Polymers under Multiple Constraints

Polymer- & Soft-Matter-Seminar

Tuesday, 15th November 2016

at: **5.15pm**

VDP4 1.27, Von-Danckelmann-Platz 4, 06120 Halle

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"Flocculation Dynamics and Reinforcement of Elastomer Nanocomposites with Conductive Fillers: A Combined Rheological and Dielectric Analysis"

The flocculation dynamics of carbon black filled elastomer melts is investigated by dielectric relaxation spectroscopy in the frequency range from 0.1 Hz up to 10 MHz during oszillatory shearing in a plate-plate rheometer [1]. It is demonstrated that during heat treatment at low strain amplitude, a pronounced flocculation of filler particles takes place leading to a successive increase of the shear modulus and dc-conductivity. Followed up by a strain sweep, the filler network breaks up and both quantities decrease simultaniously with increasing strain amplitude. Two relaxation times, obtained from a Cole-Cole fit of the dielectric spectra, are identified, which both decrease strongly with increasing flocculation time. This behaviour is analyzed in the frame of fractal network models, describing the effect of structural disorder of the conducting carbon black network on the diffusive charge transport. During flocculation, a universal scaling behaviour holds between the conductivity and the corresponding high frequency relaxation time, which fits all the measured data. The scaling exponent agrees fairly well with the prediction obtained from cluster-cluster aggregation process (CCA). It is demonstrated that the underlying basic mechanism is a change of the correlation length of the filler network, i.e. the size of the fractal heterogeneities. This decreases during flocculation due to the formation of additional conductive paths, making the system more homogeneous.

The same universal scaling behaviour, as obtained for flocculation, is found for temperature dependent dielectric measurements of the cured systems, which are heated from room temperature up to 200 °C. Thereby, the conductivity decreases significantly and the relaxation time increases, indicating that the filler network breaks up randomly due to the thermal expansion of the rubber matrix.

By referring to the structural analysis of filled elastomers, a micro-mechanical model of stress softening and filler-induced hysteresis (Dynamic Flocculation Model) has been developed [2-5]. It is based on a tube model of rubber elasticity together with a micro-mechanical model of stress induced filler cluster breakdown. The evaluation of stress softening is obtained via a pre-strain dependent hydrodynamic amplification of the rubber matrix by a fraction of rigid filler clusters with virgin filler-filler bonds. The filler-induced hysteresis is described by a cyclic breakdown and re-aggregation of the residual fraction of softer filler clusters with already broken, damaged filler-filler bonds. The model is found to be in fair agreement with experimental data obtained with carbon black and silica filled elastomers. For a finite element implementation, the concept of representative directions has been used [6, 7]. Several practical applications demonstrate that the model is well suited for predicting the stress- strain response of filler reinforced rubbers.

References:

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