

# Microstructure and Rheology of Phase-separated O/W/W Emulsions

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The microstructure and elastic modulus ( $G'$ ) of phase-separated gelatin (6 wt%) + hydroxypropyl starch (6 wt%) gels containing olive oil (0-16 wt%) dispersed as droplets in either the starch or gelatin phase were investigated. Polysorbate 20 (1 wt% of oil phase) was used to modulate the location of the oil droplets in the gels. Gelatin-starch gels showed a phase-separated microstructure composed of gelatin-rich domains with dispersed dissimilar starch inclusions. However, depending on the initial location (gelatin or starch phase) and wt% of the added oil above a critical concentration as well as the presence of polysorbate 20, the phase separation pathway was altered and evolved from nucleation and growth towards spinodal decomposition. For example, the presence of polysorbate 20 in the olive oil and subsequent addition to the gelatin phase resulted in previously-discrete starch inclusions now being interconnected leading to a bicontinuous gelatin-starch network. The presence of added oil increased the  $G'$  of all gels, with the greatest changes seen with 16 wt% added oil. Notably, with 16 wt% oil and polysorbate 20 added to the starch phase, gel  $G'$  increased ~57 % compared to the native phase-separated gelatin-starch gel. There was an ~80% increase in  $G'$  when 16 wt% oil + polysorbate 20 was added to gelatin phase whereas with no added emulsifier,  $G'$  rose by ~210 %. This strongly indicated that the oil acted as an active filler within the gel matrix. As each microstructure imparted its own rheological properties to the resulting gel, these observations demonstrated the possibility of creating a diverse group of phase-separated gel microstructures with a used-defined  $G'$ , depending on the concentration and location of the oil phase within the gel.