Spontaneous patterning of gels by confining polymer mixture to microspheres

Miho Yanagisawa
Department of Physics, Kyushu University

Polymer gels show complex surface patterns upon volume phase transition in response to external stimuli. Here we report a spontaneous patterning of gels in binary polymer solution by confining to soft microspheres: water-in-oil (W/O) microdroplets coated with a lipid layer. We encapsulated gelatin/PEG (polyethylene glycol) solution in droplets, where liquid-liquid (L-L) phase separation proceeded and then gelation of gelatin trapped the coarsening process with decreasing temperature. Deep quench generated smaller domains of gels with various shapes like network, sponge, etc. Whereas, shallow quench completed L-L phase separation before the gelation, and brought a large mono-domain in gel wetting to the droplet surface. Below the gelation temperature, smaller droplets than a critical size deformed their shapes to minimize the interfacial area between gelatin/PEG phases. The shapes of gels were determined by a force balance among bending elasticity of gel, interfacial tensions between gelatin/PEG and W/O phases. We found that the gelation increased the interfacial tension between gelatin/PEG phases. Therefore, gelation resulted in transformation of smaller droplets with a larger area-to-volume ratio to minimize the gelatin/PEG interface area against the costs of bending energy of gel and interfacial energy at W/O surface. Furthermore, the spontaneous patterns of gels were varied by changing polymer compositions and lipid species coating the droplet surface. These results demonstrate the coupling between phase separation and gelatin of polymers in droplets generates a great variety of microgel patterns.