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Recyclable crosslinked polymer networks with full property recovery made via one-step controlled radical polymerization KAILONG JIN, LINGQIAO LI, JOHN TORKELSON, Northwestern University — Rubber tires illustrate well the issues ranging from economic loss to environmental problems and sustainability issues that arise with spent, covalently crosslinked polymers. A nitroxide-mediated polymerization (NMP) strategy has been developed (see Adv. Mater. 2016, 28. 6746) that allows for one-step synthesis of recyclable crosslinked polymers from monomers or polymers that contain carbon-carbon double bonds amenable to radical polymerization. Resulting materials possess dynamic alkoxyamine crosslinks that undergo reversible decrosslinking as a function of temperature. Using polybutadiene as starting material, and styrene, an appropriate nitroxide molecule and bifunctional initiator for initial crosslinking, a model for tire rubber can be produced by reaction at temperatures comparable to those employed in tire molding. Upon cooling, the crosslinks are made permanent due to the extraordinarily strong temperature dependence of the reversible nitroxide capping and uncapping reaction. Based on thermomechanical property characterization, when the original crosslinked model rubber is chopped into bits and remolded in the melt state, a well-consolidated material is obtained which exhibits full recovery of properties reflecting crosslink density after multiple recycling steps.

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