MAKING STRONG AND TOUGH POLYMERS WITH LIGHT

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Advances in polymer chemistry over the preceding decades has resulted in the production of materials with a wide-range of properties - from super soft gels to high modulus glasses - through the control of chemical composition, molecular weight, and architecture. Among the most ubiquitous and industrially sought-after systems are semi-crystalline polymers, where self-assembly of polymer chains within crystalline lamellae endow the polymers with high tensile strength, toughness, and chemical resistance. However, despite the advantages that could be afforded in these materials when coupled with the spatiotemporal control provided by photopolymerizations, it remains an outstanding challenge to synthesize semi-crystalline polymers via photochemical methods. Recently, we developed a method for synthesizing high molecular weight semi-crystalline polymers using thiol-ene chemistry with Young's moduli and extensibilities of up to 280 MPa and 800%, respectively. Furthermore, these properties can be tuned through the combination of commercially-available dithiol thiol and diene monomers. Additionally, we show that semi-crystallinity is preserved upon the addition of small amounts of cross-linker for networks, and that these thermosets are amenable to digital light processing (DLP)-based additive manufacturing, presenting opportunities for a variety of applications requiring specified parts with excellent mechanical properties. Building on these results, current efforts in the group have explored ways to combine dynamic covalent chemistry within these materials to impart reconfigurably and recyclability to these high performance materials. We show that the outstanding mechanical properties of these materials are preserved upon the addition of dynamic thioester moieties for the realization of semi-crystalline covalent adaptable networks (CANs). Unlike their static counterparts, thiol-thioester exchange within these materials facilitates reconfigurability and recyclability that can be tuned through the network stoichiometry and the choice of nucleophilic catalyst. This work demonstrates a new avenue within the photopolymerization space towards functional, high performance polymers through simple chemistry.