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Miscibility of polyvinyl butyral/nylon 6 blends

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Abstract

This work describes the interaction of nylon6 (N6) with the random copolymer poly(vinyl butyral) (PVB) at various hydroxyl levels of residual vinyl alcohol in the PVB. The methylene and amide groups of N6 were observed to interact favorably with the PVB, resulting in thermodynamic miscibility at PVB compositions in the range of 0.50–0.65 volume fractions of vinyl alcohol in PVB. The polymers' interaction parameters were estimated to be −0.04 and 0.04 for $\chi_{\mathrm{N6/VA}}$ and $\chi_{\mathrm{N6/PVB}}$, respectively. Miscibility parameter results compared favorably with the miscibility window determined. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Miscibility; Polymer blends; Nylon

1. Introduction

Poly(vinyl butyral) (PVB) and nylon 6 (N6) are both widely used in a variety of products. PVB is a random copolymer of vinyl alcohol and vinyl butyral (VB) moieties synthesized by reacting poly(vinyl alcohol) (PVA) with butyraldehyde in an acid medium. The resulting copolymer contains unreacted vinyl alcohol (VA) groups, in amounts, which can be tailored to specific applications. Because of the presence of both hydrophilic (polar and hydrogen bonding groups) and hydrophobic groups, this copolymer has some significance in terms of potential miscibilities with other macromolecules.

The molecular structure and the effect of molecular interactions and miscibility of PVB in blends with other PVBs containing varying proportions of unreacted vinyl alcohol groups, were discussed in a previous study [1] in which the phase diagram showing the compositional ranges and limits of miscibility at various hydroxyl levels in PVB was determined, as was the reduced intramacromolecular hydroxyl butyral interaction parameter.

The random architecture and differing nature of the pendant groups in PVB suggest it is also useful to determine intermacromolecular interactions between this polymer and others. The presence of these groups also suggests the possible use of PVB as a compatibilizer in ternary systems, using concepts other than those employed with conventional compatibilizers such as maleic anhydride [2,3]. In this context, a polyamide (nylon 6) was chosen because the amide and methylene groups present are also hydrophobic, polar, and hydrogen bonding. The amide groups are known to exhibit hydrogen bonding and strong polar interaction [4–6], which may account for the relatively high melting point and tensile strength. Polyolefins and other polymers containing only hydrophobic groups are known to be immiscible with nylon 6 [6]. The present study examined PVB/N6 blends to ascertain whether there is a range of PVB structures (a "window of miscibility") that interact with nylon 6 to produce miscible blends.

2. Experimental

2.1. Materials

Nylon 6 purchased from *Sp*^{2} Scientific Products, Inc. had a relative viscosity of 30 and a glass transition temperature and melting point of 62 and 221°C, respectively. PVBs of systematically varying vinyl alcohol content were synthesized as described previously [1]. For completeness, we include the following brief description of poly(vinyl butyral) synthesis.

Polyvinyl alcohol (DP = 2000, Aldrich) containing a residual vinyl acetate content of 1% was dissolved in distilled water at ≈50°C by stirring for about 2 h to prepare an 8–10 wt% solution. The temperature of the solution was reduced to 8–12°C and a quantity of a 70% solution of nitric acid was added. To this, a pre-determined amount of butyraldehyde was added, according to the desired acetalization

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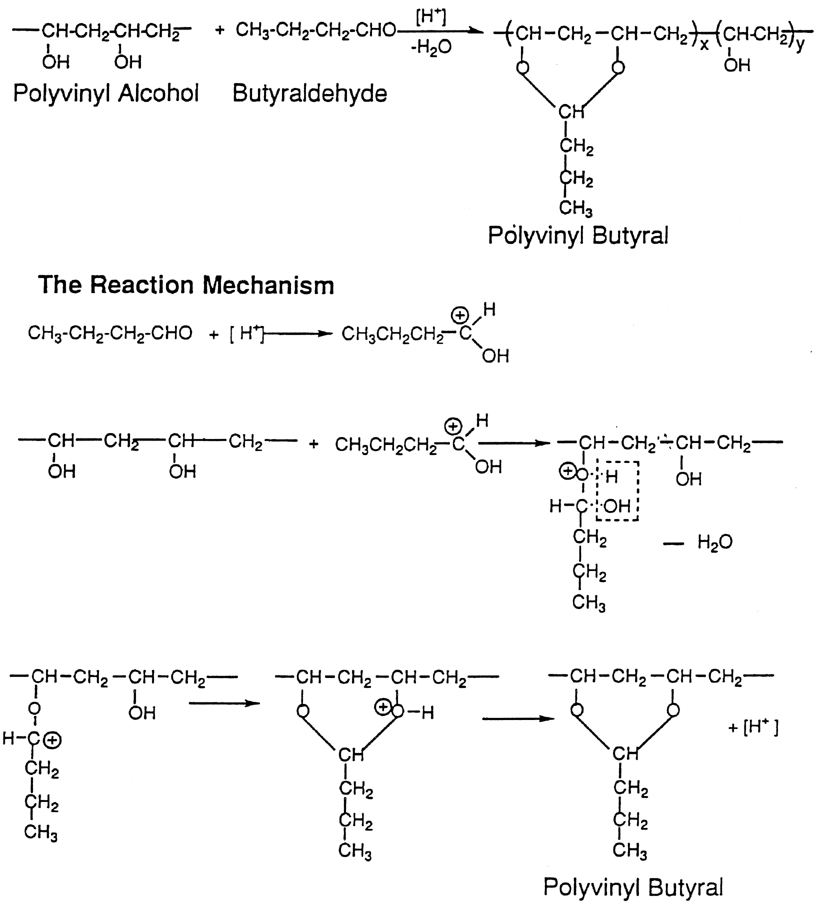


Fig. 1. Synthesis of poly(vinyl butyral).

degree, and the reaction was initially carried out at 10°C for 3 h with continuous stirring, followed by 1 h at 60–70°C to complete the acetalization reaction. The reactor contents were washed with distilled water once, neutralized with 50% sodium hydroxide to a pH of 11–12, held at this pH at 70°C for 1 h and then washed again with distilled water to a final pH of 7.5–8. The product was filtered and dried at 50°C in a vacuum oven until a constant weight of sample was achieved. GPC measurement of the PVBs indicated that no degradation occurred during the synthesis of the modified polymers. The overall reaction and reaction mechanism are shown in Fig. 1. Polyvinylbutyrals properly washed and neutralized exhibit good thermal stability up to ≥ 230°C, depending upon exposure to shear, length and extent of thermal exposure, and atmosphere (oxygen content in the atmosphere). Discoloration of PVBs is a sensitive manifestation of degradation. For all melt blending and thermal exposures, discoloration attributable to degradation was not observed to be a factor.

Table 1
Analytical results for PVBs

PVB sample no.	Vinyl acohol content of PVB			PVB T_g (°C)
	wt%	mol%	Volume fraction	
PVB14	14.2	34.8	0.12	71
PVB23	22.9	48.9	0.19	76
PVB28	28.0	55.7	0.24	–
PVB31	31.0	59.1	0.27	79
PVB36	36.4	64.8	0.32	–
PVB45	45.1	72.6	0.40	83
PVB55	55.0	79.8	0.50	85
PVB63	63.3	84.8	0.59	–
PVB70	69.5	88.0	0.65	88
PVB89	89.0	96.3	0.87	90

Analyses of PVBs with differing VA contents are shown in Table 1. The analyses were performed by titration of liberated acid following acetylation by acetic anhydride according to ASTM D-1396 (“The determination of VA content of PVBs”) [7].

The code used here to identify the samples lists the weight concentration of residual vinyl alcohol immediately following “PVB”. Thus sample “PVB14” has a vinyl alcohol content of 14 wt% and therefore 86 wt% vinyl butyral.

2.2. Preparation of PVB/nylon 6 blends

Nylon 6 and PVB’s were dried at 50°C under reduced pressure for 24 h before use. PVB/N6 blends were prepared by freeze milling the powder mixtures in liquid nitrogen with a Freezer Mill(SPEX Industries, Inc.). The mixtures

Table 2
Interaction parameter, $\chi_{VA/VB}$, of PVB/PVB blends at 180°C

Sample no.	$\phi_{1,VA}$ [a]	N [b]	χ_{crit} [c]	$\chi_{VA/VB}$ [d]
PVB14	0.12	2000	0.001	0.14
PVB23	0.19	2000	0.001	0.11
PVB28	0.24	2000	0.001	0.09
PVB31	0.27	2000	0.001	0.08
PVB36	0.32	2000	0.001	0.07
PVB45	0.40	2000	0.001	0.04
PVB55	0.50	2000	0.001	0.02
PVB63	0.59	2000	0.001	−0.03
PVB70	0.65	2000	0.001	−0.05
PVB89	0.87	2000	0.001	−0.13

[a] $\phi_{1,VA}$ is the volume fraction of vinyl alcohol for PVB 1.
[b] N is the DP of parent polyvinyl alcohol.
[c] χ_{crit} is the interaction parameter at the critical point.
[d] $\chi_{VA/VB}$ is the interaction parameter, recalculated from [1].

were then again dried for 24 h under reduced pressure at 50°C and were used to prepare the melt blends. All the latter were molded after melt mixing for 15 min at 220°C with a Mini–Max Molder(Atlas Electric Devices Co., CS-183 MMX). The rotation speed was 15 rpm.

2.3. Thermal analysis

The phase behavior of the melt blends was studied by differential scanning calorimetry (DSC:Perkin–Elmer 7) and dynamic mechanical thermal analysis (DMTA:Rheometric Scientific IV). The DSC scanning rate was 20°C/min; that of the DMTA was 2°C/min at a frequency of 10 Hz. The 50/50 (weight ratio) PVB/N6 samples were annealed at 230°C for 1 h, quenched in liquid nitrogen, and dried under reduced pressure for 24 h at 50°C. To investigate the phase behavior at lower temperatures, certain samples were annealed at 130°C for 1 h and quenched after annealing at 230°C for 1 h. The PVB31/N6 and PVB63/N6 samples with 20/80 and 80/20 weight ratios were annealed and quenched at 180°C for 1 h after annealing at 215°C for 1 h and were quenched in liquid nitrogen and dried under reduced pressure for 24 h at 50°C. All blends used in the DMTA measurements were annealed at 200°C for 1 h, quenched in liquid nitrogen and dried under reduced pressure for 24 h at 50°C.

2.4. Scanning electron microscopy

The phase structure for PVB 14, 36, 63, 89/N6 blends was investigated by means of a scanning electron microscope (SEM: JEOL LTD., JSM-35CF). For the SEM measurements, the specimens were fractured in liquid nitrogen before the etching of PVB with methanol for 20 h at room temperature. Methanol is a non-solvent for N6, but dissolved all the PVBs. The fracture surfaces of the PVB/N6 after etching were coated with gold for the SEM observation. The fracture surfaces of (PVB31/N6:80/20 wt%) and (PVB 63/N6:80/20 wt%) blends were etched with methanol for 1 min only before being coated with gold for the SEM observation. In general, the methanol etching time was chosen to produce the appropriate surface features contrast, taking into account the difference in solubility of the two polymers.

2.5. Physical property measurements

Specimens for tensile property measurements were molded at 240°C for 10 min with a Mini–Max Molder, cooled to room temperature, and dried under reduced pressure for 48 h at 50°C. The testing was done using flat dumbbell shaped samples on (Instron 4468) machine at room temperature, following the procedure described in ASTM D-638. The static load cell was 50 kN and the cross-head speed was 3.0 mm/min. Five test specimens were used for each blend and the values averaged.

2.6. Estimation of interaction parameters $\chi_{N6/VA}$ and $\chi_{N6/VB}$

The interaction parameter of a mixture of a homopolymer A_{N_1} and a copolymer $(C_yD_{1-y})_{N_2}$ from mean field theory [8] is given by:

$$\chi_{blend} = y\chi_{AC} + (1-y)\chi_{AD} - y(1-y)\chi_{CD} \quad (1)$$

where χ_{ij} are the respective segmental interactive parameters. The critical point occurs at a temperature, for which χ_{blend} equals χ_{blend}^{crit} which is given by:

$$\chi_{blend} = \chi_{blend}^{crit} = 1/2(N_1^{-1/2} + N_2^{-1/2})^2 \quad (2)$$

In this system, if there is a window of miscibility, i.e. a range of y within which miscibility occurs at a given temperature, the interaction parameters $\chi_{N6/VA}$ and $\chi_{N6/VB}$ can be estimated, since $\chi_{VA/VB}$ is known [1]. The polymerization degrees (N_i) of PVB and N6 are ~2000 and ~100, respectively. The interaction parameter $\chi_{VA/VB}$ recalculated from [1], is shown in Table 2 and is strongly composition dependent, reflecting the limitations of the mean field theory in systems where strong intermolecular specific interactions occur.

2.7. Estimation of miscibility/compatibility window from the miscibility parameter model

Another model used to estimate potential areas of compatibility/miscibility is based on the use of solubility parameters (δ) and utilizes the individual components that make up the total solubility parameter as an approach to check for possible windows of compatibility/miscibility [9,10]. The miscibility parameter is given by:

$$MP = (\delta_A^{poly} - \delta_B^{poly})^2 \quad (3)$$

where the MP is represented by appropriate combinations of the dispersive (d), polar (p), and hydrogen bonding (h), components of the solubility parameter (see Refs. [9,10]), such that for any polymer blend system the four possible MPs are: MP_{dph}, MP_{dp}, MP_{ph}, and MP_{dh}.

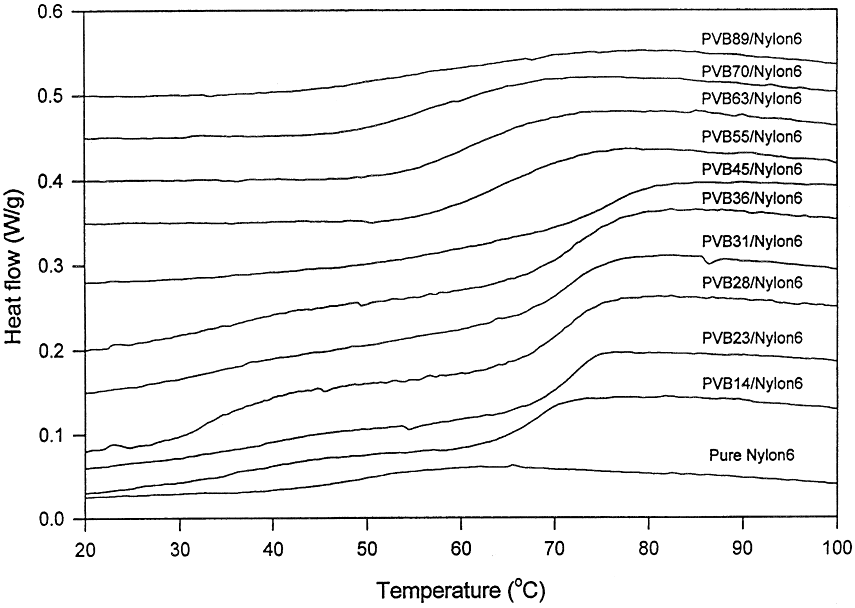


Fig. 2. DSC curves showing the glass transitions of 50/50 blends of Nylon 6 with PVB of various hydroxyl levels. Key: PVB14/Nylon6 = 50 : 50 blend of 14 wt% VA in PVB with Nylon6; PVB23/Nylon6 = 50 : 50 blend of 23 wt% VA in PVB with Nylon6; PVB28/Nylon6 = 50 : 50 blend of 28 wt% VA in PVB with Nylon6; PVB31/Nylon6 = 50 : 50 blend of 31 wt% VA in PVB with Nylon6; PVB36/Nylon6 = 50 : 50 blend of 36 wt% VA in PVB with Nylon6; PVB45/Nylon6 = 50 : 50 blend of 45 wt% VA in PVB with Nylon6; PVB55/Nylon6 = 50 : 50 blend of 55 wt% VA in PVB with Nylon6; PVB63/Nylon6 = 50 : 50 blend of 63 wt% VA in PVB with Nylon6; PVB70/Nylon6 = 50 : 50 blend of 70 wt% VA in PVB with Nylon6; PVB89/Nylon6 = 50 : 50 blend of 89 wt% VA in PVB with Nylon6.

3. Results and discussion

3.1. Thermal analysis and morphology

DSC curves showing primarily the glass transition of the 50/50 PVB/N6 blends are shown in Fig. 2. Although the results are not completely unambiguous, due at least in part to the rather ill defined T_g exhibited by N6 itself, it seems that blends of N6 and PVBs containing 14–45 wt% VA show two T_g's, while the blend of N6 and the PVB containing 55 wt% of VA has a single T_g. However, no conclusions can be drawn from the DSC results due to baseline changes and the influence of PVB on the relative proportions of amorphous/crystalline content of the nylon.

To obtain more definitive results of the miscibility behavior of these blends, dynamic mechanical thermal analysis (DMTA) was utilized. The DMTA results confirm that the 50/50 PVB/N6 blends exhibit two peaks in the loss modulus and tan δ curves up to a PVB composition of about 55 wt% of VA, whereas PVB/N6 blends in which the copolymers contain 55–70 wt% of VA exhibit a single peak. DMTA curves illustrating the effects of copolymer composition are shown in Fig. 3a–c.

The storage and the loss moduli are represented in their usual form by E' and E'', and tan δ. The multiple peaks particularly evident in the loss modulus (E'') (Fig. 3a) thus indicate that blends of N6 and PVBs containing less than 55 wt% (0.50 volume fraction) of vinyl alcohol are not thermodynamically miscible and form two phases; blends in which the PVB contains 55–70 wt% of VA exhibit a single phase (Fig. 3b).

The morphology of the samples shown in the SEM micrographs reveal a more gradual transition from two distinct phases to a single phase as the VA content of the PVBs was increased from below the lower miscibility limit ($\cong$ 55 wt% VA) through the miscible range to an apparent upper miscibility limit ($\cong$ 69 wt% VA).

Micrographs illustrating the changes are shown in Fig. 4. Protruding nylon "nodules" appear to constitute the major phase in the fractured surfaces of the immiscible N6 rich blend (PVB14/N6) indicating the pull-out of N6 from the matrix. In the miscible phase region only one phase can be observed. These results are entirely consistent with and reinforce the DSC and DMTA results.

Blends of 80/20 PVB/N6 and 20/80 PVB/N6 using PVBs of several levels of VA content were also investigated to determine how the blend ratios influence phase behavior and to allow a definitive estimate of phase boundaries. The DMTA scans and SEM results showed (within the limits of available PVB samples) the same window of miscibility that was exhibited by the 50/50 PVB/N6 blends and it was additionally noted that they were not dependent upon annealing temperature over the range examined.

A second series of 50/50 PVB/N6 blends was used to investigate melting and crystallization behavior. Melting was characterized by a series of melting endotherms that showed a gradual diminution of the nylon melting endotherm and the appearance of a second lower melting endotherm until the miscible limit was reached. At that

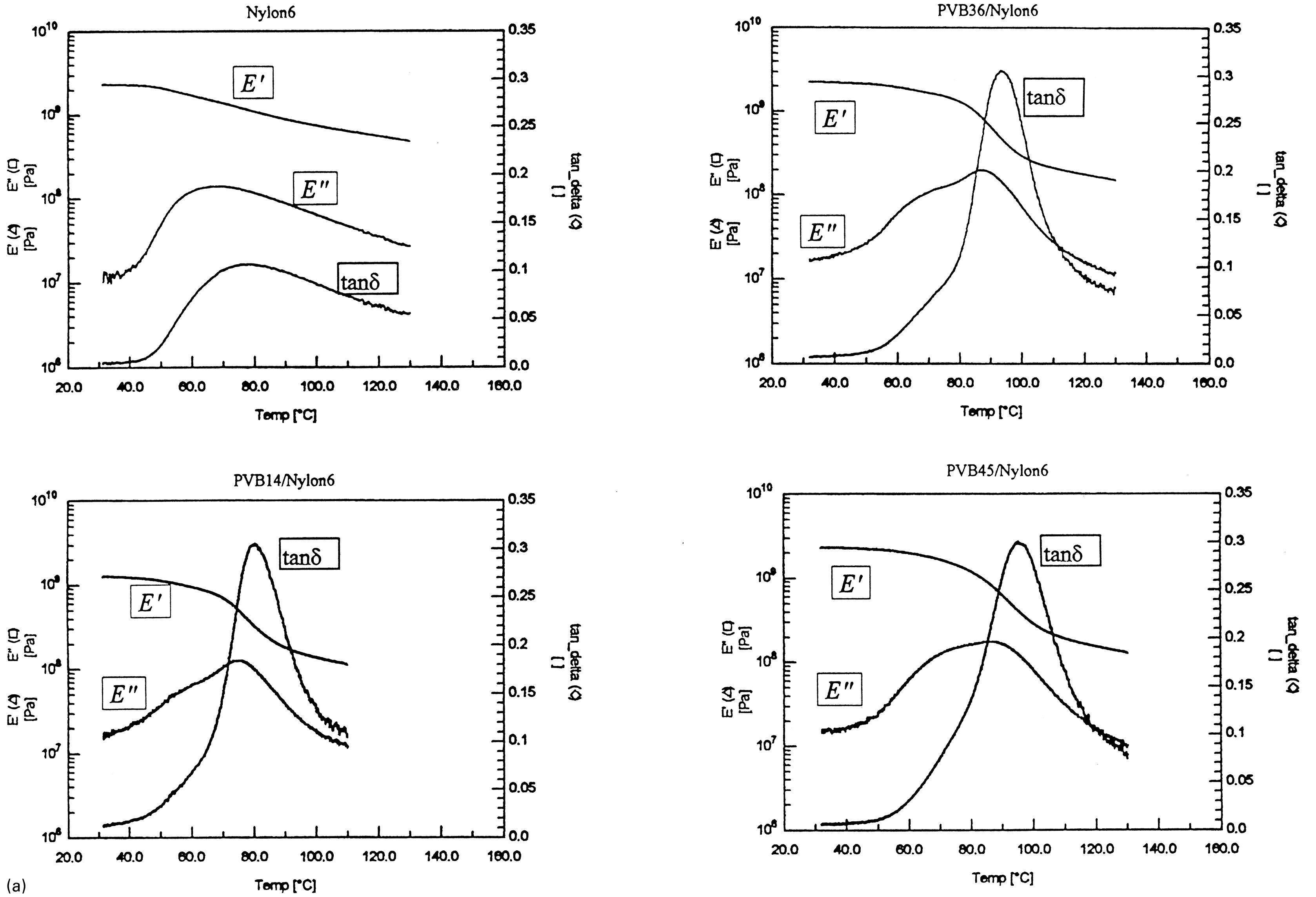


Fig. 3. Representative dynamic mechanical analysis (DMTA) curves illustrating the effects of composition on blend miscibility as indicated by moduli and tan delta responses. Key. (a) Nylon6 = polyamide: PVB14/Nylon6 = 50 : 50 blend of 14 wt% VA in PVB with Nylon6; PVB36/Nylon6 = 50 : 50 blend of 36 wt% VA in PVB with Nylon6; PVB45/Nylon6 = 50 : 50 blend of 45 wt% VA in PVB with Nylon6. (b) PVB55/Nylon6 = 50 : 50 blend of 55 wt% VA in PVB with Nylon6; PVB63/Nylon6 = 50 : 50 blend of 63 wt% VA in PVB with Nylon6; PVB70/Nylon6 = 50 : 50 blend of 70 wt% VA in PVB with Nylon 6; PVB89/Nylon6 = 50 : 50 blend of 89 wt% VA in PVB with Nylon6. (c) PVA = vinyl alcohol.

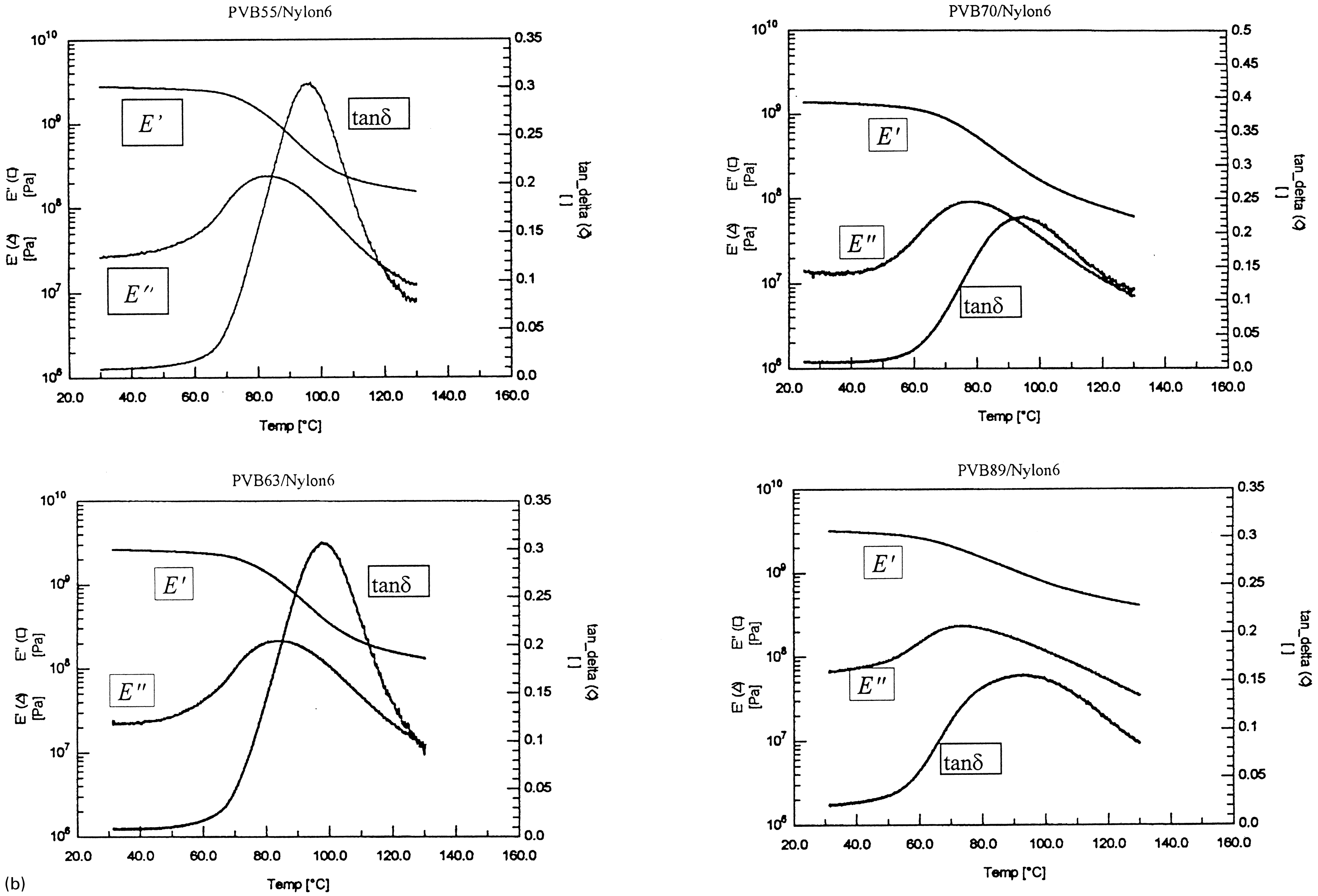


Fig. 3. (*continued*)

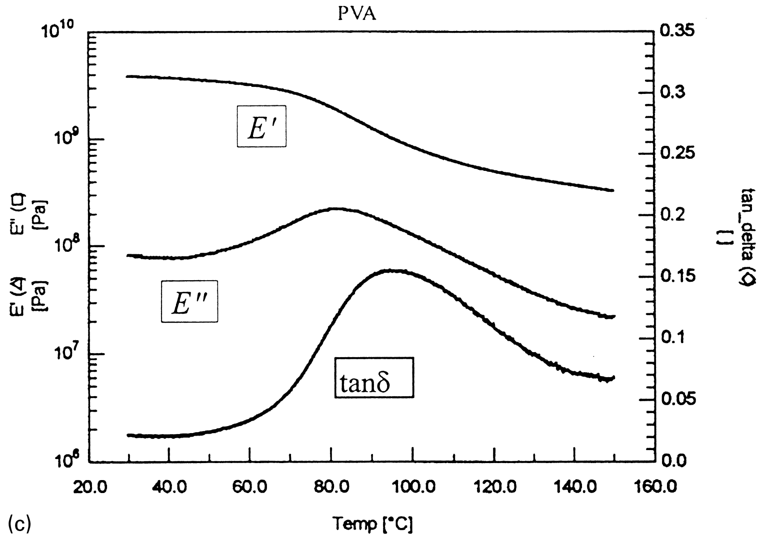
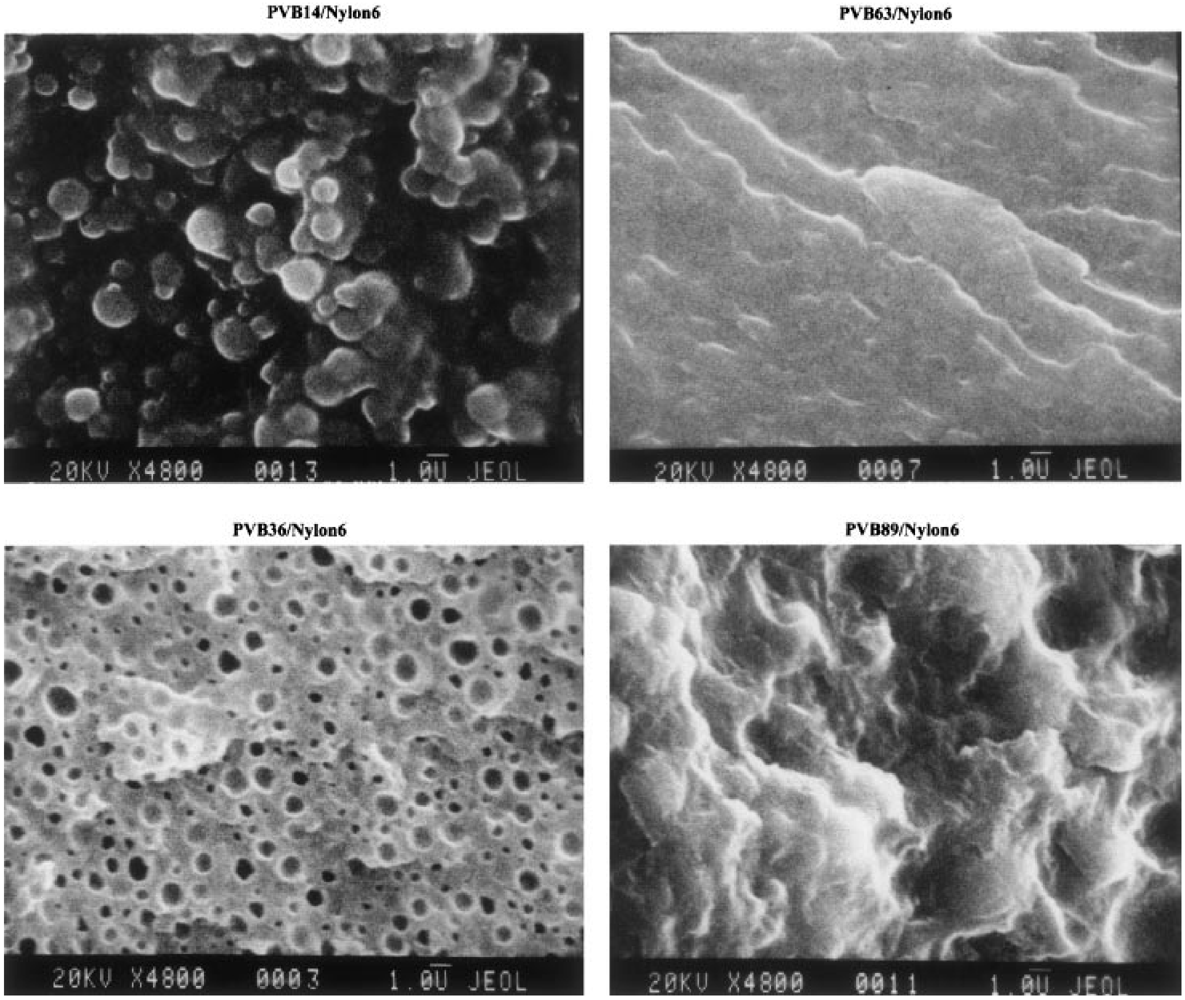


Fig. 3. (*continued*)


PVB14/Nylon6

PVB63/Nylon6

PVB36/Nylon6

PVB89/Nylon6

Fig. 4. SEM micrographs illustrating the morphology of poly(vinyl butyral)/Nylon6 blends at compositions showing immiscibility (PVB = 14 wt% VA) to compatibility(PVB = 36 wt% VA and 90 wt% VA) to miscibility (PVB = 63 wt% VA). Specimens were fractured after cooling in liquid nitrogen using DMTA samples. Fractured surfaces were etched with methanol.

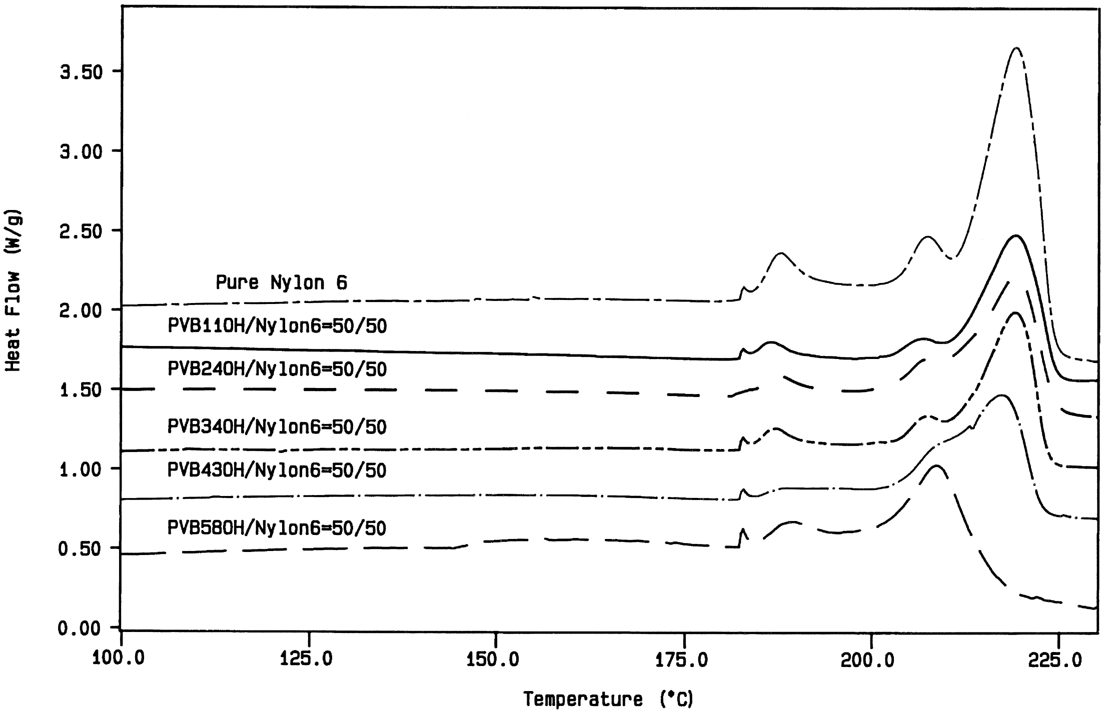


Fig. 5. Melting behavior of 50:50 blends of Nylon 6 with PVB at various hydroxyl levels. Key: PVB 11OH/Nylon6;50/50 = 11 wt% VA in PVB; 50:50 blend; PVB24OH/Nylon6;50/50 = 24 wt% VA in PVB; 50:50 blend; PVB34OH/Nylon6;50/50 = 34 wt% VA in PVB; 50:50 blend; PVB43OH/Nylon 6;50/50 = 43 wt% VA in PVB; 50:50 blend; PVB58OH/Nylon6;50/50 = 58 wt% VA in PVB; 50:50 blend.

point, only one endotherm is exhibited (Fig. 5). Allowing for the fact that only 50% of the blends consists of the crystallizable N6 component, there is no substantial change in the melting point or normalized enthalpy of fusion except for the PVB58/N6 blend composition, which, as already discussed seems to consist of a single, mixed amorphous phase. The multiple melting endotherms observed in N6 and the blends may be due to morphological effects or the presence of different crystal modifications. N6 is known to possess at least three different crystal structures, α, γ, and γ (pseudohexagonal) [11]. In the miscible blend PVB58/N6, the highest melting endotherm is completely absent.

The crystallization behavior of the blends exhibit similar trends (Fig. 6). However, the PVB43/N6 blend, which is compatible but not miscible, already exhibits a crystallization temperature some 20°C lower than those of the immiscible, incompatible blend composition. In the miscible PVB58/N6 blend, the crystallization exotherm is ill defined and occurs at a still lower temperature (170°C).

As stated and shown from DMTA, a PVB45/N6 blend is immiscible, blends PVB55/N6–PVB70/N6 are miscible, and PVB89/N6 is immiscible. These data along with the DMTA, SEM, melting and crystallization behavior, and physical properties, show the estimated lower limit of miscibility to be about 55–60 wt% of VA in PVB/N6 compositions while the upper limit of miscibility is about 70 wt% VA in PVB/N6.

Finally, Fig. 7 shows the weight fraction dependence of the melting point of N6 in a miscible blend. The dependence appears to be linear and is quite large leading to the conclusion that the effect is due to morphological rather than thermodynamic constraints.

3.2. Physical properties

The measured physical properties of the 50/50 PVB/N6 blends are listed in Table 3. The data for neat N6 and PVA

Table 3
Effect of Vinyl Alcohol content on the physical properties of 50/50 PVB/Nylon 6 blends

	Nylon6	PVB14/Nylon6	PVB31/Nylon6	PVB63/Nylon6	PVB89/Nylon6	PVA
TS, max. (MPa)	80.6 ± 5.2	80.8 ± 3.5	86.4 ± 3.7	104.1 ± 3.1	108.2 ± 3.5	109.1 ± 5.5
TS, Yield (MPa)	62.2 ± 5.2	62.4 ± 3.5	74.2 ± 3.7	85.1 ± 3.1	–	95.7 ± 5.5
TS, break (MPa)	63.2 ± 5.2	72.4 ± 3.5	74.2 ± 3.7	85.1 ± 3.1	108.2 ± 3.5	95.7 ± 5.5
Elongation, yield(%)	14.7 ± 2.2	14.1 ± 1.5	13.2 ± 1.5	9.8 ± 1.5	4.3 ± 4.5	4.2 ± 2.3
Elongation, break(%)	33.7 ± 2.2	143.1 ± 1.5	80.2 ± 1.5	35.7 ± 1.5	7.4 ± 1.5	7.5 ± 2.3
Modulus (MPa)	2633 ± 25	2632 ± 20	2633 ± 20	3508 ± 20	3509 ± 20	5261 ± 25

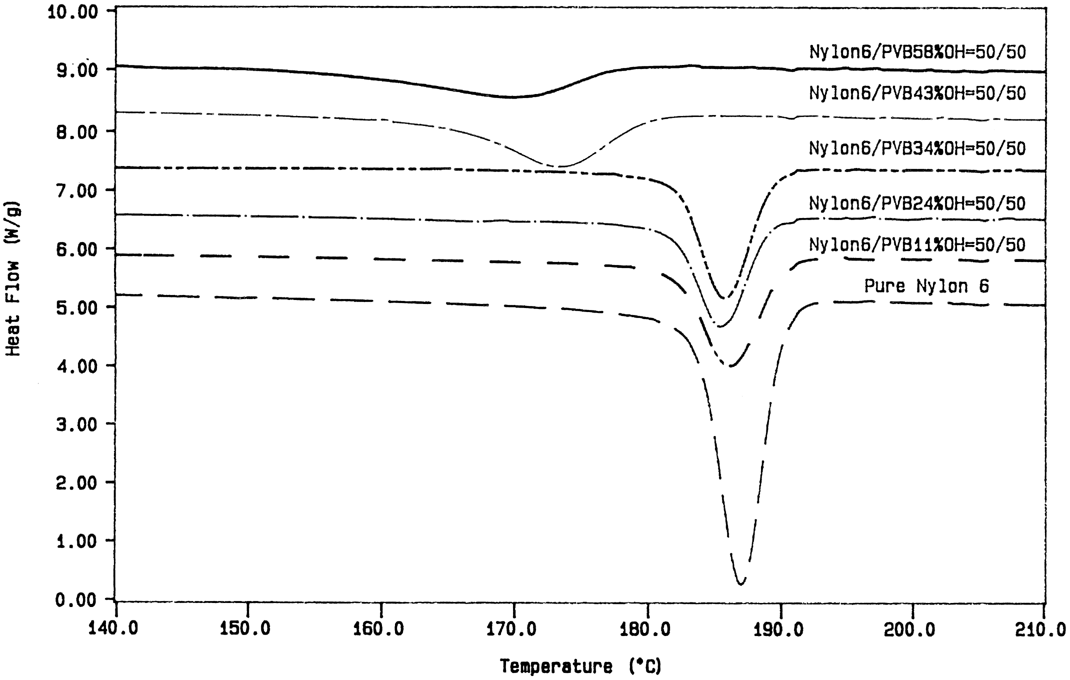


Fig. 6. Crystallization behavior of 50:50 blends of Nylon 6 with PVB at various hydroxyl levels. Key: PVB 11OH/Nylon6;50/50 = 11 wt% VA in PVB; 50:50 blend; PVB24OH/Nylon6;50/50 = 24 wt% VA in PVB; 50:50 blend; PVB34OH/Nylon6;50/50 = 34 wt% VA in PVB; 50:50 blend; PVB43OH/Nylon 6;50/50 = 43 wt% VA in PVB; 50:50 blend; PVB58OH/Nylon6;50/50 = 58 wt% VA in PVB; 50:50 blend.

are also included to complete the series and to allow comparisons.

Although the blends of PVB55/N6–PVB70/N6 posses a single amorphous phase, all of the blends posses a separate crystalline phase of N6 as expected. It is presumed that the blends of PVB14/N6 and PVB31/N6 contain two amorphous as well as a crystalline phase. At 50/50 blend compositions, a co-continuous two-phase morphology exists. However, because of the phase behavior of the blends and the presence of a crystalline phase, the morphologies are complex. From the data presented in Table 3, it can be seen that for PVB31/N6, a compatible but immiscible blend, the physical properties show some enhancement compared to N6 itself. In the case of the miscible blends, PVB55/N6 to ~PVB60/N6 (specifically PVB63/N6), the tensile strength is close to those of PVA without any loss of elongation at break compared to the nylon.

At the PVB/N6 blend composition below the level of VA in PVB that provides thermodynamic miscibility (PVB14) the properties of N6 dominate, with the exception of percentage elongation at break. This is due to the effect of the PVB which has a high elongation (~80%) at this level of VB.

For compositions of PVB in which compatibility (immiscibility) with N6 occurs, e.g. PVB89/N6, the properties, of the PVA dominate. In an intermediate range of VA in PVB (PVB31) where a number of interactions occur, the general properties including yield and elongation at break, have increased compared to N6. Similar results are found in commercial blends where thermodynamic miscibility is not achieved [12,13]. The presence of two compatible phases often leads to a blend which provides a good balance of physical properties and in some cases, better properties than from a thermodynamically miscible blend.

In the case of the PVB63/N6 miscible blend, the tensile strength is increased substantially but at the expense of % elongation yield. Thus in the case of a miscible PVB/N6 blend, the high tensile strength of VA dominates but the percentage elongation at yield and the modulus become approximately arithmetic averages of the VA and N6 values.

These results show that blends at, and slightly below, the miscibility limit, can be tailored to improve both strength and elongation of N6 without sacrificing modulus.

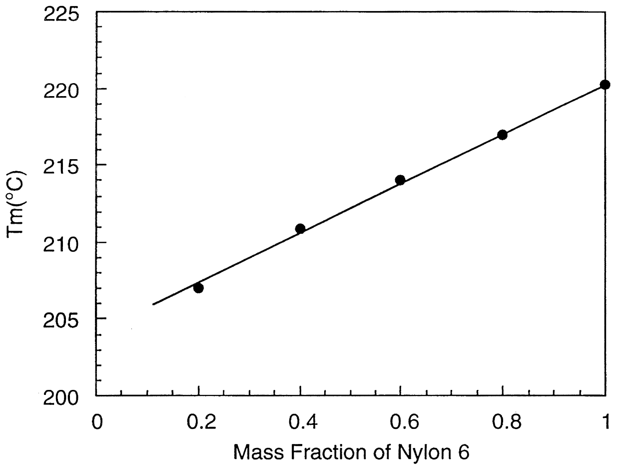


Fig. 7. Depression of melting point of Nylon 6 for a miscible blend of Nylon 6/PVB. Melting point vs. mass fraction of Nylon 6.

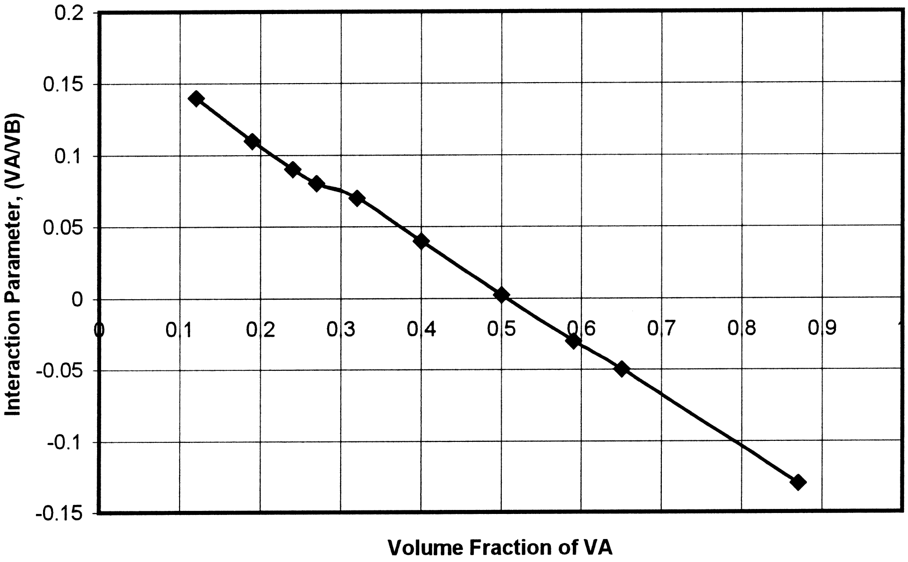


Fig. 8. Copolymer composition dependence of the interaction parameter $\chi_{VA/VB}$ as a function of volume fraction of vinyl alcohol in PVB.

3.3. Interaction parameters $\chi_{N6/VA}$ and $\chi_{N6/VB}$

The interaction parameters $\chi_{VA/VB}$ estimated in the previous work [1] (Table 2) are plotted in Fig. 8. Using the estimated miscibility limits of about 0.5 and 0.65 volume fraction of vinyl alcohol in PVB with N6 from the above results, Eqs. (1) and (2) yielded interaction parameters of: $\chi_{N6/VA} \cong -0.04$ and $\chi_{N6/VB} \cong 0.04$. These values are consistent with the experimental results since these values would account for borderline miscibility of N6 with vinyl alcohol and for its immiscibility with vinyl butyral groups. These values are also consistent with a previous report on compatibility of N6 and PVA [14] in which a positive interaction between the nylon and PVA was demonstrated but without miscibility. Presumably, this is due to energetic interactions between the VA and polyamide groups indicated by the almost neutral interaction parameter, $\chi_{N6/VA}$.

3.4. Comparison of miscibility window with the MP model

The MP model showed a large separation between MP_{dp} and the remaining MPs. The interpretation of this is that the PVB being relatively rich in VA is capable of forming inter and intra chain hydrogen bonds. Thus the MP_{dp} should be more indicative of regions that might exhibit compatibility/miscibility. The MP_{dp} indicated a region from 0.55–85 wt% VA in PVB where compatibility and/or miscibility might be expected. This is based on using a MP_{dp} value of 0.1 or less [9,10] and is illustrated in Fig. 9 where MP is plotted as a function of VA in VB content. This compares favorably with the region of miscibility determined (55–70 wt% of VA).

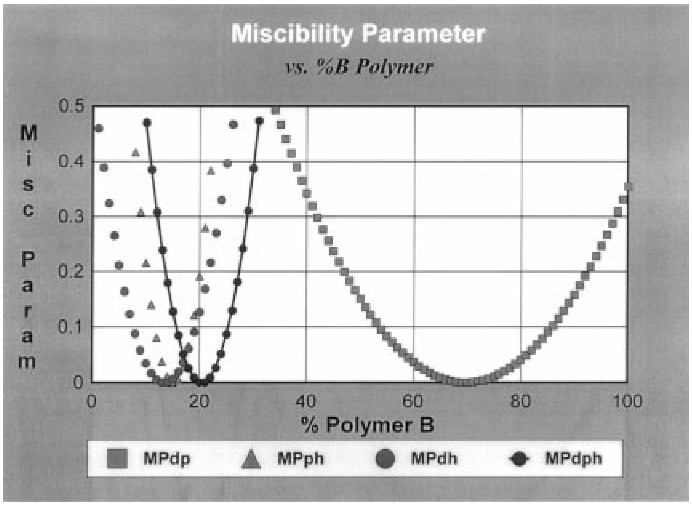


Fig. 9. Miscibility parameter (MP) vs. vinyl alcohol (%polymer B) in poly(vinyl butyral).

4. Conclusion

DMTA results for PVB /N6 blends show that a PVB containing $\cong 45$ wt% VA is immiscible, blends $\cong 55$–70 wt% VA in PVB are miscible, and a blend containing $\cong 89$ wt% VA in PVB is immiscible. These results are reinforced from DSC, SEM, melting and crystallization behavior, and physical properties, and show the estimated blend lower limit of miscibility to be about 55 wt% VA in VB while the upper limit of miscibility is about 70 wt% VA in VB. Thus blends of PVB/N6 exhibit thermodynamic miscibility in the ranges of approximately 0.5–0.65 volume fraction of vinyl alcohol in PVB.. The polymer interaction parameters, $\chi_{\mathrm{N6/VA}}$ and $\chi_{\mathrm{N6/VB}}$ were estimated using the PVB/N6 lower and upper miscibility limits and found to be $\sim$0.00 and 0.01, respectively. These values are consistent with the experimental data here and that reported previously. The MP model results also compared favorably with the miscibility window determined.

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