4th Molecular Simulations Workshop
Northeastern Regional Edition

Contents

1 Talks

1.1 Prof. Jerelle A. Joseph
Accurate Models for Understanding and Engineering Biomolecular Condensates

Princeton University

The interior of cells contains numerous components that need to be carefully organized in space to fulfill a wide range of biological functions. The most widespread form of intracellular compartments completely lack membranes. In the place of membranes, these compartments—so called biomolecular condensates—are sustained and segregated in space via phase separation, analogous to how oil drops separate when dispersed in water. Complementary to experiments, molecular modeling and simulations have surfaced as powerful tools for providing us with the missing close-up views to elucidate the physicochemical principles that shape biomolecular condensates. In this talk, I will discuss current efforts at developing quantitatively accurate residue-resolution models for studying biomolecular condensates, particularly within the paradigm of phase separation.

1.2 Prof. Nav Nidhi Rajput
Molecular Informatics for Establishing Structure-Property Relationships in Multi-component Liquid Solutions

Stony Brook University

Developing the capability to accurately predict the macroscopic properties of complex multicomponent solutions from microscopic features of molecular species is a grand challenge spanning chemistry, materials science, and engineering. Despite significant efforts undertaken in the past, the existing studies fall short of predicting the properties of multicomponent solutions due to multiple length and time scales of functional properties and a vast chemical and parameter space, which cannot be efficiently explored using trial and error based experimental approaches, brute-force computational approaches, and/or high-throughput computational approach that typically focus on just a single scale.

Driven by these needs, we developed open-source high-throughput multiscale computational infrastructures coined MISPR (Materials Informatics for Structure-Property Relationships; `https://molmd.github.io/mispr/html/index.html`).1, 2 MISPR seamlessly integrates density functional theory (DFT) calculations with classical molecular dynamics (CMD) simulations and machine learning (ML), allows 100-1000s of calculations to be performed in parallel by minimizing manual interference, and generates high fidelity databases of computational properties. MISPR includes fully automated workflows to compute electronic, thermodynamic, structural, and dynamical properties of liquid solutions. In this talk, I will demonstrate the unique features of MISPR by highlighting its different automated workflows and their application in designing optimal electrolytes for next-generation energy storage devices.1 I will then show (1) a novel "DFT-CMD-DFT" approach to accurately predict various stable species present in a multicomponent solution by analysing experimental nuclear magnetic resonance (NMR) spectra3 and (2) how MISPR can help screening thousands of potential solvents and salts to identify potential electrolyte systems.

References
1. Atwi, R.; Bliss, M.; Makeev, M.; Rajput, N. N., MISPR: an open-source package for high-throughput multiscale molecular simulations. Scientific Reports 2022, 12 (1), 15760.
2. Rasha Atwi, M. B., Nav Nidhi Rajput, https://github.com/molmd/mdproptools. 2022.
3. Atwi, R.; Chen, Y.; Han, K. S.; Mueller, K. T.; Murugesan, V.; Rajput, N. N., An automated framework for high-throughput predictions of NMR chemical shifts within liquid solutions. Nature Computational Science 2022

1.3 Prof. Peter Ravikovitch
Molecular Modeling of Materials for Adsorptive Separations

ExxonMobil

Adsorption is widely used in industry for separating molecules based on their size and interactions with porous adsorbent materials. Porous crystalline adsorbents such as zeolites, and novel metal-organic frameworks are particularly amenable to first-principles modeling and molecular simulations. The talk will present examples of how molecular modeling and simulations in combination with experiments are used in industry to develop novel materials and separation processes

1.4 Prof. Robert Riggleman
A Molecular View of Energy Dissipation and Failure in Model Polymer Networks

University of Pennsylvania

The fracture of end-linked polymer networks and gels strongly affects the performance of these versatile and widely used materials, and a molecular-level understanding of the fracture process is important to the design of new materials. The Lake-Thomas theory serves as a framework to understand and quantify the energy dissipation due to the chain scission in these materials based on an idealized picture of fracture in networks, though recent extensions of the Lake-Thomas theory have incorporated the effect of topological defects and in some examples enabled accurate prediction of the fracture. In this talk, I will describe how we use coarse-grained molecular dynamics simulations and network analysis techniques to provide a molecular view of the energy dissipated and fracture process during failure of polymer networks. In addition to the energy of the broken strand, we also consider the amount of energy released by the networks connected to the broken chain and other sources of energy dissipation due to the fracture process. Furthermore, by analyzing the topology of the cross-linked network, we are able to predict the strands most prone to failure. Our results can be used to design polymer networks and optimize the crosslinking process to tune the properties of novel materials.

2 Posters

2.1 Abdelraheem Abdallah, Nanoscale Energy Adsorption Systems (NEAS): Pressure-Driven Intrusion and Extrusion of Water in Hydrophobic Nanopores

Rutgers University
Advisor: Alexander Neimark

Suspensions of lyophobic nanoporous particles are promising materials for energy adsorption and storage. When subjected to mechanical compression, the non-wetting solvent intrudes into nanopores converting and storing the impact energy that can be further released upon solvent extrusion when the compression force is removed[1]. While various nanoscale energy adsorption systems (NEAS) have been explored experimentally, the molecular level mechanisms of the intrusion-extrusion cycles are not well-understood. Using atomistic molecular dynamics simulations (MD), we investigate the dynamics of shock impact induced intrusion and extrusion of in hydrophobic cylindrical channels using atomistic Molecular Dynamics (MD) simulations. MD simulations performed for nanopores of different sizes at quasi-static and dynamic conditions demonstrate three different stages of the compression process: (1) initial elastic compression of the solvent, (2) fluid intrusion into the pore upon reaching a threshold intrusion pressure and (3) elastic compression of the solvent upon pore-filling (see Figure 1). Extrusion of the solvent from a completely filled nanopore upon the release of pressure is initiated by the bubble nucleation. Our simulation results are correlated to the experimental observations.
1. Belogorlov, Anton A., et al. "Suspensions of lyophobic nanoporous particles as smart materials for energy absorption." Journal of Colloid and Interface Science 600 (2021): 229-242.

2.2 Avery Agles, Polysaccharide Gel Water as a Class of 'Biological Water'

Princeton University
Advisor: Ian Bourg

It is now well known that the dynamical properties of water and the resulting hydrogen bond network are disrupted by the presence of solid or large molecular interfaces, including biological interfaces like cell membranes and protein surfaces. While there has been much effort to understand this 'biological water' in the context of protein systems, little attention has been paid to polysaccharides' effect on water properties. Polysaccharides are one of the primary components of the cytosol and the extracellular matrix of both eukaryotic and prokaryotic systems. In addition, polysaccharides have found abundant uses in biomedical applications such as wound healing, drug delivery, and 3D cell culturing. In these contexts, polysaccharide gels are the medium in which and through which biomolecular processes occur. Which is to say that, in these contexts, polysaccharide gel water is a vital actor.

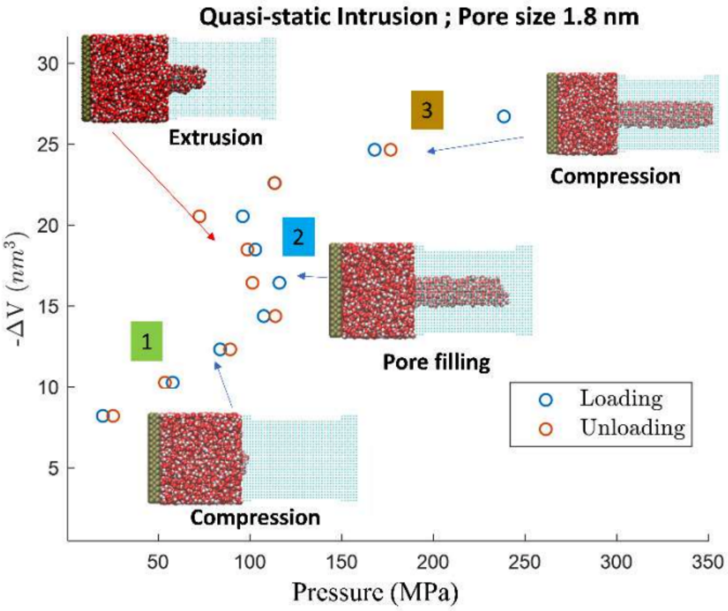

Figure 1: Intrusion-extrusion cycle in cylindrical channel: volume change as a function of fluid pressure

In this study, we focus our attention on the molecular dynamics of water within gels of the polysaccharide alginate and how those dynamics diverge from those of bulk water. The results build from and complement our previous investigation into the structure and energetics of alginate gels.1 We start with measurements of the water diffusivity as a function of water content and ion environment with the goal of determining the applicability of the core-shell model to alginate gels. We follow with measurements of the static dielectric constant of water and hydraulic permeability to understand how the collective reorientation and translation dynamics of water are impeded by the alginate network. We argue the results of this investigation provide necessary insight into the nature of biological water in all polysaccharide gels. To this end, I'll focus the discussion of these results on their implications for the various biomolecular processes occurring in the polysaccharide gels of both biological and engineered systems. References:

1. A. A. Agles and I. C. Bourg, The Journal of Physical Chemistry B, 2023, 127, 1828–1841.

2.3 Matthew Bliss, Electrolyte Effect on CO2 Electroreduction

Stony Brook University
Advisor: Nav Nidhi Rajput

The electrochemical reduction of carbon dioxide (CO2) has great potential to alleviate important environmental problems: the elevated presence of CO2 in the atmosphere and the need for fossil fuel-based production of fuels and commodity chemicals. However, the application of electrochemistry to CO2 reduction is hindered by low activities and Faradaic efficiencies. In aqueous media, the competing hydrogen evolution reaction (HER) remains an unresolved problem for selectivity. The use of nonaqueous solutions presents a possible path to mitigate this problem. In this study, we investigate the use of nonaqueous solvents by using molecular simulations to examine the effect of the solvent's chemical class on properties relevant to the CO2 reduction reaction: CO2 solubility, electrochemical stability, and viscosity.

2.4 Diana Marlén Castañeda Bagatella, Studying Carbocation Rearrangements Using Metadynamics Simulations

New Jersey Institute of Technology
Advisor: Pier Alexandre Champagne

The carbocations and their rearrangements have significant importance in synthetic organic chemistry, the biosynthesis of terpenes, and petroleum chemistry. The formation of carbocations, often from neutral electrophiles undergoing ionization, cannot be studied accurately without explicit solvation models, and as such, the activation free energies of such pathways have not been studied. To develop an approach to this problem, we first optimized our substrates of interest using density functional theory. After that, the computational approach to evaluate the reaction mechanism is developing ab initio molecular dynamics simulations (AIMD) in an explicit solvation model. With this kind of calculation, it's possible to study the energy barriers of the transformation of unimolecular reactions considering the energy from the free energy surface and not from the electronic surface and obtain accurate energies due to the known accuracy of DFT methods in organic chemistry. The results of these simulations will be combined with the investigation of the potential energy surface topography and variational transition state theory calculations to evaluate the carbocation rearrangements in many species, some of which will also be studied experimentally.

2.5 Andres Cifuentes-Lopez, An energetic Upper-Limit to the Fidleity of Ligand Identification by Bacterial Chemoreceptors

Rutgers University, Newark
Advisor: Colin Kinz-Thompson

Chemoreceptors are important transmembrane proteins that direct the swimming responses of certain bacterial species to chemical cues in the environment, such as amino acids. In particular, the Tar and Tsr chemoreceptors are well-known for facilitating responses to L-aspartate and L-serine, respectively. Tar and Tsr do that by directly binding their 'cognate' amino acids (i.e., ligands) using their ligand binding domains (LBDs). However, it is unclear how accurately Tar and Tsr are able to discriminate between the binding of their cognate versus other non-cognate ligands, and how effectively this discrimination can be transduced to their downstream signaling pathways. To answer these questions, we will calculate the difference in binding free energy of cognate versus non-cognate ligands as an energetic upper-limit on the information transduction performed by these chemoreceptors. We propose a computational workflow composed of three general stages: (1) homology modeling of LBD tertiary structures for various chemoreceptors, (2) cognate ligand docking calculations to these structures, and (3) alchemical free energy perturbation calculations using cognate and non-cognate ligands. By performing these calculations on the Escherichia coli and Salmonella entericia Tar and Tsr receptors for both L-aspartate and L-serine, the relative comparisons of our calculations provide insight into the universal molecular and energetic determinants involved in accurate chemoreception.

2.6 Nicholas Corrente, Deformation of Nanoporous Carbons Induced by Binary Adsorption: Insight from SAFT-Density Functional Theory

Rutgers University
Advisor: Alexander Neimark

Understanding the phenomenon of adsorption-induced deformation in flexible nanoporous carbons has numerous practical applications, such as mixture separations; electrode development; and hydrocarbon storage, carbon capture, and enhanced natural gas recovery. While most practical uses of nanoporous carbons involve the adsorption of mixture, theoretical and experimental works have largely been devoted to single-component adsorption.

In this work, we consider the deformation of nanoporous carbons in the context of carbon dioxide displacement of natural gas, focusing specifically on the deformation induced by adsorption of carbon dioxide and methane mixtures. We employ classical density functional theory (DFT) calculations augmented by the perturbed chain statistical associating fluid theory (SAFT)[1] equation of state to model adsorption of fluid mixtures on carbon slit pores at geologically relevant temperatures and pressures. We then utilize a thermodynamic model relating the adsorption stress to the grand thermodynamic potential of the adsorbed phase to quantify the deformation of the pore[2]. We begin with the simplest case of pure methane, and then incrementally increase the fraction of adsorbed CO2, mimicking CO2-assisted natural gas recovery techniques. We thereby determine the effect of geological conditions, pore size, and fluid composition on the adsorption-induced deformation of the nanoporous carbons.

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2. Ravikovitch, P. I.; Neimark, A. V., Density functional theory model of adsorption deformation. Langmuir 2006, 22 (26), 10864-10868.

2.7 Joy Datta, Investigating Next Generation Electrode Material for Multivalent Ion Battery

New Jersey Institute of Technology
Advisor: Dibakar Datta

As Li is a rare earth material and will become scarce in near future it will be better to shift our focus to other potential ion-based batteries. Recently, Ca has gained lots of attention for its low-cost, high energy density, and abundance. But Ca ion is bigger compared to Li-ion which needs lots of space in electrode materials to accommodate and diffuse Ca ion by maintaining cycle stability. Molybdenum vanadium oxide (MoVO) can host Ca ions because of its Micron sized shaped structure with Nanopores inside. In this work, Diffusion mechanisms of Ca ion in Trigonal MoVO structure have been studied by first-principles density functional theory. Our triangular MoVO structure consists of triangular, hexagonal, and heptagonal tunnels that help Ca ions to diffuse smoothly. The activation barrier and diffusion path for moving Ca ions from one site to another have been calculated by Nudge Elastic Band (NEB) method. This work also reported the most favorable location for Ca ions to accommodate in MoVO structure by calculating adsorption potential. Our calculations revealed that void size is an important factor for Ca ions to travel inside the MoVO structure.

2.8 Dhwanit Dave, Sequence-Tunable Phase Behavior in Designed Peptides Rationalized Using Molecular Dynamics

City University of New York
Advisor:Rein Ulijn

Research in biological condensed phases has exploded over the past decade. A conceptual framework of understanding is emerging, derived from biological, biomimetic, and synthetic sequences. However, de novo peptide condensate design remains a challenge due to incomplete understanding of the structural and interaction complexity. We designed peptide modules based on a simple repeat motif composed of tripeptide spacers (GSG, SGS, GLG) interspersed with adhesive amino-acids (R/H and Y). We show, using sequence editing and a combination of computation and experiment, that $n \rightarrow \pi^*$ interactions in GLG backbones are a dominant factor in providing sufficient backbone structure, which in turn regulates the water interface, collectively promoting liquid droplet formation. Moreover, these H(GLG)Y and R(GLG)Y condensates unexpectedly display sequence-dependent emission that is a consequence of their non-covalent network interactions, and readily observable by confocal microscopy. These condensates can be taken up by cells and can be visualized, thus holding promise for biomedical applications.

2.9 Lilia Escobedo, Identifying and Characterizing Hydrophilic Protein Surface Patches

University of Pennsylvania
Advisor: Amish Patel and Daeyeon Lee

Biofouling, the undesired formation of biofilms, can contaminate a wide variety of surfaces that operate in aqueous environments, such as medical device implants and water filtration membranes. One way to combat this issue is to increase a surface's hydration to prevent proteins from adsorbing onto the surface, which is the first step in biofilm formation. While homogeneous surface modifications, such as polar and/or zwitterionic surface coatings employ electrostatics to enhance surface hydration, more heterogeneous surface modifications derived from proteins themselves could also provide enhanced surface hydration, since proteins surfaces have evolved to resist non-specific aggregation and fouling by other proteins in the crowded cellular environment. By identifying and characterizing the most hydrophilic regions, we aim to uncover the chemical patterns responsible for protein-protein selectivity and provide a basis for the creation of non-fouling surfaces. Using indirect umbrella sampling (INDUS) molecular dynamics simulations, we have characterized the atomic-level hydrophilicity of a protein surface directly based on water affinity. Using this characterization and clustering techniques, we have identified protein surface patches of similar water affinity for various proteins, ranging from highly hydrophobic to highly hydrophilic. Our results indicate that patches of similar water affinity contain both polar and nonpolar atoms as well as amino acids of varying hydrophilicity. We have also found that proteins surface patches of similar partial charge do not have similar water affinity, indicating the importance of other factors, such as chemical patterning and surface topology on protein surface hydrophilicity.

2.10 Santiago Flores Roman, Probing Efficacy of Gassmann Theory on Nanopores by Adsorption of Gallium on Silica

New Jersey Institute of Technology
Advisor: Gennady Gor

Thermodynamic properties of fluids in the nanoscale tend to differ from their analogues in bulk, which raises uncertainty about whether Gassmann theory can predict the elastic properties of nanoconfined fluids. For example, to predict the bulk modulus of a composite, one needs to estimate the bulk modulus of the non-porous material, which may bring biased estimations of the target modulus. However, all the information the Gassmann theory needs can be obtained directly from molecular simulations. To predict the bulk modulus of the confined fluid, we reproduced gallium adsorption on silica in a grand-canonical ensemble in LAMMPS using an embedding atom model (EAM) for the fluid and an All-Atom model for the porous solid. Whereas for the elastic constants of the solid (porous and non-porous), we measured the average changes of the stress tensor in a canonical ensemble.

2.11 Rubayet Hassan, Entropy-Driven Nanomechanics of Flexible Membranes

New Jersey Institute of Technology
Advisor: Fatemeh Ahmadpoor

Biological membranes fluctuate noticeability at physiological temperature. These thermal fluctuations play a key role in many biological processes such as exo and endocytosis, membrane fusion, pore formation, cell adhesion, binding-unbinding transitions, among many others and studying the entropic factors in biological phenomena have now become the cornerstone of cell-mechanics research. These aforementioned biophysical phenomena are governed by a complex interplay between the various attractive and repulsive forces that mediate between biological structures. A key role is played by a repulsive force termed entropic pressure, the origins of which lie in the thermally excited fluctuations of membranes. The entropic force, in sharp contrast to the only other known repulsive hydration force, is decidedly long range and competes with the van der Waals force at all distances. The entropic force is predicated on a variety of physical arguments and mathematical approximations, started by Helfrich who proposed that this force scales as $1/d^3$. This power law has been later on validated using advanced theoretical and computational techniques. In the majority of the previous works— both theoretical and computational studies— the membrane is assumed to be tension free. In this work, we take cognizance of the fact that biological membranes may experience surface tension on many physiological occasions and the membrane surface tension acts as a driving force for several biological activities. We reformulate the entropic force for a fluctuating membrane with surface tension and show that entropic pressure decays with distance d exponentially when the surface tension is remarkably large. Our result is able to explain some of the experimental observations on how placing the membranes under surface tension promotes adhesion and vesicle fusions due to a significant reduction in entropic repulsive force.

2.12 Daniel Mottern, Computational Modeling of PFAS Adsorption in Metal and Covalent Organic Frameworks

New Jersey Institute of Technology
Advisor: Joshua Young

Per- and Polyfluoroalkyl substances (PFAS) are a family of several thousand chemicals that have plagued water fronts for decades, and show little sign of degradation due to the strength of their carbon-fluorine bonds. Their slow removal from the environment and the multitude of harmful health effects related with them, have led to significant research in methods for sensing these molecules, and remediating them from aqueous environments. Recently, nanoporous crystalline materials such as metal- and covalent-organic frameworks (MOFs and COFs) have gained interest as adsorbent materials for aqueous contaminants. However, despite their popularity and the urgency of the issue, experimental research has been limited to a small number of well-known MOFs, and even fewer COFs have been discussed, due to the time, cost, and risk associated with trying a new structure with little foreknowledge of how it will behave. By using molecular simulation methods, in particular Density Functional Theory (DFT), we can study the adsorption behavior of PFAS molecules on a variety of structures, and the effects of various modifications on our simulation environment.

2.13 Jason Ogbebor, Elastic Properties of Supercritical Methane in Nanopores: A Molecular Simulation Study

New Jersey Institute of Technology
Advisor: Gennady Gor

A number of thermodynamic properties of fluids are affected by confinement in nanopores. Of particular interest to this work is the compressibility. In molecular dynamics and molecular Monte Carlo simulations, calculating compressibility requires measuring fluctuations of volume (isothermal-isobaric ensemble), the internal virial (canonical ensemble), or number of particles (grand canonical Monte Carlo). This work aims to use a fourth method in the canonical ensemble: direct uniaxial compression of the fluid between two pore walls, where one wall is affixed with a spring. By perturbing the relaxed length of the spring, we aim to explicitly calculate the numerical derivative of molar volume with respect to pressure, yielding the compressibility of the fluid. To ensure that the results of this method are independent of the prescribed spring constant, multiple constants are prescribed and the results are compared. The model system is methane confined in graphitic slit pores of varying widths. The compressibility of methane in these systems has already been studied, allowing for verification of results.

2.14 Akash Pallath, Order Parameters for Enhanced Sampling of Hydrophobic Polymer Collapse

University of Pennsylvania
Advisor: Amish Patel

Conformationally flexible molecules, such as peptides and polymers, have significant importance in both biological and materials settings. In solution, these molecules often exist in two or more stable states separated by free energetic barriers. Many of these molecules have the ability to transition between these stable states in response to various stimuli, such as changes in temperature, pH, or co-solvent concentration. This property makes them useful in a broad range of applications, including catalysis and controlled drug delivery. Molecular dynamics simulations offer a systematic approach to mapping the free energy landscapes of solvated polymers and peptides, and understanding their responses to stimuli. However, even for simple hydrophobic polymers in aqueous solution, accurately sampling their conformational landscapes can be extremely hallenging, as barriers in both solute and solvent coordinates hinder transitions between their stable states.

Enhanced sampling methods such as umbrella sampling, metadynamics, and On-the-fly Probability Enhanced Sampling (OPES) bias one or more order parameters that characterize polymer conformation in an attempt to overcome these barriers and sample the underlying conformational landscape. Various studies have biased order parameters based on solute coordinates, such as radius of gyration, to characterize the free energy landscapes of coarse-grained hydrophobic polymers, often using implicit solvent treatments. However, for more accurate polymer models in explicit solvent, we show that such solute-based order parameters fail to overcome desolvation barriers in the hydrophobic collapse process, resulting in hysteresis when characterizing the underlying free energy landscape. To address this issue, we leverage our understanding of the mechanism of hydrophobic polymer collapse to develop specialized order parameters that operate on both chain and solvent coordinates, and use them in conjunction with OPES to efficiently characterize the conformational free energy landscape of hydrophobic polymers. Furthermore, we employ our techniques to quantitatively characterize the effects of temperature on the thermodynamics of collapse for one such hydrophobic polymer in water. Our methods pave the way for future work characterizing how various stimuli modulate the conformational landscapes of biomedically relevant polymers and peptides.

2.15 Shivam Parashar, Modeling Gas Adsorption within Nanopores using Mesocanonical Ensemble Simulations

Rutgers University
Advisor: Alexander Neimark

The commonly used method for simulation of adsorption isotherms is the grand canonical Monte Carlo (GCMC), which replicates the experimental conditions of isothermal adsorption measurements. However, in mesoporous and some microporous materials, pore filling involves phase transitions between vapor-like and liquid-like adsorbate states that result in hysteretic GCMC isotherms. To determine the equilibrium transition between these states, the GCMC simulations would need to be run infinitely long. The mesocanonical ensemble (MCE) was introduced to address this issue, using a gauge cell to represent a finite volume reservoir of particles that stabilizes the adsorbed phase and restricts fluctuations.[1,2] In this study, we implement the mesocanonical ensemble Monte Carlo simulation (MCE MC) method in the open-source software RASPA and validate it on MCM-41, followed by its application to seven MOFs. Our results demonstrate that the gauge cell MCE MC method accurately simulates adsorption phase transitions in nanoporous materials and is more efficient than the Widom particle insertion method in the NVT ensemble. We also discuss how to determine equilibrium transition pressure, calculate the true grand canonical isotherm, and the energy barriers separating the vapor and liquid phases. Finally, using ZIF-412 and IRMOF-74-V as examples, we explore an interesting effect of consecutive swings of the canonical isotherm when the simulation cell contains multiple pores. The gauge cell MCE MC method is an efficient alternative to other methods such as the GCMC and the transition matrix Monte Carlo (TMMC) for simulating adsorption phase transitions in nanoporous materials.

This work is supported by the NSF CBET grant 1834339.

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2. A.V. Neimark and A. Vishnyakov, A Simulation Method for the Calculation of Chemical Potentials in Small, Inhomogeneous, and Dense Systems, Journal of Chemical Physics, 2005, V. 122, 234108.

2.16 Kuldeepsinh Raj, Guiding Maps of Solvents for Lithium-Sulfur Batteries via a Computational Data-Driven Approach

Stony Brook University
Advisor: Nav Nidhi Rajput

To develop lithium-sulfur batteries into a feasible and efficient energy storage technology of the future, it is imperative to create electrolytes that can effectively regulate the dissolution of polysulfides, while also maintaining high ionic conductivity and low viscosity. Computational chemistry techniques can be utilized to understand and modify the interactions between different components at an atomic level, thus enabling the creation of electrolytes with a diverse range of desired properties. In this work, we are employing high-throughput multi-scale simulations to study various electronic, structural and dynamical properties of lithium-sulfur battery electrolytes. As a part of this work, we recently published the first open-source database known as ComBat (Computational Database for Li-S Batteries) which includes 2,000 computed quantum-chemical and molecular dynamics properties for various lithium-sulfur electrolytes that employ solvents from 16 distinct chemical classes. The research also investigates the microscopic factors responsible for the clustering of polysulfides and the various ways that electrolyte components diffuse. Contrary to popular belief that the solvent dielectric constant alone can be used to estimate the solubility of polysulfides, the study finds that multiple factors such as the solvent C/O ratio, fluorination degree, and steric hindrance effects also influence polysulfide solubility. The study also suggests using binding energies as an indicator of Li+ dissociation, which affects ionic conductivity. The findings and design guidelines provided in this research can serve as a roadmap to predict optimal electrolyte composition with multiple desired properties.

2.17 Maithreyi Ramakrishnan, Integrating Computation and Experiment in the Design of Metabolite-sequestering Peptide Nanostructures

City University of New York
Advisor:Rein Ulijn

This study aims to attain tunable multi-component peptide systems towards selective reporting or removal of neurologically significant metabolites, in particular those that are part of the tryptophan metabolic pathway. Several neurological disorders have been linked to the imbalance of metabolites such as serotonin and kynurenic acid in the gut-brain biome. To prevent this, the study focuses on the design of materials based on order vs disorder rules, as well as functional group recognition with respect to the metabolites to use molecular selectivity in sequestering them. Given the structural similarity between the metabolites, specific amino acid residues are selected based on electrostatic considerations to interact favorably with a given metabolite. Experimental and computational approaches are used to find possible candidates through which to achieve this. The experimental approach uses Dynamic Peptide Libraries (DPL) to identify peptides that assemble into favorable nanostructures in the presence of the metabolites. This involves dynamcally exchanging peptide sequences using an endoprotease such that a thermodynamic minima is achieved. This is complimented by Molecular Dynamics (MD) simulations that will investigate the extent and types of interactions these peptides have with the metabolites as well as each other. Thus, one is able to achieve a complex co-assembly system involving oppositely charged metabolites that form nanostructures with favorably interacting tetrapeptides based on pi-pi stacking and coulombic forces that is dependant on sequence. The ability of the resulting nanostructures to selectively remove and bind metabolites is currently being investigated.

2.18 Matthew Stickles, Measuring Surface Tension In Silico by the Droplet Weight Method

New Jersey Institute of Technology
Advisor: Gennady Gor

The surface tension of a liquid is one of its key properties, which affects wettability, dispersibility, and aerosol behavior. It is particularly crucial knowledge in understanding droplet formation and spreading of aerosols. Surface tension can be obtained using experimental or modeling approaches. A commonly used experimental approach is the droplet weight method. Sometimes experiment is impossible to perform due to extreme conditions, such as high and low temperature and the toxicity of the liquid. Thus, it is much more practical to perform molecular simulations. There are several viable simulation approaches, however, no other approach mimics experimentation closely. In this work we developed a simulation that will mimic the droplet weight method, in which the capillary force (due to surface tension) is balanced by the force of gravity. This approach uses a simpler fluid, like argon, which will prove that this is a viable method by which to calculate the surface tension of other liquids.

2.19 Zachariah Vicars, Understanding Surface Ice-philicity using Enhanced Sampling

University of Pennsylvania
Advisor: Amish Patel

Ice formation is a fundamental process with broad importance in fields such as atmospheric sciences, medicine, cryobiology, and transportation. Ice nucleation typically occurs heterogeneously (on a surface) due to the ability of solid surfaces to reduce nucleation barriers. However, the precise influence of surface chemistry, geometry, nanoscale texture, and other features on these barriers is complex and incompletely understood. To address this challenge, we present a robust and computationally efficient method for characterizing surface "ice-philicity" (the propensity of the surface to nucleate ice) using molecular dynamics and enhanced sampling. Using this approach, we investigate a family of model surfaces and elucidate the effects of lattice matching, surface polarity, and surface attractions on the resulting ice-philicity. Our work provides a prescription for quantifying surface ice-philicity and outlines key design principles for engineering surfaces that control ice formation.

2.20 Baris Eser Ugur, Investigation of Nuclear Quantum Effects in Condensed Phase Systems

Princeton University
Advisor: Michael Webb

Nuclear quantum effects (NQEs) play an important role in a wide range of systems, especially those involving light nuclei and low temperatures. Despite the prevalence of NQEs, computational and experimental studies investigating their impact have been limited to a narrow range of systems such as water and materials involving hydrogen ion transfer. Furthermore, present studies on NQEs demonstrate a lack of focus on the chemical features that enhance such effects and the mechanism with which they are manifested in macroscopic properties. In this study, we employ Path Integral Molecular Dynamics (PIMD) simulations to investigate the effect of NQEs on the macroscopic properties of 87 different organic liquid systems with various functional groups and molecular features, with an additional focus on the effect of deuteration on such properties. We then highlight the chemical characteristics that enhance NQEs and analyze the underlying physics determining the influence of NQEs in various macroscopic properties. Employing a second set of simulations with different force field parameters, we illustrate the force field dependence of the calculated magnitude of NQEs. Overall, this work provides a deeper understanding of how NQEs are manifested in material properties and can act as a guide to evaluate whether a specific system will require advanced simulation techniques such as PIMD to fully encapsulate the physics governing its properties.