CPP 3 Polymer Materials

Time: Monday 10:00–12:30 Room: ZEU 114

CPP 3.1 Mon 10:00 ZEU 114

Generalized Cauchy Relation For Reactive Polymers: Epoxies and Polyurethanes, a comparison — •Christelle Vergnat[1], Martine Philipp[2], Ravi Bactavatchalou[1], Jörg Baller[2], Wulf Possart[1], Ulrich Müller[1], Patrick Alnot[3], Roland Sanctuary[2], and Jan K. Krüger[1] — [1]Laboratoire Europpéen de Recherche Universitaire Saarland-Lorraine-Luxembourg Universität des Saarlandes D-66123 Saarbrücken — [2]Université de Luxembourg, 162a,Avenue de la Faiencerie L-1511 Luxembourg — [3]Université de Nancy, Bd des Aiguillettes F-54506 Vandoeuvre

The generalized Cauchy Relation c11=B c44+A for isotropic materials is a linear transformation which strongly reduces the independence of the two remaining elastic constants c11 and c44 of the isotropic state. The pre-factor B usually reflects the global symmetry and has a value of three. The parameter A seems to be indicative to the difference between global and local symmetry of the material. At sufficient high probe frequencies the generalized relation even holds for the liquid state and doesn*t change at the transition to the glassy state. As a matter of fact the generalized Cauchy relation is even maintained during the curing process of reactive polymers. Within this contribution we present Cauchy Relations for epoxies and polyurethanes. The results will be discussed in the context of local symmetry and local heterogeneity.

CPP 3.2 Mon 10:15 ZEU 114

The Chemical Glass Transition as seen by High Performance Brillouin spectroscopy: Epoxies and Polyurethanes, a comparison — •Martine Philipp[1], Christelle Vergnat[2], Ravi Bactavatchalou[2], Jörg Baller[1], Wulf Possart[2], Ulrich Müller[2], Didier Rouxel[3], Roland Sanctuary[1], and Jan K. Krüger[2] — [1]Laboratoire Européen de Recherche Universitaire Saarland-Lorraine-Luxembourg Université de Luxembourg 162a, avenue de la Faiencerie L-1511 Luxembourg — [2]Universität des Saarlandes D-66123 Saarbrücken — [3]Université de Nancy I, Bd des Aiguillettes F-54506 Vandoeuvre

The nature of the glass transition and the nature of the glassy state are still a matter of debate. There are two contradictory hypothesis which do compete: The kinetic hypothesis and the transition hypothesis. According to the kinetic hypothesis the glass transition is due to a cross-over of the relevant intrinsic alpha-relaxation time with the time constant of the measurement probe in addition to the patience of the experimentalist. It is therefore of great interest to investigate a type of glass transition which by definition does not suffer from such a cross-over between the experimental time scale and the intrinsic relaxation times. For that purpose we present in this contribution investigations of the chemically induced glass transition. As model substances we have chosen the two reactive polymers: epoxy and polyurethane. As a sensitive probe for the detection of the chemical glass transition we use so-called mode-Grüneisen parameters. These parameters have been measured with high performance Brillouin spectroscopy. The results will be compared with other physical quantities as the specific heat and the refractive index.

CPP 3.3 Mon 10:30 ZEU 114

Strain dependent structural changes of spider dragline silk observed with single fiber x-ray diffraction — •Anja Glisovic[1], Thorsten Vehoff[1], Richard Davies[2], Christian Riekel[2], and Tim Salditt[1] — [1]Institut für Röntgenphysik, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany — [2]ESRF, 6 rue Jules Horowitz, BP220, 38043 GRENOBLE CEDEX, FRANCE

Spider silk is a biopolymer which has attracted more and more attention in recent years due to its unique combination of tensile strength and viscoelastic extensibility. In order to study the correlation between structure and mechanical properties we combined stress-strain measurements on single dragline fibers with x-ray diffraction. The experiments were carried out at ESRF's microfocus beamline ID 13 in Grenoble. With this method the structural changes of the fiber's crystalline parts could be studied. The changes of the crystallites are quite small and their contribution to the mechanical properties is discussed.

— 15 min. break —

CPP 3.4 Mon 11:00 ZEU 114

High-Performance Polypropylene pipes via biaxial processing — •Ralf Kleppinger[1], Patrick Voets[2], Maria Soliman[2], Colin Morath[3], and Ian Ward[3] — [1]DSM Research, Geleen, The Netherlands — [2]SABIC EuroPetrochemicals, Geleen, The Netherlands — [3]Department for Physics and Astronomy, Leeds University, Great Britain

Besides revealing a complex polymorphism that is affected by thermal history but also specific additives, recent studies have also revealed that polypropylene reveals a unique crystallization behavior when subjected to sufficiently strong shear fields. The interplay among both is of crucial importance with respect to the morphology and finally determines macroscopic properties of the material. Using polypropylene pipes as a practical example we demonstrate how processing-induced orientation in the material can either result in deterioration or enhancement of properties. Our results demonstrate that by using the die-drawing process a significant improvement of the properties of polypropylene pipes can be achieved.

CPP 3.5 Mon 11:15 ZEU 114

Scattering from polymer networks under elongational strain — •Carsten Svaneborg[1], Ralf Everaers[1], and Gary S. Grest[2] — [1]Max-Planck-Institut für Physik komplexer Systeme, Nöthnitzer Straße 38, 01187 Dresden — [2]Sandia National Laboratories, Albuquerque, NM 87185, USA

Molecular dynamics simulations are used to sample the single-chain form factor of labelled sub-chains in model polymer networks under elongational strain. We observe very similar results for randomly cross-linked and for randomly end-linked networks with the same average strand length and see no indication of lozenge-like scattering patterns reported for some experimental systems. Our data analysis shows that a recent variant of the tube model quantitatively describes scattering in the Guinier regime as well as the macroscopic elastic properties. The observed failure of the theory outside the Guinier regime is shown to be due to non-Gaussian pair-distance distributions. Nevertheless, the results presented here [1] and in Ref. [2,3] open up the possibility to systematically link SANS studies of *microscopic* deformations to the *macroscopic* elastic properties of rubber-elastic systems.

[1] C. Svaneborg, G. S. Grest, and R. Everaers. Euro. Phys. Lett. accepted.
[2] C. Svaneborg, G. S. Grest, and R. Everaers. Phys. Rev. Lett., 93:257801, 2004.
[3] C. Svaneborg, G. S. Grest, and R. Everaers. Polymer, 46:4283, 2005.

CPP 3.6 Mon 11:30 ZEU 114

The study of the relation between cavitation and microfibrillation in HDPE using time resolved synchrotron X-ray scattering during tensile deformation — •Konrad Schneider[1], Sondes Trabelsi[2], Stamm Manfred[1], and Riekel Christian[3] — [1]Leibniz-Institut für Polymerforschung Dresden, Hohe Str. 6, 01069 Dresden — [2]Forschungszentrum Karlsruhe GmbH, Institut für Synchrotronstrahlung, ANKA, Hermann-von-Helmholtz-Platz 1, 76344 Karlsruhe — [3]European Synchrotron Radiation Facility (ESRF), BP 220, F38043 Grenoble, France

Real time synchrotron Small-Angle and Wide-Angle X-ray Scattering was performed during the tensile deformation of a high-density polyethylene copolymer. The changes of the structure in the crystalline and in the amorphous domains were followed during the three characteristic stages of the load-displacement curves: The elastic stage and the plastic stage composed of the phase of the lowering load in the force-displacement-curve and the strain hardening. Competitive phenomena like crystallite fragmentation and cavitations were found to occur simultaneously in the lowering phase of the load but at different length scale. We prove that the volume fraction of the voids increase during the fibrillation of the lamellae stacks. At increasing strain the changes of the radius of gyration of the cavities in stretching as well as in transversal direction were discussed. During strain hardening no further increase in the volume fraction of the voids was found. However, we make evidence for the establishment of a new long spacing order of the micro-block crystallites, which were stable despite the occurrence of the melting process.

CPP 3.7 Mon 11:45 ZEU 114

Investigation of Swollen Polymer/Gas-Systems: Atomistic Packing Models Based on Experimental Properties — •Martin Böhning[1], Matthias Heuchel[2], Ole Hölck[1], and Martin R. Siegert[2] — [1]Bundesanstalt für Materialforschung und -prüfung (BAM), Unter den Eichen 87, 12205 Berlin, Germany — [2]GKSS Forschungszentrum, Institut für Polymerforschung, Kantstrasse 55, 14513 Teltow, Germany

Changes in glassy polymers induced by sorption of substantial amounts of small penetrant molecules can affect the amorphous packing structure as well as the molecular mobility of the polymer chains. These changes - manifested as swelling and plasticisation phenomena - can have radical influences on relevant properties, especially concerning the gas transport behaviour in membrane or barrier applications. Based on sorption and dilation measurements of CO2 in different glassy polymers (e.g. polysulfone and a polyimide) detailed atomistic packing models have been created using MM/MD-simulation techniques. The analysis of representative models of swollen and unswollen states allows a detailed characterisation of these changes in terms of free volume distribution and molecular mobility. Solubilities and diffusivities of penetrants in these models were calculated using MD and GCMC methods. Results are evaluated in relation to phenomenological models and compared to experimental findigs in order to prove the reliability and predictive abilities of this approach.

CPP 3.8 Mon 12:00 ZEU 114

Epoxy networks — •Marian Brandau[1], Michael Schulz[2], and Steffen Trimper[1] — [1]Fachbereich Physik, Martin-Luther-Universität, Friedemann-Bach-Platz, 06108 Halle — [2]Abteilung Theoretische Physik, Universität Ulm, 89069 Ulm

We present the results of a Monte Carlo simulation for the kinetic of formation in epoxy networks. In particular, two time regimes are distinguished, namely a short time regime characterized by a radicalic polymerisation and a long time regime where slow formation processes are relevant. Whereas the first stage is reaction controlled, diffusion processes determine the second one. On a more microscopic level two kinds of molecules with different functional groups are taken into account. While the so-called cross-linkers are characterized by three or four functional groups, the chain formers are bifunctional. The aim is to find out universal properties of the kinetic of formation such as the cluster size distribution function, and the behavior in the vicinity of the percolation threshold. Moreover, we consider the light-scattering function.

CPP 3.9 Mon 12:15 ZEU 114

A non-cubic network phase in ABC copolymers via e^--tomograpy — •Gerd E Schroeder[1], Stephen T Hyde[1], Hermis Iatrou[2], Nikos Hadjichristidis[2], Satoshi Akasaka[3], and Hirokazu Hasegawa[3] — [1]Applied Maths, Research School of Physical Sciences, Australian National University, 0200 Canberra, Australia — [2]Chemistry Department, University of Athens, Panepistimiopolis, Zografou 15771, Athens, Greece — [3]Department of Polymer Chemistry, Graduate School of Engineering, Kyoto University, Katsura, Nishikyo-ku, Kyoto 615-8510 Japan

We report the discovery of a novel bicontinuous tetragonal phase in the linear ABC triblock terpolymer system polystyrene, polyisoprene and polydimethylsilocane. The data is consistent with spacegroup Fddd and is distinctly non-cubic. The channel topology of the mesophase is distinct from the cubic Gyroid and Diamond mesophases. It consists in 2 identical intertwined labyrinths with 3- *and* 4-connected nodes. Our mesophase differs from an earlier report of a copolymer phase (also in a linear terpolymer system) with the same spacegroup by Bates *et al*, who deduced a single channel morphology, based on TEM and SAXS data [1]. Our proposal is based on 3D e^--tomography data. The channel geometry is identified via a *medial surface* (MS) algorithm. The MS is a generalised channel graph consisting in surface patches rather than line segments and is, in contrast to line graphs, a complete descriptor of both topology *and* geometry.

[1] T. Epps, E. Cochran, T. Bailey, R. Waletzko, C. Hardy, and F. Bates, Macromolecules **37**, 8325–8341 (2004)