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Interpenetrating Amphiphilic Polymer Networks of Poly(2-hydroxyethyl methacrylate) and Poly(ethylene oxide)

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Interpenetrating polymer networks of poly(2-hydroxyethyl methacrylate) and poly(ethylene oxide) were synthesized by the free radical copolymerization of 2-hydroxyethyl methacrylate and ethylene glycol dimethacrylate in aqueous solutions of poly(ethylene oxide) diglycidyl ether (PEODGE), followed by amine−epoxy coupling reactions within the resultant gel network. The effects of ethylenediamine and PEODGE concentrations and coupling methods were examined and are discussed in this paper. Swelling studies and cryogenic-SEM analysis showed that these novel networks have high water intakes (equilibrium water content >3) and pore sizes on the order of up to 1 μm.

Introduction

Visually clear 2-hydroxyethyl methacrylate (HEMA) hydrogels are usually prepared by the free radical copolymerization of HEMA and a cross-linking agent at low dilutions (<40−45%[1,2]) of water. These gels have been largely employed in biomedical applications[3,4] and as separation or adsorption matrixes for various metal ions;[5,6] however, the wider usage of such gels in aqueous media is restricted because of their limited water intake.[1,7] Furthermore, the pore sizes (0.5−5 nm[8,9]) of these gels are severely limited by their high polymer contents, which lead to increasing interpenetration of polymer chains during network formation.[10]

To improve the water-sorption characteristics of these gels, monomers that are more hydrophilic than HEMA—such as poly(ethylene glycol) methacrylate,[11] vinylpyrrolidone,[12] and various ionic or zwitterionic monomers[13,14] can be used to partly replace HEMA in the reaction mixture. Although the swelling characteristics of these copolymers are generally improved, the preparations of such gels are complicated by factors which include the different relative reactivities of the monomers.[15−17]

An alternative approach to modify the swelling behavior of these networks is to prepare interpenetrating polymer networks (IPNs) of poly(2-hydroxyethyl methacrylate) (PHEMA) and polymers that are more hydrophilic than PHEMA.[18] Semi-I IPNs of PHEMA have been prepared in the presence of hydrophilic polymers such as poly(ethylene glycol),[19] poly(ethylene glycol) dimethyl ether,[20] and poly(vinylpyrrolidone).[21] These networks have improved swelling properties but limited uses in aqueous medium—because, when the networks are placed in water, the hydrophilic linear polymers can diffuse out of the gel matrix. This can result in undesirable volume transitions of the gels, and the introduction of unwanted compounds into the surrounding medium. In contrast, there have been relatively few studies on the preparation of full IPNs of PHEMA that ultilize hydrophilic polymers as the IPN agent. In these studies, the hydrophilic polymer is poly(vinyl alcohol)[22] or gelatin.[23] Because both of these polymers have limited compatibility with PHEMA, visually clear networks have only been obtained when the HEMA content of the reaction mixture is >40%.

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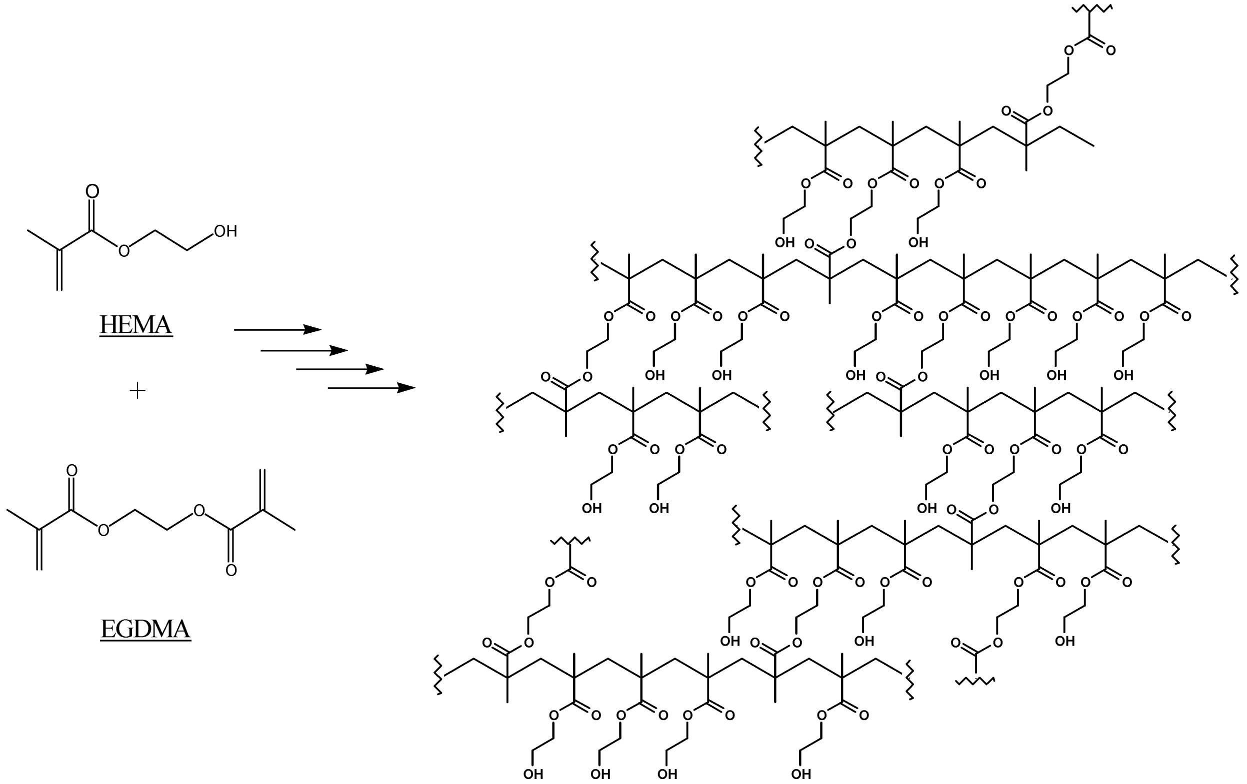
Scheme 1. Network Formation by the Free Radical Copolymerization of HEMA and EGDMA



In the present work, the synthesis of full IPNs based on cross-linked PHEMA and cross-linked poly(ethylene oxide) diglycidyl ether (PEODGE) is described; the first polymer network is formed by the free-radical copolymerization of HEMA and ethylene glycol dimethacrylate (EGDMA) (Scheme 1) and then the second network by coupling reactions between PEODGE and ethylenediamine (EDA) (Scheme 2). When compared to other existing PHEMA networks, visually clear polymers are obtained at significantly lower total monomer concentrations; the resultant networks obtained from this new approach also have very different swelling and porous properties, which can overcome some of the disadvantages described above.

Materials and Methods

Materials. HEMA (97%), EGDMA (98%), PEODGE (MW = 530), poly(ethylene glycol) (PEG; MW = 400), EDA (>99.5%), *N*,*N*,*N*′,*N*′-tetramethylethylenediamine (TEMED; >99.5%), ammonium persulfate (APS; >99.5%), isopropyl ether (IPE, >99%), and benzoic acid were purchased from Aldrich Fine Chemicals (Castle Hill, NSW, Australia). *N*,*N*-Dimethylformamide (DMF), hydrochloric acid (HCl; 37% in water), and sodium hydroxide were obtained from AJAX FineChem (Seven Hills, Australia). HEMA and EGDMA were filtered through an activated basic alumina column, distilled under reduced pressure, and stored at 4 °C. The epoxy content of the PEODGE sample was determined to be $(3.22 \pm 0.01) \times 10^{-3}$ mol/g by standard HCl titration in DMF (according to ref 24), which gave a number-average molecular weight of 620 when each poly(ethylene oxide) (PEO) chain was assumed to have two epoxy end groups. All other reagents, unless specified, were of analytical grade and were used without further purification, and distilled water was used at all times.

The monomer solutions are classified according to their monomer contents (wt % total monomers in the reaction mixture) and cross-linker content (mol % cross-linker in the monomer mixture). The PEODGE concentration of the reaction mixtures is given in weight percentage. The actual functionality (f_a) of EDA and PEODGE in the coupling reactions is defined as

$$f_a(\text{molecule A}) = \frac{\text{total number of functional group on A that can react}}{\text{total number of molecules A in the system}} \quad (1a)$$

For example, the actual functionality of EDA when reacting with PEODGE is given in the following equation:

$$f_a(\text{EDA}) = (\text{functionality of PEODGE})\frac{[\text{EDA}]}{[\text{PEODGE}]} = 2\frac{[\text{EDA}]}{[\text{PEODGE}]} \quad (1b)$$

Preparation of Semi-I IPNs. Monomer solutions (5 g) were prepared by mixing HEMA and EGDMA in the appropriate amount of PEODGE solution in water (% PEODGE is calculated according to the weight of PEODGE in the final reaction mixture) in disposable glass vials. The mixture was degassed by argon purging prior to the addition of the initiator system (0.1 mol % initiator/mol of double bonds) composed of freshly made 10% (v/v) TEMED and 10% (w/v) APS. The polymerization was then allowed to proceed at room temperature overnight under an argon atmosphere.

Preparation of Full IPNs. Semi-I IPNs prepared according to the above procedures were (1) placed in IPE solutions of EDA (50 mL, 2 mol of EDA/mol of PEODGE) for 96 h at 30 °C (method A) or (2) placed in IPE (50 mL) and equilibrated to 30 °C, after which EDA was added over 10 h, at 2 h intervals (5 × 0.2 mol of EDA/mol of PEODGE, followed by 1 mol of EDA/mol of PEODGE), and kept at constant temperature for a further 86 h (method B).

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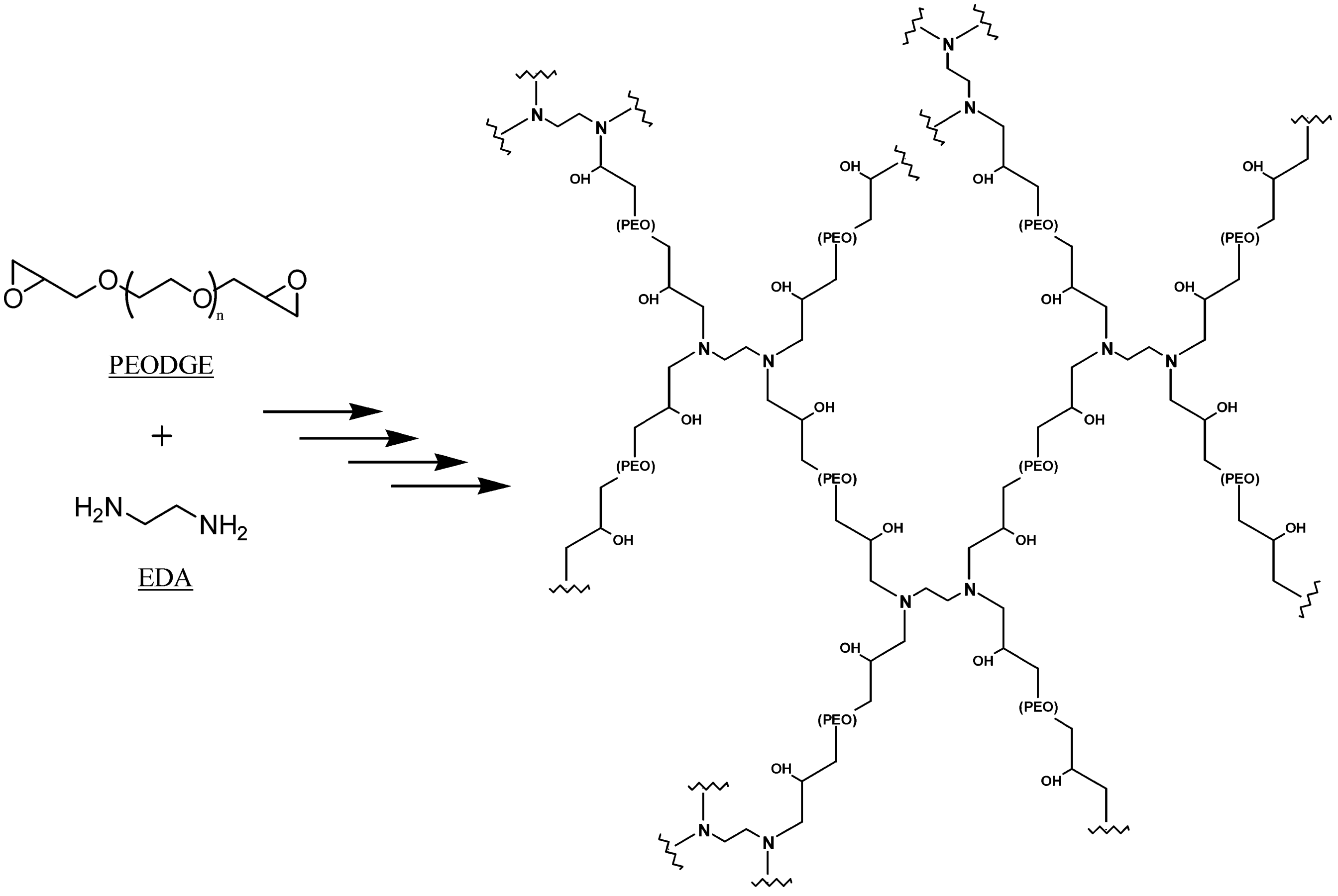
Scheme 2. Network Formation by the Coupling Reactions between PEODGE and EDA[a]



[a] PEO denotes the poly(ethylene oxide) unit of PEODGE.

Turbidity Measurements. Monomer solutions (10 g) were prepared according to the above procedure. After the addition of the initiator system, two 3.75 mL samples were pipetted into disposable cuvettes (10 × 10 × 45 mm^3), and the polymerization was then allowed to proceed at room temperature overnight under an argon environment.

Turbidity measurements of the resultant gels were made using UV−vis spectrophotometry. Distilled water was used for the baseline, and the absorbance of each gel sample was recorded at 600 nm. Turbidity (τ) is defined by the equation $I/I_o = \exp(-\tau x)$, where I_o and I are the initial and final light intensities transmitted through the sample and x is the sample length. The adsorption of the sample, A, is defined by the equation $A = \log(I_o/I)$. Therefore, the turbidity of the gel samples was finally determined by the following equation:

$$\tau = -[\ln(10^{-A})] \tag{2}$$

Swelling Studies. Polymers made according to the above procedure were immersed in water for 1 week, during which the immersing solutions were exchanged on a daily basis. The gels were then dried in a 40 °C regular oven for 1 week. The equilibrium water content (EWC) of the gels was determined according to eq 3. The gel yield of the reactions was calculated from the weight of the dried gel.

$$\mathrm{EWC} = \frac{\mathrm{weight(swollen\ gel)} - \mathrm{weight(dried\ gel)}}{\mathrm{weight(dried\ gel)}} \tag{3}$$

Cryo Scanning Electron Microscopy (Cryo-SEM) Analysis. After equilibration in water, a piece of the hydrogel (5 mm × 5 mm) was mounted vertically onto an SEM stub and cryogenically fractured in liquid nitrogen. The water from the fractured surface of the gel was sublimed at −60 °C for 60 min. The gel was then cooled to −190 °C, and images of the fractured polymer were taken using an XL30 field emission scanning electron microscope.

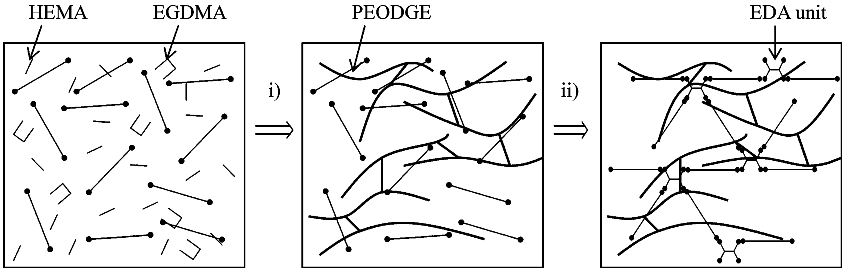
Results and Discussion

The general procedure for the preparation of the IPNs is schematically shown in Scheme 3. In the first step, HEMA and EGDMA are copolymerized in an aqueous solution of PEODGE. After the formation of the semi-I IPN, the gel is placed in an IPE solution of EDA. Upon the diffusion of EDA into the interior of the gel, the amino hydrogens can react with the epoxy rings of PEODGE, resulting in the formation of a hybrid class of sequential and simultaneous full IPN.

Semi-I IPNs of PHEMA. In previous work[7] we demonstrated that in contrast to the use of water as solvent for HEMA polymerization, the use of suitable hydroorganic mixtures as polymerization solvent can result in the formation of clear hydrogels at low initial monomer contents (<60%) due to the ability of the organic solvent to solvate the polymer chains throughout the polymerization. The organic component of these polymerization solvents can be selected on the basis of its solubility parameter (δ). It was found that, at high water dilutions, the formation of a visually clear hydrogel is promoted by the use of an organic component that has a δ similar to or lower than that of PHEMA.

Poly(ethylene oxide)s (δ = 24.2 $MPa^{0.5}$ [25]) have a δ similar to that of PHEMA (δ = 26.93 $MPa^{0.5}$ [26]); PEODGE is therefore chosen as the IPN agent because, first, it can act as part of the polymerization solvent to prepare

Scheme 3. Schematic Diagrams Representing the Preparation of pHEMA/PEO IPNs[a]



[a] (i) Semi-I IPN is formed by the free radical polymerization of HEMA and EGDMA in aqueous PEGDGE solutions. (ii) The final full IPN is prepared by coupling reactions between PEODGE and EDA, which is diffused into the gel from the immersing solution.

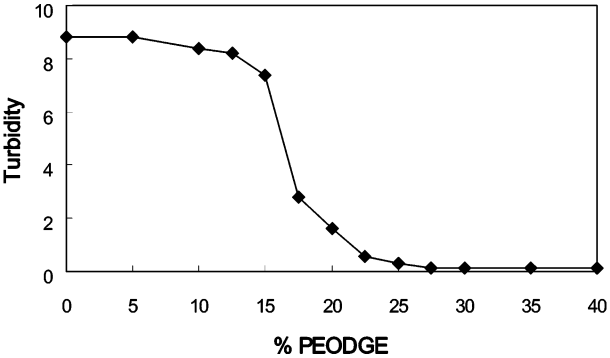


Figure 1. Turbidity measurements of semi-I IPNs of pHEMA (20 wt % total monomers, 2 mol % cross-linker) synthesized in the presence of various amounts of PEODGE.

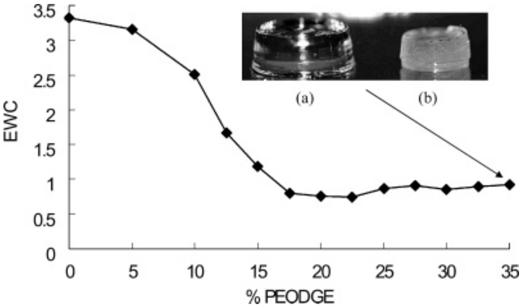


Figure 2. EWCs of semi-I IPNs of pHEMA (20 wt % total monomers, 2 mol % cross-linker) synthesized with various amounts PEODGE. The inset shows photographs of the gel network at 35% PEODGE (a) before and (b) after the solvent exchange process.

visually clear semi-I IPNs of PHEMA at low monomer contents and, second, it is well-known that the extent of swelling of a polymer is related to the difference between its δ and that of the swelling solvent.[25] The final PEO network (formed after the subsequent PEODGE coupling reactions) of the full IPNs is therefore expected to provide solvation to the PHEMA network.

Semi-I IPNs of PHEMA (20 wt % monomers, 2 mol % cross-linker) were prepared at various % PEODGE values and their turbidities measured (Figure 1) to monitor the extent of polymerization-induced phase separation in the samples, and to provide guidelines for the preparation of clear semi-I IPNs. The results demonstrate the ability of PEODGE to solvate PHEMA chains formed during the polymerization; visually clear hydrogels can be obtained when the reaction mixture contains more than 25% PEODGE.

The degree of swelling observed at equilibrium, as represented by the EWC of the polymer via eq 3, is a representation of the competition between the entropy of dilution, gained by the added volume of the polymer throughout which the solvent may spread, and the elasticity of the polymer network as well as the heat of mixing.[27,28] EWCs of water-swollen semi-I IPNs are shown in Figure 2. The degree of swelling is significantly higher when the reaction mixture contains low amounts of PEODGE; it first decreased with increasing PEODGE content until ~20%, and then remained at an approximately constant EWC value at higher PEODGE concentrations.

The swelling properties of the opaque polymers are similar to those reported in our earlier work[7] and to those of the PHEMA gels obtained by Dušek and Sedlacek[29] and Chirila et al.[2] in pure aqueous systems. Their enhanced swellings were ascribed to variations in the dimensions of the polymeric network caused by the phase separation process. More importantly, Figure 2 also demonstrates that the clear gels formed in the absence of phase separation have a narrow range of EWCs (0.8−0.95). The volume transitions of these gels are illustrated by the inset in the figure, which shows a photograph of the semi-I IPN synthesized in the presence of 35% PEODGE before (EWC = 4) and after (EWC = 0.9) the solvent exchange process. This phenomenon is attributed to changes in the δ of the swelling medium (PEODGE within the gel network has diffused out and is replaced with water).

Coupling Reactions of PEODGE and EDA. The glycidyl end groups of PEODGE are known to react with amines that contain active hydrogen atoms.[30,31] In this work, EDA—a compound which contains two primary amine groups—is used as the coupling agent for the formation of the full IPNs.

In step-growth polymerizations, the functionality of a molecule is not simply the number of functional groups

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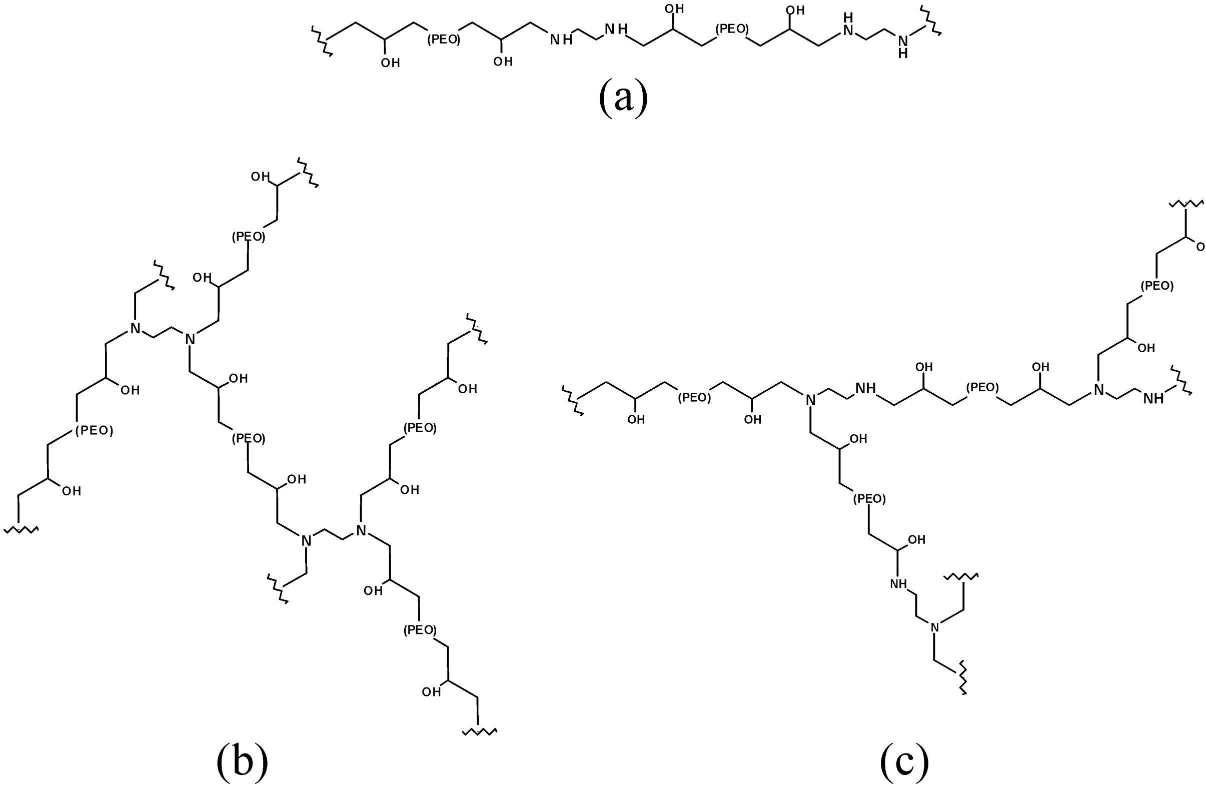
Scheme 4. Schematic Representation of the Epoxy−Amine Coupling Reaction at (a) 1:1 EDA:PEODGE, (b) 1:2 EDA:PEODGE, and (c) 3:4 EDA:PEODGE



in that molecule, but is the number of functional groups that can react in the system under consideration.[32] Therefore, although the potential functionality of EDA is 4 (active amino hydrogens) and that of PEODGE is 2 (epoxy groups), the actual functionality (f_a) of the molecules is dependent upon the ratio of the two compounds in the system and is defined according to eq 1. The formation of a three-dimensional polymer network requires the presence of branch units (units with $f_a > 2$), whereas linear polymers result when both monomers are bifunctional.

The influence of EDA on the coupling system will be discussed by examples in which f_a(PEODGE) is kept at 2. This is achieved when the systems have at least 1 mol of amino hydrogen/mol of epoxy, such that all epoxy groups on PEODGE can be reacted.

When there is 2 mol of amino hydrogen available for every mole of epoxy (i.e., 1:1 EDA:PEODGE; Scheme 4a), f_a(EDA) is decreased to 2 because only half of the amino hydrogens can react and linear polymers are therefore expected to form in the reaction mixture. On the other hand, the formation of three-dimensional networks occurs when f_a(EDA) is >2. The tightest gel network is expected to form when there is 1 mol of amino hydrogen/mol of epoxy (i.e., 1:2 EDA:PEODGE; Scheme 4b); f_a(EDA) is 4 in this system, and networks with tetrafunctional cross-linked points are expected. The porosity and flexibility of the networks are expected to increase with decreasing f_a(EDA). For example, when there is 1.5 mol of active hydrogen available for every mole of epoxy group (i.e., 3:4 EDA:PEODGE; Scheme 4c), f_a(EDA) is decreased to 3 and networks with trifunctional branched points are obtained.

To investigate the effects of PEODGE and EDA on the coupling reactions, various PEO networks were prepared (in the absence of HEMA and EGDMA) by reacting PEODGE and EDA at different dilutions of water. The products from these reactions are transparent hydrogels, which are similar to the PEO hydrogels obtained by cross-linking PEG with diisocyanates.[33]

Figure 3a shows the EWC of water-swollen PEODGE hydrogels prepared at various % PEODGE values, when the number of moles of EDA per mole of PEODGE is varied from 0.5 to 1 such that f_a(EDA) is decreased from 4 to 2 while f_a(PEODGE) is kept at 2. At the same f_a(EDA), the EWC of the gels was observed to decrease with increasing % PEODGE, which is consistent with the increasing network density of the gels. The gel yields of the reaction mixture are shown in Figure 3b, and were observed to follow a trend similar to that of the EWC values.

In contrast to the theoretical predictions, minimum EWC values were not obtained when f_a(EDA) was 4, but when it was 3.6, which can be attributed to the restricted mobility of the functional groups in the post gel reaction period. On the other hand, Figure 3b shows that (1) significant amounts of cross-linked gel products were obtained at $f_a(\mathrm{EDA}) = 2$, when linear polymers were expected, and (2) the gel yields of the reaction mixtures decreased with decreasing f_a(EDA) when the functionality was between 3.6 and 2.4, when the systems were expected to produce cross-linked polymers with different extents of swelling but similar gel yields.

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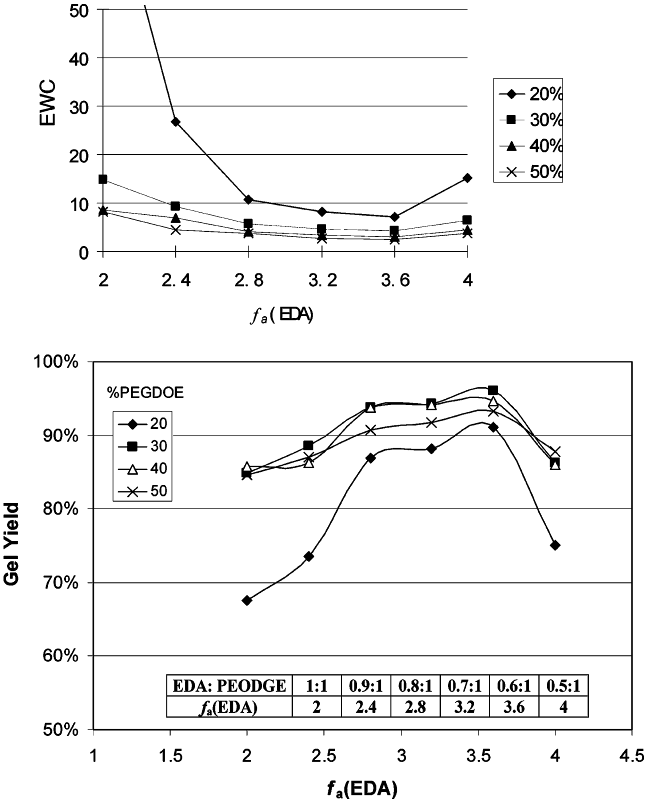

Figure 3. (a, top) EWCs of PEO hydrogels that were synthesized at 20−50% PEODGE, when EDA:PEODGE was varied from 1:1 to 0.5:1 such that f_a(EDA) was increased from 2 to 4. (b, bottom) Gel yields of PEO hydrogels that were synthesized at a specific PEODGE concentration versus the actual functionality of EDA (which corresponds to a specific DEA:PEODGE ratio). Inset table: The DEA:PEODGE ratios used correspond to the values of the actual functionality of EDA.

These two observations suggest that the calculated f_a(EDA) is in fact an average of a distribution composed of EDA units with higher and lower functionality; PEO chains coupled with EDA units of high functionalities (>2) form the gel network, while those coupled with EDA units of low functionalities (≤2) form soluble polymers which do not constitute part of the polymer network. The broad distribution of f_a(EDA) can occur if the epoxy−secondary amine reactions are significantly faster than the epoxy−primary amine reactions (i.e., a positive kinetic substitution effect[34,35]). Although it has been reported[36] that in aliphatic amines the primary and secondary amino hydrogens have closely similar reactivities, the observed positive substitution effect can be caused by other factors either thermodynamic or kinetic,[37] for example, the localization of PEODGE chains in the reaction mixture.

Full IPNs of PHEMA and PEO. To prepare the full IPNs, semi-I PHEMA networks (20 wt % monomers, 2 mol % cross-linker) were formed in the presence of 35% PEODGE and then the PEO chains coupled with EDA.

Table 1. EWCs of Full IPNs (20 mol % Monomer, 2 mol % Cross-Linker HEMA/EGDMA, 35% PEGOGE, Method A) Prepared When Various Amounts of EDA Are in the Immersing Solution

n(EDA)/ n(PEODGE)	EWC	n(EDA)/ n(PEODGE)	EWC	n(EDA)/ n(PEODGE)	EWC
0.25	2.00	1	2.87	6	3.15
0.5	2.11	2	3.10	8	3.09
0.75	2.54	4	3.14		

In the first set of experiments (method A), the gels were placed in IPE solutions which contain various amounts of EDA (0−8 mol of EDA/mol of PEODGE). Owing to the low solubility of EDA in IPE, all the IPE solutions are slightly turbid. At the end of the reactions, the initially turbid solutions became clear, which indicated the diffusion of EDA into the interior of the gel.

Polymer networks prepared according to the above procedure were equilibrated in water and their EWCs determined (Table 1). It can be seen that coupling of PEODGE chains within the hydrogel network leads to significant increases in the degree of swelling; the EWCs of the gels first increase with increasing amounts of EDA in the immersing solution and then remain approximately constant at around 3.0−3.2 when there is 2 mol of EDA/mol of PEODGE. The high water intake of these gels can be attributed to the hydrophilicity of the PEO network, and the favorable interactions between PEO and PHEMA, which reduce the hydrophobic interactions between PHEMA chains in water.

When low amounts of EDA were used (<2 mol of EDA/mol of PEODGE), IPNs with significantly lower EWCs and collapsed cores (Figure 4a) were formed because of insufficient coupling of PEODGE chains in the region; during the swelling process, the lightly branched PEODGE chains are removed from the network, which leads to gel shrinkage in the middle of the gel. This phenomenon was not observed at higher EDA concentrations, which indicates the formation of PEO networks throughout the sample.

It is interesting to note that when >1 mol of EDA/mol of PEODGE was used, significant surface cracks (layers of gels being “peeled off” from the exterior, as shown in Figure 4b) were observed in all the water-swollen IPNs. This phenomenon can be understood in terms of the diffusion process of EDA into the interior of the gel and its influence on the coupling process. The transfer rate of EDA from the bulk solution to the interior of the gel is proportional to the concentration gradient and to the area of the interface; the concentration of EDA is hence expected to be much higher at the exterior of the gel and also at the start of the reaction. The excess amount of amine group at the outer layer of the networks can lead to the formation of various PEO-based polymers that do not constitute part of the gel network because they have EDA units with reduced f_a. The observations are consistent with results obtained from the previous section, and suggest that the formation of a full PEO network throughout the IPN is favored by the slow diffusion of EDA (≥2 mol of EDA/mol of PEODGE) into the semi-I networks. It should be noted that, when compared to results obtained in the preceding section, the amounts of EDA per PEODGE used in full IPN formation are significantly higher than those used in the PEO hydrogel formation. This can be

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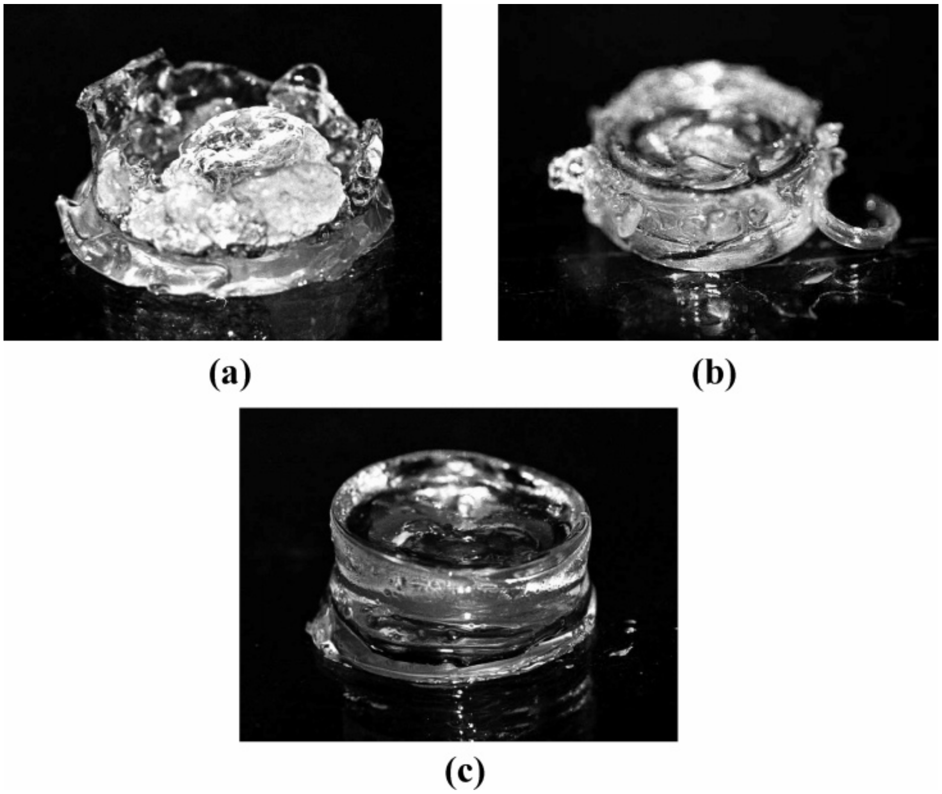

Figure 4. Images of full IPNs (20 wt % total monomers, 2 mol % cross-linker, 35% PEODGE) prepared by method A, (a) 1 mol of EDA/mol of PEODGE, (b) 2 mol of EDA/mol of PEODGE, and by method B, (c) 2 mol of EDA/mol of PEODGE.

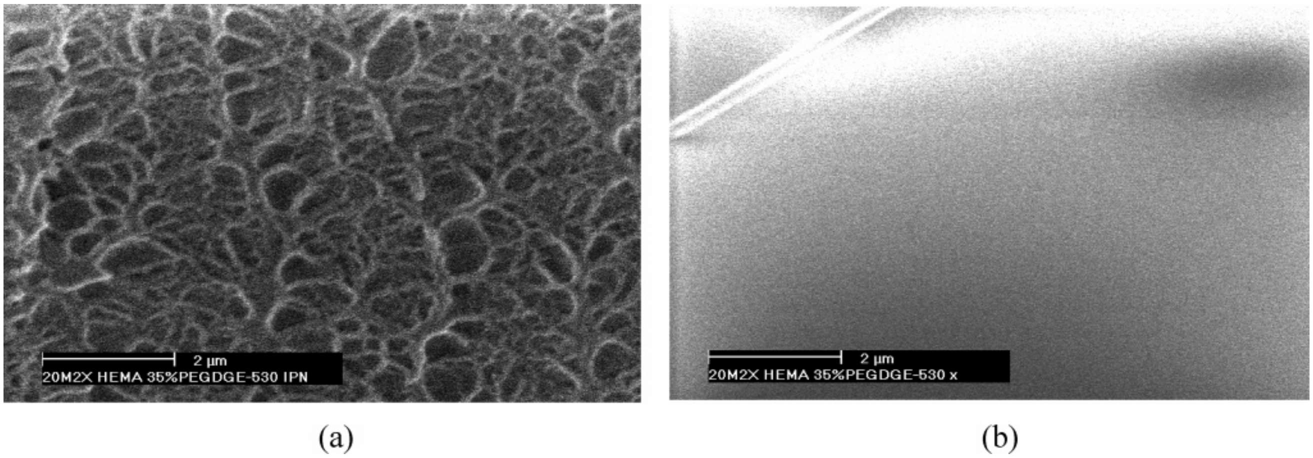

Figure 5. SEM images (10000×) of the cross-sectional interior of water-swollen (a) full and (b) semi-I IPNs of pHEMA (20 wt % total monomers, 2 mol % cross-linker, 35% PEODGE, 2 mol of EDA/mol of PEODGE, method B). The scale bars are 2 μm in both images.

attributed to the incomplete transfer of EDA from the immersing medium to the gel matrix.

In a subsequent set of experiments (method B), the semi-I network was placed in IPE, and EDA (overall 2 mol of EDA/mol of PEODGE) added into the solution at regular time intervals to reduce the transfer rate of EDA to the gel. As illustrated in Figure 4c, no visible surface cracks were observed on the network that was prepared by this approach. The EWC and gel yield (the percentages of PHEMA and PEODGE chains that are connected to the final gel network) of the network were determined to be 3.6 and 83%, respectively, which indicates the successful incorporation of PEO chains into the network. Cryo-SEM images of the water-swollen full IPN were taken—and compared to those of the corre-

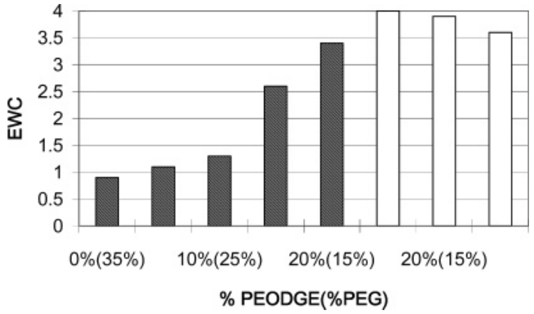

Figure 6. EWCs of full IPNs (20 mol % monomers, 2 mol % cross-linker HEMA/EGDMA, 2 mol of EDA/mol of PEODGE, method B) prepared in the presence of various PEODGE/PEG-400 mixtures. Visually clear gels are represented by solid bars and visually opaque gels by shaded bars.

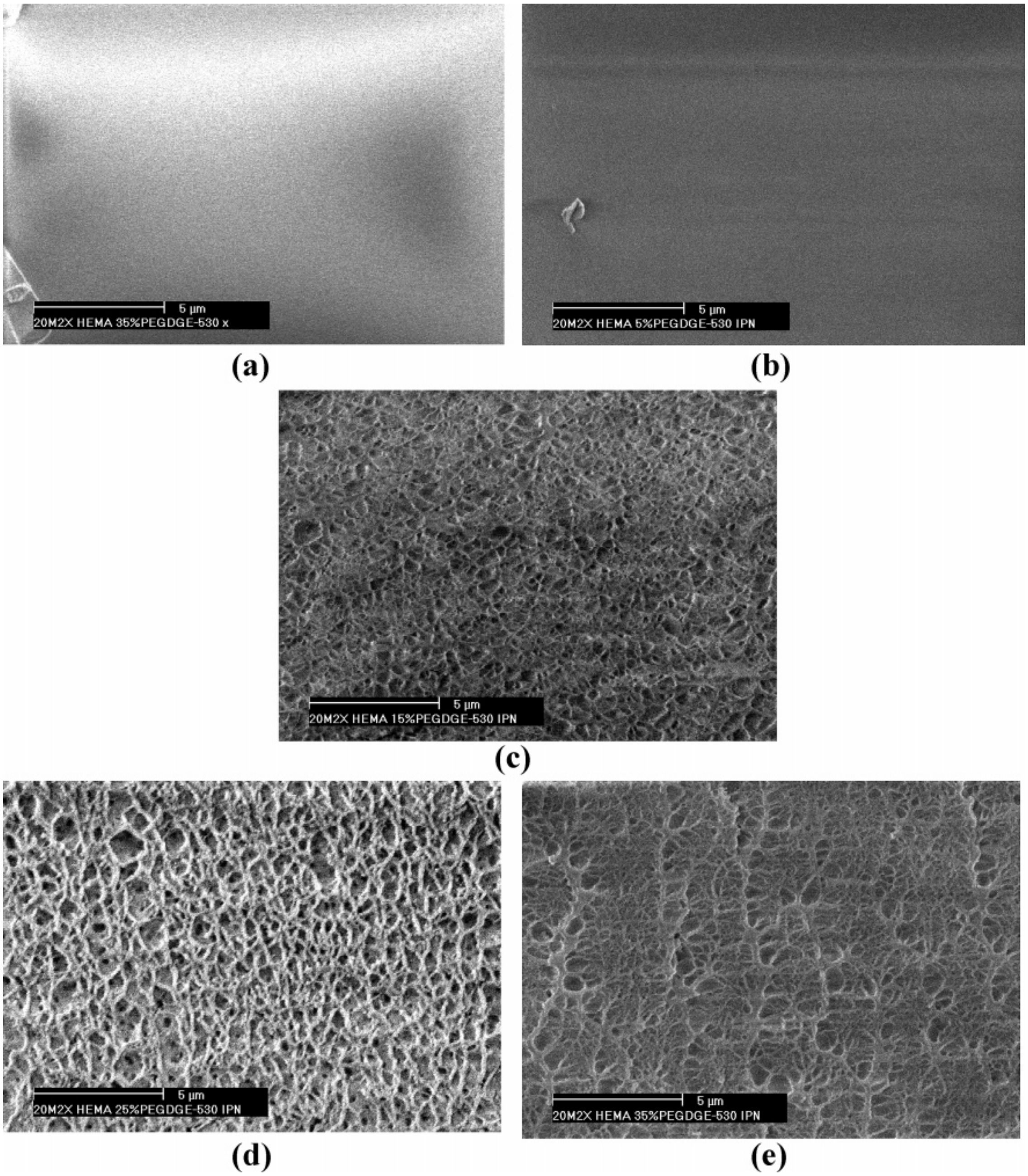

Figure 7. SEM images (5000×) of the cross-sectional interior of water-swollen full IPNs (20 wt % total monomers, 2 mol % cross-linker, 2 mol of EDA/mol of PEODGE, method B) prepared with (a) 0%, (b) 5%, (c) 15%, (d) 25%, and (e) 35% PEODGE. The scale bars are 5 μm in all images.

sponding semi-I IPN—to examine the surface morphologies and apparent pore size distributions of the gel networks (Figure 5). It can be seen clearly that, when compared to the water-equilibrated semi-I IPN sample and also to visually clear PHEMA networks obtained by conventional techniques, the full IPN has much bigger pores and a wider pore size distribution. Particularly, two distinct types of pores—macropores on the order of up to 800 nm, and micropores on the order of 50−100 nm—were observed in the images.

Results from Figure 1 show that at 20 wt % monomers and 2 mol % cross-linker, visually clear polymers of HEMA can only be formed when the reaction mixture contains more than 25% PEODGE. PEG-400, which has a structure and molecular weight similar to those of PEODGE, was therefore chosen to partly replace PEODGE in the polymerization solvent for the effects of % PEODGE on the swelling properties of the resultant polymer networks to be investigated. In the experiments, visually clear semi-I networks of PHEMA were prepared in a range of 35% (PEG-400/PEODGE) mixtures, after which the PEODGEs were coupled by method B (overall 2 mol of EDA/mol of PEODGE). PEG-400 is inert in the coupling reactions and can consequently be washed out from the network matrix during the solvent exchange process.

EWCs of the water-swollen full IPNs are shown in Figure 6; it can be seen (from the shaded bars) that the IPNs obtained at lower % PEODGE are slightly opalescent. Cryo-SEM images of the water-swollen full IPN were taken to examine the effects of % PEODGE on the surface morphologies and apparent pore size distributions of the gel networks (Figure 7). It can be seen that results obtained from the cryo-SEM analysis are consistent with those obtained from the above swelling studies; the porosity of the opalescent networks was observed to increase with increasing % PEODGE, while pore sizes on the order of 1 μm were obtained at lower % PEODGE (25%, compared with 35%) for the visually clear networks.

By comparing Figures 2 and 6, it can be seen that visually clear semi-I IPNs and full IPNs are both obtained when the reaction mixture contains ≥25% PEODGE; this highlights the different levels of solvation of the PEO chains on the PHEMA network during and after the synthesis. EWCs of the visually clear networks (25−35% PEODGE) were found to increase with decreasing % PEODGE, which is consistent with results obtained from the PEODGE−EDA coupling reactions and can be attributed to the decreased network density of the system. On the other hand, EWCs of the opalescent networks (0−20% PEODGE) were found to decrease with decreasing % PEODGE, which is consistent with the expected increasing hydrophobicity of the networks.

Conclusions

In this paper we describe the preparation of HEMA-based IPNs by the use of PEODGE as the IPN agent. It was demonstrated that visually clear networks—with higher equilibrium water contents and bigger pore sizes than those of conventional HEMA hydrogels—can be formed by preparing full IPNs of the polymer and a cross-linkable macromolecular solvent for the polymer. The properties of these networks are very different from those of the corresponding semi-I IPNs, and are shown to be dependent upon the EDA and PEO contents of the system. In general, the pore sizes of the networks were observed to decrease with increasing PEODGE content of the reaction mixture, while optimum network formation is promoted by the slow addition of EDA (with ≥2 mol of EDA/mol of PEODGE) into the semi-I networks.

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