CPP 1: Gels, Polymers Networks and Elastomers I

Time: Monday 9:30–11:15 Location: H34

Invited Talk CPP 1.1 Mon 9:30 H34

Impact of smallest loops and composition fluctuations on the structure of end-linked polymer model networks — •Michael Lang, Reinhard Scholz, and Toni Müller — Leibniz-Institut für Polymerforschung, Dresden, Germany

A self-consistent scheme of differential equations is developed for predicting the frequency of the two smallest loop defects within polymer model networks. Without any adjustable parameter, we obtain excellent agreement with Monte Carlo simulations that sample loop formation only up to the given maximum loop size. The formation of loops of second generation leads to correlations between connected junctions that cannot be treated exactly by considering statistical arguments alone, which is in contrast to reversible networks where equilibrium statistics are sufficient. These correlations and the statistics of the junctions are provided by our model. Comparison with more realistic simulation data in three dimensions indicates that composition fluctuations of cross-links and chains clearly impact network formation. The differences between the statistics of the network junctions and our mean field predictions provide insight into the size of the domains with a predominance of chains or junctions and thus, regarding the quality of the mixture. Our results are highly relevant for an accurate modeling of network structure, improved estimates of the elastic properties of polymer networks, and for advanced analysis techniques of the network structure like network disassembly spectrometry or multiple quantum nuclear magnetic resonance.

CPP 1.2 Mon 10:00 H34

Hybrid PNIPAM Films for Green Hydrogen Production — •Morgan P. Le Dû[1], David P. Kosbahn[1], Thomas Baier[1], Qi Zhong[2], Apostolos Vagias[3], Robert Cubitt[3], Narendra Chaulagain[4], Karthik Shankar[4], Hagen Übele[5], Katharina Krischer[5], and Peter Müller-Buschbaum[1] — [1]TUM School of Natural Sciences, Chair for Functional Materials, Garching, Germany — [2]Zhejiang Sci-Tech University, Hangzhou, China — [3]Institut Laue-Langevin (ILL), Grenoble, France — [4]University of Alberta, Department of Electrical and Computer Engineering, Edmonton, Canada — [5]TUM School of Natural Sciences, Nonequilibrium Chemical Physics, 85748 Garching, Germany

Platinum (Pt) doped carbon nitride (CN) is a promising photocatalyst under visible light for green hydrogen (H2) production. We develop this system in a thin polymer film to make it industrially scalable. The poly(N-isopropylacrylamide) (PNIPAM) hydrogel is used as a host matrix and water storage medium to facilitate homogeneous dispersion of the catalytic centers. The hybrid film's vertical distribution and inner microstructure are studied under in situ conditions with time-of-flight neutron reflectometry (ToF NR) and grazing incidence small angle neutron scattering (GISANS). The resulting H2 produced is measured by gas chromatography.

CPP 1.3 Mon 10:15 H34

Effect of different network topologies on swelling and mechanical properties of polyelectrolyte hydrogels — •Somesh Kurahatti, Mariano Brito, David Beyer, and Christian Holm — Institute for Computational Physics, Stuttgart, Germany

We investigate the properties of polyelectrolyte hydrogels with various network heterogeneities, particularly their swelling capacity, elastic modulus and salt partitioning. We benchmark the scaling predictions for elastic modulus and equilibrium swelling ratio of the hydrogels under various salinity conditions, verifying the theoretically predicted behavior. Decoupling the elastic modulus from the swelling behavior is beneficial for applications such as super absorbers. Here, for example, now wants to increase the swelling ratio while maintaining a reasonable mechanical strength. We therefore explore mechanical and structural properties of the hydrogels with varying topologies, namely gels with dangling ends, gels with floating chains, and bottle-brush gels in counterpart to a reference regular gel. We observe that incorporating dangling ends changed swelling ratio and bulk modulus inline with the scaling predictions, whereas bottle-brush and floating-chain gels deviate from the predictions. Specifically, floating chains resulted in higher moduli and higher swelling ratio, while bottle-brush gels resulted in lower moduli and lower swelling ratios than the regular counterpart, each maintaining the same swelling ratio and modulus. The new swelling-mechanical relations allow us to treat them in a decoupled manner via the topology variation, which turns out to be of paramount relevance in the optimization and on-demand design of hydrogels.

CPP 1.4 Mon 10:30 H34

Aqueous foams stabilized by PNIPAM microgels: A multiscale investigation — •Joanne Zimmer, Luca Mirau, Gaëtan Barth, and Regine von Klitzing — Soft Matter at Interfaces, Institute for Condensed Matter Physics, TU Darmstadt, Hochschulstraße 8, D-64289 Darmstadt

Foams possess several structural motifs on different length scales with the smallest building blocks being the foam films or even the liquid/air interface. In this work, PNIPAM microgels (MG) are used as foam stabilizers as they are highly interfacial active and temperature responsive. In addition, the MG stiffness and by this their interfacial activity can be tuned by variation of the crosslinker content. We present a multi-scale approach towards understanding how the MG properties are related to the macroscopic foam and foam film stability. Spatially resolved disjoining pressure isotherms are measured with a Thin Film Pressure Balance (TFPB). The isotherms provide information about the film thickness, surface mobility and film stability. These properties are correlated with macroscopic foam characteristics such as foamability, stability and drainage dynamics. Indeed, the results are in good agreement on all length scales: A lower crosslinker content supports foam formation due to a faster surface coverage and results in an increased foam stability, as a more rigid surface layer is formed. Increasing the amount of crosslinker in the MG induces a transition from Poiseuille flow to plug flow in the foam during drainage which coincides with an increasing mobility of the surface of a single foam film measured in a TFPB.

CPP 1.5 Mon 10:45 H34

Phase separation in elastic polymer networks — •Takahiro Yokoyama[1,2], Yicheng Qiang[3], Chengjie Luo[3], Oliver Paulin[3], David Zwicker[3], and Arash Nikoubashman[1,2] — [1]Leibniz-Institut für Polymerforschung Dresden e.V., Dresden, Germany — [2]Technische Universität Dresden, Dresden, Germany — [3]Max-Planck-Institut für Dynamik und Selbstorganisation, Göttingen, Germany

Phase separation in polymer networks, from synthetic gels to biopolymer networks, significantly influences their structural and mechanical properties. While previous studies suggest that network mechanics regulate phase separation of the network itself and the embedded (complex) liquid, the fundamental principles governing this interaction remain poorly understood. To address this knowledge gap, we used molecular dynamics (MD) simulations to explore the phase behavior of elastic polymer networks. We systematically changed the quality of the (implicit) solvent, polymer chain length, and polymer flexibility. Our simulations identified two distinct phase separation behaviors depending on the polymer flexibility: macrophase separation into a polymer-rich and polymer-poor region for flexible polymer chains, and microphase separation with finite-sized pores for networks formed by semi-flexible chains. For the latter systems, we found that the pore size decreased with increasing chain stiffness. Furthermore, the network elasticity increased with decreasing pore size, following a power law as recently proposed by field theory. This study highlights the importance of polymer stiffness in shaping the size and distribution of (biomolecular) droplets within elastic networks.

CPP 1.6 Mon 11:00 H34

Nanoscopic measurements of the water content in microgels using fluorescence lifetime imaging — •Dominik Wöll[1], Alexandre Fürstenberg[2], Jörg Enderlein[3], Oleksii Nevskyi[3], Sankar Jana[1], Hannah Höche[1], and Leon Trottenberg[1] — [1]RWTH Aachen University, Institute of Physical Chemistry, Aachen, Germany — [2]University of Geneva, Department of Physical Chemistry and Department of Inorganic and Analytical Chemistry, Genf, Switzerland — [3]Georg August University, 3rd Institute of Physics - Biophysics, Göttingen, Germany

The role of water molecules in the structure, function, and dynamics of (bio-)materials is significant, and thus, an estimation of the number of water molecules within different compartments is crucial. It has been demonstrated that the fluorescence of red emissive dyes is quenched in the presence of H_2O, with the excited energy transferred to surrounding H_2O molecules. Fluorescence lifetime imaging (FLIM) can thus be used to quantify the number of water molecules in proximity to the probing dye Atto 655, which was covalently embedded into microgels in the present study. Microgels represent an intriguing class of nanoparticles with considerable potential for applications in drug delivery and medicinal chemistry. We recorded FLIM images at varying H_2O:D_2O ratios, in both the swollen and collapsed states of the microgels at 22 °C and 40 °C, respectively. Stern-Volmer analysis enabled the calculation of the number of water molecules in the immediate vicinity of the dye molecule within the microgels at varying temperatures.