

Morphology and Mechanical Properties of Nylon 6 Toughened with Waste Poly(vinyl butyral) Film

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ABSTRACT: Phase morphology and mechanical properties of the blends of Nylon 6 with scrap poly(vinyl butyral) (PVB) film and poly[styrene-*block*-(ethylene-*co*-butene)-*block*-styrene] (SEBS) have been investigated. Scanning electron microscopic photographs revealed that the spherical PVB particles are finely and uniformly dispersed in the Nylon 6 matrix without changing the shape of the particles. The average particle sizes in all over the blend compositions for Nylon 6/PVB were slightly increased with PVB content, but the dispersed phase is tightly adhered to the matrix phase, with PVB content in the range of 20–35 wt % PVB. Elongation at break and notched Izod impact strength of all the blends were enhanced, which implies good interfacial adhesion. The rubberlike PVB film adhering to the Nylon 6 phase is suggested to give an improved impact strength and toughness. In particular, the optimum PVB content for the best impact strength is found to be in the vicinity of 20–35 wt %, and this composition exhibits better moisture resistance than the other blend compositions. All of the blends up to 35 wt % PVB show higher mechanical properties than those of Nylon 6 blended with conventional impact modifier SEBS. Thus, plasticized PVB film, which is recycled from the process of automobile safety glasses, is applicable as an impact modifier or a toughening agent of Nylon 6. © 1998 John Wiley & Sons, Inc. *J Appl Polym Sci* **67**: 1531–1540, 1998

Key words: morphology, mechanical properties, nylon 6, waste PVB film, toughening agent, impact modifier

INTRODUCTION

There is an intensive commercial interest in multiphase polymer blends or alloys, because of the potential opportunities for combining the attractive features of each material or improving the deficient characteristics of a particular material.^{1,2}

Nylons are an attractive class of polymers for engineering plastics, but they have a number of deficiencies for certain applications (e.g., brittleness, high moisture sorption, poor dimensional stability,

or marginal heat deflection temperatures). Part of the deficiencies is able to improve by blending with other plastics [such as poly(phenylene oxide),^{3,4} ABS,^{5,6} poly(olefine),^{7,8} or polycarbonate⁹] and thermoplastic elastomers.^{10,11}

Enhancement in toughness of rigid polymers by blending with suitable elastomeric components is widely practiced and continues to be of considerable scientific interest.^{12,13} There are various factors in influencing the toughness and diversity of opinion about the mechanism.^{14–16} The size of the rubber particle is one of the factors influencing the morphology of the blends. For example, it is generally recognized that rubber particles must be larger than 1 μm for toughening polystyrene,^{17,18} whereas they could be smaller than 1

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$\mu\text{m}^{19,20}$ for Nylon 6 or Nylon 66 in general. Wu¹⁴ proposed that the distance between the particles, rather than the particle size, would be the most fundamental parameter controlling the toughness of nylon. It is also recognized that some degree of adhesion or coupling between the elastomeric particles and matrix phases must exist. However, hydrocarbon elastomers do not have sufficient affinity for polymers such as nylon. This problem has been solved by incorporating functional groups into the elastomer, which can be reacted with nylon.^{14,15,19,20} In general, the impact properties of thermoplastics can be considerably enhanced by the incorporation of a dispersed elastomeric phase, although the toughening mechanism is influenced by the properties of the matrix and morphology of the blends. For example, in brittle polymers (such as polystyrene^{21,22}), rubber particles promote crazing of the matrix, whereas in semiductile polymers (such as polyamides^{19,23}) shear yielding is an usual factor in absorbing impact energy.

Recently, we have been interested in recycling the plasticized poly(vinyl butyral) (PVB) film that is collected in the course of production of automobile safety glasses. Because PVB film is very expensive, it is necessary for the scrapped film ($\sim 20\%$) to be used for the other purposes. Thus, suitable use of it has been suggested to save materials and recover the loss of waste polymer.

In this study, the possibility of the use of recycled PVB as a toughening agent or an impact modifier into Nylon 6 has been studied for replacing the role of poly[styrene-*block*-(ethylene-*co*-butene)-*block*-styrene] (SEBS) in nylon. Morphological behavior and mechanical properties have been investigated to understand the relationship between Nylon 6 matrix and PVB. These characteristics were compared with the system of Nylon 6 toughened with SEBS.

Theoretical Background

The Kerner model²⁴ relies on the assumption of perfect phase coupling, which means that the deformation applied to the matrix is transferred completely across the phase boundary. The Kerner model was originally developed for the shear modulus of a composite consisting of particle phase and polymer matrix, but the model may be useful to predict the tensile modulus of a certain class of heterogeneous blend system. The Kerner model may be written as

$$\frac{E_b}{E_m} = \frac{\varphi_m E_m + (\alpha + \varphi_d) E_d}{(1 + \alpha \varphi_d) E_m + \alpha \varphi_d E_d} \quad (1)$$

for inclusions of spherical geometry and random packing. Here, E is the tensile modulus, and φ is the volume fraction of the discrete phase. The subscripts b , m , and d refer to the blend, matrix, and disperse phase, respectively. The coupling parameter α is determined by Poisson's ratio ν of the matrix, as denoted in eq. (2)

$$\alpha = \frac{2(4 - 5\nu)}{(7 - 5\nu)}. \quad (2)$$

According to Kunori and Geil²⁵ and Nielsen,²⁶ the tensile failure of a blend is attributable to the failure of the adhesion between the disperse and continuous phases through crazing or a dewetting effect. If there is little or no adhesive force existing between the components, the tensile strength of the blend primarily depends on the continuous phase; therefore, it will reach a minimum value. Under the condition of no adhesive force between the components, the tensile strength of the blend may be represented by

$$\sigma_b = \sigma_m(1 - A_d) \quad (3)$$

where σ_b and σ_m are the tensile strength of the blend and matrix, respectively, and A_d is the fraction of the area occupied by the discrete phase in the cross section of the specimen. On the other hand, when a strong adhesive force exists between the components, the disperse phase contributes to the tensile strength of the blend, and eq. (3) may be modified as

$$\sigma_b = \sigma_m(1 - A_d) + \sigma_d A_d \quad (4)$$

in which σ_d denotes the tensile strength of the disperse phase.

According to Kunori and Geil,²⁵ when the tensile fracture loci propagate mainly through the interface, then A_d in eq. (4) varies with two-thirds power of the volume fraction (φ_d). Therefore, tensile strength of the blend may be described by

$$\sigma_b = \sigma_m(1 - \varphi_d^{2/3}) + \sigma_d \varphi_d^{2/3}. \quad (5)$$

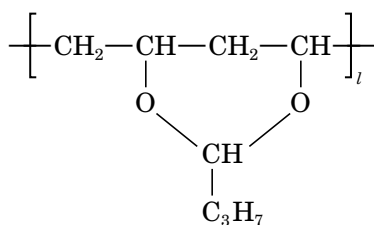
EXPERIMENTAL

Materials

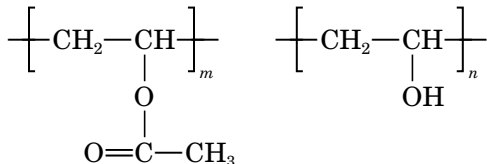
All of the materials used in this article are supplied by commercial sources. Nylon 6 is a hy-

drolitic poly(ϵ -caprolactam), of which the brand name is Toplamid 1033T supplied by Tongyang Nylon Co. Ltd. of Korea; the specific gravity is 1.14; and the melting point is 220°C.

PVB is a copolymer of vinyl butyral-*co*-vinyl alcohol-*co*-vinyl acetate as the following chemical structure where the glass transition is 70–85°C, depending on the copolymer composition. The PVB film contains ~ 28 wt % of plasticizer, whose chemical name is tetraethylene glycol diheptanoate (commercial name is 4G7). It has a molecular weight of ~ 180,000–270,000, a specific gravity of 1.07, and is supplied by Dupont Chemical Co. Ltd. under the commercial name of Butacite. The scrap of PVB used in this study is excess sheet trimmed from the edge of the laminate during the production of safety glasses in a clean room. PVB is prepared by a condensation reaction using poly-(vinyl alcohol) and butyraldehyde, and has the following chemical structure:



Poly(vinyl acetal)



Poly(vinyl acetate) Poly(vinyl alcohol)

here, $l = 76\text{--}79$, $m = 0\text{--}1$, and $n = 20\text{--}23$ wt %.

The triblock copolymer of SEBS incorporated into Nylon 6 has styrene end blocks and a hydrogenated butadiene midblock resembling an ethylene/butene copolymer. SEBS is Kraton G 1652, a commercial product of Shell Chemical Co. Molecular weight of polystyrene end-block is 7,500, and styrene content is 29% by weight.²⁷ Before every step of processing, all of the materials containing any Nylon 6 were dried for at least 24 h at 80°C in a vacuum oven to reduce moisture effect in the course of this study.

Sample Preparation

Scrapped PVB film was chopped in a high-speed blender at low temperature. Blends of Nylon 6/

PVB and Nylon 6/SEBS were prepared by melt blending in a compact mixing machine developed in our laboratory. An appropriate amount of SEBS, PVB, and Nylon 6 was mixed with a rotor speed of 60 rpm at 250°C for 15 min. A blended sample was then injected into a mold placed just below the machine. Plasticizer (28 wt %) was reduced to 7–12 wt % during the process of blend sample preparation at 250°C.

Analysis

The Jeol scanning electron microscope operated at 20 kV was used to characterize the particle size of the disperse phase and its distribution in the blend. All of the specimens were perpendicularly fractured to the stretching direction in liquid nitrogen; then, the fractured surface was sputtered with gold using a Sputter Coater for enhanced conductivity.

Stress-strain experiments were conducted on a universal testing machine of United Calibration Co. at room temperature, following the procedure described in ASTM D 638. A cross-head speed of 5 mm min⁻¹ was used for all of the measurements. For the tensile properties of wetted specimens, the specimens dried at 80°C for 24 h in a vacuum oven were immersed in distilled water at 20 ± 0.2°C for 24 h. Tensile properties of the wetted specimens were measured after water was wiped off the water of them with a cloth.

The specimens for notched Izod impact strength tests have a dimension of 52.5 × 12.7 × 3 mm, with a notch 2.5 mm in radius. The impact strength of the blends was measured using the impact testing machine of Yasuda Seiki Co. at room temperature in accordance with ASTM D 256.

RESULTS AND DISCUSSION

Dynamic Mechanical Behavior

Before disclosing our results and discussion for the morphological and mechanical investigations, it is worthwhile to introduce some preliminary study on the materials used in the course of this work.

Figure 1 depicts the DMTA dependences in bending E' [Fig. 1(A)] and in bending E'' and $\tan \delta$ [Fig. 1(B)], respectively. In Figure 1(A), three curves (a–c) indicating the PVB-related materials represent the variance of E' from 10⁹ Pa to 10⁶ Pa, implying a conventional amorphous material,

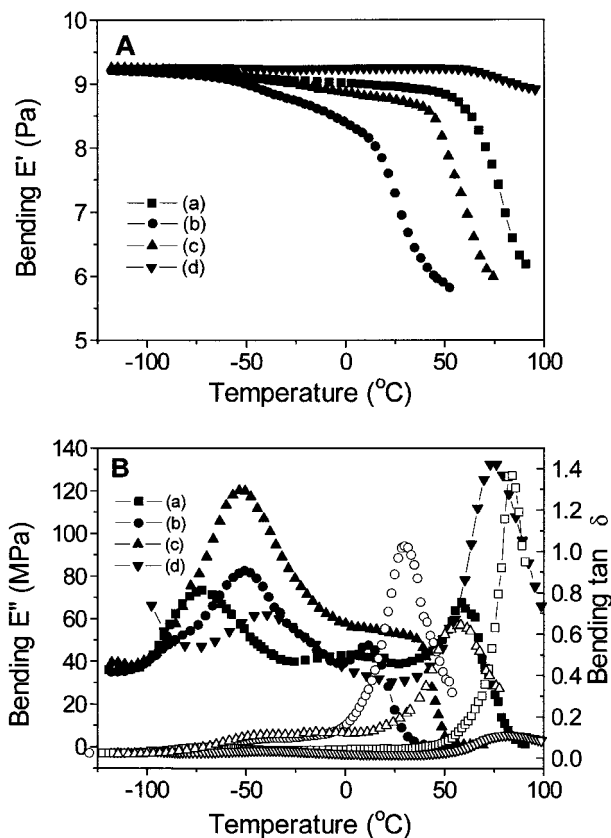


Figure 1 (A) DMTA dependences in E' : (a) PVB resin, (b) recycled PVB-molded film at 80°C, (c) recycled PVB film molded at 250°C, and (d) Nylon 6. (B) DMTA dependences in E'' and $\tan \delta$: all of the curves are the same as in (A).

whereas curve d shows a typical crystalline material. In Figure 1(B), considerable differences in thermal characteristics in the α and β relaxations between various PVB samples are observed. In each figure, curves a–d represent the DMTA dependences of virgin PVB resin, recycled PVB film (contains 28 wt % plasticizer), recycled PVB film after compression molded at 250°C (contains 12 wt % plasticizer), and Nylon 6, respectively. Specimens for curves a and b are compression-molded at 80°C and for curves c and d are at 250°C. In Figure 1(B), because β relaxation in $\tan \delta$ (with open symbol) shows a small and a broad peak, this region is also represented in E'' (with closed symbol). Curve a describing the virgin PVB resin shows two transitions in $\tan \delta$: the higher one at 84°C represents the glass transition temperature and the weak and lower one at -65°C is designated to the β transition. Here, two transitions in E'' are more obvious: the T_g and the T_β are at 59 and -72°C, respectively. Because PVB is a copoly-

mer of vinyl butyral, vinyl alcohol, and vinyl acetate, broad relaxations at low temperatures have been observed. It is interesting to analyze curves a and b in Figure 1(B) carefully. In curve b, recycled PVB film shows the glass transition temperature at 29°C in $\tan \delta$ and β relaxations at -51 to -52°C in E'' . The difference between curves a and b is the amount of existing plasticizer in curve b. The T_g of PVB is reduced to 29°C from 84°C by incorporation of a 28 wt % plasticizer. However curve c, which represents the recycled PVB film after being compression-molded at 250°C, shows the T_g at 59°C and the T_β at -53°C in $\tan \delta$. Compared with curve b, there is no difference in T_β , whereas T_g is increased $\sim 30^\circ$ due to the reduced plasticizer up to 16–20 wt % during the sample preparation. Curve d also shows two relaxations in $\tan \delta$: the higher one at 74°C, which is related to the melting behavior of the crystalline structure (this would be the softening temperature of Nylon 6); and the lower one at -39 to -41°C would be the β relaxation, which rises from the existence of moisture. All of the specimens we have used are all moisture-sensitive materials; thus, it is difficult to expect the reproducible data throughout the experiments.

For the various Nylon 6/PVB blend compositions, we have obtained three major transitions: the highest at 81–83°C in $\tan \delta$, the middle transition at 21–43°C in E'' , and the lowest at -35 to -43°C in E'' . We believe that the highest transition at $\sim 80^\circ\text{C}$ would be the softening temperature of Nylon 6. The transition around 21–43°C would give rise to the glass transition of PVB film affected by the small amount of moisture and plasticizer. The lowest transition may be interpreted as a combined peak of the β transition between Nylon 6 and PVB.

As a result of thermal properties in the blend of Nylon 6/PVB, no evidence of miscibility was observed. Thus, morphological and mechanical properties of immiscible Nylon 6/PVB have been discussed as follows.

Morphology

Scanning electron microscopic (SEM) photographs of the fractured surface of Nylon 6 blended with 10–30 wt % SEBS are shown in Figure 2. The SEBS particles are large and have been segregated into spherical domains. No morphological evidence of enhancement or good adhesion at the interface between the matrix and the disperse phase has been seen. In particular, many domains

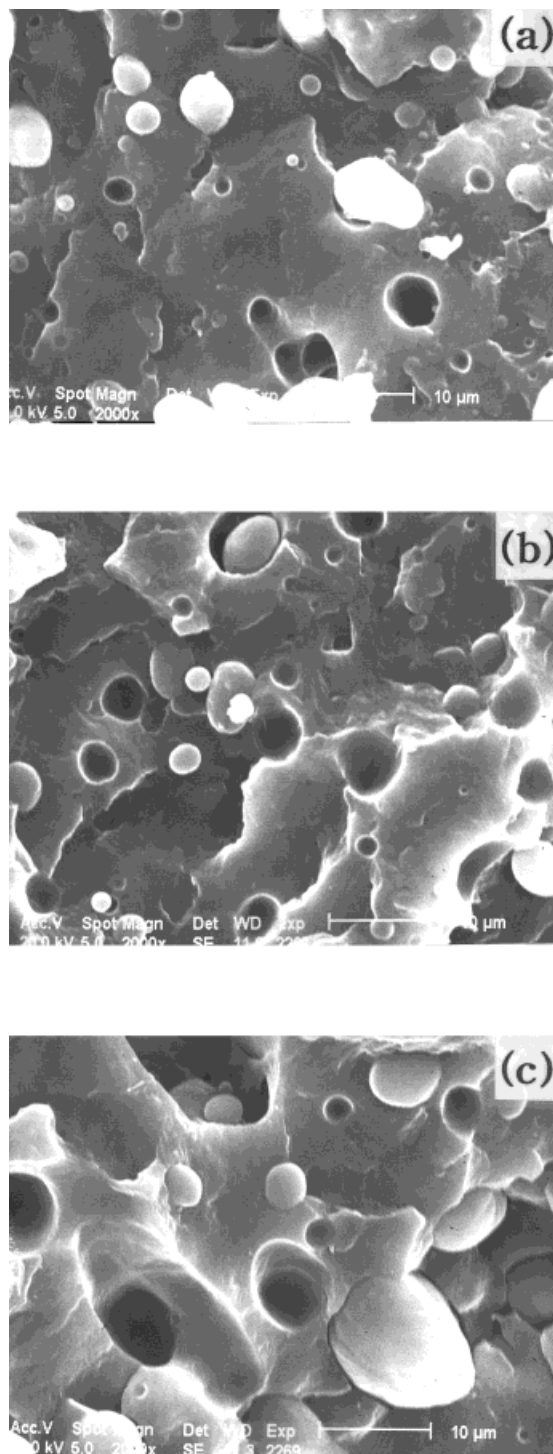


Figure 2 SEM photographs of fractured surface of Nylon 6/SEBS blends: (a) 90/10, (b) 80/20, and (c) 70/30 wt %.

of the SEBS have been pulled away from the matrix during the fracture process and then remained as empty holes. The measured maximum particle size of 90/10 wt % blend [Fig. 2(a)] is

$\sim 5 \mu\text{m}$ in diameter; however, it is gradually increased from 5 to 9 μm by incorporation of SEBS up to 30 wt %.

Figure 3 shows that the morphology of the Nylon 6/PVB blends varies with incorporation of PVB. At glance in this figure, which shows the same magnification as Figure 2, relatively smaller spherical particles of the PVB than that in the Nylon/SEBS system, are dispersed in the Nylon 6 matrix. For the blend composition containing 10 wt % PVB, the dispersed PVB particles seemed to be essentially very small [Fig. 3(b)], the maximum particle size was observed $\sim 1 \mu\text{m}$ in diameter. Then, the average particle sizes in all the blend compositions for Nylon 6/PVB of 80/20 wt % [Fig. 3(c)], 65/35 wt % [Fig. 3(d)], 60/40 wt % [Fig. 3(e)], and 50/50 wt % [Fig. 3(f)] were approximately varied between 1.5 and 2.5 μm , indicating that the size was slightly increased with PVB content, but it is still relatively small compared with the SEBS system. Distribution of the particles in Figures 3[(b)–(d)] is fairly well dispersed; on the other hand, in Figures 3(e, f), it is relatively poor with a slightly larger particle size. This may be because of the coagulation of particles during the mixing process. Nevertheless, for the whole range of blend compositions, debonding phenomena are not observed, which indicates the existence of some interfacial adhesion between the components. In the blend up to 35 wt % PVB, the disperse phase tightly adhered to the continuous phase. A certain adhesion force formed at the interface is expected to affect the morphology by lowering the interfacial tension. From comparison of the morphology of Nylon 6 blended with the recycled PVB film and conventional SEBS, the disperse phase of the former system is enhanced better than the latter. This may be due to the residual plasticizer that is possibly working as a wetting agent between Nylon 6 and PVB film.

Mechanical Properties

Stress–strain curves of the dried Nylon 6/PVB and Nylon 6/SEBS blends are shown in Figure 4(A,B). Yielding has been observed for Nylon 6 solely; however, this behavior is not observed for the rest of the blends. In addition, whole blends exhibited reduced tensile strength without the sign of necking. Gradual enhancement in elongation at break of the Nylon 6/PVB system up to 60/40 composition was observed, whereas a slight

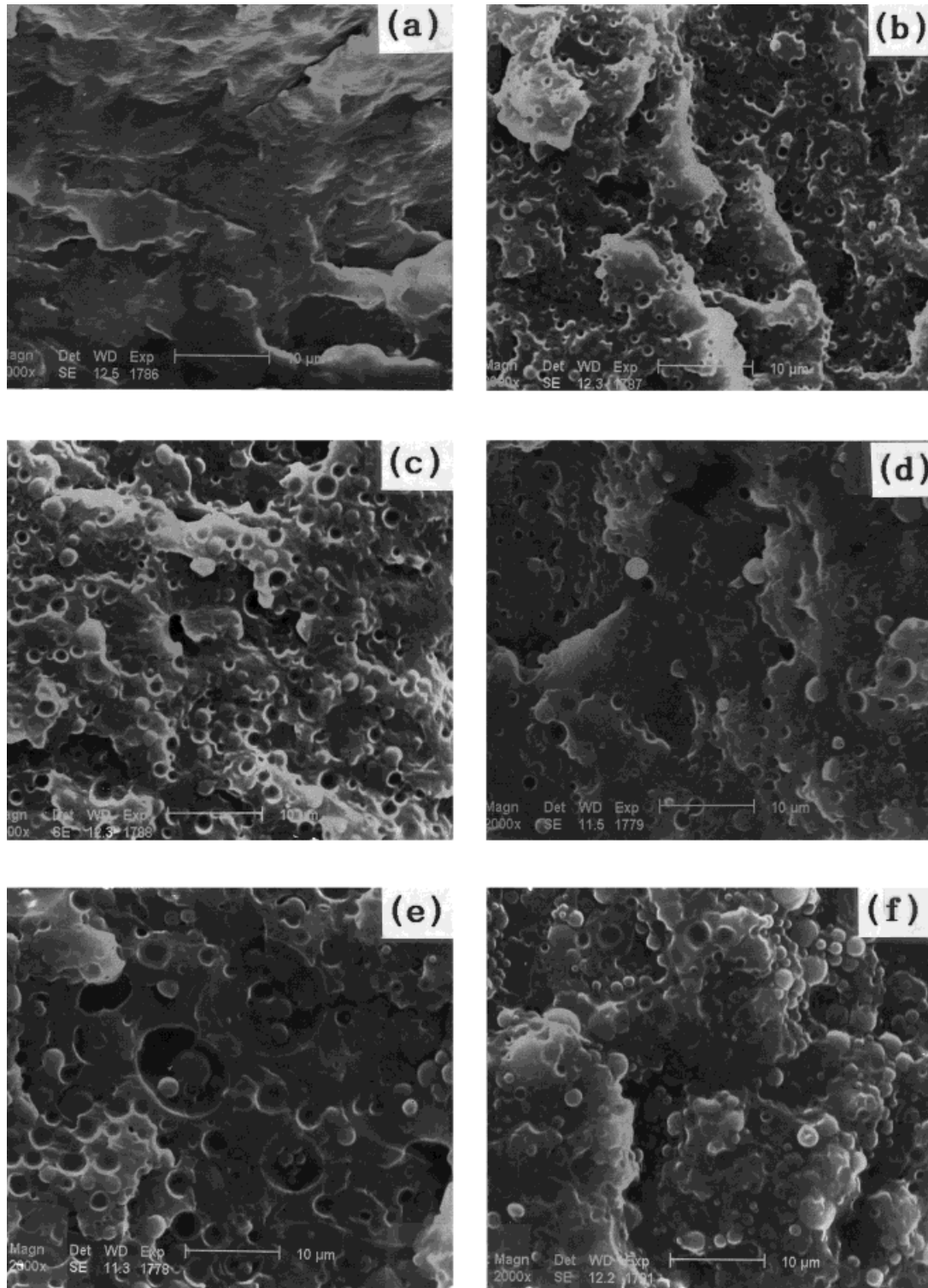


Figure 3 SEM photographs of fractured surface of Nylon 6 and Nylon 6/PVB blends: (a) Nylon 6, (b) 90/10, (c) 80/20, (d) 65/35, (e) 60/40, and (f) 50/50 wt %.

decrease was observed for the Nylon 6/SEBS system.

Based on the stress–strain curve, mechanical

properties are analyzed in terms of the tensile modulus and strength. Figure 5(A) represents the tensile modulus of the blends of Nylon 6/PVB

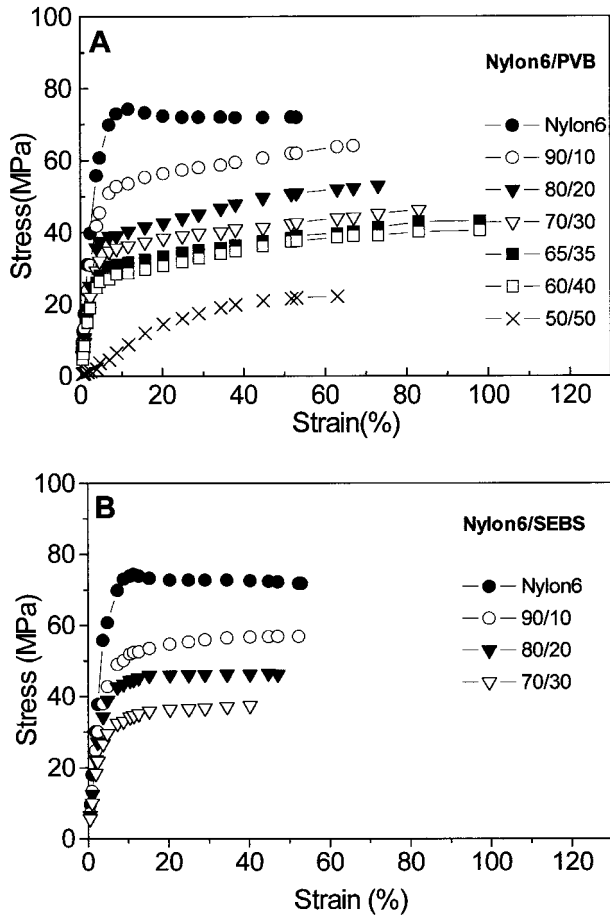


Figure 4 Stress-strain curves of Nylon 6 toughened with various contents of PVB and SEBS. (A) Nylon 6/PVB system. (B) Nylon 6/SEBS system.

(open symbol) and Nylon 6/SEBS (closed symbol) systems. Theoretical predictions on the basis of eq. (1) are used to calculate the tensile modulus, and the results are drawn with a dotted line in Figure 5. The parameters used for calculation is as follows: $E_m = 1,724.8$ MPa, $E_d = 13.2$ MPa, $\alpha = 0.75$, and $\nu = 0.44$. Poisson's ratio (ν) of Nylon 6 is taken from ref. 25. This is not critical for the application of the Kerner model, because the results prove to be rather insensitive to variations of Poisson's ratio. As the PVB content is increased, the modulus of the blend is decreased, and the experimental values agree well with the theoretical ones, thus implying that some adhesive force exists between the Nylon 6 matrix and PVB particles. On the other hand, for the blend containing 50 wt % PVB, experimental data are much lower than the theoretical prediction by the Kerner model. As observed in the SEM photographs in Figure 3(f), the dispersed particles are slightly

exposed on the surface of the continuous phase, and the particles are coagulated. This seems to indicate that the interfacial adhesion of this blend composition is relatively smaller than that of the other blend compositions due to the coagulation of the particles.

Figure 5(B) compares the experimental tensile strength of Nylon 6/PVB blend with the theoretical values calculated by eq. (5). This figure also shows the experimental data for the Nylon 6/SEBS system. The results under the experimental error agree well with the theoretical ones, but the exception is obtained in the blend containing 50 wt % PVB. This again seems to be from poor morphological behavior. As suggested in the previous section, the interfacial adhesion force is proposed from the intermolecular interaction between amide groups and vinyl acetate or the vinyl alcohol group, although the Nylon 6/PVB blend

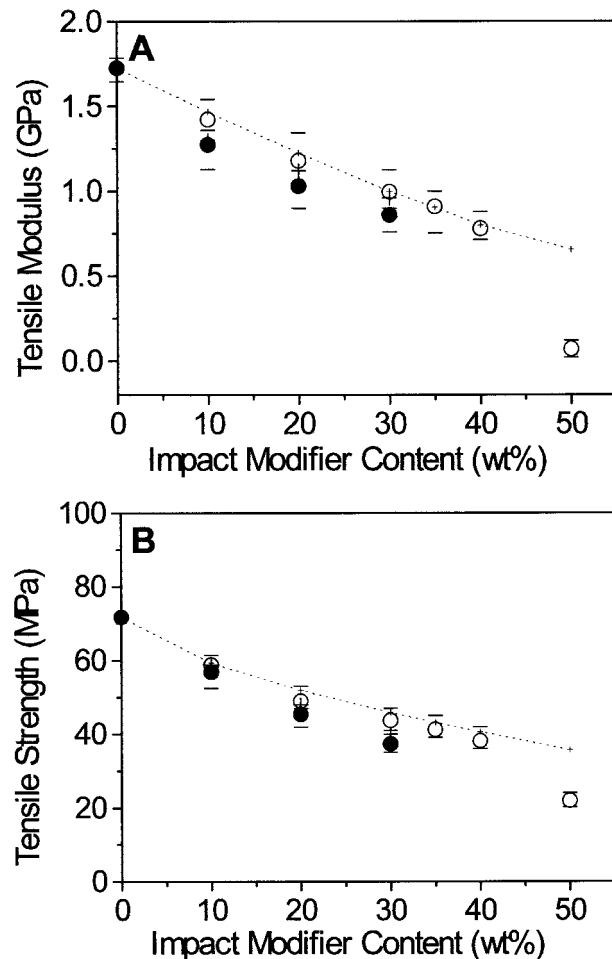


Figure 5 (A) Tensile modulus of Nylon 6 toughened with (●) SEBS and (○) PVB. (B) Tensile strength of Nylon 6 toughened with (●) SEBS and (○) PVB.

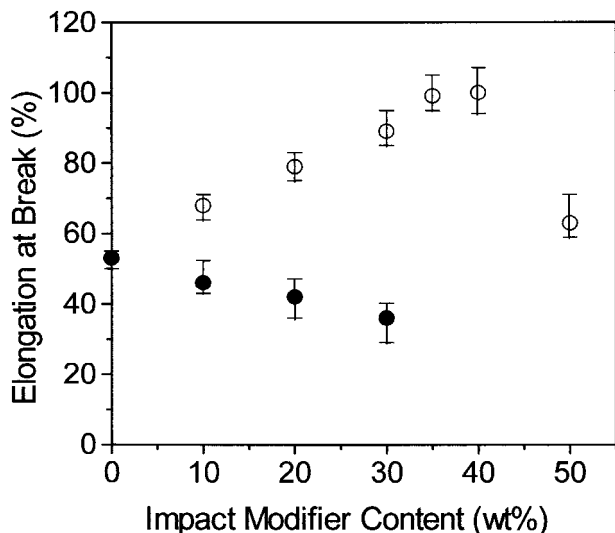


Figure 6 Elongation at break of Nylon 6 toughened with (●) SEBS and (○) PVB.

is not miscible all over the blend compositions. Nylon 6 is semicrystalline material with the melting point of 220°C, whereas PVB film is apparently a thermoplastic elastomer having the glass transition of 29°C. Blends of PVB resin and thermoplastic polyurethane were reported to be thermodynamically and mechanically miscible over a wide range of compositions.²⁹

Figures 5 and 6 compare the tensile modulus, the tensile strength, and the elongation at break between Nylon 6/PVB and those of Nylon 6/SEBS systems, respectively. Tensile modulus and strength in Nylon 6/PVB system is ~ 10–20% higher than those of the Nylon 6/SEBS system. In addition, for the Nylon 6/PVB blend, we observed a dramatic rise in elongation at break with PVB content. Nylon 6 itself showed a limited elongation at break ~ 53%, whereas the blend containing 30–40 wt % PVB showed a remarkable improvement in elongation at break up to 100%, which indicates that recycled PVB film may be used as a toughening agent.

Figure 7 compares notched Izod impact strength between the Nylon 6/PVB and Nylon 6/SEBS systems. Izod impact strength of Nylon 6 with SEBS is increased ~ 50%, with 20–30 wt % SEBS content. On the other hand, as the PVB content is increased, considerable improvement at the maximum 450% in Izod impact strength is observed. In particular, the blend containing 30–40 wt % PVB shows superior properties in elongation at break and Izod impact strength. This means that the scrapped PVB film has merit to

be used as a toughening agent or an impact modifier of Nylon 6. In the real field, synthesized SEBS has often been used for the purpose of the enhanced impact strength of Nylon 6, or maleic anhydride grafted-SEBS is a more valuable impact modifier than SEBS. Thus, recycled PVB can replace SEBS as an impact modifier of Nylon 6. Note in Figure 3(c–e) that the Nylon 6 matrix adheres to the PVB particles, indicating the existence of interfacial adhesion between the components. According to Wu,³⁰ the Izod impact strength of rubber-toughened nylon can be increased if particle size is reduced to 0.1–0.3 μm . Although the PVB particle size varies between 1 and 2.5 μm in our system, the impact strength is considerably increased up to 450%. It is likely that interparticle distance and interfacial adhesion act as an important factor for determining the toughness or brittleness in the Nylon 6/PVB system.^{19,23} In our blend system, although the average particle sizes in whole blend compositions are slightly increased with PVB content, debonding phenomena between the matrix and the dispersed phases are not observed. In the blends up to 35 wt % PVB, the dispersed phase is tightly adhered to the continuous phase. Wu^{19,30} suggested the following: if dispersed particles are sufficiently separated, the stress field around particles will be slightly affected by the presence of other particles inducing the system to be brittle. However, when the particles are close enough, the field around neighboring particles will influence the system, resulting in enhanced matrix yielding and toughness. Thus, the impact strength of the Nylon 6/

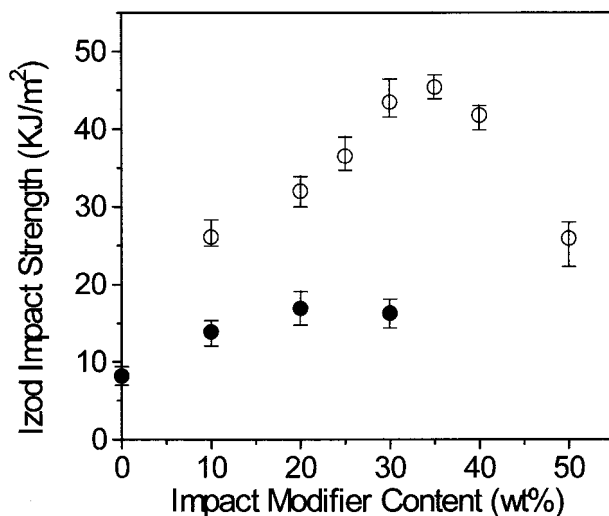


Figure 7 Izod impact strength of Nylon 6 toughened with (●) SEBS and (○) PVB at 23°C.

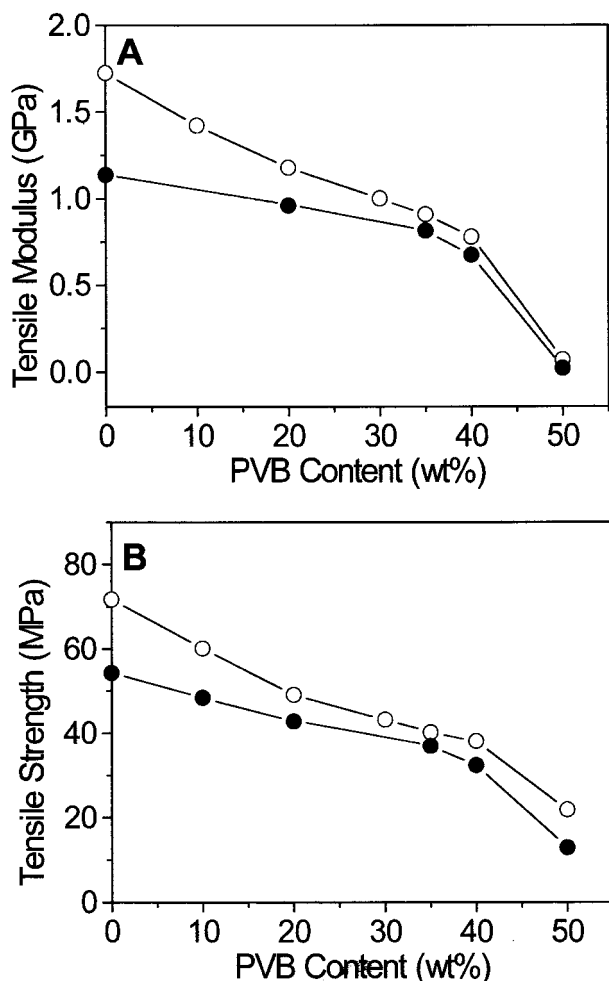


Figure 8 (A) Tensile modulus of Nylon 6 toughened with various contents of PVB. (○) Dried specimens and (●) wetted specimens. (B) Tensile strength of Nylon 6 toughened with various contents of PVB. (○) Dried specimens and (●) wetted specimens.

PVB system can be enhanced by the incorporation of rubberlike PVB that is tightly adhered to the Nylon 6 phase. If experimental error is allowed, the Nylon 6/PVB blend containing 20–35 wt % PVB shows the optimum composition for improved impact strength.

Nylons are sensitive to moisture content, and their properties after water absorption are poor compared with those of dried specimens.³¹ The strong hydrophilicity causes serious problems, especially in electrical applications. In view of hygroscopic property of Nylon 6 in wet condition, we examined the effect of PVB on the tensile properties of the blends with absorbed moisture. The results are drawn in Figure 8(A,B), where the tensile properties of both dried and wetted blends

are plotted as a function of PVB content. Tensile properties between dried and wet Nylon 6 are considerably different; those of the latter are much poorer than the former. However, for those of the blends, dried specimens show relatively higher properties than those of the wet specimens, then come close to each other when the 50/50 blend is reached. This may be due to the poor morphological structure [Fig. 3(f)], which creates a very fine void by a weak interfacial adhesion force between Nylon 6 and PVB particles. Thus, it is conceivable that moisture resistance is enhanced by the presence of PVB in the Nylon 6 matrix. In particular, the blends containing 20–35 wt % PVB are less moisture-sensitive than the other blend compositions.

CONCLUSIONS

The possibility of an impact modifier and a toughening agent of recycled PVB film in the Nylon 6 system has been investigated. For the Nylon 6/PVB blend system containing 10 wt % PVB, the dispersed PVB particles seemed to be essentially very small, as much as 1 μm . In particular, the dispersed phase is tightly adhered to the matrix phase in the range of 20–35 wt % PVB, regardless of the slightly increased particle size. Tensile modulus and strength of the Nylon 6/PVB and Nylon 6/SEBS blend systems are both lower than Nylon 6 itself. However, the former system shows higher mechanical properties than the latter. Notched Izod impact strength of the blends is enhanced up to 450%, with incorporation of PVB in the range of 10–40 wt % PVB. In addition, the Nylon 6/PVB system exhibits improved moisture resistance. Thus, recycled PVB film is applicable as a toughening agent or an impact modifier in the Nylon 6 system. In particular, the optimum blend composition would be 20–35 wt % PVB in the Nylon 6 matrix for providing a high elongation at break and an enhancement in toughness and impact strength.

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