

## Dynamic bottlebrush polymer networks: self-healing in supersoft networks

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Dynamic polymer networks are of research interest for various applications where self-healing and reprocessability are advantageous. We describe a design strategy for expanding the scope of mechanical properties of these materials by selective crosslinking of bottlebrush polymers. Well-defined bottlebrush polymers with poly(4-methylcaprolactone) side chains were cured into networks via reaction with a bis-lactone crosslinker catalyzed by a strong Lewis Acid (tin ethylhexanoate). At elevated temperatures (160 to 180 °C), residual hydroxy groups undergo transesterification reactions with sidechain esters as evidenced by stress relaxation experiments. The crosslinking density can be varied by both changing the stoichiometry of crosslinker as well as the backbone degree of polymerization, resulting in supersoft and tunable shear moduli ca. 10 – 100 kPa. The ability of these materials to self-heal was demonstrated by the re-molding of a tensile bar after rupture, with the sample recovering >85% of its original toughness over 2 cycles. In summary, design of the molecular architecture of dynamic polymer networks creates new opportunities to tailor the mechanical properties of CANs in ways that are otherwise difficult to achieve.

