STRUCTURAL RECONSTITUTION IN POLYMER MATRIX COMPOSITES AND ITS SIGNIFICANCE FOR PERFORMANCE AND FUNCTIONALITY

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Using a combination of inelastic light scattering and atomic scale simulations, we investigated the structure and properties of thermoset polymeric materials, while reversibly removed from their thermodynamic ground state. In one set of experiments we measured the adiabatic elastic modulus of epoxy *in situ*, while undergoing polymerization and cross-linking reactions. Throughout the experiment, the degree of cure was monitored using Raman light scattering (RLS), while the adiabatic modulus was measured using Brillouin light scattering (BLS). Since these are both inelastic light scattering techniques, RLS and BLS are carried out concurrently, i.e., the respective spectra were collected from the same location in the specimen and at the same time, allowing us to directly compare the modulus with the degree. As illustrate in Fig. 1, we find that the adiabatic modulus of the polymer evolves non-uniquely as a function of cure degree; it strongly depends on the cure rate.[1] We can explain the observed behavior by taking into account that two mechanisms contribute to the increase in the elastic modulus of the material during curing. First, there is the formation of covalent bonds in the network during the curing process. While the formation of these bonds lowers the electronic structure energy of the system, atoms are still somewhat removed from their ground state positions. Hence, in a second process the structure undergoes structural reconstitution toward a more optimally packed configuration of the network, which enhances non-bonding interactions. Both contributions are apparent in the adiabatic modulus derived from Brillouin scattering, as it reflects the elastic response of the polymer network in thermodynamic equilibrium. To further ascertain the notion of structural reconstitution, which is affecting only non-bonding interactions, we subject a fully cured epoxy to various degrees of strain, well within the elastic regime, using a miniature tensile tester mounted in the beam path of the light scattering setup, while simultaneously measuring the adiabatic and isothermal elastic moduli as a function of the applied strain. We find that straining this polymer network is equivalent to forming the bonds between its atoms while these are in locations displaced from the structural ground state. The fact that the polymer structure can reconstitute after undergoing appreciable deformation opens new ways of thinking about its applicability in terms of functionality and performance. Molecular simulations provide additional insights into the underlying mechanisms, and delineate the blueprints for materials design.[2]

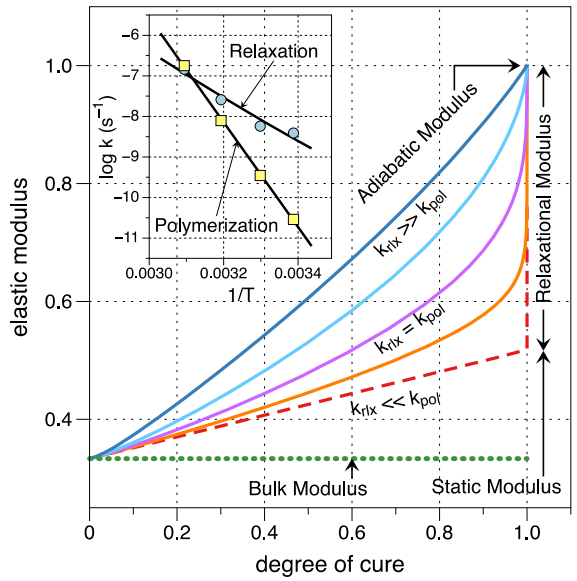

Figure 1 – Schematic of the longitudinal elastic modulus as a function of the degree of cure

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