

Damping of Vinyl Acetate-*n*-Butyl Acrylate Copolymers

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ABSTRACT: A photopolymerization process at room temperature was devised to copolymerize vinyl acetate (VAc) and *n*-butyl acrylate (BA) mainly to prepare rubber-like damping sheet bearing pressure-sensitive adhesive property in this study. The investigations using both the differential scanning calorimeter and rheometric dynamic analysis show the existence of two glass transition temperatures for each copolymer. The scanning electron microscopic pictures reveal that the degree of microphase separation increases with increasing annealing time at 70°C. It was suggested that the rubbery domain (formed by the PBA blocks) disperses in the

glassy domain (constituted by the PVAc blocks), making an effective damping entity. Excellent damping was observed for the copolymer samples, with the $\tan\delta$ peak values as high as 1.76–1.80 at a certain temperature range and with $\tan\delta > 0.3$ at quite wide temperature ranges. In addition, the copolymers containing more VAc tend to have the higher damping. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 91: 1396–1403, 2004

Key words: vinyl acetate; *n*-butyl acrylate; copolymer; damping; pressure-sensitive adhesive

INTRODUCTION

Tremendous polymers and their blends are used as dampers^{1–3} that can be applied in the aerospace, automobile, building, and house appliance industries. These polymers and blends include: polyurethane (PU)/polymethacrylates and polyacrylates interpenetrating polymer networks (IPNs)⁴; IPNs based on acrylics, vinyls and styrenic mers⁵; blends of poly(vinyl chloride) (PVC), segmented poly(ether ester) (PEE), and poly(methyl acrylate) (PMA)⁶; Urethane/acrylate simultaneous semi-IPNs⁷; blends of butyl rubber (IIR) and poly(propylene) (PP)⁸; IPN of PU-modified epoxy and PU⁹; and blends of PVC/chlorinated polyethylene/epoxidized natural rubber.¹⁰

Attempts have been made to spread the damping range to develop materials with significant damping over a fairly wide temperature,⁶ where the phase separation behavior seems to play an important role in achieving this goal. Theoretically, the loss tangent (i.e., $\tan\delta$) ($= G''/G'$ = loss modulus/storage modulus) can be simply considered as a measure of damping and has the maximum value at the glass transition temperature (T_g).^{11,12} Table I summarizes the damping property of some materials. In case phase separation oc-

curs, two or more distinct T_g s may be observed. This may result in samples with higher damping over a wide temperature range. Therefore, having material with certain phase separations by either blending or copolymerization is usually adopted in preparing good damping materials.

A photopolymerization process was conducted to prepare a rubbery-like damping sheet having pressure-sensitive property in this study. Investigations on preparation and properties for these copolymers of vinyl acetate (VAc) and *n*-butyl acrylate (BA) were therefore undertaken.

EXPERIMENTAL

Materials

The monomers VAc (99% pure) and BA (98% pure) were obtained from Lancaster Synthesis Ltd., UK. The monomer acrylic acid (AA, 98% pure) was obtained from Tedia Company, Inc. (Japan). The photoinitiator, 2,2-diethyloxy acetophenone (DEAP, 99% pure), and crosslinker, methacrylic acid glycidyl ester (GMA, 99% pure), were obtained from Tokyo Chemical Industry Co., Ltd. (Japan). All the reagents were used without further purification.

Preparations

The sample sheets were prepared by photocopolymerizing VAc, BA, and AA, using an ultraviolet (UV) lamp ["Blak-ray," model B-100AP, supplied by UVP, featuring 100 W of high-powered longwave (365 nm)]. Two pieces of glass plates, fixed by clamps and using

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TABLE I
Composition of the Damping Data for Materials Reported at Different Sources

Composition of recipes	Tan δ peak value	Tan δ peak temp. ($^{\circ}$ C)	Temp. range ($^{\circ}$ C) for tan $\delta > 0.3$	Ref.
PU/PMMA (70/30)	0.55	80	20–125	4
PU/PMA (70/30)	0.41	15	–4–46	
PU/PEMA (70/30)	0.78	60	12–127	
PU/PEA (70/30)	0.74	20	–9–100	
PMA	0.48	32	30–45	6
PVC/PEE/PMA (22.7/68.2/9.1)	0.24	65	Not available	
PVC/PEE/PMA (20.8/62.5/16.7)	0.30	40	Not available	
PVC/PEE (25/75)	0.37	48	35–65	
Urethane/acrylate (1/0)	1.3	<25	Not available	7
Urethane/acrylate (0/1)	1.7	75	48–100	
Urethane/acrylate (2/1)	0.51	63	50–80	
Urethane/acrylate (2/1)				
Urethane containing 0.01% crystallization	0.7	39	<25–75	
PP/IIR (0/100)	1.55	–35	–68–0	8
PP/IIR (50/50)	0.37	–59	–58––50	
PU-crosslink epoxy (20%)/PU (Polyol MW 4000)	0.70	7	–41.7–77	9
PU-dangled epoxy (20%)/PU (Polyol MW 400)	0.64	55	–15–>65	

Abbreviations for polymers: PU, polyurethane; PMMA, poly(methyl methacrylate); PMA, poly(methyl acrylate); PEMA, poly(ethyl methacrylate); PEA, poly(ethyl acrylate); PVC, poly(vinyl chloride); PEE, poly(ether ester); PP, polypropylene; IIR, butyl rubber (isoprene–isobutylene rubber)

silicone rubber with thickness of 0.8 mm as the spacer, formed the reaction vessel, where both inner sides of the glass plates were lined with a releasing paper and a releasing biaxially oriented polypropylene (BOPP) film, respectively. The ingredients (Table II) were mixed in a beaker before transferring into the reaction vessel by a syringe. After injection, the spacer was

sealed. The distance between the reaction vessel and the UV lamp was set to 30 cm. After 3 h of radiation, the glass plates were removed, and the sample sheets were placed in the oven at 70 $^{\circ}$ C for assigned periods of time as indicated in Table II. The conversion was determined gravimetrically, and the final conversions were usually pretty high (as high as 98–99%).

TABLE II
Recipes

Recipe no.	VAc (wt %)	BA (wt %)	GMA (wt %)	AA (wt %)	DEAP (wt %)	70 $^{\circ}$ C oven time (h)
VB11-0	50	50	1	1	1	0
VB11-12	50	50	1	1	1	12
VB11-48	50	50	1	1	1	48
VB11-72	50	50	1	1	1	72
VB11-96	50	50	1	1	1	96
VB11-120	50	50	1	1	1	120
VB11-A2	50	50	1	2	1	72
VB11-G0	50	50	0	1	1	0, 12, 48, 72
VB11-G05	50	50	0.5	1	1	72
VB11-G075	50	50	0.75	1	1	72
B1-72	—	100	1	1	1	72
VB23-72	40	60	1	1	1	72
VB32-72	60	40	1	1	1	72
V1-72	100	—	1	1	1	72

Characterizations

Data for the dynamic mechanical properties, such as storage modulus (G'), loss modulus (G'') and $\tan\delta$, were determined by using a simple parallel disk-type rheometer (RDA, RMS 800, Rheometric Scientific). Both temperature scan [with a temperature rising rate of $5^\circ\text{C}/\text{min}$, at a constant frequency of 10 Hz (i.e., 62.8 radian/s)] and frequency scan (at 30°C) were conducted.

The phase transition was analyzed by using differential scanning calorimeter (DSC, TA-2010), and samples were scanned at a heating rate of $10^\circ\text{C}/\text{min}$ from -100 – 100°C .

The phase and microphase separations of the microtomed samples were observed by using a scanning electron microscope (SEM, Topcon ABT-150).

The shear strength of sample sheets was determined using a material testing instrument (HT-9401, Hungta Co., Taiwan), with a drawing rate of 200 mm/min. Both sides of sample sheets ($L \times W = 12.7 \times 30$ mm) were attached to unreleasing BOPP films by pressing with 2 kg roller for ten times, and the overlapped area of the lap shear specimen is 12.7×30 mm. The standard test method is referred to as Chinese National Standard (CNS).¹³

To analyze the peel strength, an 180° peel test was performed according to ASTM.¹⁴ Both sides of sample sheet (100×30 mm) were attached to unreleasing BOPP films by pressing with 2 kg roller for ten times. The procedure was to separate progressively one member from the other over the adhered surfaces at a separation angle of approximately 180° and at a separation rate of 20 mm/min. The average load per unit width of bond line required was taken to be the peel strength.

A falling ball method¹⁵ was applied to determine the tack of samples. A steel ball (7/16 inch diameter) was allowed to roll down the incline (21.6° to the horizontal surface), then the distance for the ball to travel horizontally across the adhesive before stopping was taken. The reciprocal of this value is considered as a measure of the tack.

A dissolution test process was devised to determine how far the crosslink reaction progressing. In the process, 1g of cured sample was placed in 10 cm^3 of toluene at room temperature, and the time it took to dissolve completely was recorded as the dissolution time.

RESULTS AND DISCUSSION

The block copolymers are expected to form when VAc (monomer 1) and BA (monomer 2) are copolymerized, since the values of reactivity ratios deviate greatly to each other (i.e., $r_1 = 0.018$ and $r_2 = 3.48$).¹⁶

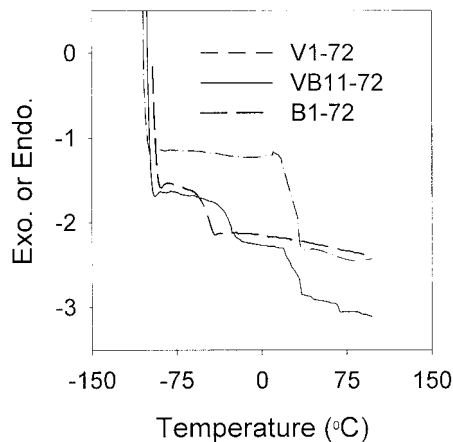


Figure 1 DSC curves [scan rate = $10^\circ\text{C}/\text{min}$ for copolymer (weight ratio of (VAc/BA) = 1/1] and its counterpart homopolymers.

Phase separation of block copolymers

It is well known that a random copolymer has a homogeneous phase and its T_g is intermediate between the T_g s of the corresponding homopolymers, and is predictable from the linear relationships such as the two-parameter empirical expression of Wood¹⁷ or the Gordon–Taylor equation.¹⁸ However, for the block copolymer, usually two T_g s are observed due to the occurrence of microphase separation. Thermodynamically, the preference of polymer molecules for their own environment (coil interpenetration) rather than for a mixed system stands for a positive ΔH (enthalpy change), outweighing the positive entropy change, ΔS (a much smaller magnitude). This leads to a positive ΔG (free energy change), and the two components will segregate and form separate phases for the block copolymer.¹⁹

The DSC and RDA results showed the existence of two glass-transition temperatures (T_g 's) for each copolymer. This reflects the occurrence of phase separation. The DSC result displays two T_g s for a copolymer with weight ratio of (VAc/BA) = 1/1, as shown in Figure 1. Two T_g s are identified by DSC for each copolymer with various weight ratios, as shown in Table III. Two T_g s for the VAc–BA copolymers are also observed by RDA (Table IV and Figure 2), but with considerable deviations from those by DSC on the values of glass-transition temperatures. It can be attributed to the tremendous deviation in thermal pulse frequency (0.1 and 62.8 radian/s, for DSC and RDA, respectively). According to the rule of thumb, the T_g value increases 6°C for each tenfold increase in the thermal frequency.

The samples were photopolymerized in bulk. The development of domain morphology (i.e., microphase separation) with increasing annealing time at 70°C is interesting. The SEM picture in Figure 3(a) shows a

TABLE III
Data of T_g Observed in DSC for Polymers with Different Weight Ratios of VAc to BA

Recipe no.	V1-72	VB32-72	VB11-72	VB23-72	B1-72
Wt. ratio (VAc/BA)	1/0	3/2	1/1	2/3	0/1
T_{g1} (°C)	—	-32	-37	-43	-53
T_{g2} (°C)	16	16	19	17	—

less distinct, more irregular structure for sample without annealing, while the striated structure becomes more evident with elapsing annealing time and eventually forms a more obvious striated pattern, as shown in Figure 3(b)–(d). Note that the annealing temperature is happened to be above the T_g of the hard segments ($\leq 68^\circ\text{C}$). The drying temperatures of solvent-cast films of A–B block copolymers were set coincidentally at slightly above the T_g of the hard segment too.^{20,21}

It is assumed the domain being the lamellar type, since the appearance of striated pattern and the equal monomer weight ratio of (VAc/BA) (VB11-G0). To have a clear conclusion, a further study using SEM and other techniques is required.

Distinguished damping performance of block copolymers

The damping of an elastomer is usually characterized by the $\tan\delta$ value, and the larger $\tan\delta$ value represents the better damping. The $\tan\delta$ peak values so far reported elsewhere are shown in Table I, mostly ranging from 0.5 to 0.7, except for the few cases having high values such as 1.55 and 1.7. Surprisingly, the $\tan\delta$ peak values can reach as high as 1.76–1.80 for copolymers prepared in this study (Table IV).

Another feature is also exhibited in Table IV. The effective damping temperature ranges (i.e., the temperature range for $\tan\delta > 0.3$) are quite broad for all the series of copolymers. On the other hand, the homopolymer (either PVAc or PBA) does not exhibit

such a continuous effective damping range from low to high temperatures, as shown in Table IV.

Curves for the RDA frequency scan at 30°C are shown in Figure 4. Copolymers containing the higher amounts of VAc have the larger $\tan\delta$ values over the range. This implies that copolymers containing the larger amount of VAc tend to have the better damping property.

It may be attributed to the phase separation effect leading to the unusual damping property. The phase (or microphase) separation has been discussed in the previous part in this report. Obviously the rubbery domain (formed by the PBA blocks) disperses in the glassy domain (constituted by the PVAc blocks), and the structure forms an effective damping entity.

When it comes to damping in applications, the temperature dependence of the damping at frequencies of the order of 100 Hz or more is also of considerable interest. However, measurement of damping data on any commercially available rheometer (such as the RDA series from Rheometrics and others) is limited to a narrow frequency range. Usually the high frequency used in RDA measurement is 100 rad/s, which is equivalent to 15.9 Hz.²² The high frequency used in our study is also restricted to 145 rad/s (i.e., 23 Hz).

In Figure 4, the $\tan\delta$ values are large at both low (0.01 Hz) and high (23 Hz) frequency ends, and the curves have minima at the middle frequencies.

As long as the time–temperature superposition applicable, the effect of frequency is the inverse of the effect of temperature, decreasing frequency being sim-

TABLE IV
Data of $\tan\delta$ Peak Value, Peak Temperature, and Temperature Range for $\tan\delta > 0.3$ for Different Samples

Recipe no.	Wt. ratio (VAc/BA)	Tan δ peak value		Tan δ peak temp. (°C)		Temp. range (°C) for $\tan\delta > 0.3$
		1st	2nd	1st	2nd	
V1-72	1/0	—	1.80	—	44	-70–-60 10–100
VB32-72	3/2	1.77	0.532	-2	62	-70–100
VB11-72	1/1	1.80	0.445	-7	62	-70–100
VB23-72	2/3	1.76	0.425	-12	68	-34–100
B1-72	0/1	2.33	—	-30	—	-47–14 71–100

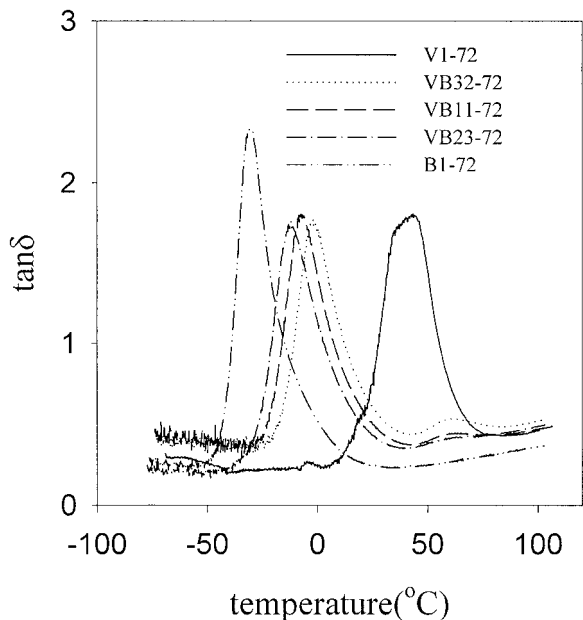


Figure 2 RDA curves (at scan rate of 5°C/min and a constant frequency of 10 Hz) of $\tan\delta$ vs temperature for copolymers with different (VAc/BA) weight ratios.

ilar to the effect of increasing temperature. The frequency scan was conducted at 30°C, which is located at the right-hand, down-hill side of the first $\tan\delta$ peak

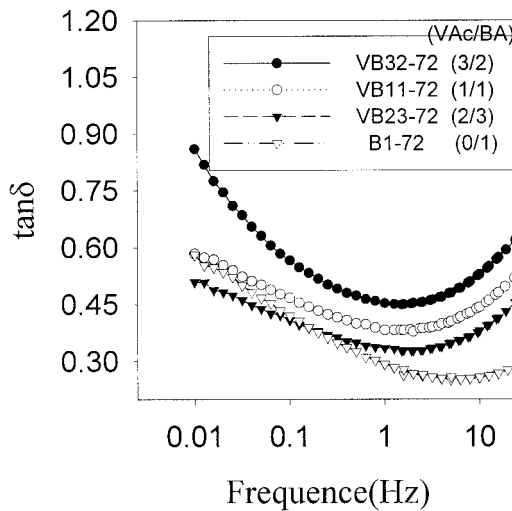


Figure 4 RDA frequency scan (at 30°C) of $\tan\delta$ for copolymer samples with various (VAc/BA) weight ratios (recipe no. B1-72, VB23-72, VB11-72, and VB32-72).

(e.g., the sample of VB11-72 on Fig. 2). Recall that increasing temperature, the $\tan\delta$ value decreases to the valley, then increases again as approaching the second $\tan\delta$ peak. The variation of $\tan\delta$ with decreasing frequency, starting from 10 Hz exactly follows the same pattern (Fig. 4).

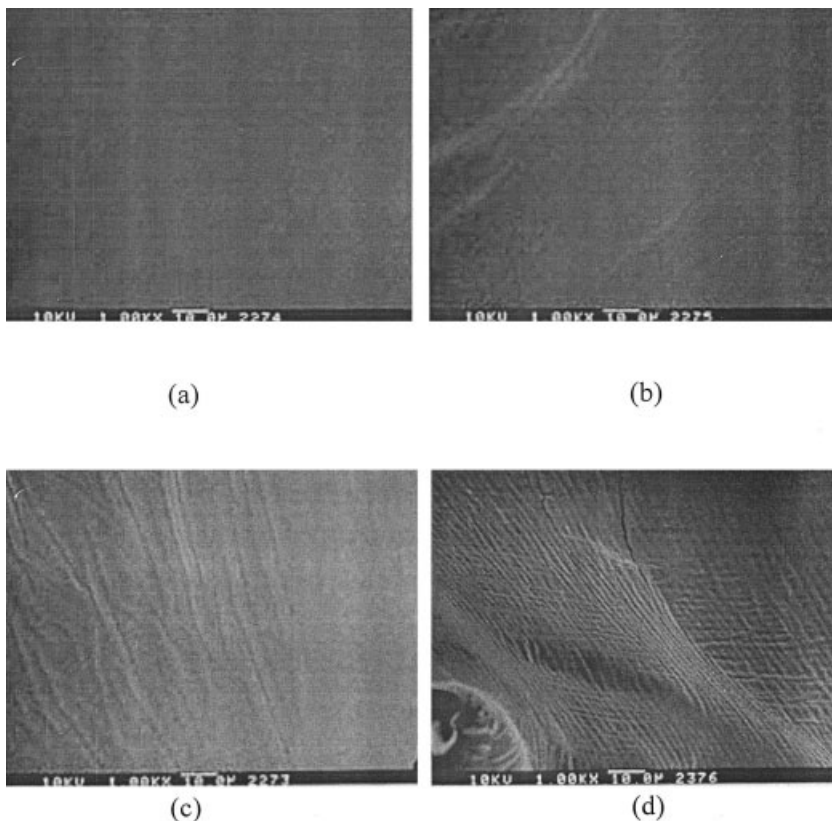


Figure 3 SEM pictures for sample prepared by using recipe no. VB11-G0 with various annealing times at 70°C: (a) 0 h, (b) 12 h, (c) 48 h, and (d) 72 h.

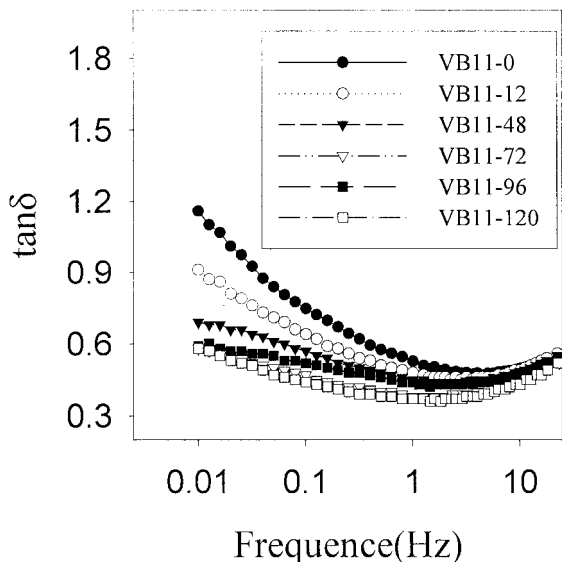


Figure 5 RDA frequency scan (at 30°C) of $\tan\delta$ for VB11 samples with various curing times.

Increasing the curing time at 70°C, the altitude of the RDA frequency scan curve decreases, as shown in Figure 5. That is, the increase of crosslink density of polymer by increasing the curing time can raise the storage modulus, and thus decrease the $\tan\delta$ value. Furthermore, the second $\tan\delta$ peak values are expected to be suppressed with increasing the curing time, since the $\tan\delta$ value at the lower frequency decreases with increasing the curing time, and the low-frequency results can be correlated to the high-temperature results. Therefore, increasing the curing time somewhat debilitates the damping.

Pressure-sensitive adhesive performance

The damping sheets prepared in this study have some pressure-sensitive adhesive (PSA) nature. The damping sheets have good pressure-sensitive performance comparable to the usual PSAs.²³ For instances, the damping sheets have better tack [rolling distance < 5 cm (Figs. 6 and 7), lower than the results showed by ref. 23 (ca. 10–20 cm)]. The peel and shear strength data are larger than those results in ref. 23, though tests might be subject to certain variations in procedure. The peel strength data in this study are between 200 and 1600 N/m, which are comparable to those of acrylic latex and staybelite ester blend PSAs (200–550 N/m), or carboxylated styrene-butadiene rubber (SBR)/resin blend PSAs (500–1100 N/m).²⁴ However, different substrates used in different systems can make considerable deviations.

Increasing the PBA content in the copolymer, the tack increases (i.e., the rolling distance decreases), the peel strength increases, and the shear strength de-

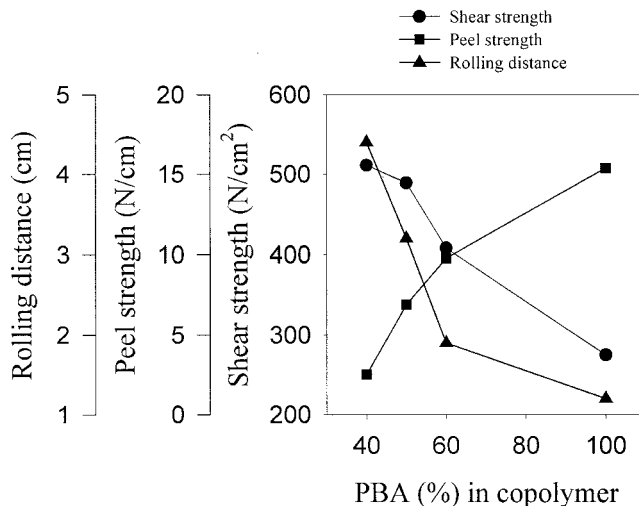


Figure 6 Pressure-sensitive adhesive performance of copolymers vs PBA (wt %) in copolymers, where all samples contained 1 wt % crosslinking agent (GMA) and were cured at 70°C for 72 h (recipe no. VB32-72, VB11-72, VB23-72, and B1-72).

creases, as shown in Figure 6. This can be attributed to the fact that having a very low T_g , the PBA blocks are relatively flexible at room temperature, and hence exhibit the higher wetting and mobility, but lower cohesion than the PVAc blocks. Note that the wetting and mobility play the important roles in deciding tack and peel strength, while the cohesion deciding the shear strength. Therefore, copolymers containing the higher amount of PBA have the better tack and peel strength, and the poor shear strength.

The balance between cohesion and adhesion affects not only the values of peel and shear strengths, but

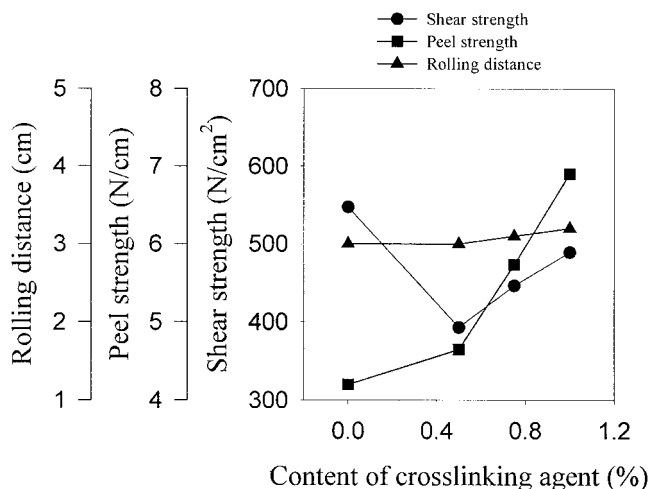


Figure 7 Pressure-sensitive adhesive performance of copolymers with (VAc/BA) weight ratio of (1/1) and various crosslinking agent contents, and cured at 70°C for 72 h [recipe no. VB11-G0(72), VB11-G05, VB11-G075, and VB11-72].

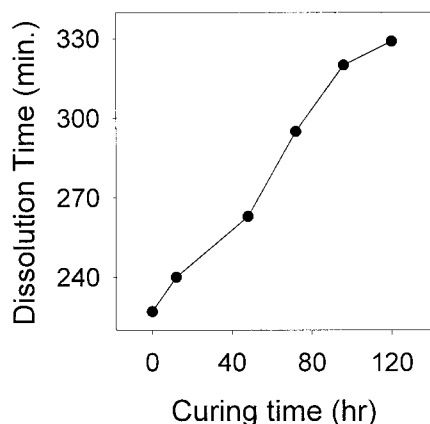


Figure 8 Samples experiencing the longer curing time show the longer dissolution time (recipe no, VB11-0, 12, 48, 72, 96, and 120).

also the detaching failure mode. If the cohesion of adhesive itself is high enough, the detaching occurs at the interface between substrate and the adhesive (i.e., an adhesive failure). On the other hand, if the cohesion is not strong enough, the failure occurs right within adhesive itself, and the "cohesive failure" happens.

In the peel test, all samples showed the cohesive failure except the one containing 40% of PBA. That is, increasing the PBA content, the cohesion is decreased, so that the detaching failure is shifted from the interface to the bulk of the adhesive. Meanwhile, in the shear test, there was only adhesive failure, and this may be attributed to the fact that the higher deformation rate is applied in the test.

Raise the crosslink density by increasing the crosslinking agent (i.e., crosslinker) content can enhance the cohesion of adhesive, and shift the detaching failure mode from cohesive failure to adhesive failure. This can be illustrated by the shear strength curve shown in Figure 7, where the cohesive failure occurs only for the sample containing no crosslinking agent, else there is adhesive failure; and the curve decreases first to the minimum (where crosslinking agent content = 0.5%), then increases again.

With decreasing the content of crosslinking agent, the peel strength decreases (where the cohesive failure mode prevails), and the tack almost keeps as constant, as shown in Figure 7.

TABLE VI
Effect of Acrylic Acid Content

Recipe No.	VB11-72	VB11-A2
Amount of AA (% wt)	1.0	2.0
Rolling distance (cm)	3.0	3.0
Peel strength (N/cm)	8.07 ^a	7.15 ^a
Shear strength (N/cm ²)	538 ^b	503 ^b

^a Cohesive failure.

^b Adhesive failure.

Increasing the curing time, the crosslink density is increased. As illustrated in Figure 8, the dissolution time increases (i.e., the degree of crosslink increases) with the curing time, but obviously values of the degree of crosslink are still limited (i.e., all the samples are still dissolvable in toluene) after curing at 70°C for the assigned period of time. The results for increasing the curing time are shown in Table V.

The tack values are all located within the error range; the peel strength increases until reaching the maximum, then decreases to a certain value, and increases again; and the shear strength increases continuously with the curing time. There is cohesive detaching failure at lower curing times, and becomes adhesive failure at certain curing times for both the peel and shear strength tests.

Increasing the acrylic acid content from 1.0 to 2.0 g in the recipe, the effect on the pressure-sensitive adhesive properties is not evident at all, as shown in Table VI.

CONCLUSIONS

The DSC and RDA results present two glass transition temperatures for each copolymer of VAc and BA. The SEM pictures show that the degree of microphase separation increases with increasing the annealing time at 70°C. It was suggested that the rubbery domain (formed by the PBA blocks) disperses in the glassy domain (constituted by the PVAc blocks), making an effective damping entity. The copolymer samples prepared exhibit significantly high damping at certain temperature range (the $\tan\delta$ peak values can

TABLE V
Curing Time Effect (Recipe VB-11 Series)

Curing time (h) at 70°C	0	12	48	72	96	100
Rolling distance (cm)	3.0	3.1	3.1	3.0	3.0	3.1
Peel strength (N/cm)	1.44 ^a	2.09 ^a	8.62 ^a	8.07 ^a	4.87 ^b	6.37 ^b
Shear strength (N/cm ²)	354 ^a	403 ^a	518 ^b	538 ^b	546 ^b	618 ^b

^a Cohesive failure.

^b Adhesive failure.

reach as high as 1.76–1.80) and pretty broad temperature ranges for $\tan\delta > 0.3$, and the copolymers containing more VAc have the larger damping. The sample sheets prepared also exhibit some pressure-sensitive adhesive nature.

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