

Title: Microstructure, swelling and deformation behavior of methacrylate hydrogels with interpenetrating network structure

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Abstract: This work is devoted to interpenetrating polymer network (IPN) hydrogels prepared by sequential processes of redox-initiated radical polymerization of the first network prepared from 2-hydroxyethyl methacrylate (HEMA), and UV-initiated radical polymerization of the second network prepared from 2-hydroxyethyl methacrylate (HEMA) or glycerol methacrylate (GMA). Microstructure, swelling and deformation responses of the IPN hydrogels and their constituent network hydrogels were tested by various techniques. The microstructure of the first poly(HEMA) network was found sensitive to polymerization conditions. A novel route for one-step synthesis of double-porous poly(HEMA) cryogel was proposed. The formation of the second poly(GMA) network in the environment of non-porous and macroporous poly(HEMA) parent networks was quantitatively studied using the ATR FTIR, UV-spectrophotometer, sol-extraction, and rheology techniques. The scattering of irradiation generated by the microstructure of parent network considerably enhanced the polymerization rate. This acceleration effect was quantified by careful optical analysis. The gelation point of the second poly(GMA) network in the environment of the parent network was detected rheologically and formation of crosslinked network structure within the IPN was confirmed. The development of inhomogeneities during the network formation studied by SWAXS was ascribed to intra- and intermolecular associations resulting in dense hydrophobic domains. DLS analysis of reaction mixtures showed the organization of HEMA or GMA molecules into nanosized hydrophobic domains when diluted with water already prior to polymerization. The poly(HEMA) parent network in IPNs exhibited surprisingly high swelling capacity when incorporated into the IPN with more hydrophilic poly(GMA). The combination of macroporous poly(HEMA) as the first network, which by itself exerts very low modulus, with poly(GMA) or poly(HEMA) as the second network offered interpenetrating polymer networks (IPNs) of superior mechanical properties compared to the properties of their constituents. The rubber elasticity theory was adapted to swollen non-porous homogeneous IPN hydrogels.

Keywords: hydrogel, interpenetrating polymer networks, *in-situ* formation, microstructure, swelling, mechanical properties