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Unique Properties of Reversibly Associating Polymer Networks[1]

MITCHELL ANTHAMATTEN, University of Rochester

Reversibly associating functional groups offer the polymer physicist with a new tool to develop stimuli-responsive polymers. Our focus has been to attach reversibly associating groups onto rubbery network polymers. Free radical copolymerization was used to synthesize a series of crosslinked poly(n-butylacrylate)s containing quadruple H-bonding ureidopyrimidinone (UPy) side-groups. Resulting elastomeric networks contain both covalent and dynamic non-covalent crosslinks, and this unique architecture is shown to affect viscoelastic behavior and mass-transport properties. Shape-memory effects are studied quantitatively using thermomechanical techniques. Experiments show how reversible interactions, such as hydrogen bonding, are capable of stabilizing mechanically strained states. Unlike conventional shape-memory polymers, these dynamic networks lack a well-defined shape recovery temperature. Instead, their shape recovery rate depends on temperature. To further study the dynamics and temperature dependence of mechanical relaxation, isothermal creep experiments and dynamic mechanical analysis were performed. Creep data, acquired at several different temperatures, are fit to a simple viscoelastic model. Fit viscosities exhibit Arrhenius-like temperature dependence with activation energies of $\sim$90 kJ/mol, which is in rough agreement with H-bond dissociation barriers. Molecular transport through dynamic networks is studied using gravimetric sorption and dye-diffusion techniques. Diffusion depends on temperature, network architecture, solute size, and the interaction between the solute and the network. Membranes with high temperature-sensitive diffusion properties may be useful in applications such as transdermal drug delivery, microfluidics, or liquid chemical separation processes.

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