Main-Chain Liquid-Crystalline Elastomers Using a Two-Stage Thiol-Acrylate Reaction

Abstract
Liquid-crystalline elastomers (LCEs) are a class of stimuli-responsive and active materials known for their unique mechanical and optical properties. These materials can demonstrate a large magnitude (~100%) of thermally-reversible shape change and actuation due to the coupling of liquid-crystalline order and network elasticity. As a result, LCEs have been proposed for a myriad of sensor and actuator applications, such as artificial muscles and tunable iris lenses. While promising materials, the practical application, scalability, and accessibility of main-chain LCEs remain a limiting barrier in the widespread use of LCEs. This presentation will introduce a novel two-stage thiol-acrylate Michael addition-photopolymerization reaction for scalable synthesis of main-chain LCEs and a programmable single-liquid crystal monodomain. First, this two-stage reaction platform will be used to investigate the effects of crosslinker functionality and concentration for unoriented polydomain LCE samples. Tri-functional crosslinkers demonstrated a lower isotropic transition temperature (Ti) compared to their tetra-functional counterparts, while the crosslinker concentration did not show an apparent influence on Ti. Conversely, crosslinker concentration was shown to be the primary factor on the glass transition behavior of the elastomer networks as well as achievable thermal actuation under a bias force. Second, the second-stage photopolymerization reaction will be used to program a permanently oriented monodomain. The influence of programming temperature and crosslinker concentration on failure strain were investigated to optimize network alignment and order parameter in the samples. It was shown that programming should be performed near the peak of tan δ to maximize failure strain and that thermal actuation increased linearly with increased applied programming strain. The shape-fixity of the second stage reaction was also measured and was consistently above 90% for all programming conditions up to 400% strain. Lastly, this presentation will cover how the spatio-temporal control of this reaction can be used to create highly mechanically anisotropic and shape-changing materials for previously unexplored biomedical applications.