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Entanglements and Elasticity in Polymer Networks[1]

MICHAEL RUBINSTEIN, University of North Carolina

We develop and solve a molecular model for nonlinear elasticity of entangled polymer networks, called non-affine slip-tube model. This model combines and generalizes several successful ideas introduced over the years in the field of rubber elasticity. Each chain passes through a sequence of slip-links. The topological constraints imposed by neighboring network chains on a given one are represented by the confining potential acting on the slip-links. This topological potential restricts fluctuations of the network chains to the non-affinely deformed confining tube and changes upon network deformation. Network chains are allowed to fluctuate and redistribute their length along the contour of their confining tubes. The dependence of the stress σ on the elongation λ is usually represented in the form of the Mooney stress $f^*(1/\lambda) = \sigma/(\lambda - 1/\lambda^2)$. We find a simple expression for the Mooney stress $f^*(1/\lambda) = G_c + G_e/(0.74\lambda + 0.61\lambda^{-1/2} - 0.35)$ where G_c and G_e are phantom and entangled network moduli. This allows analyzing the experimental data in the form of the universal plot and to obtain the two moduli G_c and G_e related to the densities of the cross-links and entanglements of the individual networks. The predictions of our new model are in good agreement with experimental data for uniaxially deformed polybutadiene, polydimethylsiloxane, and natural rubber networks. We generalize our non-affine slip-tube model to describe swelling of entangled networks in both good and theta solvents and find that non-affine effects due to entanglements decrease upon swelling. We also calculate the dependence of stress on strain in uniaxially deformed swollen networks.

[1]This work was done in collaboration with Sergey V. Panyukov.