

## Failure of Composite Hydrogels Templated via Colloidal Self-Assembly

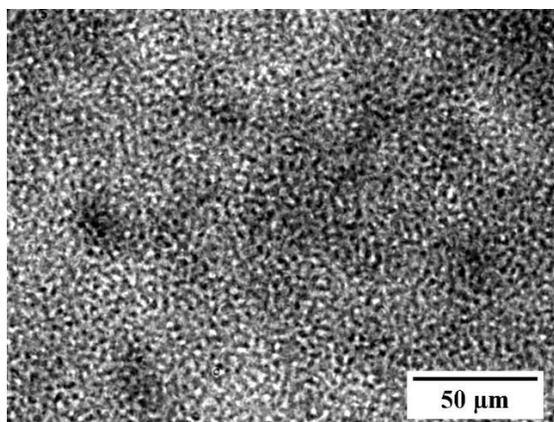
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The exploitation of heterogeneous microstructure to tune mechanical response is abundant in nature. The diversity of natural systems that control mechanics through microstructure (e.g. mussel adhesive plaques,<sup>1</sup> brain tissue,<sup>2</sup> trabecular bone<sup>3</sup>...etc.) highlights the potential of leveraging both local properties and geometry to influence material behavior. However, translating this strategy of tailoring mechanical response through complex microstructure to soft synthetic materials has been challenging. One challenge in this area has been developing systematic processing methods for creating multiphase materials at different size scales (0.1-10  $\mu\text{m}$ ).<sup>4</sup> This challenge is addressed here by employing thermoresponsive nanoemulsions to craft hydrogels interpenetrated with an assembled network of silicone oil nanodroplets (radius~25 nm) tethered together by poly(ethylene glycol) diacrylate chains.<sup>4</sup> Assembly of these colloidal networks occurs when increasing temperature causes the acrylate endgroups to go from being readily solvated by water to partitioning to the oil/water interface. This process creates assembled droplet networks that can be formed at different size scales by tuning the thermal aging time and temperature. Stabilization of these networks by photocrosslinking results in composite hydrogels with microstructures consisting of phases that are droplet-rich and droplet-poor as shown in the optical micrograph. The elastic and fracture properties of these composite hydrogels are characterized as the size of the droplet network transitions across the elastofracture length (ratio of the fracture energy to the elastic modulus) of the aqueous phase, which was measured to be 6.5  $\mu\text{m}$  via notch tests. This study deepens our understanding of the interplay between local composition and geometry to enable the design of next generation strong, tough, and resilient soft materials through the incorporation of secondary phases at controlled size scales.



Optical micrograph showing the microstructure of a composite hydrogel interpenetrated with an assembled droplet-rich hydrophobic phase.

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### References:

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