

# A New Thermo-Sensitive Hydrogel: Poly(ethylene oxide-dimethyl siloxane-ethylene oxide)/Poly(*N*-isopropyl acrylamide) Interpenetrating Polymer Networks

## I. Synthesis and Characterization

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**ABSTRACT:** Interpenetrating polymer networks (IPNs) composed of poly(ethylene oxide-dimethyl siloxane-ethylene oxide) (PEO-PDMS-PEO) ABA triblock copolymer and poly(*N*-isopropyl acrylamide) (PIPAAM) were synthesized. Transmission electron microscopy (TEM) and differential scanning calorimetry (DSC) studies of bulk IPNs revealed micro phase separation with partial phase mixing between two chemically independent networks. Aqueous equilibrium swelling curves were examined as a function of temperature. DSC thermograms of the swollen IPNs exhibited the same swelling transition temperature as that of PIPAAM, although the degree of swelling of IPNs was affected by polymer composition. These observations indicate that each network in the swollen IPNs exists independently and many have different morphology than the dried bulk state. The aqueous swelling-deswelling kinetics by temperature modulation of IPNs could be controlled by gel composition and temperatures applied.

**KEY WORDS** Interpenetrating Polymer Networks / Poly(ethylene oxide-dimethyl siloxane-ethylene oxide) / Poly(*N*-isopropylacrylamide) / Transmission Electron Microscopy / Differential Scanning Calorimetry / Equilibrium Swelling Curve / Swelling-Deswelling Kinetic / Temperature Modulation / Morphology /

Aqueous polymer solutions such as poly(methyl methacrylic acid),<sup>1</sup> poly(ethylene oxide),<sup>2</sup> poly(vinyl alcohol-ethylacetate),<sup>3</sup> and poly(*N*-isopropyl acrylamide)<sup>4</sup> exhibit phase separation at elevated temperature. This temperature is called the Lower Critical Solution Temperature (LCST) determined by relatively large negative entropy change.<sup>5-7</sup> An appropriate hydrophobic/hydrophilic (HPB/HPL) balance of polymer molecules is considered an important factor in controlling LCST of a polymer solution.<sup>8</sup> LCST behavior can be

correlated to the temperature dependence of the aqueous swelling of the crosslinked polymer.

Crosslinked poly(*N*-isopropyl acrylamide) (PIPAAM) demonstrates a sharp swelling transition (gel shrinking) around its LCST of 31—33°C,<sup>4</sup> which may be unique among thermosensitive gels due to a relatively symmetric and sharp endothermic peak at the swelling transition temperature in DSC studies.<sup>9</sup>

Recently, crosslinked PIPAAM and its copolymer with a hydrophilic (HPL) component

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have been investigated in the separation process,<sup>10</sup> immobilized enzyme activity control,<sup>11</sup> and chemical delivery of water soluble solutes.<sup>12</sup> However, because of the high water content of crosslinked PIPAAm, its mechanical strength is weak, and may limit its practical applications.<sup>13</sup>

In general, we control the swelling transition temperature and thermosensitivity of PIPAAm networks by improving mechanical properties. The first is to copolymerize PIPAAm with other hydrophobic (HPB) or HPL components.<sup>11,14</sup> Another approach is the formation of interpenetrating polymer networks.<sup>15</sup> In the first method, the swelling transition temperature as well as degree of swelling are altered by modifying the HPB/HPL balance in the PIPAAm chain. In an IPN system, the swelling transition temperature may be kept at the same temperature as that of the PIPAAm network, regardless of the content of other components, because the PIPAAm network may exist independently. Therefore, these methods may be combined to give specific swelling properties.

In this paper, the synthesis of PEO-PDMS-PEO/PIPAAm IPNs, the bulk morphology of IPNs distinguished by differential scanning calorimetry (DSC) and transmission electron microscopy (TEM), and thermosensitivity in the aqueous equilibrium swelling of the IPNs for drug delivery studies are described. The swelling-deswelling kinetics of IPNs, seen with temperature alternations past the swelling transition temperature, is also discussed.

## EXPERIMENTAL

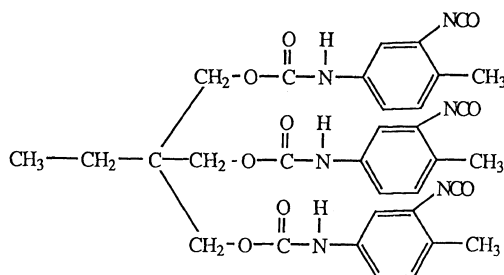
### Materials

*N*-Isopropyl acrylamide (IPAAm) ( $M_w = 113.2$ , mp = 63–65°C), ethyleneglycol dimethacrylate (EGDMA) ( $M_w = 198.2$ , bp = 83–85°C/1 mmHg), poly(ethyleneoxide) (PEO) ( $M_w = 600$ ), and 2,2'-azobisisobutyronitrile (AIBN) were purchased from Polysciences Inc. (Warrington, P.A., U.S.A.) and used

without further purification. EGDMA was used after distillation. PEO was used after dehydration by vacuum-drying at 40°C (20–30  $\mu$ mHg).

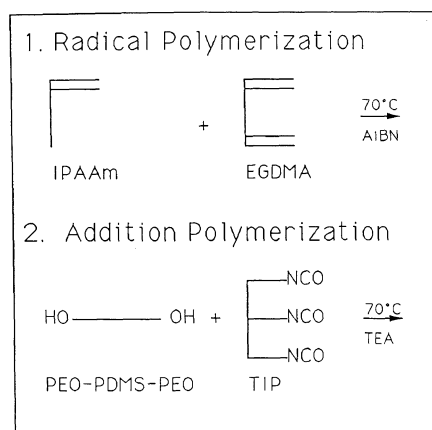
The Functionalized prepolymer of dihydroxy-terminated poly(ethylene oxide-dimethyl siloxane-ethylene oxide) (PEO-PDMS-PEO) ABA triblock copolymer ( $M_w = 2400$ , ethyleneoxide content = 50%), and poly(dimethyl siloxane) (PDMS) ( $M_w = 1200$ ) were purchased from Petrarch System Inc. (Bristol, P.A., U.S.A.), and used after dehydration.

1,1,1-Tris[*N*-(4'-methyl-3'-isocyanatophenyl)-carbamoyloxymethyl]-propane (TIP) (Colonet® L) (75 wt% solid content in ethyl acetate solution,  $M_w = 656.6$ , 13.2 wt% of iso-



**Figure 1.** Structure of 1,1,1-tris[*N*-(4'-methyl-3'-isocyanatophenyl)carbamoxyloxymethyl]-propane.

### IPNs SYNTHESIS



### Simultaneous Polymerization

**Figure 2.** Schematic diagram of IPNs synthesis.

cyanate), having the structure shown in Figure 1, was a gift from Japan Polyurethane (Tokyo, Japan).

Triethylenediamine (TEDA), a catalyst for stepwise polymerization, was purchased from Eastmann Kodak Inc. (Rochester, N.Y., U.S.A.). 1,4-dioxane was purchased from J. T. Baker Chemical Co. (Phillipsburg, N.J., U.S.A.) and used after distillation. Osmium tetroxide was purchased from Sigma Chemical Co. (St. Louis, M.O., U.S.A.).

### Synthesis

Simultaneous IPNs were obtained by the free radical polymerization of PIPAAm monomer and EGDMA monomer as a crosslinker, and stepwise polymerization of NCO groups of a trifunctional crosslinker (Colonet<sup>®</sup> L) and hydroxyl moieties associated with

PEO-PDMS-PEO prepolymer. The schematic procedure is shown in Figure 2. Two IPN systems were synthesized, consisting of PIPAAm and PEO-PDMS-PEO, respectively. In addition, homopolymer membranes composed of crosslinked PEO-PDMS-PEO, crosslinked PEO, and crosslinked PIPAAm were individually synthesized. The feed compositions of reactants are summarized in Table I.

In order to get a homogeneous solution, predetermined amounts of PEO-PDMS-PEO and IPAAm were mixed with the following: 1) EGDMA as a crosslinker (1.0 mol% of IPAAm), 2) AIBN as the initiator ( $4.12 \times 10^{-3} \text{ mol l}^{-1}$ ), 3) TIP, which was based on a one to one functional ratio for the hydroxy group of PEO-PDMS-PEO, 4) TEDA, which was 2.0 mol% isocyanate group in TIP, and 5) 1,4-dioxane (total solid feed content = vol-

**Table I.** Feed compositions for IPNs, crosslinked PEO, crosslinked PEO-PDMS-PEO, and crosslinked PIPAAm

No.	Polymer	IPAAm	EGDMA <sup>a</sup>	AIBN <sup>b</sup>	PEO-PDMS-PEO	
		g	mg	mg	g	
1	50/50 IPNs	3	53	9.1	2.31	
2	80/20 IPNs	4.8	84	9.1	0.92	
3	Crosslinked PEO-PDMS-PEO	0	0	0	4.62	
4	Crosslinked PIPAAm	6	105	9.1	0	
5	Crosslinked PEO	0	0	0	0	

No.	Polymer	PEO	Colonet <sup>®</sup> TIP <sup>c</sup>	TEDA <sup>d</sup>	Dioxan	Outlooking of swollen state
		g	g	mg	ml	at 25°C
1	50/50 IPNs	0	0.92	6.5	6	Opaque
2	80/20 IPNs	0	0.37	2.6	6	Translucent
3	Crosslinked PEO-PDMS-PEO	0	1.84	13.1	6	Transparent
4	Crosslinked PIPAAm	0	0	0	6	Transparent
5	Crosslinked PEO	3.48	3.36	23.9	6	Transparent

<sup>a</sup> 1.0 mol% of IPAAm. <sup>b</sup>  $4.12 \times 10^{-3} \text{ mol l}^{-1}$ . <sup>c</sup> Liquid amounts. <sup>d</sup> 0.2 mol% for NCO of TIP.

ume of 1,4-dioxane). The solution was degassed with dried nitrogen gas for thirty minutes and injected between two Teflon sheets. The Teflon sheets, separated by a gasket and backed by glass plates, were then clamped together. The solution as polymerized at 70°C in a constant temperature oven for three days. After cooling to room temperature, the membranes were initially immersed in 100% ethyl acetate to remove unreacted IPAAm and TIP. The membranes were then immersed in 100% methanol to remove unreacted prepolymer and other impurities. The membranes were again immersed in deionized water and equilibrated for two days. The swollen membranes were cut into 12 mm diameter disks using a cork bore. The disks were then dried at ambient temperature for one day and vacuum dried at 40°C for another day.

#### Differential Scanning Calorimetric Studies

Thermal characterization involved measurement of melting temperatures and glass transition temperatures using 30–40 mg samples of polymer or liquid with helium as a sweep gas (25 ml ml<sup>-1</sup>) at a heating rate of 20°C min<sup>-1</sup> from -140 to 170°C in a DSC (Perkin Elmer DSC-4, Norwalk, Conn., U.S.A.). The swollen gels (6–8 mg) were also studied at a heating rate of 0.31°C min<sup>-1</sup> from 27 to 35°C to investigate the swelling transition temperature of swollen IPNs.

#### Transmission Electron Microscopic Studies

Specimens for TEM studies were treated with osmium tetroxide vapor in order to selectively stain the hydrophilic microdomains. The specimens were freeze-dried at -50°C, cut into thin sections (0.5–1.0 mm thickness) and embedded in epoxy resin, modeled after Spurr's method.<sup>16</sup> The ultrathin sections were cut using an ultramicrotome (LKB Ultratome IV, Gaithersburg, M.D., U.S.A.) and the cross-sectional views of the specimens were observed by transmission electron microscopy (JEOL 100CXII, Tokyo, Japan).

#### Swelling Studies

All the IPNs and crosslinked homopolymer were put into vials filled with PBS, pH 7.4. The vials were kept in a temperature controlled water bath ranging from 10 to 60°C. The gels were removed from the vials, blotted with a paper towel to remove surface water and weighed. This procedure was repeated until swelling equilibrium was reached at a given temperature. After equilibration at one temperature, samples were re-equilibrated at other temperatures.

All samples were ambiently dried for one day and vacuum dried at 40°C for one day after the equilibrium weight was obtained at each temperature. The degree of swelling was defined as the weight of water-uptake (gram) per unit mass (gram) of dried polymer ( $W_{H_2O}/W_p$ ).

$$\frac{W_{H_2O}}{W_p} = \frac{(W_{pH_2O}) - W_p}{W_p}$$

Where:  $W_{H_2O}$  is the weight of water-uptake.  
 $W_p$  is the weight of dried polymer.  
 $W_{pH_2O}$  is the weight of the total weight of swollen gel.

For the swelling kinetic studies of the swollen IPNs, temperature modulation was conducted between 25°C, which is below the swelling transition temperature, and 33 or 40°C.

## RESULTS AND DISCUSSION

In this paper we denote these IPNs as 80/20 IPNs and 50/50 IPNs, respectively. As seen in Table I, the designation of 80/20 IPNs and 50/50 IPNs means that the feed composition ratio in the IPN system as polymer component was 80 wt% and 50 wt% of IPAAm. The bulk morphology of 80/20 IPNs and 50/50 IPNs was investigated by DSC and TEM to show incompatibilities and partial phase mixings between two chemically independent networks for each bulk IPN. However, the aqueous swelling curves as a function of temperature and DSC thermograms of IPNs showed the

**Table II.** DSC study of IPNs

No.	Polymer	PDMS	PEO	PEO		PIPAAm
		$T_g/^\circ\text{C}$	$T_g/^\circ\text{C}$	$T_{m1}/^\circ\text{C}$	$T_{m2}/^\circ\text{C}$	$T_g/^\circ\text{C}$
1	PEO ( $M_w$ 600)	—	-70	-3.4	25	—
2	Crosslinked PEO	—	24	—	—	—
3	PDMS ( $M_w$ 1200)	-120	—	—	—	—
4	PEO-PDMS-PEO ( $M_w$ 2400)	-115.2	-87.3	-22.2	18.3	—
5	Crosslinked PEO-PDMS-PEO	-115.2	-12.3	—	—	—
6	Crosslinked PIPAAm	—	—	—	—	135
7	80/20 IPNs	-111	-13	—	—	131
8	50/50 IPNs	-105	0.0	—	—	109

same swelling transition temperature as that of crosslinked PIPAAm. The degree of swelling of IPNs was controlled by IPN composition. The swelling-deswelling kinetics by temperature modulation of IPNs were studied by considering the morphology of dried bulk state which was followed with DSC and TEM.

#### *Differential Scanning Calorimetric Studies for Bulk IPNs*

Glass transition temperature ( $T_g$ ) and melting temperature ( $T_m$ ) of prepolymers and crosslinked polymers obtained by DSC are summarized in Table II.  $T_g$  was marked by finding the midpoint of the transition thermogram.  $T_m$  was defined by the peak point of the endothermic region.<sup>17</sup>

As seen in Table II, PEO-PDMS-PEO prepolymer exhibited two distinct  $T_g$ , corresponding to each component segment. However, the  $T_m$  of PEO-PDMS-PEO prepolymer shifted to a lower temperature than that of PEO prepolymer. These observations indicate that the PEO-PDMS-PEO prepolymer consists of two incompatible polymer segments and has partial phase mixing. Similar behavior has been reported by Stehlicek *et al.*<sup>18</sup> in DSC studies of nylon 12 triblock copolymer. Shalaby and Bair *et al.*<sup>19</sup> also discuss the phenomenon of partial phase mixing which may have contributed to

plasticization of nylon 12.

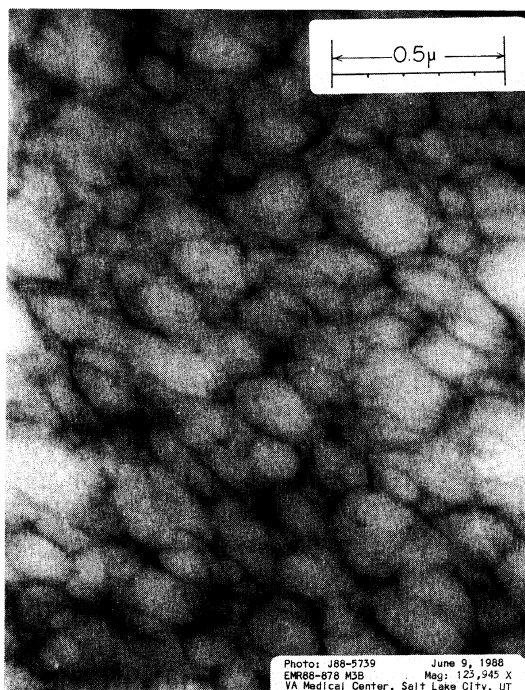
In DSC studies of crosslinked PEO and crosslinked PEO-PDMS-PEO, the  $T_m$  of PEO, which was shown with prepolymers, diminishes, and the  $T_g$  of PEO shifts to higher temperatures, indicating a loss of mobility not seen with PEO and PEO-PDMS-PEO prepolymer.

The thermograms of bulk IPNs showed three distinct  $T_g$  from PEO, PDMS, and PIPAAm phases. The  $T_g$  of PEO-PDMS-PEO increased and the  $T_g$  of PIPAAm decreased when the content of PEO-PDMS-PEO increased. These observations may also indicate phase separation with partial phase mixing seen among the three components. Klemperner *et al.*<sup>20,21</sup> similarly recognized some polymer phase mixing in polyurethane(PU)-polyacrylic acid(PA) IPNs with DSC.

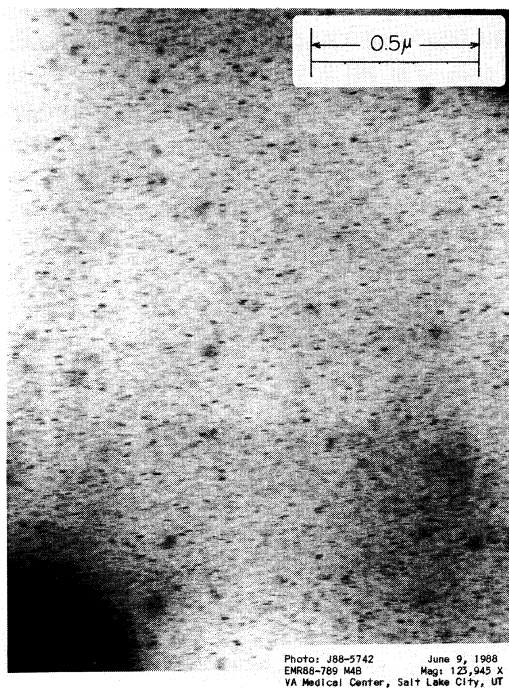
#### *Transmission Electron Microscopic Studies for Bulk IPNs*

Figures 3–5 show TEM pictures of crosslinked PEO-PDMS-PEO (Figure 3), 50/50 IPNs (Figure 4), and 80/20 IPNs (Figure 5) which were stained by OsO<sub>4</sub>. In these studies, OsO<sub>4</sub> could stain the PEO segment, although PIPAAm phase, TIP and PDMS segments were not stained.

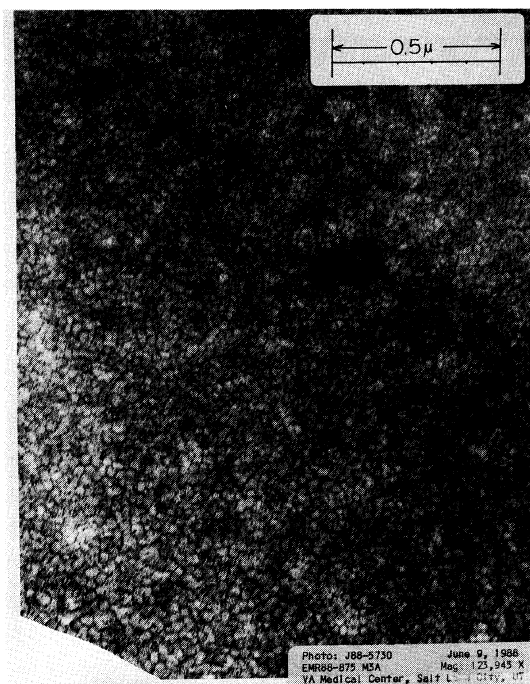
The electron micrographs of crosslinked PEO-PDMS-PEO, 50/50 IPNs, 80/20 IPNs



**Figure 3.** Transmission electron micrograph of cross-linked PEO-PDMS-PEO.



**Figure 5.** Transmission electron micrograph of 80/20 IPNs.



**Figure 4.** Transmission electron micrograph of 50/50 IPNs.

exhibited heterogeneity. Crosslinked PEO-PDMS-PEO (Figure 3) showed cellular morphology. The stained area represents the PEO phase. The unstained area represents the PDMS phase. The average diameter of cellular domains was approximately 200 to 300 Å. The electron micrographs of 50/50 IPNs (Figure 4) showed micro phase separated structures. The size of cellular domains was approximately 20 to 40 Å and the cell walls were believed to be mainly the PEO phase. The inner cell domains were believed to be mainly the PIPAAm phase and PDMS phase. Figure 5 shows an electron micrograph of 80/20 IPNs which exhibits phase inversion. The slightly stained domains (PEO phase) were surrounded by a white area which may consist of PIPAAm and PDMS phases. The PIPAAm phase, which forms a continuous phase, was believed to surround PEO domains with a concentric PDMS layer around PEO domains. The size of the PEO domains was approximately 20 Å.

From the bulk IPN morphology studies with DSC and TEM, 50/50 IPNs was believed to consist of PIPAAm domains with a PDMS concentric layer in the exterior of the domains and a PEO continuous phase, revealing partial phase mixing between the two chemical independent networks. On the other hand, 80/20 IPNs were believed to consist of PEO domains with a PDMS concentric layer around the domains and a PIPAAm continuous phase, also revealing phase mixing behavior.

#### *Differential Scanning Calorimetric Studies of Swollen IPNs*

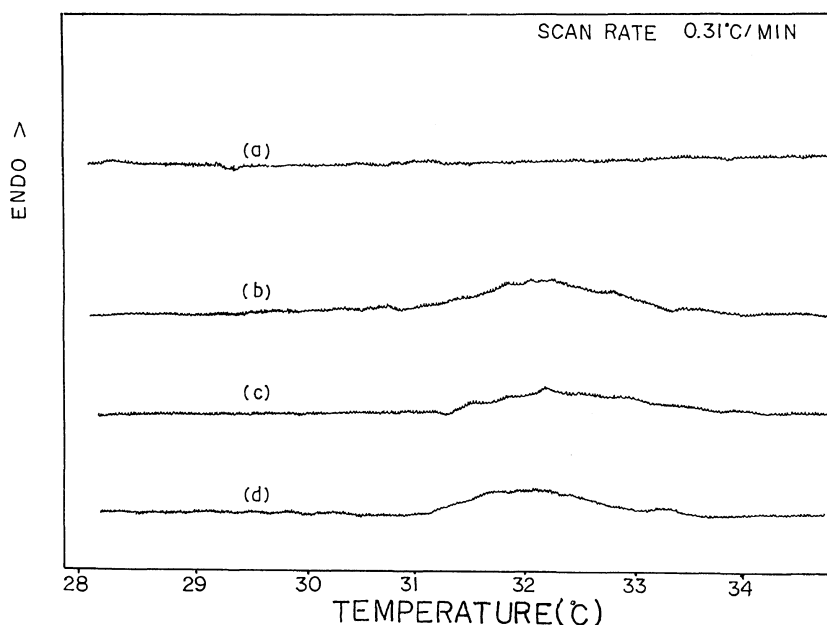
As seen in Figure 6, the transition of all swollen IPNs observed by DSC occurred at the same temperature range as crosslinked PIPAAm (31 to 33°C). The thermograms showed endothermic peaks with heating. These swelling transition phenomena may require energy to change the conformation of the gel and move into a shrinkage stage as the tem-

perature is raised.

#### *Swelling Studies (aqueous equilibrium swelling as the function of temperature)*

Figure 7 shows aqueous equilibrium swelling as a function of temperature for 50/50 IPNs, 80/20 IPNs, crosslinked PEO-PDMS-PEO and crosslinked PIPAAm. All of the IPNs and the crosslinked PIPAAm have the same transition temperature regardless of the composition of the PEO-PDMS-PEO. Bulk dried morphology, observed with DSC, indicated that the PIPAAm phase was mixed with PEO and PDMS segments. The degree of swelling of IPNs was controlled by the content of the PEO-PDMS-PEO. Crosslinked PEO-PDMS-PEO also exhibited a low degree of swelling and a slightly negative thermosensitivity, which is consistent with the properties of PEO.<sup>22</sup>

These observations suggest that the PIPAAm network in the swollen IPNs can expand due to increased compatibility with water be-



**Figure 6.** DSC thermogram of the swollen IPNs, crosslinked PEO-PDMS-PEO, and crosslinked PIPAAm: 1 (a) crosslinked PEO-PDMS-PEO; 2 (b) crosslinked PIPAAm; 3 (c) 80/20 IPNs; 4 (d) 50/50 IPNs.

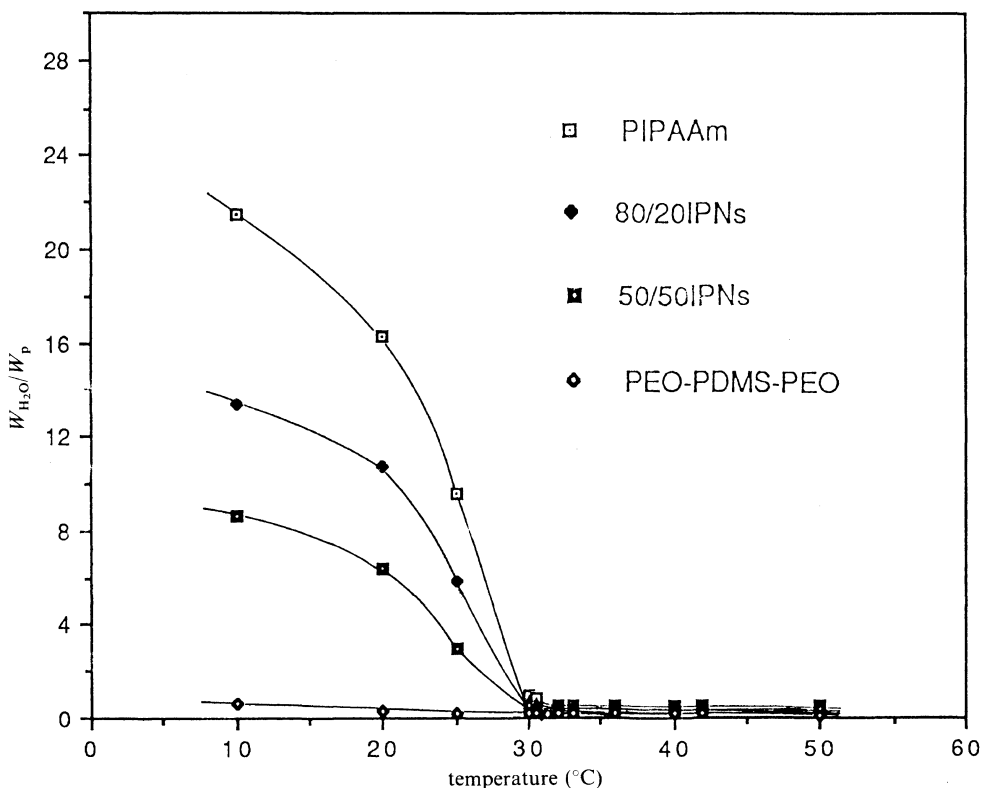


Figure 7. Equilibrium swelling curve of IPNs hydrogel as a function of temperature: a (□) crosslinked PIPAAm; b (◆) 80/20 IPNs; c (■) 50/50 IPNs; d (◇) crosslinked PEO-PDMS-PEO.

low the swelling transition temperature and exist independently. The swelling transition behavior of IPNs was in sharp contrast with the random copolymer of IPAAm, which was affected by comonomer presence.<sup>14</sup> Namely, the swelling transition temperature can be shifted to lower temperature with increasing other hydrophobic component or to a higher temperature with increasing other hydrophilic components in the case of a random copolymer. On the other hand, the swelling transition temperature of IPN can be fixed regardless of the IPN composition.

Swollen 80/20 IPNs and 50/50 IPNs were translucent and opaque, respectively.

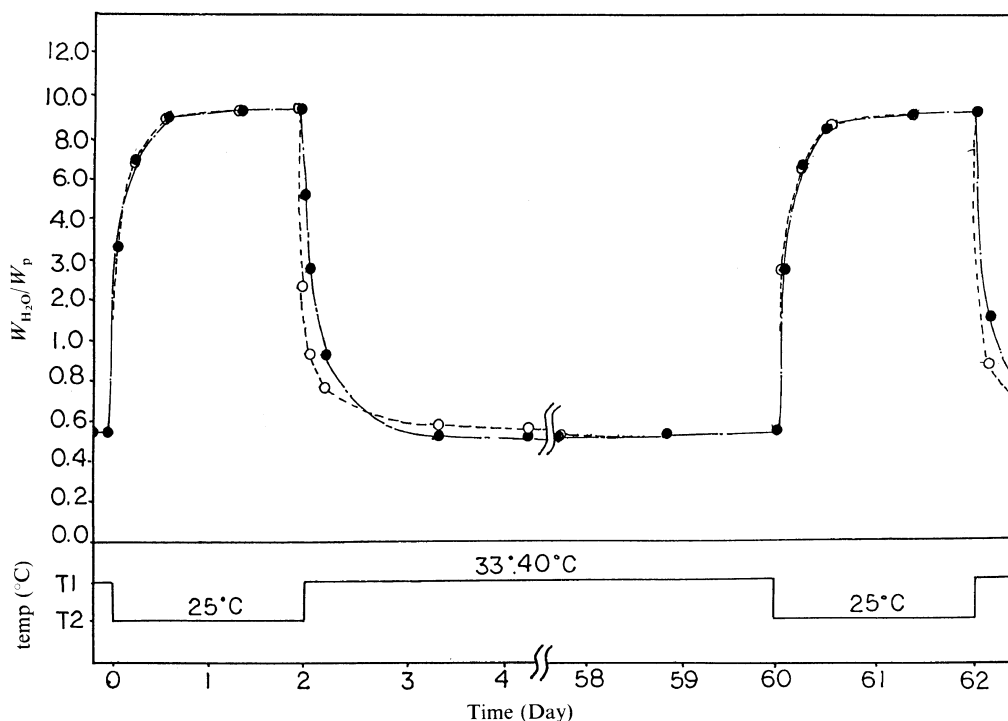
The observations of DSC and the above swelling studies indicate that each component in a swollen IPNs may be more phase-separated than in bulk IPNs.

#### Swelling Studies (swelling-deswelling kinetic studies)

The swelling-deswelling kinetics of crosslinked PIPAAm, 80/20 IPNs and 50/50 IPNs with temperature modulation between 25 and 33 or 40°C are shown in Figures 8, 9, and 10, respectively.

As seen in Figure 8, the swelling kinetics, after reaching equilibration of crosslinked PIPAAm at 33°C, took about two days to reach the next equilibrium at 25°C. Deswelling kinetics showed a large difference between modulation at 33 and 40°C. Deswelling kinetics at 40°C showed an extremely retarded pattern in reaching equilibrium. Deswelling kinetics took two days at 33°C and about 2 months at 40°C to attain the next equilibrium stage. These observations indicate that the deswelling kinetics of crosslinked PIPAAm





**Figure 8.** Swelling-deswelling kinetics of crosslinked PIPAAm by temperature modulation: a (---○---) 25–40°C; b (---●---) 25–33°C.

above its swelling transition temperature is strongly affected by temperature. Namely, deswelling kinetics at 33°C may be caused by a gentle squeezing effect due to the tender deswelling behavior of the gel surface which allows water to be expelled outside the gel. The deswelling kinetics at 40°C may be caused by a strong squeezing effect followed by dense structure formation which pretends to expel water from the deswollen gel.

In Figures 9 and 10, the swelling-deswelling kinetics of 80/20 IPNs and 50/50 IPNs demonstrated complicated behavior dependent on gel composition and temperature applied.

The swelling kinetics of 80/20 IPNs and 50/50 IPNs at 25°C required approximately the same time period as that for crosslinked PIPAAm. For deswelling at 33°C, it took about 2 weeks for the 80/20 IPNs and required about 1 month for 50/50 IPNs. At 40°C, in the deswelling kinetics of 50/50 IPNs and 80/20

IPNs, there was rapid squeezing in the initial step followed by a deswelling slower than the deswelling processes of 50/50 IPNs, 80/20 IPNs and crosslinked PIPAAm at 33°C. The deswelling rate was also affected by IPN composition. The deswelling rate for an intermediate time course became slower as PEO-PDMS-PEO content increased. Finally, these processes took approximately two months, like crosslinked PIPAAm, to attain equilibrium.

Overall, the deswelling kinetics of 80/20 IPNs was similar to those of crosslinked PIPAAm. The deswelling kinetics of 50/50 IPNs, however, showed a highly retarded pattern in the intermediate time course taken to reach equilibrium.

If the bulk morphology of IPN distinguished by TEM and DSC directly is correlated to individual phases for each swollen IPN, an interpretation of swelling-deswelling

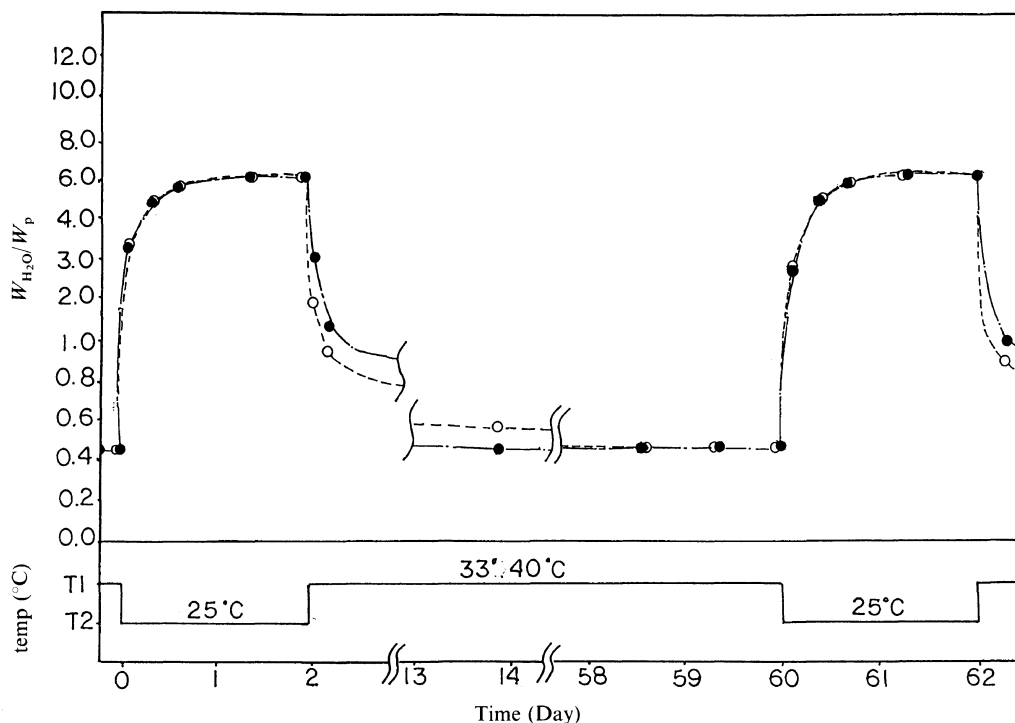


Figure 9. Swelling-deswelling kinetics of 80/20 IPNs by temperature modulation: a (---○---) 25—40°C; b (---●---) 25—33°C.

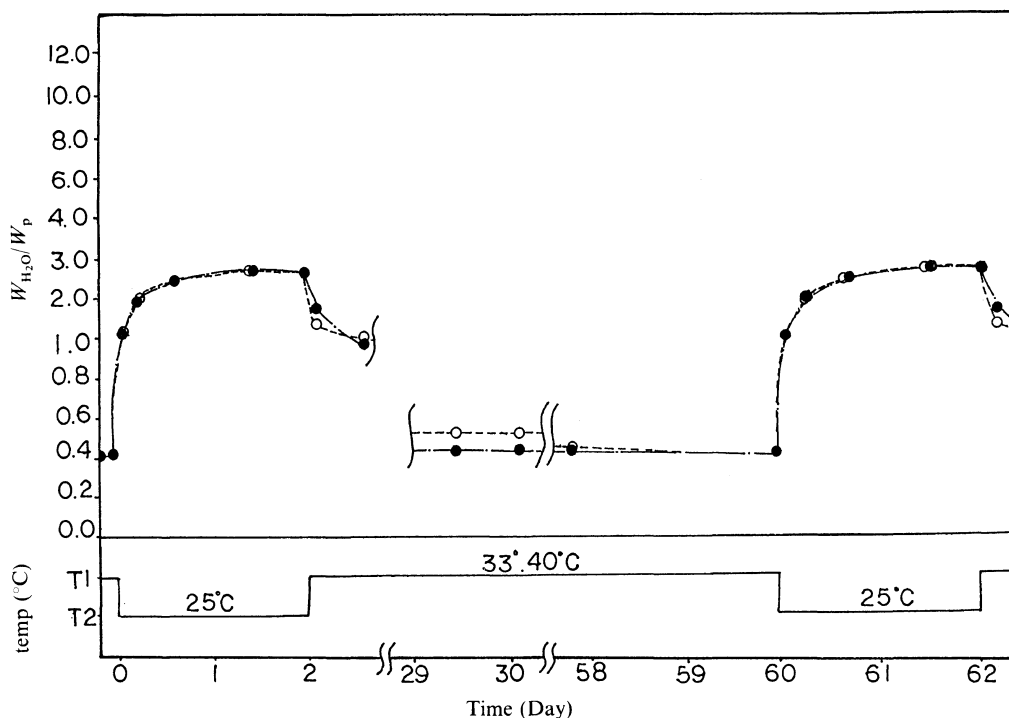
kinetics of IPNs may be possible. In the swollen state above the swelling transition temperature, morphology was assumed to resemble the dried bulk morphology.

Namely, in 50/50 IPNs, the PDMS segment content as HPB component was about 25%, if defined directly from feed composition. PDMS phase was influenced by phase mixing with HPL components. The HPL PEO segment in 50/50 IPNs formed the continuous phase. Therefore, in this swelling kinetic study, the HPB PDMS segment and phase continuity of the mixed PEO may provide a channel for water diffusion with temperature modulation at 25°C, from the equilibrium state at 33 and at 40°C if bulk morphology of 50/50 IPNs is correlated with its swollen morphology. If the pure PDMS segment forms a continuous phase or a concentric layer around or near PIPAAm domains in bulk 50/50 IPNs, the swelling kinetics should show a retarded pat-

tern due to the HPB character of the PDMS segment. As possible evidence, the appearance of 50/50 IPNs were slightly transparent or translucent at the equilibrium stage at 33 and 40°C. Even crosslinked PIPAAm appeared opaque at these temperatures.

The swelling kinetics of 80/20 IPNs may present the same pattern as that of crosslinked PIPAAm due to the phase continuity of PIPAAm in the dried 80/20 IPNs which was distinguished with TEM.

In deswelling kinetics, we again hypothesize that the morphology of each swollen IPN morphology shows a more highly phase separated structure than that of dried IPNs and has a phase relationship similar to that of dried IPNs. Namely, equilibrated swollen 50/50 IPNs at 25°C may consist of swollen PIPAAm domains, a concentric PDMS segment layer around the PIPAAm domain and a swollen PEO segment phase continuous structure to



**Figure 10.** Swelling-deswelling kinetics of 50/50 IPNs by temperature modulation: a (---○---) 25–40°C; b (---●---) 25–33°C.

explain the deswelling kinetics of 50/50 IPNs. Therefore, the phase separated PDMS segment may act as a barrier layer to prevent water from squeezing out, resulting in retarded deswelling kinetics. Similarly, the swollen 80/20 IPNs may also form a highly phase separated swollen structure which consists of a swollen PIPAAm continuous phase, slightly swollen PEO domains and a PDMS concentric layer surrounding PEO domains. The deswelling kinetics of swollen 80/20 IPNs showed a pattern similar to that of crosslinked PIPAAm with slightly retarded deswelling due to PEO and PDMS phases. In our other IPN gel study, a highly phase separated swollen structure was also recognized.<sup>15</sup>

## CONCLUSIONS

In conclusion, we synthesized PEO-PDMS-PEO/PIPAAm IPNs by simultaneous IPN po-

lymerization. From DSC, TEM, and swelling studies, it was suggested that the degree of swelling of IPNs can be controlled without changing the swelling transition temperature by IPN formulation and composition, although the bulk morphology showed some phase mixing in each component. The aqueous swelling-deswelling kinetics of IPNs was also controlled by gel compositions and temperature applied. The precise kinetic behavior may be interpreted by swollen IPN morphology hypothesized from bulk IPN morphology studies.

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