

CPP 49: Elastomers and Gels

Time: Friday 10:15–12:15

Location: H48

CPP 49.1 Fri 10:15 H48

Reinforced elastomers: Measurement of the strain at various scales and mesoscale modelling of reinforcement and non linear properties. — ●PAUL SOTTA¹, STÉPHANE DUPRES², SAMY MERABIA³, PIERRE-ANTOINE ALBOUY², and DIDIER LONG¹ — ¹Laboratoire Polymères et Matériaux Avancés CNRS/Rhodia, Saint-Fons, France — ²Laboratoire de Physique des Solides, CNRS/Université Paris 11, Orsay, France — ³Laboratoire de Physique de la Matière Condensée et Nanostructures, Université de Lyon, France

Combining mechanics, 2H NMR on stretched samples and measurement of strain-induced crystallization, we have measured the strain at various scales in polyisoprene elastomers reinforced with carbon black. We have shown that local strain inhomogeneity is more pronounced in the presence of more reinforcing carbon black grades [S. Dupres et al., *Macromolecules*, 2009, 42, 2634]. Related to these and other experimental results, we have developed a new mesoscale modelling of reinforced elastomers in which the glass transition temperature shift of the elastomer matrix close to fillers is implemented. The model gives a unified picture of an ensemble of complex phenomena in both the linear and nonlinear regimes of reinforced elastomers: reinforcement over a large temperature range, Payne and Mullins effects, plasticity [S. Merabia et al., *Macromolecules*, 2008, 41, 8252; S. Merabia et al., submitted to *J. Polym. Sci. B, Polym. Phys.*, 2009].

CPP 49.2 Fri 10:30 H48

Caloric and optical studies of the influence of fumed silica nanoparticles on the cross-linking of polydimethylsiloxane — ●ULRICH MÜLLER, BARTOSZ ZIELINSKI, MARTINE PHILIPP, ROLAND SANCTUARY, and JAN KRISTIAN KRÜGER — Université du Luxembourg, Laboratoire de Physique des Matériaux, Campus Limpertsberg, 162a avenue de la Faïencerie, L-1511 Luxembourg

Silicone rubbers based on polydimethylsiloxane (PDMS) are synthetic elastomers with outstanding properties such as chemical resistance, wide range temperature resistance, biocompatibility etc. Furthermore since the late 80s there is an increasing interest in improving the silicone rubbers properties by the introduction of fumed silica particles. Hydrogen groups on the particle's surface are known to exhibit strong interaction with the siloxane backbone of PDMS. But there are only a few studies concerning the influence of these interactions on property evolution during the cross-linking process. We showcase that fumed silica nanoparticles have a profound impact on the cross-linking of a room temperature vulcanized silicone rubber as evidenced by caloric and optical studies. Another point of view on the cross-linking process is given by the first-time implementation of temperature modulated refractometry (TMR) that allows for an estimation of the thermal expansion coefficient during isothermal processes. Caloric and optical studies show a differing behavior as a function of the nanoparticle concentration that will be in the focus of the discussion.

CPP 49.3 Fri 10:45 H48

Theory of light-induced deformation of azobenzene elastomers — ●VLADIMIR TOSHCHEVIKOV^{1,2}, MARINA SAPHIANNIKOVA¹, and GERT HEINRICH¹ — ¹Leibniz-Institut für Polymerforschung Dresden e.V., Hohe Str. 6, 01069 Dresden, Germany — ²Institute of Macromolecular Compounds, Bolshoi pr. 31, 199004 Saint-Petersburg, Russia

Photosensitive elastomers bearing azobenzene moieties in their strands are able to change the shape under light irradiation. These polymers have a fascinating potential for technical applications as smart materials (microrobots, micropumps, artificial muscles). Recently, we have proposed a microscopic theory [1] of light-induced deformations in amorphous azobenzene polymers consisting of short oligomers which bear azobenzene moieties in their side chains. In the present work we extend the theory [1] to azobenzene elastomers taking the chain structure of network strands explicitly into account. We show that similar to the amorphous azobenzene polymers the photo-elastic behaviour of azobenzene elastomers is very sensitive to their chemical structure. Depending on it, a sample can be either stretched or uniaxially compressed along the electric vector of the linearly polarized light. For some chemical structures, elongation of a sample displays a non-monotonic behaviour with the light intensity. The results of the theory

are compared with experimental data for azobenzene elastomers.

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[1] V.Toshchevnikov, M.Saphiannikova, G.Heinrich. *J. Phys. Chem. B* 2009, 113, 5032.

CPP 49.4 Fri 11:00 H48

Magnetically induced surface instabilities in a thermoreversible ferrogel — ●TOBIAS LANG¹, CHRISTIAN GOLLWITZER¹, MARINA KREKHOVA², GÜNTHER LATTERMANN², INGO REHBERG¹, and REINHARD RICHTER¹ — ¹Experimentalphysik V, Universität Bayreuth — ²Makromolekulare Chemie I, Universität Bayreuth

The viscoelastic properties of thermoreversible ferrogels [1] can be tuned via temperature variation, making them an exciting type of magnetic soft matters. We study the formation of surface instabilities in a layer of thermoreversible ferrogel when exposed to a homogeneous vertical magnetic field. The dynamics of the surface evolution is recorded using an specialized X-ray technique [2]. The ferrogel samples are characterized using rheological and magnetometer measurements. The critical magnetic field for the formation of Rosensweig cusps [3] is determined and compared to theoretical predictions [4]. In a conduction to [5] the growth rates of the emerging Rosensweig patterns are measured.

[1] G. Lattermann and M. Krekhova, *Macromol. Rapid. Commun.* 27, 1273 (2006)[2] R. Richter and J. Bläsing, *Rev. Sci. Instrum.* 72, 1729 (2001)[3] M.D. Cowley and R.E. Rosensweig, *J. Fluid Mech.* 30, 671 (1967)[4] S. Bohlius, H. Brand, H. Pleiner, and M. Gels, *Z. Phys. Chem.*, 220, 97-104 (2006)[5] C. Gollwitzer, M. Krekhova, G. Lattermann, I. Rehberg, R. Richter, *Soft Matter* 5, 2093 (2009)

CPP 49.5 Fri 11:15 H48

Segmental order of entangled polymer networks is controlled by monomer fluctuations along the confining tube — ●MICHAEL LANG and JENS-UWE SOMMER — Leibniz Institut für Polymerforschung, Hohe Straße 6, 01069 Dresden

The tube model of entangled chains is applied to compute segment fluctuations and segmental orientational order parameter in polymer networks. The latter is essential for interpreting NMR measurements of entangled polymer networks. The sliding motion of monomers along the tube axis leads to a non-homogeneous reduction of segmental order along the chain. For network strands of length N much larger than entanglement length N_e , the average segmental order decreases $\sim (N_e N)^{-1/2}$ in marked contrast to the $1/N_e$ contribution of entanglements to network elasticity. As consequence, network modulus is not proportional to segmental order in entangled polymer networks. Monte Carlo simulation results of polymer networks over a wide range of molecular weights are in quantitative agreement with the theoretical predictions. The impact of entanglements on these properties is directly tested by comparing with simulations where entanglement constraints are switched off.

CPP 49.6 Fri 11:30 H48

NMR observation of "quantized" defect structures in hydrogels — ●FRANK LANGE¹, TAKAMASA SAKAI², and KAY SAALWÄCHTER¹ — ¹Institut für Physik - NMR, Betty-Heimann-Str. 7, 06120 Halle, Germany — ²Department of Bioengineering, School of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan

We present data on a new class of hydrogels, the so called Tetra-PEG. These biodegradable gels are made from four-arm macromers with well-defined molecular weight. End-linking of the precursor macromers in semi-dilute solution results in gels with a highly homogeneous microstructure and advanced mechanical properties, in contrast to conventional hydrogels, which are usually very heterogeneous. Investigations were performed at a low-field NMR spectrometer with a proton multiple-quantum pulse sequence.

Analysis of the data shows a step-like increase of the normalized double-quantum buildup curve, which is due to a well-defined microscopic defect structure of the hydrogels. Such unique structural details are here observed for the first time. The observation becomes possible because of the unique overall homogeneity of the samples. The

residual dipolar coupling constant derived from the buildup curves is proportional to the inverse of the network chain molecular weight, and based on this we conclude that the defects are due to multi-linking between two macromers. The dependence of the amounts and types of defects on various parameters such as concentration or stoichiometry variations is investigated in detail.

CPP 49.7 Fri 11:45 H48

Coupling strength between optical polarizability and structural formation during the chemically induced gelation and glass transition in epoxies — •MARTINE PHILIPP¹, CARSTEN WEHLACK², WULFF POSSART², ULRICH MÜLLER¹, ROLAND SANCTUARY¹, and JAN KRÜGER¹ — ¹Laboratory for condensed matter physics, University of Luxembourg, Luxembourg — ²Lehrstuhl für Adhesion und Interphasen in Polymeren, Universität des Saarlandes, Germany

The chemically induced gelation and glass transition are investigated during the network formation in epoxies. The refractive index turns out to be astonishingly sensitive to structural formation close to the macroscopically isostructural phase transitions. Indeed, the evolution of the refractive index is not only determined by the mass density's behaviour, in the sense of the Lorentz-Lorenz relationship, but additional relevant contributions of the electronic polarizability at optical frequencies are evidenced. The gelation and the glass transition seem to influence the optical polarizability in strongly different manners. Topological disturbances, like nanoparticles will be shown to diminish the optical polarizability during gelation close to the sol-gel transi-

tion. High performance refractometry and infrared spectroscopy are combined as experimental techniques.

CPP 49.8 Fri 12:00 H48

Surface-near structuring of pressure sensitive adhesive films — •ALEXANDER DIETHERT¹, VOLKER KÖRSTGENS¹, DAVID MAGERL¹, YANA PEYKOVA², OLGA LEBEDEVA², NORBERT WILLENBACHER², and PETER MÜLLER-BUSCHBAUM¹ — ¹TU München, Physik Department LS E13, James-Franck-Str. 1, 85747 Garching (Germany) — ²Universität Karlsruhe (TH), Institut für Mechanische Verfahrenstechnik und Mechanik, Gotthard-Franz-Str. 3, 76131 Karlsruhe (Germany)

Pressure sensitive adhesives (PSAs) are used for many different applications, such as for example adhesive foils or binding materials. A prominent class of PSA films is based on statistical copolymers. In the presented work we focus on the PSA model system P(EHA-stat-MMA) consisting of the tacky monomer ethyl hexylacrylate and the glassy monomer methyl methacrylate. The PSA films were prepared with solution casting.

In a previous x-ray reflectivity investigation we could prove the existence of enrichment layers of one type of monomer near the sample surface. Grazing incidence small angle x-ray scattering (GISAXS) experiments now added a new aspect: in addition to the layering also lateral structures are present. The data are modeled with cylindrical PMMA objects in a PEHA matrix. In particular, the evolution of the radii and the center-to-center distance during aging of the PSA film is quantified. Furthermore, the effect of this reorganization process on the adhesive properties is monitored with mechanical tack tests.