

Physical Properties of Poly(vinyl chloride)-Grafted *N*-Isopropylacrylamide Graft Copolymers and Corresponding Polyblends

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ABSTRACT: The physical properties of poly(vinyl chloride) (PVC) and poly(*N*-isopropylacrylamide) [poly(NIPAAm)] blend systems, and their corresponding graft copolymers such as PVC-*g*-NIPAAm, were investigated in this work. The compatible range for PVC-poly(NIPAAm) blend systems is less than 15 wt % poly(NIPAAm). The water absorbencies for the grafted films increase with increase in graft percentage. The water absorbencies for the blend systems increase with increase in poly(NIPAAm) content within the compatible range for the blends, but the absorbencies decrease when the amount of poly(NIPAAm) is more than the compatible range in the blend system. The tensile strengths for the graft copolymers are larger than the corresponding blends. © 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 76: 170–178, 2000

Key words: polyblend; poly(vinyl chloride); poly(*N*-isopropylacrylamide); graft copolymer

INTRODUCTION

Polymer blends are physical mixtures of structurally different polymers that interact through secondary forces with no covalent bonding.¹ The importance of blending has increased recently because their superior properties in relation to homopolymers can be compared to those of alloys over metals. The degree of compatibility usually determines the final properties of the blend. However, most polymer-polymer pairs are incompatible or have a low degree of compatibility.² Hence, in order to improve the compatibility of the polymer blend, the graft copolymerizations for poly(vinyl chloride) (PVC) and vinyl monomers were carried out using dehydrochlorination or γ -irradiation by many researchers in the past.^{3–20} For

example, Krishnan and Krishnan,⁶ Goldberg and Vahiaui,⁷ and Singh et al.⁸ investigated the grafting of *N*-(vinyl pyrrolidine), 2-hydroxyethyl methacrylate (HEMA), and methacrylic acid onto medical PVC sheets by UV or γ -irradiation, respectively. Mukherjee et al. and Mobasher et al.^{15,16} performed the dehydrochlorination of PVC in pyridine and then grafted it with styrene and 4-methacryloxy, 2,2-hydroxybenzophenone using benzoyl peroxide (BPO) or azo-*bis*-isobutyronitrile (AIBN) as initiator. At the same time, graft copolymers for PVC grafted with functional monomers [glycidyl methacrylate (GMA) and HEMA] were successfully prepared in previous articles.^{21–25}

The degree of compatibility, predicted using the reduced viscometric method and thermodynamic principles, for PVC-poly(GMA) and PVC-poly(HEMA) blend systems were investigated in a previous study.²⁴ The results showed that the compatible ranges for PVC-poly(GMA) and PVC-poly(HEMA) blend systems are located at greater than 76% PVC and 90% PVC, respectively.

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The graft copolymerization of *N*-isopropylacrylamide (NIPAAm) onto PVC was investigated in our previous article.²⁶ Assessment of the compatibility and mechanical properties and surface properties for the PVC–poly(NIPAAm) blend systems and for the corresponding graft copolymers is the main purpose of this article.

EXPERIMENTAL

Materials

PVC resin (S-70, $DP = 1070$) was supplied by Formosa Plastic Co. (Taiwan). *N*-isopropylacrylamide (NIPAAm) monomer was supplied by TCI (Japan). Benzoyl peroxide (BPO) was recrystallized by dissolving in chloroform at room temperature and then precipitated by methanol. Dioxane, tetrahydrofuran (THF), *n*-hexane, diethyl ether, cyclohexanone, and methanol were used as received.

Preparation of Linear Poly(NIPAAm)

Linear poly(NIPAAm) was synthesized in dioxane with BPO as a free radical initiator (1.13×10^{-2} mol of BPO/mol of monomer). Dried N_2 gas was bubbled through the solution for 20 min to remove dissolved oxygen. The solution was polymerized at 80°C for 3 h under a N_2 atmosphere. The synthesized polymers were recovered by precipitation in *n*-hexane (300 mL) and purified by dissolving in tetrahydrofuran (THF, 20 mL) and reprecipitation in diethyl ether (200 mL). The polymers were filtered and dried in a vacuum overnight.

Preparation of Polymer Films

About 0.5 g of the various polymers [PVC–poly(NIPAAm) and PVC-*g*-NIPAAm] was dissolved in 10 mL of THF. The solution was poured into a Petri dish of 9 cm diameter, which was placed in a 50°C oven for 24 h to remove the solvent. The film cast by this method is referred to as standard film.

Measurements of Mechanical Properties

An Instron Universal tester Model 1130 with a load cell of 5 kg was used to study the stress–strain behavior of the samples. A gauge length of 2 cm and a strain rate of 5 cm/min were used in this study. The measurements were performed at room temperature using solution casting to ob-

tain a film specimen. The specimen dimensions were 0.5 cm width, 6 cm length, and 0.5 mm thickness. The reported datum was the average of six experimental data.

Viscometric Measurements

Viscometric measurements were carried out with an Ubbelodhe viscometer (flow time 99.55 s for pure water) at $30 \pm 0.01^\circ\text{C}$. The polyblend samples were dissolved in THF solutions of varied compositions to yield stock solutions of approximately 1.0 g of polymer per 100 mL of solvent.

Viscosity data were calculated according to Huggins' equation

$$\frac{\eta_{sp}}{C} = [\eta] + k'[\eta]^2C \quad (1)$$

where η_{sp} is the specific viscosity, C is the concentration of polymer blend, $[\eta]$ is the intrinsic viscosity, and k' is Huggins' constant.

η_{sp}/C is plotted versus C , $[\eta]$ and k' are determined, respectively, from the intercept and the slope of the straight line.

DSC Measurements

Glass transition temperature (T_g) was determined by using a Du Pont 2000 differential scanning calorimeter. The sample was 8 mg of dried film. All samples were heated up to 150°C at heating rates of 20°C/min and kept 90 s at that temperature to remove the traces of solvent, then quenched to -30°C and kept at that temperature for 5 min, then reheated to 150°C at a heating rate of 20°C/min. This procedure is first scan. The procedure was repeated once. The T_g values adopted in this article were second scanning values.

Measurements of Water Absorbency

The polymeric film (1 cm width, 2 cm length, and 0.5 mm thickness) was used to test water absorbency. The measurement of water absorbency was performed by weighing the sample, which was then immersed into 50 mL water for 24 h. The absorbency was then calculated as follows

$$\text{Absorbency} = (W_w - W_d)/W_d \quad (2)$$

where W_w is the weight of wet film and W_d is the weight of dry film.

Table I The Characteristic Data of Polymers Used for ΔH_m

Sample	Solubility Parameter (δ)	Density (ρ)	Number-Average Molecular Weight (\bar{M}_n) ^c
PVC	9.45 ^a	1.44	65,000
Poly(NIPAAm)	10.14 ^b	1.28	26,000
THF	9.1 ^a	—	—
Vinyl chloride	—	—	62.5
NIPAAm	—	—	113.16

^a The data of δ was obtained from polymer handbook.

^b The data was obtained by Hoy's method.

^c \bar{M}_n was measured by gel permeation chromatography.

RESULTS AND DISCUSSION

Compatibility of Polymer Blend Predicted by Thermodynamic Principle

According to thermodynamic principles, any solution process is governed by the free-energy relationship.

$$\Delta G = \Delta H - T\Delta S \quad (3)$$

If a polymer dissolves spontaneously, the free energy of solution, ΔG , is negative. The entropy of solution, ΔS , invariably has a positive value arising from increased conformational mobility of the polymer chains. Hence, the magnitude of the enthalpy of solution, ΔH , determines the sign of ΔG . Clearly, for the polymer to dissolve (negative ΔG), ΔH must be small. As suggested by Gee,²⁷ the equation of heat of mixing for one polymer dissolved in an organic solvent could be expressed as follows

$$\Delta H = \{V_0(\delta_0 - \delta_1)^2(1 - v_0)^2\}^{1/2} \quad (4)$$

where V and δ are the molar volume and solubility parameter of the solvent 0 and the polymer 1, and the term $(1 - v_0)$ represents the volume fraction of the polymer. Eq. (4) was extended by Schneier²⁸ as eq. (5) to include the polymer-polymer pairs interactions

$$\Delta H_{\text{mix}} = \{V_1(\delta_1 - \delta_2)^2(1 - v_1)^2\}^{1/2} \quad (5)$$

where V_1 and v_1 are the molar volume and volume fraction, respectively, of polymer 1; the term $(1 - v_1)$ represents the volume fraction of polymer 2 in the mixture; and δ_1 and δ_2 are the solubility parameters of polymer 1 and polymer 2, respectively. Because there is a given weight, X , of poly-

mer in every mixture, $(X_1/M_1)/\rho_1 = V_1$ represents the molar volume fraction of polymer 1, where M and ρ are the molecular weight of an average monomer unit and density, respectively. The volume fraction, v_1 , of polymer 1 presented in the system is given by

$$v_1 = \frac{(X_1/M_1)/\rho_1}{(X_1/M_1)/\rho_1 + (X_2/M_2)/\rho_2} \quad (6)$$

and

$$1 - v_1 = \frac{X_2 M_1 \rho_1}{X_1 M_2 \rho_2 + X_2 M_1 \rho_1} \quad (7)$$

where $X_1 + X_2 = 1$. Substituting this value into eqs. (5) and (7), eq. (8) is obtained as follows:

$$\Delta H_{\text{mix}} = \left\{ X_1 M_1 \rho_1 (\delta_1 - \delta_2)^2 \times \left[\frac{X_2}{(1 - X_2) M_2 \rho_2 + (1 - X_1) M_1 \rho_1} \right]^2 \right\}^{1/2} \quad (8)$$

In order to use eq. (8), values are needed for the solubility parameter, density, and molecular weight of an average unit of the polymer. The validity of eq. (8) was tested using the data of Bohn,²⁹ who listed the polymer pairs under the heading of first and second components. When the polymer systems are compatible, the value of ΔH_{mix} lies in the range of from 1×10^{-3} to 10×10^{-3} cal/mol.

Hence, we can predict the range of compatibility of PVC-poly(NIPAAm) blend systems from the above description and some thermodynamic parameters. Table I lists the characteristic data of the solubility parameter (δ), density (ρ), and number-average molecular weight (\bar{M}_n) for PVC and

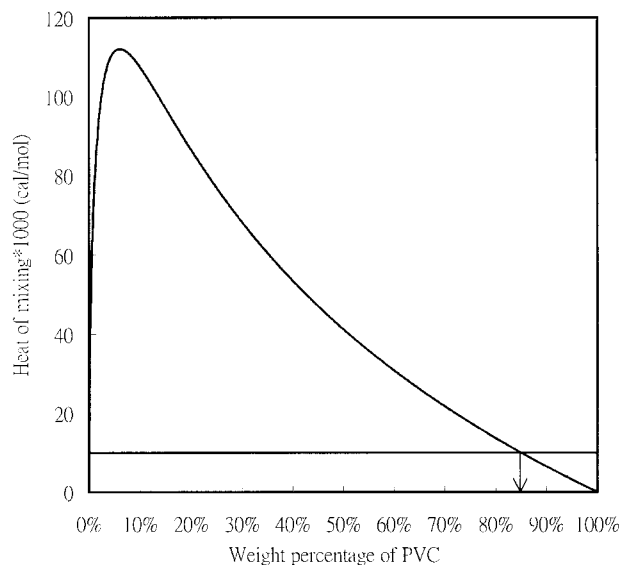


Figure 1 Heat of mixing versus percentage of PVC in PVC–poly(NIPAAm) polymer blends.

poly(NIPAAm). Figure 1 is the relationship of ΔH_{mix} and weight percentage of PVC for PVC–poly(NIPAAm). According to the above description, we know that the range of compatibility of PVC–poly(NIPAAm) blend systems is less than 15 wt % poly(NIPAAm) in the blend system.

Optical Microscopy Measurement

Figure 2 shows the microphotographs of the film by optical microscope (Nikon OPTIPHOT-100, magnification of 200). The pictures shown in Figure 2(a,b) show a homogeneous microstructure for the PVC–poly(NIPAAm) blend containing 5 wt % and 10 wt % poly(NIPAAm). But the picture shown in Figure 2(c) exhibits a significant phase separation domain for the PVC–20 wt % poly(NIPAAm) blend. Hence, the PVC–poly(NIPAAm) blend system can be confirmed to be thoroughly compatible within the range of 10 wt % to 20 wt % poly(NIPAAm). On the other hand, the films for 5, 10, and 20 wt % poly(NIPAAm) blend systems do not show any pores in microscopic observation. But when the composition of poly(NIPAAm) is more than 20 wt %, the film conspicuously appears to have many pores [see Fig. 2(d–f)].

The microphotograph for the grafted films shows no concavities (or pores) even when the grafted percentage is more than 16 wt % poly(NIPAAm)[see Fig. 2(g–i)]. This is because the NIPAAm is grafted onto PVC by chemical reaction. Comparing microphotographs in Figure 2,

the blend film appears to have pores during the range of incompatibility, but the grafted film does not so appear.

Reduced Viscosity Measurement

In order to check the compatible range obtained from the thermodynamic method and microscopy measurement, the reduced viscosity measurement technique was adopted to elucidate the compatibility of the polymer blends. The reduced viscosity method was presented in a previous report by the authors,²⁴ which noted that a linear parallel relationship was obtained for compatible blend systems [poly(GMA)–PVC]—for example, the slope of the respective blend composition in the plot of reduced viscosity (η_{sp}/C) versus polymer concentration (C) has the same value.

From the molecular-level viewpoint, the compatibility of a polymer blend is dependent on the molecular interaction between the two polymers.

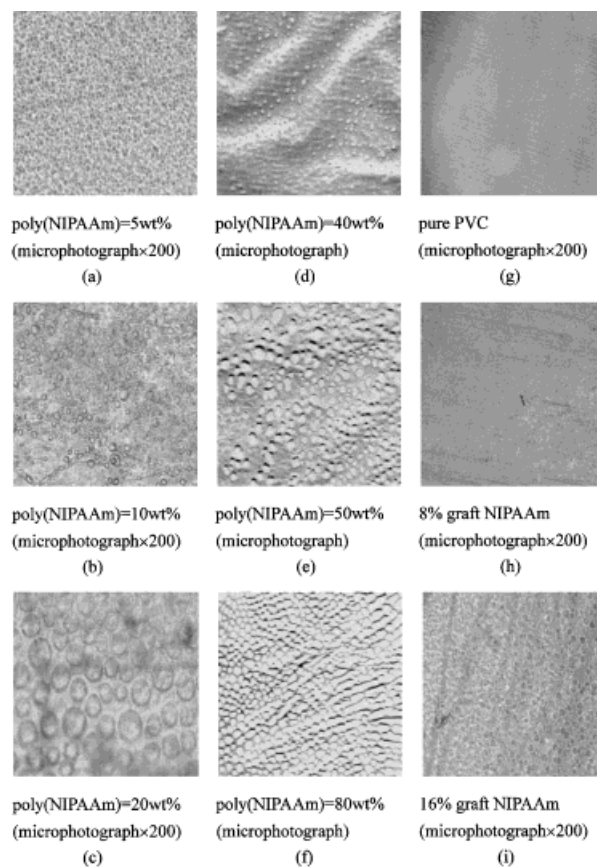


Figure 2 Microphotograph of various percentages of poly(NIPAAm) in PVC–poly(NIPAAm) blend system and PVC-g-NIPAAm copolymer.

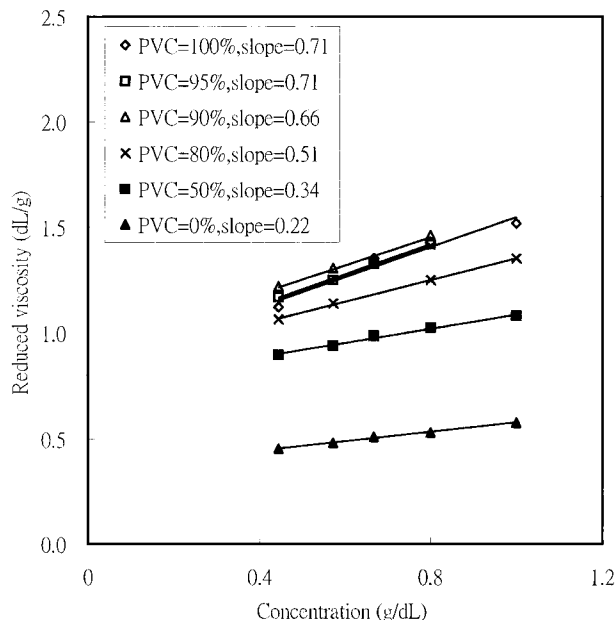


Figure 3 Plot of reduced viscosity versus concentration for PVC–poly(NIPAAm) polymer blend.

These molecular interactions would be reflected in the viscosity of the dilute solution. Hence, if the blend is a compatible system and the system has no molecular interaction in solution, then the slope of the plot of η_{sp}/C versus C for the blend should have an additivity, and if the blend is an incompatible system, then the mutual attraction force or repulsion force between the polymers would make the sum of slopes in the plot of η_{sp}/C versus C for a blend be a positive or negative deviation from linear additivity.

Therefore, here we apply the concept of the additivity law of the slope in a plot of the η_{sp}/C versus C for the polymer blend, to judge the compatibility of the polymer blends. Figure 3 shows, respectively, the relationship of the additivity of the slope in the plot of η_{sp}/C versus C for the polyblends of PVC–poly(NIPAAm) between theoretical and experimental values. The additivity law of the slope (theoretical value, S_{the}) for the plot of the η_{sp}/C versus C in a blend can be simply given by

$$S_{\text{the}} = X_1 S_1 + X_2 S_2 \quad (9)$$

where X_1 and X_2 are the weight percentages of homopolymer 1 and homopolymer 2, respectively; and S_1 and S_2 are the slopes for the plot of η_{sp}/C versus C for homopolymer 1 and homopolymer 2, respectively.

The results shown in Figures 4 indicate that the compatibility of the polymer pairs depends on the additivity of the slope in the plot of η_{sp}/C versus C for the respective polymer. At the same time, it was found that the tendency of the deviation (D) of slopes deviated from the additivity law in a way that was similar to the tendency of the change of heat of mixing (ΔH_m). Hence, the deviation of the slope from the additivity law as well as ΔH_m s for respective polymer pairs against the composition of various polyblends are plotted in Figure 5 for PVC–poly(NIPAAm) blend systems. Restated, this D value can be used to judge the compatibility of various polymer blends.

The D value for a blend can be given as follows

$$D(\%) = \frac{|S_{\text{the}} - S_{\text{exp}}|}{|S_{\text{the}}|} \times 100\% \quad (10)$$

where S_{the} is a theoretical slope obtained from the additivity law of slope in the plot of η_{sp}/C versus C for two homopolymers according to eq. (9). S_{exp} is a slope obtained from the experiment according to Huggins' equation.

According to a previous report by the authors,²⁴ the D value of the compatible range for a polyblend is lower than 15%. Applying this result, it can be inferred from Figure 5 that the com-

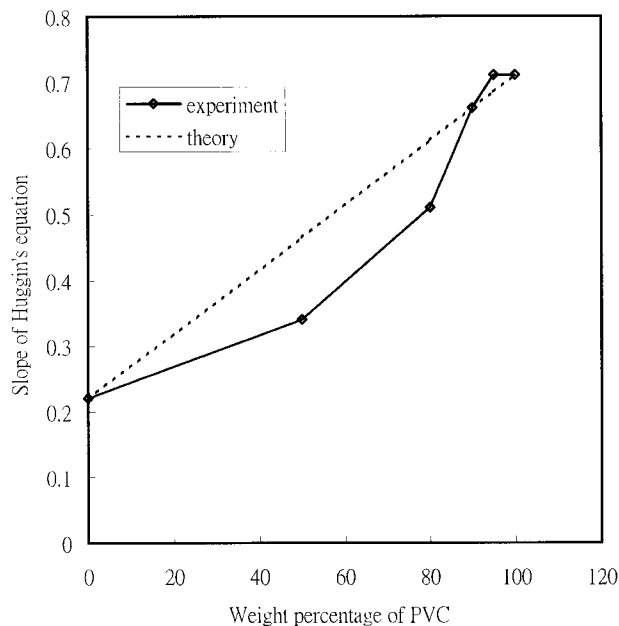


Figure 4 Comparison of additivity of slope between theory and experiment based on Huggins' equation in PVC–poly(NIPAAm) polymer blend.

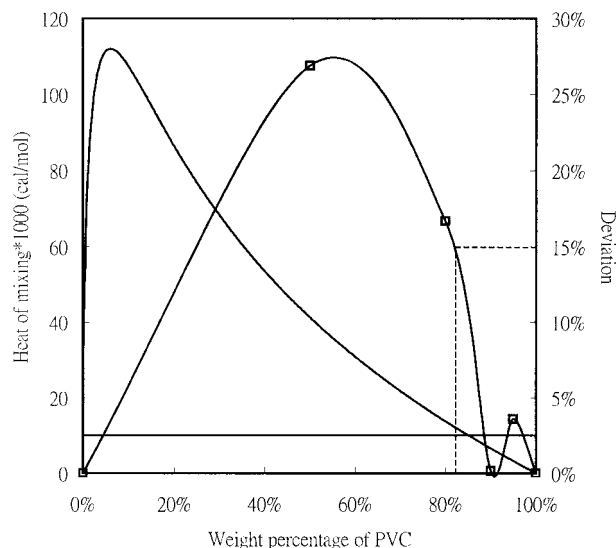


Figure 5 Comparison between deviation of additivity of reduced viscosity and heat of mixing in PVC/poly-(NIPAAm) polymer blend.

pletely compatible range is up to 18 wt % poly-(NIPAAm) for PVC–poly(NIPAAm) blend systems. The result conforms to the microscopic observation and the thermodynamic principle.

DSC MEASUREMENT (T_g TEST)

To judge the compatibility of a blend, we can observe the glass transition temperature (T_g) measured by differential scanning calorimetry (DSC). Because there is no molecular interaction or repulsion in a compatible system, the macromolecules of blend will sufficiently disperse each

other, and no phase separation occurs in the polyblend. Hence, only one T_g is observed. However, the incompatible blend would show two T_g s. The data shown in Table II for the PVC–poly-(NIPAAm) blend system only exhibit one T_g in various compositions. This result shows that the compatible range is below 20 wt % poly(NIPAAm) in the blend system. The T_g values are shown in Table II calculated using the Fox equation

$$\frac{1}{T_g} = \frac{X_1}{T_{g1}} + \frac{X_2}{T_{g2}} \quad (11)$$

where X_i is the weight fraction of polymer i) are approximately coincident with those analyzed by DSC. A single value of T_g for each polyblend provided the evidence of complete compatibility of the polymer mixture.

The data shown in Table II for PVC-*g*-NIPAAm system also exhibit only one T_g in various compositions. The range of T_g for the graft is 354~365 K. The T_g s increase with the increasing of the NIPAAm amount in the graft copolymers.

Comparing the graft system with the blend system, we find that the values of T_g for the PVC-*g*-NIPAAm copolymer are lower than those for the PVC–poly(NIPAAm) blend system.

Hydrophilicity of the Grafted Copolymers and Polymer Blends

Contact Angle Measurement

Because of hydrophilic groups (—CONH—) existing in NIPAAm, the grafted copolymer and the polymer blend will improve the hydrophilic abil-

Table II T_g s of PVC, PVC–poly(NIPAAm) Blend, PVC-*g*-NIPAAm, and Poly(NIPAAm)

Sample	NIPAAm (%)	T_g (K)	T_g (Fox) (K)
Pure PVC	0%	354	354
	5 wt %	355	356
PVC–poly(NIPAAm)	10 wt %	357	357
	15 wt %	362	359
	20 wt %	372	361
	8%	354	356
PVC- <i>g</i> -NIPAAm	12%	355	358
	16%	358	359
	18%	363	360
	20%	365	361
Poly(NIPAAm)	100%	408	408

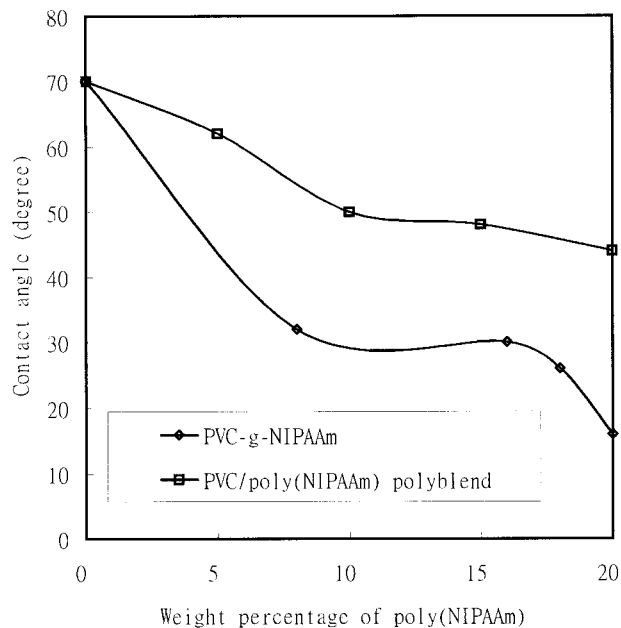


Figure 6 Comparison of contact angle between PVC–poly(NIPAAm) blends and PVC-*g*-NIPAAm grafts containing various weight percentages of NIPAAm.

ity of PVC. The improvement of hydrophilicity for PVC can be detected by contact angle between polymer film and water drop. When the hydrophilic ability of the polymer films increases, the interface tension between film and water drop increases; then the water easily permeates into the film, and the contact angle between film and water drop decreases. The results of contact angle measurements for various PVC–poly(NIPAAm) polymer blends and PVC-*g*-NIPAAm grafted copolymers are shown in Figure 6. The contact angles for PVC-*g*-NIPAAm grafted copolymers are all smaller than those for PVC–poly(NIPAAm) blends. These results show that the improvement of hydrophilicity for the grafted copolymer containing a hydrophilic side chain is better than that for the polymer blends in which hydrophilic polymer is dispersed into the PVC.

Water Absorbency Measurement

The water absorbency of sample films immersed into 30 mL water for 1 day is shown in Table III. The results indicate that the water absorbency for all grafted copolymers increases with an increase in grafted percentage of NIPAAm. This result explicitly indicates that the hydrophilic group of NIPAAm would affect the water absorbency for graft copolymer. The water absorbencies for

PVC–poly(NIPAAm) blend films increase with an increase in the poly(NIPAAm) content until 15 wt % (the compatible range). But the absorbencies for blend film decrease with further increasing of the poly(NIPAAm) in the blend system (incompatible range) when the amount of poly(NIPAAm) in the blend system is above 15 wt %. This is because the poly(NIPAAm) could dissolve into water. The dissolved amount of poly(NIPAAm) increases with the increasing of amount of poly(NIPAAm) in the blend, so the absorbency of blend film decreases with an increase of the poly(NIPAAm) content. Comparing both films, the absorbency of grafted film is higher than that of blend film.

Figure 7 show microphotographs of the cast films (which were firstly immersed into water and then removed) by optical microscopy (200 \times). There are some concavities and grooves in these films for every blend. Comparing the Figure 2 with Figure 7, it can be seen that the number of concavities for the water-immersed blend film is more than that of the standard blend film. This phenomenon proves that the poly(NIPAAm) in blend film would be dissolved into water. The result shown in Figure 7 can also explain why the absorbencies show a negative trend when the amount of poly(NIPAAm) is more than 20 wt %.

Mechanical Properties Measurement

Figure 8 show the comparison of the tensile strength for PVC-*g*-NIPAAm, and PVC–NIPAAm blends at various weight percentages of NIPAAm. The results shown in Figure 8 indicate that the tensile strength for the PVC-*g*-NIPAAm graft co-

Table III Water Absorbencies for PVC–poly(NIPAAm) Films

Sample	NIPAAm (%)	Absorbency (%)
Pure PVC	0%	0.00% \pm 0.0%
	5%	0.53% \pm 0.2%
	10%	0.98% \pm 0.6%
	15%	1.68% \pm 0.6%
	20%	0.12% \pm 0.4%
PVC–poly(NIPAAm)	30%	–4.98% \pm 2.0%
	40%	–13.25% \pm 2.5%
	50%	–23.12% \pm 0.5%
	8%	0.67% \pm 0.1%
	PVC- <i>g</i> -NIPAAm	16%
18%		2.39% \pm 0.1%
20%		3.43% \pm 0.1%

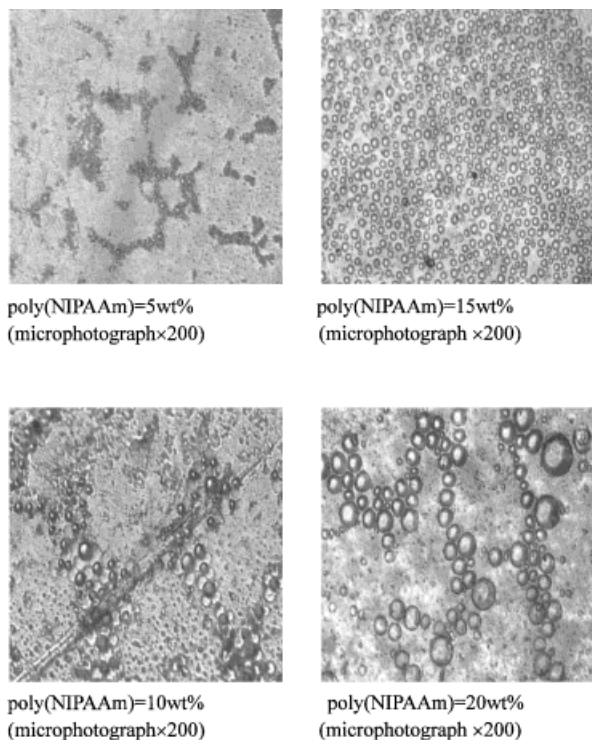


Figure 7 Microphotograph of various weight percentages of poly(NIPAAm) in PVC–poly(NIPAAm) blend system (water-immersed).

polymers are higher than those for PVC–poly(NIPAAm) blends. This result may be due to the compatibility of polymer–polymer pairs. For polymer blends the mechanical properties are profoundly affected by the properties of the blend constituents and the degree of compatibility of the polymer blends, but usually the latter is a major factor in determining the final properties of a blend. Most polymer–polymer pairs are incompatible or have narrow compatible ranges in their blends, so the polymer blends are usually replaced with a grafted copolymer to improve physical properties. The tensile strength is weaker for blends than for grafted copolymer for a narrow compatibility range system [PVC–poly(NIPAAm) pairs]. From the above results it can be concluded that because the compatible range is narrow for the polymer pair, the tensile strength for the graft copolymer is higher.

Effect of Water-Soaking on Mechanical Properties for PVC–poly(NIPAAm) Blend Films

After the film was soaked in water for 24 h and then dried in an oven overnight, the dried sample

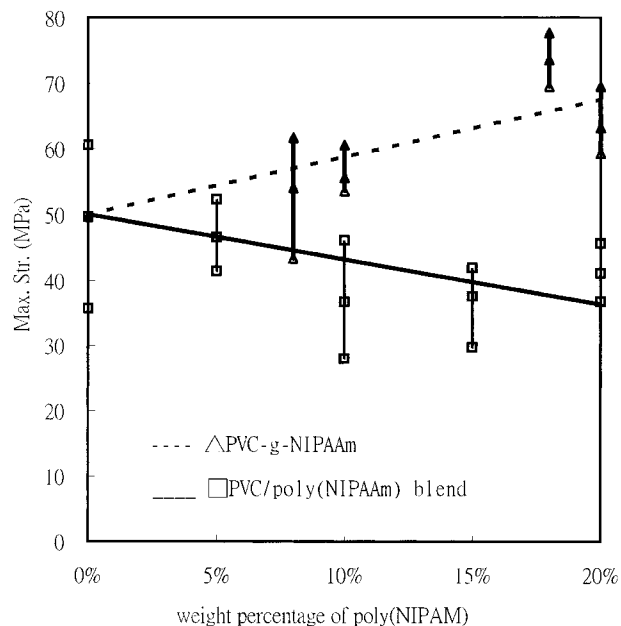


Figure 8 Comparison of tensile strength between PVC–poly(NIPAAm) blends and PVC-g-NIPAAm grafts containing various weight percentages of NIPAAm.

films were tested for their mechanical properties. The results for tensile strength, shown in Figure 9, indicate that tensile strength decreases at

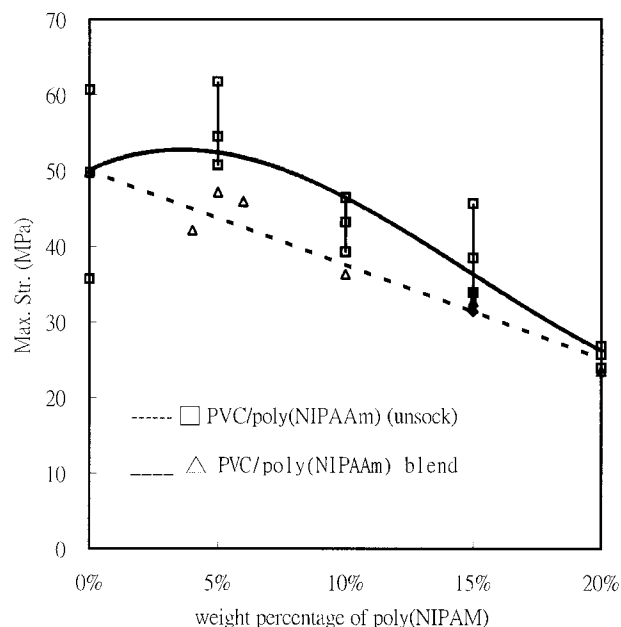


Figure 9 The comparison of tensile strength at various weight percentages of poly(NIPAAm) for PVC–poly(NIPAAm) blend after soaking in water.

higher poly(NIPAAm) percentages and that the strengths for water-soaked films are lower than for standard films. This indicates that the structure of the polyblend was destroyed because the NIPAAm component was dissolved into water.

CONCLUSION

The compatible range predicted by thermodynamic principle is 15 wt % poly(NIPAAm) in the PVC–poly(NIPAAm) blend system. This result conforms to the reduced viscosity method, microphotographic observation, and the T_g test. The water absorbency for grafted film increases with increasing grafted percentage. The absorbencies for blend systems increase with increasing blend percentage within the compatible range. But when the amount of poly(NIPAAm) is more than 15 wt %, the absorbencies decrease with an increase in the poly(NIPAAm) content. The tensile strengths for the graft (PVC-*g*-NIPAAm) are greater than those for the corresponding blends. The tensile strengths for the blends after soaking in water are lower than those for the original blends.

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