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Self-Healing of Polymer Networks with Reversible Bonds¹

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Self-healing polymeric materials are systems that after damage can revert to their original state with full or partial recovery of mechanical strength. Using scaling theory we study a simple model of autonomic self-healing of polymer networks. In this model one of the two end monomers of each polymer chain is fixed in space mimicking dangling chains attachment to a polymer network, while the sticky monomer at the other end of each chain can form pairwise reversible bond with the sticky end of another chain. We study the reaction kinetics of reversible bonds in this simple model and analyze the different stages in the self-repair process. The formation of bridges and the recovery of the material strength across the fractured interface during the healing period occur appreciably faster after shorter waiting time, during which the fractured surfaces are kept apart. We observe the slowest formation of bridges for self-adhesion after bringing into contact two bare surfaces with equilibrium (very low) density of open stickers in comparison with self-healing. The primary role of anomalous diffusion in material self-repair for short waiting times is established, while at long waiting times the recovery of bonds across fractured interface is due to hopping diffusion of stickers between different bonded partners. Acceleration in bridge formation for self-healing compared to self-adhesion is due to excess nonequilibrium concentration of open stickers. Full recovery of reversible bonds across fractured interface (formation of bridges) occurs after appreciably longer time than the equilibration time of the concentration of reversible bonds in the bulk. The model is extended to describe enhanced toughness of dual networks with both permanent and reversible cross-links.

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