MOLECULAR DYNAMICS SIMULATION OF THE DYNAMIC MECHANICAL RESPONSE OF PHOTOPOLYMERIZED METHACRYLATE NETWORKS[(I)]

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[(I)]The computational results presented have been achieved using the Vienna Scientific Cluster (VSC)

Dynamic mechanical analysis (DMA) is one of the most widely used characterization methods in polymer science, as the information obtained can be related to numerous parameters of the polymer that is tested. But proving such relations is a difficult task, for the reason that many important properties are not directly accessible experimentally. One way to study and obtain additional information on a molecular level is molecular dynamics simulations.

This work deals with the simulation of the formation of polymer networks and subsequent characterization using forced oscillation. Important for the quality of the measurement is a realistic network structure. To obtain a model of a polymer network, that is as close as possible to a real network, the simulated polymerization algorithm 'Polymatic' [1] was used. With this method a final double bond conversion of 95.25 % was calculated for TEGDMA reacted at room temperature. This network was then analyzed using a simulated DMA measurement. The low signal to noise ratio coming from thermal fluctuations was overcome by averaging adjacent data points and then fitting the known response function, similar to [2]. By repeating this measurement over a range of temperatures, it is possible to determine the temperature dependence of the storage modulus. It was found that when increasing the double bond conversion, the curve was shifted to higher modulus values. This is because reacted TEGDMA increases the crosslink density of the polymer network. The comparison with a real measurement showed a tilt and a shift towards higher temperatures. This is a result of the high oscillation frequency that is necessary because of computational limitations.

In upcoming simulations, the presented method will be applied to tackle fundamental questions concerning the influence of crosslink density, mono/multi-functional monomers/oligomers, intermolecular forces, on the thermo-mechanical properties of photocured polymers.

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[1] Theor Chem Acc 132, 1334 (2013)

[2] Phys. Chem. Chem. Phys., 2015,17, 7196-7207