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Keynote Speaker

Fernando Escobedo, Department of Chemical and Biological Engineering, Cornell University

Some lessons on the Self-assembly Engineering of Soft Materials and the Role of Entropy

The ever-expanding ability to synthesize diverse micro- and nano-particles, and polymers and oligomers with highly controlled sequences over a wide range of chemistries, has fueled the interest for their use as building blocks to assemble novel soft materials with potentially unique properties. Molecular simulations studies over the last two decades have revealed many correlations between the features of such building blocks (like shape and chemical selectivity) and the structure of the resulting self-assembled morphologies, which constitute the basis for the rules of designing novel and complex morphologies. In this presentation, I will highlight a few such lessons which our group has contributed to unveil, some of them involving the formation of nanoparticle alloys, others the formation of network phases with polyphillic molecules; some relating to thermodynamic driving forces/packing free-energy and others related to the enhancement of kinetic pathways toward crystallization through mesophase intermediates. The crucial but often overlooked role of entropy will be described through specific vignettes.

Biography

Professor Escobedo received a B.S. degree in Chemical Engineering from the University of San Agustin in Peru (1987) and worked for 5 years as an R&D engineer in a Peruvian company before coming to the U.S. for graduate studies. He received a Chemical Engineering M.S. degree from the University of Nebraska-Lincoln (1993) and the Ph.D. from the University of Wisconsin-Madison (1997). He joined the faculty of Cornell University at the end of 1998. He received the Camille & Henry Dreyfus Foundation new faculty award, 1999, the CAREER Award from the National Science Foundation, 2000, the Alfred P. Sloan Foundation fellowship, 2004, and the AIChE CoMSEF Impact Award, 2012. He also won an award for teaching excellence from the College of Engineering (2003). He currently holds the Marjorie Hart Chair of Engineering.

Invited Speakers

Chris Bartel, Department of Chemical Engineering and Materials Science, University of Minnesota, Twin Cities

Thermodynamic predictions of solid-state phase transformations during synthesis

High-throughput density functional theory (DFT) calculations have accelerated the rate at which solid-state materials can be screened for various properties of technological interest. To understand whether these hypothetical materials can be made in the lab, the current standard is to calculate their thermodynamic stability with respect to potential competing phases. While stability is an important metric, it does not determine if or how that material can be synthesized. Whereas stability (at some set of conditions) is an intrinsic property of a material, synthesizability is not a similarly straightforward binary – it depends on experimental choices such as precursors, temperature, synthesis approach, etc. This talk will discuss our recent efforts to leverage DFT-calculated thermodynamics towards the prediction of synthesis pathways in the context of traditional ceramic synthesis as well as thin film deposition.

Biography

Chris Bartel is an Assistant Professor in the Department of Chemical Engineering and Materials Science (CEMS) at the University of Minnesota. Prior to joining CEMS in 2022, he earned a PhD in Chemical Engineering from the University of Colorado and worked as a postdoctoral researcher in Materials Science at UC Berkeley. Chris now leads the "Design of Materials on Computers Lab," which leverages first-principles calculations, thermodynamic modeling, solid-state chemistry, and machine learning to accelerate the design of solid-state materials for energy-related applications. He has been recognized as an Emerging Investigator by the journal Nanoscale and honored as a Scialog Fellow in Negative Emissions Science and Sustainable Minerals, Metals, and Materials. Chris grew up near New Orleans and earned a BS in Chemical Engineering from Auburn University.

Whitney Loo, Department of Chemical and Biological Engineering, University of Wisconsin - Madison

Investigating the Thermodynamics of Single Ion Conducting Polymer Blend Electrolytes Using X-ray and Neutron Scattering

The ongoing development of rechargeable batteries with increased energy density is critical to continue the growth in the electrification of mobile technologies. To achieve these goals, new electrolytes must be developed that possess fast ion transport characterized by high ionic conductivity and high cation transference number (the fraction of ions that carry current), a combination of properties typically not found in a single material. Therefore, we propose to engineer polymer blend electrolytes as suitable electrolyte replacements. In this presentation, we will study blends of ion-containing polymer and ion-conducting polymers and determine how blend composition and polymer molecular weight affects thermodynamics. Through a combined approach leveraging X-ray and neutron scattering, we are able to resolve the nanostructure across a wide range of length-scales ranging from monomer sub-units to inter-chain interactions. By quantitatively fitting the scattering profiles, we can extract thermodynamics parameters and relevant length-scales and show how local charge correlations affect nanoscopic structure. We show that our results qualitatively agree with previously developed theory for similar systems and conclude by suggesting how molecular engineering of these blends can enable precise tuning of the strength of charge correlations and the concentration of charged species to further probe blend thermodynamics.

Biography

Whitney Loo joined the Department of Chemical and Biological Engineering as an Assistant Professor in January 2023. Whitney obtained her B.S. in Chemical Engineering from MIT. She obtained her Ph.D. in Chemical Engineering from UC Berkeley in 2020 where she worked with Nitash Balsara studying the molecular level physics of block copolymer electrolytes for Lithium metal batteries. Most recently, she is a Postdoctoral Scholar working jointly at the University of Chicago with Paul Nealey and the Molecular Foundry at Lawrence Berkeley National Lab with Ricardo Ruiz. Her postdoctoral research involves the design of novel polymers and nanofabrication techniques for block copolymer nanolithography. Her independent research group at UW-Madison focuses on designing polymers for a more sustainable future including applications in polymer electrolytes for battery applications, mechanical recycling of mixed waste plastics and advanced microelectronics manufacturing.

Janani Sampath, Department of Chemical Engineering, University of Florida

Investigating contributions from vehicular and Grotthuss transport on hydroxide ion conductivity in anion exchange membranes (AEMs)

Anion exchange membranes (AEMs) used in alkaline fuel cells rely on the selective transport of hydroxide ions through the membrane to produce electricity. The low conductivity of hydroxide ions in AEMs has prevented their widespread use and improving AEM conductivity is of significant fundamental and industrial relevance. The target for conductivity in AEMs set by the Department of Energy is typically $\geq$ 70 mS/cm at operating conditions. An understanding of the hydroxide transport mechanisms through the polymer can help in the design of membranes with improved performance. In prior work, we leveraged atomistic molecular dynamics (MD) simulations to capture correlations in hydroxide transport across four polyethylene AEMs with different functional groups. While the trends in conductivity were consistent with experiments, there was a significant difference in the magnitude between experiments and simulations. We ascribed this to the inability of our model to capture the reactive Grotthuss transport (proton hopping) mechanism which is dominant in these systems. In our current work, to account for Grotthuss transport, we use the heuristic reaction protocol REACTER that allows us to model reactions within the framework of classical simulations. We find that the simulated hydroxide conductivity is in better agreement with experimental values after the inclusion of Grotthuss transport. We extend our analysis across different AEM chemistries, including polyethylene, polysulfone, and polynorbornene, and find that the balance between vehicular and Grotthuss transport strongly depends on the polymer's microstructure and water uptake.

Biography

Janani Sampath joined the Chemical Engineering department at the University of Florida as an Assistant Professor in 2021. Prior to this, she was a postdoctoral researcher at the University of Washington and the Pacific Northwest National Laboratory, where her research involved elucidating driving forces behind biomolecular self-assembly using atomistic molecular dynamics simulations and enhanced sampling methods. She received her Ph.D. in Chemical Engineering from the Ohio State University in 2018, where she studied mechanical and interfacial structure -property relationships in ion containing polymers using coarse-grained models. She was a Research Engineer in Unilever R&D between 2010–2013. She obtained a B.E. in Chemical Engineering from R.V. College of Engineering, India, in 2010. She is a recent recipient of the Ralph E. Powe Junior Faculty Award and the ACS PRF Doctoral New Investigator Award.

LeeAnn Sager-Smith, Department of Chemistry, Saint Mary's College

Computational Exploration of Fermion-Exciton Condensates

Phenomena such as superconductivity and superfluidity arise due to a Bose-Einstein-like condensation of pairs of fermions into a single quantum state with large non-classical off-diagonal long-range order. Fermion pair condensates (FPCs)—the most-familiar of which involve the condensation of Cooper (electron) pairs—results in the superfluidity of their constituent electrons, which cause the material through which they flow to be both a perfect conductor and a perfect diamagnet. However, all currently-known superconductors condense at either too-low of temperatures or too-high of pressures to be viable. Similarly, exciton condensates (ECs) involve the condensation of particle-hole pairs (excitons) into a single quantum state to create a superfluid. This superfluidity of these relatively-light quasiparticles with comparatively-high binding energies are expected to condense at higher temperatures and involve the non-dissipative transfer of energy; however, exciton condensates often have too-short a lifetime to be easily observed in experiment. In this talk, I will introduce fermion-exciton condensates (FECs)—novel quantum states that simultaneously exhibit the character of superconducting states and exciton condensates and may demonstrate hybrid properties of both. The computational and theoretical prediction of these FECs as well as a model Hamiltonian that describes them will be introduced along with a state preparation for an FEC on a quantum device.

Biography

LeeAnn M. Sager-Smith is an Assistant Professor of Physical Chemistry at Saint Mary's College in Notre Dame, IN. After receiving her B.S. in Chemistry from Indiana University, she received a Ph.D. in Physical Chemistry from the University of Chicago where she worked with Dr. David Mazziotti. Her research focuses on introducing undergraduate researchers to explore the physics behind highly correlated molecular processes through a combination of traditional ab initio methodologies, quantum computation, and machine learning.

Alexander Pak, Department of Chemical and Biological Engineering, Quantitative Biosciences and Engineering Program, Materials Science Program, Colorado School of Mines

Advancing coarse-graining methods using statistical mechanical principles and probabilistic machine learning

Multiscale modeling and simulation techniques offer powerful tools for uncovering microscopic insights into dynamical processes that span wide spatial and temporal scales, which are often inaccessible to experiments alone. Bottom-up coarse-graining, where high-resolution atomistic data is used to derive low-resolution models based on statistical mechanical principles, is one such technique that has gained prominence across chemistry, biology, materials science, and other disciplines. Despite extensive algorithmic advances in coarse-grained (CG) modeling, conventional approaches relying on pseudo-particle representations and pairwise interactions face limitations in capturing complex, many-body, and non-Markovian behavior, particularly in implicit-solvent and low-resolution regimes. In this talk, I will present our recent efforts to address this gap by integrating strategies from the data science and machine learning communities. I will introduce a framework called Probabilistic Forecasting for Coarse-Graining (PFCG), which implicitly learns the underlying CG equations of motion, rather than conventional CG approaches that infer effective CG interactions (via the force-field paradigm). I will demonstrate the capabilities of our approach on several fast-folding mini-proteins with diverse folding kinetics, showing that PFCG accurately recovers both equilibrium free energy landscapes and time-dependent dynamical properties, a known challenge for existing CG models. Finally, I will show that one important benefit of our approach is to quantifiably characterize equilibrium versus non-equilibrium dynamics via path entropy generation rates. I will conclude with a discussion on the path forward for more accurate and transferable CG models, with ongoing work focused on scaling to larger (bio)macromolecular systems and improving both chemical and thermodynamic generalizability.

Biography

Alex is currently an Assistant Professor of Chemical and Biological Engineering at Colorado School of Mines, where he has been since January of 2021. He received his B.S. in Chemical Engineering from M.I.T and his Ph.D. in Chemical Engineering from UT Austin. His graduate research focused on fundamental charge storage mechanisms using carbon-based nanomaterials for supercapacitor applications. As a postdoc, Alex received the F32 NIH Postdoctoral National Research Service Award, which supported his transition into computational biophysics as part of the Chemistry Department at the University of Chicago. His group focuses on the development of multiscale simulation techniques and their application toward both fundamental understanding and engineered control of self-assembled complexes, including for macromolecules, polymers, and porous crystals.

Contributed Talks

Computational design and optimization of Ca-Ce-Ti-Mn oxide perovskites for thermochemical hydrogen generation, Manish Kumar, Washington University in St. Louis

Thermochemical hydrogen generation (TCH) via water splitting is a potentially carbon-free method for hydrogen generation, utilizing high-temperature redox cycles powered by concentrated solar or nuclear heat. The performance and cost of a TCH system depend on the properties of the redox-active metal oxide involved. With off-stoichiometric redox activity, a quinary oxide perovskite containing Ca and Ce on the A-site and Ti and Mn on the B-site ($Ca_{0.67}Ce_{0.33}Ti_{0.33}Mn_{0.66}O_3$) shows the potential to surpass the performance of the state-of-the-art material, CeO_2, which requires higher operating temperatures. However, $Ca_{0.67}Ce_{0.33}T_{0.33}Mn_{0.66}O_3$ is not yet the fully optimized composition of Ca-Ce-Ti-Mn oxide perovskites (CCTM).

In this talk, I will present our computational approach for optimizing the composition of CCTM using Hubbard U-corrected density functional theory (DFT). We compare the redox activity of various CCTM compositions, showing that the redox activity depends more strongly on the local environment of oxygen vacancies than on the global composition. Unlike $Ca_{0.67}Ce_{0.33}Ti_{0.33}Mn_{0.66}O_3$, Ce^{4+} is absent in most compositions considered, leading to greater involvement of Mn cations in the redox process. Our findings show that the reduction of Ce^{3+} is more delocalized than that of Mn^{4+} and Mn^{3+}. Inspired by Wexler et al., we developed a linear crystal feature model using readily accessible bulk properties like crystal bond dissociation energy and crystal reduction potential to predict the neutral oxygen vacancy formation energy. We trained the CFM on DFT calculations of oxide perovskites, significantly reducing the computational cost of screening the CCTM compositional space. Our CFM achieves a mean absolute error of approximately 0.23 eV by utilizing available reference thermochemical data, enabling us to determine the optimal composition of CCTM for TCH.

This material is based upon work supported by the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, Hydrogen and Fuel Cell Technologies Office program under Award Number DE-EE0010733.

Machine-Learning Interatomic Potentials for Predictive Simulations of Laser-Driven Silicon Nanoparticle Synthesis, Thomas Sundberg, University of Michigan

Silicon nanoparticles (Si-NPs) are critical to next-generation electronics and energy technologies due to their exceptional optoelectronic and thermal properties. However, industrial-scale production of tailored Si-NPs remains limited by trade-offs between precision and throughput in conventional synthesis approaches. Emerging shockwave-driven methods, including detonation and laser-driven compression of silicon precursors, present a promising route for rapid, large-scale synthesis, as demonstrated in recent experimental work. While analogous techniques have enabled tunable nanocarbon production, achieving precise control of Si-NP formation under shock conditions requires a thorough understanding of non-equilibrium atomic-scale dynamics during phase transformations. Molecular simulations are one method to understand these mechanisms but require interatomic potentials that exhibit quantum-mechanical (QM) accuracy with larger spatiotemporal scales. To address this challenge, we present a machine-learned interatomic potential (ML-IAP) for silicon, developed within the ChIMES framework, which achieves near-QM accuracy while enabling large-scale simulations of shock-induced phenomena. This work outlines our methodology for training set generation and validating model performance to both first principles and experimental results. Finally, we discuss the extension of the ChIMES hierarchical framework to Si-C systems, highlighting its potential to enable predictive simulations of nanomaterial synthesis across multi-component precursors.

Developing a consistent model for simulating mechanical and chemical degrees of freedom of stepping kinesin, Jutta Luettmer-Strathmann, University of Akron

Molecular motors transform chemical energy into mechanical work to perform important functions in many biological processes. One of the best studied motor proteins is kinesin-1, which steps hand-over hand along microtubules to transport cargo vesicles along the axon of nerve cells, using energy released by the hydrolysis of one ATP molecule in each step. Recent experimental work has provided much insight into the stepping process, which can be interpreted with abstract models of the underlying mechano-chemical cycle. In addition, atomistic simulations have revealed the protein's conformational changes during the stepping stages in detail. However, a consistent picture of kinesin stepping and force generation is still emerging. In particular, experimental data alone does not seem to be sufficient to distinguish between competing mechano-chemical models. One way to address this issue is through a mesoscopic model that is able to simulate many steps of the walker, including missteps, and that treats physical and chemical degrees of freedom in a thermodynamically consistent way. We have been developing a mesoscopic model for Brownian dynamics simulations of kinesin walking on a microtubule and pulling a cargo. Using a simple implementation of a chemical cycle we are able to capture some important aspects of molecular walkers. However, it is difficult to describe the full range of kinesin processes, including failed steps and detachment, in this way. In this work, we broaden the scope of the model and consider more carefully the role of the ATP molecule and its products in the description of the chemical and mechanical states of the system. Our goal is a thermodynamically consistent implementation of mechano-chemical states and transitions so that we may apply our model to more complex problems involving, for example, interactions with obstacles, other kinesins, or multiple microtubules.

DynaMate: Leveraging AI-Agents for customized research workflows, Orlando A. Mendible Barreto, University of Notre Dame

Developments related to large language models (LLMs) have deeply impacted everyday activities and are even more significant in scientific applications. They range from simple chatbots that respond to a prompt to very complex agents that plan, conduct, and analyze experiments. As more models and algorithms continue to be developed at a rapid pace, the complexity involved in building this framework increases. Additionally, editing these algorithms for personalized applications has become increasingly challenging. To this end, we present a modular code template that allows easy implementation of custom Python code functions to enable a multi-agent framework capable of using these functions to perform complex tasks. We used the template to build DynaMate, a complex framework for generating, running, and analyzing molecular simulations. We performed various tests that included the simulation of solvents and metal-organic frameworks, calculation of radial distribution functions, and determination of free energy landscapes. The modularity of these templates allows for easy editing and the addition of custom tools, which enables rapid access to the many tools that can be involved in scientific workflows.

Generalized Molecular Property Imputation Using a Flexible Transformer Architecture, Andrew B. Schofield, University of Notre Dame

In chemistry, there are numerous molecular data repositories in which much of the property data is missing. Classically, the tools to impute this missing data have operated on too simple a paradigm, only utilizing the molecular structure. In reality, there is typically additional known property information that is neglected or even missing structure information that makes this problem's traditional posing unwise or inapplicable. Here, we present a more general paradigm of property imputation that is capable of ingesting any known information about the molecule to make a prediction. A robust transformer model architecture was developed that can take a variable set of property or structure information as inputs to predict a different variable set of properties as outputs. This flexible architecture can predict a single property, the same set of multiple properties, or different sets of different properties. We examine these capabilities in multiple trials using a dataset of approximately 16M organic molecules and 23 properties. Finally, we proffer an imputation protocol

with the same architecture for use in determining missing values in a sparse dataset using only the dataset itself and demonstrate the suitability of this protocol for general imputation by two case studies with fluorine containing hydrocarbons and nonaromatic hydrocarbons.

Interpretable Visualizations of Data Spaces for Classification Problems,
Christian Jorgensen, University of Wisconsin-Madison

It is often scientifically insightful to identify correlations between the chemical structure or orientation of a molecule and its corresponding properties. However, this task is often complicated by the high dimensionality of common machine learning representations of materials and the challenges of interpreting black-box machine learning models. For regression tasks, hybrid learning approaches, such as principal covariates regression (PCovR), are incredibly useful for extracting structure-property relationships from data, as they combine unsupervised dimensionality reduction with supervised learning to identify a shared latent space that captures both chemical diversity and its relationship to a target property. Although methods such as linear discriminant analysis (LDA) offer similar capabilities for classification problems, these methods can struggle to capture intracluster variance and can overfit to training data. In this work, we extend the PCovR framework to create principal covariates classification (PCovC), a new dimensionality reduction method for identifying structure-property relationships for classification tasks, and kernel PCovC (KPCovC), a kernelized counterpart for learning nonlinear relationships. We then highlight the effectiveness of these methods in capturing chemical relationships with case studies in crystallography and molecular toxicity prediction. The flexibility of the PCovC framework suggests that it can be broadly impactful towards enabling "white box" prediction with problems with categorical labels.

Automated Prediction of the Reaction Networks Governing Electrolyte Degradation,
Esra Ulgey, Purdue University

The demand for improved Li-ion batteries (LIBs) has led to a growing interest in highly concentrated electrolytes (HCEs) as a solution to conventional LIB limitations. HCEs facilitate the formation of stable anion-derived solid electrolyte interfaces (SEI) through decreased solvent reduction reactions. However, understanding electrolyte decomposition is challenging due to the lack of an inclusive set of reaction channels available to simulate the initial steps of SEI formation. In this work, we are addressing this drawback by adapting and applying the Yet Another Reaction Program (YARP) methodology, developed by the Savoie group, focusing on solvents and dissolved anions from various salts suitable for HCEs. Reaction network prediction has the potential to reveal SEI formation mechanisms by analyzing transition states and thermodynamics, while also enhancing computational methods that integrate reaction kinetics into molecular dynamics simulations. As an initial case study, the anion-mediated reaction channels in typical Li-carbonate based electrolytes are characterized to understand electrolyte degradation mechanism. The elementary reduction pathways involving various anions and ethylene carbonate solvent have been identified, along with the corresponding activation barriers. This study is further extended to explore the reduction of HCE compatible solvents and salts, leading to their decomposition through multiple pathways and the formation of various products including experimentally identified SEI contributing compounds. The diverse decomposition mechanisms of different solvents highlighted the role of the salt in mediating electrolyte decomposition as well as the complexity of SEI formation and SEI's dependence on electrolyte components, influencing both its composition and structure. These findings offer valuable insights into SEI formation in HCEs by providing sufficient kinetic data on anion-mediated reaction channels for further investigations and contribute to the development of next-generation battery technologies.

Effect of Nonclassical Electrostatic Interaction on Colloidal Gel Structure in Ionic Liquids, Hwigwang Lim, University of Wisconsin-Madison

Ionic liquids are promising candidates for electrolytes in energy storage devices due to their ionic conductivities, as well as their high thermal and electrochemical stability and non-flammability.

Still, their leakage and dendrite formation problem need to be addressed to prevent failure of the system. Colloidal gel matrices formed with inorganic nanoparticles are a promising way to alleviate these problems by enhancing mechanical and transport properties without significantly inhibiting ionic conduction. In highly concentrated electrolytes, such as ionic liquids, it has been shown that the double-layer screening of these nanoparticles deviates from the conventional theory. However, the effect of the nonclassical behavior on collective colloidal structure is still poorly understood. Here we develop a simulation framework to effectively investigate the effect of different levels of theory on the colloidal structural and phase behavior. We take the approach of DLVO (Derjaguin, Landau, Verwey, and Overbeek) theory, which superposes van der Waals and electrostatic interactions between nanoparticles. We then modify the electrostatic term according to different theories of the electric double layer, and perform molecular dynamics simulation. We characterize the resulting structures using Topological Data Analysis (TDA). Our approach yields a cohesive simulation framework to model and to analyze the colloidal phase behavior in ionic liquids such that we can ultimately design more stable energy storage technologies.

Measuring and manipulating liquid-liquid phase separation in vapor-deposited glasses, Camille Bishop, Wayne State University

Liquid-liquid phase separation causes hierarchical organization in systems ranging from intracellular compartments to active layers of organic photovoltaic cells. In organic electronic applications, films are often confined to length scales of tens or hundreds of nanometers. Since interfaces alter thermodynamically-favored structures and kinetic mobility, deliberate confinement can be exploited to access morphologies that do not occur in the bulk, which could lead to new materials with exceptional properties. Here, we use physical vapor deposition of organic molecular blends to prepare thin, phase-separated films. AFM and polarized resonant soft X-ray scattering (PRSoXS) are used to measure the effect of deposition conditions and film thickness on the composition separation, interfacial molecular orientation, and thermal and kinetic stability of the films. Using a computational scattering simulation analysis framework, we are sensitive to gradients in composition and molecular orientation with nm-level resolution. We find that the hierarchical molecular organization strongly depends on film thickness, suggesting long-ranging gradients in structure and dynamics that cause highly cooperative organization.

Computational Design of Functionalized Cyclodextrin Probes for Selective and Sensitive PFAS Detection in Water, Siddarth Achar, University of Chicago

The widespread presence of persistent and toxic per- and polyfluoroalkyl substances (PFAS) in drinking water necessitates the development of sensitive and selective detection methods. Existing analytical techniques often suffer from high costs and complexity, while alternative sensing approaches like fluorescent and electrochemical sensors may lack the required performance. Field-effect transistor (FET) sensors offer a promising platform for rapid and portable PFAS detection, contingent on the availability of highly selective molecular probes. We have developed a Bayesian Optimization-driven computational high-throughput screening strategy to design functionalized β-cyclodextrin (CD) variants with significantly enhanced affinity and selectivity for perfluorooctanesulfonic acid (PFOS), a prevalent PFAS. Our approach involved defining a chemical design space of substituted CDs and employing biased molecular dynamics (parallel-biased metadynamics) simulations to efficiently estimate their binding free energies and selectivity against sodium dodecyl sulfate (SDS), a common interferent. Our computational screening efforts identified promising candidates with sub-nanomolar dissociation constants for PFOS (Kd = 10^{-11} M) and a selectivity (Kd(SDS)/ Kd(PFOS) = 5 x 10^4) approximately 5000 times greater than that of unmodified β-CD. We found that introducing alkyl and fluorocarbon functional groups on the primary hydroxyl side of the β-CD ring yielded the most favorable results, likely due to enhanced hydrophobic interactions with PFOS. These findings provide a compelling direction for the development of highly sensitive and selective molecular probes for real-time PFAS detection using FET sensors, offering a potentially cost-effective and field-deployable solution. Furthermore, the interpretable design

principles derived from this study can inform the development of probes for other environmentally relevant contaminants with similar chemical characteristics.

Large-scale simulation of bottlebrush block copolymers in melts using coarse-grained, implicit side-chain model, Haisu Kang, University of Illinois at Urbana-Champaign

Bottlebrush polymers have a wide materials design space, but any physical understanding of how molecular architecture affects macroscopic properties must account for many individual side-chains. We recently demonstrated that it is possible to describe a bottlebrush as a linear semi-flexible chain based on the wormlike cylinder (WLCy) model, which does not explicitly include side chains. These implicit side-chain (ISC) models drastically decrease the computational cost by directly mapping to simulations of the conformation of individual bottlebrushes. However, our original model was designed to consider solution bottlebrush assembly, and was therefore not applicable to the self-assembly of bottlebrush in melts.

In this study, we expand the capability of the ISC model to melt bottlebrush copolymer assembly. Using single-chain in mean-field (SCMF) simulations, molecular observables such as the end-end distance and radius of gyration are obtained to parameterize an ISC model with wormlike cylinder model parameters; effective Kuhn length $\lambda^{(-1)}$, cylinder length L and width D. Each architecture could be mapped to a unique set of wormlike cylinder model parameters, so that they can be represented by an ISC model consisting of N_{ISC} beads of size D with a bending parameter κ_θ related with effective Kuhn length $\lambda^{(-1)}$. The effective pairwise interaction potential for this ISC model was determined by using an iterative Boltzmann inversion (IBI) procedure to match with the structural features in the ESC model. In order to incorporate the incompatibility of different species for bottlebrush copolymer, we introduced the Flory-Huggins parameter χ into the pairwise interaction potential by considering additional SCMF simulations on bottlebrush binary blends. We found that the χ_N effect can be parameterized for different architectures and scaled from the homopolymer interaction potential. The predicted interaction potential for χ_N in ordered self-assembly structure led to predictions consistent with the self-assembled structure of bottlebrush block copolymers in experiments.

A Generalized Framework for Developing Alchemical Interaction Models for Self-assembling Polymer-grafted Nanoparticles, Melody Zhang, University of Michigan

Molecular simulation is a powerful tool for optimizing complex colloidal nanoparticle systems, but it has traditionally required a tradeoff between accuracy and computational efficiency. For example, all-atom simulations offer detailed descriptions of interparticle interaction but can be computationally expensive. Coarse-graining (CG) methods address this limitation by grouping multiple atoms into a single interaction site, preserving essential functional group information while significantly reducing cost. While CG method enables efficient study of complex colloidal nanoparticles such as polymer-grafted nanoparticles (PGNs), the high tunability of PGNs poses a challenge: identifying optimal configurations for a desired self-assembly behavior or structure requires evaluating an immense number of candidates. Inverse design methods tackles this challenge by optimizing tunable PGN design attributes, yet its success depends on an accurate description of PGN interactions with respect to inter-PGN distance and the tunable design parameter. Machine-learned interaction models (ML-IAMs) can learn these intricate and higher-dimensional interactions, making them promising tools for describing interaction landscape for inverse design methods. In this work, we introduce our framework that provides detailed descriptions of the PGN interactions by integrating a physics-informed "alchemical" dimension into our ML-IAM, thereby establishing a connection between energy profiles and tunable PGN attributes such as polymer length and grafting density. We first incorporate steered molecular dynamics (SMD) with the forward-reverse (FR) method into Zhou et al.'s grid sampling scheme to enable efficient and robust sampling of the potential of mean force (PMF) training data. Leveraging CG models for our testbed system, we employ FR-SMD to sample PMFs for CG PGNs with varying polymer lengths, which serves

as the alchemical dimension. We use ChIMES, a physics-informed ML-IAM, to learn the PMFs of the CG PGNs and demonstrate their accuracy in reproducing equilibrium thermodynamical and structural properties. We then extend ChIMES' functional form to incorporate the alchemical dimension, which we name the extended ChIMES (X-ChIMES) model. X-ChIMES generates a unified PMF landscape across all PGN configurations. We then apply the Digital Alchemy inverse design framework using the newly developed X-ChIMES model to optimize the design parameters for target crystal structures. Our analysis reveals the change in alchemical parameters as the target crystal structure varies, which sheds light on the self-assembly behavior of these PGNs. Our framework lays the foundation for advancing physics-informed CG ML-IAMs in nanomaterial design and discovery.

Coarse-grained simulations reveal ligand end group effects on nanoparticle adsorption to lipid bilayers., Carlos Huang-Zhu, University of Wisconsin-Madison

Ligand-coated nanoparticles have been shown to be promising biomedical agents. However, the exploration of the large design space associated with tuning their physicochemical properties to target specific interactions with biomolecules is limited by experimental challenges such as synthesis and characterization, as well as long timescales required by activity assays. These challenges thus limit the understanding of driving forces that promote nanoparticle adsorption, particularly how ligand end groups modulate nano–bio interactions. To address some of this challenges, molecular dynamics have been employed to simulate molecular interactions.

In this work, we employ coarse-grained simulations to simulate the adsorption of ligand-coated gold nanoparticles to single-component lipid bilayers. The modeled ligands possess a modular backbone consisting of an alkanethiol and a polyethylene glycol chain with a cationic quaternary ammonium as end group. We developed coarse-grained models of nanoparticles with different alkyl substituents on the cationic end group to probe how lipophilicity and chain architecture modulate nanoparticle adsorption. We described nanoparticle adsorption using a reaction coordinate that is a function of two collective variables, and used the string method with swarms-of-trajectories and umbrella sampling in combination with path variables to compute the potential of mean force along the minimum free energy path. We demonstrate that nanoparticle adsorption is favorable for all nanoparticles, and show that ligand lipophilicity and architecture modulate the adsorption process in nonobvious ways. Ligands of intermediate lipophilicities and ligands with branched architectures can protrude out of the ligand monolayer and intercalate into the lipid bilayer to minimize membrane disruption and lower free energy barriers. Furthermore, branched end groups lead to lower adsorption free energies by having ligands cross the bulk of the bilayer and attach to the opposite leaflet. These results show that coarse-grained simulations can be leveraged in accelerating the exploration of the large design space of nanoparticle engineering.

Effects of Chirality and Solvent Strength on Model Polycatenane Network Shapes, Ritav Das, The Ohio State University

Polycatenanes, or mechanically interlocked ring molecules, can be synthesized and are found naturally in kinetoplasts, networks of linked DNA rings formed by certain unicellular protists called kinetoplastids. The shape and size of these networks are sensitive to temperature and solvent conditions and how they are topologically linked together. We study these effects using molecular dynamics (MD) simulations with a simplified bead-spring model for rings in an implicit solvent, considering three topological types of networks under various solvent conditions. Specifically, we simulate networks of 25 (5x5), 49 (7x7), and 196 (14x14) rings linked in a square lattice with three different topological patterns, each based on two linkage types: over-under (OU) and under-over (UO). In the alternating chirality (Alt) system, a ring has the same linkage type (e.g., all OU) with its four neighbors. In the non-alternating chirality (B) system, the linkage types alternate (OU, UO, OU, UO) around the ring. Finally, in the semi-alternating chirality (C) system, a ring has two of the same type linkages in a row (OU, OU, UO, UO). The shapes of these systems in good solvent

are sensitive to their chirality, with the Alt, B, and C systems appearing bowl-like, saddle-shaped, and flat, respectively. The coil-to-globule transition occurred at approximately the same solvent parameter for all three networks; however, their average radii of gyration (Rg^2) decreased in the order C > B > Alt. These shapes were further characterized by analyzing the eigenvalues of each network's gyration tensors to find the asphericity, acylindricity, and relative shape anisotropy.

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Role of Surface Electrostatics in Particle-Polyelectrolyte Complex Coacervates, Sirikarn Phuangthong, University of Illinois Urbana-Champaign

Particle-polyelectrolyte (PE) complex coacervation is a liquid-liquid phase separation that occurs when polyelectrolytes undergo associative phase separation with an oppositely charged non-polymeric species, such as surfactant micelles, colloids, and proteins. While this phenomenon has been extensively studied due to its benefits in biological and industrial applications, the molecular understanding of particle-PE coacervation remains limited to low-linear charge density systems. Previous studies showed that correlation attractions between two charged micelles in the presence of polyelectrolytes are necessary for phase separation to occur. We further investigate these correlation attractions by exploring different particle-PE systems that can lead to complex coacervation.

In this work, we study how micelle surface charge density, polyelectrolyte sequence, and salt concentration affect correlation attractions necessary for particle-PE coacervation. By employing thermodynamic integration and the potential of mean force for Brownian Dynamic simulations, two-dimensional energy landscapes of interacting surfaces were obtained. Initial result shows that these inter-particle electrostatic interactions are short-range and directly related to the local charge environments, and by modifying properties such as the polymer charge sequence and surface charge density, a variety of local charge environments can be observed. The results of this project is then integrated into our MC-SCFT model to provide a better molecular understanding of particle-PE coacervation thermodynamics.

Atomistic Features Affecting Conductivity in Organic Mixed Ionic - Electronic Conductors, Begum Yuksel, Purdue University

Organic Mixed Ionic-Electronic Conductors (OMIECs) are materials that are composed of π-conjugated polymers and an electrolyte solution. They have interconnected electronic and ionic transport due to reversible doping by ions within the electrolyte solution and delocalized electrons within the π-conjugated polymers. Their unique structure allows them to be used in electronic devices. The conductivity of OMIECs can be improved by changing polymer morphology, which can be accomplished through doping. The morphology of π-conjugated polymers is also affected by ion-electron coupling in OMIECs. So, an optimal design that considers both ionic and electronic pathways is necessary to improve the conductivity of OMIECs. From a design perspective, changing the components of the electrolyte and potentially improving the OMIECs is highly attractive due to the relative simplicity of swapping out these components. However, even small changes in the electrolyte, such as varying the size of the anion, can have a cascade of effects on OMIEC morphology, doping efficiency, and charge transport which are still poorly understood. This work adapts and extends OMIEC coarse-grained molecular dynamics (CG-MD) simulation methods recently developed by the Savoie group to elucidate electrolyte-function relationships, and kinetic Monte Carlo (kMC) simulations to examine the anion size and side-chain design effect on the conductivity of OMIECs through calculating polaron mobility. The results demonstrate the impact of anion size on polaron mobility and polymer morphology across three different scenarios: polymers with highly polar side chains at full frequency, intermediate polar side chains at full frequency, and intermediate polar side chains at half frequency. We observe that charge transport is primarily governed by a lower side-chain frequency, which significantly reduces trap site formation.

In contrast, the effects of anion size and side-chain hydrophilicity are comparatively minor. These findings provide valuable insights for developing general design rules for the OMIECs with wider class of polymers and dopants.

A Morse Potential-Based Equation of State for Dense Fluids: A Statistical Fluid Theory Approach, Ehsan Heidaryan, Cornell University

An accurate equation of state (EOS) for dense fluids has been developed using the Morse potential. The quality of its theoretical predictions—covering coexistence densities, vapor pressures, and second-derivative thermophysical properties—is critically assessed against molecular simulations and experimental data for representative real substances. The approach rigorously applies the Barker-Henderson high-temperature perturbation expansion up to the third order in free energy for Morse fluids, yielding a significantly improved representation of dense fluid behavior compared to existing statistical fluid theories. Notably, the new EOS outperforms the SAFT-VR framework, which employs the Mie potential in both accuracy and predictive power. Moreover, by selecting a special potential range (α=4.5), it effectively maps onto the Lennard-Jones (LJ) potential, achieving a level of accuracy comparable to the empirical EOS for LJ fluids (Stephan, Staubach and Hasse, 2020). A defining strength of this new EOS is its ability to maintain high precision across a diverse range of substances, from water to molten gold. This, coupled with its superior modeling of second-derivative properties, enables a much-improved global representation of thermodynamic properties and fluid-phase equilibria within a non-polarizable framework.

Systematically Building and Optimizing a Generalized Hydrofluorocarbon Refrigerant Force Field, Montana Carlozo, The University of Notre Dame

Molecular simulation can be used to predict the thermophysical properties of hydrofluorocarbon refrigerants necessary for the design of the separation processes over a wide range of temperatures using an accurate generalized force field (FF) model. A generalized FF allows the parameters developed for a chemical environment (atom type) in one molecule to be reused in other molecules. However, the process for tuning and building a generalized FF is not standardized. In this work, we used eigen-decomposition and estimability analysis to guide atom type selection and nonlinear regression (NLR) to optimize the FF parameters. Gaussian process surrogate models were used to reduce the computational effort of NLR. Our work suggests that six optimized atom types recommended via data science techniques significantly improves on the predictions by GAFF. Moreover, our work demonstrates that adding more atom types ad-hoc (as is typically done) can potentially lead to overfit FF models.

The Capability of Machine-Learning Potentials to Reproduce Complex Interactions in Polymeric Systems Using Extrapolation, Natalie Hooven, University of Wisconsin-Madison

Machine-learning potentials have emerged as powerful tools for enabling accurate molecular simulation at greater time and length scales, previously inaccessible by putatively accurate force-fields or computationally expensive ab-initio methods. However, the accuracy of a machine-learning potential depends not only on the machine-learning infrastructure used for construction, but also on the accuracy of the training data available. An issue then arises: for complex systems, like polymers, this accurate training data is in itself prohibitive to obtain due to the spatiotemporal scales that remain inaccessible by accurate ab-initio simulation. Therefore, we must assess the ability of machine-learning potentials trained on smaller analogs to extrapolate to larger chemical species. By focusing solely on the ability of machine-learning potentials to work in extrapolative regimes for polyalkane systems, we can answer the fundamental question: what is the smallest polyalkane dataset that can be used to construct a machine-learning potential that is able to accurately reproduce complex macromolecular interactions in polymers? In seeking to answer this question, we gain fundamental insights into the datasets and machine-learning architectures that allow for robust extrapolations which can be used to design transferable potentials.

Development of ChIMES machine learned interatomic potentials for all silica MFI zeolites, Vallabh Vasudevan, University of Michigan

Ammonia, a cornerstone of global fertilizer production, is synthesized via the energy-intensive Haber-Bosch (HB) process, where a significant proportion of energy consumption arises from ammonia separation via cryogenic condensation. Current efforts to develop a more energy efficient industrial scale ammonia production process focus on developing improved catalysts and separation membranes to drive the reaction equilibrium. Zeolite membranes offer a promising alternative for energy-efficient separation due to their nanoporous frameworks, tunable pore sizes, and high thermal/chemical stability. These naturally occurring aluminosilicate materials are nanoporous, exhibiting a near continuum of pore sizes that allow for separation of target gas molecules. However, optimizing these materials requires atomic-level insights into adsorption, diffusion, and framework dynamics—challenges that remain experimentally intractable. Here, we will present a new machine-learned interatomic potential for bulk MFI zeolite based on the ChIMES physics-informed machine learned interatomic model and development framework. Our training strategy incorporates strained and compressed structural configurations to capture framework flexibility, enabling robust predictions of bulk properties (e.g., radial distribution functions, vibrational power spectra) that align closely with DFT benchmarks. This work will focus on thermal expansion behavior, a critical factor in membrane stability under industrial conditions, by leveraging large-scale molecular dynamics simulations to probe temperature-dependent structural changes in MFI zeolites. These simulations aim to predict thermal and dynamic properties critical to the intelligent design of membranes for industrial applications. We will further demonstrate the transferability of our framework to topologically similar zeolites, such as MEL. By bridging atomic-scale simulations with industrially relevant properties, this approach aims to accelerate the design of zeolite-based membranes for energy-efficient ammonia production and catalytic systems.

Anisotropic Coarse-Graining with Machine Learning Representations, Arthur Lin, University of Wisconsin-Madison

Physics-based, atom-centered machine learning (ML) representations have transformed atomistic simulations. By incorporating physical priors (e.g., symmetry invariance and many-body interactions), these representations have been successfully used as a basis for sophisticated structure-property analyses and computationally inexpensive machine-learned potentials (MLPs). However, many of these representations cannot be used for coarse-graining, as they assume particles are spherical with isotropic interactions. To include the additional prior of nonspherical particle geometry, we extended one such representation, the Smooth Overlap of Atomic Positions (SOAP), to further incorporate known anisotropy into the representation. This representation, deemed AniSOAP can then be coarse-grain molecules as geometrically accurate beads. AniSOAP is useful for both supervised and unsupervised tasks, such as learning the Gay-Berne potential, learning the energetics of unstable benzene crystals, and delineating liquid crystal phases. We further show that AniSOAP an be used to in determining optimal coarse-grained mappings, which coupled with an anisotropic potential, can lead to more accurate and physically-founded CG simulations.