THE COLLOIDAL MICROSTRUCTURE OF MECHANICALLY REINFORCED ORGANOHYDROGELS

Lilian Hsiao, Chemical & Biomolecular Engineering, North Carolina State University
lilian_hsiao@ncsu.edu

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There has been a surge of interest in composite hydrogel materials that can be spun into fiber form, because of advantages associated with increased moisture holding, mechanical enhancement, and biocompatibility. Nevertheless, a major challenge is that it is difficult to generate fibers with functional microstructure without expensive post-processing steps. We introduce a colloidal gel system comprised of nanoemulsion droplets of poly(dimethylsiloxane) (PDMS) suspended in a continuous phase comprised of a double network hydrogel precursor that contains sodium alginate and poly(ethylene glycol diacrylate) (PEGDA). The nanoemulsions undergo self-assembly at elevated temperatures to form hydrophobic networks with different length scales, which can be crosslinked by UV light due to its hydrogel phase. We find that the length scales of the colloidal nanoemulsions are tuned independently using the applied shear rate and temperature in a cylindrical spinneret, generating solid organohydrogel fibers with different microstructures. Furthermore, we show that it is possible to introduce a double network hydrogel precursor that is compatible with the self-assembling nanoemulsions, thus generating fibers with enhanced Young's modulus, fracture energy, and rupture stress. This study utilizes state-of-the-art understanding of colloidal gel rheology and hydrogel chemistry to generate new types of mechanically reinforced materials that can also be used to control drug loading and release.