Interpenetrating polymer networks (IPNs) are cross-linked polymeric materials consisting of two distinct networks overlapping in the same bulk volume without chemical connection. Physical entanglement between networks reinforces mechanical properties, and macroscopic miscibility can be enhanced. Reported IPN hydrogels in literature are either not injectable or require in situ polymerization (1), which can be problematic for in vivo applications due to reaction toxicity, healing, and/or UV exposure (2). Alternatively, hydrogels can be produced by reactive mixing of functional polymers. Using two orthogonal covalent cross-linking schemes, such as the hydrazone and thiosuccinimide reactions (3), an injectable IPN hydrogel can be produced (4).

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The PNIPAM single hydrogel model fits to a squared Lorentzian scattering model, indicating static domains of higher chain density in the network. The semi-IPN model, showing the entangled PVP network does not suppress PNIPAM domain formation.

The thiosuccinimide IPN does not fit well to either model, instead requiring a hybrid model summing the network terms from each with a common term for average chain solution, suggesting both static domains and transient clusters are present.

Hydrogel disks were flash-frozen in liquid nitrogen, cut, lyophilized, and gold-sputtered for SEM imaging. Pore sizes in the semi-IPN and IPN are intermediate between the large pores of PNIPAM and smaller pores of PVP. Additionally, the IPN pores tend to be more rounded than those of the semi-IPNs, which are more flattened. The samples’ distinctive responses to freeze-drying indirectly indicate the existence of structural differences between networks in the hydrated state.

Conclusions

- Demonstrated ability to make IPN hydrogels by kinetically orthogonal reactive mixing
- Resulting hydrogels have distinctive micro- and nano-scale structures, significant mechanical reinforcement, and modulated swelling and degradation behaviour

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