

Title SEMI-INTERPENETRATING POLYMER NETWORK
HYDROGELS BETWEEN POLYDIMETHYLSILOXANE/
POLY(ETHYLENE GLYCOL) AND CHITOSAN/
CARBOXYMETHYLCHITOSAN

Author Nantharak Rodkate

Advisor Assistant Professor Metha Rutnakornpituk, Ph.D.

Co - Advisor Uthai Wichai, Ph.D.
Chor. Wayakron Phetphaisit, Ph.D.

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ABSTRACT

Preparation and properties of semi-interpenetrating polymer networks (semi-IPNs) based on polydimethylsiloxane (PDMS)/poly(ethylene glycol) (PEG)-modified chitosan/carboxymethylchitosan (CMC) were studied. 1-20 wt% of both PDMS and PEG was interpenetrated into chitosan/CMC hydrogels having 10 wt% of hexamethylene-1,6-di-(aminocarboxysulfonate) (HDA) as a water-soluble crosslinker. PDMS was synthesized by acid-catalyzed ring-opening polymerization of octamethylcyclotetrasiloxane (D₄) in the presence of 1, 1, 3, 3-tetramethyldisiloxane endcapping agent and followed with hydrosilylation of the PDMS prepolymer with allyl alcohol to obtain hydroxyl-terminated PDMS. Chemical structure and functional groups of PDMS were elucidated by using ¹H NMR and FT-IR techniques. Two different molecular weights of PDMS (2K and 8K) and PEG (2K and 8K) were used in these current studies to investigate the effect of their chain lengths on percent crosslinking, water swelling behavior, surface morphology, tensile properties, water contact angle and water vapor permeability.

Considering first in the case of PDMS/PEG-modified chitosan, as compared to the unmodified one, percent crosslinking of the PDMS/PEG-modified chitosan dropped from 99% to 42-74% depending on molecular weight and percent of PDMS or PEG added. Further increasing percent of PDMS/PEG in chitosan even further

enhanced their equilibrium water content (% EWC). The modified chitosan having lower molecular weight PDMS/PEG showed higher % EWC as compared to the higher molecular weight ones. Scanning electron microscopy (SEM) indicated that increasing molecular weight or percent of PDMS/PEG in chitosan further enhanced microphase separation. Interpenetrating chitosan with HDA significantly improved phase homogeneity of PDMS/PEG-modified chitosan. According to differential scanning calorimetry (DSC), no phase transition was observed in PDMS-modified chitosan, while a slight shift of melting temperature of PEG phase was observed in PEG-modified chitosan. According to thermogravimetric analysis (TGA), a slight improvement toward thermal degradation was found in the PDMS/PEG-modified chitosan. It was also found that incorporation of PDMS/PEG into chitosan deteriorated tensile properties of the hydrogels.

Turning now to the case of PDMS/PEG-modified CMC, percent crosslinking of PDMS-modified CMC slightly decreased as high concentration of PDMS was incorporated. However, addition of PEG did not show an obvious impact on percent crosslinking of the modified CMC. % EWC of CMC was decreased upon addition of small amount of PDMS/PEG as compared to unmodified CMC. The values gradually increased as increasing PDMS/PEG concentrations in the hydrogels. Similarly to the cases of the modified chitosan, SEM and DSC results manifested that incorporation of HDA crosslinker significantly enhanced phase homogeneity of PDMS/PEG CMC without phase transition observed. Unmodified CMC and PDMS/PEG-modified CMC did not show a significant difference in TGA thermograms. Incorporation of PDMS/PEG into CMC significantly improved tensile strength but suppressed percent elongation of CMC hydrogels.