

A molecular simulation approach of epoxy resins: from curing to deformation and fracture

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Thermoset polymers are the first choice for a matrix material used in carbon and glass fibre reinforced plastics in high-performance applications. The simple manufacturing process, combined with a relatively low price and adaptive design possibilities, represents the major advantage over thermoplastic matrices. However, already the curing of thermosets is a complex and non-equilibrium process, which eventually explains why a comprehensive understanding of the interplay of the basic linking processes and the collective organization of a complex polymer network is still lacking. However, this is a prerequisite for rationalizing the material reaction and macroscopic properties of thermosetting polymers. Previously, it was shown that a hybrid quantum mechanical and molecular mechanical approach to cross-link a thermoset polymer is capable of reproducing experimentally determined mechanical and thermodynamic properties of an equivalent system [2]. To cross-link the thermoset, a new method was developed, which we call “smooth topology transfer”. This method allows a smooth transition from one molecular topology to another by mixing forces and energies in molecular dynamics simulations, achieving as a top-score a degree of cross-linking of 93%. Reactive attempts are further controlled by Monte Carlo selection criteria—which also take quantum mechanical corrections into account—allowing at the same time a polymer network with a minimum of residual stress. This is in-line with mechanical properties found in the literature and which are otherwise often overestimated by other cross-linking approaches. A good agreement between cross-linking degree and heat of formations at different curing temperatures was additionally found when comparing the results of the simulation with results from a Differential Scanning Calorimetry (DSC) of a commercially available similar thermosetting polymer. Only recently has it become known that thermoset polymers can undergo significant plastic deformation at certain conditions [1]. In an attempt to explain this fundamentally, based on the previously developed models for cured thermosets and a newly developed reactive force field based on a coarse-grained approach, we were able to show for the first time that plastic deformation is associated with a fairly large bond reorganization before material fractures actually occur.

References

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