Bio-based multi-responsive shape memory polymers using natural oils and cellulose nanocrystals

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Abstract: Shape Memory Polymers (SMPs) are a unique class of smart materials with the ability to deform and reform shapes under certain stimuli. These materials switch between an entropically-disfavored temporary shape and a permanent shape after introduction to the stimuli. This switch is based on a series of transitions, such as glass transition temperatures or melting points. So far, most of these materials have been prepared mainly using petroleum based polymers. We have developed a bio-based approach utilizing surface-initiated ATRP (atom transfer radical polymerization), via a combination of cellulose nanocrystals (CNCs) and soybean oil based polymers, which are crosslinked using TAD chemistry or epoxy resins, to create robust, sustainable, and potentially scalable SMPs. The resulting heterogeneous polymers and films can be easily characterized using XPS, DLS, DSC, DMA, and NMR to confirm chemical compositions and mechanical properties. The shape memory properties, resulting from both dynamic hydrogen bonding of secondary amide groups closely associated with Tg and permanent chemical crosslinking, are responsive to a variety of stimuli including temperature, water, and organic solvents with excellent shape fixity and recovery. Overall, these mechanically robust polymers show promise as biomass based SMPs to complement/replace current petroleum based counterparts.