Abstract

The combination of liquid crystals (LCs) with polymer networks results in fascinating materials in which the elasticity of the polymer network is coupled to LC order, and they hold promise for a variety of applications which depend on an optical or a mechanical response to external stimuli. Unfortunately, the methods for preparing LC networks are limited. In this work, we present two types of LC gels with novel network architectures: a physical gel made by polymer self-assembly in an LC solvent and a chemical gel made by crosslinking telechelic side-group liquid crystalline polymers (SGLCPs). The physical gel thermoreversibly transitions to a liquid on heating, and it can be uniformly aligned using electric, magnetic, or flow fields. The gel exhibits a fast electro-optical response, making it potentially viable for display applications. The physical gel also reveals novel equilibrium and dynamic properties that arise due the coupling between LC order and the polymer network. When confined between cell substrates prohibiting macroscopic shape change, it exhibits a temperature-sensitive striped texture transition. We provide a theoretical model that captures the striped texture by accounting for the compromise between nematic rubber elasticity and LC elasticity. Dynamic light scattering studies reveal two distinct director relaxation modes, in contrast to covalent gels, which show a single relaxation mode. We argue that the slower dynamics arise from the coupling between the director and the physically associated network which restructures on long time scales. We also investigate the self-assembled micellar structure of the gel via neutron scattering and rheometry studies, and these studies reveal that the gels' structure changes dramatically across the nematic-to-isotropic phase transition. Finally, we investigate model LC networks made by crosslinking telechelic SGLCPs using "click" chemistry. These chemical networks can be swollen in a small-molecule LC, and they exhibit a low-threshold electro-optic response and a temperature dependent size. These novel physical and chemical LC gels provide insight into the role of network structure in determining the material properties of liquid crystalline elastomers and gels.