CPP 21: Modeling and Simulation of Soft Matter II

Time: Tuesday 14:00–15:30 Location: H34

CPP 21.1 Tue 14:00 H34

Hard anisotropic particles: Excluded volume and second virial coefficients in arbitrary dimensions of space — •Markus Kulossa and Joachim Wagner — University of Rostock, Rostock, Germany

Precondition for investigations of phase behavior and equation-of-state data of many-particle systems are closed expressions for their free energy. The virial series gives access to the compressibility factor and therewith to the excess free energy of real supercritical fluids formed by hard, anisometric particles. Using Parson's approach, in many cases expressions solely based on the second virial coefficient accounting for the initial departure from ideal-gas behavior are used. In this contribution, we provide analytical expressions for the excluded volume of hard convex particles in arbitrary-dimensional Euclidean spaces. In addition to the detailed influence of the geometry and aspect ratio, we investigate the influence of singularities in the surface curvature. Analytical expressions for the excluded volume and second virial coefficients are of crucial importance for numerical calculation of higher-order virial coefficients. We provide so far unknown expressions for geometrical measures of uniaxial solids of revolution such as hyperellipsoids, hyperspherocylinders, hypercylinders, and hyperdoublecones in higher-dimensional Euclidean spaces.

CPP 21.2 Tue 14:15 H34

Navigating Chemical Space: An Active Learning Strategy Using Multi-Level Coarse-Graining — •Luis Walter and Tristan Bereau — ITP, Heidelberg University

Exploring the vast chemical compound space remains a significant challenge due to the immense number of possible molecules and limited scalability of conventional screening methods. To approach chemical space exploration more effectively, we have developed an active learning-based method that uses transferable coarse-grained models to compress chemical space into varying levels of resolution. By using multiple representations of chemical space with different coarse-graining resolutions, we balance combinatorial complexity and chemical detail. To identify target compounds, we first use an autoencoder to transform the discrete molecular spaces into continuous latent spaces. We then perform Bayesian optimization within these latent spaces, using molecular dynamics simulations to calculate target free energies of the coarse-grained compounds. This multi-level approach allows for an effective balance between exploration at lower and exploitation at higher resolutions. We demonstrate the effectiveness of our method by optimizing molecules to enhance phase separation in phospholipid bilayers. Our funnel-like strategy not only suggests optimal compounds, but also provides insight into relevant neighborhoods in chemical space. We show how this neighborhood information from lower resolutions can be used to guide the optimization at higher resolutions, thereby providing an efficient way to navigate large chemical spaces for free energy-based molecular optimization.

CPP 21.3 Tue 14:30 H34

Tuning Pore Size in Integral-Asymmetric, Isoporous Membranes via Bidisperse Diblock Copolymers — •Jiayu Xie and Marcus Müller — Institute for Theoretical Physics, Georg August University Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany

Integral-asymmetric isoporous membranes, widely used in water filtration, are fabricated using a combination of evaporation-induced self-assembly (EISA) and nonsolvent-induced phase separation (NIPS). Membranes must have pores of an optimal size; small enough to block target substances but not so small as to cause excessive water resistance. While pore size can technically be adjusted by varying the copolymer's degree of polymerization, this approach requires synthesizing new copolymers for each desired pore size. A more economical alternative is blending polymers, where tuning the blending ratio of different components offers a feasible way to control pore size. However, mixing different copolymers could lead to macrophase separation. In this study, we focus on bidisperse diblock copolymers, and employ self-consistent field theory and particle-based simulations to explore the range of pore sizes over which this blending strategy is effective in fabricating membranes. Specifically, we aim to (1) establish correlations between system parameters and pore size and (2) elucidate the effects of these parameters on the dynamic behavior during SNIPS and the resulting membrane morphologies. Our findings provide valuable insights for the cost-effective fabrication of filtration membranes with tailored pore sizes.

CPP 21.4 Tue 14:45 H34

Machine-Learning potentials to understand pairing and stacking at the origin of life — •Laurie Stevens[1], Riccardo Martina[2], Alberta Ferrarini[2], and Marialore Sulpizi[1] — [1]Faculty of Physics and Astronomy, Ruhr-Universität Bochum, Germany — [2]Chemical Science Department, Università degli Studi di Padova, Italy

When exploring the origin of life, one main question remains open: how did we get from single nucleotides to long RNA and DNA chains which then led to more complex biological structures, following the RNA world hypothesis. More specifically, we are interested in how the nucleotides interactions are able to promote the synthesis of long polynucleotides. Experimental studies suggest that free nucleotides in water spontaneously organize into small molecular columnar phases, promoting the ligation of nucleic acid chains. To uncover the mechanisms behind this self-assembly, we use Molecular Dynamics simulations combined with Machine-Learning approaches.

Ab initio methods are too computationally expensive for the timescale of interest and for the complexity of the investigated systems. To overcome this limitation, we use Neural Network Potentials (NNPs) trained with DeepMD-kit and reinforced by metadynamics. After mastering the static and dynamical properties of a single Adenosine Monophosphate (AMP) in water, we are now investigating the stacking and pairing interactions between several AMPs by predicting the free energy landscape of this system as a function of the relevant degrees of freedom.

CPP 21.5 Tue 15:00 H34

Influence of hydroxyl groups on the dielectric constant of polyethylene — •Roshal Perepadan Shaju[1], Guido Roma[1], and Xavier Colin[2] — [1]Université Paris-Saclay, CEA, Service de recherche en Corrosion et Comportement des Matériaux, SRMP, Gif sur Yvette, 91191, France — [2]PIMM, Arts et Métiers Institute of Technology, CNRS, CNAM, HESAM University, 151 Boulevard de L'Hôpital, 75013 Paris, France

Polyethylene (PE) is widely used as an insulation for electric cables in various environments, including nuclear power plants. PE is susceptible to chemical modifications when exposed to ionizing radiation and oxidative environments. Introducing polar functional groups to this semi-crystalline PE, such as carbonyls and hydroxyl (OH) groups, has been observed to modify its dielectric response. However, the precise influence of hydroxylation on the dielectric properties of semi-crystalline PE is still inadequately understood. In this paper, we employ DFT and Density Functional Perturbation Theory (DFPT) to systematically investigate the effect of OH groups on the dielectric properties of semi-crystalline PE. By modeling PE with distinct surface, amorphous, and crystalline regions, the complexity of its semi-crystalline nature is explored. The static and high-frequency dielectric constants of polyethylene systems with different concentrations of OH groups were determined. The orientation of the OH defect relative to various crystallographic axes is thoroughly examined in order to determine how the alignment of the defects in the crystal lattice may affect the dielectric tensor.

CPP 21.6 Tue 15:15 H34

Load-induced shear band formation in microscale epoxy materials — •Julian Konrad, Janina Mittelhaus, Bodo Fiedler, and Robert Meissner — Hamburg University of Technology

Thin epoxy films ($\approx 30\ \mu$m) exhibit unexpected ductile behavior under mechanical load, challenging the conventional view of thermoset materials as inherently brittle. This behavior is characterized by the formation of shear bands, as revealed by mechanical testing and infrared spectroscopy. Analysis of vibrational spectra shows redshifts in *para*-phenylene stretching vibrations due to bond elongation, and blueshifts in out-of-plane hydrogen vibrations, resulting from polarization effects in the aromatic backbone. Molecular dynamics simulations support these findings, revealing a cascade of molecular realignments driven by deformation. Notably, these structural changes are reversible upon heating, suggesting a form of frozen entropy elasticity rather than plasticity. To further investigate, we employed a novel method using the MACE neural network potential to compute vibrational spectra with *ab initio* accuracy. This approach captures strain-dependent spectral shifts, accounts for interface and size effects, and highlights key implications for the design of composite materials.