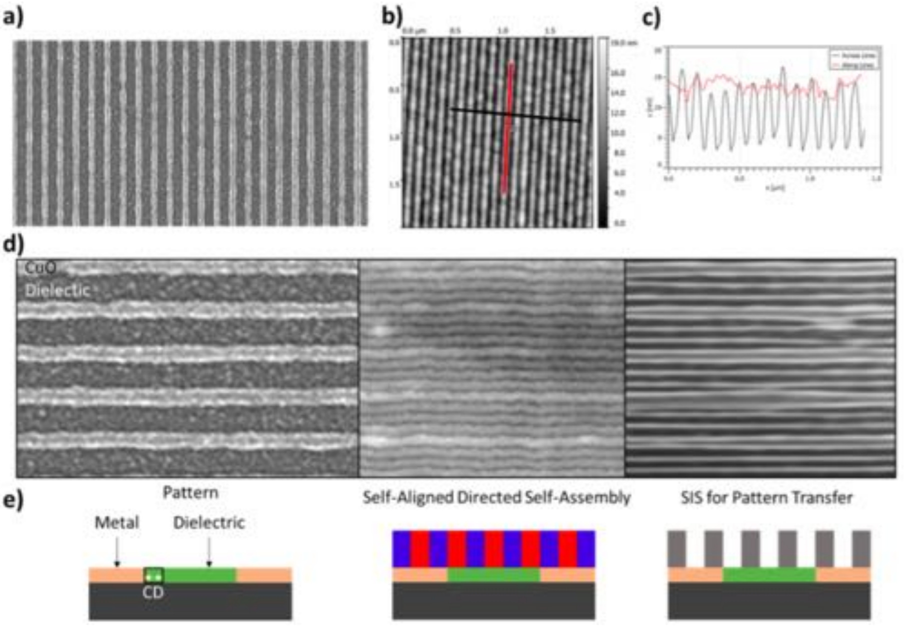


Figure 3.5: (a) Copper Oxide patter as imaged via SEM as well as (b) AFM and (c) extracted profiles. (d0 SEM metrology at each step of the self-aligned DSA process, with (e) a schematic showing the film cross section. The pitch of the guide stripe pattern (100nm) serves as an internal scale bar in all cases.

across a wide range of scientific disciplines. The **Nealey** and **de Pablo** groups previously used single crystals of blue phase (BP) liquid crystals (LCs) and optical measurement to study the soft matter analog of classic crystal-crystal phase transformations, and demonstrated that the phase transformations that arise in BP LCs are a kind of martensitic transformation. The **Nealey** group, with closely collaborating with **Kline** introduced an in-situ resonant soft X-ray scattering (RSoXS) to study in detail of martensitic transformation in single crystal BP LCs, investigating in particular how the lattice symmetry and molecular orientations change during the transformation, Figure 3.6. Based on the measurements, the **Nealey** group demonstrated that the martensitic transformation of BP LCs accompanies twinning formation, and we propose that the formation of twining is to release the residual strain caused by the rapid martensitic transformation. It is anticipated that these experiments will provide one of the most comprehensive studies yet of BP and, therefore, soft matter martensitic phase transformations. In close collaboration with NIST, Dr. Hyeongmin Jin was hired as a NIST CHiMaD Postdoctoral Fellow to be based at the IEX beamline at the APS (sector 29) and to design and build an end station for the polarization dependent RSoXS experiments.

3.4 Impact Statement for CHiMaD Phase I

In the past funding period the DSA use-case group synthesized a series of high χ block copolymers with A-b-(B-r-C) architecture, in order to process for DSA of 14 nm full pitch line and space patterns, meeting the stringent requirements for semiconductor manufacturing (Intel). Through extensive partnership with IMEC, this group has exploited TEM tomography and defect inspection tools as techniques to study defectivity, defect kinetics and hole shrink defects. Our understanding of metal - polymer interactions was further expanded during DSA process. With respect to RSoXS, the crystal reconfiguration process was successfully captured during the martensitic transformation between two blue phases by close collaboration with NIST.

3.5 Collaborations

3.5.1 NIST Collaborations

Polymeric Properties

*Jack **Douglas** (NIST), Steven **Sibener** (UC)*

There is an ongoing discussion on graphoepitaxy, dynamics of polymer structural evolution, and the effect of additives such as nanoparticles on local structure in thin polymer films with the aim to build complementing research.

In Situ Study of Blue Phase Liquid Crystal Martensitic Transformation by Resonant Soft X-ray Scattering

*Paul **Nealey** (UC), Juan **de Pablo** (UC), Joseph **Kline** (NIST), Daniel **Sunday** (NIST)*

Nealey group prepared BPLCs sample series and performed comprehensive SEM, optical microscopy, spectrometer and particularly RSoXS characterization together with NIST scientists during allotted beam times at the Advanced Light Source (ALS). The **de Pablo** group developed theory and simulations to analyze blue phase liquid crystal lattice distortion during phase transition.

3D characterization of block copolymer DSA using CDSAXS

*Paul **Nealey** (UC), Juan **de Pablo** (UC), Joseph **Kline** (NIST), Daniel **Sunday** (NIST)*

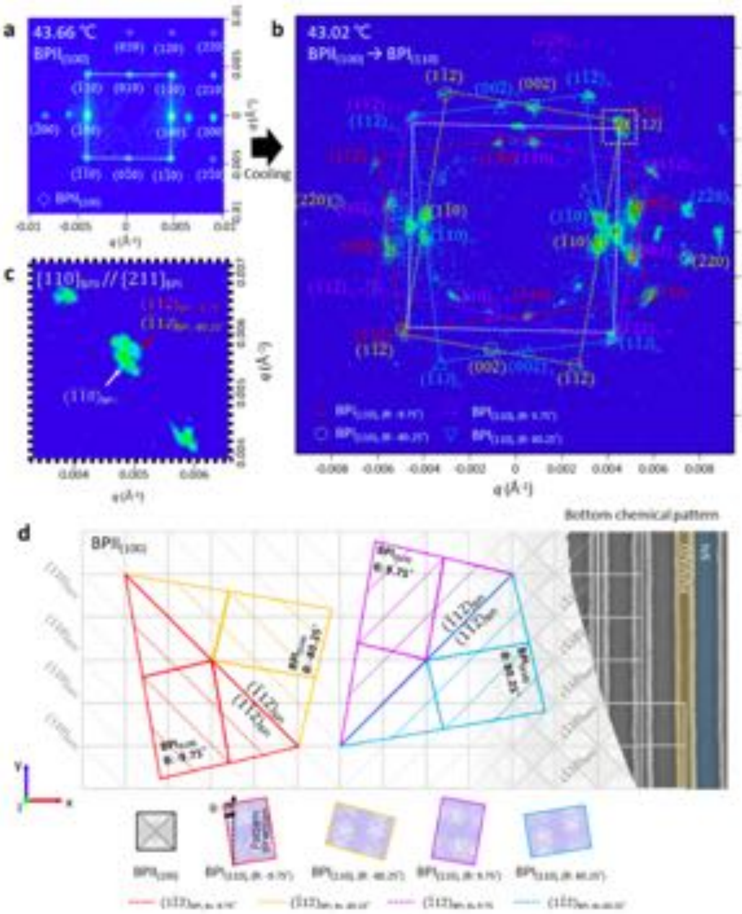


Figure 3.6: RSoXS pattern of (a) single crystal BPIII$_{(100)}$ at $43.66^{\circ}C$ and (b) poycrystalline BPI$_{(110)}$ which has distinct domains with four different in-plane orientation, θ of -9.75°, -80.25°, 9.75°, and 80.25° at 43.02°. θ is an angle difference between $[110]$ of BPI$_{(110)}$ lattice and bottom chemical pattern stripe. (c) Scattering peaks of $\{112\}_{BPI}$ plane appear near the scattering peaks of $\{110\}_{BPII}$ plane and merged each other during the transformation. (d) Schematic of BPI$_{(110)}$ lattice that martensitic transformed from single crystalline BPI$_{(100)}$.

Collaborating with **Kline** and **Sunday**, CHiMaD funded graduate students and postdocs from the **Nealey** group prepare DSA samples, perform CDSAXS experiments, and data analysis together with NIST scientists during allotted beam times at the Advanced Light Source (ALS) to characterize the 3D structure of block copolymer.

Probing the Interfacial Width of Block Copolymer Using Soft X-ray Reflectivity
*Paul **Nealey** (UC), Juan **de Pablo** (UC), Joseph **Kline** (NIST), Daniel **Sunday** (NIST)*
Nealey group collaborated with **Kline** and **Sunday** to prepared samples, performed soft X-ray reflectivity measurements at the ALS in two beam times, and analyzed data to study the interfacial width of PS-P2VP as well as PS-PMMA with ionic liquid addition.

Design and Construction of New RSOXS End Station for Sector 29 at APS, ANL
*Paul **Nealey** (UC), Joseph **Kline** (NIST), Daniel **Sunday** (NIST), Dean **Delongchamp** (NIST)*
NIST-ChiMaD postdoctoral research fellow Hyeongmin Jin designed the new end station with significant input from NIST experts, including spending significant time at NIST, to enable the key collaborative/joint project in the coming years development of polarization-dependent RSoXS techniques to characterize 3D structure and molecular orientation in soft matter systems.

3.5.2 External Collaborations

3D Characterization of Industry Relevant DSA Processes
*Paulina Rincon and Hyoseon Suh (imec, Belgium), Xiaomin Yang (Seagate), Patrick Theofanis (Intel), Paul **Nealey***
Nealey group collaborates with long-time industry partners at imec, Seagate and Intel to develop materials, processes, and 3D characterization techniques for industry-relevant DSA applications.

Synthesis and develop High-X block copolymer for directed self-assembly
*Paul **Nealey** (UC), Stuart Rowan (UC), Patrick Theofanis (Intel)*
Nealey group collaborates with Rowan (UC) group and Intel to synthesis high-χ block copolymer materials for sub-10 nm DSA.

Addition of Ionic Liquid for sub-10 nm DSA
*Paul **Nealey** (UC), Takahiro Dazai (TOK, Japan), Paulina Rincon (imec, Belgium), Roel Gronheid (imec, Belgium)*
Nealey group collaborated with TOK and imec to develop material and processes for sub-10 nm DSA using PS-PMMA with ionic liquid addition.

Complementary 3D characterization using grazing incidence small angle X-ray scattering and critical dimension small angle X-ray scattering
*Fanny Rodolakis Simoes (ANL), Jessica McChesney (ANL), Zhang Jiang, (ANL) Joseph Strzalka (ANL), Jin Wang (ANL), Nicola Ferrier (ANL), Paul **Nealey***
Nealey group collaborated with researchers at the Advanced Photon Source in Argonne National Laboratoy to develop complementary X-ray probes for the characterization of block copolymer DSA.

Development and Metrology of High-χ Materials and Process for DSA
*Raluca Tiron, Redouane Borsali and Patrick Ronnoa (CEA-LETI, CERMAN, University of Grenoble-Alpes, CNRS), Paul **Nealey***

Nealey group collaborates with researchers from CEA-LETI and University of Grenoble-Alpes at France to develop high-X materials and complementary metrologies for characterization of block copolymer DSA.

Self-Alignment of Block Copolymer Films on Graphene Nanoribbons

Steven J. ***Sibener*** *(UC), Paul* ***Nealey*** *(UC), and Michael Arnold (University of Wisconsin, Madison)*

The Arnold Group (UW) has recently developed a procedure for fabricating aligned graphene nanoribbons with a narrow size distribution. Through this collaboration, block copolymer patterns deposited on these structures has been demonstrated to align with the edges of these films. High-speed AFM allows visualization of polymer transport on the graphene films and the pattern alignment process to understand the mechanisms for alignment.

3.6 Technology Transfer

Software

Polymer Property Predictor and Database (PPPdb)

Roselyne B. Tchoua, Kyle Chard, Debra J. Audus, Logan Ward, Joshua Lequieu, Juan de Pablo, and Ian Foster

Link: `http://pppdb.uchicago.edu/`

PPPDB contains the curated and extracted values for T_g and χ from the hybrid human-automated information extraction pipeline. The data are publicly available and searchable through the website above and through the Materials Data Facility search index.

Workforce Transfer

Workforce Transfer to Industry

Following students have taken positions in the industry, directly related to their research under the Directed Self-assembly of Block Copolymer Films for Lithography Use-Case Group

- Jiaxing Ren - Lam Research
- Chun Zhou - Intel
- Hyoseon Suh - IMEC
- Soonmin Yin and Elizabeth Ashley closely collaborate with Panasonic

3.7 Publications and Presentations

5 Number of CHiMaD-supported publications in 2018 by Directed Self-assembly of Block Copolymer Films for Lithography use-case group. Please see chapter 27 for details. [27.63, 27.64, 27.65, 27.66, 27.67]

10 Number of presentations on CHiMaD supported research in 2018, please see section 26.2 for the complete list.

3.8 CHiMaD Team

DSA of Block Copolymers for Lithographic Applications **Use-Case Group Researchers**			
Name	**Position**	**Affiliation**	**Project (PI)**
Paul Nealey Juan de Pablo Steven Sibener Heinrich Jaeger	Principal Investigator Principal Investigator Principal Investigator Principal Investigator	UC/IME UC/IME UC/CHM UC/PHY	
Xiao Li	Postdoctoral Researcher	UC/IME	Directed self-assembly of liquid crystalline blue-phases into single-crystals with controllable lattice orientation *(Nealey)*
Hyeongmin Jin	Postdoctoral Researcher	UC/NIST	Development of quantitative soft x-ray probes of the three-dimensional structure of soft materials such as block copolymers and liquid crystalline systems *(Nealey)*
Chun Zhou	Graduate Student	UC/IME	Characterization of 3D structures in block copolymer in block copolymer DSA using TEM tomography *(Nealey)*
Moshe Dolejsi	Graduate Student	UC/IME	Characterization of Bulk and Microdomain Structures in Gyroid Phase Block Terpolymers with GISAXS and GIWAXS *(Nealey)*
Elizabeth Ashley	Graduate Student	UC/IME	Characterization of 3D structures in Gyroid phase block copolymer using GIWAXS *(Nealey)*
Gurdaman Khaira	Graduate Student	UC/IME	Evolutionary algorithms for materials design of DSA *(de Pablo)*
Brandon Peters	Graduate Student	UC/IME	Directed Self-Assembly of Block Copolymers *(de Pablo)*
Jonathan Raybin	Graduate Student	UC/CHM	Environmentally controlled AFM of Block Copolymer Directed Self-Assembly *(Sibener)*
Julia Murphy	Graduate Student	UC/CHM	Environmentally controlled AFM of Block Copolymer Directed Self-Assembly *(Sibener)*

4. Soft Matter Design Based on Charge Complexation

Matthew Tirrell (UC), Juan de Pablo (UC), Monica Olvera de la Cruz (NU), Kenneth Shull (NU)

Debra Audus (NIST), Vivek Prabhu (NIST), Jack Douglas (NIST), Edwin Chan (NIST)

Significance

Soft matter refers to materials where energies of order kT can influence the structure and functions of the materials. One of the main inspirations for soft matter design comes from biology, where many functional materials are built from relatively weak non-covalent interactions. Considerable success has been achieved in creating new materials based on hydrogen bonding and van der Waals interactions. In addition to these bonding modalities, and others, biology makes extensive use of electrostatic interactions, frequently mediated by multi-valent ions, including charged polymers. The Use-Case on Soft Matter Design based on Charge Complexation is a comprehensive effort to understand, and employ this understanding to design new materials based on predictive use of attractive interactions induced by multi-valent interactions in systems of charged polymers. This Use-Case comprises experimental and theoretical work measuring, simulating and modeling phase diagrams in charged polymer systems, leading to new functional materials based on charge complexation including: self-assembled nanoparticles and hydrogel formation in charged block copolymers, and organic-inorganic nanocomposites. New materials are being designed and synthesized, guided by theory and computation, for adhesives, biomaterials, drug and gene delivery, bone and tooth repair.

4.1 Design Goals

The integrated goals of this Use-Case constitute what we call the *8M program: Molecules, Mixtures, Micelles, Materials, Measurement, Moduli, Modelling, Manufacturing*. This use-case uses custom-synthesized macromolecules (M1) to study the phase diagrams of complexation in mixtures (M2) of polyelectrolytes and to exploit the micellar (M3) self-assembly possibilities resulting from complexation. Self-assembly via complexation is being used to create micelles, encapsulating materials, hydrogels, coatings, and composite materials, all based on charge complexation and ionic interactions as the underlying mechanisms. (M4). Data on phase behavior and micellization routes to new materials are input into searchable databases. Rheological measurement (M5) gives insight into moduli (M6), viscosity, and other viscoelastic properties. Predictive modelling and simulation (M7) of both phase behavior and self-assembly is increasingly advanced. This fundamental work is input into processing and eventually manufacturing, methods (M8) for soft materials based on electrostatic complexation. The ultimate goal is to write down the structure of a set of synthesizable charged macromolecules and, from that structure, be able to know with confidence the properties of the final material, and then, with deeper insight, to do the reverse. Figure 4.1 shows the system design chart for the Soft Matter Design Based on Charge Complexation use-case group.

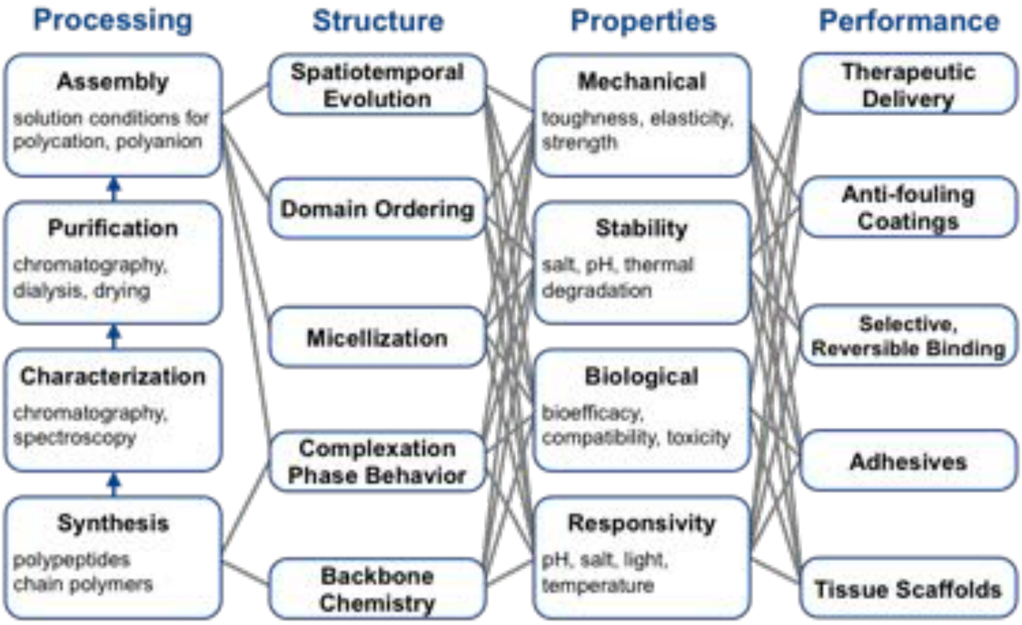


Figure 4.1: Systems design chart for soft matter design based on charge complexation.

4.2 Significant Accomplishments in 2018

Quantitative Liquid-liquid Phase Diagram of Polyglutamic Acid and Polylysine A principal accomplishment in 2018 was the publication of the quantitative liquid-liquid phase diagram of polyglutamic acid and polylysine: "Phase behavior and salt partitioning in polyelectrolyte complex coacervates", L. Li, S. Srivastava, M. Andreev, A.B. Marciel, J.J. **de Pablo** and M. **Tirrell** in Macromolecules [27.80]. The data from this study have been deposited in the CHiMaD Materials Data Facility. The important conclusion of this work, besides the materials design utility of the data themselves, is that the ancient theory by Voorn and Overbeek, valued for its simplicity and long held to be passably accurate in portraying the salt-polymer concentration phase space, but never before rigorously tested, is fundamentally wrong in not accounting properly chain connectivity and excluded volume interactions. These fundamental deficiencies predict inaccurate tie lines and binodal shapes, especially in the higher salt and higher polymer concentration regions of composition space. These data, and the physical understanding they reveal, allow us make reliable quantitative calculations moving up the "Structure" column of Figure 4.1 to design micelles and higher order self-assembled structures.

Temperature and Dynamic Effects A second significant accomplishment along these lines in 2018 relates to the top element of the "Structure" column of Figure 4.1, namely, the temperature and dynamic effects: "Effect of temperature on the structure and dynamics of triblock polyelectrolyte gels", A. Rahalkar, G. Wei, R. **Nieuwendall**, V.M. **Prabhu**, S. Srivastava, A.E. Levi, J.J. **de Pablo** and M. **Tirrell** in Journal of Chemical Physics [27.76]. In this collaborative NIST-CHiMaD study, triblock polyelectrolyte gels were characterized by small-angle neutron scattering (SANS) and dynamic light scattering (DLS). The size of the polyelectrolyte complex core does not change with temperature. However, the neutral middle block displays a temperature-dependent conformation. The liquid-like order of the complex core within the gel phase leads to stretched PEO bridging chains that approach their unperturbed dimensions with increasing concentration. A stretch ratio for bridging chains was defined as the ratio between stretched and unperturbed dimensions. A further reduction in the chain stretching occurs with increasing temperature due to solvent quality. DLS observes multiple modes consistent with a collective diffusion (fast mode) and diffusion of clusters (slow mode). The fast mode is consistent with a collective diffusion coefficient via the concentration dependence. The slow-mode diffusion coefficient decreases rapidly with total polymer concentration and is best described by the diffusion of dynamic gel clusters. These clusters appear to break up with increased temperature as revealed by the normalized diffusion constant. However, the interconnected networks persist as shown by the SANS structure factor peak.

Quantitative Understanding of Solvent Effects and Other Non-ionic Interactions in Polyelectrolyte Complexation Several collaborative studies produced significant advances in our quantitative understanding of solvent effects and other non-ionic interactions in polyelectrolyte complexation. One part of the collaboration (**Shull, Olvera de la Cruz**) focuses on measuring the interfacial adsorption of partially charged block copolymers and relating it to the competition between ionic correlations and hydrophobic interactions. The **Shull** group has used a mixed solvent system (dimethyl sulfoxide and ethylene glycol) to tailor the strength of the hydrogen-bonding interactions between partially quaternized poly(4-vinylpyridine) [QVP] and poly(methacrylic acid) [PMAA]. "Tuning the Viscoelasticity of Hydrogen-Bonded Polymeric Materials through Solvent Composition", L. Mathis, Y.Y. Chen and K.R. **Shull** in Macromolecules [27.73]. The charge introduced by the quaternization reaction enables homogeneous solutions to be formed over a large concentration range, even in the presence of attractive hydrogen-bonding interactions between the proton-donating PMAA and the proton-accepting QVP. Another CHiMaD-NIST collaboration "Influence of Ion Solvation on the Properties of Electrolyte Solutions", M. Andreev, J.J. **de Pablo**, A. **Chremos** and J.F. **Douglas** in Journal of Physical Chemistry B [27.79].

4.3 Research Accomplishments

Polyelectrolyte complexes (PECs) form upon associative phase separation of oppositely charged polymers in aqueous milieu. These materials are highly tunable, spanning from low-viscosity coacervates to glassy, high modulus solids with high water-content (30-90%), which makes them appealing for bioadhesives, food science and coatings technologies. Charged block polymers can also be engineered to self-assemble into PEC-core micelles

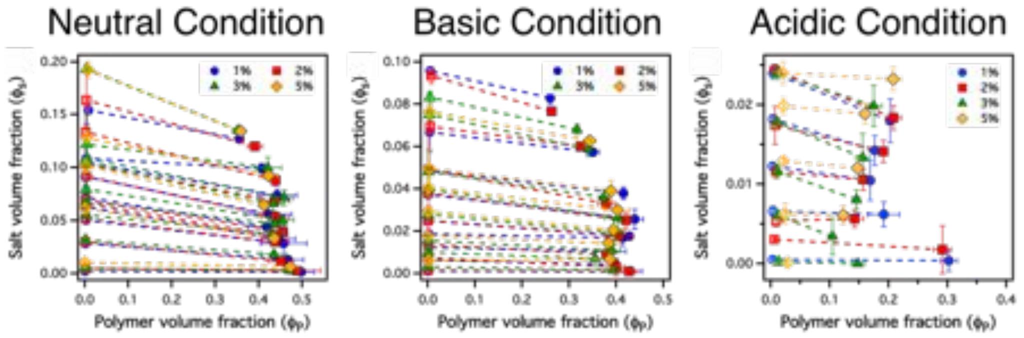


Figure 4.2: Experimentally-measured phase diagrams of PAA/PAH under neutral, basic, and acidic conditions

and hydrogels for drug delivery, regenerative medicine and theranostics. However, nearly all of the important materials attributes are currently understood only qualitatively. This year, the Soft Matter Design Based on Charge Complexation use-case group aimed to put this understanding on a more quantitative footing by generating theoretical models and corresponding thermodynamic/kinetic data to enable materials design with desired properties for end-use technologies. These overarching goals are described by a systems design chart in Figure 4.1, which illustrates the relationship between polyelectrolyte processing, structure, properties, and performance.

Phase Behavior in Charge Complexation One current focus is on precise quantitative determination of homopolymer complexes and block polyelectrolyte phase behavior. as a quantitative basis for property prediction. To clarify the misunderstanding and inconsistent experimental agreement about PECs in our field, quantitative descriptions of the phase behavior for a model polyelectrolyte pair of poly(acrylic acid) and poly(allylamine hydrochloride) have been developed, which are widely-used both in industry and academia. Although both polymers are weak polyelectrolytes, the morphology of the complexes formed by associative phase separation was solid precipitates, instead of liquid coacervate droplets. The experimental data obtained by thermogravimetric analysis under different pH environments were summarized into three binodal phase diagrams of neutral, basic and acidic conditions respectively (Figure 4.2). The phase behaviors were, to some extent, similar under neutral and basic pHs and followed the general expectations of PEC materials. However, remarkable differences were observed under the acidic conditions. The polymer content in the complex phase, instead of decreasing by addition of salt, actually increased, which were found to be caused by the hydrophobicity of the PAA hydrocarbon backbone and possible hydrogen bonding formed among the acrylic monomers. Those two factors combined might also explain the unusual high salt resistance of PAA-PAH complexes. Accordingly, besides the associative secondary phase formed by complexation, the precipitation of individual polymer is also contributing to the formation of the solid precipitate in the system.

Concurrently, experiments, theory, and simulation for block polyelectrolyte have made advances in predicting dynamic charge correlation phenomena and various thermodynamic material properties. We have established the synthetic capabilities and protocols to rapidly generate libraries of designer polyelectrolyte assemblies. An example of this is shown in Figure 4.3, depicting a representative strategy for using aqueous reversible addition-fragmentation chain transfer (RAFT) polymerization to prepare a series of styrenic diblock

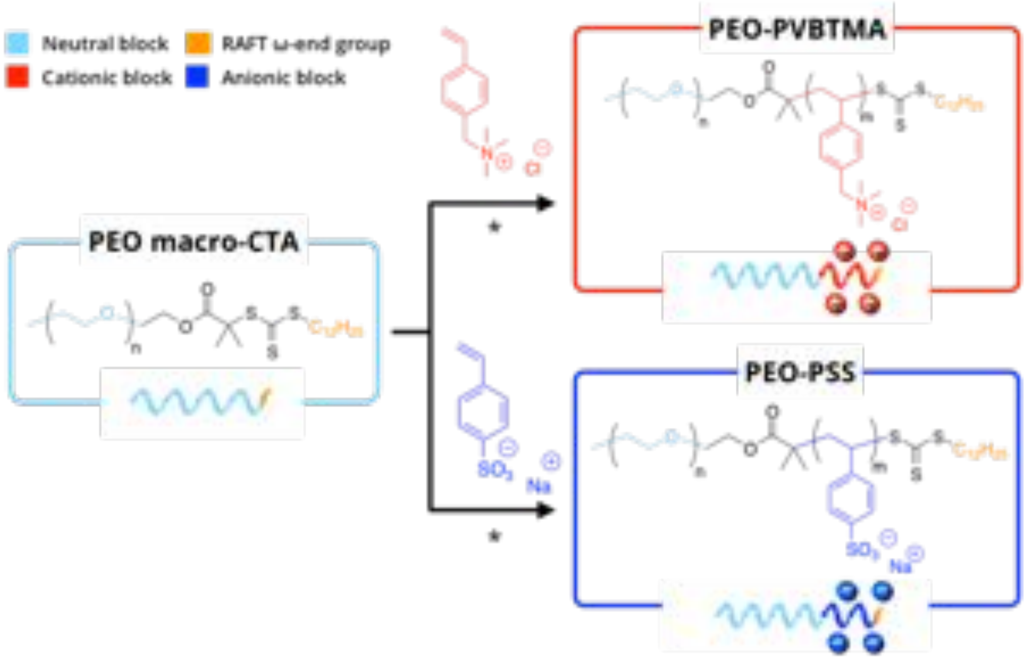


Figure 4.3: Aqueous RAFT polymerization scheme to produce gram-scale quantities of tunable designer polyelectrolytes for screening chain lengths and chemical architectures.

polycations and polyanions in a parallel synthesizer. This has expanded the ability to create materials datasets from an experimental standpoint.

Micellization and Self-assembly Via Polyelectrolyte Complexation With an initial set of well-defined materials characterized, combinations of block polyelectrolytes were systematically studied to explore the spatiotemporal formation, evolution, and disassembly dynamics of complexation-driven micelles. These aspects of polyelectrolyte solution assembly (unlike their amphiphilic block polymer counterparts) are not fully understood and lack experimental measurements to complement existing theories and simulations. To this end, synchrotron small angle X-ray and neutron scattering (i.e., SAXS and SANS) at several U.S. national labs have served as key instrumentation in pursuing these endeavors. These findings will be finalized and are poised to provide insight to predictive capabilities for ion-containing soft matter design.

In addition, we are also currently synthesizing polyester-based polyelectrolytes and their block copolymers with polyethylene oxide (PEO) to study the structure, properties, and dynamics of the bulk and complex core micelles of the electrostatically assembled materials. Our working hypothesis is that the backbone characteristics - such as hydrophobicity and rigidity - influence assembly and dynamics in the complex core micelles. To extract diffusion coefficients of the polyelectrolyte chains in the bulk complexes and complex core micelles we aim to carry out forced Rayleigh scattering experiments using block polyelectrolytes labeled with photo responsive dye. Figure 4.4 provides an overview of this synthetic platform.

The polyelectrolyte homopolymers are obtained via the ring-opening polymerization of allylated valerolactone (AVL) affording polyester prepolymers with pendant double bonds which can be subsequently functionalized with a wide variety of charged moieties using thiol-ene click chemistry. To this date, we have synthesized and purified

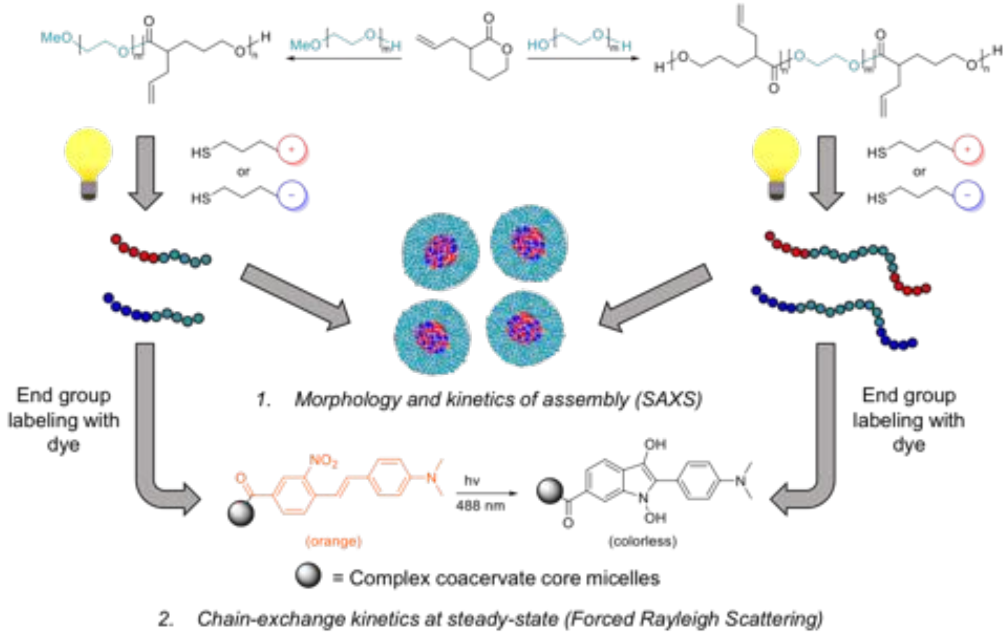


Figure 4.4: Schematic of the synthesis and characterization of block polyelectrolytes, end-functionalized to study the in situ dynamics of micelle assemblies

the allylated monomer (AVL) and polymerized it using the benign organocatalyst 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD) and benzyl alcohol as the exogenous initiator to afford molar masses of 4 and 8 kg/mol with dispersities of 1.02. Initiation from a PEO macroinitiator has not been reported, and we are at this point investigating the proper conditions for the efficient production of PEO/PAVL block copolymers. PAVL-based polyelectrolytes and their block copolymers with PEO will yield an interesting comparison to our previously studied PEO-PAGE and PEO-PS derived polyelectrolytes.

Finally, the complexation of PEC micelles formed by DNA oligonucleotides with (1) poly(lysine) (pLys) and (2) poly((vinylbenzyl) trimethylammonium chloride) (pVBTMA) was investigated. pVBTMA is a styrenic polycation with higher charge density and increased hydrophobicity compared to poly(lysine), as well as a permanent positive charge due to its quaternary amine. While these differences suggest increased complex stability, it was found that pVBTMA in fact complexes oligonucleotides more weakly than poly(lysine), as measured by stability vs monovalent salt. At physiological ionic strength, however, PEC micelles formed from both cationic blocks exhibit very similar structure-property relationships, with PEC micelle radius determined by the size of the cationic block and shape controlled by the hybridization state of the oligonucleotides. Figure 4.5 summarizes these findings. Overall, these observations narrow the design space for optimizing therapeutic PEC micelles and also provide new insights into the physics of polyelectrolyte self-assembly.

Hydrophobic and Solvation Effects Understanding the electrostatic interactions in ion-containing polymers is important to achieve better design of shape memory polymers and ion-conducting membranes for multiple energy storage and conversion applications. In molten polymers, the dielectric permittivity is low, generating strong ionic correlations that lead to clustering of the charged species. Hence, the influence of electrostatic interactions on the nanostructure of randomly charged polymers (ionomers) was studied using coarse-

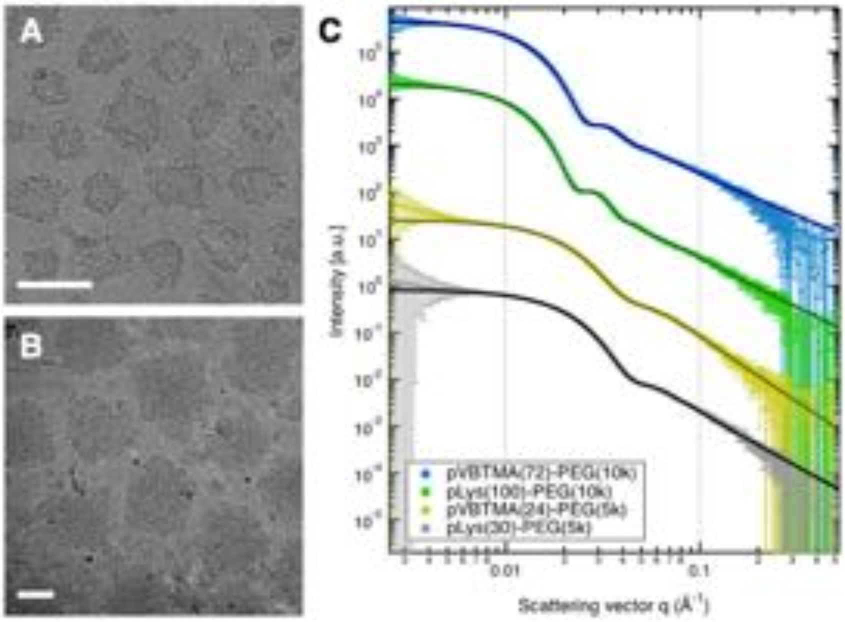


Figure 4.5: Select examples of (A)-(B) cryo imagining pLys/pVBTMA systems with oligonucleotide DNA and (C) SAXS analysis of these systems at various block lengths.

grained molecular dynamics simulations. A classic bead-spring model was applied to represent the model random ionomer melts. Pair correlation function, scattering function, and clustering of charged monomers and counter ions were studied with different concentrations of charged monomers and electrostatic interaction strengths, in order to understand electrostatic effects on ionomer chain morphology and cluster formation. Densely packed branched structures rich in charged species are found as the strength of the electrostatic interactions increases. Polydispersity in charge fraction and composition combined with ion correlations leads to percolated nanostructures with long-range fluctuations. This study identified the percolation point at which the ionic branched nanostructures percolate and offers a rigorous investigation of the statistics of the shape of the aggregates. The extra degree of freedom introduced by the charge polydispersity leads to bicontinuous structures with a broad range of compositions, similar to neutral random copolymers. These findings provide insight into the design of conducting and robust nanostructures in ion-containing polymers. In addition, this work offers insights to deouple the complicated effects due to the existence of both miscibility (characterized by Flory-Huggins parameter) and ionic interaction (characterized by Bjerrum length or electrostatic coupling parameter), elucidating the behavior of more complicated multicomponent charged copolymers, including random charged-neutral block copolymers and charged-neutral (hydrophilic-hydrophobic) random copolymers in organic solvents to construct synthetic membraneless organelles. This work was published in the journal ACS Nano [27.71].

Another efforts was the investigatation of the regimes of dielectric mismatch and ionic correlations effects in charge-containing polymer blends, using both theory and simulation techniques to study the effect of both ionic correlations and dielectric mismatch on the miscibility of an ionomer blend. Coarse-grained Molecular Dynamics (MD) simulations with explicit ions first show that ionic correlations lead to strong segregation. The MD simulations verify the thermodynamic analysis coupled with liquid state theory that includes non-linear and many body effects (Debye-Hückel Extended Mean Spherical Approxima-

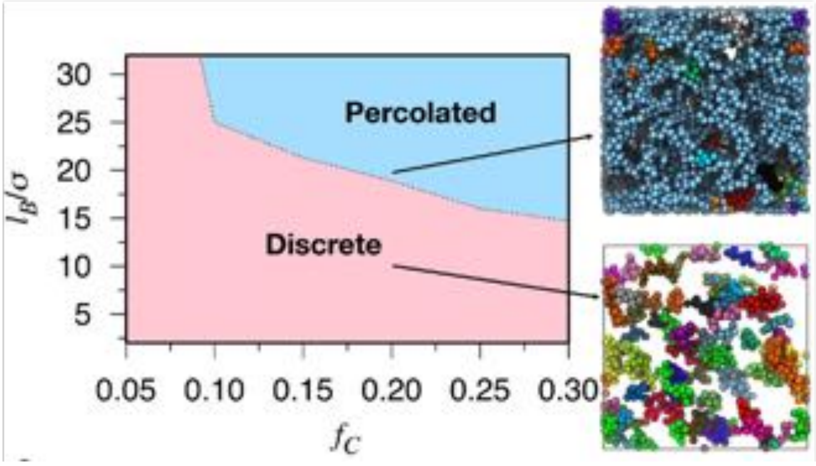


Figure 4.6: Phase diagram obtained by analyzing the distribution of ionic clusters

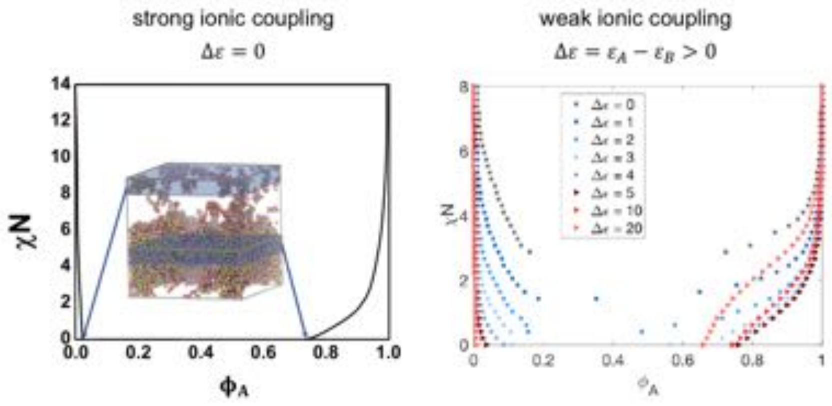


Figure 4.7: Strong ionic correlations dominate the phase behavior when dielectric mismatch is low; however, solvation effects dominate when the mismatch is high.

tion closure) that were used to study charged-neutral polymer blends. The theory was further extended to analyze the effects of the dielectric mismatch. Results show that strong correlations between ions dominate the phase behavior at low dielectric mismatch; however, when dielectric mismatch is high, solvation effects dominate, where the preference of the ions to be solvated in a higher dielectric medium is balanced by the entropic driving force for a uniform distribution of dielectric constant throughout the system. In a binary blend with no added salt, the introduction of solvation effects results in an initial decrease in miscibility followed by subsequent increase, especially for charge-rich blends. When salt is added as a third component, the effect of dielectric mismatch is significant only at low concentrations of salt. In addition, changes to the miscibility of the system due to solvation effects become dependent on the composition and dielectric constant of the blend. In regions with low charge concentration (low dielectric constant), increasing the mismatch enhances miscibility. In regions of high charge concentration (high dielectric constant), the blend becomes less miscible with increasing dielectric mismatch. This work was published in the journal Macromolecules [27.72].

Finally, it is known that the interactions between polymer molecules in solution are strongly affected by the way that the constituent polymers interact with the solvent. To address this issue, a mixed solvent system (dimethyl sulfoxide and ethylene glycol) was developed to tailor the strength of the hydrogen-bonding interactions between partially quaternized poly(4-vinylpyridine) [QVP] and poly(methacrylic acid) [PMAA]. The charge introduced by the quaternization reaction enables homogeneous solutions to be formed over a large concentration range, even in the presence of attractive hydrogen-bonding interactions between the proton-donating PMAA and the proton-accepting QVP. It was found that while the charges are important in determining the phase behavior of the system, the mechanical response is dominated by the hydrogen bonding. This work was published in the journal Macromolecules [27.73].

4.4 Impact Statement for CHiMaD Phase I

The Soft Matter Design Based on Charge Complexation use-case group has, in its first five years, made major progress on, and largely achieved many of, the objectives in the first two columns of the System Design Chart (Figure 4.1). The next five years will focus on many aspects of Properties and Performance. Data on phase behavior and self-assembly are being, and will continue to be, deposited in the Materials Data Facility to enable their widespread use in design. The scientific output has been productive as seen here and in the individual PI reports. The collaborations between the university campuses and NIST have been broadened and extended, positioning this Use-Case for further new impact in the next five years.

4.5 Collaborations

4.5.1 CHiMaD Collaborations

Interfacial Activity of Charged Block Copolymers
Kenneth ***Shull*** *(NU), Monica* ***Olvera de la Cruz*** *(NU)*
This collaboration focuses on measuring the interfacial adsorption of partially charged block copolymers and relating it to the competition between ionic correlations and hydrophobic interactions.

Phase Diagram Analysis of Polyelectrolyte Complexes
Matthew ***Tirell*** *(UC), Marius* ***Stan*** *(ANL)*
This collaboration seeks to use Bayesian analysis coupled with MCMC (Metropolis-Hastings algorithm) to optimize and evaluate the uncertainty of experimentally-measured complex phase diagrams with more meaninful confidence intervals.

4.5.2 NIST Collaborations

Impact Toughness of Polymeric Materials
Edwin ***Chan*** *(NIST), Kenneth* ***Shull*** *(NU)*
Graduate student, Shawn Chen (**Shull** group) spent 6 months at the **Chen** lab at NIST developing an instrument for measuring the ballistic impact of materials with small, micron-scale projectiles. The work complements his work on polyelectrolytes as one of the materials classes developed for impact mitigation.

Interfacial Tension of Polyelectrolyte Complex Solutions
Vivek ***Prabhu*** *(NIST), Kenneth* ***Shull*** *(NU)*
Prabhu developed techniques for measuring the very low interfacial tension values for a

polyelectrolyte complex in equilibrium with a salt solution. The **Shull** group is making comparable measurements able to access the high concentration regime.

Scaling Laws for Polyelectrolyte Complex Micelles
*Matthew **Tirell** (UC), Debra **Audus** (NIST)*
This collaboration focuses on creating a database cataloging various micellar attributes including core-size, corona size, aggregation number etc. by varying block polymer size and composition. **Audus** at NIST is leading the work on theoretical/simulation frameworks. The intent is to combine the experimental and theoretical research to achieve scaling predictions for size and other properties of polyelectrolyte complex micelles.

Temperature Effects on Structure and Dynamics of PEC Hydrogels
*Matthew **Tirell** (UC), Juan **de Pablo** (UC), Vivek **Prabhu** (NIST)*
This collaboration focuses on understanding the effects of temperature on the structure and dynamics of polyelectrolyte complex based hydrogels by synthesizing model polyelectrolytes at UC and employing neutron scattering and dynamic light scattering studies at NIST.

Phase Behavior and Salt Partitioning in Polyelectrolyte Complexes
*Matthew **Tirell** (UC), Juan **de Pablo** (UC), Vivek **Prabhu** (NIST), Debra **Audus** (NIST)*
There is an ongoing discussion between NIST and CHiMaD groups on broader issues of the proper representation of the phase behavior of polyelectrolyte complexes, experimentally and theoretically.

4.5.3 External Collaborations

Phase Behavior and Salt Partitioning in Polyelectrolyte Complexes
*Matthew **Tirrell** (UC), Juan **de Pablo** (UC), Muzhou Wang (NU)*
The **Tirrell** and **de Pablo** groups regularly hold meetings with **Shull**, **Olvera de la Cruz**, Wang (NU) groups and **Prabhu** and **Audus** on broader issues of the proper representation of the phase behavior of polyelectrolyte complexes, experimentally and theoretically. The **Tirrell** Lab including ANL staff scientist Wei Chen (ANL) is collaborating with the **Shull** Lab on producing well-defined polymers for these studies by anionic synthesis.

Polyelectrolyte Dynamics with Super-resolution Microscopy
*Matthew **Tirrell** (UC), Muzhou Wang (NU)*
Jeffrey Ting (NIST-CHiMaD postdoc) of **Tirrell** group is leading this collaboration on polyelectrolyte dynamics in polyelectrolyte complex hydrogels. We are interested in non-invasive imaging approaches to characterize polyelectrolyte dynamics through super-resolution optical microscopy.

Polymer Complexes
*Jos Zwanikken (University of Massachusetts Amherst), Monica **Olvera de la Cruz**(NU)*
This collaboration focuses on calculating the chemical potential of simple electrolytes.

Porous Polyelectrolyte Complexes
*Zhang Jiang (ANL), Kenneth **Shull** (NU)*
The aim of this collaboration is to use a salt-induced phase separation process to produce membranes for use in water purification or antifouling applications.

Block Copolymer-Based Polyelectrolytes
*Wei Chen (ANL), Kenneth **Shull** (NU)*

This collaboration focuses on the synthesis of model polyelectrolyte systems with well-controlled architectures.

Dynamics in Polyelectrolyte Complex Hydrogels
*Matthew **Tirrell** (UC), Wei Chen (ANL), Kenneth **Shull** (NU)*
This collaboration aims to producing well-defined polymers using anionic synthesis to study charge complexation in soft materials.

Metal-core Micelles for Enhanced Contrast in Liquid Cell TEM
*Matthew **Tirrell** (UC), Yuzi Liu (ANL), Shaul Aloni (Lawrence Berkeley National Laboratory), Jun Ling (Zhejiang University)*
This project explores various metal binding polymers with polyelectrolytes for metal-core micelle formation. These micelles are then used to study micelle-micelle interactions in real time via liquid cell TEM.

RAFT Polymerization in Complex Solvents
*Matthew **Tirell** (UC), Theresa Reineke (University of Minnesota)*
This collaboration focuses on new polymerization chemistry development that can be amenable to high-throughput materials development capabilities.

Diffusive Behavior of Polyelectrolyte Complex Solutions
*Matthew **Tirell** (UC), Bradley Olsen (MIT)*
This collaboration focuses on polyelectrolyte dynamics. Promising block polyelectrolytes are conjugated with fluorescent markers to conduct Forced Rayleigh Scattering experiments in the Olsen (MIT) Lab.

4.6 Technology Transfer

Software
χ-DB Database
J. De Pablo, I. Foster, D. Audus
Link: http://pppdb.uchicago.edu/
This digital handbook of properties contains 388 χ values for 120 polymers, 30 solvents. It includes 237 χ values for polymer blends of 63 unique polymers, which exceeds the 134 χ values for polymer blends of 41 unique polymers found in the Physical Properties of Polymers Handbook. The χ DB digital handbook also includes 84 newly published/measured χ values measured from 2010 to 2015 while the handbook was last published in 2007.

Data
Datasets Uploaded to Materials Data Facility in 2018

1. Phase Behavior and Salt Partitioning in Polyelectrolyte Complex Coacervates
   *Li, L; Srivastava, S; Andreev, M; Marciel, AB; **de Pablo**, JJ; **Tirrell**, MV*
   DOI: 10.18126/M2VP8X

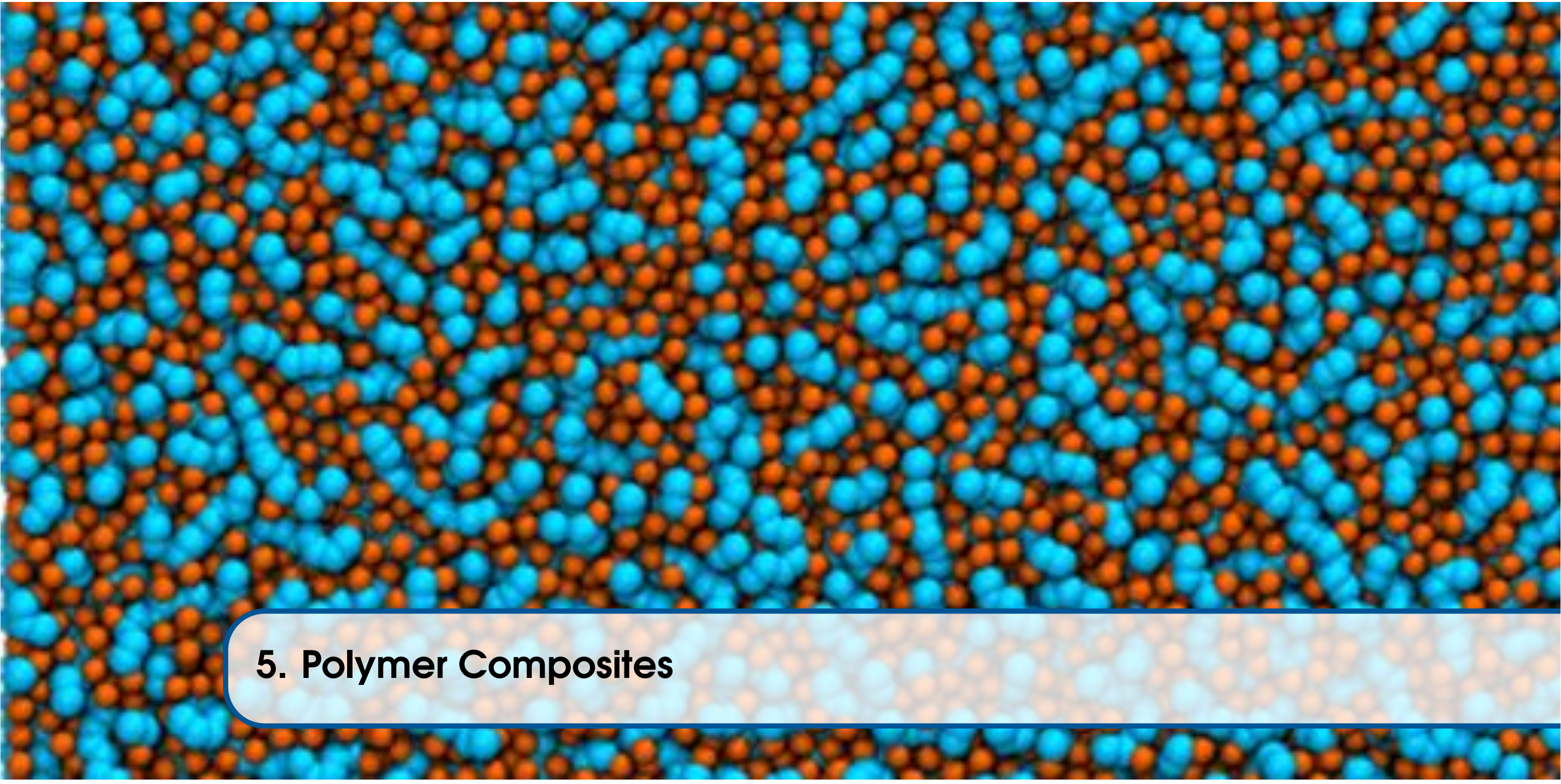
4.7 Publications and Presentations

14 Number of CHiMaD-supported publications in 2018 by Soft Matter Design Based on Charge Complexation use-case group. Please see chapter 27 for details. [27.68, 27.69, 27.106, 27.71, 27.72, 27.73, 27.74, 27.75, 27.76, 27.78, 27.79, 27.80, 27.81, 27.82]

33 Number of presentations on CHiMaD supported research in 2018, please see section 26.3 for the complete list.

4.8 CHiMaD Team

Soft Matter Based on Charge Complexation Use-Case Group Researchers			
Name	**Position**	**Affiliation**	**Project (PI)**
Matthew Tirrell Juan de Pablo Monica Olvera de la Cruz Kenneth Shull	Principal Investigator Principal Investigator Principal Investigator Principal Investigator	UC/IME UC/IME NU/MSE NU/MSE	
Jeffrey Ting	Postdoctoral Researcher	UC-NIST	Dynamics and molecular mobility in electrostatically self-assembled block copolymers *(Tirrell)*
Victor Pryamitsyn	Researcher	NU/MSE	Coarse Grained Simulations of Organic Polymer Solar Cells *(Olvera de la Cruz)*
Alexander Marras	Postdoctoral Researcher	UC/IME	Development and characterization of polymer-biomolecule assemblies *(Tirrell)*
Angelika Neitzel	Postdoctoral Researcher	UC/IME	New Syntheses and Dynamics in Polyelectrolyte Assemblies *(Tirrell)*
Ashley Guo	Graduate Student	UC/IME	Biomolecule aggregation and detection *(de Pablo)*
Jiyuan Li	Graduate Student	UC/IME	Biomaterials *(de Pablo)*
Hadi Ramezani-Dakhel	Graduate Student	UC/IME	Biomaterials *(de Pablo)*
Ha-Kyung Kwon	Graduate Student	NU/MSE	Biomaterials *(Olvera de la Cruz)*
Boran Ma	Graduate Student	NU/MSE	Polymer Blends *(Olvera de la Cruz)*
Kazi Sadman	Graduate Student	NU/MSE	Deposition, Mechanics & Equilibrium Structure of Polyelectrolyte Complexes *(Shull)*
Shawn Chen	Graduate Student	NU/MSE	Creep and Fracture of Polyelectrolyte Films in the Non-liner regime *(Shull)*



5. Polymer Composites

Kenneth Shull (NU), Sinan Keten (NU), Greg Olson (NU), Erik Luijten (NU)

Jeffrey Gilman (NIST), Frederick Phelan Jr (NIST), Jack Douglas (NIST), Stephan Stranick (NIST), Jan Obrzut (NIST), Ketan Khare (NIST), Jeremiah Woodcock (NIST), Gale Holmes (NIST), Christopher Soles (NIST), Edwin Chan (NIST), Douglas Fox (NIST)

Significance

While polymer composites are lightweight, corrosion resistant and with tailorable properties, their capabilities are vastly underutilized due to lack of: 1) understanding of multiphase composite properties under small constituent changes, 2) ability to tailor multiple physical properties simultaneously, and 3) durability predictions for long-term properties under use conditions. Tackling these challenges requires a fundamental understanding of the nature of polymer behavior near interfaces (the interphase) and ability to predict microstructure and its impact on properties. In this use case group, we pursue an integrated approach to relate macroscale polymer composite behavior to chemical constituents and kinetic behavior, and link these resources to development of high-performance modeling and predictive tools. The research includes experimental investigations mapping local physical properties, simulations and analysis to predict microstructure and interphase formation, and a data analytics approach to curate, visualize, and develop analysis and design tools.

The focus of the Polymer Matrix Materials use-case group is initially on polymer nanocomposites, with a long term vision for traditional and hybrid composites. The key of multiphase polymer response is to understand the interaction of filler particles with the matrix material and to explore the physical properties of the interphase zone. This is accomplished both by experimental efforts and physics-based modeling using Molecular Dynamics (MD) and Monte Carlo (MC) simulations to predict interphase polymer response as a function of confinement, crosslinking density, monomer and filler chemistries, filler dispersion and environmental effects such as moisture absorption. At the same time, the researchers of the Polymer Matrix Materials use-case are assembling the known data into a central resource, which will be used with supervised learning methods to identify overarching trends. Combining the physics based methodology with the curated data and tools will *provide an unprecedented ability to develop new predictive models of composite behavior.*

5.1 Design Goals

The ultimate goal for the Polymer Matrix Materials use-case is to develop databases, models and tools to enable prediction of hierarchical composite behavior based on constituent components and processing. The systems design chart of for the Polymer Composites use-case group is shown in Figure 5.1.

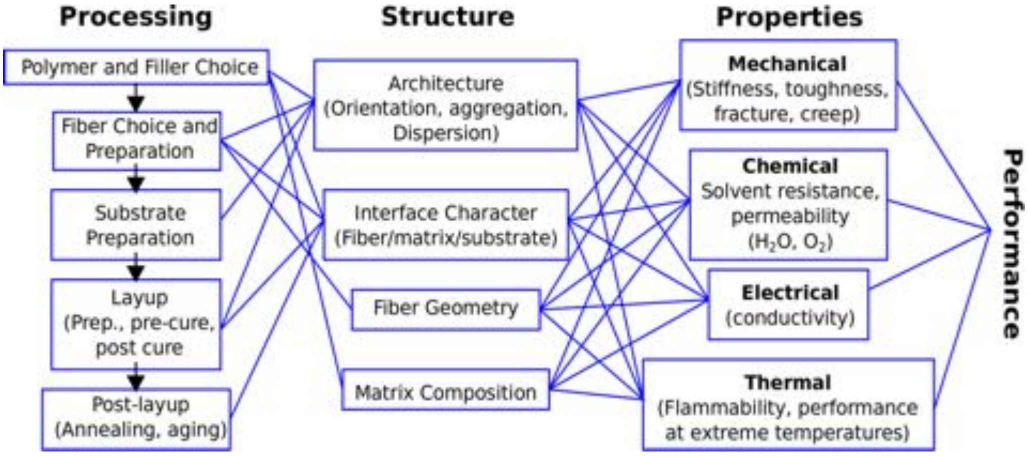


Figure 5.1: The system chart for the polymer composites use case group.

In short term this use-case aims to;

- Develop the fundamental understanding of interphase properties in cellulose-polymer nanocomposites
- Develop databases, models, and tools to manipulate and design interphase properties in cellulose-polymer nanocomposites and thus control their bulk physical properties (e.g. diffusion, modulus, T_g)
- Develop design rules to tune properties and control service life (e.g., double interphase modulus and double freeze-thaw cycles)

5.2 Significant Accomplishments in 2018

Accomplishments in 2018 involved the ongoing development and application of simulation tools for characterizing the nanoscale structure of composite resins and interfaces. These are listed in the research accomplishments section below. The Polymer Composites use-case group has also developed a new focus for the second funding cylce, which involves low temperature/high rate fracture processes. Techniques have been developed for quantifying the fracture toughness of composite resins at controlled temperatures down to $-100°C$, in addition to methods for quantifying the cure kinetics and viscoelastic properties of epoxy resins at time-scales of 60 nanoseconds, which overlaps with the time scales that can be modeled with course grain simulations preformed within the use case. In 2018 a model epoxy system was adopted following a decision that it is well suited for refining the design methodology being developed in the use case. Better characterization of key thermomechanical properties of this model system (and related systems) will lead to more efficient material-by-design processes. Despite the extensive use of molecular dynamics

(MD) simulations to predict certain properties of epoxies, all-atomistic simulations suffer from several length and time scale limitations. This restrict their potential to describe physical processes that require large temporal and spatial scales, such as phase separation phenomena and mechanical properties of resins that include long-chain crosslinkers. To address these issues, coarse-grained models of epoxy resins were developed suitable to be applied to the fracture problems relevant to this use case. In the future, simulation parameters will be refined by comparing simulation results to experimental data generated within the use case.

5.3 Research Accomplishments

Role of Water on the Behavior of Cellulose Nanocrystals Simulations of water adsorption and interfacial mechanics of cellulose nanocrystals with different surface modifications were carried out. The combined expertise within the use case, including atomistic molecular dynamics and grand-canonical Monte Carlo simulations, made it possible to efficiently examine a wide range of surface modifications as well as the underlying mechanisms that affect water adsorption. This work lead in collaboration by **Luijten** and **Keten** groups, elucidated the experimental observations reported by NIST. This work was published in ACS APPLIED MATERIALS & INTERFACES [27.60].

Mechanical Properties of Hairy Nanoparticle Assemblies Matrix-free polymer-grafted nanocrystals, also known as assembled hairy nanoparticles (aHNPs), shown in Figure 5.2a, can significantly improve the thermomechanical properties of traditional polymer nanocomposites. However, effective strategies for improving the stiffness and toughness of these nanoparticles are lacking, given the general conflicting nature of these two mechanical properties and the large number of molecular design parameters that are available. A computational framework was developed through a collaboration of **Keten** and **Chen** groups which combines multi-response Gaussian process metamodeling and coarse-grained molecular dynamics simulations to simultaneously achieve optimal mechanical properties of the polymer grafted nanoparticles within a defined parametric space. Combining data-driven, machine learning techniques with simulations can speed up parametric searches by 6-7 orders of magnitude relative to equivalent atomistic studies. The data-driven models can also populate Ashby material design plots (Figure 5.2) that, once validated by experiment, theory, or further simulations, can be used to accurately guide material selection. Taking poly(methyl methacrylate) grafted to high-aspect ratio cellulose nanocrystals as a model nanocomposite, this multi-objective design optimization framework revealed the Pareto frontier, marking the upper-bound of mechanical properties within the design parameter space. In particular, the Gaussian process metamodel was used to show that, in order to reach the Pareto frontier, a nanoparticle weight percentage greater than 60% and grafted chains longer than the critical length scale governing transition into the semi-dilute brush regime were required. This work was published in ACS Nano [27.54].

Coarse Grained Modeling of Glass-forming Liquid Soft condensed matter characteristically exhibits a strong temperature dependence of relaxation properties due to glass formation. However, the prediction of their dynamic properties remains difficult as no effective temperature transferable coarse-graining method exists. This fundamental problem has been addressed through an energy-renormalization scheme in conjunction with the localization model of relaxation. Taking ortho-terphenyl (OTP) as a model glass-forming liquid, it was shown that preserving the Debye-Waller Factor $< u^2 >$via renormalizing the cohesive

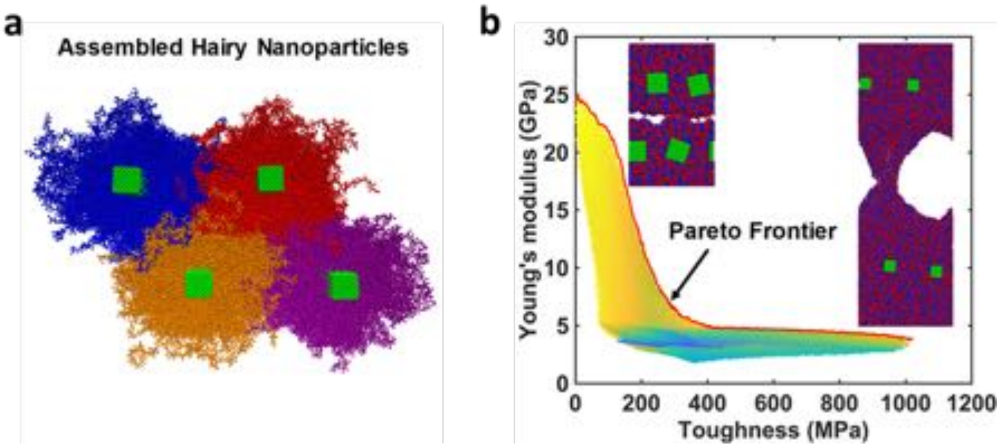


Figure 5.2: (a) Schematic of the assembled hairy nanoparticles (aHNPs) and (b) Young's Modulus vs toughness plot revealing the Pareto frontier with insets showing the range of failure from brittle to ductile observed in aHNPs.

interaction under coarse-graining allows for quantitatively accurate predictions of both short and long-time dynamics throughout the entire temperature range of glass formation. This collaborative work between **Keten**, **Douglas** and **Phelan** groups was published in Journal of Physical Chemistry B [27.61].

Coarse Grained Modeling of Epoxy Resins The superior thermomechanical properties of epoxy resins have led to their adoption in a wide range of applications, most notably as structural adhesives and matrix materials in fiber-reinforced composites. Better characterization of key thermomechanical properties can lead to more efficient material-by-design processes. Despite the extensive use of MD simulations to predict certain properties of epoxies, all-atomistic simulations suffer from several length and time scale limitations. This restrict their potential to describe physical processes that require large temporal and spatial scales, such as phase separation phenomena and mechanical properties of resins that include long-chain crosslinkers. To address these issues, the **Keten** group is working on developing coarse-grained models of epoxy resins. The preliminary results show that once the bonded potential parameters have been obtained from the corresponding atomistic trajectories using the Inverse Boltzmann Method, the same set of optimized non-bonded interaction parameters is sufficient to capture the mechanical properties of resins with different degrees of cross-linking. Furthermore, we plan to incorporate breakable bond potentials, which will enable predictive modeling of the failure behavior and mesoscale fracture mechanics of epoxy resins with greater computational efficiency.

Dynamics of Epoxy Curing The dynamic of epoxy curing was studied at room temperature, which is essential for the epoxy prepreg applying procedure. The applying of repreg requires both the viscosity and adhesion are in a right region to make sure it is viscous enough to fit the surface but sticky enough to hold itself and the structure (Figure 5.3)). The effects from molecular weight of linear cross-linker to the cross-linking dynamic were discovered. The higher molecular weight of the cross-linker of the same system, the slower the cross-linking is. Meanwhile, rheology of epoxy at cryogenic condition (e.g. in liquid nitrogen and oxygen) determines the behavior and application of epoxy composite at that condition. However, only very limited research has been done in that area. A primary research utilizing the Quartz Crystal Microbalance (QCM) technique was initiated by

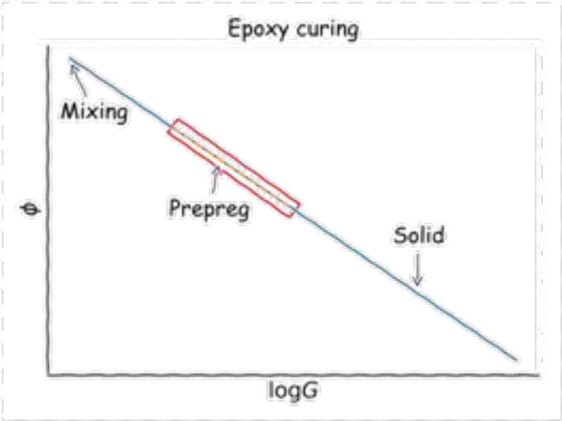


Figure 5.3: Schematic illustrated of the mechanic property-changing while the epoxy curing

Shull group and more promising results are expected in the coming year.

5.4 Impact Statement for CHiMaD Phase I

During CHiMaD's Phase I, the focus of the Polymer Composites use case group has been on the development of simulation methods and nanoscale characterization techniques that enable the composite structures to be designed more efficiently. A particular focus of these efforts has been on composite materials used for infrastructure applications, where a primary concern is maintaining material performance during long-term exposure to water. While these types of applications are still relevant to the use case, our primary focus will be on high performance composites and their performance under additional environmental extremes, including their use at very low temperatures. Many of the simulation and modeling tools needed for this effort have been developed in CHiMaD's Phase I, laying the groundwork for an increased focus on data generation in the upcoming phase.

5.5 Collaborations

5.5.1 CHiMaD Collaborations

Cellulose Nanocomposite Design

Sinan ***Keten*** *(NU), Wei* ***Chen*** *(NU)*

This collaboration investigated the design of polymer grafted cellulose nanocomposite (CNC) using molecular dynamics simulations. A metamodel was built to study the mechanical properties of PMMA-CNC nanocomposite and a statistical sensitivity analysis was conducted for interpreting the model and screening out the important/unimportant factors.

5.5.2 NIST Collaborations

Coarse Grained Modeling of Glass-forming Liquid

Sinan ***Keten*** *(NU), Jack* ***Douglas*** *(NIST), Fred* ***Phelan*** *(NIST)*

This collaborative work between **Keten**, **Douglas** and **Phelan** groups, explained in detail above, was published in Journal of Physical Chemistry B [27.61].

Epoxy Composites

Qifeng Wang (NU), Jeffrey ***Gilman*** *(NIST); Christopher* ***Soles*** *(NIST), Kenneth* ***Shull*** *(NU), Sinan* ***Keten*** *(NU)*

Correlating the quasielastic neutron scattering (QENS) at the frequency range about 200 GHz with relative lower frequency rheology from quartz crystal microbalance (QCM) at about 15 MHz. In addition, correlate both experimental results with computer simulation.

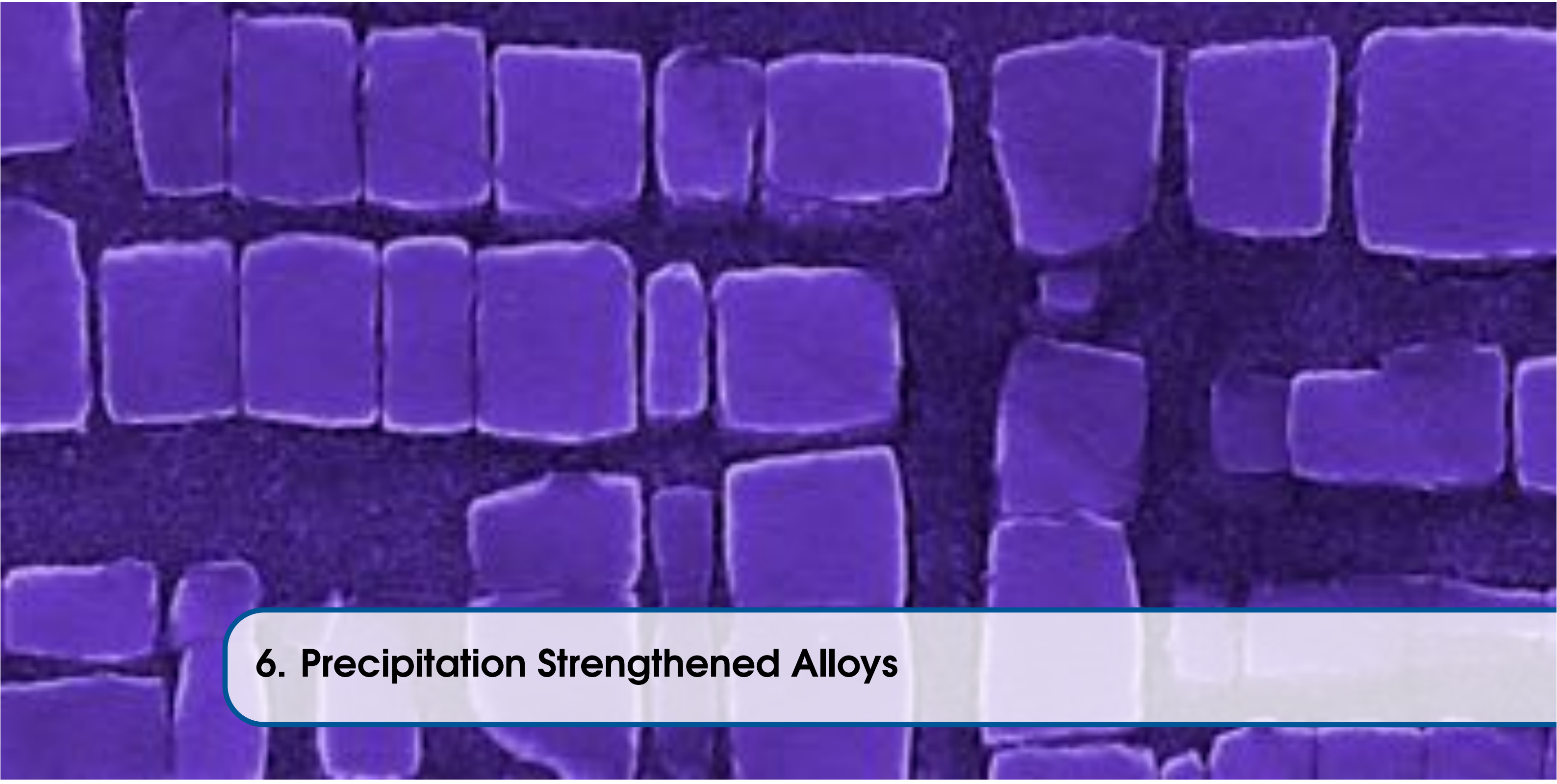
5.6 Publications and Presentations

5 Number of CHiMaD-supported publications in 2018 by Polymer Matrix Materials use-case group. Please see chapter 27 for details [27.54, 27.58, 27.59, 27.60, 27.61, 27.62].

7 Number of presentations on CHiMaD supported research in 2018, please see section 26.4 for the complete list.

5.7 CHiMaD Team

Polymer Matrix Materials Use-Case Group Researchers			
Name	**Position**	**Affiliation**	**Project (PI)**
Kenneth Shull	Principal Investigator	NU/MSE	
Sinan Keten	Principal Investigator	NU/ME	
Greg Olson	Principal Investigator	NU/MSE	
Erik Luijten	Principal Investigator	NU/MSE	
Wenjie Xia	Postdoctoral Researcher	NU-NIST	Transferable coarse-grained models for glass-forming polymers *(Keten, Douglas, Phelan)*
Qifeng Wang	Postdoctoral Researcher	NU/MSE	Carbon Fiber Reinforced Polymer Design *(Shull, Industry funded)*
Zhaoxu Meng	Postdoctoral Researcher	NU/ME	Multiscale modeling and mechanical properties of epoxy resins *(Keten)*
Xin Qin	Graduate Student	NU/ME	Coarse-grained modeling of CN neat films *(Keten)*
Nitin Hansoge	Graduate Student	NU/ME	Study of mechanical properties of polymer grafted CNC assemblies *(Keten)*
Zonghui Wei	Graduate Student	NU/PHY	Predictive modeling of gelation through machine learning *(Luijten)*



6. Precipitation Strengthened Alloys

Gregory Olson (NU), David Dunand (NU), David Seidman (NU), Yip-Wah Chung (NU), Michael Bedzyk (NU), Wing-Kam Liu (NU), Alex Umantsev (FSU), Christopher Wolverton (NU), Ankit Agrawal (NU), Alok Choudhary (NU), Olle Heinonen (ANL), Peter Voorhees (NU)

Ricardo Komai (QuesTek), Jason Sebastian (QuesTek), Jeff Doak (QuesTek), James Saal (QuesTek), Ida Berglund (QuesTek), Dana Frankel (QuesTek))

Carelyn Campbell (NIST), Ursula Kattner (NIST), Eric Lass (NIST), Shengyen Li (NIST), Kil-Won Moon (NIST), Maureen Williams (NIST), Lyle Levine (NIST), Mark Stoudt (NIST)

Significance

New capabilities in predictive design of precipitation-strengthened alloys are being applied to Co-based superalloys and high-performance Shape Memory Alloys, both employing low-misfit coherent aluminide precipitates closely analogous to the γ- γ' *Ni-based superalloys. The Co-based alloys are of interest for both high-temperature creep and fatigue resistance in turbine applications, as well as ambient temperature wear-resistance applications such as aerospace bushings to replace Be-Cu alloys. The shape memory alloys are of high interest for device and actuation applications in medicine, aerospace and automotive sectors where ultrahigh-cycle fatigue is the primary limitation of current alloys; here, efficient precipitation strengthening can eliminate the accommodation slip associated with thermoelastic martensitic transformation, standing as the principal cause of cyclic instability.*

6.1 Design Goals

Short-term goals for the Co alloys address ambient temperature applications, where the intrinsic low surface friction characteristics of Co-based alloys makes their precipitation strengthened variants of clear near-term interest as a replacement for Be-Cu aerospace

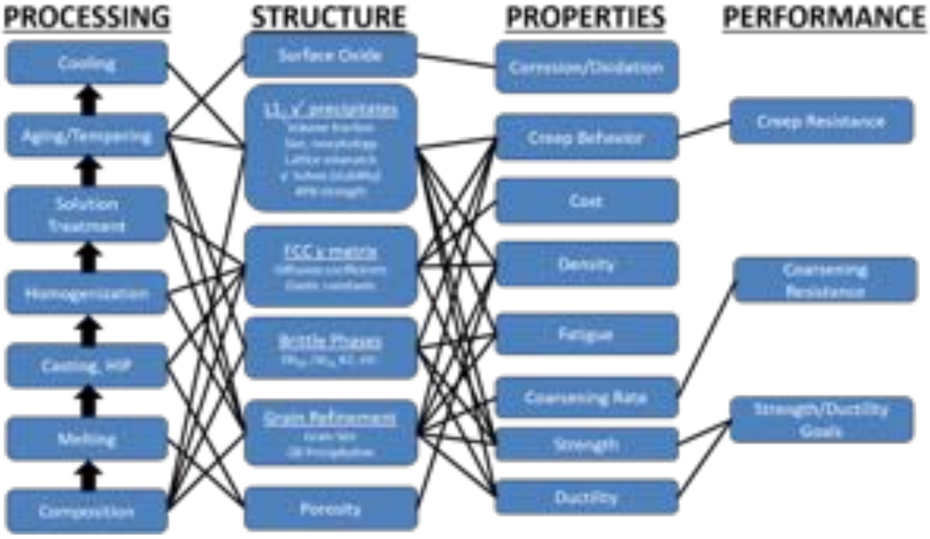


Figure 6.1: Systems design chart of Cobalt Superalloys

bushing applications of current environmental concern. Long-term goals address high-temperature applications (represented by the system chart of Figure 14.1) where metallic superalloys play a vital role in electrical power generating systems (gas and steam turbines) and jet engines. While achievement of the full set of properties necessary for these applications is an ambitious goal, the potential of the new family of Co-based superalloys to achieve the same strength levels of current Ni-based superalloys, but with a temperature capability $100-150^{o}C$ higher would enable a major advance in the fuel efficiency of gas turbines and jet engines. In support of new capabilities in additive manufacturing by 3D printing, precipitation-strengthened superalloy research also addresses process optimization in the 718+ Ni-based alloy being developed under the DARPA Open Manufacturing program, identifying processability constraints to design alloys compatible with the new manufacturing process.

The breadth of developing databases for Shape Memory alloy design support both the near-term optimization of high Pd alloys for medical applications favoring high radiopacity and low Ni content for biocompatibility (Figure 6.2), and the longer-term design of lower-cost high Zr alloys to meet the requirements of aerospace and automotive actuators. This will allow the new class of cyclically-stable alloys to have broad technological impact.

A further extension of our precipitation-strengthening research in the past year has been application of the same precipitation principles to management of thermal conductivity in multiphase thermoelectric materials under the DARPA SIMPLEX program and DARPA-MATRIX programs.

During 2018, Precipitation-Strengthened Alloys use-case group researchers have concentrated on building on their results from previous years and advancing the field in the following topics:

- Experimentation for Co-alloy fundamental data development
- Co-Based CALPHAD Database Development
- High-throughput Combinatorial Synthesis
- DFT Calculations of Diffusion in Co and Interfacial energies
- High-Performance Shape Memory Alloys

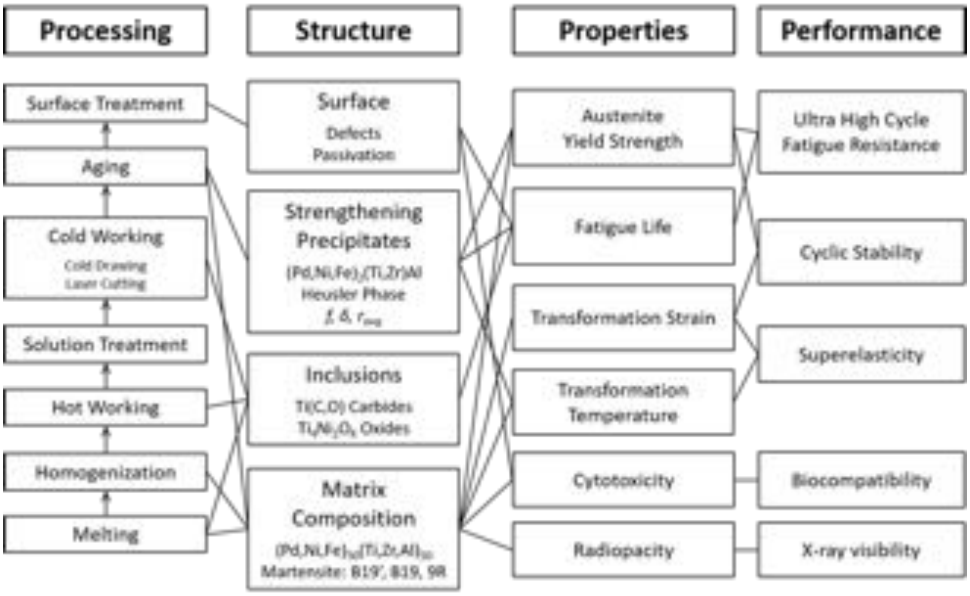


Figure 6.2: Systems design chart of High-Performance Shape Memory Alloys

- Fatigue Nucleation Modeling
- Industrial Accelerated Qualification of Materials - Co-alloy development
- Industrial Accelerated Qualification of Materials - Shape memory alloy development
- Precipitation-Strengthened Thermoelectrics and CALPHAD development
- Steel Research Group (SRG) Projects and Design Class Interactions
- First Principles Study of Al- and Mg- Alloys

A detailed explanation of research achievements in each topic can be found in the next section.

6.2 Research Accomplishments

6.2.1 Cobalt-based Superalloys

Model Alloy and Prototype Characterization Effect of Cr addition to multinary Co based superalloys (Co-Ni-Al-W-TiT-a and Co-Ni-Al-W-Ti-Mo-Nb-Ta) have been closely examined by **Dunand**, **Seidman**, and **Lass**. Contrary to literature results showing Cr deteriorates the creep performance, our study indicates Cr does not alter the creep performance in multinary Co-based superalloys, Figure 6.3. While the γ'-morphology evolves from cuboid-to-sphere due to the lattice parameter misfit reduction due to the Cr addition; therefore, its impact on creep resistance appears to be minimal. It is likely that addition of elements, such as Ti and Ta, are masking the deleterious effect of Cr. Dislocation dynamics study using TEM is being conducted to understand the effect of Cr on creep properties in multinary Co superalloys.

The recently-discovered metastable γ'-$Co_3(Ta_{0.76}V_{0.24})$-phase formed on aging in a Co-6Ta-6V (at.%) ternary alloy is stabilized by partial replacement of Ta and V with Al and Ti. In two alloys with the compositions Co-6Al-3Ta-3V and Co-5Al-3Ta-3V-1Ti with a γ plus γ' microstructure, the γ'-precipitates remain stable for up to 168 h at 850 and 900°C, with no precipitation of additional phases. Adding Ni and B and doubling the Ti concentration produces a γ/γ' superalloy, Co-10Ni-5Al-3Ta-3V-2Ti-0.04B (at.%), with γ'-precipitates, which are stable for up to six weeks of aging at 850°C, while slowly coarsening and

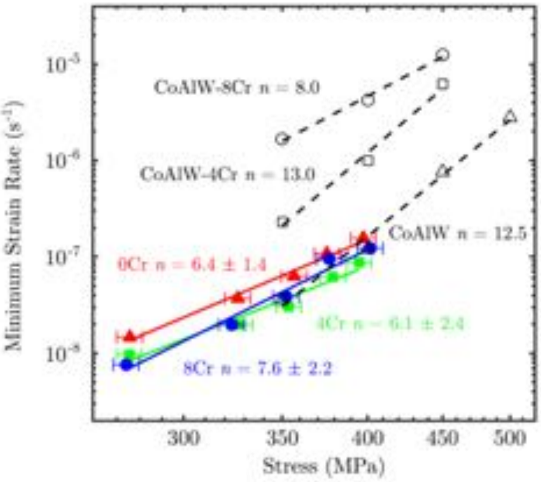


Figure 6.3: Plot of minimum strain rate as a function of applied stress for Co-Ni-Al-W-Ti-Mo-N-Ta Co-based superalloys with different concentrations of Cr. Comparisons with the archival literature data (dashed line) demonstrates that Cr does not alter the creep properties of multinary Co superalloys significantly.

coalescing to form cuboidal to elongated morphologies. After 1 day of aging at 850°C, the γ'-nanoprecipitates have the composition $(Co_{0.87}Ni_{0.17})_3(Ta_{0.42}Al_{0.23}Ti_{0.19}V_{0.15}B_{0.01})$, with Al and Ti replacing at the same rate both Ta and V in the original metastable $Co_3(Ta_{0.76}V_{0.24})$ phase. To improve the oxidation resistance, 4 at.% Cr is added to the new superalloy, resulting in a somewhat higher volume fraction of finer cuboidal γ'-precipitates after one week of aging at 850°C, but no other deleterious phases. These W- and Mo-free γ/γ' superalloys dispaly good creep resistance at 850°C, on par with two other recent Co-base γ/γ' superalloys: (i) Co-9W-9Al-8Cr (at.%), which has a higher mass density due to its high W content; and (ii) Co-30Ni-10Al-5Mo-2Nb (at.%), which has a lower density (as it is W-free) but it contains triple the Ni concentration.

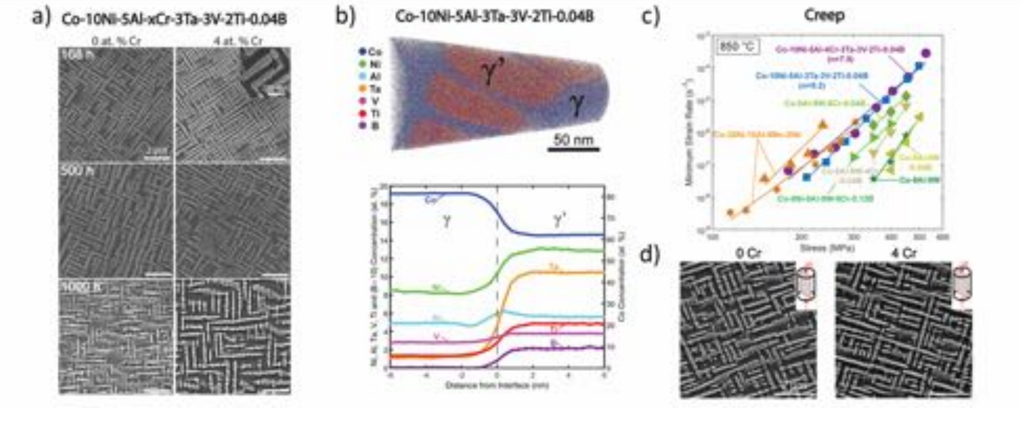


Figure 6.4: a) Microstructure evolutions as a function of time in 0Cr and 4Cr alloy after aging at 850°C; b) APT results showing partitions of constituent elements to the γ- or γ'-phase; c) creep results for 0Cr and 4Cr alloys compare to other Co-based superalloys; and d) post-creep microstructure showing minimum signs of rafting in 0Cr and 4Cr alloys.

A single-phase Co-based superalloy has been examined for mechanical properties. L26, with nominal composition Co-30Ni-11Al-5.5W-4Ti-2.5Ta-0.1B, shows a stable single-phase γ' microstructure after aging at 900°C, 1000°C, and 1100°C for 24hrs, 1 week, and 1000hrs (at 1100°C the sample was only aged for 24hrs and 1 week). This single phase γ' alloy exhibits three distinct stages of creep (Figure 6.5A) and shows creep resistance similar

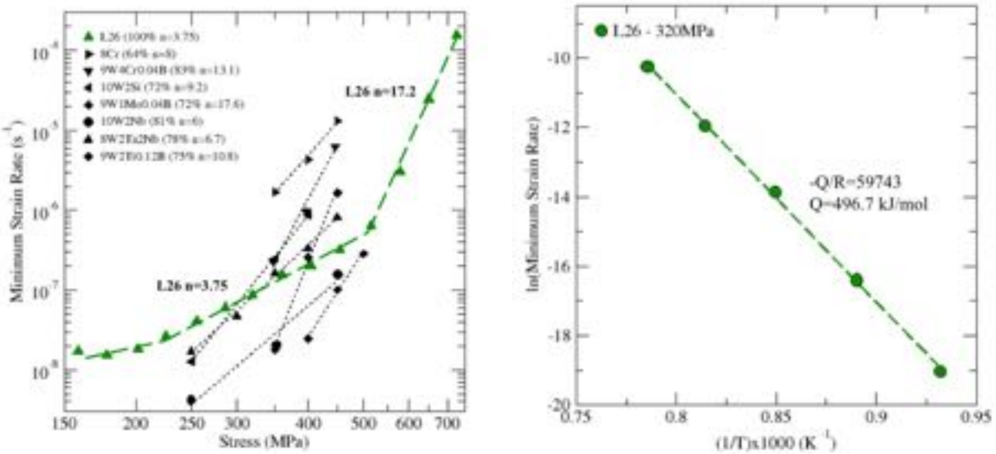


Figure 6.5: (a) Plot of minimum strain rate as a function of applied stress for pure γ' (L26) with comparative literature data (in black) of alloys with high γ' volume fractions. (b) Plot of the natural log of the minimum strain rate as a function of inverse temperature for pure γ'crept at a constant stress of 320MPa in order to calculate the activation energy (Q=496.7 kJ/mol)

to alloys with high γ' volume fractions. Figure 6.5B shows that the alloy exhibit a creep activation energy of 496.7kJ/mol at 320MPa which is slightly lower than a commercially available cobalt-base superalloy (MAR-M 302) at 525kJ/mol1.

Pre-CALPHAD Protodata: High-Throughput Combinatorial Phase Determination in Co-based systems **Bedzyk** and **Chung** have refined their methodology for generating experimental isothermal ternary phase diagrams using high-throughput combinatorial thin-film deposition and multi-modal analysis. Systems studied this year by simultaneous X-ray diffraction (XRD) and X-ray fluorescence (XRF) include Co-Mo-Re, Co-Mo-Nb, Co-Ni-Ta and Co-Ti-Ta at 900°C. The Advanced Photon Source (APS) setup at DND-CAT 5BM-C was improved by making it possible to perform high throughput measurements on a 100 mm diameter Al_2O_3 wafer with up to 2000 sample points with differing alloy mixtures. We also upgraded the vacuum furnace to accommodate these larger sapphire wafers. The samples were prepared by multi-target dc magnetron sputtering, followed by high-temperature vacuum annealing for simultaneous XRD/XRF measurements.

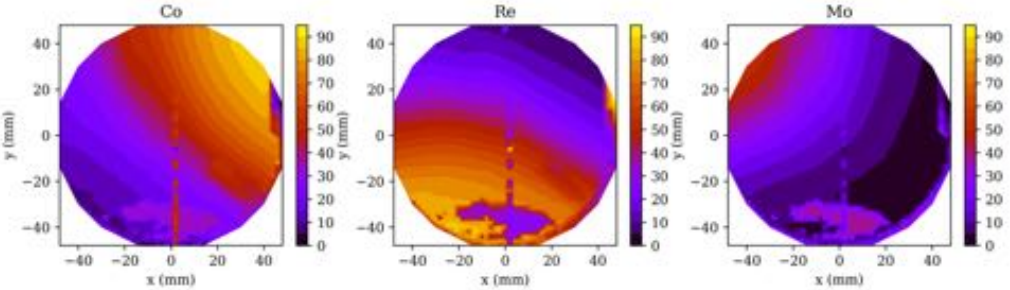


Figure 6.6: XRF determined atomic percent maps across 100 mm wafer for Co, Mo and Re.

To efficiently analyze the large data set with 2000 sampling points, each having an XRD pattern and XRF spectrum, we developed Python codes for data reduction and implemented machine learning for phase clustering. Figure 6.6 shows the XRF determined atomic-ratio

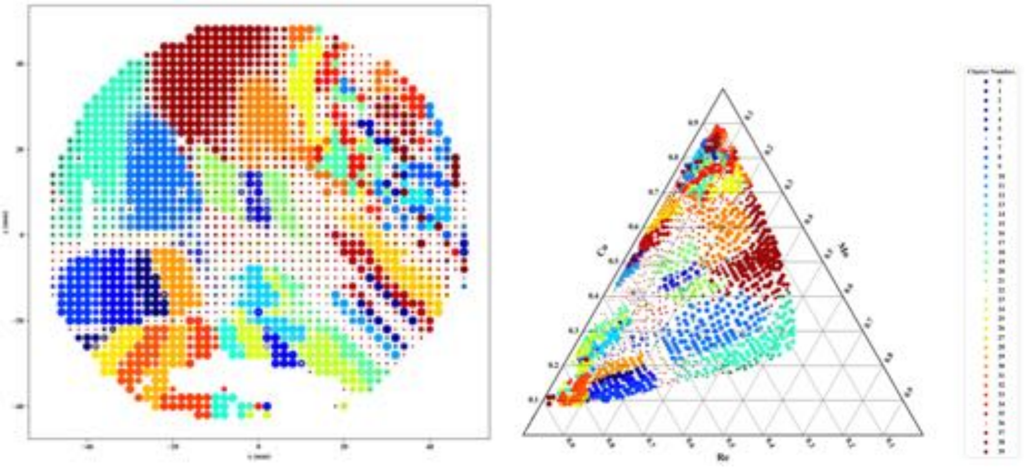


Figure 6.7: Co-Mo-Re system at 900°C. (a) XRD determined phase clusters mapped on to the 100 mm wafer. (b) shows the XRD + XRF combined determination of the phase clusters on the Gibbs triangle. The marker size is proportional to the phase fraction

maps for the Co-Mo-Re coated wafer. As can be seen the sputter deposition pattern resulted in concentration gradients of the three elements as a consequence of the three sputter source locations. Without any knowledge of the phases or materials the XRD data is clustered using machine learning, which separates out common sets of diffraction lines. Each diffraction pattern is considered as a 1500-dimensional vector corresponding to the intensity at each one of the 1500 2 values. We adapted a fuzzy c-means clustering algorithm for phase separation the resulting clusters map for the Co-Mo-Re system is shown in Figure 6.7a. This XRD determined map is combined with the XRF atomic ratios map to produce the ternary phase diagram shown in Figure 6.7b.

Bedzyk and **Chung** have maintained continuous contact with NIST collaborators (**Campbell**) in sharing results and obtaining their support and feedback in the research. They also received input from **Dunand** and **Voorhees** in choosing the systems studied, and also closely collaborating **Agrawal** to further improve the machine learning algorithms

CALPHAD Assessment The refinement of the multi-component Co-based CALPHAD thermodynamic database has been continued during 2018 by NIST CHiMaD Post-Doctoral Fellow Peisheng Wang in collaboration with **Kattner** (NIST) and **Campbell** under co-supervision of **Olson** . The major achievement during the year 2018 is the development of a new method to describe the magnetic contribution to the total energy for complex compounds, which was often ignored. Figures 6.8 show the calculated magnetic moments for the C36 phase in the Co-Ta system and calculated Co-Ta phase diagram, respectively. Compounds with magnetism transfer to the non-magnetic state according to the previous CALPHAD descriptions which is definitely not correct. In the method developed by us, all the magnetic compounds transfer to the paramagnetic state rather than the non-magnetic state. A more reliable description of the binary sub-system can lead to a more reliable prediction in higher order systems. Figure 6.9 shows the predicted isothermal section at 1073 K in the Co-Ni-Ta system, which is consistent with the experimental data by literature data and the unpublished data from the **Chung** group. The experimental studies are still ongoing. Also, the addition of the element Re was studied systematically. Several key ternary systems, Co-Ni-Re, Co-Al-Re and Co-Cr-Re have been studied. This research is still ongoing.

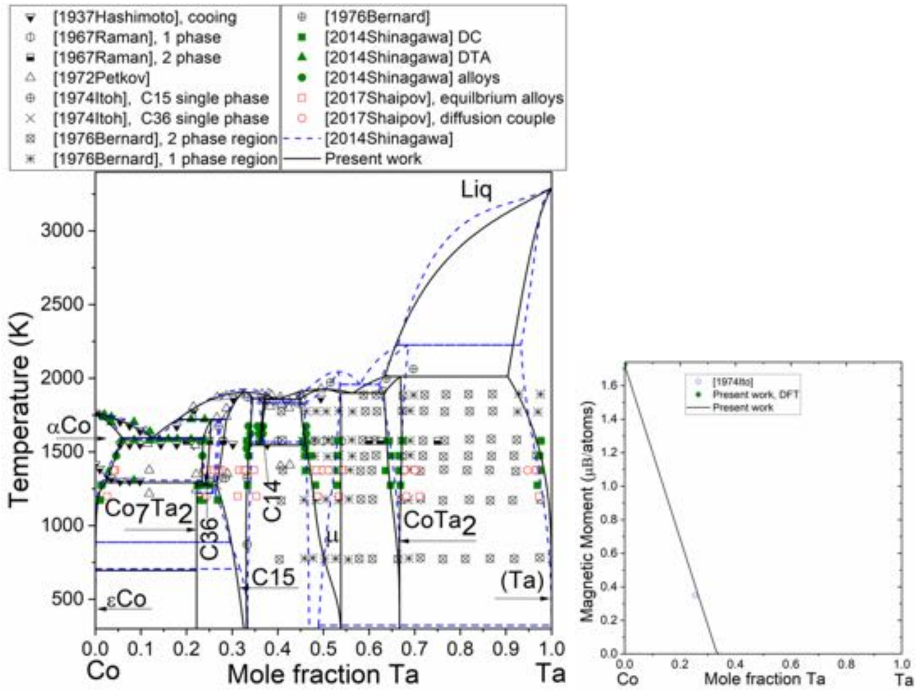


Figure 6.8: (a) Calculated Co-Ta phase diagram; (b) Calculated magnetic moment for the C36 phase in the Co-Ta system.

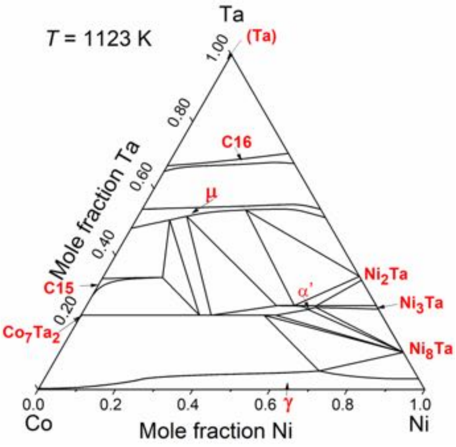


Figure 6.9: Predicted isothermal section at 1123 K in the Co-Ni-Ta system using current thermodynamic database.

Cobalt Superalloy Design Synthesis Building on our database developments, a first fully integrative CALPHAD-based design of a high-temperature Co-based superalloy was conducted in **Olson**'s MSc390 Materials Design class by a team of students coached by **Komai** and Chung (NU) of the **Dunand** group. Selecting key alloy components from attribute cross-plots developed through the ongoing collaboration of **Campbell** with Granta Design, the full thermodynamic and mobility databases were applied to achieve stable L12 dispersions, employing for the first time Re additions to reduce vacancy diffusivity for enhanced creep resistance, while retaining required Cr and Al levels for oxidation resistance. The exercise also integrated DFT estimates of APB energies of end members of the L12 Co3X system with X=Al,Cr,Ta,Ti,W by Bi-Cheng Zhou of the **Wolverton** group. The feasibility exercise identified the need to refine our molar volume databases to constrain interphase misfit to the weekly negative level desired for optimal creep performance.

6.2.2 High-Performance Shape Memory Alloys

Process-Structure-Property linkages for NiTi wire drawing and resulting fatigue strength The **Liu** group has developed a mechanistic data-driven modeling framework to connect the extrusion process modeling to fatigue crack incubation life for NiTi shape memory alloys (SMAs), Figure 6.10. This extends our Finite Elements-based work in 2017 to new, data-driven methods. The framework relies on an a priori deterministic sampling of the solution space, a K-means clustering method, and a mechanistic Lippmann-Schwinger equation solved using a self-consistent scheme. This allows for a decomposition of the computational work into an offline (one-time) solution, and an online (once per load step) solution. This method is formulated in a finite strain setting in order to model the large plastic strains that develop during metal forming processes. An efficient implementation of an inclusion fragmentation model is introduced in order to model this micromechanism in a clustered discretization. With the addition of a fatigue strength prediction method also based on data science, process-structure-property-performance relationships are predicted in the case of cold-drawn NiTi tubes. It shows inclusion fragmentation resulting in a jump in fatigue strength. Relatively large reductions are often required to achieve large gains in fatigue performance. The newly developed data-driven framework made this finding possible, by substantially decreasing the computational expense involved for each part of the simulation.

Phenomenological Model of Ductile Fracture and Fatigue In support of numerical modeling of fatigue failure, **Umantsev** has developed a Bifurcation Theory of Plasticity and Damage which includes all major components of the mechanical behavior of ductile materials and covers all regimes of viscoplastic tensile/compressive loading and unloading. The approach is based on the concepts of the internal energy and internal damage parameter. The latter characterizes a particular micromechanical mechanism of deformation and presents a convenient identifier of the stress/strain state of the material where its zero (or nearly zero) value identifies the elastic or quasi-elastic state, its positive value-the plastic state, and its critical value defines the ultimate failure of the specimen. In the present theory, we describe uniaxial tension or compression of the materials by the following expression of the internal energy:

$$u = \frac{1}{2}C(\varepsilon - \varepsilon_r)^2 + B\{\frac{1}{2}F_y(\varepsilon, \varepsilon_b)p^2 + \frac{1}{3}p^3\}$$

Here ε and p are the total strain and damage parameter, C>0 and B>0 are the elastic and plastic moduli respectively, which scale the elastic and plastic energies, F_y is the loading function, and ε_r and ε_b are the parameters, which reflect the history of the deformation

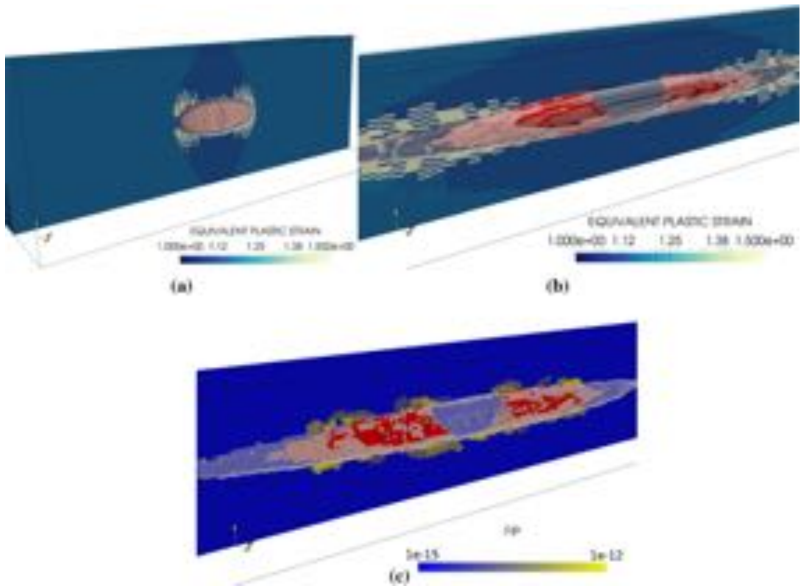


Figure 6.10: (Cold drawing and fatigue indicator parameter (FIP) computation results showing the particle fragments in red, the voids in light gray, and: a the equivalent plastic strain in the matrix in undeformed configuration, b the equivalent plastic strain in the matrix in deformed configuration after displacement reconstruction, c the FIP in the matrix on a new voxel mesh of the deformed configuration after mesh transfer. Note that the three figures are not in the same spatial scale, as section height in b, c is reduced by 45% compared to (a).

process. Transition from the elastic to plastic state in the quasi-static loading, tensile or compressive, is viewed in the theory as a transcritical bifurcation of the damage parameter while the stress/strain reversal in the plastic state (that is, unloading) is viewed as a discontinuous switch back into the quasi-elastic state. Dynamical behavior of the material in the theory has the attributes of both rate-dependent and rate-independent plasticity.

The developed approach was applied to the phenomenon of fatigue in the strain-controlled loading conditions. In Figure 6.11, the dynamic outcomes of the cyclic loading are analyzed in the phase space p(ε), (ε). The stress-strain loops (see Fig. 12a) are not completely closed but progressively rotate clockwise that is, in the sense of the material softening, which may be called rotational ratcheting. It manifests decrease of the deformation resistance of the material. In the (ε, p)-plane, see Figure 6.11b, the loops rise upward until they reach the point of failure (ε_f, p_f).

In Figure 6.12, the dynamic outcomes of the model are plotted as the functions of the elapsed time t, which helped us identify the plastic bifurcation point.

The numerical simulations demonstrated that the fatigue life of the specimen increases with the increase of the yield parameter and decreases with the increase of the work-hardening coefficient, viscosity, and frequency of cycling. The Coffin-Manson relation, see Figure 6.13, helped identify various regimes of this processes; low-cycle and high-cycle fatigue.

Quantifying Size Dependence of Fatigue Nucleant Potency in TiNi Alloys While increasing matrix strength through nanoscale dispersion is vital to cyclic stability, a large number of high cycle fatigue experiments showed that the fatigue resistance of SMAs could be improved via controlling non-metallic inclusion (NMI) size which act as the potent fatigue nucleants. A quantitative relationship, however, is still sought between the fatigue

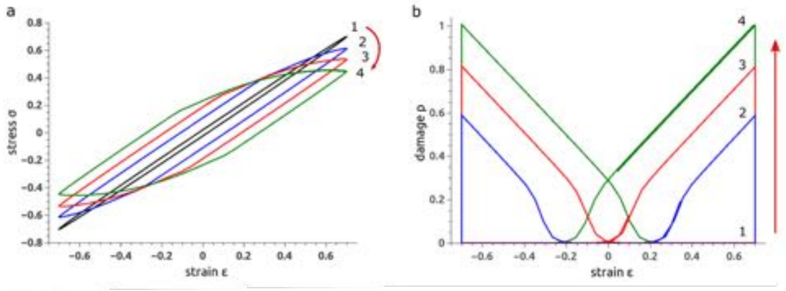
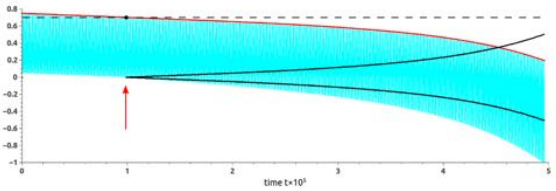


Figure 6.11: Cyclic response of the specimen in the strain-controlled fatigue experiment with the frequency $f = 1 \cdot 10^{-3}$and initial amplitude in the elastic regime. (a) (ε, σ) plane, (b) (ε, p)plane. Cycles 1,2,3,4 correspond to the initial, $430^t h$, $470^t h$ and final $495^t h$ cycles, respectively

Figure 6.12: Time evolution of the same specimen as in Figure 12. Light blue band - the loading function F_y ; red curve the yield strain $e(t)$; black curves - the back strain ε_b ; arrow - the plastic bifurcation point.



resistance and inclusion size. In the Predictive Science & Engineering Design program (PSED), a group of ICME master students supervised **Olson** investigated this problem through both empirical and computational methods. In the empirical model, Figure 6.14, fatigue crack nucleation potency (strain amplitude for 10^7 cycles) and NMI size were connected via cumulative number density (N_V) of NMI: on the one hand, they extracted the relationship between fatigue strain amplitude and crack nucleates number density from high cycle fatigue data of wire and diamond specimens; on the other hand, the direct observation of NMI from microscope enabled recovering 3D NMI size distribution with classic stereological analysis. Assuming all cracks nucleate from NMI, the fatigue strain was found to follow a -1/2 power law of NMI size: $\varepsilon_a(10^7) \sim D^{-1/2}$ In parallel with the development of the empirical model, a crystal plasticity finite element (CPFE) analysis in collaboration with the **Liu** group was also utilized to study the effect of NMI size on fatigue resistance. To simulate different inclusion sizes, they simplified the geometry of NMI clusters as two spherical inclusions connected by void and used different averaging volume sizes while keeping the mesh geometry constant. The results closely matched the -1/2 power law from the empirical model.

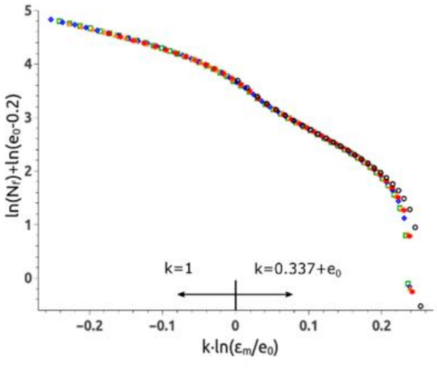


Figure 6.13: Log-log plot of the results of the effect of the initial yield parameter: $e^v_y = e_0 = 0.5$ (black), 0.75 (red), 0.85 (orange), 0.9 (blue), 1.0 (green).

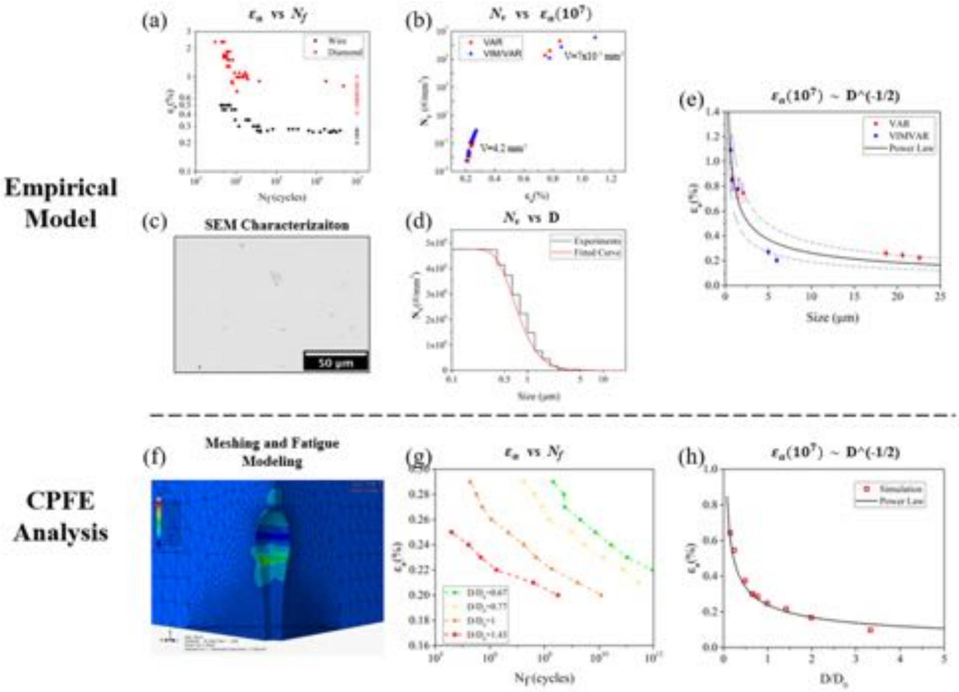


Figure 6.14: A schematic of empirical model: (a) the experimental high cycle fatigue data for wire and diamond specimens; (b) the function of crack nucleates density on fatigue strain amplitude derived from fatigue data; (c) SEM characterization of NMI; (d) the cumulative number density of NMI; (e) the -1/2 power law: $\varepsilon_a(10^7) \sim D^{-1/2}$. And the CPFE analysis results: (f) stress concentration on the interface between NMI and matrix during loading and unloading; (g) the simulated fatigue life at varying strain level for different NMI size; (h) the simulated results match the -1/2 power law.

Multicomponent Shape Memory Alloy Design **Olson** group focused on the systematic design of fatigue resistant NiTi-based multicomponent shape memory alloys, in collaboration with **Frankel**. The coherent precipitation of nanoscale L21 Heusler aluminides has already been demonstrated to provide excellent strengthening and resistance to accommodation slip. Previous research made great effort to stabilize the martensite phase by Pd additions and control the misfit between matrix and Heusler precipitates simultaneously. The softening effect of Pd additions, however, was disadvantageous to the total strength. Another martensite stabilizer, Hf, was taken to replace Pd in our new alloy design. Phase relations, precipitation kinetics, composition evolution, transformation temperatures, and crystal structures have been characterized in the NiTiHfAl system, Figure 6.15. The precipitation evolution characterized by Atom Probe Tomography (APT) revealed the partition behavior of each elements and the relatively low coarsening rates of aluminides with Hf additions, proving controlled microstructure with heat-treatment process design. And the cyclic thermal stability was greatly enhanced by aluminides precipitation during aging at 600°C and 550°C while the transformation temperatures evolved into opposite directions for these two temperatures. Mechanical tests will be utilized to characterize the effect of aluminide nano-precipitates on superelastic performance and cyclic mechanical stability.

While the primary experiments demonstrated the potential benefits of the NiTiHfAl alloy system, the predictable computational design of the final prototype requires a solid thermodynamic database. Xuyang Wang (Northwestern Polytechnic University, visiting

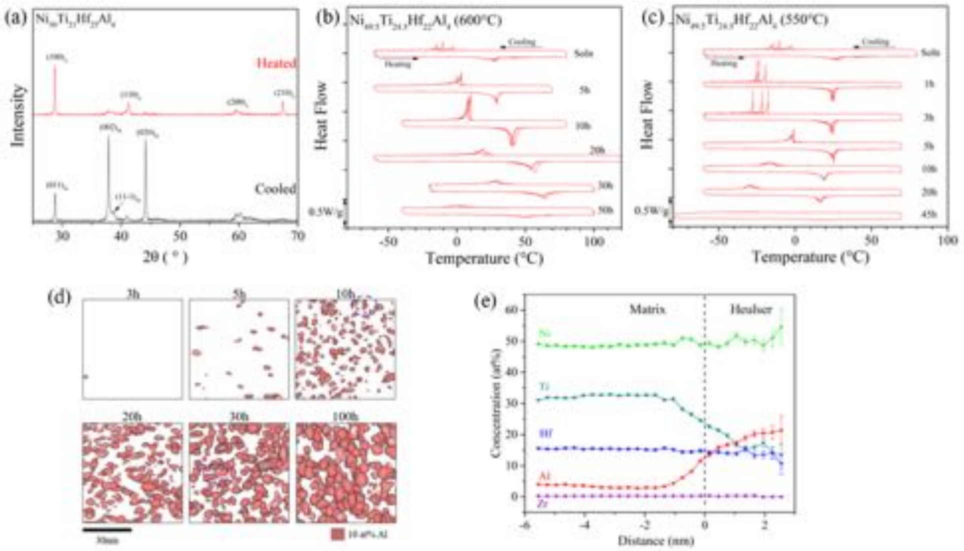


Figure 6.15: (a) The room temperature x-ray diffraction pattern of solutioned Ni50Ti21Hf25Al4 after heating/cooling process. (b, c) The DSC curves of $Ni_{49.5}Ti_{23.5}Hf_{22}Al_4$ for three cycles of cooling and heating as a function of aging treatment. (d, e) The aluminide precipitates evolution of $Ni_{50}Ti_{31}Hf_{15}Al_4$ during aging at 550°C and the element partition behavior of specimen aged for 100h.

student), collaborated with **Olson** on the reassessment of the Ni-Ti-Hf system. Three binary databases (Ni-Ti, Ti-Hf and Ni-Hf) were assessed prior to the Ni-Ti-Hf ternary system: all binary databases are optimized from published experimental data including phase diagram data, the activity coefficients, enthalpies of formation of liquid and the enthalpy of mixing of the intermediate phases. For the Ni-Ti-Hf ternary database, special attention was paid to the ternary intermediate H-phase. It was found in Ni-rich NiTiHf shape memory alloys and was considered as a line compound in our assessment. The calculation based on the database indicated H-phase doesn't occur at 900°C but can be thermodynamically stable below 600°C, which matched experimental measurements. Future database work will focus on developing Ni-Ti-Hf-Al quaternary system with efficient experimental validation.

6.2.3 Precipitation-Enhanced Thermoelectric Materials

CALPHAD Assessment of Chalcogenide Systems In further support of design of microstructured thermoelectric materials initiated under the DARPA SIMPLEX and MATRIX programs, thermodynamic and kinetic modeling of Pb based chalcogenides in the PbX (X=S,Se,Te) system have been performed **Voorhees** group. The thermodynamic and kinetic models are based on the CALPHAD method with input from first-principles calculations using density functional theory (DFT) as well as experimental data. In addition, thermodynamic assessments of the A-Q (A=Bi, Sb; Q=Te, Se) systems have been formed and a technique for modeling homologous compounds has been addressed.

Many thermodynamic assessments of the Pb-X systems use stoichiometric models to describe the Pb-X phase; however, the native defects of the Pb-X semiconductor create intrinsic carriers that play key roles in optimizing the carrier concentration of the thermoelectrics. First- principles calculations using density functional theory and the dilute

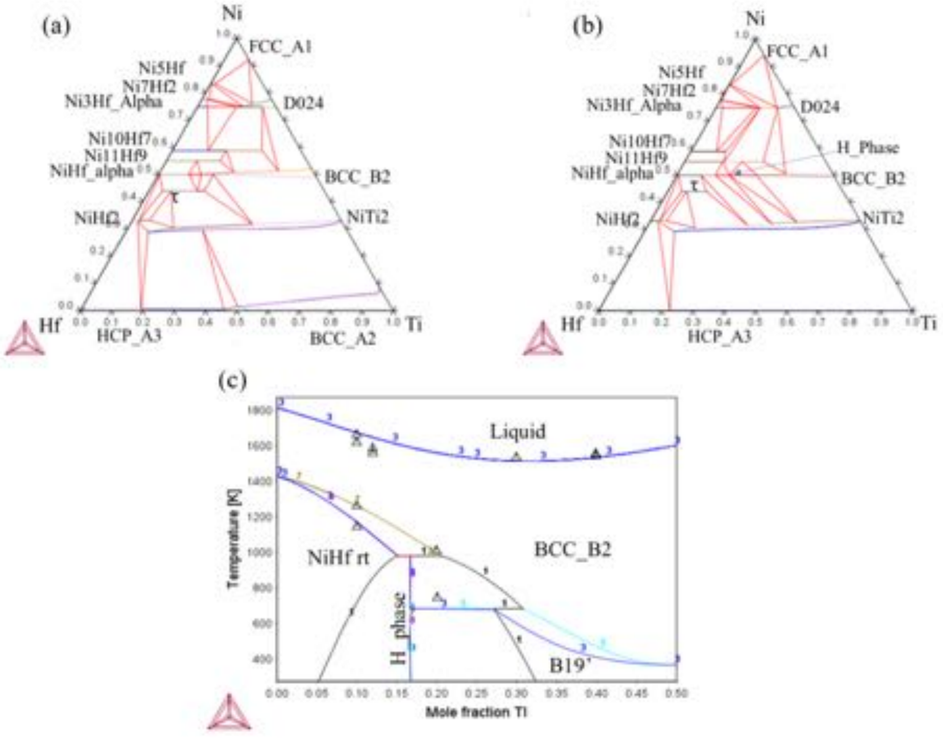


Figure 6.16: The isothermal sections of Ni-Ti-Hf ternary phase diagram at (a) 900°C and (b) 600°C; and (c) the vertical section at 50 at% Ni contents.

limit approximation have been performed to determine the dominant defects in the PbSe compound. The dominant defects were found to be Pb and Se vacancies for Se-rich and Pb-rich growth conditions, respectively. The formation energies of the defects were calculated along with the total number of defects and compared to experimental data in the literature, which agree in type, but differ in overall magnitude from the first-principles calculations.

To get a more accurate thermodynamic description of the intrinsic carriers, CALPHAD assessments have been performed using a five-sublattice (5SL) model and a simpler two-sublattice (2SL) model. Each model contains parameters used in the optimization that can be linked to physically measurable constants or values that can be derived from first-principles or experiments. The two models agree well with each other as well as the experimental and DFT data. The 2SL model was used to build a thermodynamic database for the pseudobinaries and pseudoternary Pb(S,Se,Te) system. In addition, Na doping in the PbX phase has been calculated via first- principles and incorporated into the CALPHAD models.

A kinetic assessment of the PbS-PbTe has been performed using experimental data derived from a diffusion couple. A forward simulation method was used to determine the interdiffusivities from the annealed composition profiles. The interdiffusivities were used to build a mobility database, which agrees reasonably well with the experimental data.

Lastly, an overview of how these models can be used to optimize thermoelectric materials has been given. The lattice thermal conductivity can be modeled using the Debye-Callaway model, which can be used to determine an optimal particle size for a second phase dispersion. A single parabolic band model is used to describe the transport properties dependence on the carrier concentration, which can be determined from the previously determined phase diagram. The thermodynamic and kinetic databases can then be used with PrecipiCalc to determine appropriate annealing times to achieve desired microstructure. Annealed microstructures of the Pb(Te,S) system indicate that this material does not go through homogenous nucleation but instead a discontinuous precipitation process that deserves further investigation.

Design of Multifunctional Intermetallic Thermoelectrics Further application of the tools and methods developed under the DARPA-SIMPLEX data analytics program has been conducted under the new DARPA-MATRIX initiative to design specific thermoelectric materials for new devices in collaboration of **Olson**, **Doak** and **Saal** with **Snyder**. Using pre-CALPHAD data from the DFT-generated OQMD database by Wolverton, initial design of a two-phase half-heusler intermetallic alloy demonstrated feasibility of a thermoelectric "superalloy" combining thermoelectric properties with oxidation resistance required by a specific device being developed by the United Technology Research Center (UTRC).

6.2.4 SRG Design Consortium Projects and Design Class Interactions

Northwestern's SRG materials design consortium joined the CHiMaD umbrella organization during Year One, offering the leveraging opportunity of the CALPHAD-based metals design projects with topics and sponsors listed in Figure 6.17.

2018 SRG Design Projects

- **ONR CyberAlloys** (Olson, Kontsevoi)
  - CMD of Fe & Ti alloys for blast and fragment protection
- **DOE/GM Lightweighting Initiative** (Olson, Wolverton, Voorhees)
  - CMD of cast aluminum for cylinder heads
- **ArcelorMittal AHSS** (Olson)
  - CMD of high-strength automotive Q&P TRIP steels

DMDII Rapid Qualification for Additive Manufacturing (Olson, Liu, Cao)
-ICME AIM modeling of PH48S stainless maraging steel
DARPA/Honeywell Open Manufacturing (QuesTek)
- ICME for SLM additive manufacturing of Ni 718+
DARPA-SIMPLEX: Data-Driven Discovery for Designed Thermoelectric Materials (QuesTek/CHiMaD; Olson, Voorhees, Snyder)
DARPA-MATRIX: Designed Thermoelectric Materials for Transduction Devices (QuesTek/CHiMaD; Olson, Snyder)

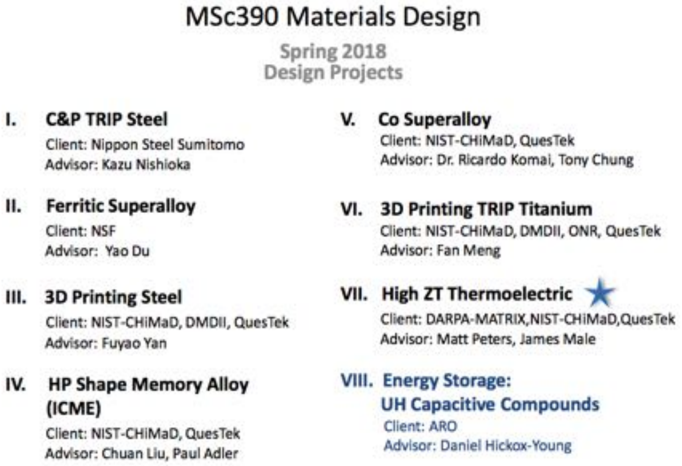
CHiMaD

Figure 6.17: 2018 SRG Design Projects

The ONR CyberAlloys project applies CALPHAD-based design to transformation toughened alloys for blast and fragment protection in both Ti-based and Fe-based systems integrating all-electron FLAPW DFT predictions of interfacial properties important to ductile fracture toughness. Efforts in 2018 have included adaptation of both alloy systems to 3D printing. In its final year, the collaboration with GM under the DOE Lightweighting Initiative has quantified the thermodynamics and kinetics of Q phase precipitation strengthening, summarized in Acta Mat publications, and applied it in design of cast Al alloys with higher temperature performance, now under further development at GM. Caterpillar collaboration under the same DOE initiative adapted multiphase precipitation concepts from our previous martensitic blast protection steel designs to demonstrate novel cast steel prototypes for lower cost crankshaft applications. Also completed in the past year, AHSS design research supported by ArcelorMittal has quantified the nonequilibrium thermodynamics of carbon partitioning in Quench & Partition martensite-austenite TRIP steels for high formability automotive sheet applications. In support of the new technology of additive manufacturing, the NIST-MSAM project completed in 2016 quantified recrystallization behavior of Ti and steel alloys, enabling a new project under the Chicago-based DMDII Manufacturing Institute which has applied the results in rapid qualification strategies demonstrated with QuesTek's Ferrium PH48S stainless maraging steel. QuesTek's collaboration with Honeywell under the DARPA Open Manufacturing initiative completed a 6-year 3-phase effort demonstrating AIM qualification of printed Ni superalloy components for aerospace applications, using data from 3 lots of alloy powder to efficiently forecast minimum properties.

Building on our CHiMaD Year One demonstration of a hybrid approach to integrating CALPHAD with datamining techniques, QuesTek secured a major DARPA-SIMPLEX consortium project with CHiMaD to apply this approach to integrated compound discovery and predictive microstructural design in thermoelectric materials. This has been followed in 2018 by application of the methods in specific designs for device applications under the DARPA MATRIX program. These projects help demonstrate the generality of the methods, tools, and database infrastructure of CHiMaD.

The SRG Annual Meeting held each March at Northwestern continues to serve as a review of CHiMaD metals activities offering interaction with a broader community. In support of NIST collaboration, NIST speakers at the 2018 SRG meeting included **Campbell** on database development and **Gilman** on Carbon Fiber Reinforced Polymer (CFRP) design. Building on the NIST-sponsored US/Japan Materials Genome Workshop held in Tsukuba in June 2015, the 2018 SRG meeting at Northwestern was followed by the 6th NU-NIMS Materials Genome Workshop. Inspired by the scope of the CHiMaD activities, the workshop emphasized new initiatives in materials informatics. Plans for enhanced collaboration between NU and NIMS continue, with the Materials Genome Workshop now an annual event.



MSc390 Materials Design

Spring 2018
Design Projects

I. **C&P TRIP Steel**
Client: Nippon Steel Sumitomo
Advisor: Kazu Nishioka

II. **Ferritic Superalloy**
Client: NSF
Advisor: Yao Du

III. **3D Printing Steel**
Client: NIST-CHiMaD, DMDII, QuesTek
Advisor: Fuyao Yan

IV. **HP Shape Memory Alloy (ICME)**
Client: NIST-CHiMaD, QuesTek
Advisor: Chuan Liu, Paul Adler

V. **Co Superalloy**
Client: NIST-CHiMaD, QuesTek
Advisor: Dr. Ricardo Komai, Tony Chung

VI. **3D Printing TRIP Titanium**
Client: NIST-CHiMaD, DMDII, ONR, QuesTek
Advisor: Fan Meng

VII. **High ZT Thermoelectric** ★
Client: DARPA-MATRIX,NIST-CHiMaD,QuesTek
Advisor: Matt Peters, James Male

VIII. **Energy Storage: UH Capacitive Compounds**
Client: ARO
Advisor: Daniel Hickox-Young

Figure 6.18: 2018 Materials Design Class Projects

The SRG and CHiMaD research projects provide a hierarchical project coaching system where graduate students serve as technical advisors to the class project teams summarized in Figure 6.18. Projects supported by CHiMaD research include the Co superalloy and shape memory alloy, as well as steel and Titanium alloy designs constrained for 3D printing. Building on our move into electronic materials represented by the High ZT Thermoelectric materials project and the broader array of design methodologies under CHiMaD development, highlighted in blue is a new project in collaboration with James Rondinelli (NU), applying novel datamining strategies to directed compound discovery for Ultrahigh-Capacitance materials.

Highlighted by the star in Figure 19, graduate student Matt Peters coached a team of undergraduate students applying his databases in the integrated CALPHAD-based design of a chalcogenide thermoelectric with optimized microstructure based on the PbTe-SrTe-PbTe system. The project placed 3rd in the 2018 ASM Undergraduate Design competition.

6.2.5 First Principles Study of Al- and Mg- Alloys

First-principles study of crystal structure and stability of T1 precipitates in Al-Li-Cu alloys Aluminum-lithium-copper alloys have a low density, high elastic modulus and high specific strength. Due to this combination of properties, alloys strengthened with the ternary (Al-Li-Cu) T1 phase have attracted a great deal of interest especially in aerospace applications. Determining the atomic structural information of the precipitate is a fundamental step in developing a basis for advanced alloy design; however, even though many experimental studies have addressed the T1 crystal structure, it remains the subject of some controversy. Here, we use density functional theory (DFT) calculations to investigate the structure and composition of the T1 phase by comparing the energetic stability of five previously-proposed models of the crystal structure of T1. The DFT formation energy of these proposed T1 crystal structures was calculated using a special quasi-random structure (SQS) approach to describe a disordered Al-Cu sub-lattice. In conflict with the experimental phase diagram, DFT calculations of all five proposed models result in an energetically unstable T1 phase. We search for a new, lower-energy structure of T1 using a cluster expansion approach, and find a new structural model with DFT energy that is stable (at T=0K), i.e., on the calculated convex hull of the Al-Li-Cu ternary system. However, this new predicted phase does not have a tie-line with Al, but the formation energy of the phase is very close to the energy required to make a tie-line with Al (E =0.013eV/atom), which could be affected by finite temperature entropic effects (i.e., vibrational entropic stabilization), [27.48].

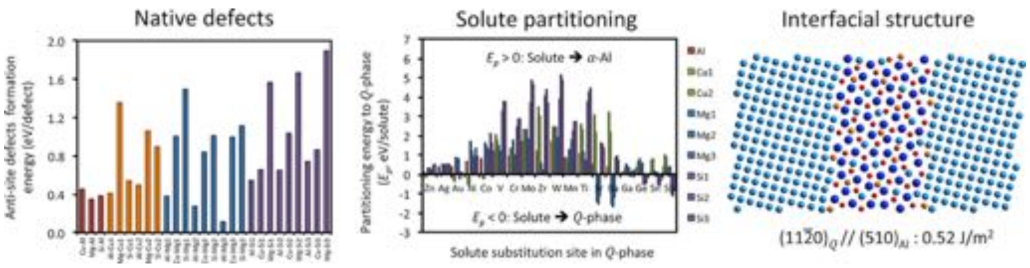


Figure 6.19: Energetics of native defects, solute partitioning, and interfacial energy of Q precipitate in Al-Cu-Mg-Si alloys

Energetics of native defects, solute partitioning, and interfacial energy of Q precipitate in Al-Cu-Mg-Si alloys The compound $Al_3Cu_2Mg_9Si_7$, is known as the Q-phase and forms as a thermodynamically stable precipitate during aging in the quaternary Al-Cu-Mg-Si system. We perform atomic-scale density functional theory (DFT) calculations of defect properties, solute partitioning, and interfacial stability of the $Al_3Cu_2Mg_9Si_7$(Q) precipitate. We find [27.44]: (i) simple native point defect (i.e., vacancies and anti-sites) thermodynamics can partially explain the experimentally observed off-stoichiometry, such as the observed variation of compositions, $Al_{3+\delta}Cu_2Mg_{9-\delta}Si_7$ (Mg-deficient and Al-rich) in experiment. (ii) Calculated solute-partitioning energies of common solutes allow us to define general rules for site-preference in the Q-phase in terms of electronic structure and atomic radius. To validate our DFT predictions, we perform atom-probe tomography (APT) experiments for six-different elements (Zn, Ni, Mn, Ti, V, and Zr). The results show that the partitioning behavior of solutes Ni, Zn, and Mn are consistent with DFT predictions, but the transition elements (Ti, V, and Zr), which are anomalously slow diffusers in Al, partition to the Q-phase in constrast to DFT partitioning energies. (iii) For the low energy interface $(11\bar{2}0)_Q//(510)_{Al}$ observed in needleshaped Q-precipitate, we survey various terminations and orientations and derive a low-energy interfacial structure. We find this low-energy interfacial model has Cu atoms nearest to the interface, which is in agreement with previous literature on Cu interfacial segregation at the Q'//α-Al interface. The computed interfacial energy (0.52 J/m^2) and the corresponding structure will be useful input to future multi-scale modeling of microstructural evolution.

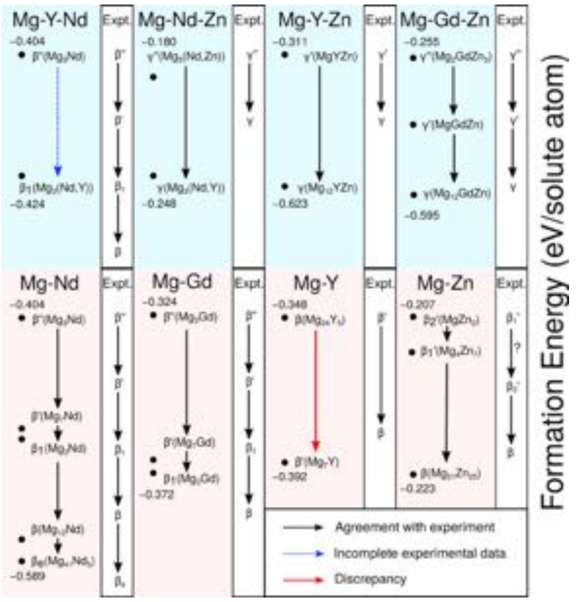


Figure 6.20: A comparison of the calculated and experimentally reported precipitation sequences. The calculated sequences in Mg-Nd, Mg-Gd, Mg-Nd-Zn, Mg-Y-Zn, Mg-Gd-Zn, Mg-Y-Nd, Mg-Zn systems fully or partially match those reported in experiments [27.42]

Crystal structure, energetics, and phase stability of strengthening precipitates in Mg alloys: A first-principles study Magnesium alloys have attracted increasing interest due to their potential use as light-weight structural materials but their application is limited by their low strength compared to conventional alloys. Age hardening is commonly employed to form strengthening precipitates in such alloys, which impedes the motion of dislocations, and leads to improved strength. However, the exact composition, crystal structure, and energetics of many of these strengthening precipitates are either unknown or not clearly resolved, making the precise engineering and design of such alloys difficult. Toward this end, we use first-principles density functional theory calculations to elucidate the crystal structures and energetics of a very large set of precipitates in magnesium alloys. For cases where the precipitate crystal structure is not known, we comprehensively search over decorations of many prototype structures, including hcp superstructures, and in addition, perform global structural optimization using the Minima Hopping Method to predict suitable crystal structures. For all the strengthening precipitates, we calculate the formation energies, construct the respective zero temperature convex hulls, and analyze their stabilities. We show that the bulk formation energies per solute atom (essentially, the solute chemical potentials) decrease along the observed sequences of precipitation, validating our calculations in Mg-Nd, Gd, Y, Y-Nd, Nd-Zn, Gd-Zn, Y-Zn, Al, Zn, Sn, Al-Ca, Ca-Zn alloy systems. In addition, we construct a monolayer model for the Guinier-Preston zones (GP zones) observed in the Mg-Nd-Zn system during early stages of age hardening, and thereby explain the formation of the γ'' ($Mg_5(Nd,Zn)$) phase from the GP zones, as observed in experiments.

6.3 QuesTek Activities

As the principal commercialization partner of CHiMaD, QuesTek Innovations is actively involved in the application of CHiMaD methods, tools and databases in a wide range of computational materials design and qualification projects for commercial and federal agency clients. In addition, QuesTek staff actively participate in CHiMaD education outreach programs, and special projects demonstrating generalization of design methods first demonstrated in metal alloys.

6.3.1 Exploration of Co-Ni-Al-X (X=Cr and Ta) System

Following the work from the 2018 MSc390 Materials Design team's determination that the description in the NIST Co TDB could be improved further, **Komai** (QT) designed model alloys that explored the Co-Ni-Al-X (X=Cr and Ta) space where the phase region is predicted differently between the QT-Co, TCNI8, and NIST-Co thermodynamic databases. Four compositions were designed and are being evaluated for solvus, phase composition, and phase fraction after 1000°C aging to supplement the NIST-Co TDB. This data will be shared amongst the cobalt Use-Case Group and future data efforts will be directed towards the improvement of the NIST-Co TDB.

A meeting at QuesTek with **Stan** explored strategies for uncertainty quantification in CALPHAD data systems in preparation for the new CHiMaD Tool Group on Materials Genome Uncertainty Quantification (UQ), and discussed strategies to expand the scope to UQ in AIM qualification estimates of minimum properties from manufacturing variation.

6.3.2 Materials Data Software

In summer of 2018, QuesTek worked to adopt the materials data software being used and developed by NIST and CHiMaD: the Materials Data Curation System (MDCS) and the Materials Data Facility (MDF). QuesTek intern Deepali Patil (Cal Poly Pomona) worked with **Komai**, **Doak**, and **Campbell**, to implement the MDCS system in-house and explore future use with datasets relevant to existing Ni-based superalloy strength modeling efforts (exploring solid solution strength data from experimental work). Furthermore, Ward (U Chicago) worked with QuesTek to perform a training in the MDF data sharing capabilities so that QuesTek's data produced under CHiMaD can be shared between collaborators such as the Cobalt thermodynamic data.

6.3.3 Additive Manufacturing

QuesTek has produced a large amount of additive manufacturing data related to cracking and printability across multiple programs, multiple AM methods, and multiple material systems, including newly designed alloys. QuesTek also has developed a method of predicting printability from a CALPHAD approach. The results across various programs (including power, speed, powder size, composition, and others) were inspected along with a failure analysis approach to examining cracked specimens. This effort was led by two QuesTek interns Jacqueline Hardin (University of Illinois at Urbana-Champaign) and Nicholas Ury (Cal Poly Pomona and a former CHiMaD REU student) under the guidance of **Komai**. The culmination of this work led to the development of a "Crack Atlas of Additive Manufacturing" guide to examining different cracking behaviors observed in AM builds across multiple alloy systems. Hardin and Ury were able to identify compositional and thermodynamic indicators of solidification and liquation cracking behavior. Furthermore, they proposed a systematic framework for a more robust printability predictor; for example, the need for a method of predicting ductility dip cracking or other solid state cracking mechanisms that are not related to liquid feeding and dendrite formation.

6.3.4 Thermoelectrics

QuesTek continued work on the microstructural design of thermoelectric compounds through both the DARPA funded MATRIX program and internal resources. Led by **Doak** in collaboration with **Olson** and Jeffrey Snyder (NU), the design of a high temperature thermoelectric compound was performed to produce a compound that was also oxidation resistant in order to perform at higher temperatures. Initial concepts were prototyped and heat treated to explore the oxidation resistance of the system. This system is a (Ti,Zr,Hf)Co(Sb,Sn) half-Heusler thermoelectric and demonstrated the ability to form a Cr-oxide after being treated at 700°C for 1 hour.

6.3.5 Polymer Martensites

In support of the new Use Case effort on CFRP composites for cryogenic applications motivated by space vehicles, led by **Shull**, QuesTek has begun exploring the concept of transformation toughened polymers. The motivation is the well-established use of dispersed metastable austenite in cryogenic steels developed in the 1940s to exploit athermal transformation yielding at cryogenic temperatures to offset the high temperature dependence of thermal strengthening in BCC steels. Building on his doctoral research experience in biological martensitic transformations, **Komai**, working with **Olson**, has assembled archival literature establishing engineering polymer systems exhibiting high-strain martensitic transformations. This includes recent work demonstrating room-temperature transformation ductilization of epoxy-based 2-phase copolymers relevant to a CFRP matrix. Research will next explore potential systems meeting the transformation entropy requirements to emulate the cryogenic steels systems.

Nealey group at UC has been performing analysis of polymer martensitic transformations, specifically exploring the liquid crystal "blue phase." A joint meeting with **Komai** and **Olson** at the CHiMaD Center initiated discussion of a more rigorous application of the crystal kinematic theory of martensitic transformation.

ASM Materials Genome Camp Komai once again served as an organizer and lead instructor at the ASM Materials Genome Camp, this year a five-day camp in comparison to last year's pilot three-day camp. Working with **Gulsoy**, **Olson**, Clay Houser (NU), and Jonathan Emery (NU), an extended course developed for teachers using the design of a self-healing metal matrix "Frankensteel" composite.

Materials Design Workshop at KTH Royal Institute of Technology, Stockholm, Sweden In collaboration with CHiMaD and QuesTek Europe AB, QuesTek Innovations continued to develop the Materials Design Workshop series for a special workshop at KTH in Stockholm, Sweden at the Hero-m center for design. **Komai**, **Gulsoy**, and graduate student Clay Houser developed and delivered a special version of the materials design workshop to enhance and encourage materials design efforts internationally.

6.4 Collaborations

6.4.1 CHiMaD Collaborations

Integrating CALPHAD and data mining for advanced steel design
*Ankit **Agrawal** (NU), Alok **Choudhary** (NU), Gregory **Olson** (NU)*
This collaboration aims to use CALPHAD to enrich steel composition and processing data with structure information to derive more reliable data-driven structure-property linkages.

Thermoelectrics data mining
*Ankit **Agrawal** (NU), Alok **Choudhary** (NU), Gregory **Olson** (NU)*
This project aims at developing predictive models for thermoelectric properties of compounds and materials. It had started with primary support from the DARPA SIMPLEX project (managed by NAISE/CHiMaD).

Phase-field rafting model in Co-based superalloys
*Olle **Heinonen** (ANL), Peter **Voorhees** (NU), David **Seidman** (NU), David **Dunand** (NU)*
This collaboration aims to develop a phase-field model to predict rafting behavior in Co-based superalloys.

Development of atom-probe data storage and publishing server
*Ian **Foster** (UC), Ben **Blaiszik** (UC), David **Seidman** (NU), David **Dunand** (NU)*
Building on the newly built data storage server and data publishing interface, the data security and backup option are currently being expanded for data produced by NUCAPT.

Additive Manufacturing Modeling
*Jian **Cao** (NU), Wing-Kam **Liu** (NU)*
This collaboration aims to generate experimental results used for validation of and input for microscale, macroscale and reduced order models for processing and mechanical properties of AM materials. This includes in-situ mechanical testing at Argonne National Laboratory's APS using Beamline 2BM to produce 3D images of the void morphology during extensile deformation - a capability available at only a few facilities worldwide. We have also conducted micro-Laue diffraction at beamline 34IDE to understand residual stress and grain orientation in 3D. Other collaboration includes using techniques such as IR imaging and SEM imaging to determine solidification/cooling rates for comparison to thermal simulations.

Predictive design of NiTi shape memory alloys
*Gregory **Olson** (NU), Paul **Adler** (NU), Wing-Kam **Liu** (NU)*
This collaboration simulated the extrusion process of a NiTi SMA wire with a mesh adaption FE code, and a finite deformation self-consistent clustering analysis code. With

these methods, wire drawing was modeled. The resulting microstructures were input to a crack nucleation life prediction model that uses the newly developed crystal plasticity self-consistent clustering analysis. This collaboration was facilitated by the interdisciplinary doctoral cluster Predictive Science and Engineering Design at Northwestern.

Phase Clustering
*Michael **Bedzyk** (NU), Yip-Wah **Chung** (NU), Ankit **Agrawal** (NU), Alok **Choudhary** (NU)*
This collaboration aims to improve the phase clustering implementation of the Machine learning technique, as discussed above.

6.4.2 NIST Collaborations

NIST CALPHAD database assessment
*Gregory **Olson** (NU), Ursula **Kattner** (NIST), Carelyn **Campbell** (NIST)*
Peisheng Wang (NIST-CHiMaD Postdoctoral Reserach Fellow), published CALPHAD database assessments supporting design of Co-based superalloys.

Thermodynamic based Multinary Co-Based Superalloys Design
*Carelyn **Campbell** (NIST), Eric **Lass** (NIST), David **Seidman** (NU), David **Dunand** (NU)*
This collaboration aims to improve alloy design by providing iterative feedback on the results of the mechanical testing and microstructural characterization, designed by **Lass**, of multinary Co based superalloys.

High throughput combinatorial synthesis and analysis of Co-based ternary alloys
*Ursula **Kattner** (NIST), Carelyn **Campbell** (NIST), Michael **Bedzyk** (NU), Yip-Wah **Chung** (NU)*
This collaboration is based on NIST PIs providing theoretical and computational support for the use-case research.

6.4.3 External Collaborations

Digital Manufacturing and Design Innovation Institute (DMDII)
*Kornel F. Ehmann (NU), Greg J. Wagner (NU), Lucia Mirabella (Siemens), Suraj R. Musuvathy (Siemens), Edward R. Slavin III (Siemens), Sanjeev K. Srivastava (Siemens), Federico M. Sciammarella (NIU), Joseph S. Santner (NIU), Jason **Sebastian** (QuesTek), David **Snyder** (QuesTek), Jiten Shah (Product Development & Analysis), Jian **Cao** (NU), Wei **Chen** (NU), Gregory **Olson** (NU), Wing-Kam **Liu** (NU)*
The external collaborators are working with a DMDII project working on AM process modeling, calibration, assessment and prediction. This work includes calibration experiments, macro-scale and micro-scale simulations for mechanical properties and processing, and characterization experiments.

Cyber-Physical Systems (CPS)
*Kornel F. Ehmann (NU), Greg J. Wagner (NU), Jian **Cao** (NU), Wing-Kam **Liu** (NU)*
This collaboration focuses on integrated systems for advanced manufacturing, connecting experiments and simulation through a multi-loop control scheme with novel data-handling framework. This connects to the Additive Manufacturing efforts.

Novel Additive Manufacturing Combining Extrusion 3D-Printing and CVD for Co-based Superalloy Micro-lattices
*Heeman Choe (Kookmin University, South Korea), David **Seidman** (NU), David **Dunand** (NU)*

This collaboration focuses on additive manufacturing of Co-based superalloy micro-lattices using a combination of extrusion 3D-printing and pack-cementation processes.

Modelling Of The Incremental Bulk Metal Forming Processes

*Alexander **Umantsev** (FSU), O. I. Bylya and Mr. B. Krishnamurthy (University of Strathclyde, UK)*

In collaboration with the Advanced Forming Research Centre at the University of Strathclyde, Glasgow, UK the Bifurcation theory was applied to the analysis of the experiments on the rotary bulk-forming processing of high-strength steel pipes. First, theoretical model for the elastic and plastic properties was fully calibrated. Then, the model was used to reproduce the experimental results. The UK group is planning to use the simulation results for the design of the new processing regimes.

Cobalt-base Superalloys from Powder Metallurgy

*Monica Campos Gomez (Universidad Carlos III de Madrid, Spain), David **Seidman** (NU), David **Dunand** (NU)*

This collaboration is developing γ'-strengthened Co-based superalloys using powder metallurgy.

Synchrotron X-ray imaging

*Tao Sun (ANL), Olle **Heinonen** (ANL), Wing-Kam **Liu** (NU)*

This collaboration focuses on quantitative analysis in additive manufacturing and intense laser melting, connecting real-time synchrotron X-ray experiments and multiphysics thermal-fluid-vaporization models.

6.5 Technology Transfer

Data

Datasets Uploaded to Materials Data Facility in 2018

1. Atom Probe Tomography Reconstruction and Analysis for the Temporal Evolution of Co-Al-W Superalloys at 650°C
   *P. Bocchini, D.-W. Chung, D. **Dunand**, D. **Seidman***
   DOI: 10.18126/M2S34X
2. Atom Probe Tomography Reconstruction and Analysis for the Temporal Evolution of Co-Al-W Superalloys at 750°C
   *P. Bocchini, D.-W. Chung, D. **Dunand**, D. **Seidman***
   DOI: 10.18126/M2WS7W
3. $\gamma+\gamma'$ Microstructures in the Co-Nb-V Ternary System
   *F. Reyes Tirado, J. Perrin Toinin, D. **Dunand***
   DOI: 10.18126/M21M0R
4. $\gamma+\gamma'$ Microstructures in the Co-Ta-V Ternary System
   *F. Reyes Tirado, J. Perrin Toinin, D. **Dunand***
   DOI: 10.18126/M2N920
5. Influence of ruthenium in a model Co-Al-W superalloy
   *S. Sauza, P. Bocchini, D.-W. Chung, D. **Dunand**, D. **Seidman***
   DOI: 10.18126/M2CS77

6.6 Publications and Presentations

13 Number of CHiMaD-supported publications in 2018 by Precipitation-strengthened Alloys use-case group. Please see chapter 27 for details. [27.44, 27.48, 27.35, 27.89, 27.90, 27.91, 27.92, 27.93, 27.95, 27.98, 27.100, 27.56, 27.42]

33 Number of presentations on CHiMaD supported research in 2018, please see section 26.5 for the complete list.

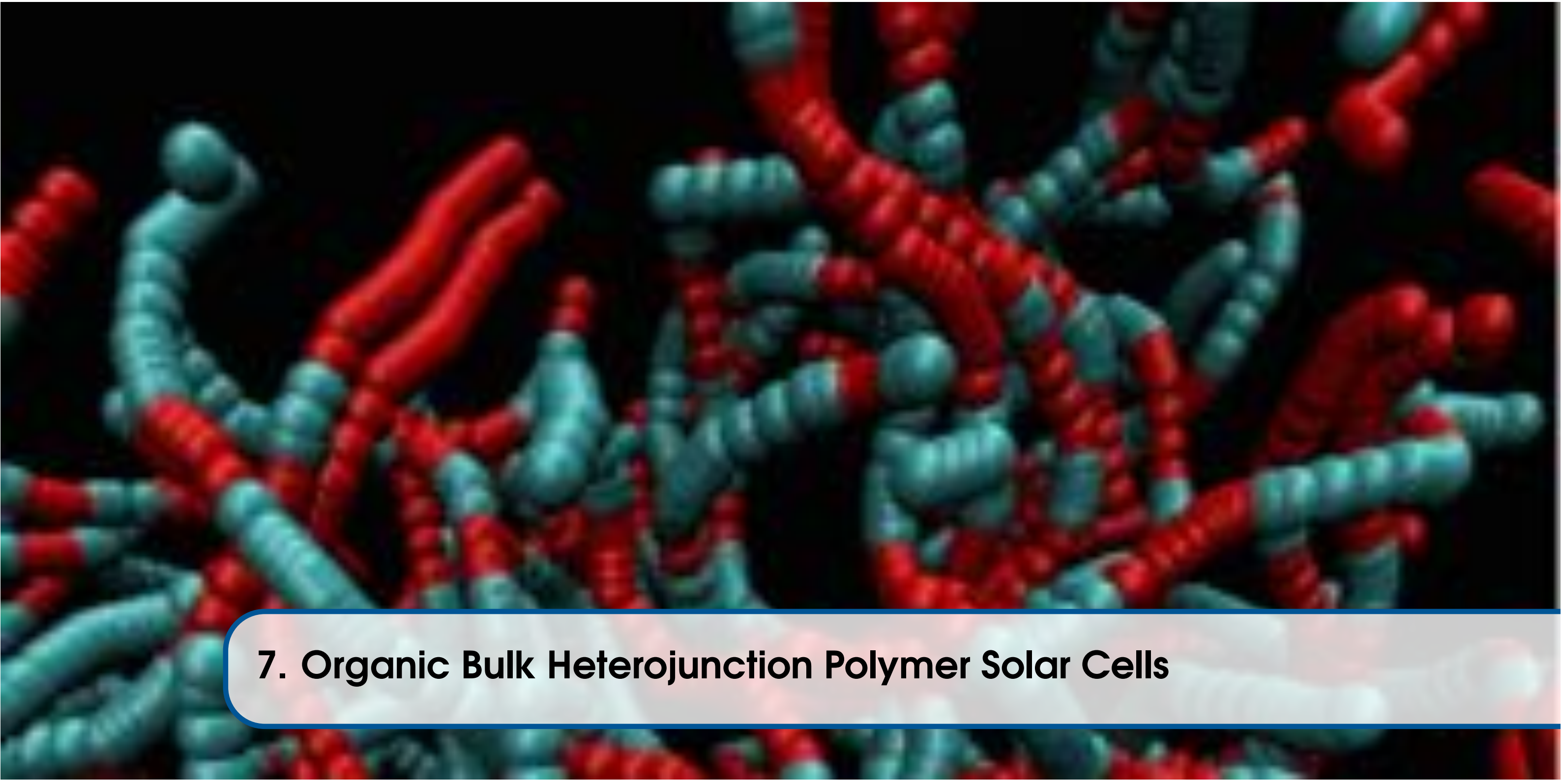
6.7 CHiMaD Team

Precipitation-Strengthened Alloys Use-Case Group Researchers			
Name	**Position**	**Affiliation**	**Project (PI)**
Gregory Olson	Principal Investigator	NU/MSE	
David Dunand	Principal Investigator	NU/MSE	
David Seidman	Principal Investigator	NU/MSE	
Christopher Wolverton	Principal Investigator	NU/MSE	
Ankit Agrawal	Principal Investigator	NU/EECS	
Alok Choudhary	Principal Investigator	NU/EECS	
Yip-Wah Chung	Principal Investigator	NU/MSE	
Michael Bedzyk	Principal Investigator	NU/MSE	
Wing Kam Liu	Principal Investigator	NU/ME	
Alexander Umantsev	Principal Investigator	FSU/PHY	
Jason Sebastian	Principal Investigator	QT	
Ricardo Komai	Principal Investigator	QT	
Jeff Doak	Principal Investigator	QT	
James Saal	Principal Investigator	QT	
Ida Berglund	Principal Investigator	QT	
Dana Frankel	Principal Investigator	QT	

Precipitation-Strengthened Alloys
Use-Case Group Researchers

Name	**Position**	**Affiliation**	**Project (PI)**
Paul Adler	S. Research Associate	NU/MSE	SMA for biomedical applications *(Olson)*
Sung-II Baik	NUCAPT Manager	NU/MSE	TEM study of dislocation dynamics in Co-based superalloys. *(Seidman/Dunand)*
Peisheng Wang	Postdoctoral Researcher	NU/NIST	Co-Alloy database development *(Olson/Kattner/ Campbell/Lass)*
K.V.L.V. Narayanachari	Postdoctoral Researcher	NU/MSE	High Throughput Combinatorial Synthesis and Multimodal Analysis of Ternary Alloys Precipitation-Strengthened Cobalt Alloys *(Chung/Bedzyk)*
Modesar Shakoor	Postdoctoral Researcher	NU/ME	NiTi wire extrusion process modeling *(Liu)*
Yan Ping Lian	Postdoctoral Researcher	NU/ME	CAFE modeling of grain growth *(Liu)*
Jacques Perrin Toinin	Postdoctoral Researcher	NU/MSE	Effect of Cr in microstructure and mechanical properties in Co-based Superalloys *(Seidman/Dunand)*
Mykola Lavrskyi	Postdoctoral Researcher	FSU/Phy	Phenomenological Model of Ductile Fracturing *(Umantsev)*
Wenkun Wu	Postdoctoral Researcher	ANL	Phase Field Modeling of Co-alloys *(Heinonen)*

Precipitation-Strengthened Alloys Use-Case Group Students			
Name	**Position**	**Affiliation**	**Project (PI)**
Fernando L. Reyes Tirado	Graduate Student	NU/MSE	Co-Alloys *(Dunand/Seidman- NSF Fellow)*
Matthew Peters	Graduate Student	NU/MSE	CALPHAD Methods for Thermoelectrics *(Voorhees - NDSEG Fellowship)*
Chuan Liu	Graduate Student	NU/MSE	SMA for biomedical applications *(Olson)*
Cheng Yu	Graduate Student	NU/ME	Fatigue life prediction for advanced materials *(Liu)*
Orion Kafka	Graduate Student	NU/ME	Fatigue life prediction for advanced materials *(Liu-NSF Fellow)*
Zheng Jia	Graduate Student	NU/ME	Mechanics of energetic materials *(Liu)*
Ding-Wen (Tony) Chung	Graduate Student	NU/MSE	Composition Design for Oxidation Resistance in Co-based Superalloys *(Dunand/Seidman)*
Francesca Long	Graduate Student	NU/MSE	Co-based Superalloys *(Dunand/Seidman)*
Daniel Ng	Undergraduate Student	NU/MSE	Effect of Cr on microstructure and mechanical properties in Co-based Superalloys *(Dunand/Seidman)*
Rachel Orenstein	Undergraduate Student	NU/MSE	Combinatorial XRD+XRF setup *(Bedzyk/Chung - Unfunded)*
Spencer Taylor	Undergraduate Student	UNT	Powder metallurgy processing of Co-based Superalloys *(Dunand/Seidman - visiting)*



7. Organic Bulk Heterojunction Polymer Solar Cells

Luping Yu (UC), Juan de Pablo (UC), Guilia Galli (UC), Tobin Marks (NU)

Lee Richter Jr (NIST), Dean Delongchamp (NIST)

Significance

Bulk heterojunction organic solar cells (OSCs) represent an alternative solar energy harvesting system, which is currently being pursued by numerous research groups world-wide. Commercial products are starting to appear in the market from several companies. This use-case group is addressing critical issues that affect the performance of OSC devices through materials design and synthesis to test new structure/function ideas, exploring new means of morphological control and its influence on OSC performance, new approaches to device optimization, data collection/analysis on new formulations and device characteristics, and on structure/property relationships. New electron donors or acceptors exhibiting superior photovoltaic properties are being developed, and their synergistic interactions are being studied. General rules for designing new polymers for use as charge relays or energy harvesting antennae in ternary OSCs are also being investigated. In close coupling to these experimental efforts, new theoretical methods are being developed to elucidate key electronic processes (electronic excitation, charge separation and transport) in our champion polymer systems and ternary polymer solar cells. To this end, large-scale modeling approaches are also being developed to simulate the morphology formation, polymer chain aggregation, and ways to control them. These studies are also closely coupled with efforts by NIST scientists to fully utilize their expertise in structural and morphological characterization of CHiMAD materials. These new materials and the basic knowledge gained will deepen our understanding of structure/property relationships and accelerate the development of OSCs towards more efficient and cost-effective commercial products.

7.1 Design Goals

The main design goal of the Organic Bulk Heterojunction Polymer Solar Cells use case group is to arrive at all-organic solar cells with reproducible efficiencies that are well above those of currently known for organic thiophene-based materials (ca. 8%). In the long term, a key aim of this group is to arrive at all-polymer organic photovoltaic materials that will obviate the need for fullerenes. The development of OPV materials to date has largely followed an Edisonian approach. This group seeks to identify key molecular and structural descriptors of performance which may be used in the future to rationally design improved materials for next generation OPV cells. To that end, the group is targeting the following activities:

- Develop novel ternary systems that can significantly enhance the power conversion efficiency (PCE) values of devices. Systematically survey the structures of third component polymers to establish structure/property relationship on energy level matching, template effects for self-assembly, and the relationship between composition and Voc values.
- Develop new types of polymers and small molecules with varied band gaps, energy levels, and optimized charge transport behaviors for use either as electron donors or acceptors in OSCs. Carry out detailed studies of structure property relationships.
- Develop efficient all-organic or all-inorganic, non-fullerene electron acceptors to replace fullerenes in OSCs.
- Develop new methods and force fields capable of fully describing aromatic semiconducting polymers.
- Model the electronic properties of these polymers based on sophisticated quantum mechanical calculations.
- Leverage NIST expertise by collaborating on several critical issues, including accurate measurement of glass transition temperatures, fullerene-polymer interaction, effect of new non-fullerene acceptors, and measurements of structure coupled to atomistic predictive simulations.

7.2 Significant Accomplishments in 2018

Relationships between single crystal structure of indacenidithienothiophene based OPV acceptor materials and their thin-film crystalline phase orientation in BHJ blends with donor polymers have been studied in detail by **Marks** group and key relationships were elucidated, significant theoretical descriptors for the blending behavior were identified and used for next-generation materials synthesis. These studies also led to unprecedented highly ordered OPV blends based on fluorinated indacenothien-othiophene acceptors with power conversion efficiencies (PCEs) in binary blends surpassing the 12% threshold and with good environmental, thermal, and microstructural stability. Accomplishment details for the various sub-projects which were enabled by CHiMaD support are outlined below.

7.3 Research Accomplishments

All-Polymer Solar Cells: Recent Progress, Challenges, and Prospects For over two decades bulk-heterojunction polymer solar cell (BHJ-PSC) research was dominated by donor:acceptor BHJ blends based on polymer donors and fullerene molecular acceptors. This situation has changed recently, with non-fullerene PSCs developing very rapidly. The power conversion efficiencies of non-fullerene PSCs have now reached $\geq 15\%$, which is far above the most efficient fullerene-based PSCs. Among the various non-fullerene PSCs, all-polymer solar cells (APSCs) based on polymer donor-polymer acceptor BHJs have attracted growing attention, due to the following attractions: (1) large and tunable light absorption of the polymer donor/polymer acceptor pair; (2) robustness of the BHJ film morphology; (3) compatibility with large scale/large area manufacturing; (4) long-term stability of the cell to external environmental and mechanical stresses. This minireview highlights the opportunities offered by APSCs, selected polymer families suitable for these devices with optimization to enhance the performance further, and discusses the challenges facing APSC development for commercial applications.

Closely packed, low reorganization energy pi-extended postfullerene acceptors for efficient polymer solar cells PNAS New organic semiconductors are essential for developing inexpensive, high-efficiency, solution-processable polymer solar cells (PSCs). PSC photoactive layers are typically fabricated by film-casting a donor polymer and a fullerene acceptor blend, with ensuing solvent evaporation and phase separation creating discrete conduits for photogenerated holes and electrons. Until recently, n-type fullerene acceptors dominated the PSC literature; however, indaceno-dithienothiophene (IDTT)-based acceptors have recently enabled remarkable PSC performance metrics, for reasons that are not entirely obvious. We report two isomeric IDTT-based acceptors 3,9-bis(2-methylene-(3-(1,1-dicyanomethylene)-benz-(5,6)indanone))-5,5,11,11-tetrakis(4-nonylphenyl)- dithieno-[2,3-d:2',3'-d']-s-indaceno[1,2-b:5,6-b']di-thiophene (ITN-C9) and 3,9-bis(2-methylene-(3-(1,1-dicyanomethylene)-benz(6,7)indanone))-5,5,11,11-tetrakis(4-nonylphenyl)- dithieno [2,3-d:2',3'-d'] -s-indaceno[1,2-b:5,6-b']dithiophene (ITzN-C9) that shed light on the exceptional IDTT properties vis-a-vis fullerenes. The neat acceptors and blends with fluoropolymer donor poly{[4,8-bis[5-(2- ethylhexyl)-4-fluoro-2-thienyl]benzo [1,2-b:4,5-b'] dithiophene2,6-diyl]-alt-[2,5-thio-phenediyl[5,7-bis(2-ethylhexyl)-4,8-dioxo4H,8H-benzo[1,2-c:4,5-c'] dithiophene-1,3-diyl]]} (PBDB-TF) are investigated by optical spectroscopy, cyclic voltammetry, thermogravimetric analysis, differential scanning calorimetry, single-crystal X-ray diffraction, photovoltaic response, space-charge-limited current transport, atomic force microscopy, grazing incidence wide-angle X-ray scattering, and density functional theory-level quantum chemical analysis. The data reveal that ITN-C9 and ITzN-C9 organize such that the lowest unoccupied molecular orbital-rich end groups have intermolecular $\pi - \pi$ distances as close as 3.31(Figure 7.1) angstrom, with electronic coupling integrals as large as 38 meV, and internal reorganization energies as small as 0.133 eV, comparable to or superior to those in fullerenes. ITN-C9 and ITzN-C9 have broad solar-relevant optical absorption, and, when blended with PBDB-TF, afford devices with power conversion efficiencies near 10%. Performance differences between ITN-C9 and ITzN-C9 are understandable in terms of molecular and electronic structure distinctions via the influences on molecular packing and orientation with respect to the electrode.

Photoactive Blend Morphology Engineering through Systematically Tuning Aggregation in All-Polymer Solar Cells Polymer aggregation plays a critical role in the miscibility of materials and the performance of all-polymer solar cells (APSCs). However, many aspects of how polymer texturing and aggregation affect photoactive blend film microstructure and photovoltaic performance are poorly understood. Here the effects of aggregation in donor C-H acceptor blends are studied, in which the number-average molecular weights (M_{ns}) of both an amorphous donor polymer, poly[4,8-bis(5-(2-ethylhexyl)thiophen -2-yl)benzo[1,2-b;4,5-b']dithiophene-2,6-diyl-alt-(4-(2-ethylhexyl)-3-fluorothieno[3,4-b]thiophene-)-2-carboxylate-2-6-diyl)] (PBDTT-FTTE) and a semicrystalline acceptor polymer, poly[N,N'-bis(2-octyldodecyl)naphthalene-1,4,5,8-bis (dicarboximide)-2,6-diyl]-alt-5,5'-(2,2'-bithiophene) (P(NDI2OD-T2)) are systematically varied. The photovoltaic performance is correlated with active layer microstructural and opto-

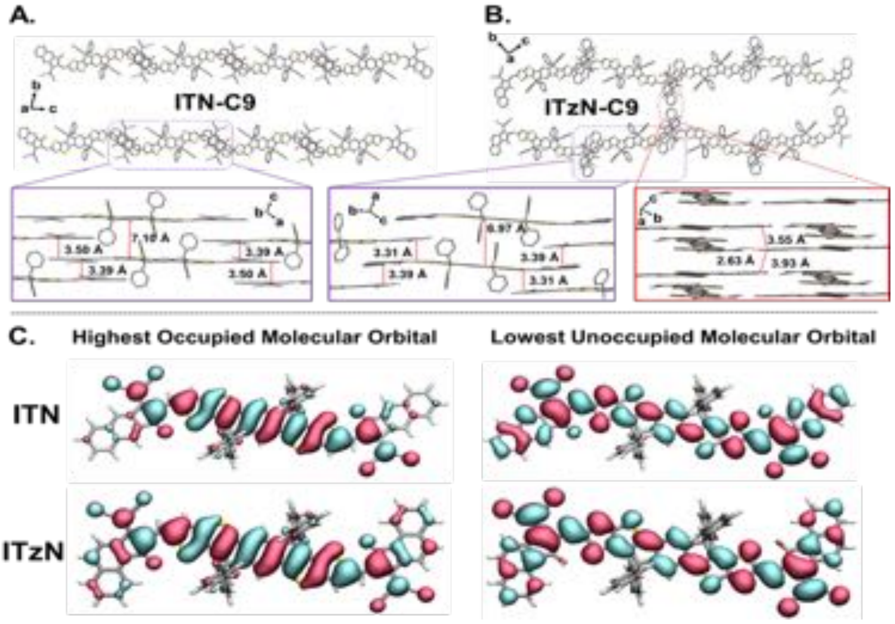


Figure 7.1: (A) Crystal packing of non-fullerene acceptors, (A) ITN-C9 and (B) ITzN-C9 with the closest $\pi - \pi$ contacts shown. (C) HOMO and LUMO contours of ITN and ITzN

electronic data acquired by in-depth transmission electron microscopy, grazing incidence wide-angle X-ray scattering, thermal analysis, and optical spectroscopic measurements. Coarse-grained modeling provides insight into the effects of polymer aggregation on the blend morphology. Notably, the computed average distance between the donor and the acceptor polymers correlates well with solar cell photovoltaic metrics such as short-circuit current density (J_{sc}) and represents a useful index for understanding/predicting active layer blend material intermixing trends. Importantly, these results demonstrate that for polymers with different texturing tendencies (amorphous/semicrystalline), the key for optimal APSC performance, photovoltaic blend morphology can be controlled via both donor and acceptor polymer aggregation.

Hole-Transfer Dependence on Blend Morphology and Energy Level Alignment in Polymer: ITIC Photovoltaic Materials Bulk-heterojunction organic photovoltaic materials containing nonfullerene acceptors (NFAs) have seen remarkable advances in the past year, finally surpassing fullerenes in performance. Indeed, acceptors based on indacenodithiophene (IDT) have become synonymous with high power conversion efficiencies (PCEs). Nevertheless, NFAs have yet to achieve fill factors (ffs) comparable to those of the highest-performing fullerene-based materials. To address this seeming anomaly, this study examines a high efficiency IDT-based acceptor, ITIC, paired with three donor polymers known to achieve high ffs with fullerenes, PTPD3T, PBTI3T, and PBTSA3T. Excellent PCEs up to 8.43% are achieved from PTPD3T:ITIC blends, reflecting good charge transport, optimal morphology, and efficient ITIC to PTPD3T hole-transfer, as observed by femtosecond transient absorption spectroscopy. Hole-transfer is observed from ITIC to PBTI3T and PBTSA3T, but less efficiently, reflecting measurably inferior morphology and nonoptimal energy level alignment, resulting in PCEs of 5.34% and 4.65%, respectively. This work demonstrates the importance of proper morphology and kinetics of ITIC → donor polymer hole-transfer in boosting the performance of polymer:ITIC photovoltaic bulk heterojunction blends.

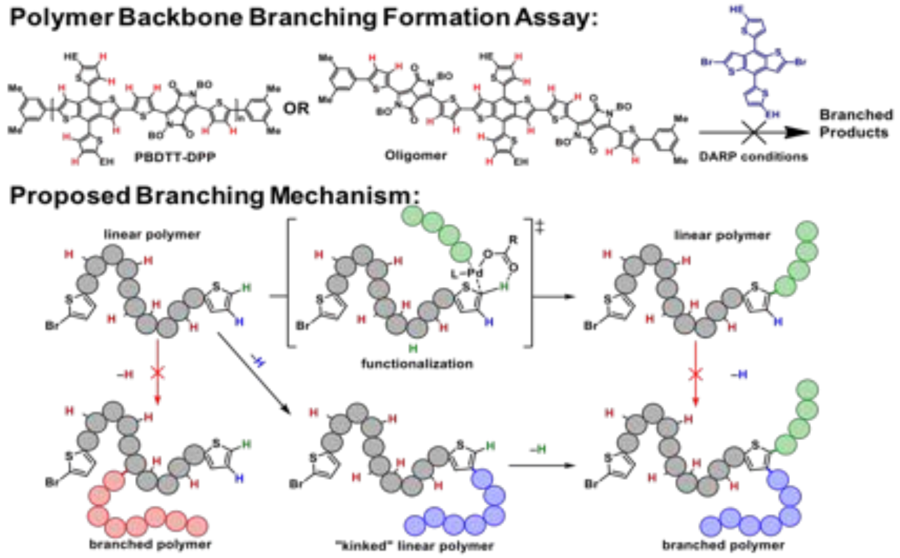


Figure 7.2: Definitive elucidation and suppression of branch formation pathways in the direct C-H arylation polymerization (DARP) pathway to a photovoltaic polymer. Suppressing these pathways greatly enhances photovoltaic efficiency

Defect Identification and Suppression in the Environmentally Benign Tin-Free Catalytic Synthesis of Photovoltaic Copolymers Direct C-H arylation polymerization (DARP) holds great promise for the green, atom-efficient synthesis of π-conjugated copolymers for use in high-performance polymer solar cells (PSCs). However, C-H arylation regioselectivity control for monomers containing multiple reactive aryl C-H bonds is not well understood, and nonselective reactivity results in material defects with unknown effects on PSC performance. Here, the effects of reaction conditions on copolymer molecular mass, dispersity, and PSC performance as well as defect formation pathways occurring during the DARP synthesis of an archetypal benzodithiophene-alt-diketopyrrolopyrrole copolymer, PBDTT-DPP, are scrutinized. Small molecule model studies analyzed by HPLC-HRMS elucidate the effects of DARP conditions on trace chemical defect (primarily hydrodehalogenation and β-C-H arylation) formation. Copolymer branching arising from nonselective β-C-H arylation of monomers at the polymer chain end is identified as the principal photovoltaically deleterious defect. Fine-tuning the DARP reaction conditions reduces branching densities to below 1%, with an exceptional C-H regioselectivity of >110:1. The optimal copolymers achieve superior PSC performance versus defect-rich DARP-derived copolymers and rival those from Stille polycondensations.

Performance, Morphology, and Charge Recombination Correlations in Ternary Squaraine Solar Cells Ternary bulk-heterojunction organic solar cells (BHJ-OSCs) are demonstrated by combining two squaraine donors (USQ3OH and IDPSQ) having complementary optical absorption and PC71BM as the acceptor. While the corresponding binary cells exhibit maximum power conversion efficiencies (PCEs) of 4.65% (IDPSQ binary) and 6.85% (USQ3OH binary), the ternary cells of weight composition IDPSQ:USQ3OH:PC71BM = 0.15:1.0:3.0 (15%TB, TB = ternary blend) exhibit a PCE of 7.20%, which is the highest known value to date for a squaraine OSC. Single crystals of both squaraines and space-charge-limited current (SCLC) measurements explain the efficiency difference between the binary cells. SCLC measurements and transmission electron microscopy imaging of the ternary devices indicate that the charge mobility slightly increases and the BHJ domain size optimizes for the 15%TB device vs that based on the USQ3OH blend. Grazing incidence wide-angle X-ray scattering data reveal that enhanced $\pi-\pi$ stacking and larger correlation lengths can be achieved after thermal annealing of the ternary blend film. Charge recombination measurements demonstrate that IDPSQ can be incorporated into the blend without increasing charge recombination. Finally, flexible OSCs on PET (polyethylene terephthalate) with a PCE of ∼4.5% were fabricated. This study demonstrates that readily accessible squaraine cores represent a viable choice for the design of new organic solar cell donor materials.

Enhancing Indacenodithiophene Acceptor Crystallinity via Substituent Manipulation Increases Organic Solar Cell Effciency We have compared and contrasted the structural, physical, photophysical, and BHJ photovoltaic properties of an ITIC series, ITIC-CX, in which the n-alkyl substituent dimensions are incremented from n-propyl to n-hexyl to n-nonyl. The ITIC-CX acceptors and their blends with two selected donor polymers (vide infra) were characterized by optical absorption spectroscopy, cyclic voltammetry (CV), differential scanning calorimetry (DSC), atomic force microscopy (AFM), X-ray diffraction (XRD), grazing incidence wide-angle X-ray scattering (GIWAXS), space-charge limited current (SCLC) mobility measurements, photovoltaic response, and PV response as a function of light intensity. The PSC performance of the ITIC-CX series is evaluated in blends with the large-bandgap, high-effciency donor polymers poly{[4,8-bis[5-(2-ethylhexyl)-2-thienyl]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl]-alt-[2,5-thio-phenediyl[5,7-bis(2-ethylhexyl)-4,8-dioxo-4H,8H-benzo- [1,2-c:4,5-c'] dithiophene-1,3-diyl]]} (PBDB-T), and the fluorinated analogue having higher crystallinity and a lower-lying HOMO, poly{[4,8-bis[5-(2- ethylhexyl)-4-fuoro-2-thienyl]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl]-alt-[2,5-thiophenediyl[5,7-bis(2-ethylhexyl)-4,8-dioxo- 4H, 8H- benzo[1,2-c:4,5-c']-dithio-phene-1,3-diyl]]} (PBDB-TF). The results show that ITIC-C9 achieves higher PSC performance than ITIC-C6 with both PBDB-T and PBDB-TF.

Sustainable Post-Fullerene Solar Cells via Direct C-H Arylation Polymerization of Benzodithiophene based Donor Copolymers An environmentally benign direct C-H arylation polymerization (DARP) process enables the tin-free synthesis of a donor-acceptor benzodithiophene-based copolymer series, PBDT(Ar)-FTTE, with systematically varied macromolecular heteroaryl substituents for next-generation sustainable, non-fullerene acceptor (NFA)-based polymer solar cells (PSCs). The photovoltaic response of these copolymers is evaluated in bulk heterojunction (BHJ) blends with the indaceno-dithienothiophene (IDTT)-based acceptor ITIC-Th. The resulting PSCs exhibit power conversion efficiencies (PCEs) greater than 8%, excellent photo-stability versus their fullerene-based counterparts, and an ambient shelf life greater than one year without encapsulation. The BHJ blend morphologies are characterized by SCLC, AFM, GIWAXS, R-SoXS, and NEXAFS measurements which demonstrate the beneficial effects of smaller well-blended copolymer domains on PSC performance. This is the first report of PSCs based on DARP-derived copolymers and NFAs.

Homologous 2D Halide Perovskites: How Thick Can They Get? In the fast-evolving field of halide perovskite semiconductors, the two-dimensional (2D) perovskites (A')2(A)n-1MnX3n+1, where A = Cs+, CH3NH3+, HC(NH2)2+, A'= ammonium cation acting as spacer; M = Ge2+, Sn2+, Pb2+ and X = Cl-, Br-, I-, have recently made a critical entry. The n value defines the thickness of the 2D layers which controls the optical and electronic properties. The 2D perovskites have demonstrated preliminary optoelectronic device lifetime superior to their 3D counterparts. They have also attracted fundamental interest as solution-processed quantum wells with structural and physical properties tunable via chemical composition, notably by the n value defining the perovskite layer thickness. The higher members (n>5) have not been documented and there is a keen interest and important scientific questions to determine fundamental limits n. Therefore, before we can further develop and utilize these materials in a technologically meaningful sense, it is imperative to understand their thermodynamic stability and fundamental synthetic limitations and the derived structure-function relationships. We report the first effective synthesis of the highest iodide n-members yet, namely (CH3(CH2)2NH3)2-(CH3NH3)5Pb6I19 (n = 6) and (CH3(CH2)2NH3)2(CH3NH3)6Pb7I22 (n = 7), and confirm the crystal structure with single-crystal X-ray diffraction and provide indirect evidence for "(CH3(CH2)2NH3)2(CH3NH3)8Pb9I28" ("n=9"). Direct HCl solution calorimetric measurements show the compounds with n > 7 have unfavorable enthalpies of formation (δHf), suggesting the formation of higher homologues to be challenging. Finally, we report preliminary n-dependent solar cell efficiency in the range of 9-12.6% in these higher n-members, highlighting the strong promise of these materials for high performance devices.

A Chemically Doped Naphthalenediimide-Bithiazole Polymer for n-Type Organic Thermoelectrics The synthesis of a novel naphthalenediimide (NDI)-bithiazole (Tz2)-based polymer [P(NDI2OD-Tz2)] is reported, and structural, thin-film morphological, as well as charge transport and thermoelectric properties are compared to the parent and widely investigated NDI-bithiophene (T2) polymer [P(NDI2OD-T2)]. Since the steric repulsions in Tz2 are far lower than in T2, P(NDI2OD-Tz2) exhibits a more planar and rigid backbone, enhancing $\pi - \pi$ chain stacking and intermolecular interactions. In addition, the electron-deficient nature of Tz2 enhances the polymer electron affinity, thus reducing the polymer donor-acceptor character. When n-doped with amines, P(NDI2OD-Tz2) achieves electrical conductivity ($\approx$0.1 S cm-1) and a power factor (1.5 μW m-1 K-2) far greater than those of P(NDI2OD-T2) (0.003 S cm-1 and 0.012 μW m-1 K-2, respectively). These results demonstrate that planarized NDI-based polymers with reduced donor-acceptor character can achieve substantial electrical conductivity and thermoelectric response.

7.4 Collaborations

7.4.1 CHiMaD Collaborations

Coarse Grain Modeling of All-Polymer Solar Cell Materials
*Monica **Olvera de la Cruz** (NU), Tobin **Marks** (NU)*
This collaboration models donor polymer-acceptor polymer blends to understand molecular mass effects on blend rheology, mixing, microstructure, and photovoltaic response.

X-ray Scattering of Photoactive Organic Thin Films
*Michael **Bedzyk** (NU), Lin Chen (NU), Tobin **Marks** (NU)*
This collaboration analyzes structural order in bulk-heterojunction organic photovoltaic materials by x-ray scattering.

Theoretical guidance in experimental design by modeling effects of molecular packing on charge-transport properties
*Guilia **Galli** (UC), Tobin **Marks** (NU)*

This collaboration studies of charge transport using Marcus theory, predicting the hole mobilities for fluorinated diketopyrrolopyrrole monomers and polymers.

Development of New Donor and Acceptor Building Blocks
*Luping **Yu** (UC), Tobin **Marks** (NU)*
Synthesis and characterization of bulk-heterojunction structures composed of UC high-performance acceptors with NU high-performance donors and vice versa.

7.4.2 NIST Collaborations

Grazing Incidence Wide-Angle X-Ray Scattering (GIWAXS), Near Edge X-Ray Absorption Spectroscopy, and Resonant Soft X-Ray Scattering (RSoXS)
*Lee **Richter** (NIST), Dean **Delongchamp** (NIST), Subhrangsu **Mukherjee**, Tobin **Marks** (NU)*
Photoactive bulk heterojunction blends are morphologically complex systems. Understanding the basic principles of materials "mixing" at the nanoscale level is a critical for fundamental knowledge to advance the performance and manufacturability of organic solar cells. The aforementioned techniques are extremely important for the optimization of blend morphology, understanding microstructure, elucidating chemical structure - photovoltaic performance relationships, and guiding the synthesis of next-generation materials.

Thermal Analysis of All-Polymer Solar Cell Materials
*Chad **Snyder** (NIST), Tobin **Marks** (NU)*
Investigation of the thermal properties of donor polymer-acceptor polymer blends to understand molecular mass effects on blend rheology, mixing, microstructure, and ultimately photovoltaic response.

Design. Synthesis, Processing, and Industrial Evaluation of Semiconducting Polymers and Donor-Acceptor Blends for Photovoltaic Polymers
*Antonio Facchetti (flexterra Inc.; Raynergy Inc.), Tobin **Marks** (NU)*
This collaboration focuses on the design and synthesis of n- and p-type semiconducting polymers for OPVs. flexterra and Raynergy provide expertise in scale-up coating, printing, and evaluation of organic solar cell modules made with Northwestern materials.

Sample Evalutation
*Lee **Richter** (NIST), Luping **Yu** (UC)*
Samples produced by Yu group at UC were evaluated by Richter group. Towards this goal, gold standard SANS measurements were performed to validate MD polymer folding and solubility calculations and the thermal properties of Polymer-Polymer OPV active layers were evaluated.

7.4.3 External Collaborations

Microstructure Analysis of Bulk Heterojunction Photovoltaic Blends
*Vinayak Dravid (NU), Tobin **Marks** (NU)*
This collaboration focuses on the analysis of phase separation, microstructure, and local atomic composition in bulk heterojunction photovoltaic blends by advanced electron microscopy techniques.

GIWAX measurements
*Wei Chen (ANL), Luping **Yu** (UC)*
This collaboration focuses on GIWAX measurements of polymers for OPV.

Charge transfer in single molecular acceptors
*John Grey (University of New Mexico), Luping **Yu** (UC)*
This collaboration investigated the effect of Conformational flexibility on Electronic Coupling and Charge Transfer Character in Single Propeller-Shaped Perylene Diimide Tetramer Arrays.

Two photon absorption in electron acceptors
*Theodore Goodson (University of Michigan), Luping **Yu** (UC)*
This collaboration focused on studies of Charge Transfer and Aggregation Effects on the Performance of Planar vs Twisted Nonfullerene Acceptor Isomers for Organic Solar Cells.

7.5 Technology Transfer

Patent
Antiambipolar heterojunctions from solution-processed semiconductors
A. Facchetti, T. J. Marks, Y. Xia, M. Drees, F. Melkonyan, W. Zhao
Date: 2018
Patent Number: U.S. Patent No. 9,929,725 (2018)
This technology describes a scalable, easily implemented solution process for fabricating opto-electronic devices for energy conversion/current generation such as in a solar energy cell.

7.6 Publications and Presentations

14 Number of CHiMaD-supported publications in 2018 by Organic Bulk Heterojunction Polymer Solar Cells use-case group. Please see chapter 27 for details [27.106, 27.107, 27.108, 27.109, 27.110, 27.111, 27.112, 27.113, 27.114, 27.115, 27.116, 27.117, 27.118, 27.119].

6 Number of presentations on CHiMaD supported research in 2018, please see section 26.6 for the complete list.

7.7 CHiMaD Team

Organic Bulk Heterojunction Polymer Solar Cells Use-Case Group Researchers			
Name	**Position**	**Affiliation**	**Project (PI)**
Luping Yu Juan de Pablo Guilia Galli Tobin Marks	Principal Investigator Principal Investigator Principal Investigator Principal Investigator	UC/CHEM UC/IME UC/IME NU/MSE	
Matthew Goldey	Postdoctoral Researcher	UC/IME	Electronic structure models of semiconducting small molecules and polymers for organic photovoltaics *(Galli)*
Ferdinand Melkonyan	Research Assistant Professor	NU/CHEM	Sulfonamide Organic Photovoltaic Building Blocks; Diacetylenes as Organic Photovoltaic Building Blocks *(Marks)*
Gang Wang	Postdoctoral Researcher	NU/CHEM	GIWAXS Characterization; Polymer Processing *(Marks)*
Daniel Reid	Graduate Student	UC/IME	Simulation of OPV materials *(de Pablo)*
Donglin Zhao	Graduate Student	UC/CHEM	OPV devices based on non-fullerene electron acceptors *(Yu)*
Valerii Sharapov	Graduate Student	UC/CHEM	OPV devices based on ternary system. *(Yu)*

III

CHiMaD Seed Groups

8. Additive Manufacturing

Jian Cao (NU), Wing Kam Liu (NU)

Lyle Levine (NIST), Li Ma (NIST), Carelyn Campbell (NIST), Mark Stoudt (NIST)

8.1 *Significance & Research*

Additive manufacturing breaks three fundamental manufacturing tradeoffs: capital versus scale, capital versus scope, and capital verses complexity. It also significantly reduces material waste by placing materials only where needed. Widespread acceptance of additive manufacturing will increase supply chain efficiency, reduce time to market, move from mass production to mass customization, and sustain the environment. This will spur a technology renaissance by bridging manufacturing of all scales with material design and will broaden participation by disseminating accessibility to cutting-edge manufacturing processes.

The technique development and modeling efforts of the Additive Manufacturing seed group complement the efforts on Alloy Design for Additive Manufacturing under Precipitation Strengthened Alloys use-case group, which are summarized in Chapter 6.

8.2 Directed Energy Deposition (DED)

Research accomplishments described below include **Cao** Group's prediction of thermal history using machine learning, in-situ X-ray monitoring of directed energy deposition process (DED), and life cycle analysis of dies repaired by additive manufacturing, specifically, by DED.

Prediction of thermal history in the directed energy deposition process using recurrent neural networks Due to the limited number of experiments and the omission of crucial time-series features of the directed energy deposition (DED) process such as the toolpath, which directly affects thermal history and hierarchical microstructure in additively-manufactured materials, current data-driven models fail to provide a profound understanding of this process. To address this gap, one research accomplishment is a recurrent neural network

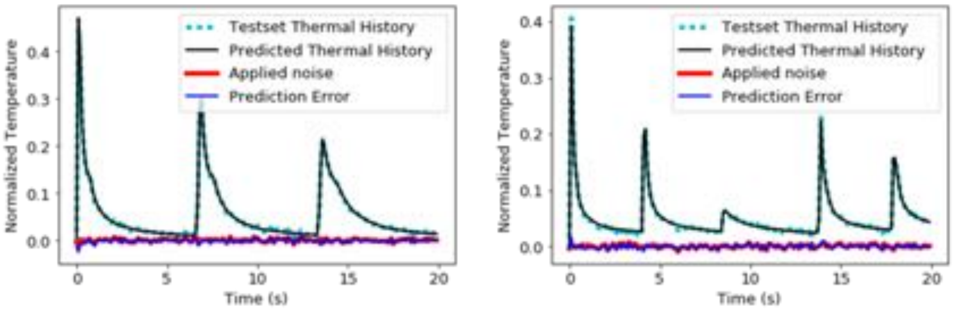


Figure 8.1: Evaluation of the stacked RNN model on the test dataset for two random points over 20 s of the DED process; Comparison of the model prediction (black line) and the test-set value (cyan dashed line) for the thermal history of a point in a thin-wall build with uni-directional toolpath (left) and a cylindrical build circular toolpath (right)

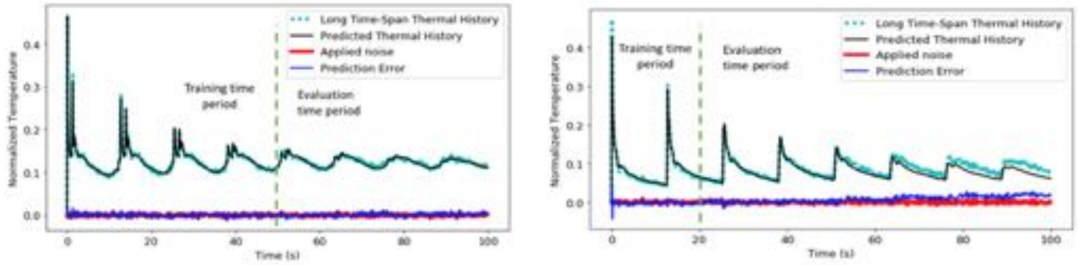
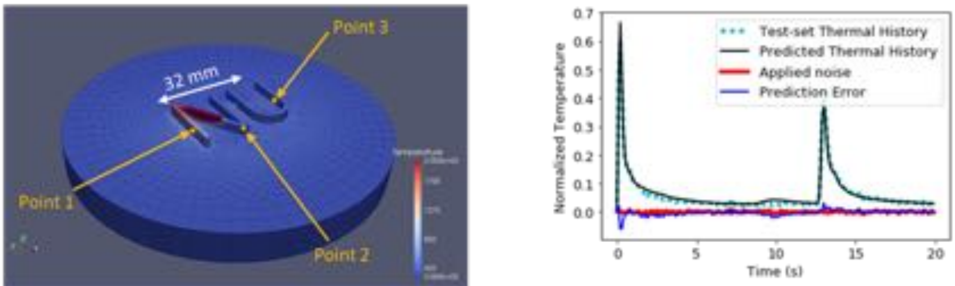


Figure 8.2: Evaluation of the stacked RNN model on the test dataset for 100 s, while trained on 20 s (top) and 50 s (bottom) of the process; Comparison of the model prediction (black line) and the test-set value (cyan dashed line) for the long time span thermal history of a point in a cubic build with zigzag toolpath

(RNN) structure to predict the thermal history of any given localized area within a DED build in a many-to-many configuration. This approach is well-suited for DED processes since it can accurately calculate the high-dimensional thermal history of the builds in a computationally efficient manner.

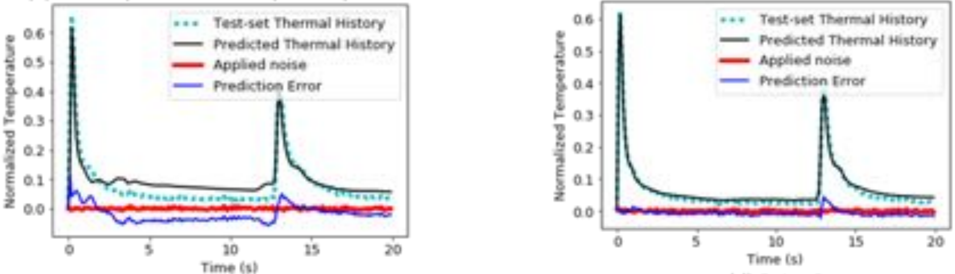
The stacked RNN structure has GRU formulation that allows for thermal history prediction at any given spatial point of a DED build. The results show that the model reaches an MSE of $2.97e^{-5}$ on a test dataset after a 100 epoch training. Additionally, two overarching tests for predicting the thermal history over a longer time span and non-trained geometries are examined, showing the potential of RNN models to predict complex behaviors in AM processes, considering that the accuracy of the model can be further improved by increasing training epochs and geometry types included in the database. (Figures 8.1-8.3)

In-situ high-speed X-ray monitoring of the directed energy deposition process The DED process adds flexibility, in terms of locally varying powder materials, to the ability of building components with complex geometry. Although the process is promising, porosity is common in a built component, hence decreasing fatigue life and mechanical strength. The understanding of the physical phenomena during the interaction of a laser beam and powder-blown deposition is limited and requires in-situ monitoring to capture the influences of process parameters on powder flow, absorptivity of laser energy into the substrate, melt pool dynamics and porosity formation. This study introduces a piezo-driven powder deposition system that allows for imaging of individual powder particles



(a) NU-shape build and inspected points

(b) Point 1



(c) Point 2

(d) Point 3

Figure 8.3: Evaluating the trained model on a dissimilar geometry; The NU-shape build geometry and the inspected point locations (a), comparative figures for the points 1, 2 and 3 between model prediction and the test-set (b), (c), and (d), respectively. The toolpath of this build goes from the bottom left to the upper right side of the letter N and then moves from the upper left to the upper right side of the letter U

that flow into a scanning melt pool. Here, in-situ imaging of the powder-blown additive manufacturing process of Ti-6Al-4V powder particles is the first of its kind and reveals how laser-matter interaction influences powder flow and porosity formation.

This study used a high-resolution, time-resolved X-ray imaging technique at the Advanced Photon Source at Argonne National Laboratory to evaluate the interactions between the laser beam, flying particles, and the melt pool behavior during rapid solidification in DED. High-speed X-ray images reveal the laser-matter interactions in various modes of DED processing can aid in the validation of thermal, thermo-fluid dynamic and thermo-mechanical models. In the case of gravity-fed, low powder mass flow, the laser-induced vapor plume scatters particles away from the melt pool with velocities of up to 10 m/s. Surface tension of the melt pool entrains particles on the surface to enter the melt pool or ejects hot particles. Various mechanisms allow for individual in-flight particles to enter the melt pool, including particle interactions and localized pressure gradients from a scanning laser. Particles that flow into the melt pool influence the stability of the laser-induced cavity and the surrounding melt pool. Porosity continually forms, stirs within the melt pool, shrinks into the liquid phase of the substrate, or is constrained at the solidification boundary. Changes in mass entering or ejecting out of the melt pool influence the resulting cooling rates of the melt pool, which can reach up to 10^6 K/s. (Figures 8.4-8.7)

Repairing Automotive Dies with Directed Energy Deposition: Industrial Application and Life Cycle Analysis Powder-based additive manufacturing technologies are developing rapidly. To assess its applicability, comparison of performance and environmental impacts of additive technologies and conventional techniques must be performed. Toyota manufactures over two million aluminum four-cylinder engines in the United States each year

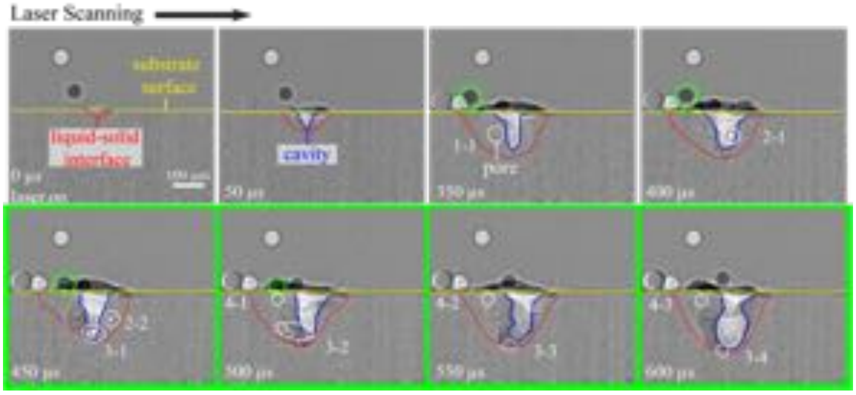


Figure 8.4: Cavity, melt pool and porosity evolution as mass is added due to surface tension in an experiment with a laser power of 250 W and a scan speed of 100 mm/s. All frames are processed images.

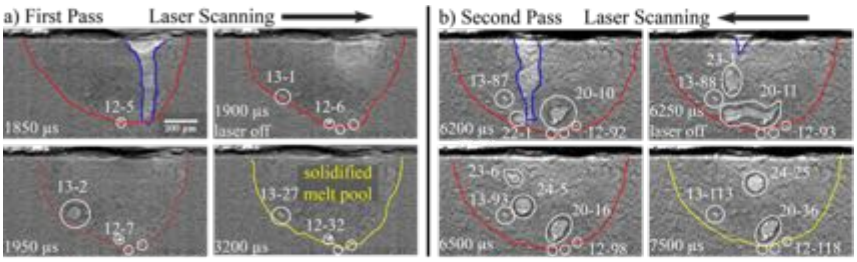


Figure 8.5: Evolution of keyhole porosity after the laser is turned off and until the substrate is solidified for two passes. The experimental conditions are 150 W laser power, 100 mm/s, 400 V voltage input into the piezo-driven powder delivery system with a pulse frequency of 100 Hz, duty cycle of 10% for each pulse

via die casting. The original dies used in this process last 12,000 cycles before a repair is needed. A die repaired via the traditional repair process, tungsten inert gas (TIG) welding, only lasts an average of 2,500 cycles. A hybrid repair process involving machining away the damaged areas and then rebuilding them additively via powder-blown directed energy deposition (DED) has been developed. The repair was performed in the DMG MORI LaserTec 65 3D hybrid additive and subtractive machine. (Figure 8.8)

The die repaired via DED resulted in the same life as the original die, 12,000 cycles. The use of DED repairs eliminated the need for emergency repairs and non-scheduled downtime on the line because the DED repaired dies only require the planned periodic repairs every 10,000 cycles. Life cycle analyses were conducted comparing the traditional welding repair process to the DED repair process. The Eco-Indicator 99 scores for the proposed DED repair method, and an impact breakdown shows that impacts are highest on fossil fuels (score of 0.29), followed by respiratory effects (score of 0.24), and carcinogenics (score of 0.04). Ionizing radiation and ozone layer depletion impacts are negligible (Figures 8.9-8.10). Together, the fossil fuels and respiratory effects impacts account for 86% of the total Eco-Indicator 99 score. In general, electricity use during the repair process and gas atomization in producing H13 powder are associated with higher impacts than other processes. Energy use during the DED repair process dominates many of the impact categories. Electricity used during the repair process accounts for 52% of the respiratory effects impact, 50% of the carcinogenics impact, 46% of the climate change impact, 47%

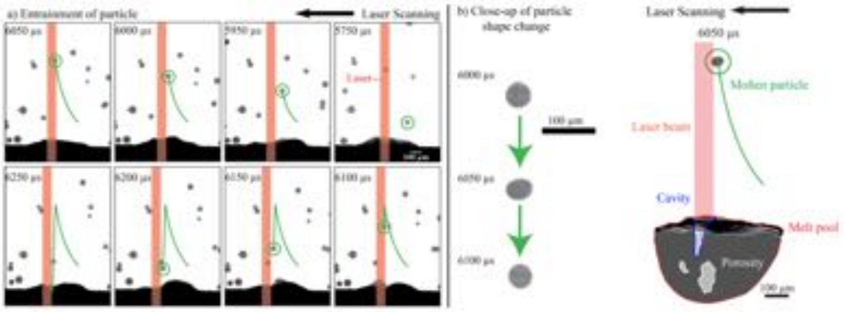


Figure 8.6: (Left) Example of vapor entrainment during a keyhole mode experiment where the laser power is 150 W and the scan speed is 100 mm/s where a) shows the particle behavior as the laser moves from right to left and b) the change in particle shape as the laser hits the in-flight particle; (Right) Example of vapor-driven entrainment of a single particle from left. When the laser is attenuated, the cavity within the melt pool collapses, forming porosity.

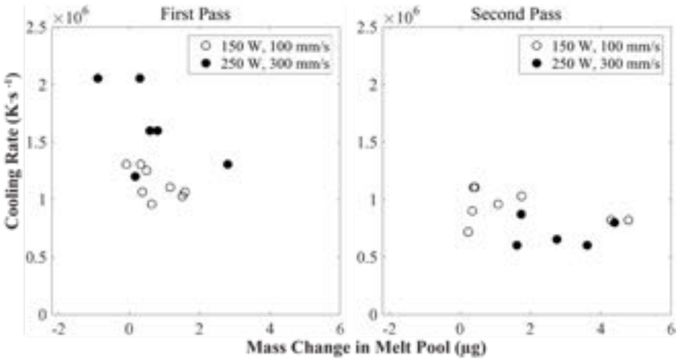


Figure 8.7: Cooling rate with the change of mass in the melt pool for each pass during each keyhole mode experiment. Each data point represents a pass in one experiment

of the ecotoxicity impact, 49% of the land occupation impact, 43% of the acidification and eutrophication impact, and 38% of the mineral extraction impact. Results show that the DED repair process results in significantly less damage to the assessed impact categories except for ionizing radiation. Therefore, it can be concluded that the DED repair process could lessen most environmental impacts compared to traditional welding repair. Further work towards increasing energy and material efficiencies of the method could yield further reductions in environmental impacts.

Cooling Rate Effect on Tensile Strength of Laser Deposited Inconel 718

The thermal history generated by the additive manufacturing process influences the resulting material properties. Although trends exist between solidification rate and microstructure, solidification rate is not enough to predict final microstructure and thus mechanical properties. The purpose of this study is to relate the combined effects of solidification time and cooling time of the built material to its final ultimate tensile strength. Cooling time was defined as the time from when the location of interest last passes through $1,200°C$ to when it reaches $400°C$.

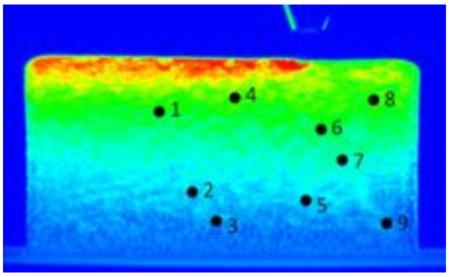


Figure 8.12: Temperature measurement of locations of interest on wall

Figure 8.8: (Left) Die Condition prior to repair; (Right) Repaired die.

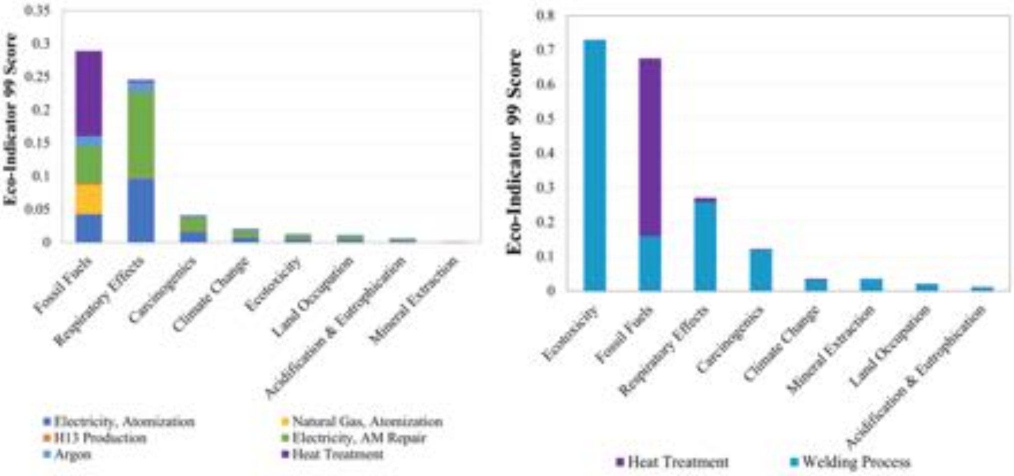


Figure 8.9: (Left) Eco-Indicator 99 Scores for the proposed DED repair method. Ozone layer depletion and ionizing radiation impacts were negligible; (Right) Eco-Indicator 99 Scores for the conventional welding repair method. Ionizing radiation and ozone layer depletion impacts were negligible.

Nine locations on a laser deposited IN718 thin wall were studied in detail to understand the effect of cooling rate on tensile strength. Tensile samples were machined at these locations. (Figure 8.12)

The thermal histories of the locations of interest were compared with build geometry and the ultimate tensile strength of that location. An inverse proportional relationship was seen between the distance of the location of interest from the substrate and the cooling time (Figure 8.11 - Left).

A trend was also seen linking increased surface temperature and increased solidification time. Weighted Cooling And Solidification Time (WCAST) was defined as the sum of weighted normalized solidification time and the normalized cooling time. Ultimate tensile strength was seen to decrease as WCAST increased (Figure 8.11 - Middle). Optical microscopy images of the build microstructure confirm that longer cooling and solidification times lead to coarser microstructures, which may cause the lower tensile strengths measured (Figure 8.11 - Right).

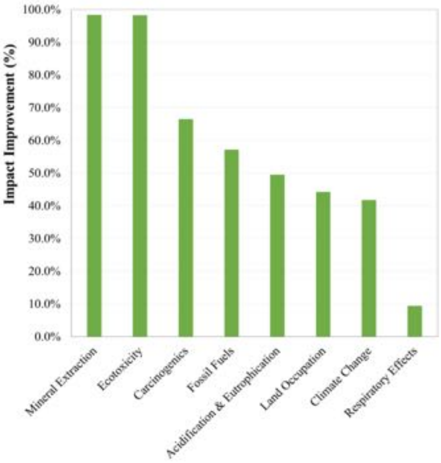


Figure 8.10: Impact improvement of the DED repair method compared to the conventional welding repair method

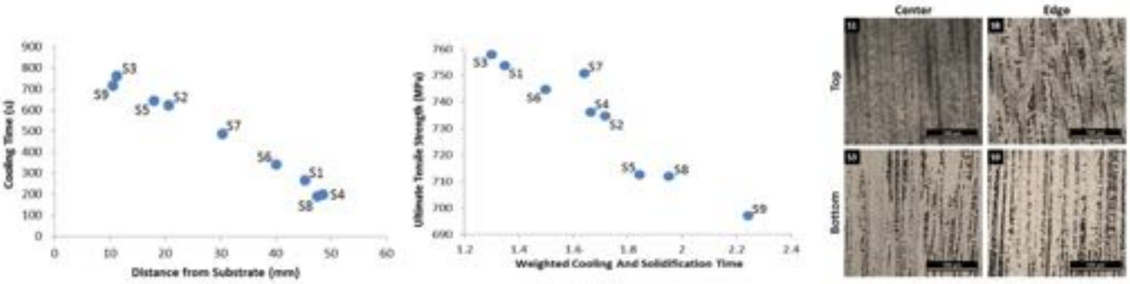


Figure 8.11: (Left) Cooling time versus distance from substrate; (Middle) Ultimate tensile strength versus WCAST; (Right) Optical microscopy images showing microstructures at different locations of the sample

8.3 High-dimensional process-structure-properties linkages in AM

Liu Group developed a novel workflow to produce, visualize, and design high-dimensional process-structure-property linkages in AM through the utilization of multiphysics modeling, experimental measurements, and data science techniques, Figure 8.13. An application of this workflow to quantify and design microstructure and properties was demonstrated. The high-dimensional datasets, which include laser power, mass flow rate, energy density, cooling rate, dilution, dendrite arm spacing, and microhardness, have been derived by physics-based models and experiments. The profound relations among P-S-P data have been visualized by self-organizing map (SOM). The important relationship trends have been found. Multiple objective optimization examples are proposed and solved to seek desired process parameters

8.4 Additive Manufacturing Benchmark Challenge

In the additive manufacturing benchmark (AM-BENCH) experiments, melt pool geometry, cooling rates, surface topography, and dendritic microstructure in laser melting Inconel 625 plate were used to challenge and validate the thermal computational models that must incorporate the pertinent fundamental physics. To this end, three thermal models incorporating different physics are compared with the experimental data by **Liu** group. It is identified that the heat convection enhanced by the thermocapillary flow inside the molten pool and heat loss caused by vaporization play pivotal roles to guarantee the accuracy

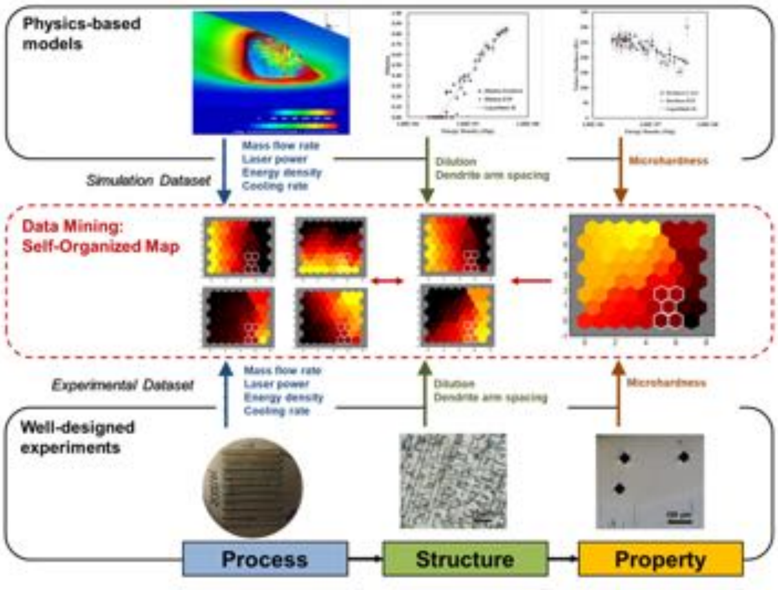


Figure 8.13: A schematic description of the workflow typically employed in current computational efforts (top row) and experimental efforts (bottom row), and how this can be augmented with a data mining approach to recover high-value P-S-P linkages of interest to material innovation efforts.

of the predictions and should be considered in the thermal model. Zhengtao Gan (NU), Yanping Lian (NU), Stephen E. Lin (NU), Kevontrez K. Jones (NU), Wing Kam **Liu**, and Gregory J. **Wagner** won (1st Place) the following AM-Bench 2018 Benchmark Challenge Awards with this work: (1) Best modeling results predicting the cooling rates of three single laser tracks on a bare IN625 plate; (2) Best modeling results predicting the grain structure of three single laser tracks on a bare IN625 plate; (3) Best modeling results predicting the dendritic microstructure of three single laser tracks on a bare IN625 plate.

8.5 Impact Statement for CHiMaD Phase I

The research detailed above are steps towards quick qualification of components, an increase of process autonomy, true integration of design and manufacturing, and ultimately a release of this technology from the hands of a few to the hands of many.

8.6 Collaborations

8.6.1 CHiMaD Collaborations

Experimental and computational investigations of AM Specimens
*Gregory J. Wagner (NU), Wing Kam **Liu** (NU), Kornel F. Ehmann (NU) and Jian **Cao** (NU)*
Graduate students have been working closely together to link experimental and computational results of specimens created via additive manufacturing.

Additive Manufacturing Modeling
*Jian **Cao** (NU), Wing-Kam **Liu** (NU)*
This collaboration aims to generate experimental results used for validation of and input for microscale, macroscale and reduced order models for processing and mechanical properties of AM materials. This includes in-situ mechanical testing at Argonne National

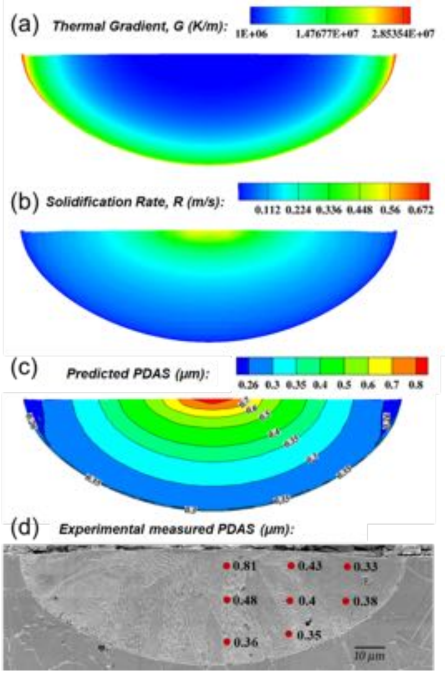


Figure 8.14: Distribution of thermal gradient, solidification rate, predicted primary dendrite arm spacing (PDAS), and experimental measured PDAS at the cross-section for case B (195 W, 800 mm/s): (a) thermal gradient, (b) solidification rate, (c) predicted PDAS, and (d) experimental measured PDAS.

Laboratory's APS using Beamline 2BM to produce 3D images of the void morphology during extensile deformation - a capability available at only a few facilities worldwide. We have also conducted micro-Laue diffraction at beamline 34IDE to understand residual stress and grain orientation in 3D. Other collaboration includes using techniques such as IR imaging and SEM imaging to determine solidification/cooling rates for comparison to thermal simulations.

8.6.2 NIST Collaborations

AM Research Findings Collaboration

*Lyle **Levine** (NIST), Li **Ma** (NIST), Carelyn **Campbell** (NIST), Mark **Stoudt** (NIST), Jian **Cao** (NU)*

The NIST and CHiMaD researchers regularly exchange of research findings.

Additive manufacturing

*Lyle **Levine** (NIST), Yan Lu (NIST), Jason Fox (NIST), Paul Witherell (NIST), Gregory Wagner (NU), Wing-Kam **Liu** (NU)*

Discussions on the recent developments in AM and the groups' contributions are ongoing. This collaboration intends to quantify uncertainties in methods commonly used for AM process modeling. This work incorporates the use of data-analytics methodology, calibration experiments, and AM process models ranging from low-fidelity to high-fidelity.

8.6.3 External Collaborations

Digital Manufacturing and Design Innovation Institute (DMDII)

*Kornel F. Ehmann (NU), Greg J. Wagner (NU), Lucia Mirabella (Siemens), Suraj R. Musuvathy (Siemens), Edward R. Slavin III (Siemens), Sanjeev K. Srivastava (Siemens), Federico M. Sciammarella (NIU), Joseph S. Santner (NIU), Jason **Sebastian** (QuesTek), David **Snyder** (QuesTek), Jiten Shah (Product Development & Analysis), Jian **Cao** (NU),*

Wei ***Chen*** *(NU), Gregory* ***Olson*** *(NU), Wing-Kam* ***Liu*** *(NU)*
The external collaborators are working with a DMDII project working on AM process modeling, calibration, assessment and prediction. This work includes calibration experiments, macro-scale and micro-scale simulations for mechanical properties and processing, and characterization experiments.

Life Cycle Analysis of Additive Manufacturing
Fengqi You (Cornell University), Jian ***Cao*** *(NU)*
This collaboration involves an analysis of life cycles for additively manufactured alloys.

8.7 Publications and Presentations

11 Number of CHiMaD-supported publications in 2018 by Additive Manufacturing seed group. Please see chapter 27 for details. [27.8, 27.9, 27.40, 27.11, 27.12, 27.13, 27.8, 27.9, 27.94, 27.97, 27.96]

8 Number of presentations on CHiMaD supported research in 2018, please see section 26.7 for the complete list.

8.8 CHiMaD Team

Additive Manufacturing Seed Group Researchers			
Name	**Position**	**Affiliation**	**Project (PI)**
Jian Cao Wing Kam Liu	Principal Investigator Principal Investigator	NU/ME NU/ME	
Wentao Yan	Postdoctoral Researcher	NU/ME	CFD simulation of additive manufacturing process *(Liu)*
Zhengtao Gan	Postdoctoral Researcher	NU/ME	CFD simulation of additive manufacturing process *(Liu)*
Jennifer Bennett	Graduate Student	NU/ME	Macro Thermal Control of Additive Manufacturing *(Cao, Ehmann (NU))*
Sarah Wolff	Graduate Student	NU/ME	Macro Thermal Control of Additive Manufacturing *(Cao, Ehmann (NU))*

9. Uncertainty of Thermodynamics Data and Phase Equilibria

Marius Stan (ANL)

Ursula Kattner (NIST)

9.1 Significance & Research

The goal of the seed group was to create the necessary knowledge, analysis, and software that helps researchers determine the optimal phase diagram and associated thermodynamic data. To that end, we have advanced the mathematical treatment of phase equilibrium uncertainty and have developed the methodology and computational tools necessary to update phase equilibria and thermodynamic models with the necessary uncertainty intervals. This will immediately benefit CHiMaD projects that involve analysis of material thermodynamic data and models.

Development of robust approaches to select, calibrate, and quantify the uncertainty of thermodynamic property models Thermodynamic property models are a critical component in important technological applications, including the calculation of phase diagrams (CALPHAD) and phase field simulations of microstructure evolution during material processing. These models must capture the material physics from a wide range of experimental and simulated datasets, and have good extrapolative capabilities. Unfortunately, the development of such models is subject to many challenges, including the presence of outliers, conflicting datasets, inaccurate error bars and thermodynamic inconsistencies. Furthermore, databases and journal articles often provide thermodynamic property models without parameter uncertainties or confidence intervals, precluding the assessment of the potential reliability of competing models [2]. While researchers have employed a variety of statistical techniques in addressing some of these issues, no comprehensive framework yet exists. In this work, we propose a novel Bayesian framework for calibrating, selecting and quantifying the uncertainty of thermodynamic property models along with easily implemented approaches for addressing the common issues previously mentioned. We demonstrate the efficiency of the framework by creating a thermodynamic property model for Hafnium metal.

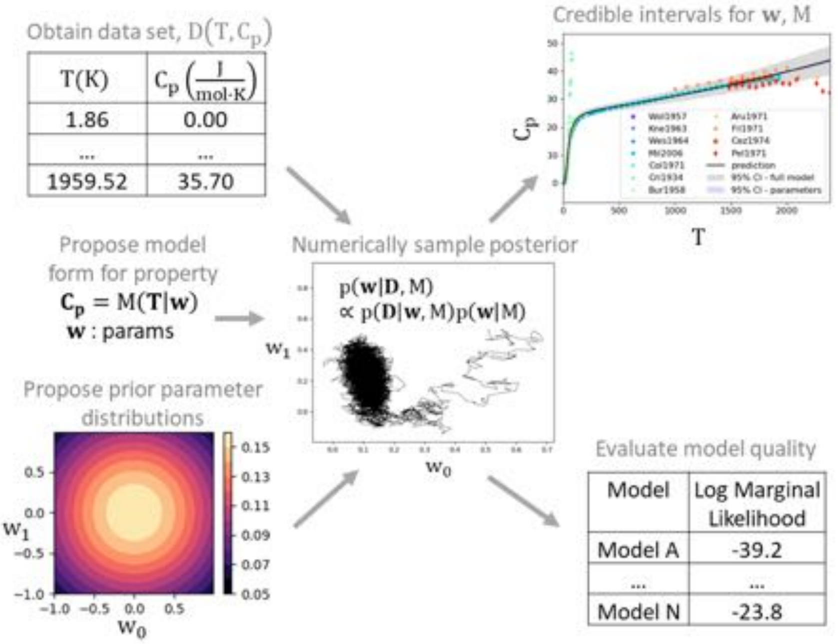


Figure 9.1: A schematic illustrating the Bayesian approach to model selection, calibration and uncertainty quantification

Figure 9.1 portrays the workflow used in this work that includes model calibration, selection and uncertainty quantification through Bayesian statistics. Starting at the top left of the figure, the base data are obtained and a model or set of potential models is proposed. Then the expert picks reasonable probability distributions for the model parameters (the prior) and for the data given the model (the Likelihood), based on physical understanding of the phenomena. Given the model, prior and Likelihood probability distributions, the distribution of the model parameters given the observed data (the posterior) can be computed according to Bayes' Theorem. In this work, the posterior distribution is computed using cutting edge numerical techniques. At the right of Figure 9.1, credible intervals are computed for the parameters (which are then propagated through to the model prediction) and model selection is performed via marginal Likelihood. This flexible Bayesian framework enables solutions to the problems mentioned earlier through careful selection of the form of the Likelihood function. Additionally, we use a novel approach to incorporate diverse measurements of different thermodynamic quantities into the same thermodynamic property model without incurring errors introduced by converting all quantities to the same basis (for example differentiating enthalpy measurements to obtain specific heat). Figure 9.2 presents the specific heat models for the alpha, beta and liquid phases of Hafnium metal resulting from this Bayesian workflow alongside the models of two previous assessments. This work provides guidelines to the CALPHAD and greater material science community to enable the development of more effective and transparent property models.

9.2 Impact Statement for CHiMaD Phase I

This seed group created an expanding community of scientists interested in uncertainty quantification that extends beyond thermodynamic applications. We trust that our innova-

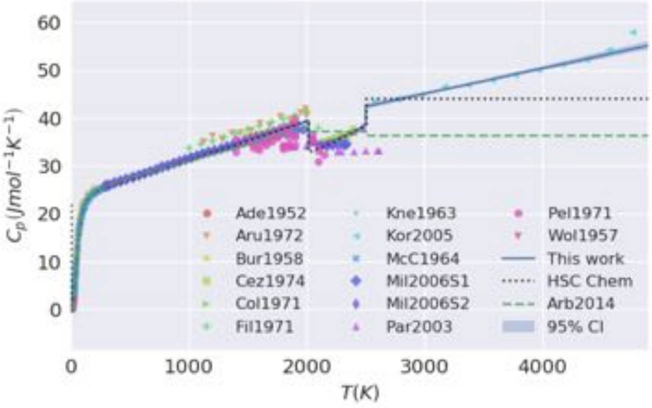


Figure 9.2: A schematic illustrating the Bayesian approach to model selection, calibration and uncertainty quantification

tive methodology will be adopted by many CHiMaD use-case groups and will become a method of choice for NIST and the materials science community in general.

9.3 Collaborations

9.3.1 CHiMaD Collaborations

Phase Field Simulations

*Olle **Heinonen** (ANL), Peter **Voorhees** (NU), Marius **Stan** (ANL)*

Stan group is an ongoing participant of the CHiMaD Phase Field Methods Workshops, where they provide expertise in testing of the community benchmark problems.

Materials Data Facility

*Marius **Stan** (ANL), Ian **Foster** (UC), Ben Blaiszik (UC)*

The collaboration between the seed group and the MDF further explores the data management of thermodynamic data.

9.3.2 NIST Collaborations

Uncertainty of thermodynamic data and phase diagrams

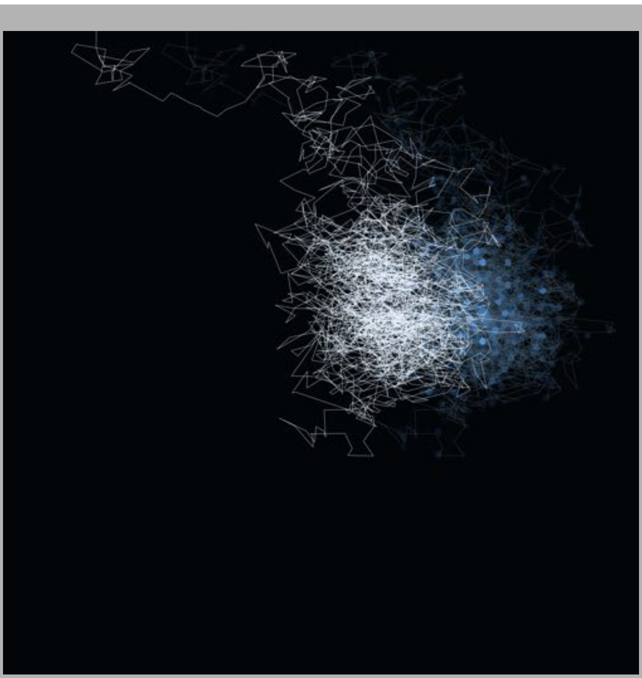
*Ursula **Kattner** (NIST), Carelyn **Campbell** (NIST), Marius **Stan** (ANL)*

The ongoing NIST-CHiMaD collaboration focuses on evaluating uncertainty of free energies and binary phase diagrams.

9.4 Publications and Presentations

2 Number of CHiMaD-supported publications in 2018 by Uncertainty of Thermodynamics Data and Phase Equilibria seed group. Please see chapter 27 for details. [27.14, 27.15]

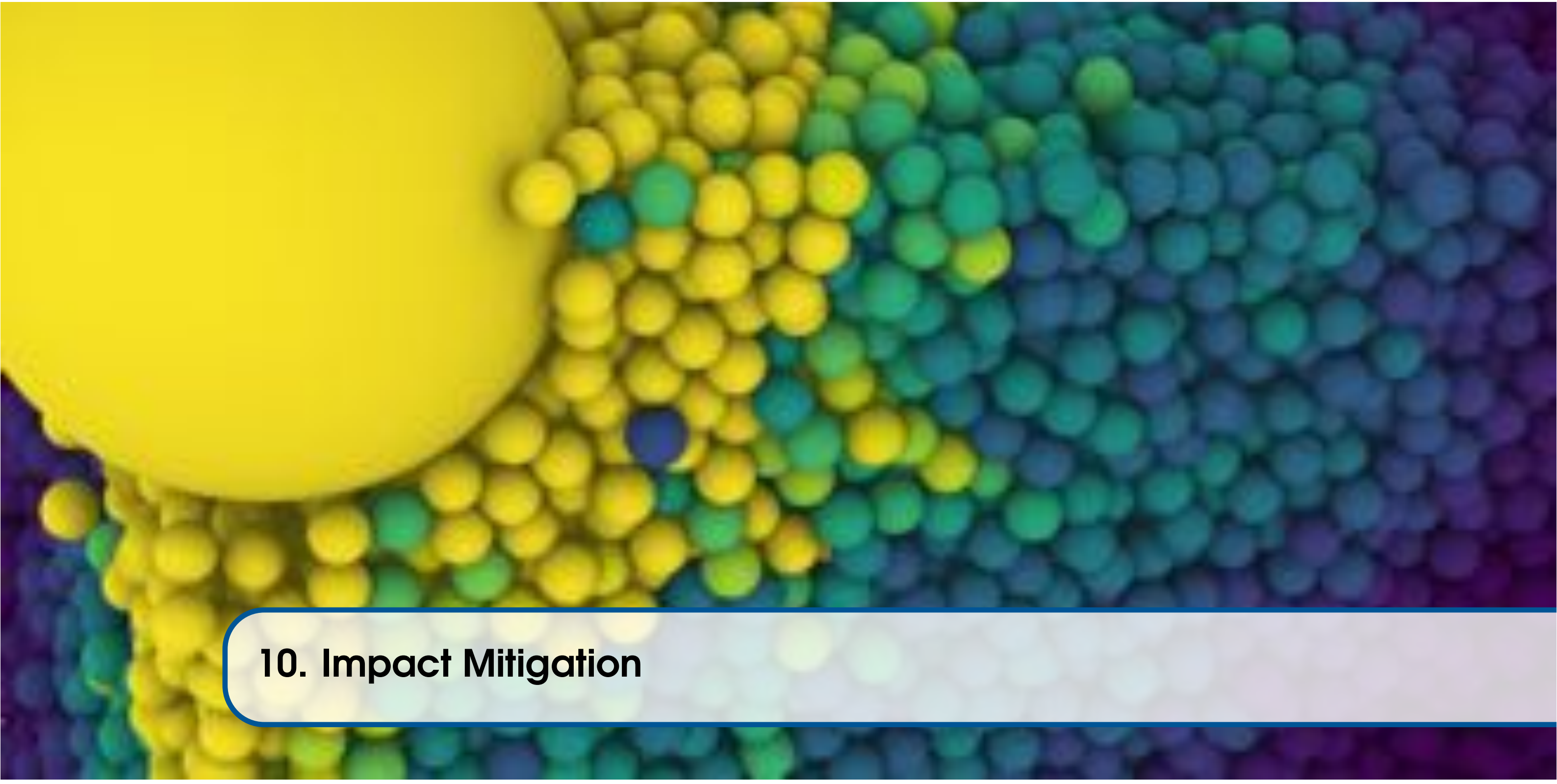
11 Number of presentations on CHiMaD supported research in 2018, please see section 26.8 for the complete list.



Header Image: Visualization of a Markov Chain Monte Carlo simulation for two thermodynamic properties and their resultant distribution [N. Paulson]

9.5 CHiMaD Team

Uncertainty of Thermodynamics Data and Phase Equilibria Seed Group Researchers			
Name	**Position**	**Affiliation**	**Project (PI)**
Marius Stan	Principal Investigator	ANL/MCS	
Noah Paulson	Postdoctoral Researcher	ANL/GSS	Uncertainty of Thermodynamics Data and Phase Equilibria *(Stan)*



10. Impact Mitigation

Heinrich Jaeger (UC), Sidney Nagel (UC), Juan de Pablo (UC)

Edwin Chan (NIST), Aaron Forster (NIST), Christopher Soles (NIST)

Significance

This Seed Group investigates new platforms for next-generation impact-protection materials in safety-critical applications. The damaging effects of blunt impact on human physiology and health have been most notable in the manifestation of the high rates of mild traumatic brain injury (mTBI) and chronic traumatic encephalopathy (CTE) in athletes. Impacts to soft tissues also are the basis for many chronic joint and musculoskeletal diseases. In team sports athletes rely on pads of geometrically structured, non-linear, time-dependent polymeric materials (e.g. elastomers, foams, thermoplastics, gas bladders, etc.), to absorb energy, limit momentum transfer, and reduce force at velocities that can exceed 45 m/s and energies up to 75 J. In many cases such materials are clearly inadequate, and there is a pressing need to reduce impact injuries to the head and body from a wide variety of threats in sporting, industrial, and military applications. Advances in protective materials have been incremental due to a lack of dynamic material property data and a lack of predictive models to spur innovation.

10.1 Design Goals

The design goals of this Seed group are to develop innovative material systems for impact mitigation that will be superior to those available today, and to do so by relying on predictive models coupled to high-precision experimentation in the context of both fluid and solid shock absorbing systems

Particle-fluid Hybrid Systems. A first promising materials platform is based on particle-fluid hybrid systems. These can easily conform to arbitrary surfaces and confining volumes in their unstressed state, and they are highly effective in dissipating energy through frictional contacts when they deform plastically under impacts. In dense suspensions of non-Brownian particles in a liquid, impact leads to a dynamic transformation from a fluid-like to a solid-like state, triggered by rapidly moving solidification, or jamming, fronts. These jamming fronts are investigated in both experiments and simulations. In dry particulate systems, where the interstitial fluid is air, shock fronts generated by impact were studied in simulations and experiments.

Architected materials. A second promising platform is based on architected materials that can be designed to guide stress transmission. A particularly interesting class of such materials are disordered networks that exhibit auxetic response to applied strain, i.e., exhibit a negative Poisson ratio.

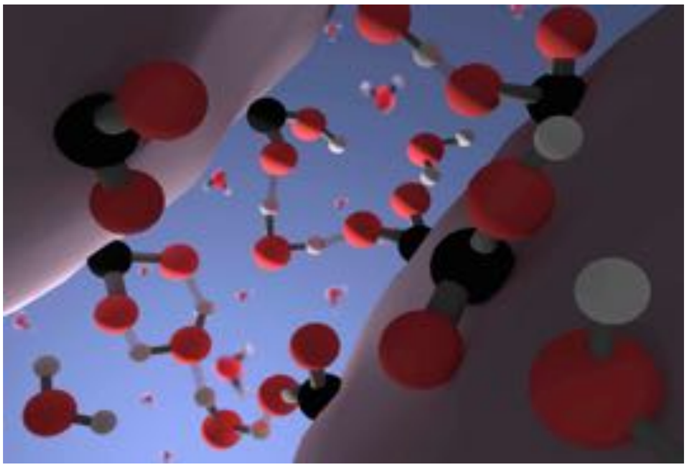


Figure 10.1: Two micron-size PMMA/ITA particles in water at close approach, with interparticle hydrogen bonding between COOH surface groups. [27.16]

10.2 Significant Accomplishments in 2018

Particle-fluid Hybrid Systems Using dense aqueous suspensions as a model for particle-fluid hybrid systems we demonstrated that shear jamming in can be controlled by tuning the propensity for hydrogen bonding between particles. This constitutes the first direct link between reversible shear jamming, a macroscale rheological phenomenon, and particle surface chemistry, which operates on the scale of nanometers. This discovery opens up new opportunities for designing rate-dependent impact mitigation behavior via particle surface functionalization. The results were recently published in Nature Materials: N. James, E. Han, R. A. Lopez de la Cruz, J. Jureller, and H. M. **Jaeger**, Interparticle hydrogen bonding can elicit shear jamming in dense suspensions, Nature Materials 17, 965-970 (2018) [DOI: 10.1038/s41563-018-0175-5].

10.3 Research Accomplishments

Dry Granular Materials TFrictional interaction in dry granular materials provide an excellent energy dissipation mechanism. However, current attempts at developing unified granular drag laws vary widely in form and parameterization, and are frequently only applicable in a specific experimental context. We seek to understand the physical processes involved in impact into a granular material for a wide range of velocities and impact modalities. Our approach combines molecular dynamics simulations with constant-speed penetration experiments. Our detailed, fully three-dimensional simulations agree quantitatively with experimental results, probe grain-scale effects that are otherwise difficult to image, and reveal competing time- and depth-dependent effects that challenge existing models.

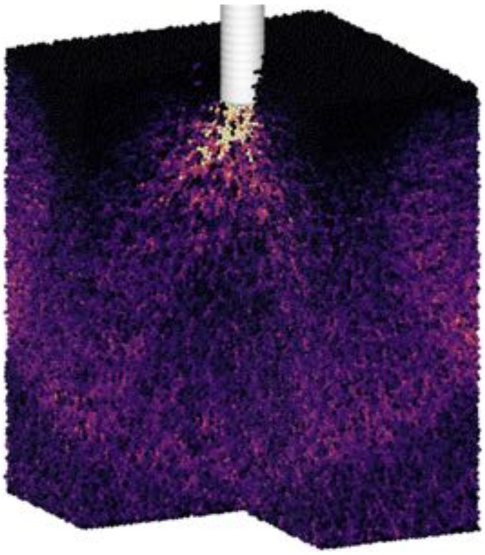


Figure 10.2: Snapshot from a three-dimensional molecular dynamics simulation of impact into a bed of granular material. Color indicates the magnitude of the local particle velocity.

During this reporting period, we identified a nonlinear stress peak during the early stages of impact, whose magnitude scales with impact speed and which is closely tied to the speed of sound in the granular aggregate. Following this peak the force on the projectile was found to increase linearly with penetration depth, while its magnitude is independent of velocity. The simulations showed that the initial peak is due to a local compression of grains upon first impact, which then propagates acoustically through the material (Figure 10.2). After the initial compression, the grains dilate to allow flow and produce a steady-state flow field around the intruder, yielding the velocity-independent linear force. We are currently characterizing the spatial extent and most important features of this flow region, which will provide insights into granular impact at high speeds and also indicate ways in which the impact characteristics of a granular material can be tuned.

Network-based Architeched Materials We investigated elastic networks as a means to direct, and potentially re-direct, stress propagation following an impact. To this end, auxetic materials are particularly promising. These materials are characterized by a negative Poisson ratio. As Poisson's ratio approaches the lower mechanical limit of -1 for isotropic materials, auxetic materials show enhanced resistance to impact and shear making them suitable for applications ranging from robotics to impact mitigation. However, past experimental efforts aimed at reaching the -1 limit have resulted in highly anisotropic materials, showing a negative Poisson's ratio only when subjected to deformations along specific directions. Isotropic auxetic designs have only attained modestly negative values of Poisson's ratio, or have led to solutions that cannot be manufactured in three dimensions.

We developed a design strategy to create *isotropic* auxetic structures from disordered networks (Figure 10.3), which allows for materials with Poisson's ratios as low as -0.98 and a significantly enhanced shear modulus. The materials conceived through this approach were successfully fabricated in laboratory experiments (see below) and confirmed to behave as predicted. Distinct trends in network structure and bond strengths were found to correlate with Poisson's ratio, shedding light on the motifs which lead to auxetic behavior. The ideas or concepts introduced here can be generalized to higher dimensions, wide ranges of material classes, and a wide spectrum of length scales, thereby providing a general platform that could impact technology.

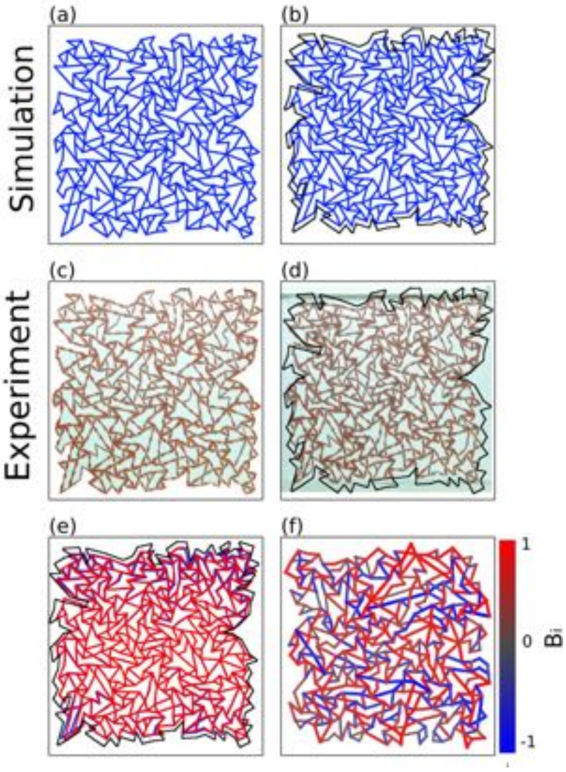


Figure 10.3: Isotropic auxetic networks in simulation and experiment. (a) Uncompressed simulation of auxetic network predicted to show a Poisson's ratio of approximately -1. (b) Simulated network uniaxially strained to 3.6% with outline of uncompressed network shown in black. (c) Uncompressed experimental realization of identical network. (d) Compressed experimental network at strain 3.6% with outline of uncompressed network shown in black. (e) Overlay of experimental (red) and simulated (red) networks at strain 3.6%. (f) Bond-level contributions to bulk modulus B in network under uniform compression, as determined by computing the virial coefficient contributions of each bond. Contributions are normalized by the largest value of the individual values B_i. [manuscript in preparation: Formation of Isotropic Auxetic Networks using Materials Optimization Strategies, DR Reid, N Pashine, AS Bowen, AJ Liu, SR **Nagel**, JJ **de Pablo**]

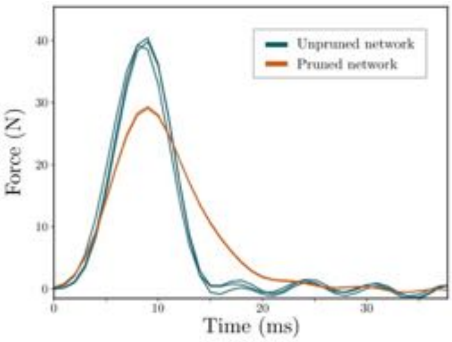


Figure 10.4: Comparison of transmitted impact force through unpruned and pruned disordered network structures, 3D-printed from elastic material. The pruned network exhibited auxetic behavior.

For testing the force transmission through auxetic structures, we fabricated networks by 3D-printing and performed impact tests. This effort was led by **Chan** (NIST), who spent time at University of Chicago during the summer of 2018. We measured the stress-strain responses of the networks while impacting them with constant speed or fixed initial momentum. We tested two types of networks: pruned (auxetic) and unpruned (ordinary elastic) (Figure 10.4). The unpruned networks were used as the control, and the pruned networks were generated from the unpruned networks by removing certain bounds so that the Poisson's ratio is lowered, as was confirmed by the uniaxial compression tests. Previous experimental and numerical work focused on quasi-static deformations of such networks, while we investigated the dynamic load response. Importantly, we found that pruned, auxetic networks generate a more desirable load response, by significantly lowering the peak transmitted force and spreading out the time over which this force acts.

10.4 Impact Statement for CHiMaD Phase I

This Seed Group initiated and implemented a concerted effort to explore new approaches for next-generation impact mitigating materials, focusing on two particularly promising platforms: particle-fluid hybrid systems and architectured network-based structures. In concentrated particle suspensions we discovered how impact can trigger a fluid-to-solid transformation that is reversible, proceeds via rapidly propagating shear jamming fronts, and can be tuned via the particles' surface chemistry. Investigating architectured network-based structures we introduced a new class of disordered three-dimensional networks that can be designed to exhibit auxetic load response. In contrast to regular auxetic lattices, which are fixed in their response and typically are auxetic along certain loading directions only, these networks are tunable and isotropic.

10.5 Collaborations

10.5.1 NIST Collaborations

Development of Novel Metrologies

*Aaron **Forster** (NIST), Edwin **Chan** (NIST), Heinrich **Jaeger** (UC)*

This collaboration focuses on complementing NIST and CHiMaD efforts in developing metrologies for multiaxial dynamic loading coupled with distributed force measurement, and volumetric exterior strain measurements. As a part of this mission, Edwin **Chan** (NIST) spent 6 weeks at the **Jaeger** laboratory at University of Chicago during the summer of 2018.

10.6 Publications and Presentations

5 Number of CHiMaD-supported publications in 2018 by Impact Mitigation seed group. Please see chapter 27 for details. [27.16, 27.17, 27.18, 27.19, 27.20]

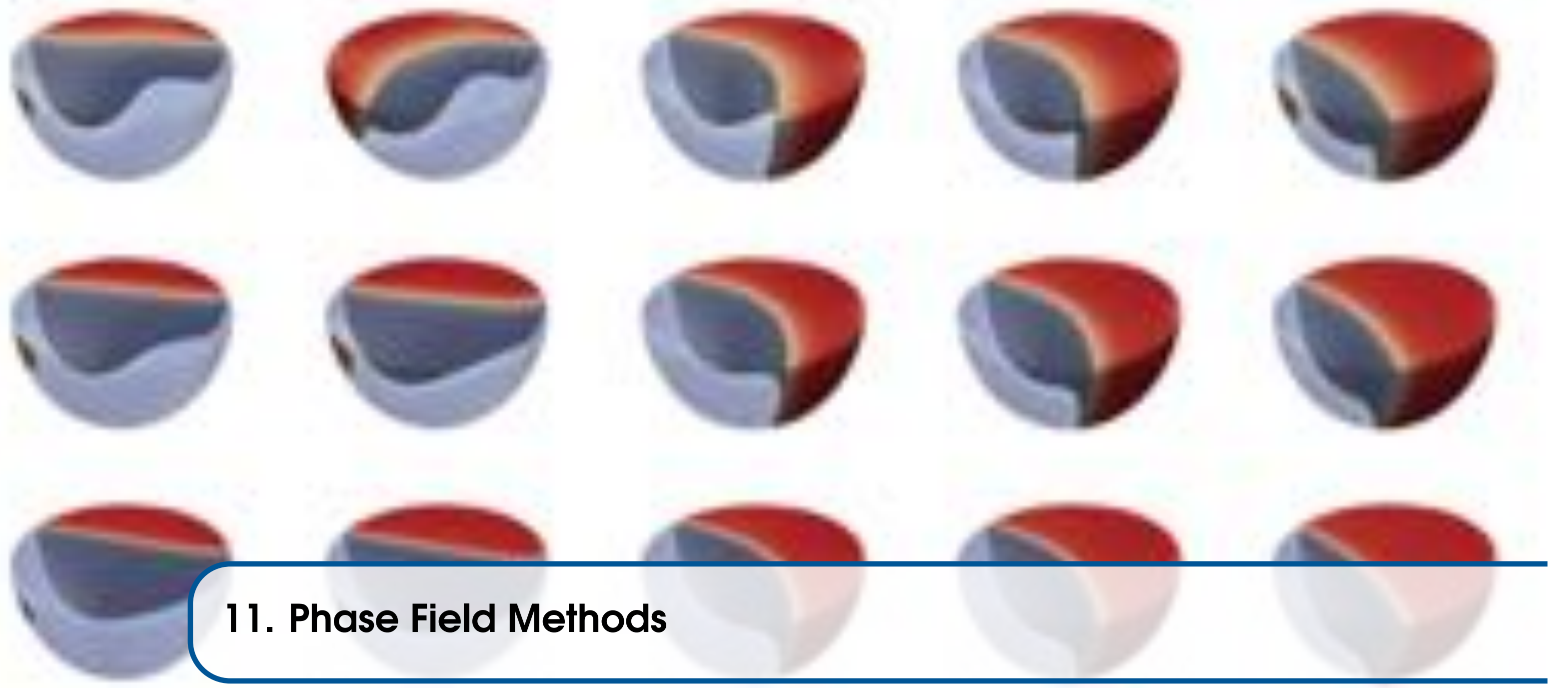
1 Number of presentations on CHiMaD-supported research given in 2018 by Impact Mitigation seed members, please see section 26.9 for the complete list.

10.7 CHiMaD Team

Impact Mitigation Seed Group Researchers			
Name	**Position**	**Affiliation**	**Project** (PI)
Juan de Pablo	Principal Investigator	UC/IME	
Sidney Nagel	Principal Investigator	UC/PHY	
Heinrich Jaeger	Principal Investigator	UC/PHY	
Nidhi Pashine	Graduate Student	UC/PHY	Designing allostery-inspired response in mechanical networks *(Nagel)*
Daniel Reed	Graduate Student	UC/IME	Designing user-specified mechanical responses in network based materials *(de Pablo)*
Leah Roth	Graduate Student	UC/PHY	Impact mitigation using granular materials *(Jaeger)*

IV

CHiMaD Tools & Facilities



11. Phase Field Methods

Olle Heinonen (NU), Peter Voorhees (NU), David Chopp (NU), Barry Smith (ANL)

James Warren (NIST), Jonathan Guyer (NIST)

Significance

The Phase Field Methods Tool Group is aimed at developing quantitatively predictive computational phase field methods, including phase field crystal models, and to develop benchmark problems for phase field modeling against which developers can test and benchmark codes. This latter goal also involves building and encouraging sustained involvement from the phase field community, especially code developers but also graduate students and post-doctoral researchers, by organizing and running workshops on phase field methods and setting up web pages for benchmark problems and community involvement and feedback. The benchmark problems also serve as an educational vehicle in graduate courses.

11.1 Goals

The goal of this tool group is broadly to develop quantitatively predictive phase field models and efficient algorithms, and to apply them to a range of materials problems, such as grain growth, solidification, and coarsening

11.2 Significant Accomplishments in 2018

Model development. A parallel implementation of a locally adaptive time stepping (LATS) method for the Cahn-Hilliard equation has been tested and shown to be unconditionally stable, and the results verified against other methods. Some minor development issues remain, and the implementation is expected to be completed in the spring of 2019.

CHiMaD Phase Field Methods Workshops. The tool group lead the organization of two CHiMaD Phase Field Methods Workshops in February and September of 2018, making it 7 workshops organized since the inception of the series in 2015. The first workshop focused on Benchmark problems in Method of Manufactured Solutions, led by Steven DeWitt (University of Michigan), and in coupled Cahn-Hilliard-electrostatic problem, led by Andrea Jokisaari (Idaho National Laboratory). The workshop also featured other presentations on GPU-based Phase Field modeling and Uncertainty Quantification, the latter led by the respective CHiMaD seed group. The second workshop focused on nucleation modeling in Phase Field, and featured lectures by Tamas Pusztai and Laszlo Granasy (Wigner Research Centre for Physics, Hungary) on the subject, as well as presentations by Steven DeWitt, Andrea Jokisaari, and Larry Aagesen (Idaho National Laboratory). At the workshop, revisions to the coupled Cahn-Hilliard-electrostatic Benchmark problem were also discussed, and participants presented solutions to a version sent out in advance to the workshop.

There is ongoing work to plan the upcoming workshop to be held in April of 2019 which will focus on nucleation.

PFHub: CHiMaD Phase Field Website. Significant work was also done by (Wheeler) on upgrading the CHiMaD Phase Field Methods website. Modifications were aimed at simplifying uploads and use as well as moving its hosting site. Currently the website can be accessed at `https://pages.nist.gov/pfhub/`

11.3 Research Accomplishments in 2018

Phase Field Benchmark Problems. A second paper on Phase Field Benchmark Problems was published [27.21], consisting of two problems: (a) dendritic growth in two dimensions, (b) coupling the Cahn-Hilliard equation to elastic degrees of freedom by examining a precipitate in a matrix. The dendritic growth problem highlights that a numerical solution is rather sensitive to the time-stepper used, while the coupled Cahn-Hilliard-elastic problem exploits phenomena of elastic instability and bifurcations that may occur depending on materials properties, initial conditions, and initial precipitate size.

A third set of Benchmark Problems has been designed and tested, and a manuscript is under preparation. This third set focuses the following problems: (a) two-dimensional Stokes flow in a channel with constriction, (b) a coupled Cahn-Hilliard-electrostatic (Poisson) problem that is a simple model of electrochemical systems. The Stokes problem highlights the need to handle boundary conditions at the in-flow and out-flow boundaries, as well as meshing at the constriction. In the Cahn-Hilliard-Poisson problem, issues with how to couple a solver for the long-range electrostatic fields generated by the diffusing charge density to the Cahn-Hilliard solver are explored.

Locally Adaptive Time Stepping (LATS) Method for Cahn-Hilliard Equation. The parallel Locally Adaptive Time Stepping (LATS) method is nearing completion, and is expected to be completed by Spring 2019. The temporal adaptivity is working as anticipated and is illustrated in Figure 11.1. The method remains unconditionally stable and the computed results between the parallel distributed algorithm and a standard serial implementation match very well. The only remaining issue for completion is to deal with situations where an interface passes through a corner formed by multiple subdomains. If the neighboring subdomain where an interface will pass into is using a large time step, it occasionally does not anticipate the necessary time step reduction when the interface crosses through the junction diagonally. Multiple solutions are being explored to resolve this issue.

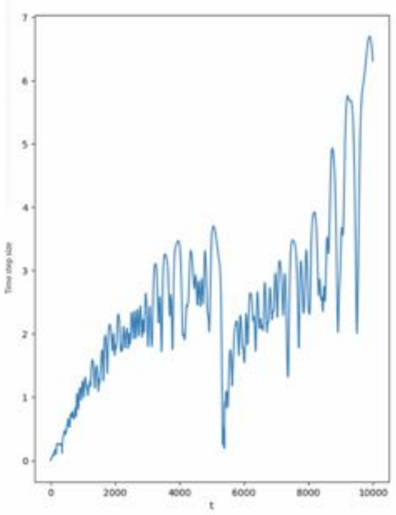


Figure 11.1: Representative time step sizes for a subdomain using the LATS method. The adaptive time steps grow as the two phases in the Cahn-Hilliard equation coalesce and become more regular. Time steps will drop during significant topological changes and then recover.

Fully Time-Dependent Coarsening Solver. The numerical method has been fully implemented in both 2D and 3D, and is currently in the testing phase. Testing is incomplete because the original test problem appears to not be well-posed, i.e. it does not have a unique solution. Without that, obtaining numerical convergence will not be possible. Parameter selections are being examined, in particular the chemical potentials chosen at the interface, to find a well-posed test problem that can also be solved analytically in order to better test the validity of the code.

Phase Field Crystal Method for Highly Anisotropic Interfaces Highly anisotropic interfaces play an important role in the development of material microstructure. Using the diffusive atomistic phase-field crystal (PFC) formalism, we determine the capability of the model to quantitatively describe these interfaces [27.24]. Specifically, we coarse grain the PFC model to attain both its complex amplitude formulation and its corresponding phase-field limit. Using this latter formulation, in one-dimensional calculations, we determine the surface energy and the properties of the Wulff shape. We find that the model can yield Wulff shapes with missing orientations, the transition to missing orientations, and facet formation. We show that the corresponding phase-field limit of the complex amplitude model yields a self-consistent description of highly anisotropic surface properties that are a function of the surface orientation with respect to the underlying crystal lattice. The phase-field model is also capable of describing missing orientations on equilibrium shapes of crystals and naturally includes a regularizing contribution. We demonstrate, in two dimensions, how the resultant model can be used to study growth of crystals with varying degrees of anisotropy in the phase-field limit, see Figure 11.2

Phase Field Modeling of Multicomponent Eutectic Solidification We applied our model for multicomponent eutectic solidification to multicomponent Al alloys [27.26]. The microstructure developed by two-phase lamellar eutectics $\alpha - \theta'$ Al-Cu and Al-Cu-Ag alloys is analyzed. A model of two-phase eutectic growth in multicomponent alloys is used to determine the scaling law of the eutectic microstructure using the alloy thermophysical properties. The application of the model to these alloys shows that the addition of Ag to Al-Cu alloys does not significantly change the length scale of the microstructure, which agrees with previous experimental studies. This is explained by the combined phenomena of the decrease in interface energies with the addition of Ag and the superheating of the $\alpha-$phase interface induced by the Ag composition profile. The theory can be used to

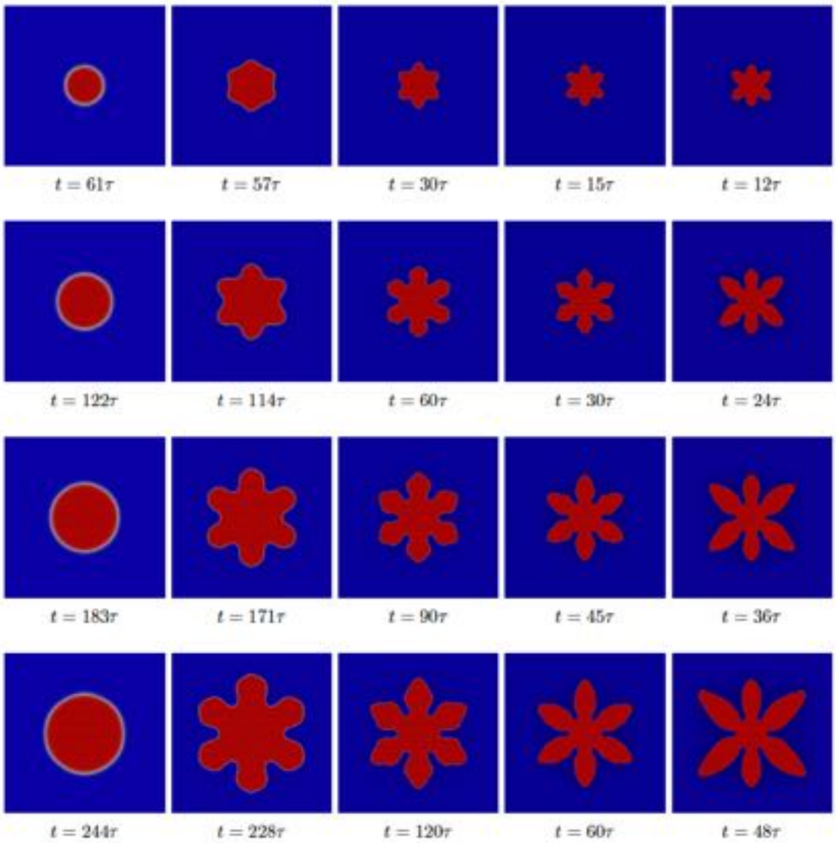


Figure 11.2: Dendritic solidification of the effective phase-field model demonstrating effects of highly anisotropic surfaces as function of temperature. Each column of images represents a different temperature, which decreases from left to right. Time increases from top to bottom, where we have attempted to match time according to similar solid fractions, terminating at an approximate solid fraction of 0.12. Red represents the solid phase, while blue is zero representing the liquid phase and black the density peaks. Evident is the early onset of the dendritic morphology for decreasing temperature, we also observe that due to increasing anisotropy, the form and shape of the dendrites also change. Specifically, we note that the primary arms become finer, almost needlelike, with decreasing temperature, and the interfaces become smoother eliminating fluctuations that would eventually cause the emergence of side branching.

design alloys to yield a given scale of the eutectic microstructure.

Machine Learning and 3D Atom Probe Tomography. In collaboration with researchers at Argonne National Laboratory as well as with **Dunand** and **Seidman** from Precipitation-Strengthened Alloys Use-Case Group and with partial funding from CHiMaD a machine-learning based method was developed to extract interfaces from 3D Atom Probe Tomography data and to do quantitative analysis of interfacial properties (eg thickness and composition); a manuscript is under preparation.

11.4 Impact Statement for CHiMaD Phase I

The work on Phase Field Benchmark Problems is now starting to gain attraction and citations, and achieving the goals set out by the tool group in testing codes and training practitioners. The Benchmark Problems are now used in teaching graduate students at, eg, the University of Florida and at the Indian Institute of Science in Bangalore, India.

11.5 Collaborations

11.5.1 Internal Collaborations

Multi-Scale Modeling of Co-Al-W Alloys

*Olle **Heinonen** (ANL), Peter **Voorhees** (NU), David **Dunand** (NU), David **Seidman** (NU), Chris **Wolverton** (NU), Barry **Smith** (ANL)*

This collaboration focuses on complementing the efforts of the Precipitation Strengthened Alloys use-case group and utilizes phase field methods to determine the morphology of precipitates in Co alloys; a computational model for the equilibrium shape of γ' inclusions in a γ-phase matrix of Co-W-Al alloys. **Dunand** (NU), **Seidman** (NU), **Wolverton** (NU) groups provided the inputs such as interface structure, compositions, interfacial energies for the modeling lead by **Voorhees** (NU) and **Heinonen** (ANL) groups while **Smith** (ANL) was consulted for numerical and applied math issues that arose during the modeling.

Locally Adaptive Time Stepping (LATS) method

*David **Chopp** (NU), Peter **Voorhees** (NU)*

This collaboration aims to implement a parallel Locally Adaptive Time Stepping (LATS) Method for the Cahn-Hilliard equation using test cases. **Voorhees** (NU) group provided the **Chopp** (NU) group problems for validating their completed LATS algorithm.

Fully Time-Dependent Coarsening Solver

*David **Chopp** (NU), Peter **Voorhees** (NU)*

This collaboration aims to implement a fully time-dependent coarsening solver using test cases.

Modeling of Additive Manufacturing

*Peter **Voorhees**, Stephen Davis (NU)*

The goal of this work is to model the evolution of solid-liquid interfacial morphology under rapid solidification and fluid flow conditions relevant to additive manufacturing.

CALPHAD Modeling of Thermoelectrics

*Jeff Snyder (NU), Peter **Voorhees**, Gregory **Olson** (NU)*

This project is focused on modeling the thermodynamics and phase transformation kinetics in Pb-based chalcogenides.

11.5.2 NIST Collaborations

Phase Field Benchmark Problems

*Olle **Heinonen** (ANL), Peter **Voorhees** (NU), James **Warren** (NIST), Jonathan **Guyer** (NIST), Daniel **Wheeler** (NIST), Trevor **Keller** (NIST)*

This collaboration aims to develop phase field benchmark problems for the global phase field community. To date, four CHiMaD Phase field workshops have been organized to engage the community for their input in developing the benchmark problems. A website has been developed by Wheeler (NIST) as a part of this collaboration *(https://pages.nist.gov/chimad-phase-field/#benchmarks)*. The latest benchmark problems were published on Journal of Computational Materials Science [27.21].

11.6 Technology Transfer

Community Resource
CHiMaD Phase Field Website
D. Wheeler (NIST), Olle ***Heinonen*** *(ANL), Peter* ***Voorhees*** *(NU), James* ***Warren*** *(NIST), Jonathan* ***Guyer*** *(NIST)*
Link: `https://pages.nist.gov/pfhub/`

CHiMaD Phase Field Website is a resource for the phase field community to compare and contrast phase field codes and libraries with an aim to improve community code collaboration. The first set of benchmark problems for phase field methods, which grew out of the CHiMaD Phase Field workshops I-III and constructed with feedback from the workshops was published in *Computational Materials Science* as an Editor's Choice publication. The second set of benchmark problems for phase field methods were published in *Computational Materials Science* in 2018 [27.21]. The benchmark problems, together with one set of solutions, are also posted on the CHiMaD Phase Field Website. It is expected that these benchmark problems will be a significant resource for developers of phase field codes - the problems are already being used in college courses taught by CHiMaD Phase Field Methods workshop contributors such as by Mike Tonks (Florida State Universiy) and Abhik Choudhary (Indian Institute of Science, Bangalore, India). The website is also being used at Chemomechanics Summer School (Maria-Laach, Germany) and Global Initiative of Academic Networks (Gian) Lecture Series (IIT Bombay, India). A third set of benchmark problems are in preparation and will be tested during the workshops organized in 2019.

Community Resource
Global Initiative of Academic Networks (Gian) Lecture Series
Peter ***Voorhees*** *(NU)*
Link: `https://www.youtube.com/watch?v=3Nt5hS8S2qY`

Peter **Voorhees** was invited to Indian Institute of Technology (IIT) Bombay to deliver a Gian lecture series - an initiative by Government of India - on Phase Field Methods. The lecture series, which included a discussion of CHiMaD's Phase Field community resources, are posted publicly on YouTube. The link to the first lecture is provided above.

Data
Datasets Uploaded to Materials Data Facility in 2018

1. Phase field modeling data for published Benchmarks Problems 3 and 4
   A.M. Jokisaari (INL), P. W. ***Voorhees****, J.E.* ***Guyer****, J.A.* ***Warren****, and O.G.* ***Heinonen***
   DOI: 10.18126/M2QS6Z

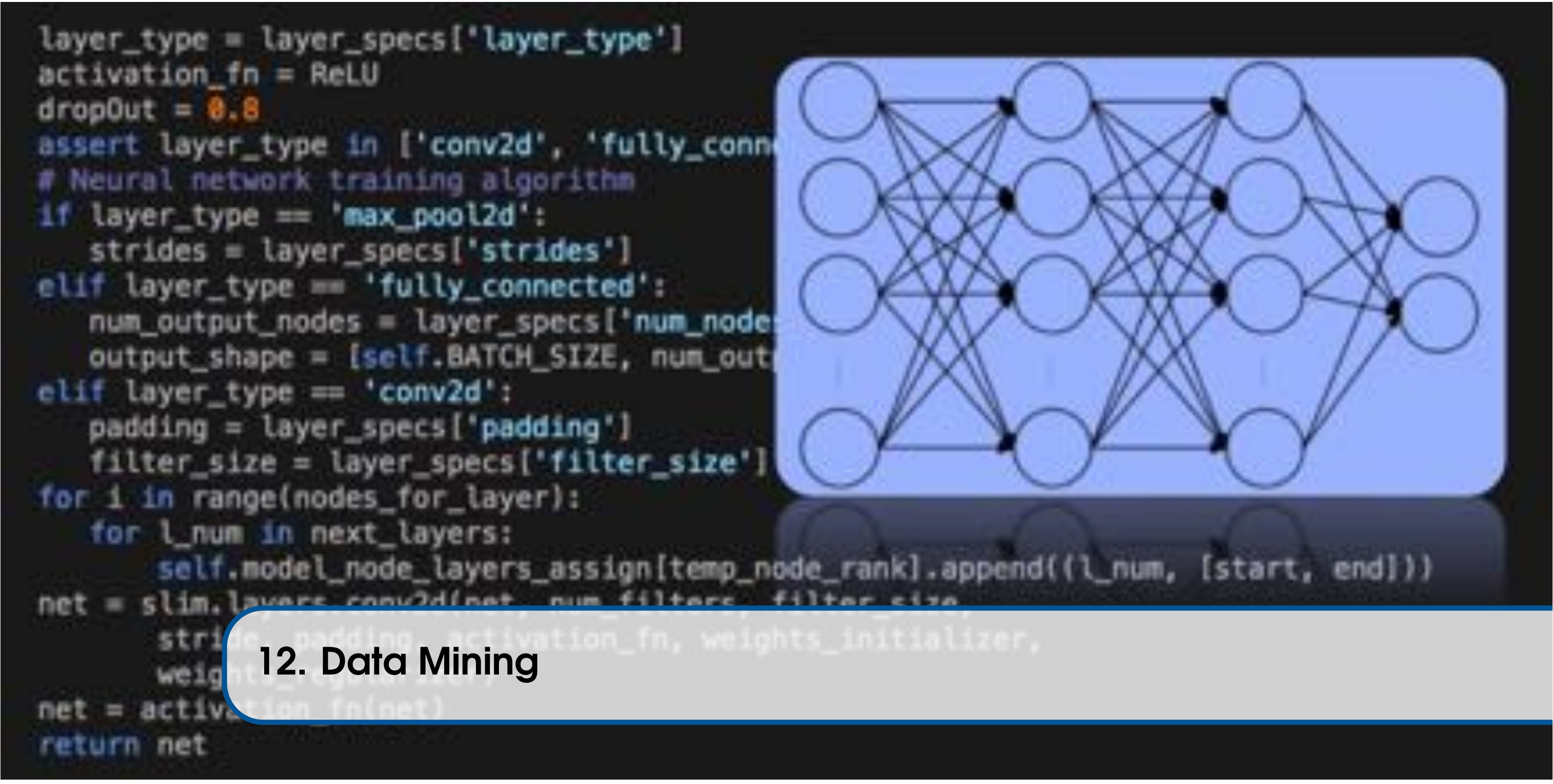
11.7 Publications and Presentations

8 Number of CHiMaD-supported publications in 2018 by Phase Field Methods seed group. Please see chapter 27 for details [27.21, 27.22, 27.23, 27.24, 27.25, 27.26, 27.27, 27.28].

10 Number of presentations on CHiMaD supported research in 2018, please see section 26.10 for the complete list.

11.8 CHiMaD Team

Phase Field Methods Group Researchers			
Name	**Position**	**Affiliation**	**Project (PI)**
Olle Heinonen Peter Voorhees David Chopp Barry Smith	Principal Investigator Principal Investigator Principal Investigator Principal Investigator	ANL/NAISE NU/MSE NU/ESAM ANL/NAISE	
Nana Ofori-Opoku	Postdoctoral Researcher	NU-NIST	Phase field crystal *(Voorhees, Warren)*
Wenkun Wu	Postdoctoral Researcher	ANL/MCS	Phase field benchmark problems, Modeling of microstructural evolution of Co-based super alloys *(Heinonen)*
Tiberiu Stan	Postdoctoral Researcher	NU/MSE	Phase field methods *(Voorhees)*
Matthew Peters	Graduate Student	NU/MSE	CALPHAD Methods for Thermoelectrics *(Voorhees)*
Narut Sereewattanawoot	Graduate Student	NU/ESAM	PLATS Methodology *(Chopp)*



```
layer_type = layer_specs['layer_type']
activation_fn = ReLU
dropOut = 0.8
assert layer_type in ['conv2d', 'fully_conn
# Neural network training algorithm
if layer_type == 'max_pool2d':
   strides = layer_specs['strides']
elif layer_type == 'fully_connected':
   num_output_nodes = layer_specs['num_node
   output_shape = [self.BATCH_SIZE, num_out
elif layer_type == 'conv2d':
   padding = layer_specs['padding']
   filter_size = layer_specs['filter_size']
for i in range(nodes_for_layer):
   for l_num in next_layers:
       self.model_node_layers_assign[temp_node_rank].append((l_num, [start, end]))
net = slim.layers.conv2d(net, num_filters, filter_size,
       stride, padding, activation_fn, weights_initializer,
       weig
net = activation_fn(net)
return net
```

12. Data Mining

Alok Choudhary (NU), Ankit Agrawal (NU), Wei Chen (NU), Chris Wolverton (NU)

Carelyn Campbell (NIST), Kamal Choudhary (NIST), Brian DeCost (NIST), Martin Green (NIST), Jason Hattrick-Simpers (NIST), Gilad Kusne (NIST), Shengyen Li (NIST), Nam Nguyen (NIST), Andrew Reid (NIST), Francesca Tavazza (NIST)

Significance

The CHiMaD data mining team is working towards making significant advances in data-driven informatics to accelerate materials discovery. In this era of big data, we are being bombarded with huge volumes of data from a variety of different sources (experiments, simulations) at unprecedented rates in practically all fields of science and engineering, and materials science is no exception. Advanced techniques for data-driven analytics are needed to analyze all this data in ways that can help extract meaningful information and knowledge from it, and thus contribute to accelerating materials discovery and the vision of Materials Genome Initiative. The diversity and expertise within the CHiMaD team provides unique opportunities for collaborations among materials scientists and computer scientists to realize this goal. This requires designing and developing specific analytics and data mining approaches for problems in materials science, and evaluating the resulting insights/discoveries via necessary experiments and simulations. The long-term vision is to work with NIST to enable data-to-discovery (or actionable insights) at unprecedented latency through summary statistics, query, reporting, analysis, bottom-up as well as hypothesis-driven discoveries and data mining on various heterogeneous and big databases that are complex, high dimensional, structured and semi-structured.

12.1 Design Goals

The design goals of the Data Mining use-case group for this reporting period were four-fold:

- Continue to make progress in the ongoing collaborative materials informatics projects from prior years

- Design and develop software deploying materials informatics for the materials community to use
- Make efforts towards validating the predictions from our data mining models via experiments
- Explore new projects and collaborations to solve important problems in materials science that can benefit from data-driven informatics

12.2 Significant Accomplishments in 2018

Following are the significant accomplishments of the data mining team in the past year:

- ElemNet - a deep neural network for learning materials properties using only raw elemental compositions as input has been developed; results were published in Scientific Reports. In addition, an ElemNet-based deep transfer learning approach was developed to build predictive models on JARVIS, Materials Project, and experimental data in collaboration with NIST.
- A deep adversarial learning methodology for microstructure design has been developed; results were published in Journal of Mechanical Design.
- Advanced predictive models for steel fatigue strength prediction has been developed; results were published in International Journal of Fatigue.
- CheMixNet - a set of neural networks for predicting chemical properties using both SMILES and molecular fingerprints as input has been developed; results were accepted at NIPS MLMM 2018.
- A CHiMaD-NIST-SLAC collaboration has demonstrated that the iterative machine learning guided high throughput experiments can accelerate discovery of metallic glasses; results were published in Science Advances.
- A framework was established for designing bulk metallic glasses in collaboration with Liquidmetal Technologies; results were published in Acta Materialia.
- A recurrent neural network for predicting thermal history in Direct Energy Deposition processes has been developed in a cross-use-case collaboration with Additive Manufacturing seed group; results were published in Manufacturing Letters.
- A Bayesian optimization framework for data-driven materials design has been developed.
- A deep transfer learning based predictive model for crack detection in infrastructure images has been developed.
- An approach for integrating domain knowledge into deep learning models so that it could be applied even for small datasets has been established through a CHiMaD-NIST-WVU collaboration including **Reid** from NIST.
- Design principles for building deep regression networks with numerical vectors as input has been developed, and applied to materials datasets with composition- and structure-based inputs.
- Convolution neural networks for extracting and representing structure information of inorganic compounds for machine learning were developed.
- Predictive models for martensite start temperature were developed and deployed in collaboration with Precipitation-strengthened Alloys group
- As part of a CHiMaD-NIST collaboration, predictive models for work-to-necking of Ni-superalloys were developed and deployed.
- A deep learning approach was developed to directly analyze warped XRD images for phase identification, as a part of a CHiMaD-NIST collaboration.

- A fuzzy clustering methodology for phase clustering experimental isothermal ternary phase diagrams of Co-superalloys developed in collaboration with Precipitation-Strengthened Alloys group.

12.3 Research Accomplishments

ElemNet: Deep Learning the Chemistry of Materials from Only Elemental Composition Conventional machine learning approaches for predicting material properties from elemental compositions have emphasized the importance of leveraging domain knowledge when designing model inputs. A collaboration between **Agrawal, Choudhary, Wolverton** groups and Ward (UC) have lead to the development of a deep neural network for learning materials properties such as formation energy, using only raw elemental compositions as input (see Figure 12.1). This work has demonstrated that by using a deep learning approach, it is possible to bypass such manual feature engineering requiring domain knowledge and achieve much better results, even with only a few thousand training samples. The designed and implemented a deep neural network model, *ElemNet*, automatically captures the physical and chemical interactions and similarities between different elements using artificial intelligence which allows it to predict the materials properties with better accuracy and speed. The speed and best-in-class accuracy of *ElemNet* enabled us to perform a fast and robust screening for new material candidates in a huge combinatorial spa space; where we predicted hundreds of thousands of chemical systems that could contain yet-undiscovered compounds. *ElemNet* is available as part of our online formation energy predictor tool (available at http://info.eecs.northwestern.edu/FEpredictor; accessed more than 2000 times as of December 2018), and a manuscript based on this work has been published in Scientific Reports [27.31].

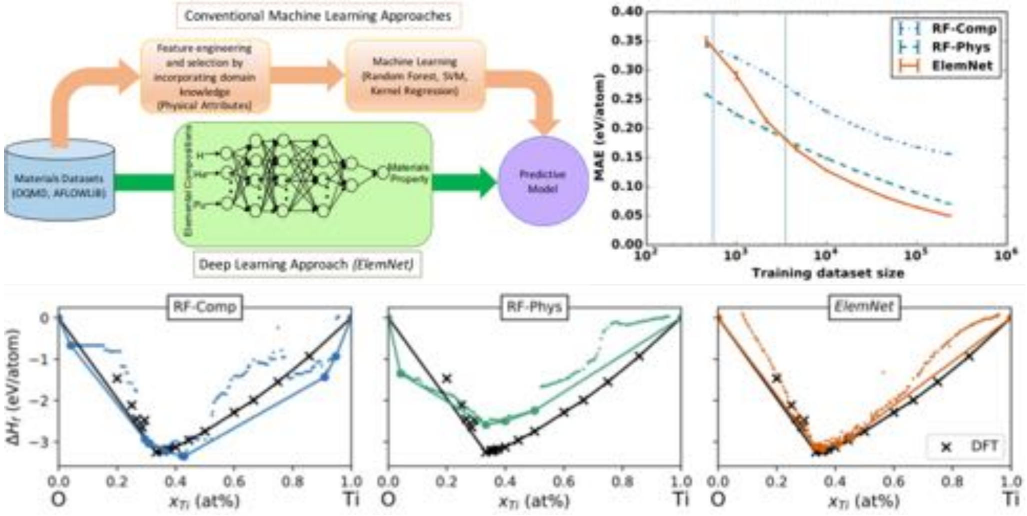


Figure 12.1: *ElemNet*: A deep learning approach to bypass manual feature engineering for predictive modeling of materials properties directly from raw elemental composition (top-left); Model accuracy with increasing training data (top-right); Predicted phase diagrams from a hold-out test of Ti-O binary system (bottom). RF-Comp and RF-Phys denote Random Forest model on raw compositions and composition-derived physical attributes respectively. The black lines in the bottom charts show the OQMD convex hull.

ElemNet-based Transfer Learning on JARVIS As an extension of the development of

ElemNet, a collaborative project between **Agrawal, Choudhary**, **Choudhary, DeCost, Tavazza, Campbell** investigated its deep-learning application to smaller datasets. Since *ElemNet* is trained on a relatively big database (*OQMD*) and has demonstrated to have self-learned some key chemistry knowledge such as element similarity and interaction, it is predicted to be a good starting point for building accurate models on other materials datasets which may not be large enough for direct application of deep learning. This collaboration demonstrated that the large DFT-computational datasets can be leveraged together with the existing experimental data using artificial intelligence to build significantly robust prediction models that are closer to the experimental observations using the concept of transfer learning. It was found that *ElemNet*-based transfer learning can yield up to 40% more accurate predictive models on *JARVIS, Materials Project*, and a small experimental database.

Microstructural Materials Design via Deep Adversarial Learning Methodology Identifying the key microstructure representations is crucial for Computational Materials Design (CMD). However, existing microstructure characterization and reconstruction (MCR) techniques have limitations to be applied for microstructural materials design. Some MCR approaches are not applicable for microstructural materials design because no parameters are available to serve as design variables, while others introduce significant information loss in either microstructure representation and/or dimensionality reduction. In collaboration **Agrawal, Choudhary, Chen**, and Brinson (Duke University) groups, have developed a deep adversarial learning methodology that overcomes the limitations of existing MCR techniques. In the proposed methodology (see Figure 12.2), generative adversarial networks (GAN) are trained to learn the mapping between latent variables and microstructures. Thereafter, the low-dimensional latent variables serve as design variables, and a Bayesian optimization framework is applied to obtain microstructures with desired material property. Due to the special design of the network architecture, the proposed methodology is able to identify the latent (design) variables with desired dimensionality, as well as capturing complex material microstructural characteristics. The validity of the proposed methodology is tested numerically on a synthetic microstructure dataset and its effectiveness for microstructural materials design is evaluated through a case study of optimizing optical performance for energy absorption. Additional features, such as scalability and transferability, are also demonstrated in this work. In essence, the proposed methodology provides an end-to-end solution for microstructural materials design, in which GAN reduces information loss and preserves more microstructural characteristics, and the GP-Hedge optimization improves the efficiency of design exploration. This work has been published in Journal of Mechanical Design.

Predicting Fatigue Strength of Steel Alloys Based on Ensemble Data Mining **Agrawal** and **Choudhary** groups have further built on their prior collaboration with the Precipitation-Stregthened Alloys use-case group on steel fatigue strength prediction by comparing 40 supervised modeling configurations on NIMS dataset, including ensemble modeling techniques, resulting in more accurate predictive models that are significantly better (statistically) than prior models (see Figure 12.3). Further, feature analysis helped identify processing and composition parameters most influential to fatigue strength. Although all processing parameters were highly correlated with fatigue strength, the most influential ones were found to be related to tempering, carburization, diffusion, through hardening, and normalization (in that order). The latest models are made available in our online steel fatigue strength predictor (available at http://info.eecs.northwestern.edu/SteelFatigueStrengthPredictor;

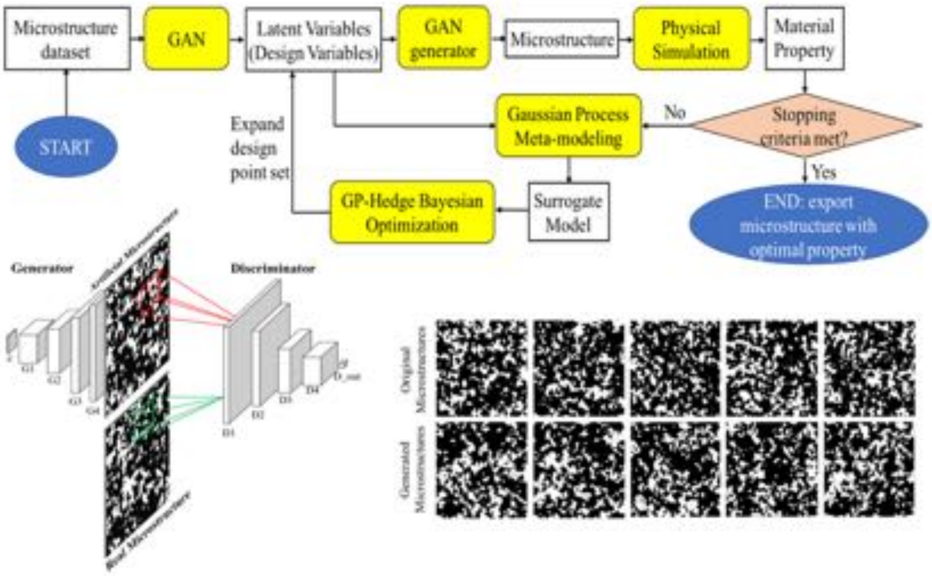


Figure 12.2: The flowchart of the proposed deep learning based microstructure design methodology (top); Generative adversarial network (GAN) for microstructure design (bottom-left); Examples of original (training) microstructures and microstructures produced by the generative adversarial networks (GAN) generator (bottom-right).

accessed more than 1500 times as of December 2018). This work has been published in International Journal of Fatigue [27.29].

CheMixNet: Predicting Chemical Properties using Multiple Molecular Representations
A collaboration between **Agrawal** and **Choudhary** groups lead to the development of *CheMixNet* - a set of neural networks for predicting chemical properties by using simplified molecular-input line-entry system (*SMILES*) notations as sequence inputs and molecular fingerprints as vector inputs (see Figure 12.4). *SMILES* is a linear representation of chemical structures which encodes the connection table, and the stereochemistry of a molecule as a line of text with a grammar structure denoting atoms, bonds, rings and chains, and this information can be used to predict chemical properties. Molecular fingerprints are representations of chemical structures, successfully used in similarity search, clustering, classification, drug discovery, and virtual screening and are a standard and computationally efficient abstract representation where structural features are represented as a bit string. Both *SMILES* and molecular fingerprints are different representations for describing the structure of a molecule, and there exist several predictive models for learning chemical properties using one of these representations. Our work has improved upon the existing state-of-the-art approach of directly learning from vector representations such as molecular fingerprints or chemical text representations such as *SMILES* by harnessing the network structure of both forms of representations. *CheMixNet* is a multi-input-single-output architecture that learns chemical properties from a mix of intermediate features learned from two different input representations - *SMILES* as sequences and molecular fingerprints as vector inputs. We have demonstrated the efficacy of *CheMixNet* architectures by evaluating them on six different datasets. The proposed *CheMixNet* models not only outperform the candidate neural architectures such as contemporary fully connected networks that use molecular fingerprints and 1-dimensional convolutional neural network (CNN) and

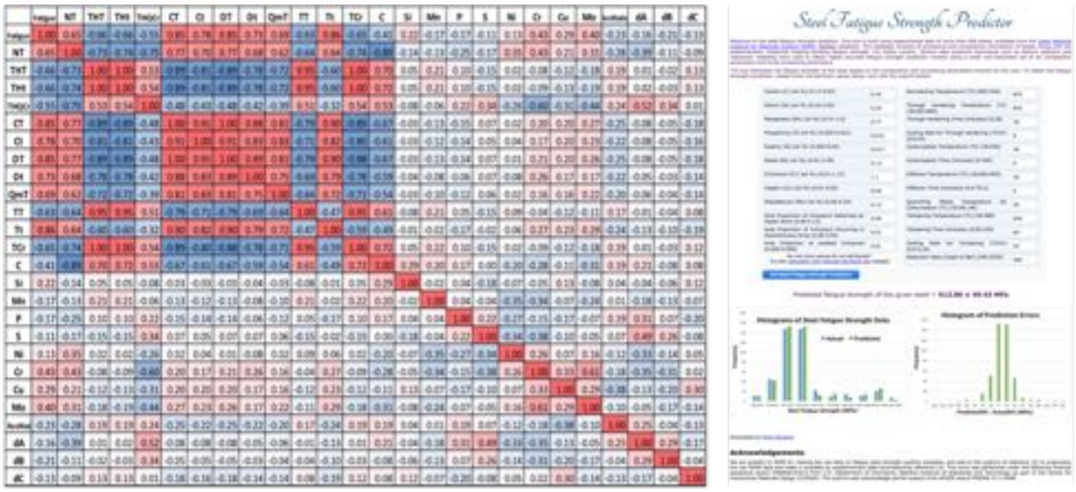


Figure 12.3: Intra-feature correlation heat map with positive and negative correlations in red and blue respectively (left); A screenshot of the deployed steel fatigue strength predictor (right).

recurrent neural networks (RNN) models trained on *SMILES* sequences, but also other state-of-the-art architectures such as Chemception and Molecular Graph Convolutions. This work was accepted and presented at the Neural Information Processing Systems (NIPS) 2018 workshop on Machine Learning for Molecules and Materials [27.32].

Accelerated Discovery of Metallic Glasses Through Iteration of Machine Learning and High-throughput Experiments With more than a hundred elements in the periodic table, a large number of potential new materials exist to address the technological and societal challenges we face today; however, without some guidance, searching through this vast combinatorial space is frustratingly slow and expensive, especially for materials strongly influenced by processing. A collaboration between **Wolverton**, **Hattrick-Simpers** and Mehta (SLAC) trained a machine learning (ML) model on previously reported observations, parameters from physiochemical theories, and make it synthesis method-dependent to guide high-throughput (HiTp) experiments to find a new system of metallic glasses in the Co-V-Zr ternary. Experimental observations are in good agreement with the predictions of the model, but there are quantitative discrepancies in the precise compositions predicted. These discrepancies were used to retrain the ML model. The refined model has significantly improved accuracy not only for the Co-V-Zr system but also across all other available validation data. The refined model was then used to guide the discovery of metallic glasses in two additional previously unreported ternaries. Although this approach of iterative use of ML and HiTp experiments (see Figure 12.5) has guided us to rapid discovery of three new glass-forming systems, it has also provided a quantitatively accurate, synthesis method-sensitive predictor for metallic glasses that improves performance with use and thus promises to greatly accelerate discovery of many new metallic glasses. This discovery paradigm is applicable to a wider range of materials and should prove equally powerful for other materials and properties that are synthesis path-dependent and that current physiochemical theories find challenging to predict. This work was published in Science Advances [27.63] and featured in Forbes ("Scientists Use Artificial Intelligence To Discover New Materials") and The Verge ("How AI is helping us discover materials faster than ever), the links to the latter can be found under the technology transfer section.

A Machine Learning Approach for Engineering Bulk Metallic Glass Alloys Bulk metallic

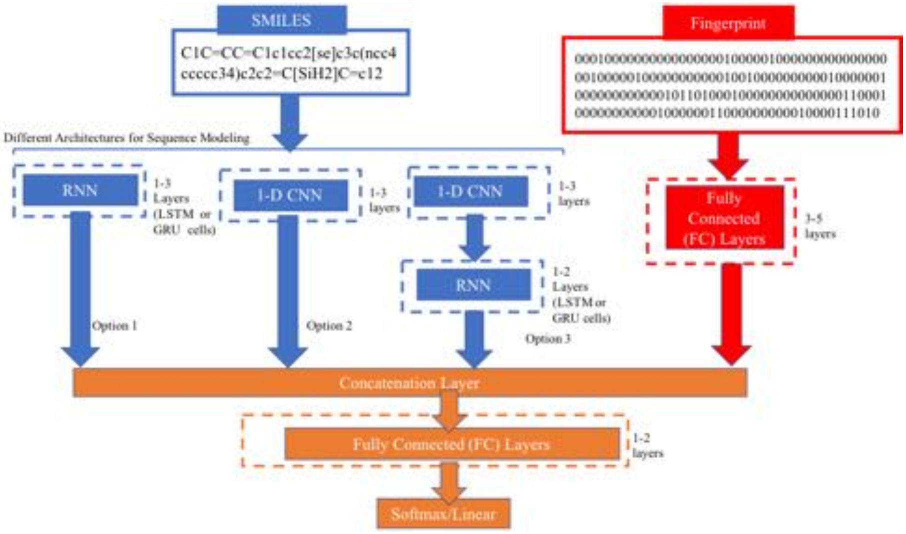


Figure 12.4: *CheMixNet*: A deep learning approach to learn from multiple molecular representations: SMILES and Fingerprints. The blue branch represents candidate neural network options - convolutional and/or recurrent networks for learning from *SMILES* sequences, and the red branch represents fully connected neural network for learning from fingerprints. Finally, fully connected layers learn the final output label from the concatenation of intermediate features (illustrated in orange) of the two branches.

glasses (BMGs) are a unique class of materials that are gaining traction in a wide variety of applications due to their attractive physical properties. One limitation to the wide-scale use of these materials is the lack of predictable tools for understanding the relationships between alloy composition and ideal properties. To address this issue, **Wolverton**, Ward (UC) and Liquidmetal Technologies, in collaboration, developed a framework for designing metallic glasses using machine learning (ML) models that predict three key properties of candidate BMG compositions: ability to exist in an amorphous state, critical casting diameter (Dmax), and supercooled liquid range (ΔTx). These models take only the composition of the alloy as input, and were created from a database of more than 8000 metallic glass experiments assembled from several dozen papers and handbooks. The collaboration employed these ML models to optimize the properties of existing commercial alloys and found, experimentally, several of the ML-predicted compositions can form glasses and exceed existing alloys in one of the two design variables, ΔTx (see Figure 12.6). This work was published in Acta Materialia [27.102].

Predicting Thermal History in Directed Energy Deposition Processes using Recurrent Neural Networks Directed Energy Deposition (DED) is a growing additive manufacturing technology due to its superior properties such as build flexibility at multiple scales and limited waste. However, both experimental and physics-based models have limitations in providing accurate and computationally efficient predictions of process outcomes, which is essential for real-time process control and optimization. The collaboration with Additive Manufacturing seed group (**Cao**) developed a recurrent neural network (RNN) structure with a Gated Recurrent Unit (GRU) formulation for predicting the high-dimensional thermal history in DED processes with variations in geometry, build dimensions, toolpath strategy, laser power and scan speed. The results indicate that the model can accurately predict the thermal history of any given point of the DED build on a test-set database with

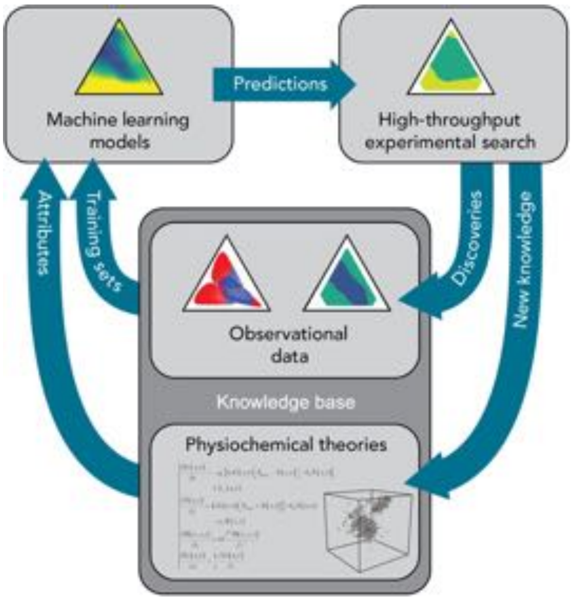


Figure 12.5: Schematic depiction of a paradigm for rapid and guided discovery of materials through iterative combination of machine learning with high-throughput experimentation.

limited training. The model's general applicability and ability to accurately predict thermal histories has been demonstrated through two overarching tests conducted for long time spans and non-trained geometries. This collaboration initiated as a Predictive Science and Engineering Design (PSED) cluster project at Northwestern. A manuscript based on this work was published in Manufacturing Letters [27.40].

Bayesian Optimization Framework for Data-Driven Materials Design Materials design and discovery have been formulated as optimization problems with the goal of achieving desired material properties or device performance. One challenge of solving material design problems using optimization is the co-existence of both qualitative and quantitative design variables associated with material compositions, microstructure morphology, and processing conditions. The **Chen** group has developed a Bayesian optimization-based materials design framework that utilizes existing materials databases and sequentially explores the unknown design space via Bayesian inference. To address the challenge of mixed types of design variables, we have developed a novel latent variable model for GP-based simulation response surface modeling with both quantitative and qualitative factors. The approach maps the qualitative factor levels to a corresponding set of 2D latent numerical variable values, so that distances in the latent variable space account for response correlations across levels of the qualitative factors. The developed approach is flexible enough to accurately capture complex correlations of many qualitative factors. In addition, the estimated latent variables provide insight into the relationship between the levels of a factor, e.g., how far apart the material performance with respect to the choice of the type of filler materials in nanocomposite design. By integrating the latent variable GP model into the Bayesian optimization framework, we overcome the difficulty of using BO for problems with qualitative input factors. As shown in Figure 12.7, a Bayesian Optimization framework for data-driven materials design proposed consists four major steps: (1) Step 1 (materials database) involves creating a materials database (both physical and computer data) based on the information gathered from literature and simulations (2) Step 2 (machine learning and Bayesian inference) fits a machine learning model using the available dataset and provides uncertainty quantification of model prediction based on the nature of data, (3) Step 3 ("on-demand" design exploration) makes inference about

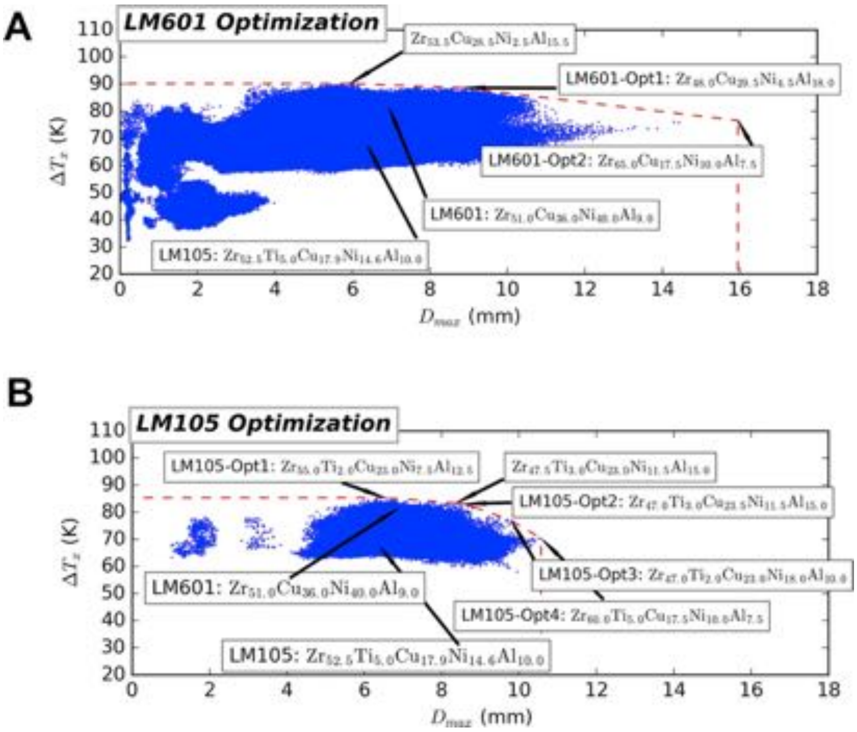


Figure 12.6: Machine-learning-predicted properties of alloys evaluated during the optimization of two established BMG alloys: LM601 and LM105. The red dashed line in each plot represents the Pareto surface of the predicted alloys, which was used to identify alloys with optimal levels of critical casting diameter (Dmax) and supercooled liquid range (ΔTx). The properties, names, and compositions of alloys tested in this work are labeled with arrows.

where to sample the next point (either physical or computer experiments) based on the uncertainty in the current fitted model and the design objective, and (4) Step 4 (augmenting dataset) evaluates the chosen design point(s) to augment the materials database and update the metamodel. As this procedure keeps iterating, more sample points will be sequentially added to improve the metamodel prediction and identify the global optimum solution. This approach has been successfully applied to optimizing light absorption of the quasi-random solar cell by designing the structure and selecting materials simultaneously and identifying the best combinatorial solution of the hybrid organic-inorganic perovskite.

Crack Detection in Infrastructure Images using VGG-based Transfer Learning In collaboration **Agrawal** and **Choudhary** groups have developed a simplified crack detection model from UAV images of civil infrastructure through the application of deep transfer learning approach, building on the transfer learning approach used last year for automated pavement crack detection. For transfer learning, a deep Convolutional Neural Network VGG-16 trained on the "big data" ImageNet database was used, which contains millions of images, and transfer that learning to automatically detect cracks in infrastructure images. Results were published in [27.36].

Learning Crystal Plasticity from Discrete Dislocation Dynamics using Deep Learning In collaboration, **Agrawal**, **Choudhary**, **Reid**, **Campbell** and Papanikolaou (West Virginia University) groups, developed an approach for integrating domain knowledge into deep learning models so that it could be effectively applied even for small datasets. More specifically, two-point correlation function, which is used to capture the spatial information

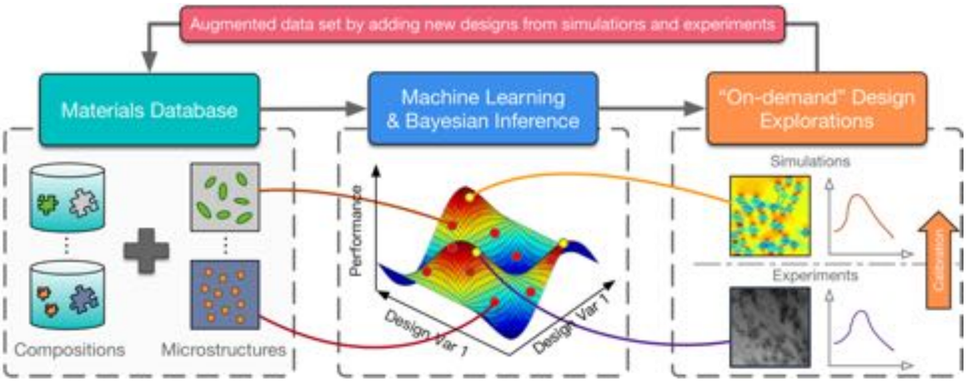


Figure 12.7: Bayesian optimization framework for data-driven materials design.

from data, is used as the domain knowledge to be integrated in the training of the proposed hybrid convolutional neural network. The efficacy of the proposed approach is evaluated through a case study where initial strain deformation level is predicted given resulting microstructural total strain profiles of samples. The analysis suggests that integrating domain knowledge into deep learning can not only improve the model's performance for small dataset, but also make the results more explainable based on domain knowledge.

Individual Residual Learning for Crystal Structure Aware Formation Enthalpy Prediction **Agrawal** and **Choudhary** groups have developed design principles for building deep regression networks composed of fully connected layers for data mining problems with numerical vectors as input. A novel deep regression network with individual residual learning was introduced where the proposed architecture leverages shortcut connections after each layer such that each layer learns the residual mapping between its output and input. As an illustrative example, the efficacy of the proposed networks was examined for learning properties of materials from numerical attributes derived from their composition and crystal structure, using the materials data in the Open Quantum Materials Database (OQMD) and the Materials Project. It was found that the use of individual residual learning leads to better convergence compared to the existing approach of using shortcut connections after stacking of multiple layers while maintaining the same number of parameters, and has the potential to reduce the model error by more than 40%.

Convolutional Neural Networks to Predict Properties of Inorganic Compounds Incorporating structural information in machine learning (ML) models of materials properties has been shown to improve the predictive accuracy of the resulting models. The **Wolverton** group has demonstrated a novel use of convolution neural networks (CNN) to extract information about the crystal structure of inorganic compounds. The CNN-extracted structural properties are then combined with composition-dependent elemental properties to form the material representation for the ML model. This method is illustrated using datasets consisting of high-throughput density functional theory (DFT) data of formation energies of compounds. The accuracy of different representations of the compound crystal structure are critically examined as input for the CNN. The ML models are trained on $\sim 200,000$ entries taken from the Open Quantum Materials Database and evaluate the predictive accuracy on a test set of $20,000$ compounds. The predictive accuracy of our CNN model for formation energies is compared with other recently-proposed structural representations, e.g., those based on Voronoi tessellation.

Martensite Start Temperature Prediction In continuation of the collaborative steel data mining efforts with Precipitation-Strengthened Alloys group from prior years **Agrawal**, **Choudhary**, and **Olson**, have developed accurate decision tree based models for martensite start (MsT) temperatures using data of more that 800 steel alloys. The model consists of a decision tree with different linear models at leaf nodes. Compared to prior composition-based models for MsT prediction ($R2 < 0.7$), the developed models are highly accurate ($R2 > 0.9$). The MsT predictive model is deployed online and is available at http://info.eecs.northwestern.edu/MsTpredictor.

Ni Super Alloys Property Prediction In continuation of the collaborative efforts from prior years, **Agrawal**, **Choudhary**, **Campbell** and **Li** have developed accurate data mining models for predicting work-to-necking of a given Ni-Al-Cr superalloy, using data of more than 200 Ni-Al-Cr superalloys simulations done at NIST. Current analysis has resulted in accurate models for predicting work-to-necking with $R2 > 0.85$ and MAE $< 1.5MPa$, and the corresponding predictive model is deployed online and is available at http://info.eecs.northwestern.edu/NiSuperalloy.

Mining X-ray Diffraction Images for Phase Identification using Convolution Neural Networks X-ray diffraction (XRD) is a widely used technique to determine the atomic-scale structures to understand the composition-structure-property relationship of materials. The current approach for the analysis of XRD patterns involves conversion to 1D (intensity-2Θ) patterns; this leads to the loss in the peak characteristics present in the warped (2D) XRD pattern. The collaboration between **Agrawal**, **Choudhary**, **Kusne** and **Nguyen** aims to eliminate such loss of information by directly leveraging the warped 2D XRD patterns through a deep learning solution. The developed CNN model takes the warped XRD patterns as inputs and predicts the phase region labels. The primary challenge is the presence of irregular background of high intensity. To remove the background, some novel methods based on convolution are devised and a peak area detection network based on slope filters is introduced. The proposed methods are evaluated using two sets of XRD patterns collected from SLAC and Bruker D-8 for the SnTiZn composition space; each set contains 177 experimental XRD patterns. It is found that background removal techniques can boost the classification performance.

Phase Clustering in Co Super Alloys using Fuzzy Clustering In a new collaboration with the Precipitation-Strengthened Alloys group, **Agrawal**, **Choudhary**, **Bedzyk**, and **Chung** characterized high-throughput combinatorial thin-film libraries of X-ray diffraction (XRD) and X-ray fluorescence (XRF) using a customized set up at the Advanced Photon Source (APS), DND-CAT 5BM-C. A methodology was developed for phase clustering experimental isothermal ternary phase diagrams of Co-super alloys using materials independent methods to cluster the XRD data generated on Co-super alloys like Co-Ni-Ta, Co-Ti-Ta, Co-Mo-Nb and Co-Mo-Re. The clustering is implemented using machine learning techniques. Each diffraction pattern is considered as a multi-dimensional vector corresponding to the intensity at each one of the 1500 Q values. The code separates out common sets of diffraction lines. A fuzzy c-means clustering algorithm was adapted for phase separation the resulting clusters map for the Co-Ni-Ta system. This XRD determined map is combined with the XRF atomic ratios map to produce the ternary phase diagram (see Figure 12.8).

Interplay of cation and anion redox in $Li_4Mn_2O_5$ cathode material and prediction of improved Li-4(Mn,M)(2)O-5 electrodes for Li-ion batteries Significant research effort has focused on improving the specific energy of lithium-ion batteries for emerging applications, such as electric vehicles. Recently, a rock salt-type $Li_4Mn_2O_5$ cathode

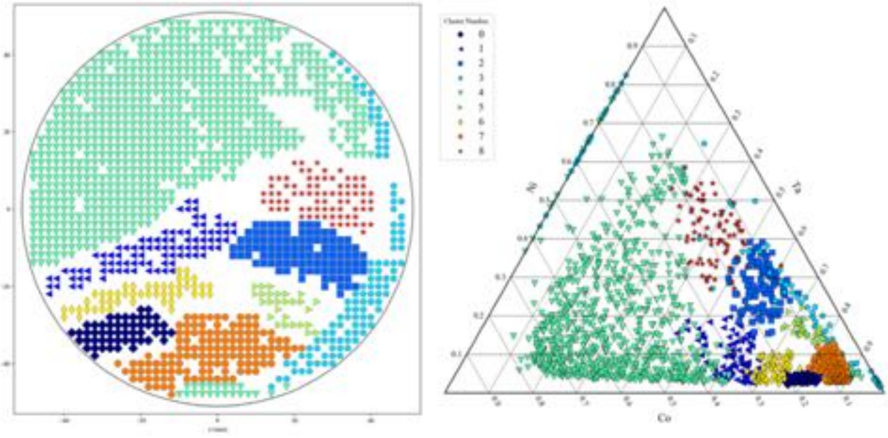


Figure 12.8: Phase clustered Co-Ni-Ta system at 900°C. The XRD determined phase clusters mapped on to the 100 mm wafer, x and y-axis indicating the location on the wafer in millimeter (left); The phase clustered XRD data combined with XRF determined atomic ratios mapped on the Gibbs triangle. The markers indicating the pure-phase regions (right)

material with a large discharge capacity ($\sim 350 mA \cdot hour g^{-1}$) was discovered. However, a full structural model of $Li_4Mn_2O_5$ and its corresponding phase transformations, as well as the atomistic origins of the high capacity, warrants further investigation. The **Wolverton** group used first-principles density functional theory (DFT) calculations to investigate both the disordered rock salt-type $Li_4Mn_2O_5$ structure and the ordered ground-state structure. The ionic ordering in the ground-state structure is determined via a DFT-based enumeration method. Using both the ordered and disordered structures to interrogate the delithiation process, it was found that it occurs via a three-step reaction pathway involving the complex interplay of cation and anion redox reactions: (i) an initial metal oxidation, $Mn^{3+} \rightarrow Mn^{4+}(Li_xMn_2O_5, 4 > x > 2)$; (ii) followed by anion oxidation, $O^{2-} \rightarrow O^{1-}(2 > x > 1)$; and (iii) finally, further metal oxidation, $Mn^{4+} \rightarrow Mn^{5+}(1 > x > 0)$. This final step is concomitant with the Mn migration from the original octahedral site to the adjacent tetrahedral site, introducing a kinetic barrier to reversible charge/discharge cycles. Armed with this knowledge of the charging process, high-throughput DFT calculations were used to study metal mixing in this compound, screening potential new materials for stability and kinetic reversibility. It is predicted that mixing with $M = V$ and Cr in $Li_4(Mn,M)_2O_5$ will produce new stable compounds with substantially improved electrochemical properties. Results were published in Science Advances [27.45]

New Two-Dimensional Thermoelectric Material with Ultralow Thermal Conductivity
A high power factor and low lattice thermal conductivity are two essential ingredients of highly efficient thermoelectric materials. Although monolayers of transition-metal dichalcogenides possess high power factors, high lattice thermal conductivities significantly impede their practical applications. The **Wolverton** group's first-principles calculations show that these two ingredients are well fulfilled in the recently synthesized Pd_2Se_3 monolayer, whose crystal structure is composed of $[Se_2]^{2-}$ dimers, Se^{2-} anions, and Pd^{2+} cations coordinated in a square-planar manner. The detailed analysis of third-order interatomic force constants reveals that the anharmonicity and soft phonon modes associated with covalently bonded $[Se_2]^{2-}$ dimers lead to ultralow lattice thermal conductivities in Pd_2Se_3 monolayers (1.5 and 2.9 $Wm^{-1}K^{-1}$ along the a- and b-axes at 300 K, respectively),

which are comparable to those of high-performance bulk thermoelectric materials such as PbTe. Moreover, the pudding-mold type band structure, caused by Pd^{2+} (d^8) cations coordinated in a square-planar crystal field, leads to high power factors in Pd_2Se_3 monolayers. Consequently, both electron- and hole-doped thermoelectric materials with a considerably high-zT can be achieved at moderate carrier concentrations, suggesting that Pd_2Se_3 is a promising two-dimensional thermoelectric material. These results suggest that hierarchical chemical bonds, that is, coexistence of different types of chemical bonds, combined with a square-planar crystal field is a promising route for designing high-efficiency thermoelectric materials. This work was published in Chemistry of Materials [27.43].

MRS Bulletin Issue on "Data-centric science for materials innovation" With the development of high-speed computers, networks, and huge storage, researchers can utilize a large volume and wide variety of materials data generated by experimental facilities and computations. The emergence of these big data and advanced analytical techniques has opened unprecedented opportunities for materials research. The discovery of many kinds of materials, such as energy-harvesting materials, structural materials, catalysts, optoelectronic materials, and magnetic materials, have been greatly accelerated through high-throughput screening. The utility of data-centric science for materials research is likely to grow significantly in the future. Unraveling the complexities inherent in big data could lead to novel design rules as well as new materials and functionalities. The September 2018 issue of MRS Bulletin [27.49] focused on these data-centric topics in materials innovation, editors: I. Tanaka, K. Rajan, and C. **Wolverton**.

12.4 Impact Statement for CHiMaD Phase I

As we envisioned five years ago, the CHiMaD data mining team has put CHiMaD and NIST at the forefront of materials informatics and data science. This is particularly evident with the visibility and impact of numerous cross-domain collaborations, publications in top venues/journals (both in CS and MS), software tools, etc. that would not have been possible without CHiMaD. In addition, we have been able to inspire a lot of graduate students and young researchers both in CS and MS, to learn and develop interdisciplinary materials informatics skills, who will become future leaders in the field. The international recognition and acclaim that the CHiMaD data mining team has been receiving is extremely encouraging. As an example, even computer scientists in the team have delivered numerous invited lectures/keynotes in top materials science conferences all over the world, invited to path finding workshops organized by federal agencies, and so on. Finally, we are seeing a tremendous amount of interest from the industry to adopt these AI/ML techniques in their workflows, with a surge in the number of collaboration requests, many of which have not only led to external projects/funding, but more importantly are enabling to significantly expand the impact of data-driven materials science, and accelerate the discovery & deployment of advanced materials in the society, which is the vision of MGI.

12.5 Collaborations

12.5.1 CHiMaD Collaborations

Data mining for Additive Manufacturing

*Ankit **Agrawal** (NU), Alok **Choudhary** (NU), Jian **Cao** (NU)*

This collaboration with the additive manufacturing seed group aims to develop data mining models for predicting temperature profiles during direct energy deposition process.

Phase Clustering

Ankit ***Agrawal*** *(NU), Alok* ***Choudhary*** *(NU), Michael* ***Bedzyk*** *(NU), Yip-Wah* ***Chung*** *(NU)*
This collaboration with the precipitation-strengthened alloys use-case group aims to develop clustering techniques on 1-D X-ray diffraction data (peaks) to identify phases.

Thermoelectrics Data Mining

Ankit ***Agrawal*** *(NU), Alok* ***Choudhary*** *(NU), Greg* ***Olson*** *(NU)*
Stemming from a collaboration which started with the thermoelectrics-focused DARPA SIMPLEX project, and resulting in CHiMaD's thermoelectrics seed group, this project aims at developing predictive models for thermoelectric properties of compounds and materials.

DFT-OQMD data mining

Chris ***Wolverton*** *(NU), Ankit* ***Agrawal*** *(NU), Alok* ***Choudhary*** *(NU), Logan Ward (UC)*
This collaboration involves several projects and is geared towards applying data mining techniques for predictive modeling of various materials properties using the OQMD database.

Phase-field modeling of Co-based superalloys

Olle ***Heinonen*** *(ANL), Chris* ***Wolverton*** *(NU)*
This ongoing collaboration uses DFT to calculate input parameters required by phase-field modeling, such as interfacial energies and elastic constants.

Cellulose Nanocomposite (CNC) Design

Sinan ***Keten*** *(NU), Wei* ***Chen*** *(NU)*
This collaboration investigated the design of polymer grafted cellulose nanocomposite (CNC) using molecular dynamics simulations. A metamodel was built to study the mechanical properties of PMMA-CNC nanocomposite and a statistical sensitivity analysis was conducted for interpreting the model and screening out the important/unimportant factors.

12.5.2 External Collaborations

Data-driven Tools for Accelerating Materials Property/Structure Prediction

Samsung Advanced Institute of Technology (SAIT) , Chris ***Wolverton*** *(NU)*
A project with Samsung was initiated to develop machine learning models applied to dielectric materials. The materials representations developed will be used in the group's ML work, and also the structural descriptors such as Voronoi Tesselation, and CGCNN models to apply these ML tools to search for high-dielectric gate materials. Samsung has previous experience with applying neural nets to organic molecule datasets, and we are collaborating with them to determine how to apply these NN approaches to inorganic, crystalline datasets.

Materials Informatics: Large Databases and Machine Learning for Materials Design and Discovery

Toyota Research Institute, Chris ***Wolverton*** *(NU)*
This collaboration expands the OQMD via calculations of complex properties (phonons, defects) and more accurate methods, for a sizeable (but not complete) set of $10^3 - 10^4$ compounds. The machine learning approaches to predict these properties will allow the

"completion" of these datasets across a much wider range of $\sim 10^6 - 10^8$ compounds and materials at a significantly reduced computational cost.

Deep Learning 3-Phase Polymer Systems

Craig Burkhart (Goodyear Tire Company), Wei ***Chen*** *(NU)*
This collaboration applied the developed deep learning algorithm for the 3-phase polymer nanocomposite systems.

Digital Manufacturing and Design Innovation Institute (DMDII)

Kornel F. Ehmann (NU), Greg J. Wagner (NU), Lucia Mirabella (Siemens), Suraj R. Musuvathy (Siemens), Edward R. Slavin III (Siemens), Sanjeev K. Srivastava (Siemens), Federico M. Sciammarella (NIU), Joseph S. Santner (NIU), Jason ***Sebastian*** *(QuesTek), David Snyder (QuesTek), Jiten Shah (Product Development & Analysis), Jian* ***Cao*** *(NU), Wei* ***Chen*** *(NU), Gregory* ***Olson*** *(NU), Wing-Kam* ***Liu*** *(NU)*
The external collaborators are working with a DMDII project working on AM process modeling, calibration, assessment and prediction. This work includes calibration experiments, macro-scale and micro-scale simulations for mechanical properties and processing, and characterization experiments.

Creation of NanoMine Data Resource and Materials Informatics for Polymer Nanocomposites Analysis and Design

Cate Brinson (Duke University), Wei ***Chen*** *(NU), Linda Shradler (RPI)*
This collaboration was towards creation of the Nanomine data resource including data schema, knowledge graph ontology, and microstructure characterization & reconstruction and data analysis tools. Built upon the Nanomine data resource, case study for nanodielectrics in capacitor has been created to demonstrate the integration of the processing-structure and structure-property relationships to develop optimized materials for achieving multiple desired properties.

Steel data mining

Ankit ***Agrawal*** *(NU), Alok* ***Choudhary*** *(NU), Greg* ***Olson*** *(NU), Abhinov* ***Saboo*** *(QT), Wei Xiong (University of Pittsburgh)*
This collaboration aims to develop predictive models for properties of steel, as well as use CALPHAD to enrich steel composition and processing data with structure information to derive more reliable data-driven structure-property linkages.

12.5.3 NIST Collaborations

Accelerated Discovery of Metallic Glasses

Jason ***Hattrick-Simpers*** *(NIST), Apurva Mehta (SLAC), Chris* ***Wolverton*** *(NU)*
This collaboration focuses on the discovery of new metallic glasses with machine learning and high-throughput experimentation.

Crystal Plasticity data mining

Ankit ***Agrawal*** *(NU), Alok* ***Choudhary*** *(NU), Andrew* ***Reid*** *(NIST), Papanikolaou (West Virginia University)*
This is a collaboration with NIST and West Virginia University that aims at developing data-driven techniques on discrete dislocation dynamics simulation images to identify prior deformation history.

Superalloys data mining

Ankit ***Agrawal*** *(NU), Alok* ***Choudhary*** *(NU), Carelyn* ***Campbell*** *(NIST), Shengyen* ***Li***

(NIST)
This project aims at using data-driven techniques to predict work-to-necking of a Ni superalloy, and subsequently discover superalloys with optimal properties.

X-ray Diffraction image mining
Gilad ***Kusne*** *(NIST), Nam* ***Nguyen*** *(NIST), Jason* ***Hattrick-Simpers*** *(NIST), Martin* ***Green*** *(NIST), Ankit* ***Agrawal*** *(NU), Alok* ***Choudhary*** *(NU)*
This project aims at using data-driven techniques for noise/background removal from XRD images, as well classification of XRD images into different phase categories.

JARVIS-DFT data mining
Kamal ***Choudhary*** *(NIST), Francesca* ***Tavazza*** *(NIST), Brian* ***DeCost*** *(NIST), Carelyn* ***Campbell*** *(NIST), Ankit* ***Agrawal*** *(NU), Alok* ***Choudhary*** *(NU)*
This collaboration aims at applying transfer learning techniques for predicting materials properties on JARVIS-DFT dataset.

Florescence Life Time
Jeff ***Gilman*** *(NIST), Wei* ***Chen*** *(NU)*
This collaboration intends to analyze fluorescence life time data. NIST colleagues are currently making improvements on data collection.

CNT Polymer Composite Design
Bharath ***Natarajan*** *(NIST), Wei* ***Chen*** *(NU)*
This collaboration characterized CNT polymer composites using Spectral Density Function (SDF) method.

12.6 Technology Transfer

Software
Martensite Start Temperature (MsT) Predictor
Ankit ***Agrawal****, Wei Xiong, Greg* ***Olson****, Alok* ***Choudhary***
Distribution: Public Beta
Access Link: `http://info.eecs.northwestern.edu/MsTpredictor`
This is an online software tool that estimates the martensite start temperature (MsT) of a given steel based on its composition parameters entered by the user. The predictive models deployed in this tool were built on data provided by Xiong (University of Pittsburgh). All the models and software for the online tool have been developed by **Agrawal**.

Software
Ni-Al-Cr Superalloy Property Predictor
Ankit ***Agrawal****, Shengyen* ***Li****, Carelyn* ***Campbell****, Alok* ***Choudhary***
Distribution: Public Beta
Access Link: `http://info.eecs.northwestern.edu/NiSuperalloy`
This is an online software tool that estimates the work-to-necking (WTN) of a given Ni-Al-Cr superalloy at a given temperature based on its composition parameters entered by the user. The predictive models deployed in this tool were built on data provided by **Li** and **Campbell** at NIST. All the models and software for the online tool have been developed by **Agrawal**.

Community Resource
Podcast: Role of AI in Materials Science
Chris ***Wolverton***

Access Link: `https://citrine.io/category/podcasts/`
Chris **Wolverton** was featured in a recent Citrine podcast on the role of AI and machine learning in materials datasets. The podcast is available on the Citrine website (link provided above), and is also available for listen/download on iTunes, Stitcher, and a few other podcasting services.

Community Resource
Public Outreach: Accelerated Discovery of Metallic Glasses
*Chris **Wolverton**, Jason **Hattrick-Simpers***
The NIST-CHiMaD collaborative research on *accelerated discovery of metallic glasses through iteration of machine learning and high-throughput experiments* was prominently featured in high-profile articles on:

- Forbes, "Scientists Use Artificial Intelligence To Discover New Materials" `https://www.forbes.com/sites/meriameberboucha/2018/04/22/scientists-use-artificial-intelligence-to-discover-new-materials/#12b60feb38c4`
- The Verge, "How AI is Helping Us Discover Materials Faster Than Ever" `https://www.theverge.com/2018/4/25/17275270/artificial-intelligence-materials-science-computation`

Data
Datasets Uploaded to Online Resources in 2018

1. Microstructure Characterization and Reconstruction images and tools
   *Iyer A.; **Chen**, W.*
   Link: `http://nanomine.northwestern.edu:8000/`

12.7 Publications and Presentations

26 Number of CHiMaD-supported publications in 2018 by Data Mining Tool group. Please see chapter 27 for details [27.29, 27.30, 27.31, 27.32, 27.33, 27.34, 27.35, 27.36, 27.37, 27.38, 27.39, 27.40, 27.102 27.43, 27.45, 27.46, 27.63, 27.49, 27.50, 27.51, 27.52, 27.53, 27.54, 27.55, 27.56, 27.57].

24 Number of presentations on CHiMaD supported research in 2018, please see section 26.11 for the complete list.

12.8 CHiMaD Team

Data Mining Use-Case Group Researchers			
Name	**Position**	**Affiliation**	**Project (PI)**
Alok Choudhary Ankit Agrawal Christopher Wolverton Wei Chen	Principal Investigator Principal Investigator Principal Investigator Principal Investigator	NU/EECS NU/EECS NU/MSE NU/ME	
Wei-keng Liao	Research Faculty	NU/EECS	DFT data mining *(Choudhary, Agrawal)*
Kasthurirangan Gopalakrishnan	Research Faculty	NU/EECS	Infrastructure crack detection using deep transfer learning *(Choudhary, Agrawal)*
Arindam Paul	Graduate Student	NU/EECS	Chemical Property Prediction *(Choudhary, Agrawal)*
Dipendra Kumar Jha	Graduate Student	NU/EECS	DFT data mining *(Choudhary, Agrawal)*
Zijang Yang	Graduate Student	NU/EECS	Crystal plasticity data mining *(Choudhary, Agrawal)*
Sean Griesemer	Graduate Student	NU/MSE	Discovering Crystal Structures in High Throughput *(Wolverton)*
Cheol Woo Park	Graduate Student	NU/MSE	Machine Learning for Materials Discovery *(Wolverton)*
Soo Kim	Graduate Student	NU/MSE	DFT data mining *(Wolverton)*
Vinay Hegde	Graduate Student	NU/MSE	DFT data mining *(Wolverton)*
Yichi Zhang	Graduate Student	NU/ME	Bayesian optimization for materials design *(Chen)*
Akshay Iyer	Graduate Student	NU/ME	NanoMine data resource tools and web development *(Chen)*
Xiaolin Li	Graduate Student	NU/ME	Deep Learning methods *(Chen- external industry funding: Goodyear)*
Tianyu Huang	Graduate Student	NU/ME	Metamodeling and statistical analysis of polymer nanocomposite systems *(Chen- external funding: DOE/Ford ICME)*

13. Materials Data Facility

Ian Foster (UC/ANL), Benjamin Blaiszik (UC/ANL), Steven Tuecke (UC/ANL), Kyle Chard (UC), Jim Pruyne (UC), Logan Ward (UC) , Marcus Schwarting (ANL), Jonathan Gaff (UC)

Chandler Becker (NIST), Sherif Youssef (NIST), Alden Dima (NIST), Robert Hanisch (NIST), Mary Brady (NIST), Raymond Plante (NIST), Zachary Trautt (NIST), Kimberly Tryka (NIST), James Warren (NIST), Carelyn Campbell (NIST), Martin Green (NIST), Aaron Kusne (NIST), Nhan Nguyen (NIST), Jason Hattrick-Simpers (NIST), Joshua Martin (NIST)

Significance

At a high level, the goals of Materials Data Facility (MDF) include streamlining and automating data sharing, discovery, access, and analysis by: 1) Simplifying data publication, regardless of data size, type, and location; 2) Enabling automated data and metadata ingest to simplify discovery, usage, and mixing of distributed materials datasets; 3) Unifying search of disparate materials data sources from both MDF and other community sources; and 4) Connecting the growing materials data ecosystem with shared data extractors and interfaces to facilitate cross-deposit of data and metadata. By achieving these goals, MDF will create a set of unique data resources and interfaces to encourage broader data reuse, promote improved replicability practices, and enable vast new machine learning opportunities within CHiMaD and in the broader community.

13.1 Goals

A summary of the Materials Data Facility (MDF) service ecosystem is provided in (Figure 13.1), showing how researchers can: 1) deposit data from many common locations (e.g., *Globus, Google Drive*, and *4CeeD*, Figure 13.1-*Left*); 2) have that data indexed and enriched by the *MDF Connect* service (Figure 13.1-*Center*) and 3) register that data with many different services (MDF Data Publication and Discovery services, *Citrination, NIST MRR*, Figure 13.1-*Right*). Using MDF services, researchers can publish datasets as bundles

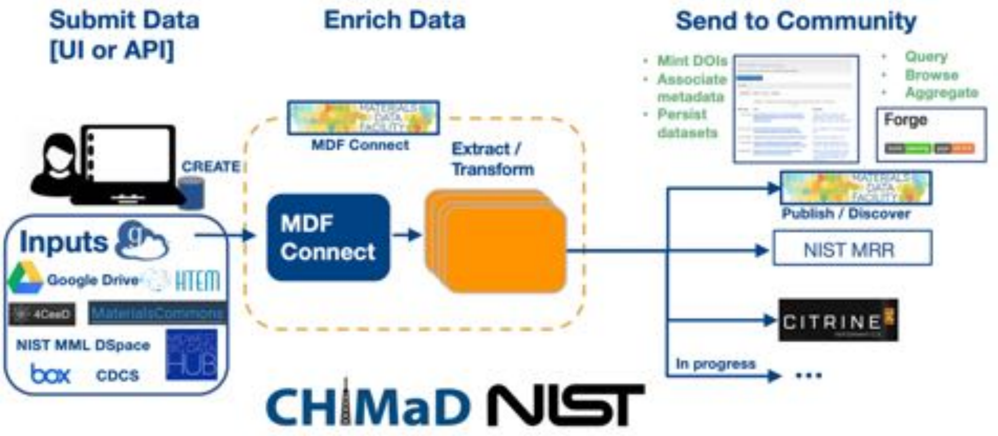


Figure 13.1: MDF Data ecosystem and MDF Connect prototype flow. The user specifies a minimal amount of metadata, including authors, title, and data locations and sends a request for dataset creation to MDF Connect. MDF Connect assembles the data from the enumerated data locations and performs a series of extractions and transformations on the data. The data are then shared with the community resources enumerated by the user.

of data and metadata, leveraging distributed storage systems. Users may query, browse, aggregate, and download data and metadata from the data discovery service through a web UI, an API, or a Python interface. Importantly, the MDF index contains searchable information about many datasets external to the MDF ecosystem providing researchers a unique way to search across community data resources.

13.2 Accomplishments in Research and Feature

13.2.1 MDF Data Publication and Discovery

The MDF team has continued to operate and maintain the MDF Data Publication and Discovery services. The MDF Data Publication service enables publication of datasets up to many terabytes in size and many millions of files. We have successfully published 112 datasets, representing over 200 authors, and including 43 datasets with CHiMaD researchers (Figure 13.2). In 2018 we saw a dramatic increase in datasets being deposited that are critical to machine learning studies in materials. MDF also continued to develop, extend, and maintain the MDF Data Discovery service and the Forge Python client that has been described in previous reports. The Forge client provides programmatic access allowing researchers to search, aggregate, and download data from MDF. The code to Forge is openly available on Github, and the client can be installed via the Python Package Index (PyPI) from Mac/Windows/Linux command line using the PyPI installer as: $ pip install mdf_forge.

13.2.2 MDF Connect

Researchers today generate data from many different instruments and computational resources, store that data in many locations, and have many choices of data repositories for storing these data during and after completion of their research. To begin simplifying this ecosystem, we have developed and deployed a prototype service called MDF Connect (Figure 13.1). MDF Connect is a data service built to facilitate simple, automated or

Figure 13.2: Cumulative statistics through 2018 for the MDF data publication service. MDF contains 112 datasets, 43 of which are affiliated with CHiMaD PIs.

form-driven, submission of datasets into MDF Publish and Discover, enrichment of the data through material-specific metadata extraction, and automation of data and extracted metadata registration in a variety of community data services. Another key feature of MDF Connect is the integration of MDF Connect with cloud storage providers (*Google Drive, Box, Dropbox*), distributed storage solutions (*Globus*), repositories (*NIST MML DSpace, Figshare, Zenodo*), and material-specific services (*4CeeD, Materials Commons*) to enable researchers to directly import their data from its current location without added complexity. These key integrations are described in the subsequent section.

MDF Connect Integrations

We have developed integrations to a variety of data sources that will simplify the way users can make data available as input to MDF. We have built integrations to *Dropbox, Figshare,* and *Amazon S3*. Additionally, we have built integrations to allow deposition of data from *Globus, Google Drive*, and *Box* - three common data storage locations we identified as being used by researchers to share files. These integrations (Figure 13.3) will allow users to send data to MDF Connect from the locations where they are already performing their collaborative file sharing. Further, we have prioritized the development of integrations to emerging NSF and DOE-funded materials-specific data platforms (e.g., *4CeeD* and *Materials Commons* respectively). The *4CeeD* integration has been deployed within *4CeeD*, and their users are now able to bundle their project data and send that data bundle to MDF Connect after filling out a minimal set of metadata.

MDF Connect Data Indexers

MDF Connect is able to parse metadata from CALPHAD TDB files, electron microscopy formats (e.g., dm3, dm4, TIF, etc.), CIF, VASP, generic image, and Citrine-format CSV files automatically. If the user supplies a mapping of the data, MDF Connect can also parse from JSON, CSV, YAML, XML, and Excel files, as well as metadata stored in filenames (with a user supplied regex). When a file is parsed, basic file information (e.g., file name, file size, file checksum) is also recorded.

The MDF team is working closely with developers from NREL's High-throughput experimental database (HTEM), Citrine Informatics, and 4CeeD/Brown Dog to standardize and share scientific data type extractors. This collaborative project, tentatively called **MaterialsIO**, is available on Github at `https://github.com/materials-data-facility/MaterialsIO`. MDF is leading an effort to form a working group, including participants from NIST, to develop appropriate standards to facilitate sharing and reuse of these extractors. This group will be convened by the end of 2019.

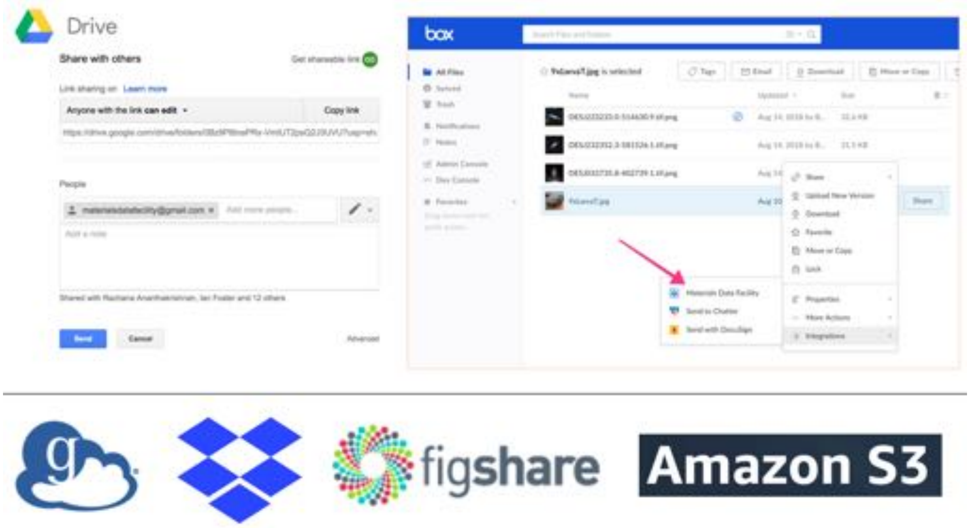


Figure 13.3: MDF Connect data input integrations. (top-left) To use data from Google Drive, a user shares the data with the MDF Google user. (top-right) To use data from Box, a user installs the MDF Connect Box app and then can click a file or a folder to send to MDF Connect. (bottom) MDF Connect also supports direct connection to Globus, Dropbox, Figshare, Amazon S3, and others.

13.3 Access Information

Data publication and data discovery services are now available generally (i.e., to CHiMaD researchers and researchers in the broader materials community). *MDF Connect* is in alpha testing, and is available to CHiMaD researchers for testing purposes. Researchers interested in using MDF services within their personal research, research group, or larger collaborations can visit `https://www.materialsdatafacility.org` for more information or contact the MDF project PIs. All codes described here (and more) are publicly available within project-specific repositories at `https://github.com/materials-data-facility`.

13.4 Collaborations

13.5 NIST Collaborations

Search for Thermoelectric Compositionally-Complex Alloys

*Ian **Foster** (UC), Apurva Mehta (SLAC), Bryce Meredig (Citrine), Chistopher **Wolverton** (NU), Jeffrey **Snyder** (NU), Jason **Hattrick-Simpers** (NIST), Joshua **Martin** (NIST)*
Members of the MDF team have started a newly DOE-funded collaboration with members of NIST and CHiMaD on discovering thermoelectric materials with High-Throughput Experiment (HTE) and machine learning. The project, which started in October 2018, will combine the HTE and thermoelectric expertise of NIST/CHiMaD with the data science experience of the MDF team.

Phase Field Hub (PFHub) and MDF Integrations

*Ian **Foster** (UC), Ben **Blaiszik** (UC), Raymundo Arroyave (Texas A&M University), Daniel **Wheeler** (NIST), Trevor **Keller** (NIST), James **Warren** (NIST)*
The MDF team is working with the NIST/CHiMaD Phase Field Community Hub to simplify their data ingest processes and to enable PFHub data assets to be more easily

discovered by the community. Additionally, through this work, MDF is working to publish a >50 TB dataset generated at Texas A&M University.

High-throughput Experimental Materials Science Virtual Laboratory
*Ian **Foster** (UC), Ben **Blaiszik** (UC), Zachary **Trautt** (NIST), Martin **Green** (NIST), Aaron **Kusne** (NIST), Jason **Hattrick-Simpers** (NIST), Nhan **Nguyen** (NIST)*
MDF will take part as a node in the Materials Science Virtual Laboratory (MSVL) that is led by NIST. The MSVL will leverage MDF and other *Globus* services to provide key aspects of the data infrastructure for the evolving project.

Cloud Deployment of a MDF Materials Resource Registry (MRR)
*Ian **Foster** (UC), Ben **Blaiszik** (UC), Lauro **Bartolo** (NU), Chandler **Becker** (NIST), Sharief **Youssef** (NIST), Alden **Dima** (NIST), Robert **Hanisch** (NIST), Mary **Brady** (NIST), Ray **Plante** (NIST), Zachary **Trautt** (NIST), Kimberly **Tryka** (NIST), James **Warren** (NIST)*
MDF has worked with NIST to deploy a second MRR instance hosted in the AWS cloud. This instance has been deployed, has been populated with MDF records, and has been used to demonstrate cross-harvesting between the NIST MRR and the MDF MRR.

Polymer Property Predictor and Database (PPPdb) MDF Integrations
*Ian **Foster** (UC), Ben **Blaiszik** (UC), Debra **Audus** (NIST), Juan **de Pablo** (UC)*
The MDF team is working with the PPPdb team to regularly index the contents and to make the compiled polymer properties programmatically accessible.

Polymer Pilot Project: Polymer Data Special Issue Calls
*Laura **Bartolo** (NU), Begum **Gulsoy** (NU), Ian **Foster** (UC), Benjamin **Blaiszik** (UC), Alden **Dima** (NIST), Chandler **Becker** (NIST), Frederick **Phelan** (NIST), James **Warren** (NIST)*
MDF data infrastructure is being leveraged to simplify data deposit for the upcoming polymer data special issue calls in 2019 for the journals Macromolecules and ACS Macro Letters. More information on the pilot project can be found under CHiMaD Data Efforts, Chapter 19.

NIST-CHiMaD Materials Microscopy Data Workshop Co-Planning
*Laura **Bartolo** (NU), Begum **Gulsoy** (NU), Ian **Foster** (UC), Benjamin **Blaiszik** (UC), Carelyn **Campbell** (NIST), June **Lau**, John Henry **Scott**, Zachary **Trautt**, Marcus Hanwell (KitWare), William Harris (Zeiss Microscopy)*
MDF team members **Foster** and **Blaiszik** were part of the organizing committee for the NIST-CHiMaD Materials Microscopy Data Workshop held at Northwestern in October 25-28 2018. More information on this workshop can be found under CHiMaD Data Efforts, Chapter 19.

13.6 External Collaborations

Integrative Materials Design (IMAD): Midwest Big Data Spoke Joint Proposal
*Ian **Foster** (UC/ANL), Benjamin **Blaiszik** (UC), Laura **Bartolo** (NU), Peter **Voorhees** (NU), Dane Morgan (UW-Madison), John Allison (UM-Ann Harbor), Dallas Trinkle (UIUC)*
This continuing collaboration by materials researchers at UC, NU, Argonne, UIUC, UW-Madison, UM-Ann Arbor aims to connect various efforts across the Midwest, including industrial, to build a variety of partnerships and leverage shared data services and capabilities in their materials design and research efforts. More information on IMAD can be found under CHiMaD Data Efforts, Chapter 19.

13.7 Future Outlook

In 2019, MDF research activities will focus on automation, integration, and extraction. Automation will include developing methods for continuous data refinement, extraction of metadata indexed in MDF, and data capture at the point of generation via integration with other services (*MDCS*). Integration with community data services, experimental facilities (e.g., NIST and Advanced Photon Source (APS) beamlines) and computing facilities will expand data acquisition and the use base, and provide opportunities for long-term sustainability. Extraction will involve automated identification of metadata from diverse materials file types and output formats, for use in MDF and other community efforts (e.g., *Materials Commons, 4CeeD, MATIN, NIST MRR*, and *Citrine*).

Growing MDF capabilities in the coming year will allow each CHiMaD use case to make gathered datasets available internally and to the community through web and programmatic interfaces. For example: (1) Polymer databases: We will continue to work to index and share metadata from NanoMine and PPPdb, and will publish and index datasets from articles in the upcoming polymer special issue journal calls of Brinson (Duke), **de Pablo**, **Foster**, and **Bartolo**. (2) Phase Field Methods: MDF will support this case, and the broader phase field community, by publishing and indexing challenge submissions (i.e., test results, timestep images, meshes) for greater replicability and transparency. (3) Data Mining: We will leverage MDF data services and clients to create unique training sets. MDF has prepared a joint proposal with PIs **Choudary** and **Agrawal** to seed a new data mining effort that leverages MDF capabilities and also would generate data relevant to CHiMaD.

13.8 Technology Transfer

Data Services
Materials Data Facility
I. Foster, S. Tuecke, B. Blaiszik, K. Chard, J. Pruyne, M. Ondrajcek
Access Link: `https://materialsdatafacility.org`

Data
Sample Datasets Uploaded to Materials Data Facility in 2018
***denotes CHiMaD affiliated dataset*

1. NaxCoO2 phase stability and hierarchical orderings in the O3/P3 structure family
   Kaufman, Jonas L.; Van der Ven, Anton [UCSB]
   DOI: 10.18126/M2505P
2. Dataset for Polyelectrolyte Complexation of Oligonucleotides by Charged Hydrophobic - Neutral Hydrophilic Block Polymers
   Marras, Alexander E.; Vieregg, Jeffrey R.; Ting, Jeffrey M.; Rubien, Jack D.; Tirrell, Matthew V. [University of Chicago]
   DOI: 10.18126/M2QW8R**
3. Data for Overcoming the Overcoming the Memory Bottleneck in Auxiliary Field Quantum Monte Carlo Simulations with Interpolative Separable Density Fitting
   Malone, Fionn D.; Shuai, Zhang; Miguel, Morales. A [Lawrence Livermore National Laboratory]
   DOI: 10.18126/M29S67

4. Hot-electron mediated ion diffusion in proton-irradiated magnesium oxide
   Lee, Cheng-Wei; Schleife, Andre, [University of Illinois at Urbana-Champaign]
   DOI: 10.18126/M2C35K
5. Dataset for An efficient hybrid orbital representation for quantum Monte Carlo calculations
   Luo, Ye; Esler, Kenneth P.; Kent, Paul R. C.; Shulenburger, Luke [Oak Ridge National Laboratory]
   DOI: 10.18126/M2S64Z
6. Vacancy Mediated and Interstitial Solute Transport in Zr from Density Functional Theory Calculations
   Jain, Abhinav C. P.; Burr, Patrick A.; Trinkle, Dallas R. [University of Illinois at Urbana-Champaign]
   DOI: 10.18126/M2R04K
7. GISAXS Data for Polymer Thin Films and Gratings
   Dolejsi, Moshe; Bowen, Alec; de Pablo, Juan; Nealey, Paul [University of Chicago]
   DOI: 10.18126/M25630**
8. A Machine Learning Approach for Engineering Bulk Metallic Glass Alloys *Ward, Logan; O'Keeffe, Stephanie C.; Stevick, Joesph; Jelbert, Glenton R.; Aykol, Muratahan; Wolverton, Chris [Northwestern University]*
   DOI: 10.18126/M2662X**
9. Detection of Open Loop Defects in STEM Images of Irradiation-Damaged Alloys - Source Code for Detection and Image Dataset
   Li, Wei; Field, Kevin G.; Morgan, Dane [University of Wisconsin-Madison]
   DOI: 10.18126/M2692Z
10. Accelerated Discovery of Metallic Glasses through Iteration of Machine Learning and High-Throughput Experiments
    Fang, Ren; Ward, Logan; Williams, Travis; Laws, Kevin J.; Wolverton, Christopher; Hattrick-Simpers, Jason; Mehta, Apurva [SLAC National Accelerator Laboratory]
    DOI: 10.18126/M2B06M**
11. The Third Sandia Fracture Challenge
    Kramer, Sharlotte; Boyce, Brad; Jones, Amanda; Gearhart, Jhana; Salzbrenner, Brad [Sandia National Laboratory]
    DOI: 10.18126/M26D20
12. Spatially Heterogeneous Dynamics in a Metallic Glass Forming Liquid Imaged by Electron Correlation Microscopy *Zhang, Pei; Maldonis, Jason; Liu, Ze; Schroers, Jan; Voyles, Paul [University of Wisconsin-Madison]*
    DOI: 10.18126/M2GW5F

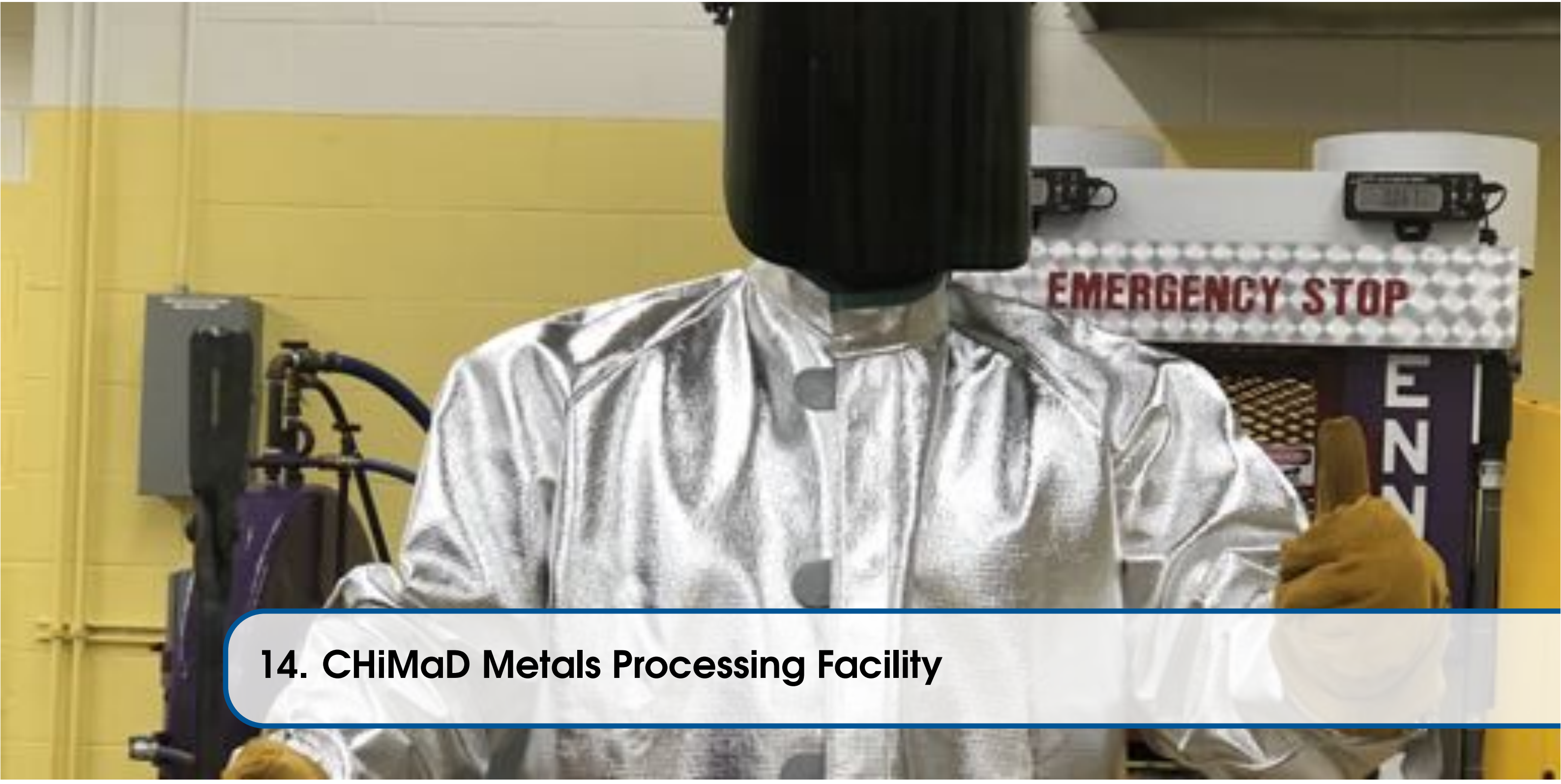
13.9 Publications and Presentations

8 Number of CHiMaD-supported publications in 2018 by Materials Data Facility Developers. Please see chapter 27 for details. [27.63, 27.101, 27.102, 27.103, 27.104, 27.105, 27.31, 27.34]

35 Number of presentations on CHiMaD supported research in 2018, please see section 26.12 for the complete list.

13.10 CHiMaD Team

Materials Data Facility Developers		
Name	**Position**	**Affiliation**
Ian Foster	Principal Investigator	UC/CS
Steven Tuecke	Deputy Director	UC/CI - ANL/MCS
Benjamin Blaiszik	Software Developer	UC/CI - ANL/MCS
Kyle Chard	Senior Researcher/Fellow	UC/CI - ANL/MCS
Jim Pruyne	Senior Software Developer	UC/CI
Michal Ondrejcek	Research Programmer	UIUC/NCSA
Logan Ward	Postdoctoral Researcher	UC/CS
Marcus Schwarting	Researcher	ANL



14. CHiMaD Metals Processing Facility

Facility Manager: Paul Adler (NU)
1801 Maple Avenue, Evanston, IL, USA 60201

Significance

The paradigm of computational design of material systems has been shown to significantly reduce time from ideation to component qualification. Despite the advantage of rapid prototyping via virtual simulation some experimentation is still required for model refinement. Initial testing typically involves production of buttons of desired composition mainly to verify predicted phase fractions, chemical partitioning, and cast microstructures. Scale-up manufacture of designed alloys will involve processing effects that greatly influence the final microstructural and thus property gradients. The goal of this effort is to construct a metal processing laboratory capable of capturing these effects for the expressed purpose of further refinement of computational models of promising systems.

14.1 CHiMaD Metals Processing Facility

The CHiMaD Metals Processing Facility (CMPF) construction was completed in March 2016. In February 2018, became a part of the Northwestern University's NUCORE system. As such, the CMPF metal-processing capabilities are available to non-profit academics and for-profit commercial organizations. Cost structures are commensurate with established standards for each category of user although we can generally accommodate customers budgetary constraints. The equipment is new or refurbished with data monitoring and acquisition systems built in depending on the specific unit. The equipment meets or exceeds all current governmental and societal safety guidelines. CMPF hosts a cadre of highly trained personnel to optimize successful metal processing efforts.

14.1.1 Currently Available Equipment

- Buehler AM/0.5 Arc Melter
- Fenn 4F-4 Rotary Swager - refurbished in 2016
- Fenn 051 2/4 Hi Rolling Mill - refurbished in 2016
- AIP6-45H Hot Isostatic Press (HIP)
- Lindberg 51863 Box Furnace
- Materials Research Furnaces (MRF) Quenching/Deformation Vacuum Furnace
- SentroTechST-1600C-666 Box Furnace
- Struers Labotom-5 Cut-off Saw
- Econoline 101696WD-A Direct Pressure Sand Blaster

Figure 14.1: CMPF Facility

14.2 Technology Transfer

Website

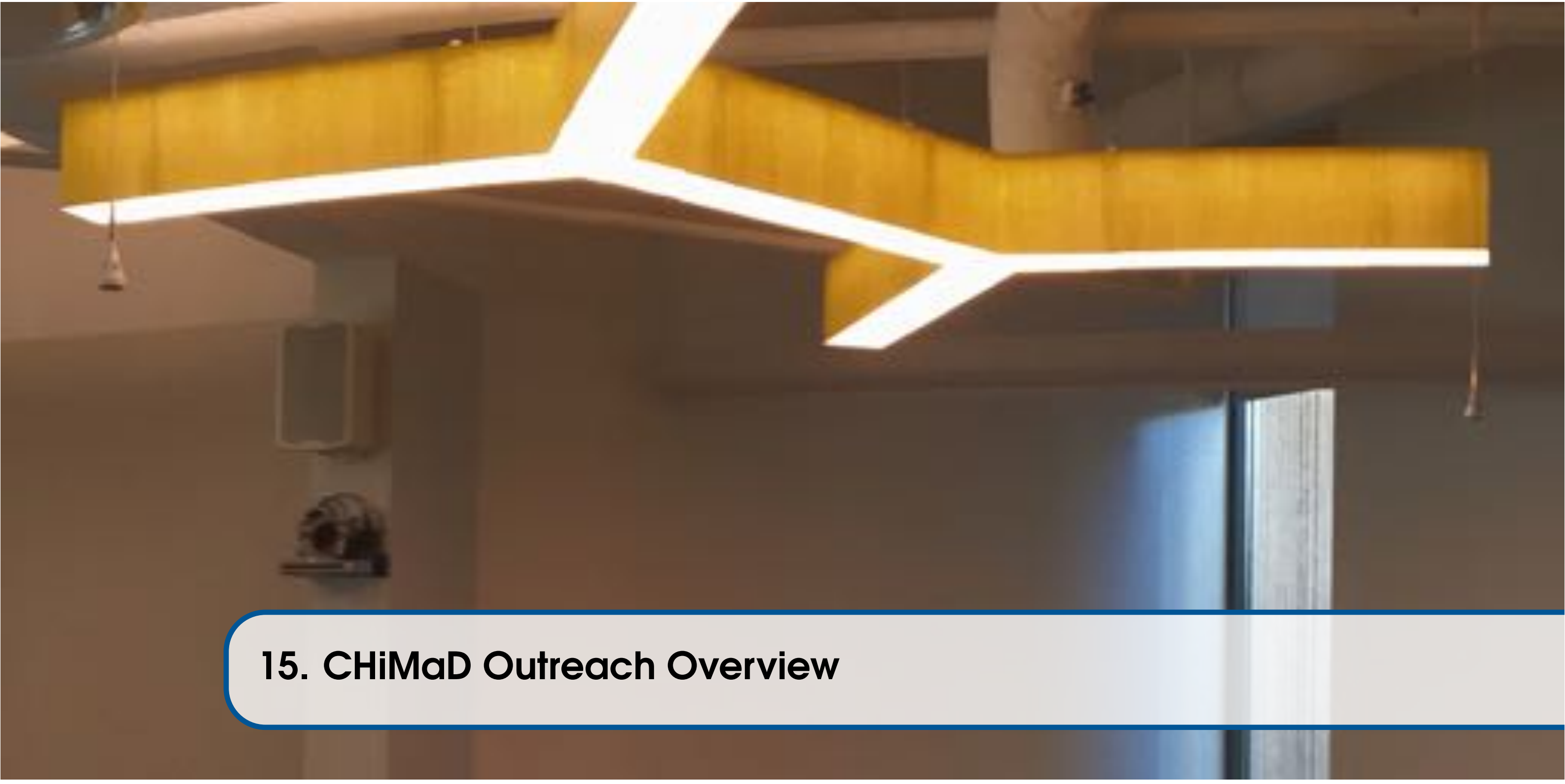
CHiMaD Metals Processing Facility

Access Link: `https://facilities.research.northwestern.edu/browse-facilities/chimad-metals-processing-facility-cmpf`

Official website for the CMPF allowing internal and external users to reserve the facility equipment.

V

Outreach



15. CHiMaD Outreach Overview

Begum Gulsoy (NU)

CHiMaD Outreach program and initiatives are developed to meet the goals of the MGI, with an emphasis on training the current and next generation of government, industrial and academic researchers with the right tools to make MGI a sustainable success. The major goals of the CHiMaD Outreach are as follows:

- Equipping the next generation workforce with concepts and tools needed to accelerate materials design.
- Engaging academia, government and industry in materials design and building a design-focused communication between the different entities.
- Enhancing the knowledge of the current workforce, in academia, government, and industry by making the existing and new design tools and concepts developed accessible.

The CHiMaD Outreach Program focuses on *Training, Community Building, Best Practices* and *Diversity* for Undergraduate and Graduate students, Postdoctoral Researchers, Research Faculty and Professional as well as Industry, Figure 15.1.

Towards these goal, in 2018, CHiMaD organized an MGI Seminar series, welcoming an international group of researchers to present about their MGI-related efforts as well as other international MGI initiatives they are a part of. This series of presentations were broadcast to all CHiMaD partner institutions. In accordance with the goal of equipping the next generation of researcher and in collaboration with ASM International and its Educational Committee, CHiMaD continued the dissemination of some of the most significant current materials design software to engineering universities in the U.S. where the universities are expected to build these tools into their undergraduate curriculum. Since the inception of this initiative, 17 universities were awarded the Materials Genome Toolkit. CHiMaD also took the lead in the organization of various high-impact workshops aiming at community building and bridging for collaborations and open-access exchange of knowledge on their topics of interest as well as initiated Materials Data and Database focused working groups and meeting series. In 2018, CHiMaD has continued improving its outreach activities for under represented groups, working with high-school students through the University of

Figure 15.1: CHiMaD Outreach Program is structured to address the *Training, Community Building, Best Practices* and *Diversity* in academia, government and industry.

Chicago Collegiate Scholars Program Summer School, and undergraduate level students through CHiMaD Research Education for Undergraduates (CHiMaD REU) Program and NIST-Summer Undergraduate Research Fellowship (SURF) programs and inspiring them to join the field. These activities were complemented by outreach to high-school science educators through ASM Materials Genome Teachers Camp. This chapter, therefore, provides an overview of the CHiMaD Outreach activities in 2018.

16. CHiMaD Supported MGI Seminars and Webinars

All CHiMaD seminar abstracts, presenter biographies and event fliers are available on the CHiMaD Event Archives at http://chimad.northwestern.edu/news-events/Event_Archives.html

02/08/18 - *CHiMaD HQ & Broadcast*
Materials Genome Engineering in China: The Beijing Advanced Innovation Center and the National Key R&D Programs
Dawei Zhang, Yu Yan
Professors, University of Science and Technology, Beijing, China

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02/09/18 - *CHiMaD HQ & Broadcast*
Integrated Computational Materials Engineering at QuesTek Innovations
Jason Sebastian
President, QuesTek Innovations LLC

02/22/18 - *CHiMaD HQ & Broadcast*
What Insights into Additive Manufactured Materials will Exascale Computing Enable?
James Belak
Senior Scientist, Lawrence Livermore National Laboratory

04/11/18 - *CHiMaD HQ & Broadcast*
Engineering Science for Society - with Materials Solutions
Judith Todd
Department Head of Engineering Science and Mechanics, Penn State University

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05/01/18 - *CHiMaD HQ & Broadcast*
From the Efficient Exploration of the High Entropy Alloy Space to Path Planning in Functionally Graded Materials: Alloy Design as a Constraint Satisfaction Problem
Raymundo Arroyave
Professor, Texas A&M University

05/03/18 - *CHiMaD HQ & Broadcast*
NIST Measurement Activities for Materials Performance Under Extreme Conditions

Steven Mates
National Institute of Standards and Technology

05/10/18 - *CHiMaD HQ & Broadcast*
Machine Learning Enabled New Materials Discovery
Taylor Sparks
Assistant Professor, University of Utah

05/14/18 - *CHiMaD HQ & Broadcast*
Multiscale Modelling of Precipitation Hardening in Al-Cu Alloys
Javier Llorca
Professor, Polytechnic University of Madrid

06/25/18 - *CHiMaD HQ*
Designing Materials to Revolutionize and Engineer our Future
John Schlueter
Program Director, DMREF, National Science Foundation

08/20/18 - *CHiMaD HQ & Broadcast*
Connecting Kinetic Monte Carlo Simulations and Experimental Characterization Techniques to Elucidate Processing Structure-Property Relationships in Nanostructured Semiconductor Devices
Michael Heiber
NIST-CHiMaD Postdoctoral Research Fellow, CHiMaD

10/17/18 - *CHiMaD HQ & Broadcast*
A Practical Tutorial for Simple and Reproducible Materials Informatics
Logan Ward
Postdoctoral Researcher, University of Chicago

11/15/18 - *CHiMaD HQ & Broadcast*
Bifurcation Theory of Plasticity, Damage and Failure
Alexander Umantsev
Professor, Fayetteville State University

17. CHiMaD Materials Design Training

Begum Gulsoy (NU), Gregory Olson (NU), Ricardo Komai (QT)

17.1 CHiMaD Materials Design Training Workshops

Background

Designing novel materials of specific properties for a particular application requires simultaneously utilizing physical theory, advanced computational methods and models, materials properties databases and complex calculations. This approach stands in contrast to the traditional trial-and-error method of materials discovery. CHiMaD focuses this approach on the creation of novel hierarchical materials which exploit distinct structural details at various scales, from the atomic on up, to obtain enhanced properties. The center's research focuses on both organic and inorganic advanced materials in fields as diverse as self-assembled biomaterials, smart materials for self-assembled circuit designs, advanced ceramics and metal alloys.

Scope and Format

The CHiMaD Materials Design Training aims to introduce and further the attendees' knowledge of the Materials by Design philosophy and related practical design concepts. During the workshop, small, diverse groups with varying types of expertise are lead through discussions on building System Design Charts while identifying unique design goals and strategies. The hands-on approach allows to showcase the strength of accommodating different types and even levels of expertise for design.

17.1.1 CHiMaD Materials Design Training Workshop at Northwestern

One of the goals of the CHiMaD Materials Design Training Workshop series is to enhance the understanding of Materials by Design concepts by CHiMaD graduate students, post-doctoral researchers and principal investigators, with an emphasis on better understanding how and where their research fits within the design goals of the use case, tool and seed groups they a part of. These workshops also welcome any interested faculty or researchers who are interested learning more about CHiMaD and its design philosophy.

Towards this goal, the first CHiMaD Materials Design Training Workshop of 2018 was organized at Northwestern on March 1, 2018 by **Komai** and **Gulsoy**. During this workshop, 14 CHiMaD student and postdoctoral researchers representing *Precipitation Strengthened Alloys*, *Low-Dimentional Nanoelectronic Materials*, *Polymer Coposites* use-case groups and *Data Mining* tool group had a chance to explore the Materials by Design concepts in a similar hands-on fashion and worked on building existing systems design charts as well as in some cases enhancing those previously built by CHiMaD students and postdoctoral researchers, Figure 17.1. These students and postdocs who are mostly new to the center were joined by those who have previously taken this training in an effort to smoothly transition the materials design and system knowledge within a use-case. Such cases are also a good example of using different levels of expertise in refining the systems design charts.

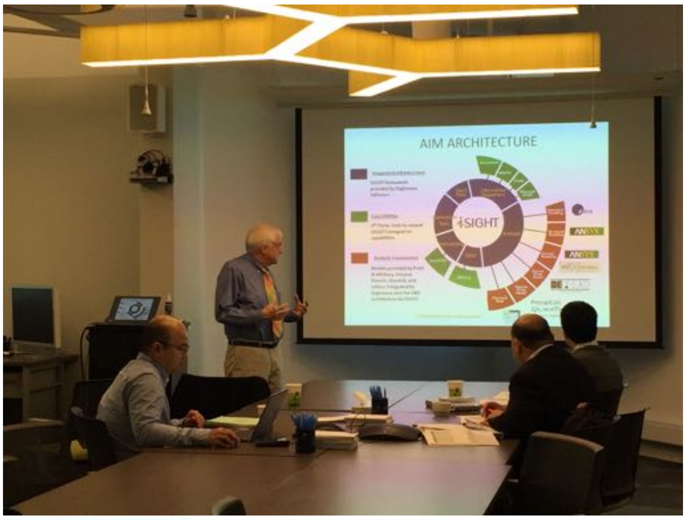
Figure 17.1: *(Top)* CHiMaD Materials Design Training Workshop at Northwestern on March 1, 2018 was attended by CHiMaD students and postdoctoral researchers *(Bottom)* The training provides a hands-on experience to attendees through small working groups focusing on different material systems.

A follow-up CHiMaD Materials Design Training Fall Workshop was organized at Northwestern on November 27, 2018 by **Komai**, **Gulsoy** and **Houser** building on the material from earlier in the year. The specific focus of this workshop was to refine the previously built *System Design Charts* as well as introduce the concept of *Calculation Flow Block Diagrams*. During the workshop, members of the **Lauhon** research group, representing the *Low-Dimensional Nanoelectronic Materials* use-case group built a visual representation of their research methodology through *Calculation Flow Block Diagrams* with an aim to incorporate it into their everyday research decisions.

17.1.2 CHiMaD Materials Design Training Workshop at University of Chicago

To address the logistics of being a multi-institutional Center, a following CHiMaD Materials Design Workshop was organized at University of Chicago on March 2, 2018 by **Komai** and **Gulsoy**. During this workshop, 7 CHiMaD student and postdoctoral researchers representing *Soft Matter Design using Charge Complexation*, *Directed Self Assembly of Block Copolymers for Lithographic Applications* and *Uncertainty Quantification in Computational Thermodynamics* use-case and seed groups, had a chance to explore the Materials by Design concepts in a similar hands-on fashion and worked on building existing systems design charts as well as in some cases enhancing those previously built by CHiMaD students and postdoctoral researchers, Figure 17.2. These students and postdocs who are mostly new to the center were joined by those who have previously taken this training in an effort to smoothly transition the materials design and system knowledge within a use-case.

Figure 17.2: *(Top)* CHiMaD Materials Design Training Workshop at University of Chicago on March 2, 2018 was attended by CHiMaD students and postdoctoral researchers *(Bottom)* The training provides a hands-on experience to attendees through small working groups focusing on different material systems.



Meetings	SRG	NU-NIMS	
Workshops	Materials Design	Gear Steels	AIM Methodology
Customized Labs	NU-MAT 390: Materials Design (2 weeks)		

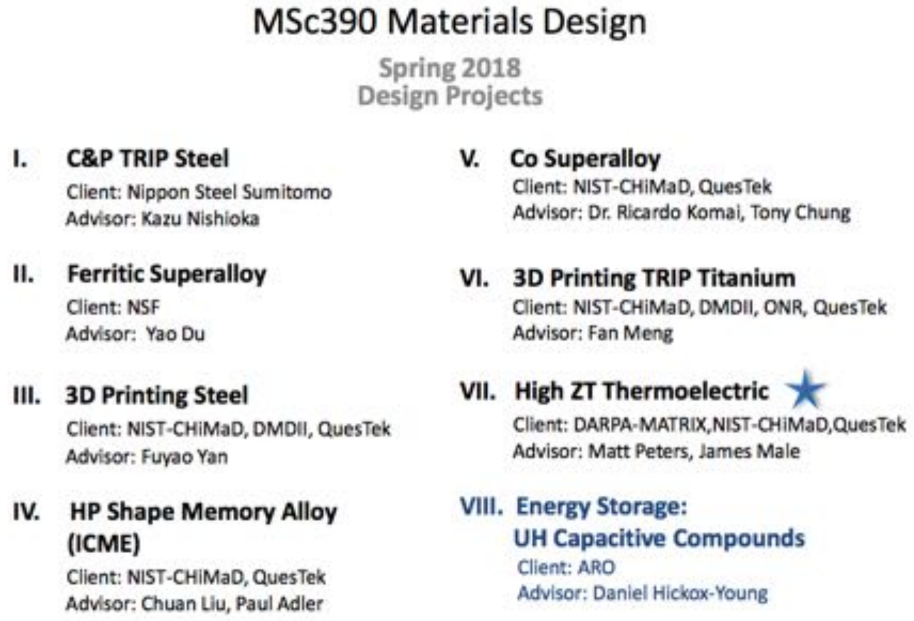
Figure 17.3: The pilot CHiMaD Materials Design Training for Industry was held for ITOCHU Techno-Solutions Corporation of Japan as a 3-week program incorporating material-specific workshops as well as CHiMaD Meetings into the training.

17.1.3 CHiMaD Materials Design Training Workshops for Industry - Pilot

In 2018, CHiMaD lead the organization of the pilot CHiMaD Materials Design Training for Industry between March 26-April 13, 2018 with **Olson**, **Komai**, **Gulsoy** as intructors. A three-week training program was developed for ITOCHU Techno-Solutions Corporation of Japan incorporating material-specific workshops as well as CHiMaD Meetings into the training. During the duration of the training, the industrial researchers attended three workshops, content of which was designed with their topics of interest: (1) CHiMaD Materials Design Training; and (2) Gear Steel Workshop (3) AIM Methodology Workshop, Figure 17.3. They also attended two CHiMaD meetings, learning about design advancements for a broader range of materials: (1) Annual SRG Meeting; (2) NU-NIMS Materials Genome Workshop. To complement the presented knowledge, they also were lead through hands-on materials design labs using ThermoCalc software. Future trainings are planned for 2019 with other industrial clients.

17.1.4 CHiMaD Materials Design Training Workshop with Hero-m, Sweden

CHiMaD, QuesTek Europe AB and QuesTek Innovations, in collaboration, continued to develop the Materials Design Workshop series for a special workshop at KTH Royal Institute of Technology (KTH) in Stockholm, Sweden at the Hero-m center for design. On November 13-14, 2018, KTH's Hero-m Center, CHiMaD, QuesTek Innovations and QuesTek Europe jointly lead the organization of a Materials Design Workshop in Stockholm, Sweden to demonstrate the use of ICME methods for materials design and provide hands on experience for attendees. The workshop aimed to accelerate the design process of the four design projects in Hero-m with the exclusive attendance of members and scientific advisory board of the Hero-m Center, as well as to further the collaboration and synergy between Hero-m and CHiMaD. The workshop focused on generating a *system design chart*



MSc390 Materials Design

Spring 2018
Design Projects

I. **C&P TRIP Steel**
Client: Nippon Steel Sumitomo
Advisor: Kazu Nishioka

II. **Ferritic Superalloy**
Client: NSF
Advisor: Yao Du

III. **3D Printing Steel**
Client: NIST-CHiMaD, DMDII, QuesTek
Advisor: Fuyao Yan

IV. **HP Shape Memory Alloy (ICME)**
Client: NIST-CHiMaD, QuesTek
Advisor: Chuan Liu, Paul Adler

V. **Co Superalloy**
Client: NIST-CHiMaD, QuesTek
Advisor: Dr. Ricardo Komai, Tony Chung

VI. **3D Printing TRIP Titanium**
Client: NIST-CHiMaD, DMDII, ONR, QuesTek
Advisor: Fan Meng

VII. **High ZT Thermoelectric** ★
Client: DARPA-MATRIX,NIST-CHiMaD,QuesTek
Advisor: Matt Peters, James Male

VIII. **Energy Storage: UH Capacitive Compounds**
Client: ARO
Advisor: Daniel Hickox-Young

Figure 17.4: The pilot CHiMaD Materials Design Training for Industry was held for ITOCHU Techno-Solutions Corporation of Japan as a 3-week program incorporating material-specific workshops as well as CHiMaD Meetings into the training.

for each project while learning from experienced instructors from QuesTek and CHiMaD. This workshop also acted as a "training the trainers" opportunity for CHiMaD; Clay Houser (Olson Group, NU) lead the discussion on Calculation Flow Block Diagrams as a first-time instructor. He will be taking a leadership position in future CHiMaD Materials Design Training workshops, starting in February 2019.

17.2 MSc 390: Materials Design Course at Northwestern

Materials Design has historically been a part of the core curriculum at Northwestern University. MSc: 390 Materials Design course offered by Gregory **Olson** every year builds teams of undergraduate and graduate students focusing on materials design problems for real engineering applications, lead by PhD students and professionals. In the past, the course hosted international students, NIST postdoctoral fellows, and industry professionals as team coaches alongside the PhD students and Research professionals at Northwestern.

In 2018, SRG and CHiMaD research projects provided again a hierarchical project coaching system where graduate students serve as technical advisors to the class project teams as summarized in Figure 17.4. Projects supported by CHiMaD research include the Co superalloy and shape memory alloy, as well as steel and Titanium alloy designs constrained for 3D printing. Building on our move into electronic materials represented by the High ZT Thermoelectric materials project and the broader array of design methodologies under CHiMaD development, highlighted in blue is a new project in collaboration with James Rondinelli (NU), applying novel data mining strategies to directed compound discovery for Ultrahigh-Capacitance materials.

Highlighted by the star in Figure 17.4, CHiMaD graduate student Matt Peters (Voorhees Group, NU) coached a team of undergraduate students applying his databases in the integrated CALPHAD-based design of a chalcogenide thermoelectric with optimized microstructure based on the PbTe-SrTe-PbTe system. The project placed 3^{rd} in the 2018 ASM Undergraduate Design competition.

18. Workshops and Meetings

The agendas for CHiMaD-organized and supported workshops are available on the CHiMaD Event Archives at http://chimad.northwestern.edu/news-events/Event_Archives.html

18.1 CHiMaD Phase Field Methods Workshop Series

January 20-22, 2018 & September 25-27, 2018 Organizers: Olle ***Heinonen*** *(ANL), Peter* ***Voorhees*** *(NU), James* ***Warren*** *(NIST), Jonathan* ***Guyer*** *(NIST)*

Scope
The goal of the CHiMaD Phase Field Methods Workshop series is to develop benchmark problems for the global phase field community and to engage this elite team of invited Phase Field experts and framework developers for in providing input during this development.

The Phase Field tool group, along with its NIST collaborators, lead the organization of two CHiMaD Phase Field Methods Workshops in February and September of 2018, making it 7 workshops organized since the inception of the series in 2015, Figure 18.1. The first workshop focused on Benchmark problems in Method of Manufactured Solutions, led by Steven DeWitt (University of Michigan), and in coupled Cahn-Hilliard-electrostatic problem, led by Andrea Jokisaari (Idaho National Laboratory). The workshop also featured other presentations on GPU-based Phase Field modeling and Uncertainty Quantification, the latter led by the respective CHiMaD seed group. This workshop was attended by 22 researchers. The second workshop focused on nucleation modeling in Phase Field, and featured lectures by Tamas Pusztai and Laszlo Granasy (Wigner Research Centre for Physics, Hungary) on the subject, as well as presentations by Steven DeWitt (University of Michigan), Andrea Jokisaari, and Larry Aagesen (Idaho National Laboratory). At the workshop, revisions to the coupled Cahn-Hilliard-electrostatic Benchmark problem were also discussed, and participants presented solutions to a version sent out in advance to the workshop.This workshop was attended by 38 researchers, making it the highest attended workshop to date in the series.

There is ongoing work to plan the upcoming workshop to be held in April of 2019

Figure 18.1: *(Top)* 6^{th} (January 20-22, 2018) and *(Bottom)* 7^{th} (September 25-27, 2018) CHiMaD Phase Field Methods Workshops were attended by phase field experts from around the world

which will focus on nucleation. For more information on CHiMaD Phase Field Method Workshops and the organizing tool group, please refer to Section 11 of this report.

18.2 Advanced Polymer Characterization Workshop by TOSOH Bioscience

January 23, 2018 | Organizers: Jeffrey Ting (CHiMaD), Begum ***Gulsoy*** *(NU)*

Scope

Bring together academic and local industrial parties in enhancing their knowledge of experimental methodology

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GPC experts from TOSOH Bioscience lead the Advanced Polymer Characterization Workshop for 15 attendees from academia and local industry. Topics covered included: (1) Fundamentals of Triple Detection in Both Ambient and High Temperature GPC Analysis; (2) GPC Method Development for Challenging Applications and; (3) Introduction to Tosoh Bioscience and EcoSEC GPC Systems. This workshop was recommended and co-organized by NIST-CHiMaD Postdoctoral Research Fellow, Jeffrey Ting.

18.3 34th Annual SRG Meeting

March 26-27, 2018 | Organizer: Gregory ***Olson***

Scope

Northwestern's SRG materials design consortium, founded in 1985, joined the CHiMaD umbrella organization during Year One, offering the leveraging opportunity of the CALPHAD-based metals design projects with topics and sponsors listed in Figure 18.2.

The ONR CyberAlloys project applies CALPHAD-based design to transformation toughened alloys for blast and fragment protection in both Ti-based and Fe-based systems integrating all-electron FLAPW DFT predictions of interfacial properties important to ductile fracture toughness. Efforts in 2018 have included adaptation of both alloy systems to 3D printing. In its final year, the collaboration with GM under the DOE Lightweighting Initiative has quantified the thermodynamics and kinetics of Q phase precipitation strengthening, summarized in Acta Mat publications, and applied it in design of cast Al alloys with higher temperature performance, now under further development at GM. Caterpillar collaboration under the same DOE initiative adapted multiphase precipitation concepts from our previous martensitic blast protection steel designs to demonstrate novel cast steel prototypes for lower cost crankshaft applications. Also completed in the past year, AHSS design research supported by ArcelorMittal has quantified the nonequilibrium thermodynamics of carbon partitioning in Quench & Partition martensite-austenite TRIP steels for high formability automotive sheet applications. In support of the new technology of additive manufacturing, the NIST-MSAM project completed in 2016 quantified recrystallization behavior of Ti and steel alloys, enabling a new project under the Chicago-based DMDII Manufacturing Institute which has applied the results in rapid qualification strategies demonstrated with QuesTek's Ferrium PH48S stainless maraging steel. QuesTek's collaboration with Honeywell under the DARPA Open Manufacturing initiative completed a 6-year 3-phase effort demonstrating AIM qualification of printed Ni

2018 SRG Design Projects

- **ONR CyberAlloys** (Olson, Kontsevoi)
  - CMD of Fe & Ti alloys for blast and fragment protection
- **DOE/GM Lightweighting Initiative** (Olson, Wolverton, Voorhees)
  - CMD of cast aluminum for cylinder heads
- **ArcelorMittal AHSS** (Olson)
  - CMD of high-strength automotive Q&P TRIP steels

DMDII Rapid Qualification for Additive Manufacturing (Olson, Liu, Cao)

-ICME AIM modeling of PH48S stainless maraging steel

DARPA/Honeywell Open Manufacturing (QuesTek)

- ICME for SLM additive manufacturing of Ni 718+

DARPA-SIMPLEX: Data-Driven Discovery for Designed Thermoelectric Materials (QuesTek/CHiMaD; Olson, Voorhees, Snyder)

DARPA-MATRIX: Designed Thermoelectric Materials for Transduction Devices (QuesTek/CHiMaD; Olson, Snyder)

CHiMaD

Figure 18.2: *2018 SRG Design Projects*

superalloy components for aerospace applications, using data from 3 lots of alloy powder to efficiently forecast minimum properties.

The SRG Annual Meeting held each March at Northwestern continues to serve as a review of CHiMaD metals activities offering interaction with a broader community. Since coming under the CHiMaD umbrella, the meeting is broadcast via *Bluejeans* providing colleagues who cannot attend on-site an opportunity to attend virtually. In 2018, 112 attendees registered to attend the meeting on-site, while 26 registered to attend virtually, with industrial representatives from over 20 companies. A total of 27 presentations were delivered during the two-day meeting. In support of NIST collaboration, NIST speakers at the 2018 SRG meeting included **Campbell** on database development and **Gilman** on carbon fiber reinforced polymer (CFRP) design.

For more information, please refer to chapters 6 and 17.

18.4 6th NU-NIMS Materials Genome Workshop

March 28, 2018 | Organizer: Gregory ***Olson****, Koichi Tsuchiya (NIMS), Ikumu Watanabe (NIMS)*

Scope

Providing an overview of efforts at Northwestern and National Institute of Materials Science (NIMS) in Japan, with an emphasis on building enahanced collaborations between the two institutions

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Building on the NIST-sponsored US/Japan Materials Genome Workshop held in Tsukuba in June 2015, the 2018 SRG meeting at Northwestern was followed by the 6th NU-NIMS Materials Genome Workshop. There were 19 presentations were delivered during the day-long meeting, welcoming 25 attendees including Japan and Sweden, Figure 18.5. Inspired by the scope of the CHiMaD activities, the workshop emphasized new initiatives in materials informatics. Plans for enhanced collaboration between NU and NIMS continue, with the Materials Genome Workshop now an annual event.

Figure 18.3: *6th NU-NIMS Materials Genome Workshop* was attended by an international group of researchers representing USA, Japan and Sweden.

18.5 CHiMaD Annual Review Meeting

August 16-17, 2018 | Organizer: CHiMaD Leadership

Scope

The The CHiMaD Annual Review Meeting is the technical overview of the Center for Hierarchical Materials Design. It annually brings together CHiMaD principal investigators, postdoctoral researchers and students from Northwestern University, University of Chicago, Argonne National Laboratory as well as partners from QuesTek Innovations and ASM International along with collaborators from National Institute of Standards and Technology. This meeting also acts as a platform for the CHiMaD Technical Advisory Board to review and provide feedback on the CHiMaD activities and for industrial partners to explore the capabilities of and possible partnerships with CHiMaD.

The CHiMaD Annual Meeting is attended by CHiMaD researchers, postdocs, students, NIST collaborators, Technical Advisory Board members, CHiMaD's industrial partners, and prospective industrial companies. The attendance at the meeting has been consistently over 100 attendees. A poster session is also organized as a part of this meeting to allow for collaborators and industry professionals to connect with CHiMaD students and postdocs. In 2018, the CHiMaD Annual Meeting was held as a review meeting towards the renewal of the Center, Figure 18.4.

Figure 18.4: In 2018, the CHiMaD Annual Meeting was held as a review meeting towards the renewal of the Center.

18.6 Thermoelectrics Seminar

August 2, 2018 | Organizer: Jeffrey Snyder

Scope

Thermoelectric materials could play a key role in meeting increasing global energy needs, but are currently limited due to their low efficiency. Current research is dedicated to investigating methods for improving their overall efficiency.

This internationally attended seminar organized by Jeffrey Snyder (NU) compliments CHiMaD efforts of the Thermoelectric Materials Seed Group. The seed group members were present for the discussion.

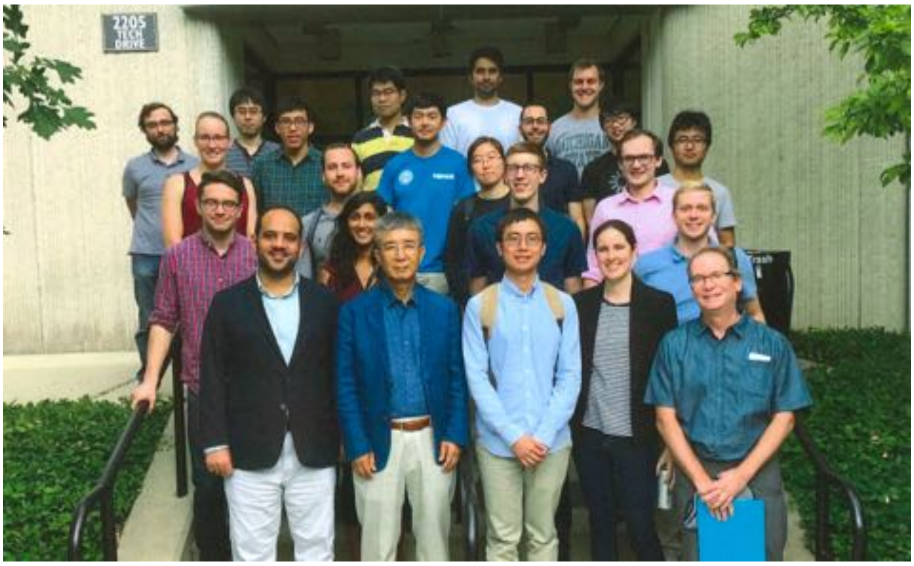
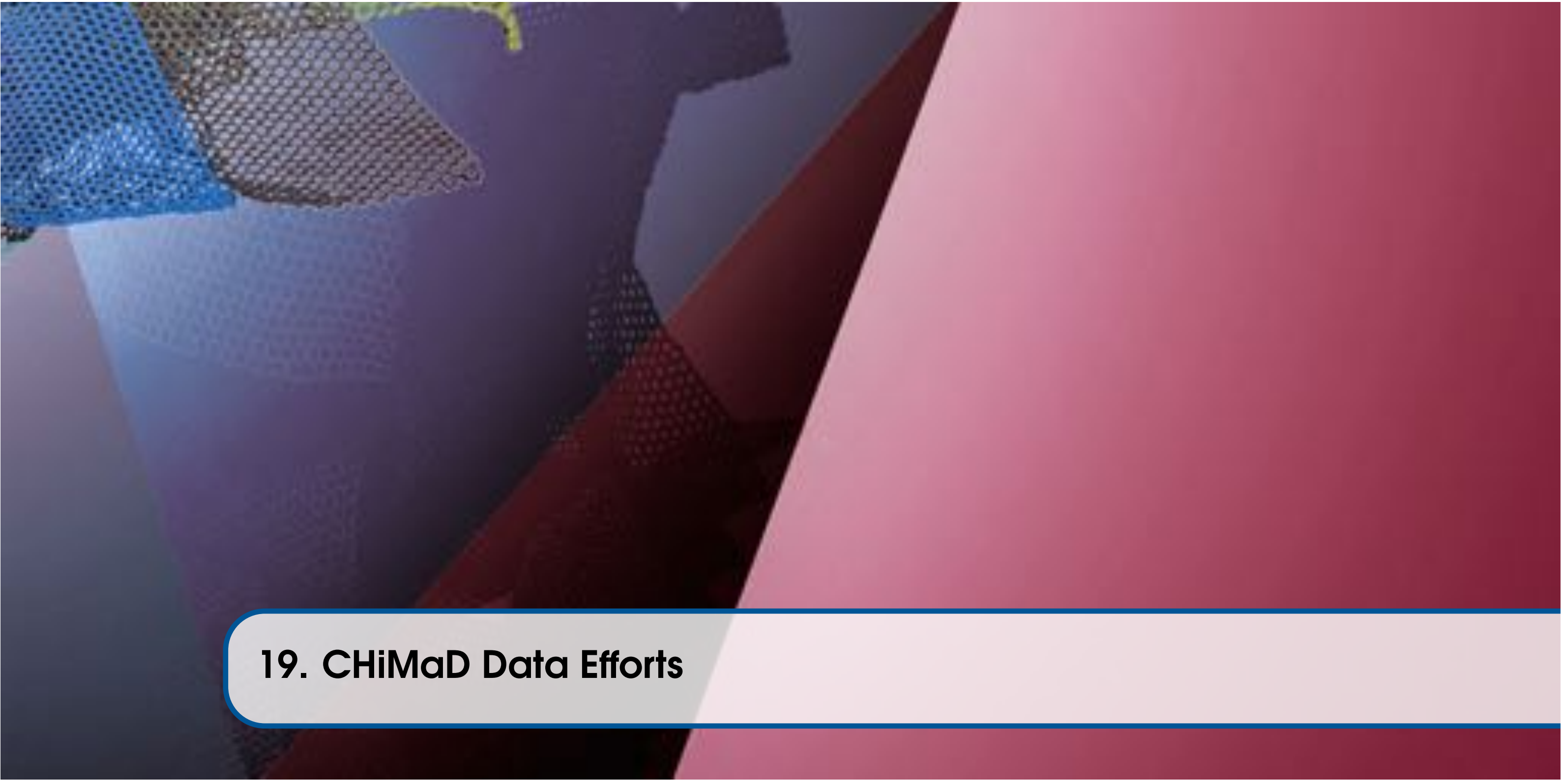


Figure 18.5: In 2018, the CHiMaD Annual Meeting was held as a review meeting towards the renewal of the Center.



19. CHiMaD Data Efforts

Laura Bartolo (NU), Begum Gulsoy (NU), Ian Foster (UC), Ben Blasizik (UC), David Seidman (NU), Juan de Pablo (UC)

James Warren (NIST), Carelyn Campbell (NIST), Zachary Trautt (NIST), June Lau (NIST), John Henry Scott (NIST), Robert Hanisch (NIST), Chandler Becker (NIST), Frederick Phelan (NIST), Debbie Audus (NIST), Ursula Kattner (NIST), Kenneth Kroenlein (NIST), Shengyen Li (NIST), Raymond Plante (NIST), Alden Dima (NIST), Sharief Youssef (NIST)

Significance

Digital data and a materials innovation infrastructure are foundational components of the Materials Genome Initiative for accelerating discovery of new materials and reducing costs of advanced manufacturing. NIST plays a central role in the Federal government on the establishment of the materials innovation infrastructure to enable effective data exchange; best practices for data quality; and analytics for data-driven materials science. As the NIST Center of Excellence for Hierarchical Materials Design, CHiMaD concentrates its data efforts on collaboration, contribution and implementation to data exchange, data management practices, and data-driven materials science analytics to meet the goals of the Center and NIST.

19.1 Goals

Effective data tools, best practices, and standards are required for CHiMaD and others in the materials community to capitalize on the benefits of a robust materials innovation infrastructure to reach the goals of MGI. To meet this need, CHiMaD works in concert with NIST and others in the materials community on development and adoption of tools, best practices, and standards. During the year of 2018 CHiMaD has continued to focus its data efforts for data management integration into scientific practices in two primary areas: (1) workshops and training sessions with the CHiMaD Use Groups to increase awareness and knowledge of the current materials data landscape; and (2) collaborations across the

Figure 19.1: NIST-CHiMaD Materials Microscopy Data Conference was attended by participants from academia, government labs, industry.

Center, with NIST, and with others in the materials community to increase the potential for impact and adoption of promising advances.

To support data tool development, best practices, and standards, CHiMaD has held multiple workshops and training activities as well as been invited to contribute to data efforts in the materials community over the past year.

19.2 2018 NIST-CHiMaD Materials Microscopy Data Conference

On October 25-26, 2018 CHiMaD hosted the *NIST-CHiMaD Materials Microscopy Data Conference* at CHiMaD Headquarters, Figure 19.1. Organized by representatives of NIST, Carelyn **Campbell**, John Henry **Scott**, June **Lau** and Zachary **Trautt**, and CHiMaD, Begum **Gulsoy**, Ben **Blaiszik**, Ian **Foster**, and Laura **Bartolo**, as well as Industry, Marcus Hanwell (KitWare), William Harris (Zeiss Microscopy) the aim of the 1.5 day workshop was to galvanize community actions to address opportunities, gaps, and implementation of evolving best practices through forward-looking *Plenary Talk, Rapid Talks* and *Small Group sessions* organized around instrument data workflow.

Small Group session Focus:

- **Sample History**: Outline important sample history (generation & processing) and sample preparation information to be included. (*Leader; Carelyn* ***Campbell***)
- **Metadata**: Define key concepts for materials microscopy data and the relationships between the concepts. (*Leader; John Henry* ***Scott***)

- **Workflow**: Identify easy materials data capture for two different types of microscopy instruments. (*Leader; June* ***Lau***)

Workshop participants included 36 attendees from academia, government labs, industry. Industrial attendees included representatives from Zeiss Microscopy, Kitware, Hummingbird Scientific, Jeol USA, Citrine Informatics. A representative from each of the Big Data Spoke IMaD partners and from the Midwest Big Data Hub were also present. Follow up conference calls are being scheduled to take place in March and April 2019 to continue the discussions of the Sample working group. The second Workshop in the series will be held at NIST CCoE (Rockville, MD) on May 15-16, 2019.

Figure 19.2: Survey results from 2018 CHiMaD hosted the NIST-CHiMaD Materials Microscopy Data Conference.

Online resources from the 2018 Workshop:

- **Workshop agenda and presentations:**
  `http://chimad.northwestern.edu/news-events/CHiMaD_Data_Database_ Efforts.html`
- **Short video on the Workshop:**
  `https://northwestern.box.com/s/gi15cwrwlqoj6xef33tir3a70zvw7ijk`
- **Short video on the Workshop small group sessions:**
  Sample History, Metadata, and Workflow
  `https://northwestern.box.com/s/a45mibkc1uq9aywksiuot904dycwknf7`

A survey results collected from 27 of 36 workshop participants revealed a successful workshop; Figure 19.2

19.3 Best Practices: NIST/CHiMaD Prototype Polymer Data Project

The *Polymer Data Project* aims to establish a prototype interoperable infrastructure for Polymer Data where all can connect together in a pilot project including researchers, databases, journals, and tools to publish, search and analyze. The planning group, lead by CHiMaD, includes representatives from American Chemical Society, Argonne National Laboratory, Citrine Informatics, NIST, Northwestern University, University of Chicago, University of Delaware, Royal Society of Chemistry, Solvay, Yale University and University of Vermont. The project involves CHiMaD's Polymer Predictive Properties and

Database (PPPDB) hosted by University of Chicago and NanoMine, currently lead by Brinson at Duke University, along with two major publishers and a subset of their polymer journals. Towards fulfilling the scope of the Polymer Data Project, **American Chemical Society**, and its *Macromolecules* and *ACS Macro Letters* journals are hosting a virtual issue with the theme collection title, "Structure Property Processing Relationships in Polymer Nanocomposites" with guest editors, Cate Brinson (Duke University) and Linda Schadler (University of Vermont). **The Royal Society of Chemistry** and its *Molecular Systems Design & Engineering* is hosting a virtual issue with the theme collection title: "Charge Transporting Nanostructured Polymers for Electrochemical Systems", with guest editors, Shrayesh Patel (IME, Chicago) and Nitash Balsara (UC Berkeley). The deadline to submit to articles and accompanying data was December 2018. In NanoMine (ACS) and Polymer Property Predictor and Database/Materials Data Facility (RSC), authors were encouraged to submit data into their data repositories into by using:

- Customizable templates with support from the repositories
- NIST Materials Science/Polymer Core High Level descriptors
- Persistent Identifiers for submitted data (Handles as temporary IDs during review and DOIs as permanent IDs after acceptance of paper)

Based on the efforts of the Polymer Data Project, CHiMaD participants (Debra **Audus**, NIST; Laura **Bartolo**, CHiMaD/NU; Shrayesh Patel, UC) have been invited to take part in an NSF FAIR (Findable, Accessible, Interoperable, Reuseable) Hackathon to be held February 2019 in Washington.

19.4 Contributions to NIST and Other Community Data Efforts

CHiMaD's Data Efforts were invited to participate and present in numerous materials data events throughout 2018 which are briefly summarized below:

- *High-Throughput Experimental Materials Collaboratory* (HTE-MC), 2/28/18 - 3/2/18 was held at NIST for presentations and discussions involving the deployment of a federated network of high-throughput experimental (synthesis and characterization) tools, which are integrated with a materials data infrastructure.
- *Configurable Data Curation System Annual Convention* (CDCS Con), 7/30-31/18 aimed to improve collaboration and communication within the materials community to share solutions developed outside of NIST as well as to roadmap future efforts at NIST with regard to materials data infrastructure.
- *Glass/Ceramics Data Science Workshop*, 2/6/18 was sponsored by ACerS and held at Nexight Group Office to build a community of data people and ceramic and glass people for building tools, vocabulary, and network to effectively work together.
- *Machine Learning in Science and Engineering (Materials Track)*, 6/6-8/18 was sponsored by Carnegie Mellon University and Georgia Tech to bring together leaders from a variety of disciplines to present leading-edge research on applications of machine learning on science and industry.
- CHiMaD was nominated and elected to become an Advisory Board member of the NSF sponsored OpenKIM (`https://openkim.org`), represented by Laura **Bartolo**.

19.5 NSF Midwest Big Data Spoke on Integrative Materials Design (IMaD)

CHiMaD leads the NSF Big Data project, IMaD, under the direction of PIs Ian **Foster** and Ben **Blaiszik** (UC). As part of the CHiMaD Data Efforts, collaborations with the Midwest Big Data Hub (MBDH) involved the participation of MBDH in the NIST/CHiMaD Materials Microscopy Data Workshop as well as contributions by CHiMaD of data resources and science communication to MBDH. Through the IMaD/NU Science Communication Intern, a series of short videos have been produced and distributed on the IMaD Partners' research and impacts on materials development: CHiMaD, University of Illinois at Urbana Champagne (UIUC), University of Michigan, and the University of Wisconsin:

- CHiMaD / NUCAPT: Northwestern Center for Atom Probe Tomography
  `https://northwestern.box.com/s/npdck14h0wg5o84l9gvxjyco0dazt94y`
- University of Illinois at Urbana Champagne / 4CEED
  `https://northwestern.box.com/s/4d29ka8sm5lyrfcin2aoal5jxvmnmpnq`
- University of Michigan / PRISMS' Materials Commons
  `https://northwestern.box.com/s/enfu2swuvu98kfpo98vogsipmmkq40uy`

19.6 Impact Statement for CHiMaD Phase I

During Y1-5, CHiMaD has established itself as a recognized community contributor and collaborator on the promotion, adoption, and implementation of materials related data tool development, best practices, and standards. Its workshops and training efforts have been well received and well attended. It has also established a customary pattern to work with other recognized materials science leaders in co-hosting materials data events to maximize the reach and impact in the materials community. Through its individual and joint efforts, CHiMaD continues to add its expertise and commitment to promote scientific discovery through open and FAIR data processing and to serve as a prototype research center model for integrating data management into scientific practice.

20. Educational Outreach with ASM International

Begum Gulsoy (NU), Gregory Olson (NU), Ricardo Komai (QT)

Larry Berardinis (ASM), Scott Henry (ASM)

20.1 ASM Materials Genome Toolkit

In 2015, ASM International and its affiliate members administered a competition among undergraduate universities in the U.S., awarding six schools with state-of-the-art materials design software and related materials property databases. The competition was announced in early 2015 and broadly publicized via direct email, on the ASM and CMD Network websites, through several ASM newsletters, and in a press release issued to various news outlets and materials-oriented professional societies. Schools competed on the basis of a proposal stating how they planned to integrate the Materials Genome Toolkit software into their undergraduate curriculum and in the context of the annual ASM Undergraduate Design Competition. A review committee convened by the ASM Action in Education Committee, the subcommittee on Computational Materials Engineering, evaluated the entries, ranking them according to several factors, including the degree to which the proposed plan balanced theory and application in the use of the software.

To date, 18 undergraduate-serving engineering institutions have received the CHiMaD-sponsored three-year, multi-user license, giving hundreds of students and faculty across the U.S. access to the latest Thermo-Calc thermodynamics code, DICTRA multi-component diffusion code, TC-PRISMA precipitation simulator, and several thermodynamic and mobility databases. In 2018, four MGI Toolkits were awarded to successful proposals by Worcester Polytechnic Institute, Louisiana Technological University, Montana Technological University and Georgia Institute of Technology. Since 2017, an expanded interest by the broader materials community resulted in the awarding of successful proposals from both Materials Science and Mechanical Engineering departments. Figures 20.1 and 20.2 provide a complete list of winning schools and the lead principal investigators of the proposals between 2015-2018. The official 2018 press-release by ASM can be accessed through the following link: `https://www.asminternational.org/news/industry/-/journal_content/56/10192/36303247/NEWS`.

ASM MGI Toolkit Awardees 2016-2018
Worcester Polytechnic Institute (*2018 Awardee*) *Lead PI: Danielle Cote, Department of Materials Science and Engineering*
Montana Technological University (*2018 Awardee*) *Lead PI: Jerome Downey, Metallurgical and Materials Engineering Department*
Louisiana Technological University (*2018 Awardee*) *Lead PI: Kasra Momeni, Department of Mechanical Engineering*
Georgia Institute of Technology (*2018 Awardee*) *Lead PI: Rampi Ramprasad, Department of Materials Science and Engineering*
University of Pittsburgh (*2017 Awardee*) *Lead PI: Wei Xiong, Department of Materials Science and Engineering*
Wright State University (*2017 Awardee*) *Lead PI: Raghavan Srinivasan, Department of Mechanical and Materials Engineering*
University of Florida (*2017 Awardee*) *Lead PI: Michele Manuel, Department of Materials Science and Engineering*
Missouri University of Science & Technology (*2016 Awardee*) *Lead PI: Joe Newkirk, Department of Metallurgical and Materials Engineering* 3^{rd} Place, 2016 ASM Undergraduate Design Competition
Case Western Reserve University (*2016 Awardee*) *Lead PI: David Matthiesen, Department of Materials Science and Engineering*
Oregon State University (*2016 Awardee*) *Lead PI: Julie D. Tucker, Department of Mechanical, Industrial and Manufacturing Engineering*

Figure 20.1: List of ASM MGI Toolkit Awardees 2016-2018. The awarded schools regularly take top three spots in the ASM Undergraduate Design Competition since the inception of the award.

ASM MGI Toolkit Awardees - Renewal Program
California State Polytechnic University in Pomona (*2015 Awardee*) *Lead PI: Vilupanur Ravi, Chemical and Materials Engineering* 1st Place, 2015 ASM Undergraduate Design Competition 1st Place, 2016 ASM Undergraduate Design Competition
Carnegie Mellon University (*2015 Awardee*) *Lead PI: Bryan Webler, Department of Materials Science and Engineering* 2nd Place, 2015 ASM Undergraduate Design Competition
Michigan Technological Institute (*Pilot Program*) *Lead PI: Paul Sanders, Department of Materials Science and Engineering* 3rd Place, 2015 ASM Undergraduate Design Competition 3rd Place, 2017 ASM Undergraduate Design Competition
Alfred University (*2015 Awardee*) *Lead PI: S. K. Sundaram, Department of Materials Science and Engineering*
University of Maryland (*2015 Awardee*) *Lead PI: Yifei Mo, Department of Materials Science and Engineering*
Virginia Polytechnic Institute and State University (*2015 Awardee*) *Lead PI: Alan Druschitz, Department of Materials Science and Engineering* 1st Place, 2017 ASM Undergraduate Design Competition
Colorado School of Mines (*2015 Awardee*) **(*Awarded through LIFT*)** *Lead PI: Kip Findley, Department of Metallurgical and Materials Engineering*
Fayetteville State University (*2015 Awardee*) **(*Awarded through CHiMaD*)** *Lead PI: Alexander Umantsev, Department of Physics*

Figure 20.2: The licences of the ASM Toolkit 2015 Awardees were up for renewal in 2018

The CHiMaD-sponsored ASM Materials Genome Toolkit program is also made possible by Thermo-Calc Software, which generously offered an initial 85% discount on what will ultimately be twelve sets of tools worth more than \$1.6 million. The software distribution program addresses several CHiMaD objectives by helping the next generation of scientists and engineers become proficient with materials-by-design tools and techniques and by contributing to efforts that are advancing the collective knowledge on material systems and methodologies.

In 2018, as part of a continued partnership, Thermo-Calc Software, ASM and CHiMaD have also facilitated a program to incentivise the *renewal* of the expiring toolkit licenses. On 06/30/18, the MGI toolkit licenses of the 2015 awardees was set to expires. Under the new program, the six universities were offered a 50% match towards the discounted cost of extending their licenses for another 3-years. To date, four of the six schools have taken advantage of this new incentive and renewed their toolkit licences: Michigan Technological Institute, Virginia Technological Institute, Carnegie Mellon University and California State Polytechnic University in Pomona.

In 2019, ASM will continue to conduct the Materials Genome Toolkit competition with an aim to award up to three more undergraduate-serving engineering schools with state-of-the-art materials design software; and the renewal program with an aim to keep the tools integrated into the U.S. undergraduate engineering cirriculum. ASM is actively promoting the contest to faculty and students through email, newsletters, web posts, community forums, AM&P magazine, and a broadcast press release.

20.2 ASM Materials Genome Camp for High-School Educators

To achieve a higher-impact outreach to high school students as well as underrepresented minorities, CHiMaD has partnered with ASM International and QuesTek Innovations to host the first ASM Materials Genome Camp for High School Educators in the summer of 2017. The three-day pilot camp hosted 20 educators from six U.S. states and Canada. Upon its success, the camp was converted to a full-time (five-day) camp for the summer of 2018 by ASM. A U.S.-standard teaching material set is in preparation by volunteer educators from the first camp along with a publication in preparation by the organizers.

In 2018, the ASM Materials Genome Camp for High School Educators was attended by 8 educators from 3 different states and Canada, Figure 20.3. The Camp was lead by **Komai** who served as an organizer and a lead instructor. Together with **Gulsoy**, **Olson** and Clay Houser (NU), and Jonathan Emery (NU), an extended course developed was for teachers using the design of a self-healing metal matrix "Frankensteel" composite, Figure 20.4. During the five-day Camp, the educators designed the alloy composition for "Frankensteel" through experiment and computational methods (ThermoCalc), prepared the samples, tested mechanical properties of the sample and observed self-healing overnight. This year, the Camp also served as an opportunity to train the trainers as Clay Houser (NU) and Jonathan Emery (NU) are expected to become instructors for various CHiMaD outreach activities in 2019.

Figure 20.3: 2018 ASM Materials Genome Camp for High School Educators

Figure 20.4: CHiMaD-lead ASM Materials Genome Camp for High School Educators puts an emphasis on providing educators with a hands-on experience of the course material.

21. Outreach to Under Represented Groups

Begum Gulsoy (NU), Paul Nealey (UC), Juan de Pablo (UC), Ricardo Komai (QT)

Carelyn Campbell (NIST), James Warren (NIST)

21.1 Partnership with The University of Chicago Collegiate Scholars Program

In partnership with the University of Chicago Collegiate Scholars Program that aims to prepare Chicago Public Schools students in grades 10-12 for admission and success at colleges and universities, CHiMaD launched the *Molecular Engineering* course in summer 2016, the first engineering course offering since the program was founded in 2003. The 6-week engineering course is designed to illustrate the concepts and principles of polymeric, composite, and nanoscale materials and demonstrate the relationship between molecular structures and material properties in the context of real-world applications, with an emphasis on materials design. Students learn the fundamentals of materials science and molecular engineering, including material types and properties, design of materials and experiments, and material characterization and analysis, through lectures, hands-on experience, reading assignments, and technical communication practice, Figure 21.1. Techniques for measuring material properties and monitoring experimental processes are integrated throughout the course by use of a variety of laboratory instruments and tools. The students have shown great interest and enthusiasm in participating in the lab experiments and discussions. They have learned the key elements of a comprehensive material design process and important material systems, including polymers, composite materials, nanomaterials and semiconductors, in a stimulating and constructive environment. In 2018 different modules were taught by Paul **Nealey**, Juan **de Pablo** and Kazi Sadman (Shull Group, NU). Many of the students have decided to further study science and engineering in college and potentially pursue a career in these fields. In 2017, one of the attendees applied to Northwestern University to pursue engineering and work with CHiMaD directly as a result of this program, he ultimately accepted a position as an undergraduate researcher at UC Berkeley and is currently pursuing engineering.

The 6-week *Engineering Core Course* took place between June 25-August 3, 2018, as two 2-hour sessions each week, and was attended by 17 high-school students enrolled in the program.

Figure 21.1: CHiMaD-lead UCCSP engineering course is designed to illustrate the concepts and principles of polymeric, composite, and nanoscale materials and demonstrate the relationship between molecular structures and material properties in the context of real-world applications, with an emphasis on materials design

21.2 CHiMaD Research Experience for Undergraduates Program

To encourage engineering undergraduates to further pursue their education in materials science and design-related fields, CHiMaD launched its Research Education for Undergraduates (CHiMaD-REU) program in summer 2016. To date, eight students have completed the program. In 2018, the following three students were accepted into the program:

- Thomas Wu *(Shull Group, NU)*
- Katherine Su *(Hersam Group, NU)*
- Nicholas Ury *(QuesTek Innovations)*

The 2018 students have served to strengthen the ties between the CHiMaD-REU program and the NIST-SURF program and to support ongoing research collaborations between NIST and CHiMaD. Thomas Wu and Katherine Su completed the NIST-SURF program in 2017 under the guidance of CHiMaD's NIST PIs **Prabhu** and **Davydov**. The 2018 CHiMaD REU program facilitated a continuation of the research collaborations between the NIST-CHiMAD. Thomas Wu and Katherine Su completed the CHiMaD-REU program under the guidance of **Shull** and **Hersam** groups, completementing their 2017 summer research. Nicholas Ury, on the other hand, completed the CHiMaD REU program in 2017 and was accepted to be summer intern at QuesTek Innovations for the summer of 2018, partially supported by CHiMaD and working partially on CHiMaD-related QuesTek projects.

Brittany Nez, one of the 2017 CHiMaD-REU URM students who completed the program has been offered and accepted a six-month internship at QuesTek Innovations in 2018 and was later hired as a full time Materials Application Engineer at the company.

21.3 Support for NIST-Summer Undergraduate Research Fellowship Program

NIST colleagues led by **Campbell**, and supported by **Warren**, has been offering summer opportunities to undergraduate students under the NIST-Summer Undergraduate Research Fellowship (NIST-SURF) Program on their collaborative research topics with CHiMaD. Since the inception of the partnership in 2016, CHiMaD has placed 12 students to the SURF program. In 2018, the following SURF projects were carried out by CHiMaD students:

- Daniel Ng (NU) [**Stoudt** / Li]: *Metals Additive Manufacturing: What did we really make - Decoding the interfaces*
- David Yoon (NU) [**Salipante**]
- Eric Anderson (NU) [**Nieuwendaal**]: *Simulating REDOR NMR curves from atom positions that result from MD simulations*
- Rachel Orenstein (NU) [**Trautt**]: *Building the "self-driving" Laboratory*
- Ryan Zambrotta (NU) [**Phelan**]: *Molecular Dynamics in Mechanophore Functionalized Epoxy Networks*
- Leah Borgsmiller (NU) [**DeLongchamp** / Heiber]: *Characterizing the Competition Between Charge Extraction and Recombination in Organic Solar Cells and Contributing to a Curated Dataset for Machine Learning*
- Samantha Halam (FSU) - matched to a non-CHiMaD related project

Among these students, Daniel Ng (NU) was a returning student to the SURF program after having completed under the guidance of **Stoudt** and Li in 2017, where he co-authored 2 publications with the NIST PIs - one of the papers published in 2018 [27.j.]. Upon successful completion of the 2018 program, we was awarded the 2018 NIST MML Student Intern Accolade for his *outstanding contributions in determining the time-temperature-transformation diagram for the formation of the delta phase in additive manufactured Ni-base superalloy, IN625*. Another NIST publication from 2017 based on this summer work has also been recognized as *Editor's Choice* in *Metallurgical and Materials Transactions A* in 2018.

21.4 Support for 2018 TMS Diversity Summit

The Minerals, Metals, and Materials Society (TMS) initiated the *Summit on Diversity in the Minerals, Metals, and Materials Professions (DMMM)* in 2014. The biennial Summit was co-hosted by CHiMaD in 2016 at Northwestern University. CHiMaD has continued to support the TMS initiative in its third edition, hosted by University of Santa Barbara on July 23-24, 2018. This years theme was to *Engage - Measure - Transform*. The Summit's scope aligns with CHiMaD Outreach program goals towards underrepresented minorities in;

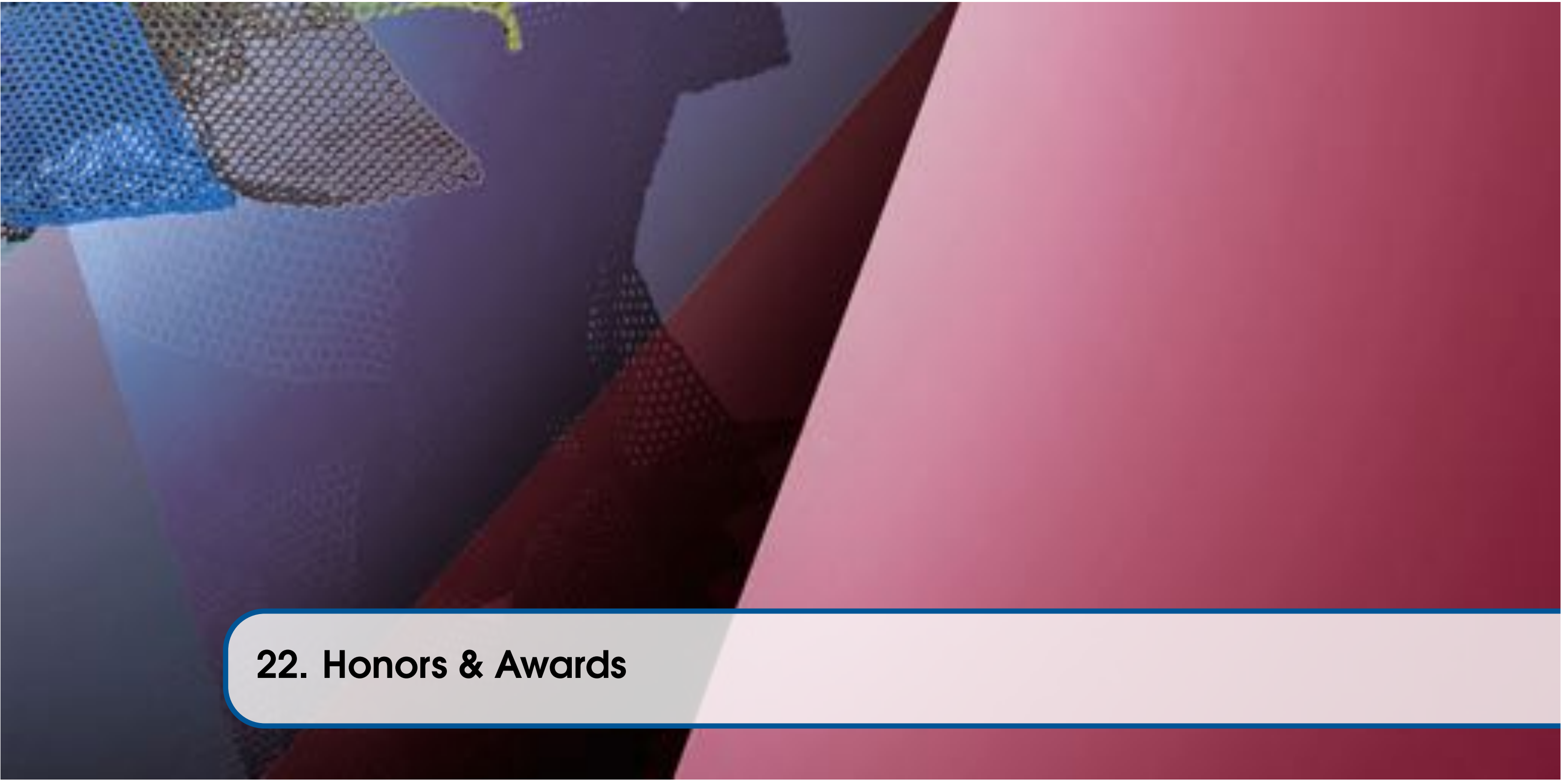
- Strategies for increasing underrepresented minority (URM) engagement in the STEM professions
- Best practices for building a welcoming and inclusive workplace culture
- Addressing factors behind the self-induced glass ceiling and supporting new pathways to career fulfillment

21.5 ASM Materials Genome Camp for High-School Educators

The Camp is discussed in detail in Section 20.2 of this report.

VI

Other



22. Honors & Awards

Awards received by CHiMaD PIs and Researchers in 2018

Wei Chen, *Northwestern University*

- Ver Steeg Faculty Award, Northwestern University

Mark Hersam, *Northwestern University*

- National Academy of Inventors (NAI) Fellow
- Web of Science Highly Cited Researcher
- Advanced Materials Hall of Fame
- Kay Malmstrom Lecture in Physics, Hamline University
- Plenary Speaker, International Graphene Innovation Conference, Xi'an China
- Plenary Speaker, Carbon-Rich Molecules to Carbon-Based Materials Conference, Nassau, Bahamas
- Plenary Speaker, International Conference on Nanoscience and Technology, Bangaluru, India

Wing Kam Liu, *Northwestern University*
with *Zhengtao Gan, Yanping Lian, Stephen E. Lin, Kevontrez K. Jones, Wing Kam Liu, and Gregory J. Wagner*

- 1st Place, Best modeling results predicting the cooling rates of three single laser tracks on a bare IN625 plate, AM-Bench 2018 Benchmark Challenge
- 1st Place, Best modeling results predicting the grain structure of three single laser tracks on a bare IN625 plate, AM-Bench 2018 Benchmark Challenge
- 1st Place, Best modeling results predicting the dendritic microstructure of three single laser tracks on a bare IN625 plate, AM-Bench 2018 Benchmark Challenge

Heinrich Jaeger, *University of Chicago*

- Sewell Avery Distinguished Service Professorship in Physics, University of Chicago
- Elected Member, American Academy of Arts and Sciences

David Seidman, *Northwestern University*

- Elected Member, National Academy of Engineering
- Gold Medal, ASM International - 2019
- Elected Member, European Union Academy of Sciences

Tobin Marks, *Northwestern University*

- Lee Fong Award and Lectureship, National Taiwan University
- CPSE Award and Lectureship, First National Meeting of the Swedish Chemical Society
- Foreign Member, Accademia Nazionale dei Lincei (Italian National Academy of Sciences)
- Clarivate Analytics Citation Laureate

Steven Sibener, *University of Chicago*
- Director of The James Franck Institute, University of Chicago

Marius Stan, *Argonne National Laboratory*
- Doctor Honoris Causa, Politehnica University of Bucharest, Romania

Peter Voorhees, *Northwestern University*
- Moore Distinguished Scholar, California Institute of Technology

Chris Wolverton, *Northwestern University*
- Fellow, American Society for Metals (ASM International)

Jennifer Bennett, *Cao Group, Northwestern University*
- Northwestern McCormick Terminal Year Fellowship

Hadalia Bergeron, *Hersam Group, Northwestern University*
- First Place, Graduate Research Competition, 2018 Society of Women Engineers Midwest Conference

Ding-Wen Chung, *Dunand/Seidman Group, Northwestern University*
- Department of Energy (DOE) Science Graduate Student Research (SCGSR) award

Nitin Hansoge, *Keten Group, Northwestern University*
- Best Poster Award, 5^th Annual Computational Research Day, Northwestern University

Lu Li, *Tirrell Group, University of Chicago*
- 3^rd place, Student Research Presentation Competition, Annual Conference of Chinese American Chemical Society in the Great Lakes Area

Alexander Marras, *Tirrell Group, University of Chicago*
- Successful User Proposal at Advanced Photon Source at Argonne National Laboratory: "Characterizing micellar assembly of oligonucleotides with synthetic designer polyelectrolytes using SAXS"
- Successful User Proposal at Molecular Foundry at Lawrence Berkeley National Laboratory: "A real-time investigation into the ordering of polyelectrolyte complex micelles via in situ TEM"

Julia Murphy, *Sibener Group, University of Chicago*
- Best Presentation Award, 2018 Women in Science Conference, Notre Dame

Angelika Neitzel, *Tirrell Group, University of Chicago*
- Selected for Future Faculty Workshop: Developing Diverse Leaders for Tomorrow program, University of Delaware

Fernando L. Reyes Tirado, *Dunand/Seidman Group, University of Chicago*
- Honorable Mention, NUANCE Image Contest for his work "Branching Out"

Jeffrey Ting, *NIST-CHiMaD Postdoctoral Fellow, Tirrell Group, University of Chicago*
- Class of 2018 ACS PMSE Future Faculty Scholar program, American Chemical Society
- Selected for ACS Postdoc 2 Faculty Workshop in Boston, American Chemical Society
- Selected for Future Faculty Workshop: Developing Diverse Leaders for Tomorrow program, University of Delaware

- Recognized by the University of Chicago Office of Research and National Laboratories for the establishment of the Joint Research Safety Initiative, serving as Vice President
- Successful User at Oak Ridge National Laboratory, High Flux Isotope Reactor: "Chain Exchange Kinetics in Polypeptide-Based Non-Viral Gene Delivery Systems"
- Successful User Proposal, NIST Center for Neutron Research: "Revealing Molecular Exchange Kinetics in Polyelectrolyte Complex Micelles"

Sarah Wolff, *Cao Group, Northwestern University*

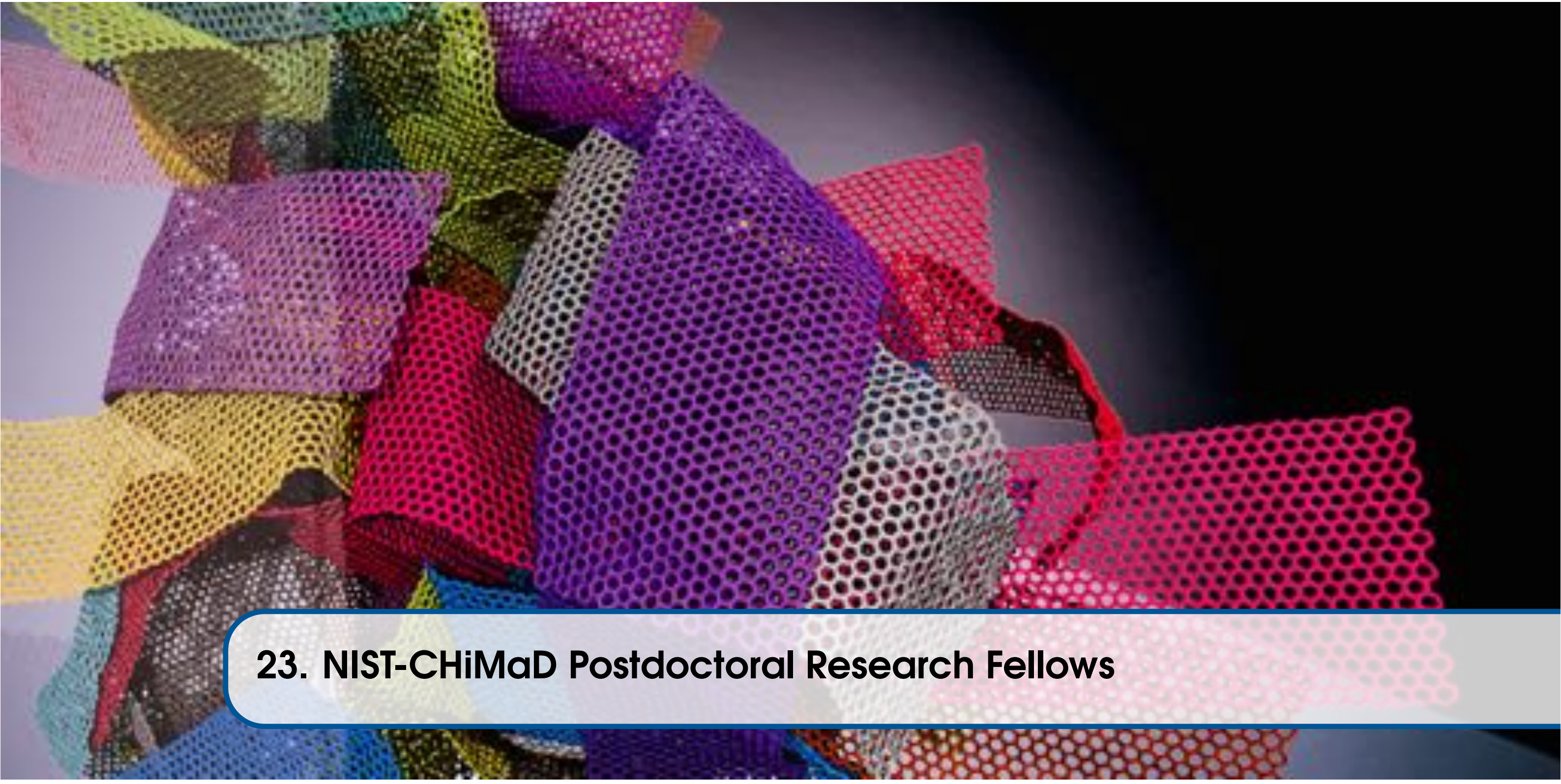
- Argonne National Laboratory Named Director's Enrico Fermi Fellowship

Hao Wu, *Tirrell Group, University of Chicago*

- Successful User at Oak Ridge National Laboratory, High Flux Isotope Reactor: "Chain Exchange Kinetics in Polypeptide-Based Non-Viral Gene Delivery Systems"
- Successful User Proposal, NIST Center for Neutron Research: "Revealing Molecular Exchange Kinetics in Polyelectrolyte Complex Micelles"

Chun Zhou, *Nealey Group, University of Chicago*

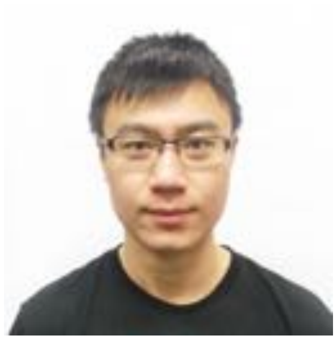
- 2018 Hiroshi Ito Memorial award, SPIE Advanced Lithography Conference - to recognize the best student paper presented in the Patterning Materials conference for *Studying the effects of chemistry and geometry on DSA hole-shrink process in three dimensions* co-authored by T. Kurosawa, T. Dazai, J. Doise, J. Ren, C. Bezik, T.Segal-Peretz, A.Yamazaki, P. R. Delgadillo, J. De Pablo, P. Nealey



23. NIST-CHiMaD Postdoctoral Research Fellows

23.1 Wenjie Xia (CHiMaD)

Polymer Composites
Supervisors: Sinan Keten (NU), Frederick Phelan (NIST), Jack Douglas (NIST)



Bio *Dr. Wenjie Xia finished his Ph.D. from the Department of Civil and Environmental Engineering at Northwestern University in 2016. Dr. Xia recently joined the Department of Civil and Environmental Engineering at North Dakota State University (NDSU) as a tenure-track assistant professor starting Aug. 2018. During his doctoral study, his research focused on computational mechanics and multi-scale modeling of polymer nanomaterials. After his Ph.D., he was awarded a NIST-CHiMaD Postdoctoral Fellowship to develop scale-bridging techniques and computational models for investigation of soft matters. He has received the Water P. Murphy Fellowship and been selected as the finalist for the American Physical Society Frank J. Padden Jr Award for excellence in graduate polymer research. He has also received the Chinese government award for outstanding students abroad.*

23.1.1 Research Accomplishments

Dr. Xia's research has developed the energy-renormalization approach to coarse-graining polymers with different glass-forming properties (i.e., fragility), that can reproduce the temperature-dependent dynamics over a wide temperature range. The established CG modeling framework through Dr. Xia's efforts is salient not only for polymers, but also for many other classes of glass, such as biomolecules, metallic glasses, and even simple molecular liquids (Figure 23.1), where cooperative dynamics and glass-forming process occur upon supercooling. This represents critical progress for building computation-based materials-by-design framework for design and development of advanced glass-forming materials with tunable properties, such as fragility, relaxation dynamics and activation energy.

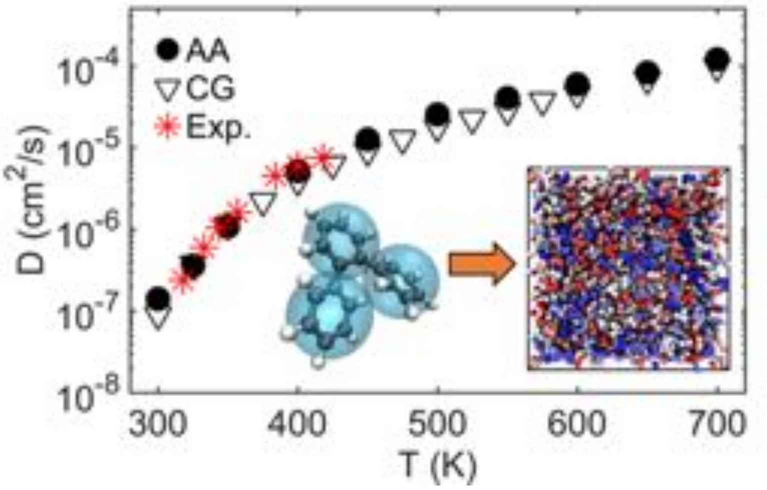


Figure 23.1: Temperature transferable coarse-graining of small molecule glass-forming liquid via energy-renormalization approach.

In another research work, Dr. Xia in collaboration with Dr. Jack Douglas (NIST) explore the structural and dynamic properties of bulk materials composed of graphene nanosheets using coarse-grained molecular dynamics simulations (Figure 23.2). Remarkably, D. Xia's results show clear evidence that bulk graphene materials exhibit a fluid-like behavior similar to linear polymer melts at elevated temperatures and that these materials transform into a glassy-like "foam" state at temperatures below the glass-transition temperature of these materials. Distinct from an isolated graphene sheet, which exhibits a relatively flat shape with fluctuations, it is found that graphene sheets in a melt state structurally adopt more "crumpled" configurations and correspondingly smaller sizes, as normally found for ordinary polymers in the melt. Upon approaching the glass transition, these two-dimensional polymeric materials exhibit a dramatic slowing down of their dynamics that is likewise similar to ordinary linear polymer glass-forming liquids. These findings show that graphene melts have interesting lubricating and "plastic" flow properties at elevated temperatures, and suggest that graphene foams are highly promising as high surface filtration materials and fire suppression additives for improving the thermal conductivities and mechanical reinforcement of polymer materials.

23.1.2 Collaborations

Temperature Transferable Coarse-graining of Polymer Dynamics

Wenjie Xia (CHiMaD), Frederick ***Phelan*** *(NIST), Jack* ***Douglas*** *(NIST)*

This collaboration focuses on development of scale-bridging technique for simulating polymer at extended time and length scales. The developed coarse-grained model through a novel approach, called the energy renormalizing approach, can capture the dynamics of the representative polymer system over a wide range of temperatures.

Figure 23.2: This image depicts a three-dimensional graphene melt composed of nanosheets, which characteristically exhibits a glass-transition behavior analogous to ordinary polymers. [Authors: Wenjie Xia, Fernando Vargas-Lara, Sinan Keten, Jack F. Douglas]

23.1.3 Publications and Presentations

4 Number of CHiMaD-supported publications in 2018. Please see chapter 27 for details [27.54, 27.58, 27.59, 27.61].

2 Number of presentations on CHiMaD supported research in 2018, please see section 26.13 for the complete list.

23.2 Richard Sheridan (CHiMaD)

Polymer Composites
Supervisors: Kenneth Shull (NU), Jeffrey Gilman (NIST), Cate Brinson (Duke)

Bio Dr. Richard Sheridan finished his Ph.D. in 2012 at the University of Colorado at Boulder in the Chemical and Biological Engineering Department. The topic of his dissertation was the applications and rheology of thermoreversible Diels-Alder networks. Following a postdoctoral term at NIST, including a NIST-NRC Postdoctoral Fellowship, he received the NIST-CHiMaD Postdoctoral Fellowship to engage in experimental studies of fiber reinforced composites, particularly fatigue, damage, and properties at the fiber matrix interface and interphase.

23.2.1 Research Accomplishments

Dr. Sheridan's core areas of interest include polymer science, surface science and metrology, hard-soft material interfaces, and functional composites. Recently, it is important for a material to bring more to an application than its basic properties, like modulus or fracture toughness. Materials can be functional, whether the function is environmental sensing, self-healing, self-assembly, or stimulus response. Dr. Sheridan aims to create new polymeric and composite materials that take advantage of multiple phases and mechanically activated chemical ("mechanophore") dopants to create designed functional material components that are resistant to damage and fatigue.

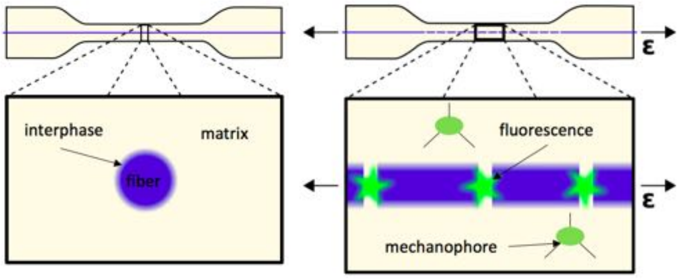


Figure 23.3: An single fiber embedded dogbone provides a platform for measurement of interphase properties when sectioned *(Left)* or elongated to fragmentation *(Right)*. The sectioned specimens reveal the interphase modulus and chemistry, while the fragmented samples show the interfacial shear strength and, when augmented with fluorescent mechanophores, regions damaged by the fragmentation events.

Directly measuring the strength of interfaces and interphase in composite materials remains a challenge. In collaboration with **Holmes** at NIST, Dr. Sheridan gained expertise in the execution and interpretation of the single-fiber fragmentation test (SFFT), which is used to compare interfacial shear strengths. The collaboration resulted in new ideas for increasing the speed and consistency of the test, including the design and construction of "Snappy," a modern single-fiber fragmentation device.

SFFT can also function as a platform for other experiments. In collaboration with **Wood-**

cock at NIST, SFFT was performed using a mechanophore-doped epoxy resin. The mechanophores became fluorescent at locations where the fibers fragmented, recording the shockwaves resulting from the energy release by each break. The position and fluorescence lifetime of the activated fluorophores was recorded with nanoscale precision on the advanced fluorescence lifetime imaging microscopes in the laboratory of **Stranick** at NIST. This work has been submitted to Nature Materials.

Along with the strength of the fiber-matrix interface/interphase, Dr. Sheridan made advances in the measurement of interphase modulus. Along with refining the process of ion beam milling AFM nanoindentation measurement of the sub- 100 nm region of perturbed material, he collaborated with **Oleshko** to perform a complimentary interphase S/TEM study to reveal correlations between the bulk plasmon peak and nanoindentation modulus.

Manipulating the composite interphase is the next step forward in advanced composite design. To that end, Dr. Sheridan has developed an efficient and reproducible lab-scale method of coating fibers with cellulose nanocrystals. These nanocrystals have been functionalized by **Fox** at American University to give chemical compatibility with an epoxy matrix and may deliver extra toughness directly to the composite interphase.

23.2.2 Collaborations

In situ Nanoimprint Decay Measurement
Richard Sheridan (CHiMaD), Sonal Bhadauria (Akron), Alamgir Karim (Houston), and Chris ***Stafford*** *(NIST)*
Graduate student making thin thermoplastic films imprinted with a nanoscale pattern and observing the decay in amplitude of the pattern to infer stabilization by nanomaterial filler.
Correlation of AFM modulus and S/TEM EELS plasmon shift in organic materials
Richard Sheridan (CHiMaD), Vladimir ***Oleshko*** *(NIST), Bharath* ***Natarajan*** *(NIST)*
NIST staff experts in TEM measurements and SEM/FIB manipulation working to correlate EELS plasmon shift with modulus, focusing on glass-epoxy interphase.

Interphase effects on adhesion in fiber reinforced composites
Richard Sheridan (CHiMaD), Gale ***Holmes*** *(NIST)*
NIST staff expert in single fiber fragmentation working to determine interphase shear strength and improve the single fiber fragmentation test.

Cellulose nanocrystal coatings for fiber reinforcement
Richard Sheridan (CHiMaD), Doug ***Fox*** *(American), Jan* ***Obrzut*** *(NIST), Bharath* ***Natarajan*** *(NIST)*
Experts in the functionalization, assembly and measurement of cellulose nanocrystal suspentions and materials.

23.2.3 Publications and Presentations

4 Number of CHiMaD-supported publications in 2018. Please see chapter 27 for details [27.54, 27.58, 27.59, 27.61].

2 Number of presentations on CHiMaD supported research in 2018, please see section 26.13 for the complete list.

23.3 Hyeong Min Jin (CHiMaD)

Directed Self Assembly of Block Copolymers for Lithographic Applications
Supervisors: Paul Nealey (UC), Joseph Kline (NIST)

Bio Dr. Hyeong Min Jin received his Ph.D. from the Materials Science and Engineering Department, KAIST in 2017 for investigating the directed molecular self-assembly by photo-thermal effect and practical applications for block copolymer nanopatterning. In 2018, he joined the Paul Nealey group at University of Chicago and was awarded a NIST-CHiMaD postdoctoral research fellowship to develop resonant soft x-ray scattering (RSoXS) to probe three-dimensional order and disorder in soft matter system.

23.3.1 Research Accomplishments

Dr. Jin is working to build the resonant soft x-ray scattering (RSoXS) chamber, which enable to probe directed self-assemblies of block copolymer systems and liquid crystals, at Sector 29 of the Advanced Photon Source (APS) at ANL. The chamber has been assembled and is currently commissioning. In the meantime, in collaboration with **Kline**, he has successfully observed the martensitic phase transformation in the blue phase (BP) liquid crystals (LCs) by RSoXS at ALS, Berkeley National Laboratory (BNL). BP LCs are chiral liquid crystalline phases in which the liquid crystal molecules spontaneously form double-twist cylinders and these cylinders pack to form one of two fluidic lattices with different cubic symmetries - the so-called BPI for the body-centered cubic structure and BPII for the simple cubic structure. Recently, it was demonstrated that the phase transformation between BPI and BPII occurred in a diffusionless manner, with characteristics reminiscent of traditional martensitic transformations in atomic crystals by optical measurement and computational simulations. However, the in-plane lattice orientation of the BP LCs upon transformation, as well as the molecular orientations within the fluidic lattice, were not possible to determine. From the RSoXS measurement during the phase transformation from single crystalline BPII$_{(100)}$ to BPI$_{(110)}$, we could clearly index scattering patterns and thus see clear details of lattice transition. BPII$_{(hkl)}$ denotes a BPII lattice oriented with (hkl) plane parallel to the surface. The POM measurements confirmed that the BPI$_{(110)}$ forms a macroscopic crosshatched structure during the martensitic transformation from a single crystal BPII$_{(100)}$, but it was unclear why this structure is formed. Based on these preliminary measurements, we propose that this crosshatched structure is due to the formation of twin lamellae to release the residual strain caused by the rapid martensitic transformation (Figure 23.4). These insights will provide a better understanding of martensitic transformation in the fluidic lattice system.

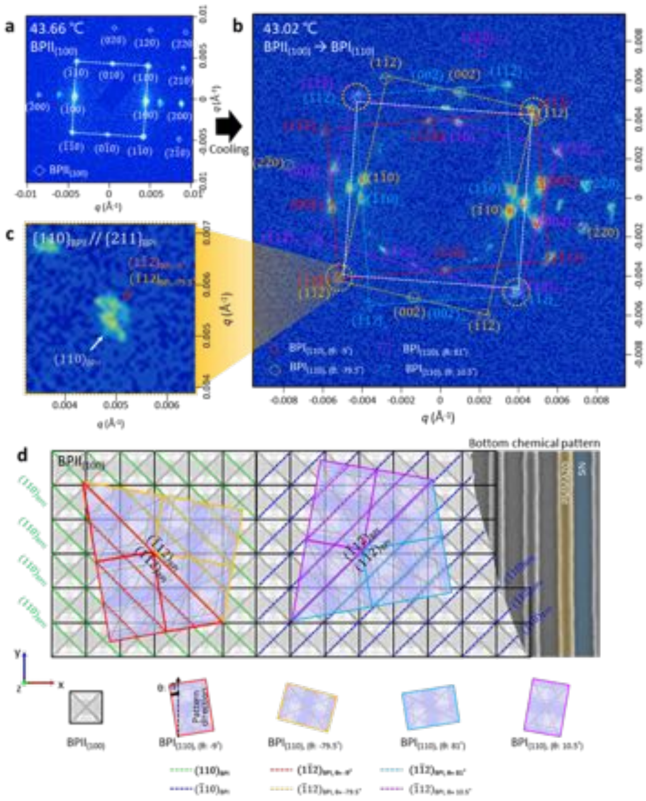


Figure 23.4: RSoXS pattern of (a) single crystal BPIII$_{(100)}$ at 43.66°C and (b) poycrystalline BPI$_{(110)}$ which has distinct domains with four different in-plane orientation, θ of $-9.75°$, $-80.25°, 9.75°$, and 80.25° at 43.02°. θ is an angle difference between [110] of BPI$_{(110)}$ lattice and bottom chemical pattern stripe. (c) Scattering peaks of $\{112\}_{BPI}$ plane appear near the scattering peaks of $\{110\}_{BPII}$ plane and merged each other during the transformation. (d) Schematic of BPI$_{(110)}$ lattice that martensitic transformed from single crystalline BPI$_{(100)}$.

23.3.2 Collaborations

RSoXS measurement of the phase transformation between BP LCs

Hyeong Min Jin (CHiMaD), Joseph ***Kline*** *(NIST)*

During the collaboration, martensitic transformations between BP LCs were investigated by RSoXS at the BL 11.0.1.2, ALS, BNL. After the measurement,**Kline** (NIST) also provided a lot of advice on analysis

Fabrication of BP LC "sandwich" cells for RSoXS measurement

Hyeong Min Jin (CHiMaD), Xiao Li (UC), James A. Dolan (UC/ANL)

This collaboration focuses on the fabrications the BP LC "sandwich" cells for the RSoXS measurement. Dr. Xiao has successfully demonstrated a single crystalline BP LC by directed self-assembly and BP LC "sandwich" cells were successfully invented by this collaboration.

23.4 Peisheng Wang (CHiMaD)

Precipitation Strengthened Alloys
Supervisors: Greg Olson (NU), Carelyn Campbell (NIST), Ursula Kattner (NIST), Kil-Won Moon (NIST)

Bio *Dr. Wang's main research interest lies in thermodynamic calculations (CALPHAD). He received his Ph.D. in 2012 from Central South University, China for experimental determination of Mg phase diagram and thermodynamic calculations. He was then a postdoctoral researcher at TU Clausthal, Germany between 2012-2014. During his postdoctoral work he was interested in Heterogeneous Nucleation and Microstructure Formation of Al-Cu-Ni alloys and a new function to describe the Gibbs energy of liquid phase. In 2015 he was awarded a NIST-CHiMaD Postdoctoral Research Fellowship to further CALPHAD methods under Precipitation-Strengthened Alloys use-case group.*

23.4.1 Research Accomplishments

Data from DFT calculations without spin-polarization is frequently used in literature to describe the "non-magnetic" part in CALPHAD which is not correct. It is because the CALPHAD description of a magnetic phase without considering the magnetic contribution to the Gibbs energy describes the paramagnetic state rather than the non-magnetic state. Based on that, a new method was developed to describe the magnetic contribution to the total energy in CALPHAD based on DFT calculation. Figure 23.5 shows the calculated enthalpies of formation and calculated magnetic moments for the sigma phase, respectively. The present CALPHAD description can well described the DFT spin-polarized calculation and paramagnetic calculation using atomistic calculation.

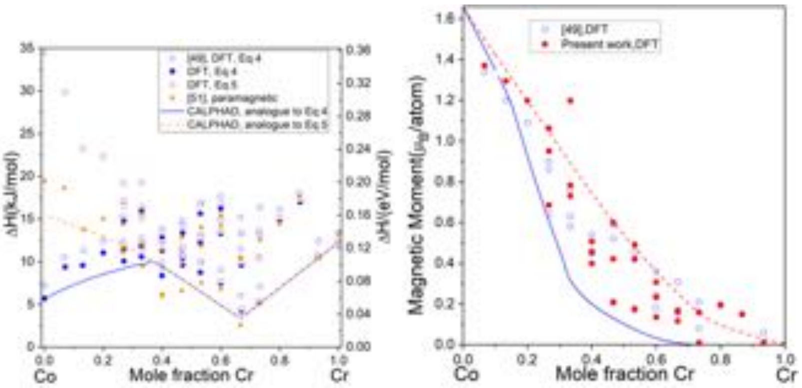


Figure 23.5: *(Left)* CALPHAD calculated enthalpy of formation in Joule per mole of atoms for the σ phase at 298.15 K compared with DFT results. For comparison with the DFT results the interaction parameters were removed from the description of the σ phase. *(Right)* Calculated magnetic moment of the σ phase at 298.15 K. The symbols show the average magnetic moment from the DFT calculations for the individual end-members.

23.4.2 Collaborations

Cobalt-based Thermodynamic Database

Peisheng Wang (CHiMaD), Carelyn ***Campbell*** *(NIST), Ursula* ***Kattner*** *(NIST), Kil-Won* ***Moon*** *(NIST), Eric* ***Lass*** *(NIST)*

This collaboration involves utilizing the co-based thermodynamic database built by Wang to establish a cobalt diffusion mobility database.

23.4.3 Publications and Presentations

1 Number of CHiMaD-supported publications in 2018. Please see chapter 27 for details [27.89].

1 Number of presentations on CHiMaD supported research in 2018, please see section 26.13 for the complete list.

23.5 Qifeng Wang (NU)

Polymer Composites - Industry Supported Fellow
Supervisor: Kenneth Shull (NU)

Bio Dr. Qifeng Wang earned his Ph.D. degree of polymer chemistry and physics at Jilin University, China. He is currently a postdoctoral research fellow in Shull group in Department of Materials Science and Engineering at Northwestern University. His research focuses on high frequency rheology of epoxy composite at extreme conditions such as cryogenic condition or high temperature, polyelectrolyte complex, and adhesion of soft materials.

23.5.1 Research Accomplishments

The dynamic of epoxy curing was studied at room temperature, which is essential for the epoxy prepreg applying procedure. The applying of repreg requires both the viscosity and adhesion are in a right region to make sure it is viscous enough to fit the surface but sticky enough to hold itself and the structure (Figure 23.6). The effects from molecular weight of linear cross-linker to the cross-linking dynamic were discovered. The higher molecular weight of the cross-linker of the same system, the slower the cross-linking is. Meanwhile, rheology of epoxy at cryogenic condition (e.g. in liquid nitrogen and oxygen) determines the behavior and application of epoxy composite at that condition. However, only very limited research has been done in that area. A primary research with the developed Quartz Crystal Microbalance (QCM) technique was initiated and more promising results are expected in the upcoming year.

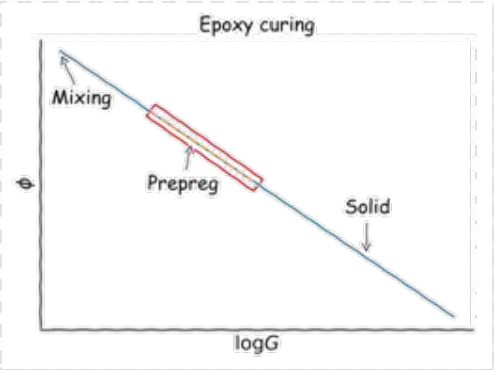


Figure 23.6: Schematic illustrated of the mechanic property-changing while the epoxy curing

23.5.2 Collaborations

Epoxy Composites
Qifeng Wang (NU/NIST), Jeffrey ***Gilman*** *(NIST); Christopher* ***Soles*** *(NIST), Kenneth* ***Shull*** *(NU), Sinan* ***Keten*** *(NU)*
Correlating the quasielastic neutron scattering (QENS) at the frequency range about 200 GHz with relative lower frequency rheology from quartz crystal microbalance (QCM) at about 15 MHz. In addition, correlate both experimental results with computer simulation.

23.5.3 Technology Transfer

Software

Quartz Crystal Microbalance (QCM) Data Collection and Analysis Software

Qifeng Wang (NU), Meghan Yang (NU), Kenneth Shull (NU)

Link: `https://github.com/zhczq/QCM_py`

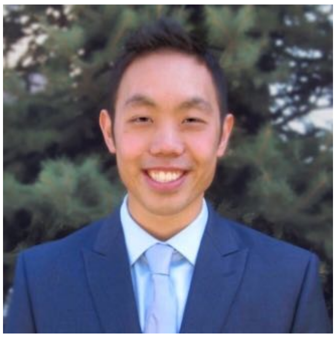
A program for the high frequency rheology study with quartz crystal microbalance (QCM) was developed. The current version of the program, developed by Wang (NU), utilizes Python and functionalizes with data acquisition (QCM data and temperature) and data analysis. The program make helps analyze epoxy at various conditions. This program aims to make it easier for the researchers to utilize the Quartz Crystal Microbalance (QCM) for mechanical studies in addition to the traditional film mass study. This open source repository is on Github.

23.5.4 Publications

1 Number of CHiMaD-supported publications in 2018. Please see chapter 27 for details [27.62].

23.6 Jeffrey Ting (CHiMaD)

Soft Matter Design based on Charge Complexation
Supervisors: Matthew Tirrell (UC), Vivek Prabhu (NIST), Jack Douglas (NIST), Debra Audus (NIST), Kenneth Kroenlein (NIST)



Bio Dr. Ting received his B.S. in Chemical Engineering in 2011 at the University of Texas and his Ph.D. in Chemical Engineering in 2016 from the University of Minnesota, where he worked with Frank Bates and Theresa Reineke on synthesizing tunable polymers for oral drug delivery. Dr. Ting was a recipient of the L. E. and D. H. Scriven Fellowship, NSF Graduate Research Fellowship, and Minnesota Doctoral Dissertation Fellowship. His work was recognized by the first annual 2015 AIChE Pharmaceutical Discovery, Development and Manufacturing Student Award. Currently, Dr. Ting is a NIST-CHiMaD Postdoctoral Fellow in Matthew Tirrell's group, where he is exploring dynamics and self-assembly of designer polyelectrolyte complexes. He was recently inaugurated into the Class of 2018 ACS PMSE Future Faculty Scholars

23.6.1 Research Accomplishments

Dr. Ting, in collaboration, has established the synthetic capabilities and protocols to rapidly generate libraries of designer polyelectrolyte assemblies. This has expanded the ability to create materials datasets from an experimental standpoint. Figure 23.7 shows a representative strategy for using aqueous reversible addition-fragmentation chain transfer (RAFT) polymerization to prepare a series of styrenic diblock polycations and polyanions in a parallel synthesizer.

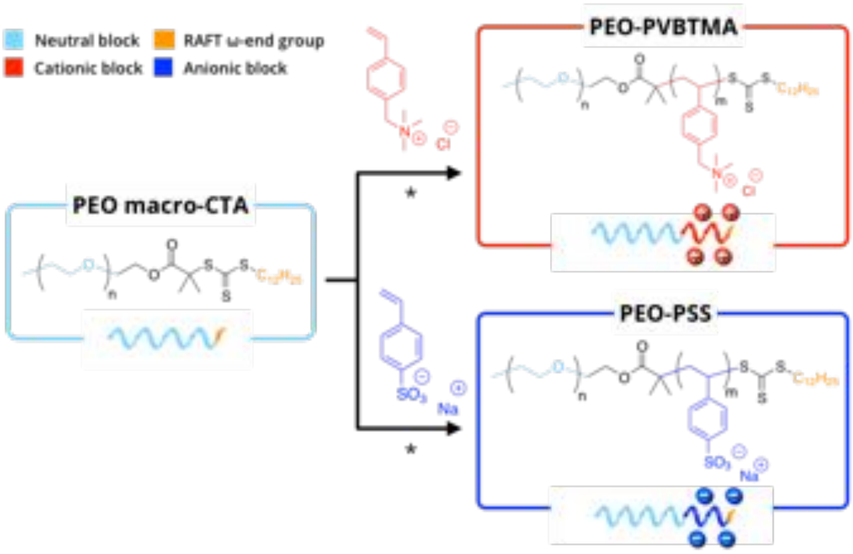


Figure 23.7: Aqueous RAFT polymerization scheme to produce gram-scale quantities of tunable designer polyelectrolytes for screening chain lengths and chemical architectures.

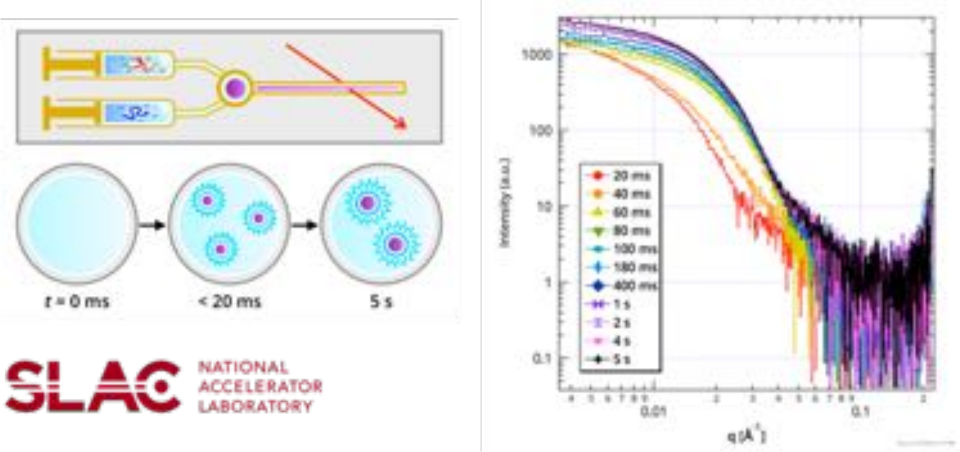


Figure 23.8: The formation kinetics of polyelectrolyte-core micelles is being investigated using time-resolved SAXS with a stopped-flow apparatus. Data collected at the Stanford Synchrotron Radiation Lightsource in SLAC National Accelerator Laboratory.

With an initial set of well-defined materials characterized, combinations of block polyelectrolytes were systematically studied to explore the spatiotemporal formation, evolution, and disassembly dynamics of complexation-driven micelles. These aspects of polyelectrolyte solution assembly (unlike their amphiphilic block polymer counterparts) are not fully understood and lack experimental measurements to complement existing theories and simulations. To this end, synchrotron small angle X-ray and neutron scattering (i.e., SAXS and SANS) at several U.S. national labs have served as key instrumentation in pursuing these endeavors. We report promising preliminary results on each of these fronts as 2018 accomplishments and, moving forward, aim to incorporate these findings into new predictive capabilities for ion-containing soft matter design.

First, poly(ethylene oxide)-block-poly((vinylbenzyl) trimethylammonium chloride)) (PEO-b-PVB) and poly(sodium acrylate) (PAA) were used as a model system for investigating the in situ formation and growth kinetics of complex-core micelles. Figure 23.8 shows an illustration of the stopped-flow apparatus that enables time-resolved small angle X-ray scattering with millisecond resolution. From the SAXS overlay, initial particles have already assembled by $\sim$20 ms. After 40 ms, a distinct change in the scattering curve indicates a transition in the assembly structure, followed by increased growth by higher scattering intensities. We are in the process of further analyzing this data, but the initial results of PEO-b-PVB/PAA micelles show the first example of understanding the structural pathway of complex-core micelle formation in aqueous self-assembly.

Next, we examined chain exchange kinetics using poly(ethylene oxide)-block-poly(lysine) (PEO-b-PLK) and the hydrogenated and deuterated forms of poly(sodium 4-vinylbenzene sulfonate) (*h*-PSS and *d*-PSS, respectively). Figure 23.9 shows a representative summary of how the time-resolved small angle neutron scattering profiles evolved. Surprisingly, the process of exchange was immensely slow, even under extreme conditions applied in attempts of accelerating the phenomena by affecting ionic strength or solvent quality (e.g., 2 M NaCl or 70°C). Thus, these particles appear to be extremely robust, and the only likely chain exchange occurs upon fission/fusion events (more rigorous analysis of this is underway). To the best of our knowledge, this is the first attempt at quantifying the chain exchange mechanism of complex-core micelles. As a comparison, utilizing deuterated

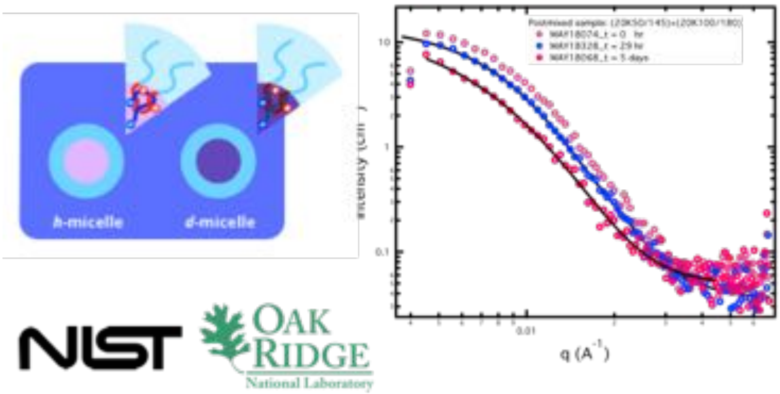


Figure 23.9: The formation kinetics of polyelectrolyte-core micelles is being investigated using time-resolved SAXS with a stopped-flow apparatus. Data collected at the Stanford Synchrotron Radiation Lightsource in SLAC National Accelerator Laboratory.

Figure 23.10: The disassembly and relaxation properties of polyelectrolyte-core micelles are being studied as a function of sodium chloride concentration using SAXS. Data was collected at the Advanced Photon Source in Argonne National Laboratory.

forms of PEO-PVB and PAA are currently being pursued.

Finally, we have led efforts to understand the effects of salt on processing the reorganization and disassembly of (1) PEO-PVB/PEO-PSS nanoaggregates and (2) PEO-PVB/PAA complex-core micelles, Figure 23.10. In the former case, time-resolved dynamic light scattering, SAXS, and cryogenic imaging enabled us to map the various relaxation pathways for far-from-equilibrium polyelectrolyte complex systems. In the latter system, the mean aggregation number per micelle and water content of the core are being determined as a function of salt. Collectively, these studies can provide metrics of stability and resistance of complex-core micelles to varying ionic conditions found in biological environments, increasing their viability as robust nanocarriers for various biomolecules.

23.6.2 Collaborations

Imaging Polyelectrolyte Dynamics with Super-resolution Microscopy

Jeffrey Ting (CHiMaD), Matt ***Tirrell*** *(UC), Muzhou Wang (NU)*

To complement scattering information on polyelectrolyte phase morphologies and address gaps in our understanding of polyelectrolyte dynamics, we are exploring non-invasive imaging approaches through super-resolution optical microscopy.

Diffusive Behavior of Polyelectrolyte Complex Solutions

Jeffrey Ting (CHiMaD), Matt ***Tirrell*** *(UC), Bradley Olsen (MIT)*

We are conjugating promising block polyelectrolytes with fluorescent markers to conduct Forced Rayleigh Scattering experiments in the Olsen Lab.

RAFT Polymerization in Complex Solvents

Jeffrey Ting (CHiMaD), Matt ***Tirrell*** *(UC), Stuart Rowan (UC), Theresa Reineke (Minnesota)*

To further advance the ease and versatility of RAFT polymerization towards high-throughput materials development capabilities, we are investigating controlled polymerization in tandem with enzymatic deoxygenation in a series of ill-defined aqueous solvents.

Model Proteins Constructs to Study Protein-Chaperone Interactions

Jeffrey Ting (CHiMaD), Matt ***Tirrell*** *(UC), Raphael Lee (UC)*

In biology, the current mechanism of how protein chaperones stabilize proteins remains unclear; we are pursuing the development of a model block polymer system to elucidate structure-property relationships towards new biomaterials that exploit identified design parameters.

Rheology, Scattering, and Dynamics of Polyelectrolyte Complex Gels

Jeffrey Ting (CHiMaD), Matt ***Tirrell*** *(UC), Vivek* ***Prabhu*** *(NIST), Debra* ***Audus*** *(NIST), Jack* ***Douglas*** *(NIST), Kenneth* ***Shull*** *(NU)*

This collaboration aims to use the unique capabilities of NIST to quantify polyelectrolyte systems using rheology and small-angle neutron scattering (Rheo-SANS), fluorescence correlation spectroscopy (FCS), and quartz crystal microbalance (QCM).

Phase Behavior and Salt Partitioning in Polyelectrolyte Complexes

Jeffrey Ting (CHiMaD), Matt ***Tirrell*** *(UC), Juan* ***de Pablo****, Vivek* ***Prabhu*** *(NIST), Debra* ***Audus*** *(NIST)*

This collaboration has extended the understanding of the fundamental phase behavior of polyelectrolyte complexes through active discussions with NIST and CHiMaD group members. Specifically, the effects of hydrophobicity have been examined in a new system using a suite of solid- and solution-state characterization tools.

23.6.3 Publications and Presentations

3 Number of CHiMaD-supported publications in 2018. Please see chapter 27 for details [27.77, 27.78, 27.82].

19 Number of presentations on CHiMaD supported research in 2018, please see section 26.13 for the complete list.

23.7 Michael Heiber (CHiMaD)

Organic Bulk Heterojunction Polymer Solar Cells
Supervisors: Dean DeLongchamp (NIST), Luping Yu (UC)

Bio Dr. Heiber obtained a B.S. in Materials Science and Engineering at the University of Illinois at Urbana-Champaign in 2007 and a Ph.D. in Polymer Science from the University of Akron in 2012. Through previous postdoctoral positions working with Prof. Vladimir Dyakonov at the University of Würzburg, Prof. Carsten Deibel at Chemnitz University of Technology, and Prof. Thuc-Quyen Nguyen at the University of California, Santa Barbara, he has developed a broad set of computational and experimental skills for investigating the physics of organic semiconductors. In July 2017, he joined the DeLongchamp group at NIST as a NIST-CHiMaD postdoctoral fellow where he has been working to combine advanced materials morphology measurements, kinetic Monte Carlo simulations, and electrical device characterization techniques to reveal the complex processing-structure-property relationships in organic solar cells.

23.7.1 Research Accomplishments

In pursuit of more accurate and reliable measurements for the complex charge transport and recombination processes that occur in bulk heterojunction OPVs, Dr. Heiber developed a new measurement technique called impedance-photocurrent device analysis (IPDA). A critical benefit of the technique is that it allows the determination of several detailed electrical characteristics of real devices under steady state, maximum power operating conditions. Using this technique, we fabricated and characterized a broad range of polymer:fullerene OPVs and demonstrated unprecedented self-consistency between the determined charge carrier mobility and bimolecular charge recombination coefficient with the device fill factor, as shown in Figure 23.11. This work is now published in Energy and Environmental Science. Building on this effort, I have constructed a new, highly automated IPDA measurement setup at NIST. Working with a CHiMaD funded NIST Summer Undergraduate Research Fellow from Northwestern University, I have developed the new IPDA setup to have significantly reduced measurement uncertainty and higher throughput that allows the ability to quantify the uncertainty due to sample variability. Our first case study using the new measurement setup will be published in early 2019.

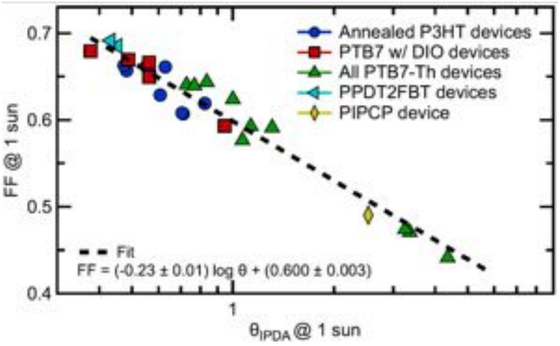


Figure 23.11: Solar cell fill factor (FF) under 1 sun illumination as a function of the competition factor () determined using the impedance-photocurrent device analysis (IPDA) technique.

To build more quantitative structure-property relationships that can capture how the detailed

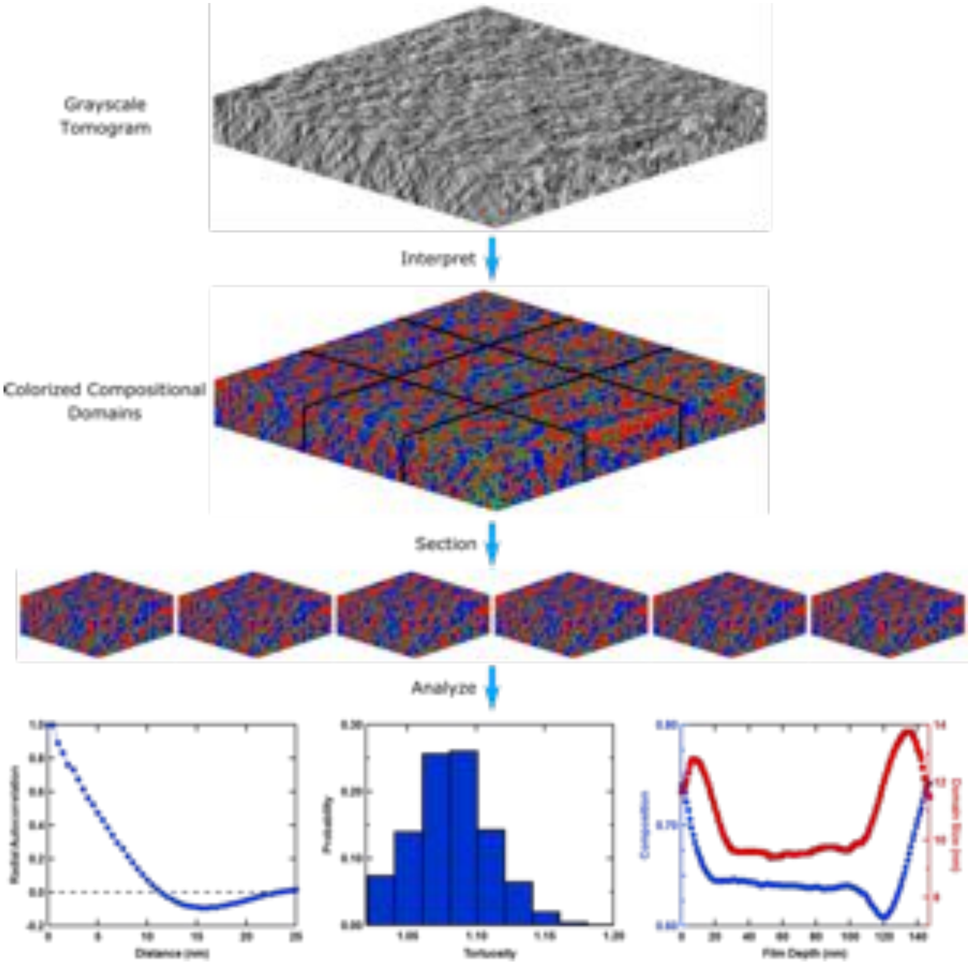


Figure 23.12: Illustrated workflow for importing, interpreting, and analyzing electron tomography data.

mesoscale phase morphology impacts the charge transport and recombination processes in bulk heterojunction OPVs, I developed an open-source software tool (*Ising_OPV*) for importing, interpreting, and analyzing experimental electron tomography datasets and then preparing morphology sets for kinetic Monte Carlo simulations, as illustrated in Figure 23.12. This software tool is now published in the Journal of Open Source Software [27.86]. In conjunction with the morphology software tool, I have also been developing an open-source kinetic Monte Carlo software tool (*Excimontec*) for simulating a wide variety of organic electronic device measurements and phenomena. This final stable release of this software tool, and our first case study using these state-of-the-art software tools to perform long-range charge transport simulations on morphology sets derived from electron tomography measurements will be published in early 2019.

23.7.2 Collaborations

Simulation and Modeling of Organic Semiconductor Devices

Michael Heiber (CHiMaD), Carsten Deibel, (Chemnitz University of Technology)

This collaboration focusses on compiling a comprehensive review of efforts using kinetic Monte Carlo and drift-diffusion simulation techniques to understand and develop organic electronic devices, including solar cells, light emitting diodes, and transistors.

Quantifying Charge Carrier Extraction and Recombination in Organic Solar Cells under Operating Conditions
Michael Heiber (CHiMaD), Thuc-Quyen Nguyen (University of California, Santa Barbara)
Development of a new measurement technique, impedance-photocurrent device analysis, for quantitatively measuring the competition between charge carrier extraction and recombination in bulk heterojunction organic solar cells under operating conditions.

Integrating Advanced Morphology Measurements and Kinetic Monte Carlo Simulations to Elucidate Structure-Property Relationships in Bulk Heterojunction OPVs
*Michael Heiber (CHiMaD), Andrew A. **Herzing** (NIST), Lee **Richter** (NIST), Dean **DeLongchamp** (NIST)*
Development and use of a unique multidisciplinary approach that brings together state-of-the-art simulation and experimental morphology characterization techniques to develop improved physical models for long-range charge transport in bulk heterojunction OPVs.

Understanding Phase Morphology Evolution during Thermal Annealing and its Impact on Charge Transport and Charge Recombination in Small Molecule OPVs
*Michael Heiber (CHiMaD), Alex **Bourque** (NIST), Andrew A. **Herzing** (NIST), Lee **Richter** (NIST), David Jones (UMelbourne), Dean **DeLongchamp** (NIST)*
Developing new, automated impedance-photocurrent device analysis measurement setup at NIST and quantifying how charge transport and charge recombination kinetics evolve following thermal annealing of BQR:PC71BM blends.

23.7.3 Technology Transfer

Software
Ising_OPV
Michael Heiber (CHiMaD)
Date: 11/30/2018
Distribution: v4.0 Final Release
Link: `https://github.com/MikeHeiber/Ising_OPV`
The Ising_OPV software package can be used to create and analyze bulk heterojunction morphologies for further use in kinetic Monte Carlo simulation tools, such as Excimontec. This package implements an Ising-based model to quickly and efficiently generate three-dimensional bulk heterojunction morphologies on a cubic lattice in a parallel computing environment. In addition, morphologies derived from other simulations models or experimental measurements can be imported into the tool. Generated or imported morphologies are then rigorously analyzed to determine important morphological features such as the domain size, tortuosity, and the interfacial area to volume ratio. This work was published in Journal of Open Software [27.86].

Software
KMC_Lattice
Michael Heiber (CHiMaD)
Date: 11/01/2018
Distribution: v2.0 Open Beta
Link: `https://github.com/MikeHeiber/KMC_Lattice`
KMC_Lattice is an object-oriented C++ software package that contains a general framework for lattice kinetic Monte Carlo (KMC) simulations. This framework consists of a number of utility functions and base classes that must be extended to create a fully operational KMC simulation. The goal of this package is to be robust, reliable, and flexible

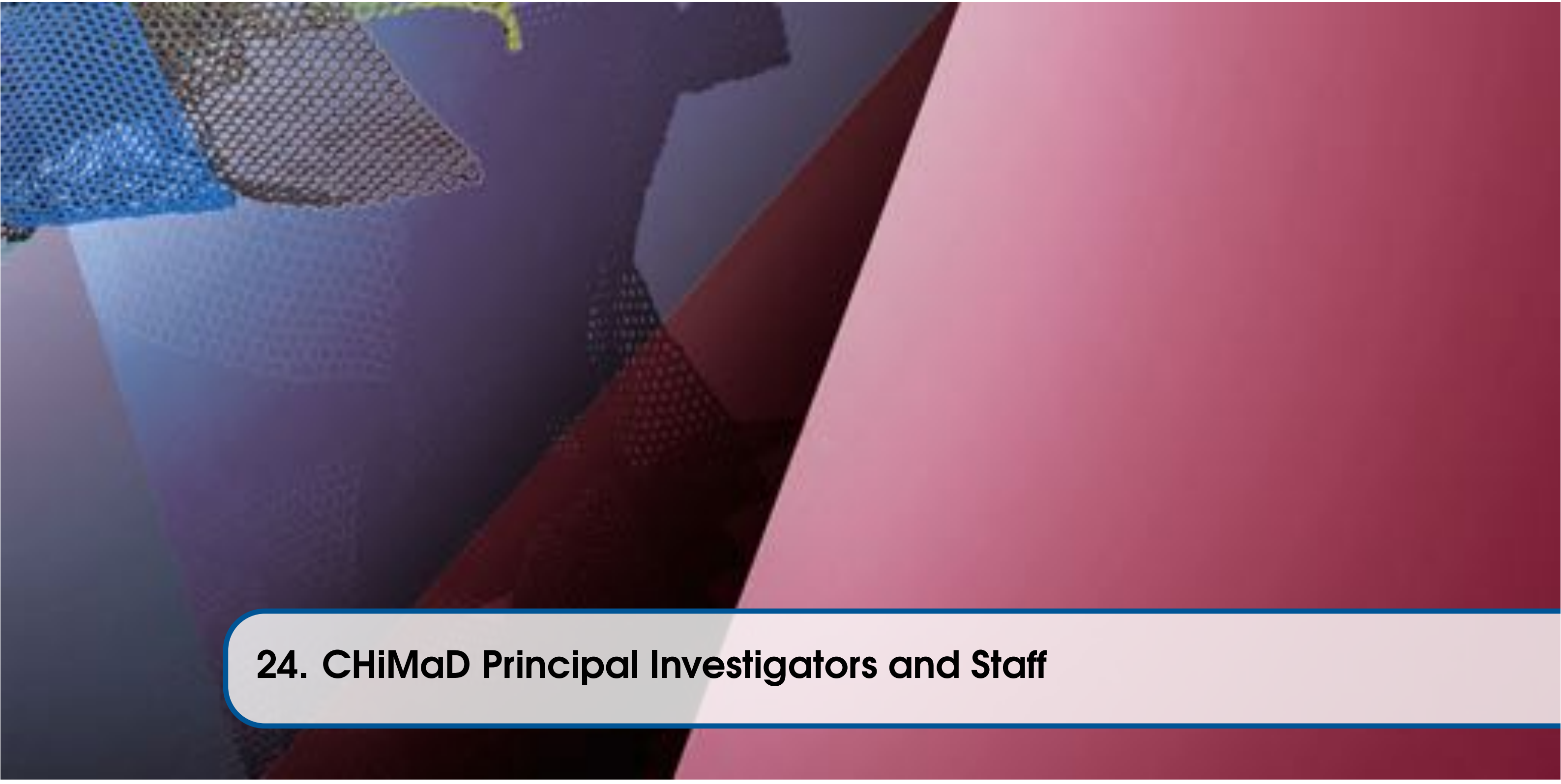
so that users can easily develop KMC simulations for a wide variety of different scientific problems without the need to start from scratch. This work was published in Journal of Open Software [27.88].

Software
Excimontec
Michael Heiber (CHiMaD)
Date: 11/01/2018
Distribution: v1.0 Open Beta
Link: `https://github.com/MikeHeiber/Excimontec`
Excimontec represents an honest effort to bring the community together around a well-tested, optimized, reliable, and accessible open-source tool for performing KMC simulations of organic electronic devices. The software is being developed in modern C++ and is optimized for efficient execution on high performance computing clusters using MPI. This software package uses object-oriented design and extends the KMC_Lattice framework.

23.7.4 Publications and Presentations

4 Number of CHiMaD-supported publications in 2018. Please see chapter 27 for details [27.85, 27.86, 27.87, 27.88].

5 Number of presentations on CHiMaD supported research in 2018, please see section 26.13 for the complete list.



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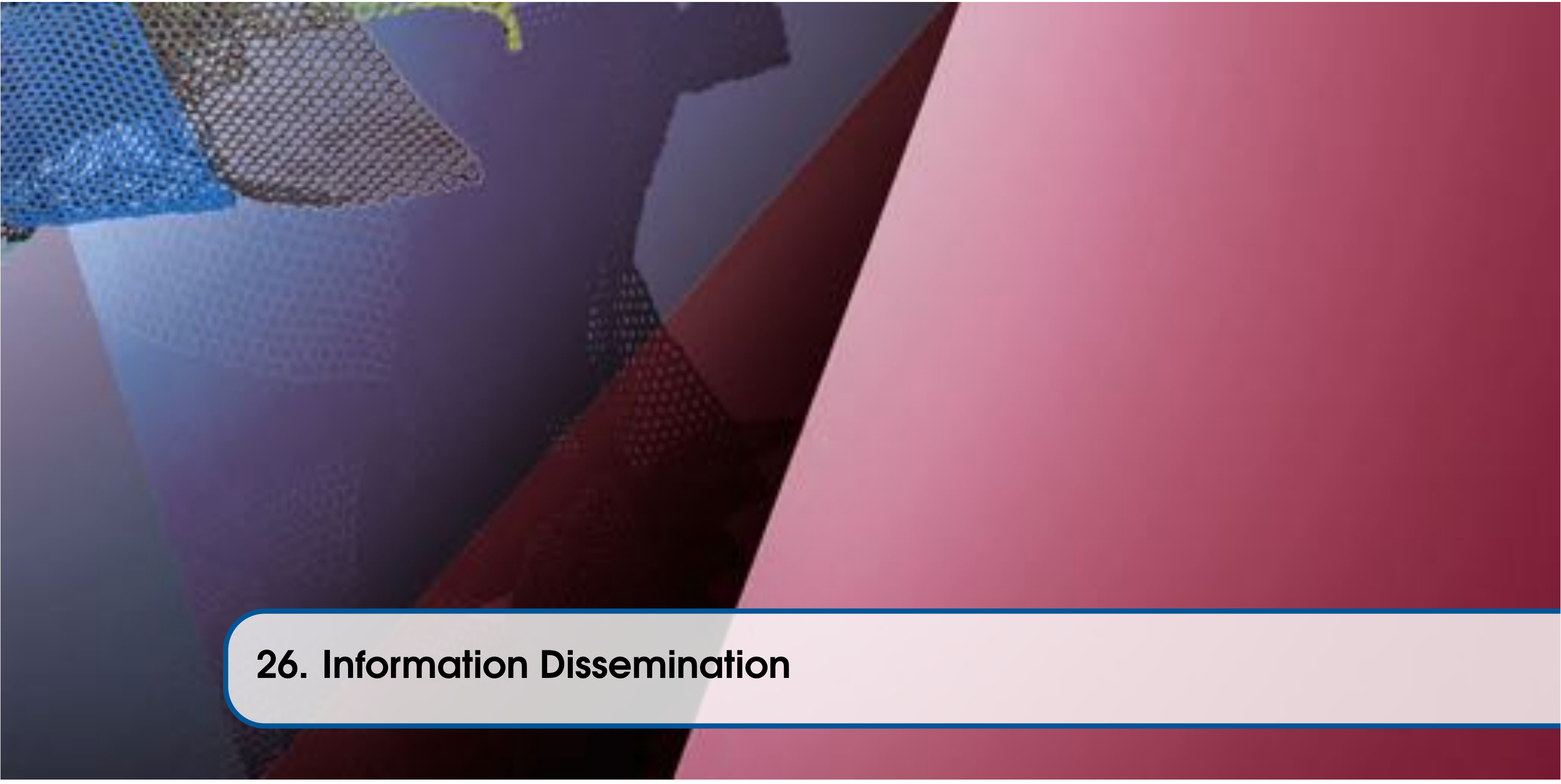


26. Information Dissemination

26.1 Low-Dimensional Nanoelectronic Materials

1. **M. C. Hersam** *(Invited)* "Nanostructured graphene-coated cathodes for high-performance lithium-ion batteries", Materials Research Society Fall Meeting, Boston, Massachusetts, USA (11/29/18)
2. **M. C. Hersam** *(Invited)* "Controlling and tailoring the electronic properties of chemically reactive 2D materials," Materials Research Society Fall Meeting, Boston, Massachusetts, USA (11/28/18).
3. **M. C. Hersam** *(Invited)* "Mixed-dimensional heterostructures as a platform for beyond von Neumann computing," Nanotechnology-Enabled Beyond-von-Neumann Computing Tutorial Workshop, Adelphi, Maryland, USA (11/13/18).
4. **M. C. Hersam** *(Plenary)* "Mixed-dimensional van der Waals heterostructures for electronic and energy applications," Hamline University Kay Malmstrom Lecture in Physics, Saint Paul, Minnesota, USA (11/9/18).
5. **M. C. Hersam** *(Invited)* "Atomically precise characterization and control of emerging 2D materials," Hamline University Physics Seminar Series, Saint Paul, Minnesota, USA (11/8/18).
6. **M. C. Hersam** *(Invited)* "Nanostructured graphene-coated cathodes for high-performance lithium-ion batteries," 14th Lithium Battery Materials and Chemistries Conference, Arlington, Virginia, USA (11/2/18).
7. **M. C. Hersam** *(Invited)* "Two-dimensional transition metal dichalcogenide memristors and memtransistors," Non-Volatile Memory Technology Symposium, Sendai, Japan (10/23/18).
8. **M. C. Hersam** *(Invited)* "Molecular engineering of chemically reactive two-dimensional materials," Frontiers of Molecular Engineering Symposium, Chicago, Illinois (9/28/18).
9. **M. C. Hersam** *(Invited)* "Introducing the nemtransistor: A potential foundational circuit element for neuromorphic computing," Illinois Ignite, Rosemont, Illinois, USA(9/27/18).
10. **M. C. Hersam** *(Plenary)* "Printable carbon and related two-dimensional nanomaterial inks for electronic and energy applications," International Graphene Innovation Conference, Xi'an China (9/19/18).
11. **M. C. Hersam** *(Invited)* "Mixed-dimensional van der Waals heterostructures for

electronic and energy applications," Shaanxi University of Science and Technology Seminar Series, Xi'an, China (9/18/18).

12. **M. C. Hersam** *(Invited)* "Printable two-dimensional nanomaterial inks for electronic and energy applications," PPG Coatings Innovation Center Seminar Series, Pittsburgh, Pennsylvania, USA (9/13/18).
13. **M. C. Hersam** *(Invited)* "Mixed-dimensional van der Waals heterostructures for electronic and energy applications," Micron Technology Seminar Series, Boise, Idaho, USA (9/7/18).
14. **M. C. Hersam** *(Invited)*"Mixed-dimensional van der Waals heterostructures for electronic and energy applications," Boise State University Materials Science and Engineering Seminar Series, Boise, Idaho (9/7/18).
15. **M. C. Hersam** *(Invited)* "Printable two-dimensional nanomaterial inks for electronic and energy applications," 256th American Chemical Society Meeting, Boston, Massachusetts, USA (8/19/18).
16. **M. C. Hersam** *(Invited)* "Nanoelectronic material inks for electronic devices and batteries," presented orally by M. C. Hersam at Huawei University Days, Schaumburg, Illinois, USA (8/2/18).
17. **M. C. Hersam** *(Invited)* "Interfacial engineering of two-dimensional nanoelectronic heterostructures," Northwestern University/Tel Aviv University Workshop on Recent Advances in Materials Science and Engineering, Evanston, Illinois, USA (7/17/18).
18. **M. C. Hersam** *(Invited)* "Surface and interface engineering of van der Waals heterostructures," International Conference on Chemical Bonding, Lihue, Hawaii (7/13/18).
19. **M. C. Hersam** *(Invited)* "Printable carbon and related two-dimensional nanomaterial inks for electronic and energy applications," XIV International Conference on Nanostructured Materials, Kowloon, Hong Kong (6/25/18).
20. **M. C. Hersam** *(Invited)* "Mixed-dimensional van der Waals heterostructures for electronic and energy applications," Structures and Materials Intelligence Seminar, McLean, Virginia, USA (6/13/18).
21. **M. C. Hersam** *(Plenary)* "Surface and interface engineering of van der Waals heterostructures," Carbon-Rich Molecules to Carbon-Based Materials Conference, Nassau, Bahamas (6/8/18).
22. **M. C. Hersam** *(Invited)* "Transition metal dichalcogenide memristors and memtransistors," 45th International Symposium on Compound Semiconductors, Cambridge, Massachusetts, USA (5/31/18).
23. **M. C. Hersam** *(Invited)* "Printable two-dimensional nanomaterial inks for flexible electronics and photonics," 233rd Electrochemical Society Meeting, Seattle, Washington, USA (5/15/18).
24. **M. C. Hersam** *(Invited)* "Nanostructured graphene-coated cathodes for high-performance lithium-ion batteries," 233rd Electrochemical Society Meeting, Seattle, Washington, USA (5/15/18).
25. **M. C. Hersam** *(Invited)* "Self-aligned short-channel heterojunction devices based on carbon nanotubes and related nanoelectronic materials," 233rd Electrochemical Society Meeting, Seattle, Washington, USA (5/15/18).
26. **M. C. Hersam** *(Invited)* "Enhanced nanostructured Li-ion battery cathode materials via conformal graphene dispersion," International Battery Seminar and Exhibit, Fort Lauderdale, Florida, USA (3/28/18).
27. **M. C. Hersam** *(Invited)* "Mixed-dimensional van der Waals heterostructures for

electronic and energy applications," IISER Pune Physics Department Seminar Series, Pune, India (3/22/18).

28. **M. C. Hersam** *(Plenary)* "Mixed-dimensional van der Waals heterostructures," International Conference on Nanoscience and Technology, Bangaluru, India (3/21/18).
29. **M. C. Hersam** *(Invited)* "Mixed-dimensional van der Waals heterostructures for electronic and energy applications," Missouri State University Campus Wide Seminar Series, Springfield, Missouri, USA (3/2/18).
30. **M. C. Hersam** *(Invited)* "Improving lithium ion batteries with nanostructured electrodes," Missouri State University Physics, Astronomy, and Materials Science Seminar Series, Springfield, Missouri, USA (3/2/18).
31. **M. C. Hersam** *(Invited)* "Printable two-dimensional nanomaterial inks for electronic and energy applications," Graphene for US International Conference, New York, New York, USA (2/22/18).
32. **M. C. Hersam** *(Invited)* "Mixed-dimensional van der Waals heterostructures," University of Colorado Materials and Nanoscience Seminar Series, Boulder, Colorado, USA (2/19/18).
33. **M. C. Hersam** *(Invited)* "Mixed-dimensional van der Waals heterostructures," Washington University Institute of Materials Science and Engineering Seminar Series, St. Louis, Missouri, USA (2/5/18).
34. **M. C. Hersam** *(Invited)* "Mixed-dimensional van der Waals heterostructures for electronic and energy applications," University of Washington Materials Science and Engineering Seminar Series, Seattle, Washington, USA (1/22/18).
35. **L. J. Lauhon** *(Invited)* "Studies of Carrier Generation, Separation, and Transport in Mixed-Dimensional Heterojunctions by Scanning Photocurrent Microscopy" , Materials Research Society Fall Meeting, Boston, MA, USA (11/26/18).
36. **L. J. Lauhon** *(Invited)* "Variable Doping of MoS2 by Atomic Layer Deposition of Molybdenum Oxides of Controlled Stochiometry" , AiMES/Electrochemical Society Joint Meeting, Cancun, Mexico (10/2/18)
37. **L. J. Lauhon** *(Invited)* "Non-Planar Mixed-Dimensional Heterostructures for Conventional and Unconventional Computing" , Fudan University, Shanghai, China (8/31/18).
38. **L. J. Lauhon** *(Invited)* "Non-Planar Mixed-Dimensional Heterostructures for Conventional and Unconventional Computing" , Nanjing University, Nanjing, China (8/30/18).
39. **L. J. Lauhon** *(Invited)* "Non-Planar Mixed-Dimensional Heterostructures for Conventional and Unconventional Computing" , Wuhan University of Technology, Wuhan, China (8/28/18).
40. **L. J. Lauhon** *(Invited)* "Non-Planar Mixed-Dimensional Heterostructures for Conventional and Unconventional Computing" , 4th International Advances in Functional Materials Conference, Nanjing, China (8/27/18).

26.2 Directed Self-assembly of Block Copolymer Films for Lithography

1. **J. Murphy**, J. G. Raybin, M. Dolejsi, P. F. Nealey, S. J. Sibener, Time-Resolved Atomic Force Microscopy Imaging Studies of Thermal Fluctuations in Confined Block Copolymer Nanopatterns, Women in Chemistry Conference, South Bend, INWomen in Chemistry Conference, South Bend, IN, USA (10/6/18)
2. **J. Raybin**, J. G. Murphy, M. Dolejsi, S. J. Sibener, *(Poster)* Direct Imaging of

Interfacial Fluctuations in Confined Block Copolymer with In Situ Atomic Force Microscopy, AVS Symposium, Chicago, IL, USA (9/6/18)

3. **J. Raybin**, J. G. Murphy, J. Ren, M. Dolejsi, X. Chen, R. Gronheid, P. F. Nealey, S. J. Sibener, Video-Rate AFM Imaging of Block Copolymer Patterns under Environmental Control, EMRS Conference, Strasbourg, France (6/18/18)
4. **M. Dolejsi**, P. Nealey , Utilization of metal - polymer interactions for self-aligned directed self-assembly of device relevant features, 2018 SPIE Advanced Lithography, San Jose, CA, USA, (2018)
5. **X. Li**, Martinez-Gonzalez JA., de Pablo JJ., Nealey PF. Directed self-assembly of blue-phase liquid crystals by chemically patterned surfaces. (Invited talk) SPIE OPTO, San Francisco, CA, USA (1/27/18)
6. **X. Li**, J.A. Martinez-Gonzalez, J.J. de Pablo, P.F. Nealey, Chemical patterns for directed self-assembly blue phase liquid crystals with equilibrated morphologies, APS Meeting, Los Angeles, CA, USA (3/5/18)
7. **C. Zhou**, T. Kurosawa, T. Dazai, P.F. Nealey, Studying the effects of chemistry and geometry on DSA hole-shrink process in three dimensions, 2018 SPIE Advanced Lithography, San Jose, CA, USA (2018)
8. **J. Li**, P. Rincon-Delgadillo, H.S. Suh, G. Mannaert, P.F. Nealey, Kinetics of Defect Annihilation in Chemo-epitaxy DSA. DSA Symposium, Sapporo, Hokkaido, Japan (2018)
9. **J. Li**, C. Zhou, X. Chen, P. Rincon-Delgadillo, P.F. Nealey, Directed Self-Assembly of Triblock Copolymers for Sub-10 nm Nanofabrication using Polymeric Additives. SPIE Advanced Lithography, San Jose, CA, USA (2018)
10. **J. Li** , T. Deng, X. Liu, N. Scherer, P.F. Nealey, DNA-Directed Assembly of Gold Nanoparticle Heterodimer Arrays with Well-Controlled Sub-5 nm Gaps. Bulletin of the American Physical Society, Los Angeles, CA, USA (2018)

26.3 Soft Matter Design Based on Charge Complexation

1. **B. Ma**, T. D. Nguyen, V. A. Pryamitsyn and M. Olvera de la Cruz, Electrostatic effect on nanostructure and dynamics in random ionomers, Materials Research Society, Phoenix, AZ, (4/2/18)
2. **V. Pryamitsyn**, M. Olvera de la Cruz, Physical origin and the architectural pathway to the "inverted" phases of the micro-segregated diblock copolymers, APS March Meeting, Los Angeles, CA, USA (3/5/18)
3. **H-K Kwon**, K. Shull, M. Olvera de la Cruz, Effect of Charge on Interfacial Activity and Micelle Formation of Ion-containing Block Copolymers at the Oil-Water Interface, APS March Meeting, Los Angeles, CA, USA (3/5/18)
4. **K. Sadman**, Q. Wang, Y. Chen, B. Keshavarz, Z. Jiang, K. Shull, Influence of hydrophobicity on polyelectrolyte complexation, American Chemical Society Annual Meeting, New Orleans, LA, USA (2018)
5. **D. Delgado**, K. Sadman, Q. Wang, K. Shull, Teaching a new dog old tricks: Phase inversion in polyelectrolytes, American Chemical Society Annual Meeting, New Orleans, LA, USA (2018)
6. **K. Shull** *(Invited)*, Charges, Hydrogen bonds and Salt: How to Design Functional Polyelectrolyte Complexes, Department Colloquium, Department of Mechanical Sciences, University of Illinois, IL, USA (9/4/18)
7. **K. Shull** *(Invited)*, Charges, Hydrogen bonds and Salt: How to Design Functional

Polyelectrolyte Complexes, Department Colloquium, Michigan Technological University, IL, USA (9/13/18)

8. **K. Shull** *(Invited)*, Polyelectrolyte Complex Membranes for Water Purification, Northwestern-Tel Aviv University Workshop, Northwestern University, Evanston, IL, USA (7/16/18)
9. **K. Shull** *(Invited)*, Mechanics of Polyelectrolyte Complexes, Deformation, Yield and Fracture of Polymers Conference, Herleen, Netherlands, (4/18)
10. **J. Ting**, H. Wu, A. Herzog-Arbeitman, J. Mitchell, S. Meng, M. Tirrell, "A Materials Genome Approach for Enabling Designer Block Polyelectrolytes" American Institute of Chemical Engineers (AIChE) Annual Meeting (10/28/18)
11. **L. Li**, S. Srivastava, M. Andreev, A. Marciel, J. Ting, J. de Pablo, M. Tirrell, "Phase Behavior and Salt Partitioning in Polyelectrolyte Complexes" American Institute of Chemical Engineers (AIChE) Annual Meeting (10/28/18)
12. **H. Wu**, J. Ting O. Werba, M. Tirrell, "Mechanism of Dissociation Kinetics in Polyelectrolyte Complex Micelles" American Institute of Chemical Engineers (AIChE) Annual Meeting (10/28/18)
13. **A. Marras**, J. Ting, J. Vieregg, J. Rubien, M. Tirrell, "Characterizing Micellar Assembly of Oligonucleotides with Polyelectrolytes" American Institute of Chemical Engineers (AIChE) Annual Meeting (10/28/18)
14. **H. Wu**, J. Ting, O. Werba, S. Meng, M. Tirrell, "Non-equilibrium phenomena in self-assembled polyelectrolyte complexes", International Symposium on Polyelectrolytes, Wageningen, Netherlands, (9/2018)
15. **J. Ting** "Materials Genome Approach Enables Designer Polyelectrolyte Complexes" American Chemical Society (ACS): PMSE Future Faculty Symposium, (9/18)
16. **J. Ting**, H. Wu, A. Herzog-Arbeitman, J. Mitchell, O. Werba, S. Meng, A. Neitzel, A. Marciel, M. Tirrell, "Rapid Discovery and Development of Designer RAFT Polyelectrolytes" American Chemical Society (ACS) Fall Meeting, Boston MA, USA, (09/2018)
17. **J. Ting**, H. Wu, M. Tirrell, "Life of the Polyelectrolyte: Complexation, Evolution, and Disassembly" American Chemical Society (ACS) Fall Meeting, Boston MA, USA, (09/2018)
18. **A. Marras**, J. Ting, J. Vieregg, J. Rubien, M. Tirrell, "Characterizing Micellar Assembly of Oligonucleotides with Polyelectrolytes using SAXS" American Chemical Society (ACS) Fall Meeting, Boston MA, USA, (09/2018)
19. **A. Marras**, M. Tirrell, "First Impressions: CV, Resume, & Cover Letter" Sharma's Soft Matter ODES-Lab Mentored Professional Development Seminar Series, (09/18)
20. **J. Ting**, M. Tirrell, M. "Soft Matter Design Based on Charged Complexation" Sharma's Soft Matter ODES-Lab Mentored Professional Development Seminar Series (09/18)
21. **J. Ting** *(Invited)*, "Molecular Engineering New Polymers for Designer Drug Delivery" DePaul University, Chicago, IL, USA (06/18)
22. **J. Ting**, H. Wu, O. Werba, A. Herzog-Arbeitman, S. Meng, M Tirrell, "Kinetic Pathways in Self-Assembled Polyelectrolyte Complexes" Polymer Physics Gordon Research Conference (06/18)
23. **J. Ting**, H. Wu, O. Werba, A. Herzog-Arbeitman, S. Meng, M Tirrell, "Kinetic Pathways in Self-Assembled Polyelectrolyte Complexes" Polymer Physics Gordon Research Seminar, South Hadley, MA, USA (06/18)
24. **Wu, H.** J. Ting O. Werba, S. Meng, M. Tirrell, "Non-Equilibrium Assembly and Ki-

netic Pathways in Polyelectrolyte Complexes" National Graduate Research Polymer Conference, South Hadley, MA, USA (06/18)

25. **S. Meng**, H. Wi, J. Ting, M. Tirrell, "Structure Dynamics, Conformation, Rheological Properties and Salt Responsivity of Polyelectrolyte Complexes" National Graduate Research Polymer Conference, Minnesota, MN, USA (06/18)
26. **J. Ting**, H. Wu, A. Herzog-Arbeitman, J. Mitchell, S. Meng, M. Tirrell, "A Materials Genome Approach to Designer Polyelectrolyte Complexes" National Graduate Research Polymer Conference, Minnesota, MN, USA (06/18)
27. **L. Li**, S. Srivastava, M. Tirrell, J.J. de Pablo "Phase Behavior and Industrial application of Polyelectrolyte Complexes" Chinese American Chemical Society in the Great Lakes Area: Annual Conference (5/18)
28. **A. Herzog-Arbeitman**, J. Ting, H. Wu, O. Werba, S. Srivastava, M. Tirrell, "Synthesis and structural Investigation of tailored polyelectrolyte complexes" American Chemical Society (ACS), New Orleans, LA, USA (3/13)
29. **J. Ting**, H. Wu, O. Werba, D. Schneiderman, A. Herzog-Arbeitman, J. Mitchell, S. Srivastava, M. Tirrell, M."Solution Assembly and Behavior of Modular RAFT Block Polyelectrolytes" American Physics Society (APS) March Meeting, Los Angeles, CA, USA (03/05/18)
30. **H. Wu**, J. Ting, O. Werba, S. Srivastava, M. Tirrell, "Polyelectrolyte Complex Micelles: Formation, Dissociation, and Chain Exchange Kinetics" American Physics Society (APS) March Meeting, Los Angeles, CA, USA (03/05/18)
31. **H. Wu**, J. Ting, S. Srivastava, M. Tirrell, "Structural Evolution and Phase Behaviors of Electrostatic Macromolecular Assemblies" American Physics Society (APS) March Meeting, Los Angeles, CA, USA (03/05/18)
32. **M. Andreev,** M, L. Li, S. Srivastava, M. Tirrell, A. Chremos, V. Prabhu, J. Douglas, J. de Pablo, "Coarse-grained model for polyelectrolyte complexation" American Physics Society (APS) March Meeting, Los Angeles, CA, USA (03/05/18)
33. **L. Li**, S. Srivastava, M. Tirrell, J.J. de Pablo "Phase Behavior and Salt Partitioning in Polyelectrolyte Complexes" American Physics Society (APS) March Meeting, Los Angeles, CA, USA (03/05/18)

26.4 Polymer Composites

1. **N. Hansoge**, S. Keten, Mechanical Properties of Hairy Nanoparticle Assemblies, Computational Research Day, Northwestern University, Evanston, IL, USA (4/10/18)
2. **S. Keten**, Mechanical Properties of Hairy Nanoparticle Assemblies, 18th U.S. National Congress for Theoretical and Applied Mechanics (USNCTAM), Chicago, IL, USA (6/5/18)
3. **S. Keten**, Mechanical Properties of Hairy Nanoparticle Assemblies, Society of Engineering Science (SES), Madrid, Spain (10/11/18)
4. **W. Xia**, "Achieving temperature transferable coarse graining of glass-forming polymers via energy renormalization", APS March Meeting, 2017, Los Angeles, CA, USA (03/05/18)
5. **W. Xia**, "Achieving temperature transferable coarse graining of glass-forming polymers via energy renormalization", WCCM, 2018, New York City, NY, USA (07/23/2018)
6. **R. Sheridan**, "Multi-Scale Metrology for Visualization and Characterization of Interphase Failure", Adhesion Society conference, San Diego, CA, USA (2/26/2018)

7. **R. Sheridan**, "Sensing interphase damage from fiber fracture with a fluorescent mechanophore", Mach Conference, Annapolis, MD, USA (4/4/2018)

26.5 Precipitation Strengthened Alloys

1. **G. Olson** *(Invited)*, "High-Temperature Aluminum," ONR Design Project Review, Arlington, VA, USA (1/8/18)
2. **G. Olson** *(Invited)*, "Genomic Materials Design," Honeywell National Security Campus, Kansas City MO, USA (2/2/18)
3. **G. Olson** *(Invited)*, "The Dragonslayer Sword Project," Torske Norse Club, Elks Club, Des Plaines IL, USA (3/10/18)
4. **G. Olson** *(Invited)*, "Genomic Materials Design: From CALPHAD to Flight," Hume/Rothery Symposium, TMS Annual Meeting, Phoenix AZ, USA (3/12/18)
5. **G. Olson** *(Invited)*, "Genomic Materials Design: From CALPHAD to Space," China Materials Design Conference, TMS Annual Meeting, Phoenix AZ, USA (3/12/18)
6. **G. Olson** *(Invited)*, "Materials Design: Leading by Example," Leadership Symposium, TMS Annual Meeting, Phoenix AZ, USA (3/13/18)
7. **G. Olson** *(Invited)*, "DragonSlayer: The First 20 Years," TMS Bladesmithing Symposium, TMS Annual Meeting, Phoenix AZ, USA (3/14/18)
8. **G. Olson** *(Invited)*, "Alloy Design for 3D Printing," Constellium Technical Advisory Board, Augsburg, Germany (4/24/18)
9. **G. Olson** *(Invited)*, "Perspective on Materials at Northwestern," Hilliard Alumni Symposium, Northwestern University, Evanston, IL, USA (5/19/18)
10. **G. Olson** *(Invited)*, "Materials by Design: Past & Future," CTC Materials Design Workshop, Tokyo, Japan (5/29/18)
11. **G. Olson** *(Invited)*, "Materials Design Overview," Kobelco Seminar, Tokyo, Japan (5/30/18)
12. **G. Olson** *(Invited)*, "Bulk Metallic Glass Design," Exmet Seminar, TCSAB, Stockholm, Sweden (6/12/18)
13. **G. Olson** *(Invited)*, "Advances in Materials Design," Sandvik Steel R&D Center, Sandvik, Sweden (6/13/18)
14. **G. Olson** *(Invited)*, "Genomic Materials Design: From CALPHAD to Flight," ICAMS 10 Year Review Keynote, Bochum, Germany (6/25/18)
15. **G. Olson** *(Invited)*, "Materials by Design," DLR Materials Center, Cologne, Germany (6/28/18)
16. **G. Olson** *(Invited)*, "Genomic Materials Design: From CALPHAD to Flight," Materials Genome Engineering Center Inaugural Meeting, USTB, Beijing, China (7/10/18)
17. **G. Olson** *(Invited)*, "Genomic Materials Design," CISRI Research Center Seminar, Beijing, China (7/11/18)
18. **G. Olson** *(Invited)*, "Materials Design Overview," Opening Lecture, ASM Materials Genome Teachers Camp, Northwestern University, Evanston, IL, USA (7/16/18)
19. **G. Olson** *(Invited)*, "Next Gen Hull Steels," Steel Performance Initiative Planning Meeting, NSWCCD, Bethesda MD, USA (10/22/18)
20. **G. Olson** *(Invited)*, "Genomic Materials Design: The Ferrous Frontier," Keynote Lecture, 2nd DoD Steel Summit, NSWCCD, Bethesda MD, USA (10/23/18)
21. **G. Olson** *(Invited)*, "Printable High/Temperature Aluminum by Design," Constellium Technical Advisory Board, Northwestern University, Evanston, IL, USA (10/26/18)
22. **G. Olson** *(Invited)*, "Materials Genomics: From CALPHAD to Flight," Honeywell

National Security Campus Seminar, Kansas City MO, USA (11/1/18)

23. **G. Olson** *(Invited)*, "CyberAlloys 2020: Naval Materials by Design," ONR Structural Metals Program Review, Tyson Corners MD, USA (12/5/18)
24. **G. Olson** *(Invited)*, "The New Materiallurgy: Genomic Materials Design," ASM Fellows Summit Keynote, Marco Island FL, USA (12/6/18)
25. **D.-W. Chung**, D. S. Ng, Jacques P. Toinin, E. A. Lass, D. N. **Seidman**, D. C. **Dunand**, "Effects of Cr on microstructural evolution, oxidation, and creep behavior in multicomponent Co-based γ'-strengthened superalloys", 6th International Workshop on Co-base Superalloys 2018, Royal School of Mines, Imperial College, London (6/9/18)
26. **W-K Liu** *(short course)*, M. Bessa, "Mechanistic Data-driven Multiscale Analysis and Applications", 18th U.S. National Congress for Theoretical and Applied Mechanics, Chicago, IL, USA (6/5/18)
27. **Zhengtao Gan**, Y. Lian, S. Lin, C. Yu, O. Kafka, G. J. Wagner, W-K Liu, "Multiscale Process-Structure Modeling Framework in Directed Energy Deposition of Ni-based Superalloy", 18th U.S. National Congress for Theoretical and Applied Mechanics, Chicago, IL, USA (6/5/18)
28. **Orion L. Kafka**, C. Yu, M. Shakoor, W-K. Liu, "Data-driven modeling of microstructure shape influence on high cycle fatigue life of NiTi wire", 4th International Congress on 3D Materials Science, Helsingor, Denmark, June 10-12 2018.
29. **W-K Liu** *(invited)* "Data-driven Multiscale Modeling and Design of Advanced Material Systems",13th World Congress in Computational Mechanics, New York, NY, USA, (7/22/18)
30. **W-K Liu** *(short-course)* C.T. Wu, Z. Liu, "Mechanistic Data-driven Multiscale Analysis and Applications", 13th World Congress in Computational Mechanics, New York, NY, USA (7/22/18)
31. **W-K Liu** *invited* "Data-driven Multiscale Modeling and Design of Advanced Material Systems", 13th World Congress in Computational Mechanics, New York, NY, USA (7/22/18)
32. **C. Yu**, M. Shakoor, O. L. Kafka, W-K. Liu, "Data-driven reduced-order modeling of microvoid evolution for ductile fracture", 13th World Congress in Computational Mechanics, New York, NY, USA (7/22/18)
33. **A. Umantsev**, NIST-MML/DOE-AMO Workshop, Gaithersburg, MD, USA (7/12/18)
34. **A. Umantsev** *Keynote*, A Phenomenological Model of Ductile Fracturing and Fatigue International Conference Plasticity, Damage & Fracture, San Juan, Puerto Rico (1/6/18)
35. **A. Umantsev**, The 55th Annual Meeting of the Society of Engineering Sciences, University Carlos III of Madrid, Madrid, Spain (10/12/18)

26.6 Organic Bulk Heterojunction Polymer Solar Cells

1. **T. Marks**, *(Invited)*, Inaugural Sigma-Aldrich Lecturer in Materials Chemistry, University of California Santa Barbara, 2018
2. **T. Marks**, *(Invited)*,Dow Distinguished Lecturer, Stanford University, CA, USA 2018
3. **T. Marks**, *(Invited)*,Davis Distinguished Lecturer, North Carolina State University, North Carolina, USA 2018
4. **T. Marks**, *(Invited)*,Arduengo Distinguished Lecturer, University of Alabama, Al-

abama, USA 2018

5. **T. Marks**, *(Invited)*,COPE Distinguished Lecturer, Georgia Institute of Technology, Atlanta, Georgia, USA 2018
6. **T. Marks**, *(Invited)*,Nobel Research Center Lecturer, Gwangju Institute of Science and Technology, Korea, 2018

26.7 Additive Manufacturing

1. **Wolff, S**; Wu, H; Parab N; Zhao C; Sun, T; Cao, J, "In-situ Monitoring of Solidification during Powder-deposition Based Additive Manufacturing", 2018 TMS Annual Meeting and Exhibition, Phoenix, AZ, USA
2. **Bennett, JL**; Kafka, OL; Liao, H; Wolff, SJ; Cheng, P; Hyatt, G; Ehmann, K; Cao, J, "Cooling rate effect on tensile strength of laser deposited Inconel 718", SME - NAMRC 45, Texas A&M University, College Station, TX, USA
3. **W-K Liu** *(invited)*, M. Shakoor, C. Yu, J. Gao, O. L. Kafka, W. Yan, "Proces-structure-properties relationships in additive manufacturing" NU-NIMS Materials Genome Workshop, Northwestern University, Evanston, IL, USA (3/28/18)
4. **Zhengtao Gan**, Y. Lian, S. Lin, C. Yu, O. Kafka, G. J. Wagner, W-K Liu, "Multiscale Process-Structure Modeling Framework in Directed Energy Deposition of Ni-based Superalloy", 18th U.S. National Congress for Theoretical and Applied Mechanics, Chicago, IL, USA (6/5/18)
5. **Orion L. Kafka**, S. J. Wolff, C. Yu, P. Cheng, J. L. Bennett, W-K Liu, "Deformation of Voids in Additively Manufacture Metal: In-situ Measurements and Mechanical Modeling", 18th U.S. National Congress for Theoretical and Applied Mechanics, Chicago, IL, USA (6/5/18)
6. **W-K Liu** *(invited)* "Additive Manufacturing Benchmarks 2018", Gaithersburg, Maryland, MD, USA (6/18/18)
7. **L-E Lindgren** *(short-course)*, W-K. Liu, A. To, A. Rollett, "Validated Simulation of Additive Manufacturing on Various Materials Physics and Scales", 13th World Congress in Computational Mechanics, New York, NY, USA (7/22/18)
8. **Z. Gan**, Y. Lian, W-K. Liu, "Predictive Multiphysics Process-Structure Models for Additive Manufacturing", 13th World Congress in Computational Mechanics, New York, NY, USA (7/22/18)

26.8 Uncertainty of Thermodynamics Data and Phase Equilibria

1. **M. Stan** *(Keynote)*, N. H. Paulson and E. Jennings, Improved Thermodynamic Models of Complex Materials, Big Data Challenges for Predictive Modeling of Complex Systems Conference, Hong Kong (11/26/18).
2. **N. H. Paulson** *(Invited)*, E. Jennings, and M. Stan, Bayesian Analysis of Thermodynamic Data, SIAM Conference on Mathematical Aspects of Materials Science, Portland, OR, USA (7/9/18).
3. **N. H. Paulson** *(Invited)*, E. Jennings and M. Stan, A Bayesian Framework for the Development of Thermodynamic Property Models with Quantified Uncertainty, 47th CALPHAD International Conference, Juriquilla Mexico (5/28/18).
4. **N. H. Paulson**, N. B. Gould, A. Yanguas-Gil, M. De La Cinta Lorenzo Martin, D. Singh, A. Greco and M. Stan, Reduced-Order Modeling of Selective Laser Sintering, Additive Manufacturing Conference, Argonne, IL, USA (5/22/18).

5. **M. Stan** *(Invited)*, Future Knowledge: Humans and Machines, Future Compute: A celebration of the Computation Institute, University of Chicago, Chicago, IL, USA (5/16/18).
6. **M. Stan** *(Keynote)*, Computational Science, Art and Society, 30th International Conference on Parallel Computational Fluid Dynamics, Parallel CFD2018, Indianapolis, IN, USA (5/15/18).
7. **N. H. Paulson**, E. Jennings and M. Stan, The Development of the Thermodynamic Property Models with Quantified Uncertainty, seminar at the National Institute of Standards and Technology (NIST), Gaithersburgh. MD, USA (5/10/18).
8. **M. Stan** *(Keynote)*, Computational Materials Science: Humans and Machines, Seminar, Notre Dame University, Indiana, IN, USA (5/4/18).
9. **M. Stan** *(Invited)* and N. H. Paulson, Uncertainty of Thermodynamic Data for Materials Design, TMS Annual Meeting & Exhibition, Phoenix, AZ, USA (3/11/18).
10. **M. Stan**, Computational Thermodynamics: Humans and Machines, TMS Annual Meeting & Exhibition, Phoenix, AZ, USA (3/1/18).
11. **Y. Ghadar**, L. Gallington, L. Skinner, J.K. Weber, Sergey V. Ushakov, A. Novrotsky, A. Vazquez Mayagoitia, and M. Stan, Determining the Structural Properties of Hafnia Utilizing Reactive Force Fields, American Physical Society March Meeting, Los Angeles, CA, USA (3/5/18).

26.9 Impact Mitigation

1. **H. Jaeger** *invited* Architectured Particulate Materials, IUTAM Symposium on Architectured Materials Mechanics, Gleacher Center, Chicago, IL, USA (9/17/18)

26.10 Phase Field Methods

1. **O.G. Heinonen and A.M. Jokisaari** Status of Benchmarks and Future Needs or Directions, CHiMaD Phase Field Methods Workshop 6, CHiMaD HQ, Evanston, IL, USA (2/20/18)
2. **O.G. Heinonen** Uncertainty Quantification, Design of Experiment, ANOVA Tutorial, CHiMaD Phase Field Methods Workshop 6, CHiMaD HQ, Evanston, IL, USA (2/20/18)
3. **O.G. Heinonen and A.M. Jokisaari** Benchmark Problem Updates , CHiMaD Phase Field Methods Workshop 7, CHiMaD HQ, Evanston, IL, USA (9/25/18)
4. **P. Voorhees** *(Invited)* Phase Field Methods, GIAN Lecture Series, Indian Institute of Technology Bombay, Mumbai, India (3/18)
5. **P. Voorhees** *(Invited)* Computational Materials Science using Phase Field Methods, Chemomechanics Summer School, Maria-Laach, Germany (6/18)
6. **P. Voorhees** *(Invited)* The Phase Field Method: Crystal Structures and Facets, Multiscale Materials Modeling, Osaka, Japan (10/18)
7. **P. Voorhees** *(Invited)* In-situ Characterization of Phase Transformations: Dendritic Solidification, Materials Science and Engineering 2018, Darmstadt, Germany (10/18)
8. **T. Stan**,Y. Sun, K. Elder, X. Xiao, M. Rappaz, P. Voorhees*Poster Presentation* 4D X-Ray Tomography Investigation of Dendrite Evolution in Aluminum Alloys, 3DMS 2018, Helsingor (Elisnore), Denmark (6/11/2018)
9. **T. Stan**,Y. Sun, K. Elder, X. Xiao, M. Rappaz, P. Voorhees 4D X-Ray Tomography Investigation of Dendrite Evolution in Aluminum Alloys, 3DMS 2018, Helsingor

(Elisnore), Denmark (6/13/2018)

10. **T. Stan**,Y. Sun, K. Elder, X. Xiao, M. Rappaz, P. Voorhees*Poster Presentation* 4D X-Ray Tomography Investigation of Dendrite Evolution in Aluminum Alloys, NUPF Current Research & Future Careers Symposium, Chicago, IL, USA (8/30/2018)

26.11 Data Mining and Analytics

1. **A. Choudhary** *(Invited)* Accelerating Materials Design and Discovery with Data Science and Machine Learning, INTERNATIONAL ADVANCED RESEARCH WORKSHOP ON HIGH PERFORMANCE COMPUTING, Cetraro, Italy (7/2/18)
2. **A. Agrawal**, Materials Informatics and Big Data: Realization of "Fourth Paradigm" of Science in Materials Science, ACS Midwest Regional Meeting (MWRM 2018), Ames IA, USA. (10/23/18)
3. **A. Agrawal** *(Invited)* Materials Informatics and Big Data: Realization of "Fourth Paradigm" of Science in Materials Science, XXVII International Materials Research Congress (MRS-Mexico), Cancun, Mexico (9/20/18)
4. **A. Agrawal** *(Invited)* Materials Informatics and Big Data: Realization of "Fourth Paradigm" of Science in Materials Science, Artificial Intelligence for Materials Science Workshop (AIMS), National Institute of Standards and Technology, Gaithersburg, MD, USA 9/7/18
5. **A. Agrawal** *(Invited)* Materials Informatics and Big Data: Realization of "Fourth Paradigm" of Science in Materials Science, 13th World Congress on Computational Mechanics (WCCM), New York City, NY, USA (7/25/18)
6. **A. Agrawal** *(Invited)* High Performance Data Mining: An Essential Paradigm for Interdisciplinary Big Data Analytics, Tata Consultancy Services Visit, TRDDC/TCS, Pune, India (7/18/18)
7. **A. Agrawal** *(Invited)* Materials Informatics and Big Data: Realization of "Fourth Paradigm" of Science in Materials Science, Toyota Visit, Toyota Motor Corporation, Toyota-shi, Japan (6/13/18)
8. **A. Agrawal** *(Invited)* Materials Informatics and Big Data: Realization of "Fourth Paradigm" of Science in Materials Science, Machine Learning in Science and Engineering, Carnegie Mellon University, Pittsburgh PA, USA. (6/8/18)
9. **A. Agrawal**, Deep Transfer Learning Based Pavement and Structural Health Monitoring, Machine Learning in Science & Engineering, Carnegie Mellon University, Pittsburgh PA, USA (6/8/18)
10. **A. Agrawal** *(Invited)* Materials Informatics and Big Data: Realization of "Fourth Paradigm" of Science in Materials Science, Machine Learning in Materials Science Workshop, University of Utah, Salt Lake City, UT, USA (4/6/18)
11. **A. Agrawal**, Materials Informatics and Big Data: Realization of "Fourth Paradigm" of Science in Materials Science, MRS Spring Meeting & Exhibit, Phoenix AZ, USA. (4/4/18)
12. **A. Agrawal** *(Invited)* Data-Driven Approaches for Steel Fatigue Strength Prediction, TMS 2018: Fatigue in Materials: Fundamentals, Multiscale Modeling and Prevention - Data-driven Investigations of Fatigue, Phoenix AZ, USA (3/12/18)
13. **A. Agrawal** *(Invited)* Materials Informatics and Big Data: Realization of "Fourth Paradigm" of Science in Materials Science, Materials Science Division Colloquium, Argonne National Laboratory, Lemont IL, USA (2/15/18)
14. **A. Paul**, CheMixNet: Mixed DNN Architectures for Predicting Chemical Proper-

ties using Multiple Molecular Representations, 2018 NIPS workshop on Machine Learning for Molecules and Materials (MLMM 2018), Montreal, Canada (12/18)

15. **C. Wolverton** *(Invited, Short Course Instructor)* TMS Machine Learning for Materials Science 2018, Pittsburgh, PA, USA (9/25/18)
16. **W. Chen** *(Invited)* Design of Manufacturable Multiscale Structures using Robust Topology Optimization, IUTAM Symposium on When topology optimization meets additive manufacturing - theory and methods, Dalian, China, (10/8/18)
17. **W. Chen** *(Semi-Plenary)* A Data-Driven Stochastic Multiscale Analysis Framework for Design of Microstructural Material Systems, WCCM (World Congress of Computational Mechanics), New York, NY (7/23/18)
18. **W. Chen** *(Keynote)* Multiscale and Multidimensional Quantification and Propagation of Manufacturing Induced Uncertainty, ISFA (International Symposium on Flexible Automation), Kanazawa, Japan, (7/15/18)
19. **W. Chen** *(Invited)* Multiscale and Multidimensional Quantification and Propagation of Uncertainty in Integrated Computational Materials Engineering, Mathematics Colloquium, Illinois Institute of Technology, (11/12/18)
20. **W. Chen** *(Invited)* Computational Design and Data Science for Accelerating the Innovation of Microstructural Material Systems, Mechanical and Civil Engineering Colloquium, John Hopkins University (4/26/18)
21. **W. Chen** *(Invited)* Computational Design and Data Science for Accelerating the Innovation of Microstructural Material Systems, School of Mechanical Engineering, Shanghai Jiao Tong University, China, (4/12/18)
22. **W. Chen** *(Invited)* Design Engineered Materials and Structural Systems, School of Mechanical Engineering, Dalian University and Technology, China, (4/10/18)
23. **W. Chen**, Li, X., Yang, Z., Brinson, L.C., Choudhary, A.N., Agrawal, A. *(Invited)* A Deep Adversarial learning Methodology for Designing Microstructural Materials Systems, ASME 2018 International Design Engineering Technical Conferences & Computers and Information in Engineering Conference, DETC2018/DAC-85633, August 26-29, Quebec City, Quebec, Canada.(9/26/18)
24. **W. Chen**, Ghumman, U.F., Iyer, A., Dulal, R., Wang, A., Munshi, J., Chien, T., Balasubramanian, G. *(Invited)* A Spectral Density Function Approach for Design of Organic Photovoltaic Cells, ASME 2018 International Design Engineering Technical Conferences & Computers and Information in Engineering Conference, DETC2018-86154, Quebec City, Quebec, Canada. (9/26/18)

26.12 Materials Data Facility

1. **I. Foster**, R. Wagner, N. Saint, E. Blau, Chard, Y. Nand Babuji, L. Ward, B. Blaiszik, M. Papka "Building an ALCF Data Service: Interactive, scalable, reproducible data science", SuperComputing 2017, Nov. 14, 2017, Denver, CO, USA
2. **B. Blaiszik**, I. Foster, L. Ward, J. Gaff et al. "Building an end-to-end Data Ecosystem to Support Materials Science Research", Materials Science Division Colloquium, Dec. 14, 2017, Lemont, IL, USA
3. **L. Ward**. "Deep Learning and Materials Design." Argonne Deep Learning Summit, Lemont, IL, USA
4. **B. Blaiszik**, I. Foster, L. Ward, J. Gaff et al. "Building an end-to-end Data Ecosystem to Support Materials Science Research", CHiMaD Phase Field Workshop, Feb. 22, 2018, Evanston, IL, USA

5. **I. Foster**, "Going Smart and Deep on Materials, Materials and Manufacturing Innovation Keynote Session", TMS Annual Meeting, March 13, 2018, Phoenix, AZ, USA
6. **I. Foster**, S. Tuecke. Globus 2018, Beyond File Transfer: GlobusWorld Keynote, April 25, 2018, Chicago, IL, USA
7. **I. Foster** "The Materials Data Facility and DLHub", Department of Energy EMN Machine Learning/Data Science Meeting, May 2, 2018, Livermore, CA, USA
8. **L. Ward**. "Machine Learning for Materials Design." Machine Learning for Materials Science, May, 3, 2018, Espoo, Finland
9. **B. Blaiszik**, I. Foster, L. Ward, J. Gaff et al. "Building an end-to-end Data Ecosystem to Support Materials Science Research", CHiMaD Phase Field Workshop, Feb. 22, 2018, Evanston, IL, USA
10. **L. Ward**. "Machine Learning for Materials Design." CECAM Workshop on Machine Learning at Interfaces, June, 6, 2018, Lausanne, Switzerland
11. **I. Foster**."Data Automation at Light Sources: Experiments and Lessons Learned", International Conference on Synchrotron Radiation Instrumentation, June 12, 2018, Taipei, Taiwan, USA
12. **B. Blaiszik** I. Foster, L. Ward, J. Gaff et al. "Building an end-to-end Data Ecosystem to Support Materials Science Research", Computation Energy and Life Sciences Coffee Hour Talk, June. 13, 2018, Lemont, IL, USA
13. **B. Blaiszik**, I. Foster, L. Ward, J. Gaff et al. "Building an end-to-end Data Ecosystem to Support Materials Science Research", Argonne Summer Student Seminar, June. 21, 2018, Lemont, IL, USA
14. **B. Blaiszik**, I. Foster, L. Ward, J. Gaff et al. "Building an end-to-end Data Ecosystem to Support Materials Science Research", Midwest Big Data Hub and National Data Service Data Science Tools & Methods Workshop, July 11, 2018, Rosemont, IL, USA
15. **I. Foster**, "A Global Research Data Platform", Midwest Big Data Hub and National Data Service Data Science Tools & Methods Workshop, July 11, 2018, Rosemont, IL, USA
16. **B. Blaiszik**, I. Foster, L. Ward, J. Gaff et al. "Building an end-to-end Data Ecosystem to Support Materials Science Research", Gaps and Opportunities for ML and Geostatistics in Geospatial Aspects of Natural Science Domains (Ecology, Hydrology, Soil Science and Agriculture), Aug 2, 2018, Lemont, IL, USA
17. **L. Ward**. "Machine Learning and DFT for Materials Discovery" Artificial Intelligence in Materials Science , August 7, 2018, NIST, Gaithersburg, MD, USA
18. **B. Blaiszik**, I. Foster, L. Ward, J. Gaff et al. "Building an end-to-end Data Ecosystem to Support Materials Science Research", Workshop on Tools and Approaches for Publishing Reproducible Research, Sept 13, 2018, Rosemont, IL, USA
19. **L. Ward**. "Reproducibility in Materials Data Analytics" Workshop on Tools and Approaches for Publishing Reproducible Research, Sept 13, 2018, Rosemont, IL, USA
20. **L. Ward**. "Systems Briefing: Community Materials Databases". Gap Analysis: Materials Discovery through Data Science at Advanced User Light Sources, Oct 4, 2018, Albuquerque, NM, USA
21. **L. Ward**. "A Practical Tutorial for Simple and Reproducible Materials Informatics". CHiMaD Materials Genome Initiative Seminar Series, Oct 16, 2018, Evanston, IL, USA

22. **B. Blaiszik**, J. Gaff, I. Foster. *(webinar)* "Integrative Materials and Design (IMaD): Helping to Connect the Materials Data Community". South Big Data Hub Seminar Series, Nov 2, 2018.
23. **B. Blaiszik**, J. Gaff, I. Foster. "Integrative Materials and Design (IMaD): Helping to Connect the Materials Data Community". Midwest Big Data Hub All-hands Meeting, Nov 6, 2018, Cleveland, OH, USA
24. **L. Ward**. "Systems Briefing A: Materials Databases." Gap Analysis: Materials Discovery through Data Science at Advanced User Light Sources, Oct 2, 2018, Santa Fe, NM, USA
25. **L. Ward**. "Deep Learning, HPC, and Data for Materials Design." Big Data and Extreme Computing 2, Nov 28, 2018, Bloomington, IN, USA
26. **L. Ward** *(webinar)* "Machine Learning for Materials Design." Webinar to WastePD Collaboration, Dec 12, 2018, Columbus, OH, USA

26.12.1 Globus World Tour Events Featuring MDF Capabilities

27. **S. Tuecke**, V. Vasiliadis, R. Ananthakrishnan "GlobusWorld Tour - Stanford" Feb 9. 2018, Harvard University, Cambridge, MA, USA
28. **I. Foster**, S. Tuecke, V. Vasiliadis, R. Ananthakrishnan "GlobusWorld Tour - New Zealand" Feb 12-14. 2018, Wellington, New Zealand
29. **S. Tuecke**, V. Vasiliadis, R. Ananthakrishnan "GlobusWorld Tour - NC State" March 27. 2018, Harvard University, Cambridge, MA, USA
30. **S. Tuecke**, V. Vasiliadis, R. Ananthakrishnan "GlobusWorld Tour - NYSERNet" May 1-2. 2018, Syracuse, NY, USA
31. **S. Tuecke**, V. Vasiliadis, R. Ananthakrishnan "GlobusWorld Tour - NCSA" June 11-12. 2018, University of Illinois at Urbana-Champaign, Urbana, IL, USA
32. **S. Tuecke**, V. Vasiliadis, R. Ananthakrishnan "GlobusWorld Tour - NCAR" Sept 5. 2018, National Center for Atmospheric Research, Boulder, CO, USA
33. **S. Tuecke**, V. Vasiliadis, R. Ananthakrishnan "GlobusWorld Tour - Oregon" Oct 23. 2018, University of Oregon, Eugene, OR, USA
34. **S. Tuecke**, V. Vasiliadis, R. Ananthakrishnan "GlobusWorld Tour - Botswana" Nov 9. 2018, Gaborone, Botswana
35. **I. Foster**, "Building the Modern Research Data Portal", October 18, 2017, eResearch Conference, Brisbane, Australia

26.13 NIST-CHiMaD Postdoctoral Researchers

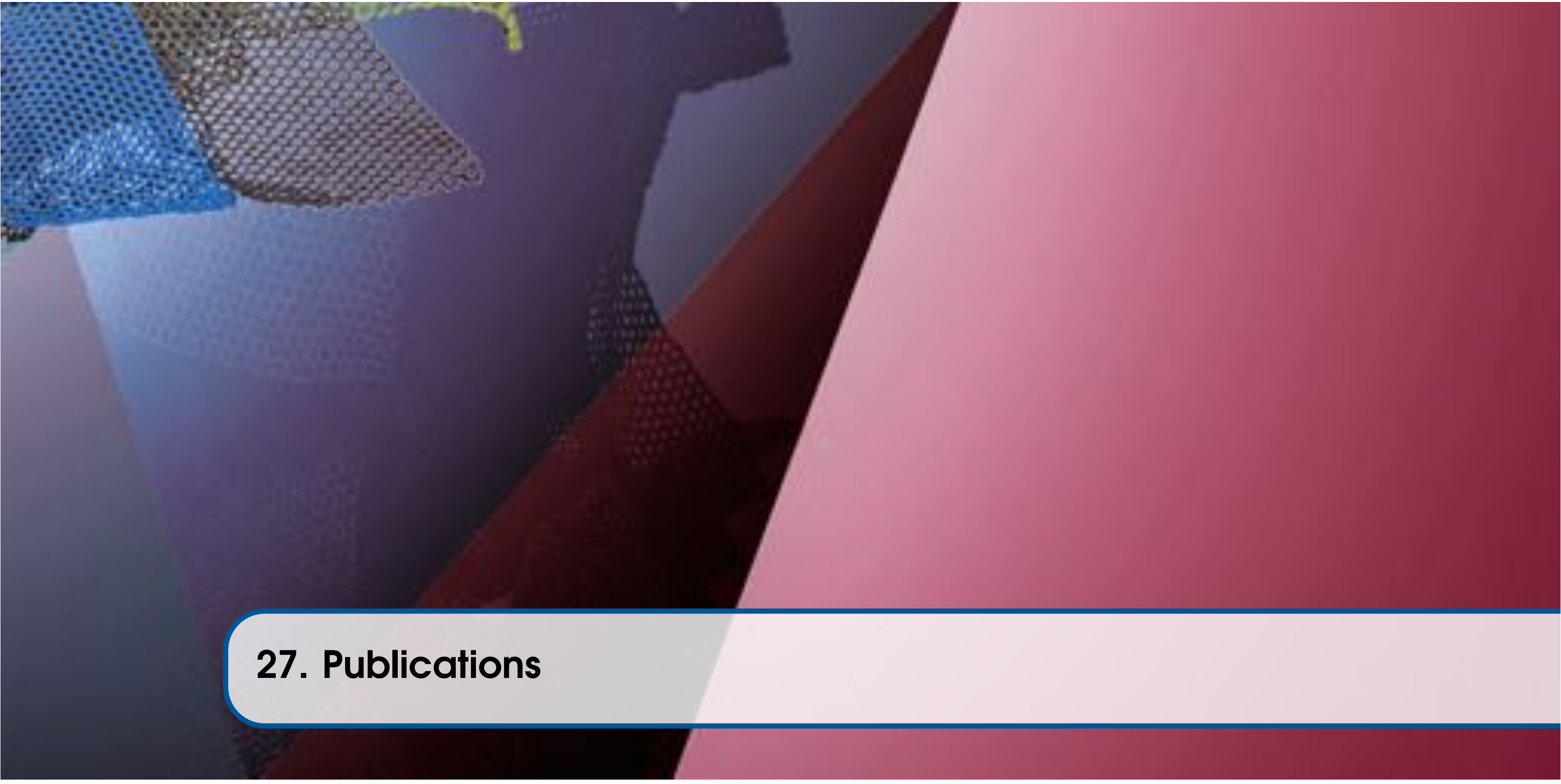
1. **W. Xia**, "Achieving temperature transferable coarse graining of glass-forming polymers via energy renormalization", APS March Meeting, Los Angeles, CA, USA (03/05/18)
2. **W. Xia**, "Achieving temperature transferable coarse graining of glass-forming polymers via energy renormalization", WCCM, New York City, NY, USA (07/23/2018)
3. **R. Sheridan**, "Multi-Scale Metrology for Visualization and Characterization of Interphase Failure", Adhesion Society conference, San Diego, CA, USA (2/26/2018)
4. **R. Sheridan**, "Sensing interphase damage from fiber fracture with a fluorescent mechanophore", Mach Conference, Annapolis, MD, USA (4/4/2018)
5. **P. Wang**, "Thermodynamic database for the Co-Al-W-Ni-Ti-Ta-Cr-Re superalloy system", TMS Annual Meeting, Pheonix, AZ, USA (3/11/18)
6. **J. Ting**, H. Wu, A. Herzog-Arbeitman, J. Mitchell, S. Meng, M. Tirrell, "A Materials

Genome Approach for Enabling Designer Block Polyelectrolytes" American Institute of Chemical Engineers (AIChE) Annual Meeting (10/28/18)

7. **L. Li**, S. Srivastava, M. Andreev, A. Marciel, J. Ting, J. de Pablo, M. Tirrell, "Phase Behavior and Salt Partitioning in Polyelectrolyte Complexes" American Institute of Chemical Engineers (AIChE) Annual Meeting (10/28/18)
8. **H. Wu**, J. Ting O. Werba, M. Tirrell, "Mechanism of Dissociation Kinetics in Polyelectrolyte Complex Micelles" American Institute of Chemical Engineers (AIChE) Annual Meeting (10/28/18)
9. **A. Marras**, J. Ting, J. Vieregg, J. Rubien, M. Tirrell, "Characterizing Micellar Assembly of Oligonucleotides with Polyelectrolytes" American Institute of Chemical Engineers (AIChE) Annual Meeting (10/28/18)
10. **H. Wu**, J. Ting, O. Werba, S. Meng, M. Tirrell, "Non-equilibrium phenomena in self-assembled polyelectrolyte complexes", International Symposium on Polyelectrolytes, Wageningen, Netherlands, (9/2018)
11. **J. Ting** "Materials Genome Approach Enables Designer Polyelectrolyte Complexes" American Chemical Society (ACS): PMSE Future Faculty Symposium, (9/18)
12. **J. Ting**, H. Wu, A. Herzog-Arbeitman, J. Mitchell, O. Werba, S. Meng, A. Neitzel, A. Marciel, M. Tirrell, "Rapid Discovery and Development of Designer RAFT Polyelectrolytes" American Chemical Society (ACS) Fall Meeting, Boston MA, USA, (09/2018)
13. **J. Ting**, H. Wu, M. Tirrell, "Life of the Polyelectrolyte: Complexation, Evolution, and Disassembly" American Chemical Society (ACS) Fall Meeting, Boston MA, USA, (09/2018)
14. **J. Ting**, M. Tirrell, M. "Soft Matter Design Based on Charged Complexation" Sharma's Soft Matter ODES-Lab Mentored Professional Development Seminar Series (09/18)
15. **J. Ting** *(Invited)*, "Molecular Engineering New Polymers for Designer Drug Delivery" DePaul University, Chicago, IL, USA (06/18)
16. **J. Ting**, H. Wu, O. Werba, A. Herzog-Arbeitman, S. Meng, M Tirrell, "Kinetic Pathways in Self-Assembled Polyelectrolyte Complexes" Polymer Physics Gordon Research Conference (06/18)
17. **J. Ting**, H. Wu, O. Werba, A. Herzog-Arbeitman, S. Meng, M Tirrell, "Kinetic Pathways in Self-Assembled Polyelectrolyte Complexes" Polymer Physics Gordon Research Seminar, South Hadley, MA, USA (06/18)
18. **Wu, H.** J. Ting O. Werba, S. Meng, M. Tirrell, "Non-Equilibrium Assembly and Kinetic Pathways in Polyelectrolyte Complexes" National Graduate Research Polymer Conference, South Hadley, MA, USA (06/18)
19. **S. Meng**, H. Wi, J. Ting, M. Tirrell, "Structure Dynamics, Conformation, Rheological Properties and Salt Responsivity of Polyelectrolyte Complexes" National Graduate Research Polymer Conference, Minnesota, MN, USA (06/18)
20. **J. Ting**, H. Wu, A. Herzog-Arbeitman, J. Mitchell, S. Meng, M. Tirrell, "A Materials Genome Approach to Designer Polyelectrolyte Complexes" National Graduate Research Polymer Conference, Minnesota, MN, USA (06/18)
21. **A. Herzog-Arbeitman**, J. Ting, H. Wu, O. Werba, S. Srivastava, M. Tirrell, "Synthesis and structural Investigation of tailored polyelectrolyte complexes" American Chemical Society (ACS), New Orleans, LA, USA (3/13)
22. **J. Ting**, H. Wu, O. Werba, D. Schneiderman, A. Herzog-Arbeitman, J. Mitchell, S. Srivastava, M. Tirrell, M."Solution Assembly and Behavior of Modular RAFT Block

Polyelectrolytes" American Physics Society (APS) March Meeting, Los Angeles, CA, USA (03/05/18)

23. **H. Wu**, J. Ting, O. Werba, S. Srivastava, M. Tirrell, "Polyelectrolyte Complex Micelles: Formation, Dissociation, and Chain Exchange Kinetics" American Physics Society (APS) March Meeting, Los Angeles, CA, USA (03/05/18)
24. **H. Wu**, J. Ting, S. Srivastava, M. Tirrell, "Structural Evolution and Phase Behaviors of Electrostatic Macromolecular Assemblies" American Physics Society (APS) March Meeting, Los Angeles, CA, USA (03/05/18)
25. **M. C. Heiber** *(Poster)*, A. A. Herzing, L. J. Richter, D. M. DeLongchamp "Elucidating Structure-Property Relationships for Charge Carrier Mobility and Mobility Relaxation in Organic Semiconductor Blends using Kinetic Monte Carlo Simulations Informed by TEM Tomography", NIST Sigma Xi Postdoctoral Poster Presentation, Gaithersburg, MD, USA (2/14/2018)
26. **M. C. Heiber**, A. A. Herzing, L. J. Richter, and D. M. DeLongchamp, "Elucidating Structure-Property Relationships for Charge Carrier Mobility in Organic Semiconductor Blends Using Kinetic Monte Carlo Simulations Informed by TEM Tomography", APS March Meeting 2018, Los Angeles, CA, USA (3/7/2018)
27. **M. C. Heiber**, "Elucidating the Complex Processing-Structure-Property Relationships in Bulk Heterojunction OPVs by Measuring the Competition Between Charge Extraction and Recombination under Operating Conditions", MRS Spring Meeting 2018, Phoenix, AZ, USA (4/3/2018)
28. **M. C. Heiber** *(Invited)*, "Integrating Advanced Morphology Measurements, Kinetic Monte Carlo Simulations, and Electrical Device Characterization to Elucidate Processing-Structure-Property Relationships in OPVs", Los Alamos National Laboratory, Los Alamos, NM, USA (7/2/2018)



27. Publications

CHiMaD Supported Publications in 2018

1. Kang, J.; Balla, I.; Liu, X.; Bergeron, H.; Kim, S.; **Wolverton**, CM.; **Hersam**, MC Selective Transfer of Rotationally Commensurate MoS2 from an Epitaxially Grown van der Waals Heterostructure CHEMISTRY OF MATERIALS. Accepted Manuscript (2018) [DOI: 10.1021/acs.chemmater.8b03128] *Partial support*

2. Moody, MJ; Henning, A; Jurca, T; Shang, JY; Bergeron, H; Balla, I; Olding, JN; Weiss, EA; **Hersam**, MC; Lohr, TL; **Marks**, TJ; **Lauhon**, LJ Atomic Layer Deposition of Molybdenum Oxides with Tunable Stoichiometry Enables Controllable Doping of MoS2 CHEMISTRY OF MATERIALS Vol 30 Pg 3628-3632 (2018) [DOI: 10.1021/acs.chemmater.8b01171]. *Primary support*

3. Zhong, CM; Sangwan, VK; Wang, C; Bergeron, H; **Hersam**, MC; Weiss, EA Mechanisms of Ultrafast Charge Separation in a PTB7/Monolayer MoS2 van der Waals Heterojunction JOURNAL OF PHYSICAL CHEMISTRY LETTERS Vol 9 Pg 2484-2491 (2018) [DOI: 10.1021/acs.jpclett.8b00628]. *Partial support*

4. Henning, A; Sangwan, VK; Bergeron, H; Balla, I; Sun, ZY; **Hersam**, MC; **Lauhon**, LJ Charge Separation at Mixed-Dimensional Single and Multilayer MoS2/Silicon Nanowire Heterojunctions ACS APPLIED MATERIALS & INTERFACES Vol 10 Pg 16760-16767 (2018) [DOI: 10.1021/acsami.8b03133]. *Partial support*

5. Sangwan, VK; Lee, HS; Bergeron, H; Balla, I; Beck, ME; Chen, KS; **Hersam**, MC Multi-Terminal Memtransistors from Polycrystalline Monolayer Molybdenum Disulfide NATURE Vol 554 Pg 500-504 (2018) [DOI: 10.1038/nature25747]. *Partial support*

6. Sangwan, VK; Beck, ME; Henning, A; Luo, JJ; Bergeron, H; Kang, JM; Balla, I; Inbar, H; **Lauhon**, LJ; **Hersam**, MC Self-Aligned van der Waals Heterojunction Diodes and Transistors NANO LETTERS Vol 18 Pg 1421-1427 (2018) [DOI: 10.1021/acs.nanolett.7b05177]. *Partial support*

7. LaMountain, T; Bergeron, H; Balla, I; Stanev, TK; **Hersam**, MC; Stern, NP Valley-Selective Optical Stark Effect Probed by Kerr Rotation PHYSICAL REVIEW B Vol 97 Ar 45307 (2018) [DOI: 10.1103/PhysRevB.97.045307]. *Partial support*

8. Yan, WT; Lin, S; Kafka, OL; Yu, C; Liu, ZL; Lian, YP; Wolff, S; **Cao**, J; Wagner, GJ; **Liu**, WK Modeling process-structure-property relationships for additive manufacturing FRONTIERS OF MECHANICAL ENGINEERING Vol 13 Is 4 Pg 482-492 (2018) [DOI: 10.1007/s11465-018-0505-y]*Partial support*

9. Yan, WT; Lin, S; Kafka, OL; Lian, YP; Yu, C; Liu, ZL; Yan, JH; Wolff, S; Wu, H; Ndip-Agbor, E; Mozaffar, M; Ehmann, K; **Cao**, J; Wagner, GJ; **Liu**, WK Data-driven multi-scale multi-physics models to derive process-structure-property relationships for additive manufacturing COMPUTATIONAL MECHANICS Vol 61 Is 5 Pg 521-541 (2018) [DOI: 10.1007/s00466-018-1539-z]*Partial support*

10. Mozaffar, M; Paul, A; Al-Bahrani, R; Wolff, S; **Choudhary**, A; **Agrawal**, A; Ehmann, K; **Cao**, J, Data-driven prediction of the high-dimensional thermal history in directed energy deposition processes via recurrent neural networks, MANUFACTURING LETTERS Vol 18 Pg 35-39 (2018) [DOI: 10.1016/j.mfglet.2018.10.002]*Partial support*

11. Bennett, JL; Kafka, OL; Liao, H; Wolff, SJ; Cheng, P; Hyatt, G; Ehmann, K; **Cao**, J, Cooling rate effect on tensile strength of laser deposited Inconel 718, PROCEDIA MANUFACTURING Vol 26 Pg 912-919 (2018) [DOI: 10.1016/j.promfg.2018.07.118] *Partial support*

12. Bennett, JL; Garcia, D; Kendrick, M; Hartman, T; Hyatt, G; Ehmann, K; You, F; **Cao**, J Repairing automotive dies with directed energy deposition: industrial application and life cycle analysis JOURNAL OF MANUFACTURING SCIENCE AND ENGINEERING (2018) [DOI: 10.1115/1.4042078]*Partial support*

13. Wolff, SJ; Wu, H; Parab N; Zhao C; Ehmann, K; Sun, T; **Cao**, J, In-situ high-speed X-ray imaging of piezo-driven directed energy deposition additive manufacturing SCIENTIFIC REPORTS (accepted)*Partial support*

14. **Stan**, M.; Sarrao, JL, Sarrao, Mesoscale Challenges: Modeling Microstructure Evolution, in "Handbook of Materials Modeling", 2nd Edition, Springer (2018)*Partial support*

15. **Stan**, M. More Resilient Infrastructure through Materials Design, Infrastructure Intelligence, (2018) *Partial support*

16. James, NM; Han, ED; de la Cruz, RAL; Jureller, J; **Jaeger**, HM Interparticle hydrogen bonding can elicit shear jamming in dense suspensions NATURE MATERIALS Vol 17 Is 11 Pg 965- (2018) [DOI: 10.1038/s41563-018-0175-5] *Partial support*

17. Han, ED; Wyart, M; Peters, IR; **Jaeger**, HM Shear fronts in shear-thickening

suspensions PHYSICAL REVIEW FLUIDS Vol 3 Is 7 Ar 73301 (2018) [DOI: 10.1103/PhysRevFluids.3.073301] *Partial support*

18. Reid, DR; Pashine, N; Wozniak, JM; **Jaeger**, HM; Liu, AJ; **Nagel**, SR; **de Pablo**, JJ Auxetic metamaterials from disordered networks PROCEEDINGS OF THE NATIONAL ACADEMY OF SCIENCES OF THE UNITED STATES OF AMERICA Vol 115 Is 7 Pg E1384-E1390 (2018) [DOI: 10.1073/pnas.1717442115] *Partial support*

19. **Jaeger**, HM; Murphy K; Roth L Architectured Particulate Materials in IUTAM Symposium Architectured Materials Mechanics, Chicago, IL, ed. T. Siegmund and F. Barthelat, Purdue University Libraries Scholarly Publishing Services, vol. (2018) [https://docs.lib.purdue.edu/iutam/presentations/abstracts/37] *Partial support*

20. Murphy K; **Jaeger** HM, Designed to Fail: Granular Plasticity and Particle Shape in IUTAM Symposium Architectured Materials Mechanics, Chicago, IL, ed. T. Siegmund and F. Barthelat, Purdue University Libraries Scholarly Publishing Services, vol. (2018) [https://docs.lib.purdue.edu/iutam/presentations/abstracts/53] *Partial support*

21. Jokisaari, AM; **Voorhees**, PW; **Guyer**, JE; **Warren**, JA; **Heinonen**, OG Phase field benchmark problems for dendritic growth and linear elasticity COMPUTATIONAL MATERIALS SCIENCE Vol 149 Is 336 Pg 347 (2018) [DOI:10.1016/j.commatsci.2018.03.015] *Primary support*

22. Kowal, KN; Davis, SH; **Voorhees**, PW Thermocapillary instabilities in a horizontal liquid layer under partial basal slip JOURNAL OF FLUID MECHANICS Vol 855 Pg 839-859 (2018) [DOI: 10.1017/jfm.2018.684] *Primary support*

23. Kowal, KN; Davis, SH; **Voorhees**, PW. Instabilities in rapid directional solidification under weak flow. PHYSICAL REVIEW E 96, no. 6 062802 (2017) [DOI: 10.1103/PhysRevE.96.062802] *Primary support*

24. Ofori-Opoku, N; **Warren**, JA; **Voorhees**, PW Self-consistent modeling of anisotropic interfaces and missing orientations: Derivation from phase-field crystal PHYSICAL REVIEW MATERIALS Vol 2 Is 8 Ar 83404 (2018) [DOI: 10.1103/PhysRevMaterials.2.083404] *Primary support*

25. Poulsen, SO; **Voorhees**, PW Smoothed Boundary Method for Diffusion-Related Partial Differential Equations in Complex Geometries INTERNATIONAL JOURNAL OF COMPUTATIONAL METHODS Vol 15 Is 3 Ar 1850014 (2018) [DOI: 10.1142/S0219876218500147] *Primary support*

26. Senninger, O; Peters, M; **Voorhees**, PW Two-Phase Eutectic Growth in Al-Cu and Al-Cu-Ag METALLURGICAL AND MATERIALS TRANSACTIONS A-PHYSICAL METALLURGY AND MATERIALS SCIENCE Vol 49A Is 5 Pg 1692-1707 (2018) [DOI: 10.1007/s11661-018-4488-4] *Primary support*

27. Zhang, J; Zhang, Y; Ludwig, W; Rowenhorst, D; **Voorhees**, PW; Poulsen, HF; Three-dimensional grain growth in pure iron. Part I. statistics on the grain level , ACTA MATERIALA Vol 156, Is 1 Pg 76-85 (2018) [DOI:10.1016/j.actamat.2018.06.021] *Primary support - not acknowledged*

28. Peters, MC; **Doak**, JW; **Saal**, JE; **Olson**, GB; **Voorhees**, PW; Using First-Principles Calculations in CALPHAD Models to Determine Carrier Concentration of the Binary PbSe Semiconductor JOURNAL OF ELECTRONIC MATERIALS Pg 1-13. (2018) [DOI: 10.1007/s11664-018-6819-z]

29. **Agrawal**, A; **Choudhary**, A; An online tool for predicting fatigue strength of steel alloys based on ensemble data mining INTERNATIONAL JOURNAL OF FATIGUE Vol 113 Pg 389-400 (2018) [DOI: 10.1016/j.ijfatigue.2018.04.017] *Primary support*

30. Yang, Z; Li, X; Brinson, LC; **Choudhary**, A; **Chen**, W; **Agrawal**, A Microstructural Materials Design via Deep Adversarial Learning Methodology JOURNAL OF MECHANICAL DESIGN Vol 140 Is 11 Pg 10 (2018) [DOI: 10.1115/1.4041371] *Primary support*

31. Jha, D.; Ward, L.; Paul ,A.; Liao, W.-K.; **Choudhary**, A.; **Wolverton**, C.; **Agrawal** A. ElemNet: Deep Learning the Chemistry of Materials From Only Elemental Composition, SCIENTIFIC REPORTS, Is 8, Ar 17593 (2018) [DOI: 10.1038/s41598-018-35934-y] *Primary support*

32. Paul, A; Jha, D; Al-Bahrani, R; Liao, W; **Choudhary**, A; **Agrawal**, A CheMixNet: Mixed DNN Architectures for Predicting Chemical Properties using Multiple Molecular Representations PROCEEDINGS OF 2018 NIPS WORKSHOP ON MACHINE LEARNING FOR MOLECULES AND MATERIALS (MLMM 2018) 2018 [arXiv: 1811.08283] *Primary support*

33. Li, X; Yang, Z; Brinson, LC; **Choudhary**, A; **Agrawal**, A; **Chen**, W A Deep Adversarial Learning Methodology for Designing Microstructural Material Systems PROCEEDINGS OF THE ASME 2018 INTERNATIONAL DESIGN ENGINEERING TECHNICAL CONFERENCES (IDETC 2018) (2018) [DOI: 10.1115/DETC2018-85633] *Primary support*

34. Kim, K; Ward, L; He, J; Krishna, A; **Agrawal**, A; **Voorhees**, P; **Wolverton**, C Machine learning accelerated high-throughput materials screening: Discovery of novel quaternary Heusler compounds PHYSICAL REVIEW MATERIALS B Vol 2 Is 12 Pg 123801 (2018) [DOI: 10.1103/PhysRevMaterials.2.123801] *Primary support*

35. Furmanchuk, A; Saal, JE; Doak, JW; Olson, GB; **Choudhary**, A; **Agrawal**, A Prediction of seebeck coefficient for compounds without restriction to fixed stoichiometry: A machine learning approach JOURNAL OF COMPUTATIONAL CHEMISTRY Vol 39 Is 4 Pg 191-201 (2018) [DOI: 10.1002/jcc.25067] *Primary support & CHiMaD DARPA-SIMPLEX*

36. Gopalakrishnan, K; Gholami, H; Vidyadharan, A; **Choudhary**, A; **Agrawal**, A

Crack Damage Detection in Unmanned Aerial Vehicle Images of Civil Infrastructure Using Pre-trained Deep Learning Model INTERNATIONAL JOURNAL FOR TRAFFIC AND TRANSPORT ENGINEERING Vol 8 Pg 1 (2018) [DOI: 10.7708/ijtte.2018.8(1).01] *Partial support*

37. Jha, D; Singh, S; Al-Bahrani, R; Liao, WK; **Choudhary**, A; De Graef, M; **Agrawal**, A Extracting Grain Orientations from EBSD Patterns of Polycrystalline Materials Using Convolutional Neural Networks MICROSCOPY AND MICROANALYSIS Vol 24 Is 5 Pg 497-502 (2018) [DOI: 10.1017/S1431927618015131] *Partial support*

38. Yang, ZJ; Yabansu, YC; Al-Bahrani, R; Liao, WK; **Choudhary**, AN; Kalidindi, SR; **Agrawal**, A Deep learning approaches for mining structure-property linkages in high contrast composites from simulation datasets COMPUTATIONAL MATERIALS SCIENCE Vol 151 Pg 278-287 (2018) [DOI: 10.1016/j.commatsci.2018.05.014] *Partial support*

39. Paul, A; Acar, P; Liu, RQ; Liao, WK; **Choudhary**, A; Sundararaghavan, V; **Agrawal**, A Data Sampling Schemes for Microstructure Design with Vibrational Tuning Constraints AIAA JOURNAL Vol 56 Is 3 Pg 1239-1250 (2018) [DOI: 10.2514/1.J056170] *Partial support*

40. Mozaffar, M; Paul, A; Al-Bahrani, R; Wolff, S; **Choudhary**, A; **Agrawal** A; Ehmann, K; **Cao**, J Data-driven prediction of the high-dimensional thermal history in directed energy deposition processes via recurrent neural networks MANUFACTURING LETTERS Vol 18 Pg 35-39 (2018) [DOI: 10.1016/j.mfglet.2018.10.002] *Partial support*

41. Ward, L; O'Keeffe, SC; Stevick, J; Jelbert, GR; Aykol, M; **Wolverton**, C A machine learning approach for engineering bulk metallic glass alloys ACTA MATERIALIA Vol 159 Pg 102-111 (2018) [DOI: 10.1016/j.actamat.2018.08.002] *Partial support*

42. Wang, DS; Amsler, M; Hegde, VI; **Saal**, JE; Issa, A; Zhou, BC; Zeng, XQ; **Wolverton**, C Crystal structure, energetics, and phase stability of strengthening precipitates in Mg alloys: A first-principles study ACTA MATERIALIA Vol 158 Pg 65-78 (2018) [DOI: 10.1016/j.actamat.2018.07.041] *Partial support*

43. Naghavi, SS; He, JG; Xia, Y; **Wolverton**, C Pd2Se3 Monolayer: A Promising Two-Dimensional Thermoelectric Material with Ultralow Lattice Thermal Conductivity and High Power Factor CHEMISTRY OF MATERIALS Vol 30 Is 16 Pg 5639-5647 (2018) [DOI: 10.1021/acs.chemmater.8b01914] *Partial support*

44. Kim, K; Bobel, A; Brajuskovic, V; Zhou, BC; Walker, M; Olson, GB; **Wolverton**, C Energetics of native defects, solute partitioning, and interfacial energy of Q precipitate in Al-Cu-Mg-Si alloys ACTA MATERIALIA Vol 154 Pg 207-219 (2018) [DOI: 10.1016/j.actamat.2018.05.031] *Partial support*

45. Yao, ZP; Kim, S; Michel, K; Zhang, YS; Aykol, M; **Wolverton**, C Stability and conductivity of cation- and anion-substituted LiBH4-based solid-state electrolytes

PHYSICAL REVIEW MATERIALS Vol 2 Is 6 Ar 65402 (2018) [DOI: 10.1103/PhysRevMaterials.2.065402] *Partial support*

46. Yao, ZP; Kim, S; He, JG; Hegde, VI; **Wolverton**, C Interplay of cation and anion redox in Li4Mn2O5 cathode material and prediction of improved Li-4(Mn,M)(2)O-5 electrodes for Li-ion batteries SCIENCE ADVANCES Vol 4 Is 5 Ar eaao6754 (2018) [DOI: 10.1126/sciadv.aao6754] *Partial support*

47. Ren, F; Ward, L; Williams, T; Laws, KJ; **Wolverton**, C; **Hattrick-Simpers**, J; Mehta, A Accelerated discovery of metallic glasses through iteration of machine learning and high-throughput experiments SCIENCE ADVANCES Vol 4 Is 4 Ar eaaq1566 (2018) [DOI: 10.1126/sciadv.aaq1566] *Partial support*

48. Kim, K; Zhou, BC; **Wolverton**, C First-principles study of crystal structure and stability of T-1 precipitates in Al-Li-Cu alloys ACTA MATERIALIA Vol 145 Pg 337-346 (2018) [DOI: 10.1016/j.actamat.2017.12.013] *Partial support*

49. Tanaka, I; Rajan, K; **Wolverton**, C, Data-centric science for materials innovation MRS BULLETIN Vol 43 Is 9 Pg 659-663 (2018) [DOI: 10.1557/mrs.2018.205] *Partial support*

50. Zhao, H., Wang, Y., Lin, A., Hu, B., Yan, R., McCusker, J., **Chen**, W., McGuinness, D.L., Schadler, L., and Brinson, L.C., NanoMine Schema: An Extensible Data Representation for Polymer Nanocomposites, APL MATERIALS, Vol 6, Is 11, Pg 111108 (2018); https://doi.org/10.1063/1.5046839 *Partial support*

51. **Chen**, W., Schadler, L., Brinson, C., Wang, Y., Zhang, Y., Prasad, A., Li, X., and Iyer, A., Materials Informatics and Data System for Polymer Nanocomposites Analysis and Design, chapter for Big, Deep, and Smart Data in the Physical Sciences (2018) *Partial support*

52. Li, XL; Zhang, YC; Zhao, H; Burkhart, C; Brinson, LC; **Chen**, W A Transfer Learning Approach for Microstructure Reconstruction and Structure-property Predictions SCIENTIFIC REPORTS Vol 8 Ar 13461 (2018) [DOI: 10.1038/s41598-018-31571-7] *Partial support*

53. Zhang, Yi., Tao, S., **Chen**, W., and Apley, D., A Latent Variable Approach to Gaussian Process Modeling with Qualitative and Quantitative Factors, TECHNOMETRICS, in revision, 2018.*Partial support*

54. Hansoge, NK; Huang, TY; Sinko, R; Xia, WJ; **Chen**, W; **Keten**, S Materials by Design for Stiff and Tough Hairy Nanoparticle Assemblies ACS NANO Vol 12 Is 8 Pg 7946-7958 (2018) [DOI: 10.1021/acsnano.8b02454] *Partial support*

55. Wang, YX; Zhang, YC; Zhao, H; Li, XL; Huang, YH; Schadler, LS; **Chen**, W; Brinson, LC Identifying interphase properties in polymer nanocomposites using adaptive optimization COMPOSITES SCIENCE AND TECHNOLOGY Vol 162 Pg 146-155 (2018) [DOI: 10.1016/j.compscitech.2018.04.017] *Partial support*

56. Bostanabad, R; Zhang, YC; Li, XL; Kearney, T; Brinson, LC; Apley, DW; Liu, WK; **Chen,** W Computational microstructure characterization and reconstruction: Review of the state-of-the-art techniques PROGRESS IN MATERIALS SCIENCE Vol 95 Pg 1-41 (2018) [DOI: 10.1016/j.pmatsci.2018.01.005] *Partial support*

57. Huang, Y., Zhao, H., Wang, Y., Ratcliff, T., Breneman, C., Brinson, L.C., **Chen**, W., and Schadler, L.S., Predicting the Breakdown Strength and Lifetime of Nanocomposites using a multi-scale modeling Approach, JOURNAL OF APPLIED PHYSICS, Vol 122, Pg 066101, (2017) *Partial support*

58. Xia, WJ; Vargas-Lara, F; **Keten**, S; **Douglas**, JF Structure and Dynamics of a Graphene Melt ACS NANO Vol 12 Is 6 Pg 5427-5435 (2018) [DOI: 10.1021/acsnano.8b00524] *Primary support*

59. Song, J; Hsu, DD; **Shull**, KR; **Phelan**, FR; **Douglas**, JF; Xia, WJ; **Keten**, S Energy Renormalization Method for the Coarse-Graining of Polymer Viscoelasticity MACROMOLECULES Vol 51 Is 10 Pg 3818-3827 (2018) [DOI: 10.1021/acs.macromol.7b02560] *Primary support*

60. Wei, ZH; Sinko, R; **Keten**, S; **Luijten**, E Effect of Surface Modification on Water Adsorption and Interfacial Mechanics of Cellulose Nanocrystals ACS APPLIED MATERIALS & INTERFACES Vol 10 Is 9 Pg 8349-8358 (2018) [DOI: 10.1021/acsami.7b18803] *Primary support*

61. Xia, WJ; Song, J; Hansoge, NK; **Phelan**, FR; **Keten**, S; **Douglas**, JF Energy Renormalization for Coarse-Graining the Dynamics of a Model Glass-Forming Liquid JOURNAL OF PHYSICAL CHEMISTRY B Vol 122 Is 6 Pg 2040-2045 (2018) [DOI: 10.1021/acs.jpcb.8b00321] *Primary support*

62. Sadman, K.; Wang, Q.; **Shull**, K.; Guanidinium can Break and Form Strongly Associating Ion-Complexes, ACS MACRO LETTERS Vol 8 Pg 117-122 (2019) [DOI:10.1021/acsmacrolett.8b00824] *Primary support*

63. Ren, JX; Zhou, C; Chen, XX; Dolejsi, M; Craig, GSW; Delgadillo, PAR; Segal-Peretz, T; **Nealey**, PF, Engineering the Kinetics of Directed Self-Assembly of Block Copolymers toward Fast and Defect-Free Assembly ACS APPLIED MATERIALS & INTERFACES Vol 10 Is 27 Pg 23414-23423 (2018) [DOI: 10.1021/acsami.8b05247] *Partial support*

64. Zhou C; Kurosawa T; Dazai T; Doise J; Ren JX; Bezik C; Segal-Peretz T; Yamazaki A; Gronheid R; Rincon-Delgadillo P; **de Pablo** J; F. **Nealey** P; Studying the effects of chemistry and geometry on DSA hole-shrink process in three-dimensions, JOURNAL OF MICRO/NANOLITOGRAPHY MEMS, and MOEMS Vol 17 Is 3, 031203 (2018) [DOI: 10.1117/1.JMM.17.3.031203] *Partial support*

65. Dolejsi M., **Nealey** P., Utilization of metal-polymer interactions for self-aligned directed self-assembly of device relevant features, JOURNAL OF MICRO/ NANOLI-

TOGRAPHY MEMS MOEMS Vol 17 Is 3 031204 (2018) [DOI: 10.1117/1.JMM.17.3.031204] *Partial support*

66. Andrich P., Li J., Liu X., Heremans F. J., **Nealey** P. F., Awschalom D. D., Microscale-Resolution Thermal Mapping using a Flexible Platform of Patterned Quantum Sensors, NANO LETTERS, Vol 18 Is 8 Pg 4684-4690 (2018) [DOI: 10.1021/acs.nanolett.8b00895] *Partial support*

67. Li, J; Zhou, C; Chen, X; Rincon-Delgadillo, P; **Nealey**, P; Directed Self-Assembly of Triblock Copolymers for Sub-10 nm Nanofabrication using Polymeric Additives. SPIE PROCEEDINGS Vol 10586 (2018) [DOI: 10.1117/12.2296857] *Partial support*

68. Wu, HX; Li, HH; Solis, FJ; **de la Cruz**, MO; **Luijten**, E Asymmetric electrolytes near structured dielectric interfaces JOURNAL OF CHEMICAL PHYSICS Vol 149 Ar 164701 (2018) [DOI: 10.1063/1.5047550] *Partial support*

69. Prusty, D; Pryamitsyn, V; **de la Cruz**, MO Thermodynamics of Associative Polymer Blends MACROMOLECULES Vol 51 Pg 5918-5932 (2018) [DOI: 10.1021/acs.macromol.8b00661] *Partial support*

70. Wang, G; Eastham, ND; Aldrich, TJ; Ma, BR; Manley, EF; Chen, ZH; Chen, LX; **de la Cruz**, MO; Chang, RPH; Melkonyan, FS; Facchetti, A; **Marks**, TJ Photoactive Blend Morphology Engineering through Systematically Tuning Aggregation in All-Polymer Solar Cells ADVANCED ENERGY MATERIALS Vol 8 Ar 1702173 (2018) [DOI: 10.1002/aenm.201702173] *Partial support*

71. Ma, BR; Nguyen, TD; Pryamitsyn, VA; **de la Cruz**, MO Ionic Correlations in Random Ionomers ACS NANO Vol 12 Pg 2311-2318 (2018) [DOI: 10.1021/acsnano.7b07432] *Primary Support*

72. Kwon, H; Ma, B; **de la Cruz**, MO; Determining the regimes of dielectric mismatch and ionic correlations effects in ionomer blends MACROMOLECULES Vol 52 Is 2 Pg 535-546 [DOI:10.1021/acs.macromol.8b02376] *Partial support*

73. Mathis, L; Chen, YY; **Shull**, KR Tuning the Viscoelasticity of Hydrogen-Bonded Polymeric Materials through Solvent Composition MACROMOLECULES Vol 51 Is 11 Pg 3975-3982 (2018) [DOI: 10.1021/acs.macromol.8b00344] *Partial support*

74. Karabin, NB; Allen, S; Kwon, HK; Bobbala, S; Firlar, E; Shokuhfar, T; **Shull**, KR; Scott, EA Sustained micellar delivery via inducible transitions in nanostructure morphology NATURE COMMUNICATIONS Vol 9 Ar 624 (2018) [DOI: 10.1038/s41467-018-03001-9] *Partial support*

75. Sadman, K.; Wiener, C.G.; Weiss, R.A.; **White**, C.C.; **Shull**, K.R.; Vogt, B.D.; Quantitative Rheometry of Thin Soft Materials Using the Quartz Crystal Microbalance with Dissipation, ANALYTICAL CHEMISTRY Vol 90 Pg 4079-4088 (2018) [DOI: 10.1021/acs.analchem.7b05423] *Partial support*

76. Rahalkar, A; Wei, GM; **Nieuwendaal**, R; **Prabhu**, VM; Srivastava, S; Levi, AE; **de Pablo**, JJ; **Tirrell**, MV Effect of temperature on the structure and dynamics of triblock polyelectrolyte gels JOURNAL OF CHEMICAL PHYSICS Vol 149 Is 16 Ar 163310 (2018) [DOI: 10.1063/1.5035083] *Partial support*

77. Wu, H; Ting, JM; Werba, O; Meng, SQ; **Tirrell**, MV Non-equilibrium phenomena and kinetic pathways in self-assembled polyelectrolyte complexes JOURNAL OF CHEMICAL PHYSICS Vol 149 Is 16 Ar 163330 (2018) [DOI: 10.1063/1.5039621] *Partial support*

78. Ting, JM; Wu, H; Herzog-Arbeitman, A; Srivastava, S; **Tirrell**, MV Synthesis and Assembly of Designer Styrenic Diblock Polyelectrolytes ACS MACRO LETTERS Vol 7 Is 6 Pg 726-733 (2018) [DOI: 10.1021/acsmacrolett.8b00346] *Partial support*

79. Andreev, M; **Prabhu**, VM; **Douglas**, JF; **Tirrell**, M; **de Pablo**, JJ Complex Coacervation in Polyelectrolytes from a Coarse-Grained Model MACROMOLECULES Vol 51 Is 17 Pg 6717-6723 (2018) [DOI: 10.1021/acs.macromol.8b00556] *Partial support*

80. Li, L; Srivastava, S; Andreev, M; Marciel, AB; **de Pablo**, JJ; **Tirrell**, MV Phase Behavior and Salt Partitioning in Polyelectrolyte Complex Coacervates MACROMOLECULES Vol 51 Is 8 Pg 2988-2995 (2018) [DOI: 10.1021/acs.macromol.8b00238] *Partial support*

81. Marciel, AB; Srivastava, S; **Tirrell**, MV Structure and rheology of polyelectrolyte complex coacervates SOFT MATTER Vol 14 Is 13 Pg 2454-2464 (2018) [DOI: 10.1039/c7sm02041d]*Partial support*

82. Schneiderman, DK; Ting, JM; Purchel, AA; Miranda, R; **Tirrell**, MV; Reineke, TM; Rowan, SJ Open-to-Air RAFT Polymerization in Complex Solvents: From Whisky to Fermentation Broth ACS MACRO LETTERS Vol 7 Is 4 Pg 406-411 (2018) [DOI: 10.1021/acsmacrolett.8b00069] *Partial support*

83. Sheridan R; **Gilman** JW; Busel JP; Hartman D; **Holmes** G; Coughlin D; Kelley P; O'Donnell, WR; Nanni, A; Troutman, D; Harris, RW; Gutierrez, J; Bakis, C; Holmes, S; Moser, R; Lackey, E; Fekete, J; Watson, S; Kim, J-H; **Forster**, A; Krishnamurthy, A; **Natarajan**, B. Road mapping workshop report on overcoming barriers to adoption of composites in sustainable infrastructure. NIST SPECIAL PUBLICATION 1218 (2017) [DOI: 10.6028/NIST.SP.1218] *Partial support*

84. Wang P; Peters, M; **Kattner**, U; **Choudhary**, K; Olson, G; Thermodynamic analysis of the topologically close packed σ phase in the Co-Cr system, INTERMETALLICS, Vol 105 Pg 13-20 (2019) [DOI: 10.1016/j.intermet.2018.11.004] *Primary Support*

85. Heiber, MC; Okubo, T; Ko, S-J; Luginbuhl, BR; Ran, NA; Wang, M; Wang, H; Uddin, MA; Woo, HY; Bazan, GC; Nguyen, T-Q; ENERGY & ENVIRONMENTAL SCIENCE, Measuring the Competition between Bimolecular Charge Recombination and Charge Transport in Organic Solar Cells under Operating Conditions, Vol 11 Pg

3019 (2018) [DOI: 10.1039/C8EE01559G] *Partial support*

86. Heiber, MC; Ising_OPV v4.0: Experimental Tomography Data Import, Interpretation, and Analysis, THE JOURNAL OF OPEN SOURCE SOFTWARE Vol 3 Is 31 Pg 1072 (2018) [DOI: 10.21105/joss.01072] *Primary Support*

87. Heiber, MC; Wagenpfahl, A; Deibel, C; Advances in Modeling the Physics of Disordered Organic Electronic Devices *In Handbook of Organic Materials for Electronic and Photonic Devices*, 2nd ed. (Woodhead, 2018) [Ebook ISBN: 9780081022856, Hardcover ISBN: 9780081022849] *Primary Support*

88. Heiber, MC; KMC_Lattice v2.0: An Object-Oriented C++ Library for Custom Kinetic Monte Carlo Simulations THE JOURNAL OF OPEN SOURCE SOFTWARE Vol 4 Is 33 Pg 1168 (2019) [DOI: 10.21105/joss.01168] *Primary Support*

89. Wang, P; Kosmann, J; **Kattner**, UR; Palumbo, M; Hammerschmidt, T; **Olson**, GB; Thermodynamic assessment of the Co-Ta system, CALPHAD Vol 64 Pg 205-212 (2019) [DOI: 10.1016/j.calphad.2018.12.002] *Primary Support*

90. Bocchini, PJ; **Dunand**, DC; Dislocation dynamics simulations of precipitation-strengthened Ni-and Co-based superalloys, MATERIALA Vol 1 Pg 211-220 (2018) [DOI: 10.1016/j.mtla.2018.05.010] *Partial support*

91. Sauza, DJ; **Dunand**, DC; Noebe, RD; **Seidman**, DN; γ'-(L12) Precipitate Evolution during Isothermal Aging of a Co-Al-W-Ni Superalloy, ACTA MATERIALA Vol 164 Pg 654-662 (2019) [DOI: j.actamat.2018.11.014] *Partial support*

92. **Lass**, EA; Sauza, DJ; **Dunand**, DC; **Seidman**, DN, Multicomponent γ'-strengthened Co-based superalloys with increased solvus temperatures and reduced mass densities, ACTA MATERIALA Vol 147 Pg 284-295 (2018) [DOI: 10.1016/j.actamat.2018.01.034] *Partial support*

93. Reyes Tirado, FL; Toinin, JP; **Dunand**, DC, $\gamma + \gamma'$ microstructures in the Co-Ta-V and Co-Nb-V ternary systems, ACTA MATERIALA Vol 151 Pg 137-148 (2018) [DOI: 10.1016/j.actamat.2018.03.057] *Partial support*

94. Yan, WT; Lian, YP; Yu, C; Kafka, OL; Liu, ZL; **Liu**, WK; Wagner, GJ An integrated process-structure-property modeling framework for additive manufacturing COMPUTER METHODS IN APPLIED MECHANICS AND ENGINEERING Vol 339 Is Pg 184-204 (2018) [DOI: 10.1016/j.cma.2018.05.004] *Primary support*

95. Kafka, OL; Yu, C; Shakoor, M; Liu, ZL; Wagner, GJ; **Liu**, WK Data-Driven Mechanistic Modeling of Influence of Microstructure on High-Cycle Fatigue Life of Nickel Titanium JOM Vol 70 Is 7 Pg 1154-1158 (2018) [DOI: 10.1007/s11837-018-2868-2] *Primary support*

96. Lian, YP; Lin, S; Yan, WT; **Liu**, WK; Wagner, GJ A parallelized three-dimensional cellular automaton model for grain growth during additive manufacturing COMPU-

TATIONAL MECHANICS Vol 61 Is 5 Pg 543-558 (2018) [DOI: 10.1007/s00466-017-1535-8] *Partial support*

97. Yan, WT; Qian, Y; Ge, WJ; Lin, S; **Liu**, WK; Lin, F; Wagner, GJ Meso-scale modeling of multiple-layer fabrication process in Selective Electron Beam Melting: Inter-layer/track voids formation MATERIALS & DESIGN Vol 141 Is Pg 210-219 (2018) [DOI: 10.1016/j.matdes.2017.12.031] *Primary support*

98. Liu, ZL; Fleming, M; **Liu**, WK Microstructural material database for self-consistent clustering analysis of elastoplastic strain softening materials COMPUTER METHODS IN APPLIED MECHANICS AND ENGINEERING Vol 330 Is Pg 547-577 (2018) [DOI: 10.1016/j.cma.2017.11.005] *Partial support*

99. Shakoor, M; Kafka, OL; Yu, C; **Liu**, WK Data science for finite strain mechanical science of ductile materials. COMPUTATIONAL MECHANICS Pg 1-13 (2018) [DOI: 10.1007/s00466-018-1655-9] *Primary support*

100. Liu Z, Kafka OL, Yu C, **Liu** WK Data-Driven Self-consistent Clustering Analysis of Heterogeneous Materials with Crystal Plasticity. In: Onate E., Peric D., de Souza Neto E., Chiumenti M. (eds) Advances in Computational Plasticity. Computational Methods in Applied Sciences, vol 46. Springer, Cham (2018) [DOI: 10.1007/978-3-319-60885-3_11] *Partial support*

101. Meredig, B; Antono, E; Church, C; Hutchinson, M; Ling, J; Paradiso, S; **Blaiszik**, B; **Foster**, I; Gibbons, B; **Hattrick-Simpers**, J; Mehta, A; Ward, L; Can Machine Learning Identify the Next YBCO? Examining Extrapolation Performance for Materials Discovery. MOLECULAR SYSTMEMS AND ENGINEERING (2018) [DOI: 10.1039/C8ME00012C] *Partial support*

102. Ward, L; Aykol, M; **Blaiszik**, B; **Foster**, I; Meredig, B; **Saal**, J; Suram, S; Strategies for accelerating the adoption of materials informatics MRS BULLETIN Vol 43 Is 9 Pg 683-689 (2018) [DOI:10.1557/mrs.2018.204] *Partial support*

103. Ward, L; Dunn, A; Faghaninia, A; Zimmermann, N.E.R. ; Bajaj, S; Wang, Q; Montoya, J; Chen, J; Bystrom, K; Dylla, M; Chard, K; Asta, M; Persson, K.A; Snyder, G.J; **Foster**, I; Jain A; Matminer: An Open Source Toolkit for Materials Data Mining COMPUTATIONAL MATERIALS SCIENCE Vol 152 Pg 60-69 (2018) [DOI: 10.1016/j.commatsci.2018.05.018] *Partial support*

104. Ananthakrishnan, R; **Blaiszik**, B; Chard, K; Chard, R; McCollam, B; Pruyne, J; Rosen, S; Tuecke, S; **Foster** I; Globus Platform Services for Data Publication In Proceedings of the Practice and Experience on Advanced Research Computing, p. 14. ACM (2018) [DOI: 10.1145/3219104.3219127] *Partial support*

105. **Blaiszik**, B; Chard, K; Chard, R; **Foster** I; Ward, L; Data Automation at Light Sources SYNCHROTRON RADIATION INSTRUMENTATION June (2018) [DOI: 10.1063/1.5084563] *Partial support*

106. Wang, G; Melkonyan, F.S; Facchetti, A; **Marks**, T.J.; All-Polymer Solar Cells: Recent Progress, Challenges, and Prospects. ANGEWANDTE CHEMIE INT. ED. In press (2018) [DOI: 10.1002/ange.201808976] *Partial support*

107. Swick, SM; Zhu, WG; Matta, M; Aldrich, TJ; Harbuzaru, A; Navarrete, JTL; Ortiz, RP; Kohlstedt, KL; Schatz, GC; Facchetti, A; Melkonyan, FS; **Marks**, TJ Closely packed, low reorganization energy pi-extended postfullerene acceptors for efficient polymer solar cells PNAS Vol 115, E8341-E8348 (2018) [DOI: 10.1073/pnas.1807535115] *Partial support*

108. Wang, SH; Sun, HD; Erdmann, T; Wang, G; Fazzi, D; Lappan, U; Puttisong, Y; Chen, ZH; Berggren, M; Crispin, X; Kiriy, A; Voit, B; **Marks**, TJ; Fabiano, S; Facchetti, A A Chemically Doped Naphthalenediimide-Bithiazole Polymer for n-Type Organic THERMOELECTRICS ADVANCED MATERIALS Vol 30 1801898 (2018) [DOI: 10.1002/adma.201801898] *Partial support*

109. Wang, G; Eastham, ND; Aldrich, TJ; Ma, BR; Manley, EF; Chen, ZH; Chen, LX; **de la Cruz**, MO; Chang, RPH; Melkonyan, FS; Facchetti, A; **Marks**, TJ Photoactive Blend Morphology Engineering through Systematically Tuning Aggregation in All-Polymer Solar Cells Advan. Energy Mater. 8, 1702173 (2018) [DOI: 10.1002/aenm.201702173] *Partial support*

110. Eastham, ND; Logsdon, JL; Manley, EF; Aldrich, TJ; Leonardi, MJ; Wang, G; Powers-Riggs, NE; Young, RM; Chen, LX; Wasielewski, MR; Melkonyan, FS; Chang, RPH; **Marks**, TJ Hole-Transfer Dependence on Blend Morphology and Energy Level Alignment in Polymer: ITIC Photovoltaic Materials ADVANCED MATERIALS Vol 301 704263 (2018) [DOI: 10.1002/adma.201704263] *Partial support*

111. Aldrich, T.J.; Dudnik, A.S.; Eastham, N.D.; Manley, E.F.; Chen, L.X.; Chang, R.P.H.; Melkonyan, F.S.; Facchetti, A.; **Marks**, T.J.; Defect Identification and Suppression in the Environmentally Benign Tin-Free Catalytic Synthesis of Photovoltaic Copolymers MACROMOLECULES 2018, Vol 51 Is 22 Pg 9140-9155 [DOI: 10.1021/acs.macromol.8b02297] *Partial support*

112. Chen, Y.; Yang, L.; Wu, J.; Wang, G.; Huang, W.; Melkonyan, F.; Lu, Z.; Huang, Y.; **Marks**, T.J.; Facchetti, A.; Performance, Morphology, and Charge Recombination Correlations in Ternary Squaraine Solar Cells, Chem. Mater. 30, 6810-6820 (2018). [DOI: 10.1021/acs.chemmater.8b02746] *Partial support*

113. Chen, Y.; Wang, G.; Yang, L.; Wu, J.; Melkonyan, F.; Huang, Y.; Lu, Z.; **Marks**, T.J.; Facchetti, A.; Novel Unsymmetrical Squaraine-Based Small Molecules for Organic Solar Cells, JOURNAL of MATERIALS CHEMISTRY C Vol 6 Is 4 Pg 847-854 (2018) *Partial support*

114. Soe, C.M.M.; Nagabhushana, GP.; Shivaramaiah, R.; Tsai, H.; Nie, T.; Blancon, J.-C.; Melkonyan, F.; Cao, D.H.; Traore, B.; Pedesseau, L.; Kepenekian, M.; Katan, C.; Even, J.; **Marks**, T.J.; Navrotsky, A.; Mohite, A.D.; Stoumpos, C.C.; Kanatzidis,

M.G.; Homologous 2D Halide Perovskites: How Thick Can They Get? PNAS *In Press* (2018) *Partial support*

115. Walwark, DJ; Datko, BD; Wu, QH; Neshchadin, A; Berrens, ML; **Yu**, LP; Grey, JK Conformational Flexibility Determines Electronic Coupling and Charge Transfer Character in Single Propeller-Shaped Perylene Diimide Tetramer Arrays JOURNAL OF PHYSICAL CHEMISTRY C Vol 122 Is 41 Pg 23261-23270 (2018) [DOI: 10.1021/acs.jpcc.8b05709] *Partial support*

116. Carlotti, B; Cai, ZX; Kim, H; Sharapov, V; Madu, IK; Zhao, DL; Chen, W; Zimmerman, PM; **Yu**, LP; Goodson, T Charge Transfer and Aggregation Effects on the Performance of Planar vs Twisted Nonfullerene Acceptor Isomers for Organic Solar Cells CHEMISTRY OF MATERIALS Vol 30 Is 13 Pg 4263-4276 (2018) [DOI: 10.1021/acs.chemmater.8b01047] *Partial support*

117. Sharapov, V; Wu, QH; Neshchadin, A; Zhao, DL; Cai, ZX; Chen, W; **Yu**, LP High Performance Ternary Organic Solar Cells due to Favored Interfacial Connection by a Non-Fullerene Electron Acceptor with Cross-Like Molecular Geometry JOURNAL OF PHYSICAL CHEMISTRY C Vol 122 Is 21 Pg 11305-11311 (2018) [DOI: 10.1021/acs.jpcc.8b03013] *Partial support*

118. Cai, ZX; Zhao, DL; Sharapov, V; Awais, MA; Zhang, N; Chen, W; **Yu**, LP Enhancement in Open-Circuit Voltage in Organic Solar Cells by Using Ladder-Type Nonfullerene Acceptors ACS APPLIED MATERIALS & INTERFACES Vol 10 Is 16 Pg 13528-13533 (2018) [DOI: 10.1021/acsami.8b01308] *Partial support*

119. Wu, QH; Zhao, DL; Goldey, MB; Filatov, AS; Sharapov, V; Colon, YJ; Cai, ZX; Chen, W; **de Pablo**, J; **Galli**, G; **Yu**, LP Intra-molecular Charge Transfer and Electron Delocalization in Non-fullerene Organic Solar Cells ACS APPLIED MATERIALS & INTERFACES Vol 10 Is 12 Pg 10043-10052 (2018) [DOI: 10.1021/acsami.7b18717] *Partial support*

Noted Other NIST Publications Influenced by CHiMaD in 2018

a. A. Rani, K. DiCamillo, S. Krylyuk, R. Debnath, P. Taheri, M. Paranjape, C.E. Korman, M.E. Zaghloul, and A.V. **Davydov**, "Control of polarity in multilayer MoTe2 field-effect transistors by channel thickness", Proc. SPIE 10725, Low-Dimensional Materials and Devices 2018, 1072515 (11 September 2018); [DOI: 10.1117/12.2503888]

b. F. Zhang, S. Krylyuk, H. Zhang, C.A. Milligan, D.Y. Zemlyanov, L.A. Bendersky, A.V. **Davydov**, and J. Appenzeller, Nature Materials (accepted 2018).

c. A.V. **Davydov**, S. **Krylyuk**, H. Zhang, L.A. Bendersky, B. **Burton**, A. Rani, I. Kalish, A. Bruma, B. Kalanyan, H. Hill, A. Hight Walker, M. Zaghloul, S. Oliver, P. Vora, F. Zhang, J. Appenzeller, H. Bergeron, M. Beck, M. Hersam, "Control of conductivity type and semiconductor-to-metal phase transition in MoTe2 single crystals", MRS Fall Meeting, Boston Nov 25, 2018 (ID 3039731)

d. K. Alberi, M.B. Nardelli, A. Zakutayev, L. Mitas, S. Curtarolo, A. Jain, M. Fornari, N. Marzari, I. Takeuchi, M.L. **Green**, M. Kanatzidis, M.F. Toney, S. Butenko, B. Meredig, S. Lany, U. **Kattner**, A. **Davydov**, E.S. Toberer, V. Stevanovic, A. Walsh, N.-G. Park, A. Aspuru-Guzik, D.P. Tabor, J. Nelson, J. Murphy, A. Setlur, J. Gregoire, H. Li, R. Xiao, A. Ludwig, L.W. Martin, A.M. Rappe,S-H. Wei, and J. Perkins, The 2019 materials by design roadmap, J. Phys. D, 52 (2019) 013001

e. H.M. Hill, A.F. Rigosi, S. **Krylyuk**, J. Tian, N.V. Nguyen, A.V. **Davydov**, D.B. Newell, and A.R. Hight Walker, Comprehensive optical characterization of atomically thin NbSe2, Physical Review B, 98 (2018) 165109.

f. D. Ruzmetov, M.R. Neupane, A. Herzing, T.P. O'Regan, A. Mazzoni, M.L. Chin, R.A. Burke, F.J. Crowne, A.G. Birdwell1, D.E. Taylor, A. Kolmakov, K. Zhang, J.A. Robinson, A.V. **Davydov** and T.G. Ivanov, Van der Waals interfaces in epitaxial vertical metal/2D/3D semiconductor heterojunctions of monolayer MoS2 and GaN, 2D Materials, 5 (2018) 045016

g. O.B. Aslan, I.M. Datye, M.J. Mleczko, K.S. Cheung, S. **Krylyuk**, A. Bruma, I. Kalish, A.V. **Davydov**, E. Pop, and T.F. Heinz, Probing the Optical Properties and Strain-Tuning of Ultrathin Mo1-xWxTe2, Nano Lett., 18 (2018) 2485

h. K.H. Smithe, A.V. Krayev, C.S. Bailey, H.R. Lee, E. Yalon, O.B. Aslan, M.M. Rojo, S. **Krylyuk**, P. Taheri, A.V. **Davydov**, T.F. Heinz, and E. Pop, Nanoscale Heterogeneities in Monolayer MoSe2 Revealed by Correlated Scanning Probe Microscopy and Tip-Enhanced Raman Spectroscopy, ACS Appl. Nano Mater. 1 (2018) 572

i. A.Y. Koposov, B.T. Spann, A. Bruma, P. Schuele, K.Bertness, and A. **Davydov**, High-Temperature Photoluminescence in Colloidal "Quasi" 2D Materials, J. Phys. Chem C 122 (2018) 5119.

j. **Stoudt**, M.R., **Lass**, E.A., Ng, D.S. et al. Metall and Mat Trans A (2018) 49: 3028. https://doi.org/10.1007/s11661-018-4643-y

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