

## CPP 5: Polymer Networks and Elastomers I

Time: Monday 9:30–12:45

Location: PC 203

**Invited Talk**

CPP 5.1 Mon 9:30 PC 203

**Strain-controlled criticality governs the nonlinear mechanics of fibre networks** — ●ABHINAV SHARMA<sup>1,2</sup>, ALBERT LICUP<sup>2</sup>, KARIN JANSEN<sup>3,4</sup>, ROBBIE RENS<sup>2</sup>, MICHAEL SHEINMAN<sup>2</sup>, JORDAN SHIVERS<sup>5</sup>, JINGCHEN FENG<sup>5</sup>, GIJSJE KOENDERINK<sup>3</sup>, and FRED MACKINTOSH<sup>2,5</sup> — <sup>1</sup>Leibniz Institute for polymer research, Dresden — <sup>2</sup>Vrije Universiteit Amsterdam — <sup>3</sup>AMOLF, Amsterdam — <sup>4</sup>University of Manchester, UK — <sup>5</sup>Rice University, Houston, USA

Disordered fibrous networks are ubiquitous in nature as major structural components of living cells and tissues. The mechanical stability of networks generally depends on the degree of connectivity: only when the average number of connections between nodes exceeds the isostatic threshold are networks stable. On increasing the connectivity through this point, such networks undergo a mechanical phase transition from a floppy to a rigid phase. However, even sub-isostatic networks become rigid when subjected to sufficiently large deformations. To study this strain-controlled transition, we perform a combination of computational modelling of fibre networks and experiments on networks of type I collagen fibres, which are crucial for the integrity of biological tissues. We show theoretically that the development of rigidity is characterized by a strain-controlled continuous phase transition with signatures of criticality. Our experiments demonstrate mechanical properties consistent with our model, including the predicted critical exponents.

CPP 5.2 Mon 10:00 PC 203

**Rheology of cross-linked polymer networks** — ●JAKOB LÖBER — Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

Polymer networks and gels may exhibit complex rheological behavior as e.g. an increase of the shear modulus in time known as rheological aging, shear rejuvenation, and a yield stress. I present a model for cross-linked polymer networks which treats the number of cross-links as a thermodynamic variable. This number may change due to a chemical reaction modeled as a rate equation, with a chemical equilibrium constant depending on mechanical deformations. Thus applying e.g. a large shear deformation affects the number of cross-links, leading to nonlinear threshold behavior reminiscent of shear rejuvenation and yield stress.

CPP 5.3 Mon 10:15 PC 203

**Separation of entropy and energy elastic contributions and the strain induced crystallization during the deformation of rubber** — ●KONRAD SCHNEIDER — Leibniz-IPF Dresden, Germany

Matrix-filler-interaction as well as strain induced crystallization (SIC) are main processes, responsible for the outstanding mechanical performance of natural rubber. By IR thermography combined with synchrotron x-ray diffraction it is possible to separate different mechanisms of deformation, reinforcement and failure in unfilled as well as filled rubber systems. Some recent measurements will be presented and discussed in detail.

CPP 5.4 Mon 10:30 PC 203

**Shear deformation of entangled and unentangled polymer networks: A Monte-Carlo-Study** — ●TONI MÜLLER<sup>1,2</sup>, MICHAEL LANG<sup>1</sup>, and JENS-UWE SOMMER<sup>1,2</sup> — <sup>1</sup>Leibniz-Institut für Polymerforschung Dresden — <sup>2</sup>Technische Universität Dresden

Using a highly efficient GPU version of the bond-fluctuation model, we study the elastic properties of end-linked 4-functional model networks where we can switch off either entanglements or both entanglements and excluded volume. With that we can separate the contribution of the connectivity and the topological constraints to the shear modulus and analyze them as a function of the strain in a simple shear deformation in our computer simulations. For the connectivity contribution, we take into account network defects, finite cyclic structures and the effect of excluded volume. The splitting of the contributions to the shear modulus from the connectivity and the entanglements is assumed in theory and it is checked explicitly with our simulation data. We estimate the entanglement degree of polymerization and compare its scaling with existing studies confirming the theoretical prediction,  $N_e(\phi) = N_e(1)\phi^{1.3}$ .

CPP 5.5 Mon 10:45 PC 203

**The elasticity of phantom networks with cyclic and linear defects** — ●MICHAEL LANG — Institut Theorie der Polymere, Leibniz Institut für Polymerforschung Dresden, Hohe Straße 6, 01069 Dresden, Germany

The resistor network analogy is used to compute the phantom modulus of realistic polymer networks containing finite cycles. We correct a recently published approach [1], connect with results on the distribution of cyclic defects, and generalize to networks of arbitrary junction functionality containing both cyclic and linear defects. It turns out that the impact of finite cyclic structures on rubber elasticity was previously [1] overestimated and that a consideration of the smallest dangling loops in combination with a precise estimate of conversion is sufficient in most cases for estimating the phantom modulus of the network.

[1] M. Zhong, R. Wang, K. Kawamoto, B. D. Olsen, J. A. Johnson. *Science*, 353:1264–1268, 2016.

**15 min. break**

CPP 5.6 Mon 11:15 PC 203

**Network formation and dynamics in H-bonding telechelic polymers: competition between association time and structural relaxation** — ●MARTIN TRESS<sup>1</sup>, KUNYUE XING<sup>1</sup>, PENGFEI CAO<sup>2</sup>, SHIWANG CHENG<sup>1</sup>, TOMONORI SAITO<sup>2</sup>, VLADIMIR NOVIKOV<sup>1</sup>, and ALEXEI SOKOLOV<sup>1,2</sup> — <sup>1</sup>University of Tennessee Knoxville, Department of Chemistry, Knoxville, Tennessee, USA — <sup>2</sup>Oak Ridge National Lab, Chemical Sciences Division, Oak Ridge, Tennessee, USA

Reversible bonds between polymer chains can form supra-molecular networks which exhibit extraordinary mechanical properties. In fact, reversible bonds are a promising route to functional materials with self-healing properties. We study short telechelic polymers with H-bonding end-groups of different interaction strength and backbone flexibility. The glass transition temperature of flexible polydimethyl siloxanes (PDMS) does not vary with H-bond strength, but differs strongly from methyl-terminated PDMS. At the same time,  $T_g$  of the much stiffer telechelic polypropylene glycol (PPG) depends significantly on the H-bond strength. In contrast, viscosity strongly depends on the H-bond strength in the PDMS series while it remains almost the same in PPG with different end groups. Complementary measurements of shear modulus and dielectric relaxation indicate that these observations can be explained by competing lifetimes of supra-molecular associations and structural relaxations. Only if the association lifetime exceeds the characteristic segmental or chain relaxation time, the glass transition or viscosity will be affected by chain associations. Otherwise the chain end associations do not influence significantly  $T_g$  or viscosity.

CPP 5.7 Mon 11:30 PC 203

**Molecular properties and growth conditions associated with PSS diffusion during annealing in polyelectrolyte multilayers** — ●ANNKATRIN SILL, AMIR AZINFAR, SVEN NEUBER, PETER NESTLER, and CHRISTIANE A. HELM — Inst. f. Physics, Greifswald University, Germany

During annealing of polyelectrolyte multilayers in concentrated solutions (1 M NaCl) interdiffusion of polyelectrolytes occurs. We investigate the interdiffusion perpendicular to the substrate using neutron reflectivity and selectively deuterated polyanions (poly(styrene sulfonate), PSS). Multilayers formed at 10 mM consist of flatly adsorbed chains. The diffusion constant of PSS can be tuned by five orders of magnitude; it decreases exponentially with the degree of polymerization of PDADMA (poly(diallyldimethylammonium)). Multilayers formed at 100 mM NaCl consist of interdigitated chains. When the degree of polymerization of PDADMA exceeds the one of PSS, the diffusion constant drops suddenly by three orders of magnitude and remains low. Such sudden transitions are better known from network than from polymer theory.

CPP 5.8 Mon 11:45 PC 203

**Application of the dynamic flocculation model for investigations of silica filled rubbers** — ●DAVID LOCKHORN and MANFRED KLÜPPEL — Deutsches Institut für Kautschuktechnologie e.V., Eupener Straße 33, D-30519 Hannover, Germany

The dynamic flocculation model (DFM) was introduced to describe the strong non-linear stress-strain behavior of filler reinforced rubbers.

The model is able to describe stress-softening as well as hysteresis by a combination of well-established concepts for rubber elasticity and a micromechanical approach for the dynamic filler flocculation in strained rubbers. The stress value at a given strain leads back to an ongoing breakdown and reaggregation of filler clusters. Multihysteresis measurements of silica filled natural rubber (NR) and styrene-butadiene rubber (SBR) compounds will be described by the DFM and some of the physically meaningful parameters will be interpreted with respect to reinforcement in NR/Silica compounds. Besides this, the physicals as well as the modulus with increasing strain which is a measure for the Payne effect will be shown for the used model compounds. The results lead to the conclusion that there is a good coupling between silica and NR especially for the coupling agent silane as expected. But by looking at the Payne effect there is an unexpected high value for the coupling agent compounds in NR. This indicates that the micro-dispersion is not sufficient.

CPP 5.9 Mon 12:00 PC 203

**Modeling magnetic elastomers as coatings with controllable hydrophobicity** — ●PEDRO A. SANCHEZ<sup>1</sup>, ELENA MININA<sup>1</sup>, SOFIA S. KANTOROVICH<sup>1,2</sup>, and ELENA YU. KRAMARENKO<sup>3</sup> — <sup>1</sup>Computational Physics, University of Vienna, Vienna, Austria — <sup>2</sup>Ural Federal University, Ekaterimburg, Russia — <sup>3</sup>Moscow State University, Moscow, Russia

Magnetic elastomers are hybrid materials consisting of a soft matrix of polymers with a high volume fraction of embedded magnetic micro- and/or nanoparticles. These materials are elastic enough to experience strong structural changes as a response to external magnetic fields. This makes them promising candidates for a broad range of applications.

One particularly interesting application of magnetic elastomers is their use as thin coatings that provide a fine control of the hydrophobicity of their free surface, as a consequence of the strong changes in its roughness induced by the application of proper external fields.

In this contribution we present a minimal computer simulation model of a magnetic elastomer thin coating that captures the dependence of its surface roughness on the elastic properties of the polymer matrix and the magnetic interactions. This modeling approach may contribute to the optimum design of these systems.

CPP 5.10 Mon 12:15 PC 203

**FORC diagrams in different systems of magnetic elas-**

**tomers studying by molecular dynamics simulations** — ●ALLA DOBROSEDOVA<sup>1</sup>, PEDRO A. SANCHEZ<sup>2</sup>, and SOFIA KANTOROVICH<sup>1,2</sup> — <sup>1</sup>Ural Federal University, Ekaterinburg, Russia — <sup>2</sup>University of Vienna, Vienna, Austria

Magnetic elastomers are the systems consisting of magnetic particles distributed in a nonmagnetic elastic matrix. We use the FORC (first-order reversal curves) diagrams to study how the matrix influences on internal magnetic interactions. We use the Molecular Dynamics Simulations to model the different systems. We consider fixed nonmagnetic particles which are connected with dipolar ones by elastic interactions. We study the systems with translational constraints only as well as the systems with translational and rotational constraints. The first step is to get the main hysteresis loop of this system in computer simulations (the direction and intensity of an external magnetic field are changing). After that we need to perform the computer simulations to obtain the first-order reversal curves. In the result, we have dependence of FORC diagrams on coercive and reversal fields. We use classical method [C. R. Pike et al., J. Appl. Phys. 85, 6660 (1999)] to get the FORC diagrams. The research was supported by the Ministry of Education and Science of the RF (project 3.1438.2017/4.6) and Austrian Science Fund (FWF, START-Project No. Y 627-N27).

CPP 5.11 Mon 12:30 PC 203

**Marginally compact hyperbranched macromolecular trees** — ●MAXIM DOLGUSHEV — Laboratoire de Physique Théorique de la Matière Condensée, Université Pierre et Marie Curie, Paris, France

This contribution presents our recent studies [1,2] on fractal hyperbranched trees with a Gaussian chain statistics, which are marginally compact. Marginal compactness means that in the  $d = 3$  dimensional space the average size  $R$  of the trees follows  $N \sim R^3$  (where  $N$  is the molecular mass of the tree), and at the same time for their surface  $A$  the relation  $A \sim N$  holds. We show that albeit the self-contact density  $\rho_c$  diverges for marginally compact objects logarithmically with the molecular weight  $N$ , this issue can be overcome by introducing linear spacers. Indeed, the spacers of length  $S$  yield a  $\log(N/S)/S^{1/2}$  behaviour, so that the strong decay with  $S$  bits rapidly the logarithmic divergence [1]. Another recipe for suppression of the self-contact density  $\rho_c$  is introduction of local stiffness [2].

[1] M. Dolgushev, J. P. Wittmer, A. Johner, O. Benzerara, H. Meyer, and J. Baschnagel, *Soft Matter* 13, 2499-2512 (2017).

[2] M. Dolgushev, A. L. Hauber, P. Pelagejcev, and J. P. Wittmer, *Phys. Rev. E* 96, 012501 (2017).