Evolution of Entanglements During Crazing of Glassy Polymers

TING GE, MARK O. ROBBINS, Johns Hopkins University, ROBERT HOY, Yale University, STEFANOS ANOGIANNAKIS, CHRISTOS TZOUMANEKAS, DOROS THEODOROU, National Technical University of Athens — Craze formation increases the fracture energy of glassy polymers by orders of magnitude. The polymer volume expands by an extension ratio which is assumed to be determined by the entanglement network. We test this assumption with molecular simulations that use the Contour Reduction Topological Analysis (CReTA) algorithm to follow topological constraints (TCs) associated with the entanglement network. The TCs are identified with contacts between chains after applying CReTA. Within systematic errors, crazing does not change the number of TCs or the distribution of chemical distances between them. Moreover, about 75% of the contacts remain between the same chains at nearly the same location. The 25% of contacts that change do not reflect a comparable loss of entanglements. Instead, small displacements within the tube change which chains contact after CReTA. This interpretation is tested by adding fixed crosslinks to a sparse entanglement network and crazing preoriented samples.

This material is based upon work supported by NSF Grant DMR 108474.

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Date submitted: 19 Nov 2010
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