Designing functional performance in liquid crystalline elastomers through correlation of fundamental material properties

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Liquid crystalline elastomers (LCEs) are functional materials capable of undergoing large deformations in response to stimuli such as heat or light. The complex shape changes exhibited by LCEs are founded in the ability to direct long-range ordering of liquid crystalline moieties within a polymer network. Upon application of stimulus, the liquid crystalline order is disrupted, causing the polymer network to contract along the initially aligned axis. This intrinsic muscle-like response of these smart materials makes them desirable for use in application such as soft robotics and biomimetic actuation. In this work, fundamental properties of liquid crystalline elastomers are first explored through a lens of materials chemistry and physics. Correlations are drawn between molecular structure, polymer network topology, and stimuli-responsive behavior in order to tune the thermomechanical and photomechanical actuation response of LCEs. Then, moving beyond programming actuation to *re*programming of LCEs, dynamic covalent chemistry is incorporated in photoresponsive LCEs such that dynamic chemistry and photo-actuation are each activated in distinct steps using orthogonal stimuli. These materials are used to demonstrate reprogrammable deformation and retention of actuated states as an example of how fundamental structure-property correlations can be used as tools for designing targeted actuation in LCEs.