Lamellar Bilayers as Reversible Sacrificial Bonds to Toughen Hydrogel

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A novel anisotropic hydrogel with a unidirectionally aligned membrane-like lamellar bilayer structure of macroscopic size is synthesized in a one-pot polymerization reaction from a polymerizable surfactant (dodecylglyceryl itaconate:DGI) and acrylamide in presence of a crosslinker. Prior to the polymerization, by applying shear flow to the precursor solution, thousands of lamellar bilayers of self-assembled DGI are aligned in one direction parallel to the substrate surface. Polymerized lamellar bilayers are stacked periodically and entrapped in the polyacrylamide network. This hydrogel shows one-dimensional swelling, strong anisotropy in the elastic modulus, and beautiful structural color. Owing to the softness and large deformability of the gel, the color of the gel can be reversibly tuned by the compressive strain over a wide wavelength range, which might be used as a soft tactile sensor, that is able to detect a complicated force field, and deformation based color display.

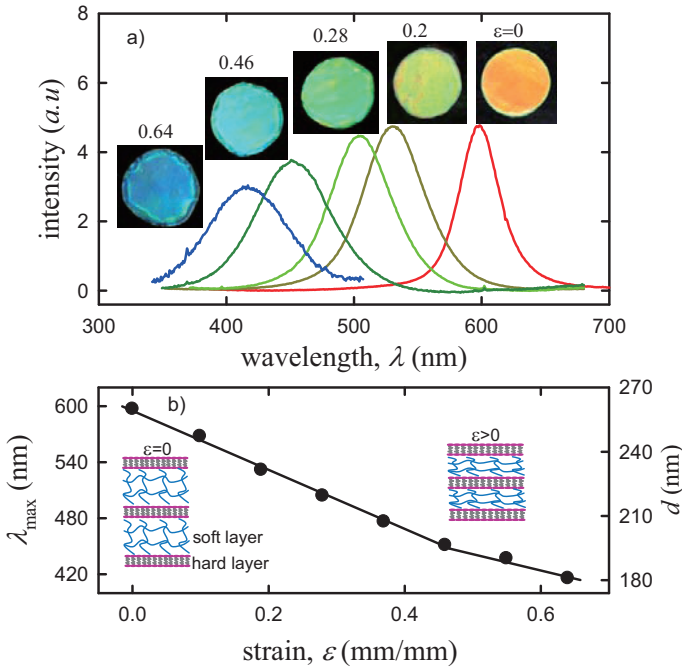

Figure 1. a) Shift of the gel color from red to blue upon gradual increasing of compressive strain (right to left) perpendicular to the lamellar layer and their corresponding reflection spectrum. b) Compressive strain dependence of the wavelength maximum (λmax) and lamellar distance (d).

The stratified lamellar bilayers not only diffract light to exhibit magnificent structural color, but also serve as a reversible sacrificial bond that dissociate upon deformation and give rise to the excellent mechanical functions such as high toughness, self-recovery, and persisting fatigue resistance. Both the molecular dissociation and lipid-like mobile nature of bilayers dramatically enhance the resistance to crack propagation by suppressing the stress concentration at the crack tip with the formation of extra-ordinary blunting. The unique toughening phenomena might allow deep insight into the toughening mechanism of the hydrogel-like soft materials such as biological soft tissues.

Reference

Md. A. Haque, G. Kamita, T. Kurokawa, K. Tsujii, J. P. Gong, Advanced Materials, 22(45), 5110-5114(2010).