

Effects of Miscibility and Viscoelasticity on Shear Creep Resistance of Natural-Rubber-Based Pressure-Sensitive Adhesives

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ABSTRACT: Natural rubber (NR) was blended in various ratios with 29 kinds of tackifier resins. Miscibilities of all the blend systems were illustrated as phase diagrams. From these blend systems, we selected 8 systems having typical phase diagrams (completely miscible, immiscible, lower critical solution temperature [LCST] types) and carried out measurements of shear creep resistance (holding power). Holding time was recorded as required time for the pressure-sensitive adhesive (PSA) tape under shear load to completely slip away from the adherend. Holding time of miscible PSA systems tended to decrease as the tackifier content increased. This is attributable to a decrease in plateau modulus of the PSA with increasing tackifier content. There was rather large difference in holding time by tackifier among the miscible PSA systems; the reason for this is also considered to be a difference in plateau modulus. Holding time of an immiscible PSA system scarcely changed by tackifier content. But in another immiscible system, holding time tended to increase with increasing tackifier content. In fact, in the case of immiscible PSAs, the effect of tackifier content on holding time was different from tackifier to tackifier. This may be caused by difference in extent of phase separation. © 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 75: 1535–1545, 2000

Key words: natural rubber; tackifier; pressure-sensitive adhesive (PSA); holding power; plateau modulus

INTRODUCTION

Pressure-sensitive adhesive (PSA) tapes can paste on various adherends under light pressure in very short time without heating or heavy pressure. The bonds formed are strong enough to use

temporarily. However, PSAs sometimes need durability for a long time, especially the resistance to shear force. For example, PSAs are used for bookbinding, upholstery, and pasting papers on the wall.¹

The shear creep test indicates the resistance of an adhesive to shear force. It is measured by the static shear test that determines the time (holding time) required to completely pull away a defined area of a tape from the test panel under a

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Table I Tackifiers Used for Measurement of Holding Power

Phase Diagram	Raw Materials	Commercial Names of Tackifiers	T_g (°C)	M_n	Main Components
Completely miscible	Rosin or terpenes	Estergum HP ^a	54.4	685	Pentaerythritol ester of hydrogenated rosin
		Superester A-75 ^a	46.6	682	Disproportionated rosin esterified by glycerol, diethyleneglycol
		YS polystar T-130 ^b	78.7	765	Terpene phenolic copolymer (phenol 25%)
LCST	Rosin or terpenes	Clearon K-4090 ^b	41.8	793	Hydrogenated terpene resin
	Petroleum	ESCOREZ 5320 ^c	75.2	395	Hydrogenated petroleum resin
Completely immiscible	Rosin or terpenes	Polypale ^d	60.2	442	25% polymerized rosin
		Estergum AAG ^a	75.7	921	Glycerol ester of rosin
	Petroleum	Kristalex 1120 ^d	68.1	873	Polymer from pure aromatic monomer

^a Supplied by Arakawa Chemical Co., Ltd. (Osaka, Japan).

^b Supplied by Yasuhara Chemical Co., Ltd. (Fuchu-shi, Hiroshima, Japan).

^c Supplied by Tonex Co., Ltd. (Kawasaki-shi, Japan).

^d Supplied by Hercules Co., Ltd. (Wilmington, DE).

constant shear force. Generally, shear creep resistance (holding power) is related to low rate (long time) and large deformation process. PSAs are highly viscoelastic, and their practical performances have intimate correlation with the viscoelasticity of the bulk of the adhesive, and with the surface properties of the adhesive and the adherend. It is generally admitted that shear creep resistance is strongly dependent on the dynamic mechanical properties of PSAs.²⁻⁴ Moreover, miscibility between the components has a great influence on performances of PSAs. However, there are few studies that explain the relationship of miscibility, viscoelasticity, and shear creep resistance of PSAs.

In this article, effects of miscibility of PSA components and dynamic mechanical properties on shear creep resistance are discussed.

EXPERIMENTAL

Natural rubber (NR) was used as the elastomer and tackifier resins used were Superester A-75,

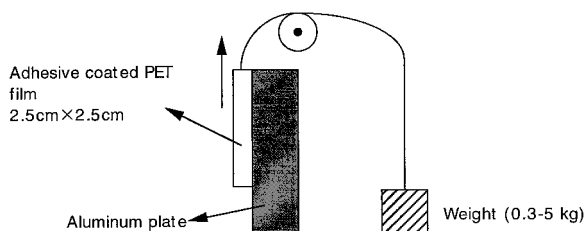


Figure 1 Measuring method of shear creep resistance.

Estergum HP, YS polystar T-130, ESCOREZ 5320, Clearon K-4090, Polypale, Kristalex 1120, and Estergum AAG. The characteristics of these tackifiers are listed in Table I. NR was blended with each tackifier in toluene solutions in nine different solute weight ratios (9 : 1 ~ 1 : 9).

PSA sheets for the measurement of shear creep resistance were prepared by coating the blend solutions on corona-treated polyethylene terephthalate (PET) films of 25 μm thickness, using our laboratory coating device. The thickness of the PSA layer was adjusted to 20 μm (dry state) by a film thickness gauge. The PSA sheets thus obtained were air dried at room temperature for 48 h and then dried in a vacuum oven at 40°C for 48 h. Dried PSA sheets were pressed onto release

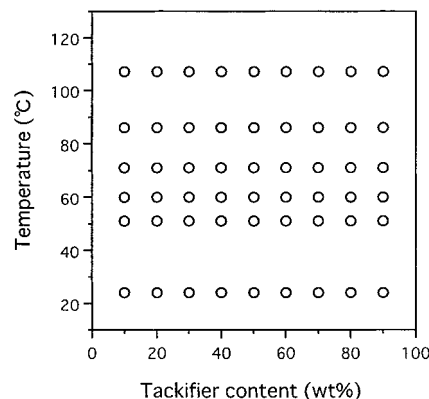


Figure 2 Phase diagram of NR/Superester A-75 system. \circ , transparent.

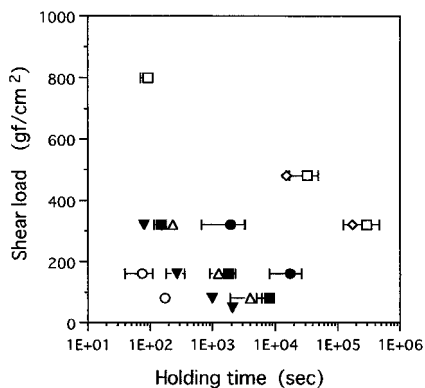


Figure 3 Holding power of NR/Superester A-75 blends at 30°C. Tackifier content: □, 0%; ◇, 10%; ●, 20%; △, 30%; ■, 40%; ○, 50%; ▼, 60%.

coating papers using a 2-kg roller. The sheets were seasoned at 20°C and 65% relative humidity (RH) for more than 14 days, and then cut into strips of 3 cm × 8 cm (PSA tapes). The adherends were aluminum plates that had pasting area of 2.5 cm × 2.5 cm. They were cleaned with toluene beforehand. The PSA tape was pressed on the pasting area of the aluminum plate with a 2-kg roller.

The specimen was fixed vertically in the Six-specimens Creep tester (type CP6-L-500, Orientec Co. Ltd., Tokyo, Japan) and the static shear force was loaded to the tape parallel to the surface of bonding as shown in Figure 1. The holding time was recorded as the time required for the adhesive tape to completely slip away from the aluminum plate. The measurements were performed

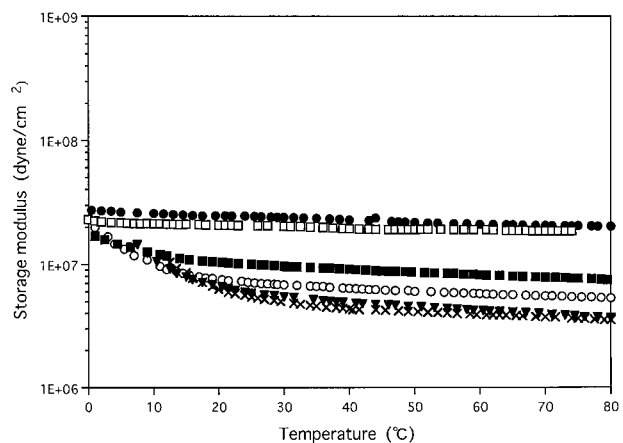


Figure 4 Shear storage moduli of NR/Superester A-75 blends. Tackifier content: □, 0%; ●, 20%; ■, 40%; ○, 50%; ▼, 60%; ×, 70%.

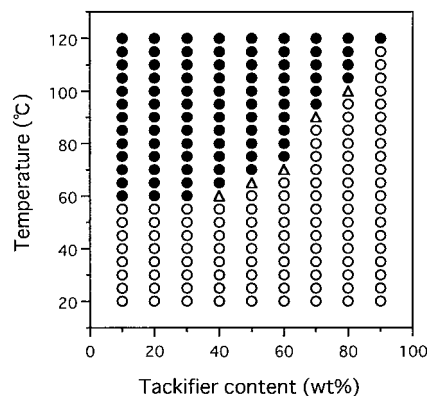


Figure 5 Phase diagram of NR/ESCOREZ 5320 system. ○, transparent; ●, opaque; △, semitransparent.

under various shear force (0.3 ~ 5 kg) and at four different temperatures (30, 40, 50, 60°C), but most of the measurements were carried out at 30°C. Measurements were conducted twice or more for the same PSA composition and under the same conditions, and the average holding time was calculated.

Dynamic mechanical properties of PSAs were measured by Rheovibron DDV III (Toyo-Baldwin Co., Ltd., Tokyo, Japan) in the shear mode at the frequency of 11 Hz and heating rate of about 1°C/min from -50°C to 80°C. PSA plates for the measurement were 1 cm × 1 cm × (2-3 mm) prepared by casting PSA solutions in dish-shaped release coating papers. Before the measurement, two PSA plates were inserted between the steel plates of the jig, and the specimen was attached to the machine.

PSA samples for microscopic observation were prepared by coating PSA solutions on slide

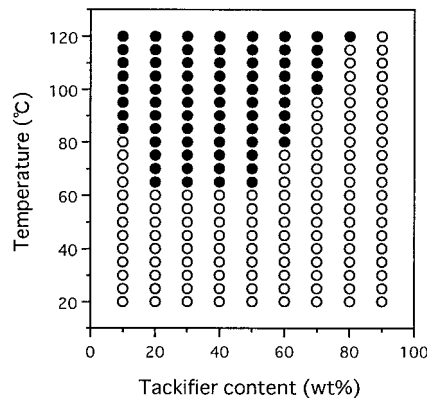


Figure 6 Phase diagram of NR/Clearon K-4090 system. ○, transparent; ●, opaque.

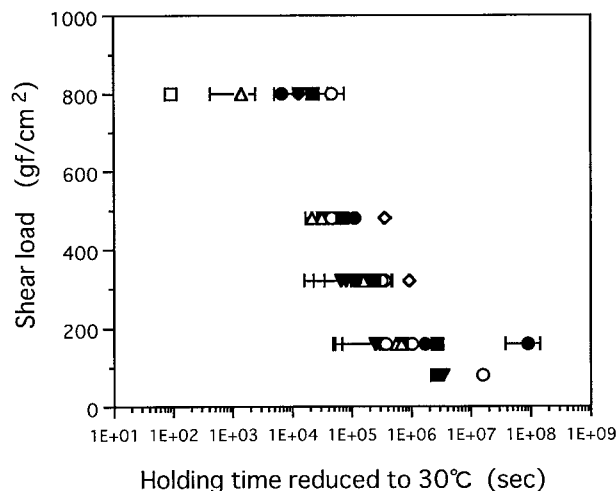


Figure 7 Holding power of NR/ESCOREZ 5320 blends. Tackifier content: \square , 0%; \diamond , 10%; \bullet , 20%; \triangle , 30%; \blacksquare , 40%; \circ , 50%; \blacktriangledown , 60%.

glasses and by vacuum drying. Photographs of the samples were taken with a camera (Olympus PM-10AK) equipped with a microscope (Olympus BX50) under magnification of 400-fold at room temperature.

RESULTS AND DISCUSSION

Shear Creep Resistance of Miscible PSAs

NR/Superester A-75 system shows the phase diagram of completely miscible (miscible at any

temperature and blend ratio of experiment) as shown in Figure 2. Holding power of NR/Superester A-75 system at 30°C is shown in Figure 3. Holding time decreases rapidly as the shear load increases. Holding time of the system tends to decrease as the tackifier content increases. This is attributable to decrease in plateau modulus of the blends with increasing tackifier content, as shown in Figure 4.² As will be discussed in detail in the latter part of this paper, plateau modulus often indicates the shear storage modulus at the temperature of minimum in $\tan \delta$. But plateau modulus in the following sentences mainly indicates the modulus at about 70°C if the temperature of minimum in $\tan \delta$ is lower than it, because holding power was measured at 30°C or higher temperatures (anyway, the difference is very small). According to Toyama,⁵ in the miscible PSAs, tackifier is mixed with NR at the molecular level, thus the tackifier molecules act as rollers to reduce density of entanglements of NR molecules, which is reflected to plateau modulus. All the PSA samples of NR/Superester A-75 system (except for NR 100%) broke in cohesive failure.

NR/ESCOREZ 5320 and NR/Clearon K-4090 systems show phase diagrams of lower critical solution temperature (LCST) type, but are miscible at 30–50°C as shown in Figures 5 and 6. Holding powers and storage moduli of these systems are shown in Figures 7–10. In the case of NR/ESCOREZ 5320 system, holding time measured at 40, 50, and 60°C is reduced to the values at 30°C using time-temperature superposition

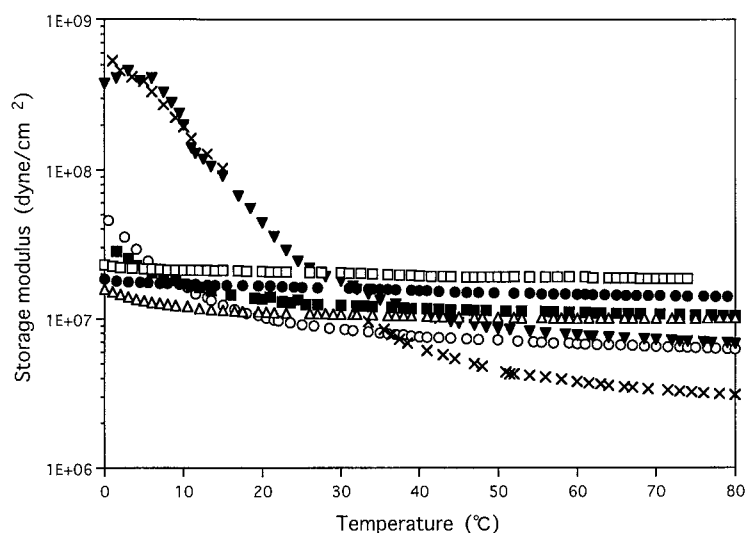


Figure 8 Shear storage moduli of NR/ESCOREZ 5320 blends. Tackifier content: \square , 0%; \bullet , 20%; \triangle , 30%; \blacksquare , 40%; \circ , 50%; \blacktriangledown , 60%; \times , 70%.

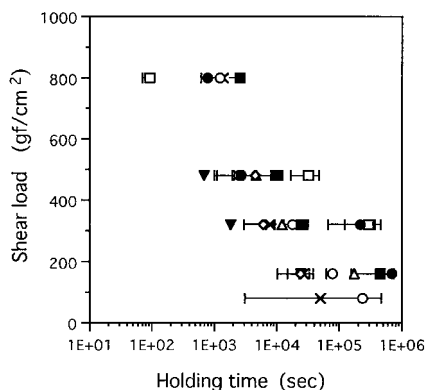


Figure 9 Holding power of NR/Clearon K-4090 blends at 30°C. Tackifier content: □, 0%; ◇, 10%; ●, 20%; △, 30%; ■, 40%; ○, 50%; ▼, 60%; ×, 70%.

principle.⁶ Similar to NR/Superester A-75 system, holding time of NR/ESCOREZ 5320 and NR/Clearon K-4090 systems tends to decrease as the tackifier content increases, though some inversions are seen under large shear load. It is also caused by decrease in plateau modulus of these blends with increasing tackifier content. There was a rather large difference in holding time by tackifier among the miscible PSA systems. As a whole, the holding time of NR/Clearon K-4090 blends is a little shorter than that of NR/ESCOREZ 5320 blends, and the holding time of NR/Superester A-75 system is much shorter than that of these two systems. The reason for this is also considered to be a difference in plateau mod-

ulus as shown in Figure 11; the order of plateau modulus is NR/Superester A-75 < NR/Clearon K-4090 < NR/ESCOREZ 5320 systems. PSAs of NR/ESCOREZ 5320 and NR/Clearon K-4090 blends often caused interfacial failure, especially for the samples containing 10–20% of tackifier or under large shear load. At the tackifier content of 10–20% it took very long time for the PSA tape to begin to slip, but once the PSA film began to slip, it slipped away from the aluminum plate in a relatively short time. The PSA tapes containing 50% or more tackifier began to slip as soon as the tests started, but took a long time to slip away from the adherend.

Similar results were obtained from other miscible NR-based PSAs: holding powers of other miscible PSAs had a tendency to decrease as the tackifier content increased. It was because plateau modulus decreased with increasing tackifier content. Holding time was in the following order at the same tackifier content under the same conditions as a whole; NR/ESCOREZ 5320 \approx YS polystar T-130 > Clearon K-4090 \gg Superester A-75 \approx Estergum HP blends. Most of the samples of NR/YS polystar T-130 and NR/Estergum HP blends showed cohesive failure.

The tendency that holding time decreases with increasing tackifier content is true of only miscible NR-based PSAs. Holding time of miscible acrylic PSAs tends to be longer as the tackifier content increases.³ In the case of block copolymer type of PSAs (SIS, SBS), holding time decreases

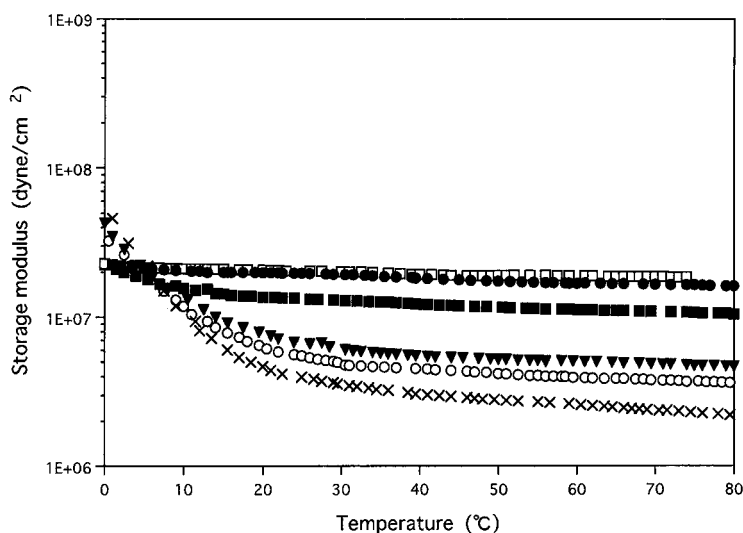


Figure 10 Shear storage moduli of NR/Clearon K-4090 blends. Tackifier content: □, 0%; ●, 20%; ■, 40%; ○, 50%; ▼, 60%; ×, 70%.

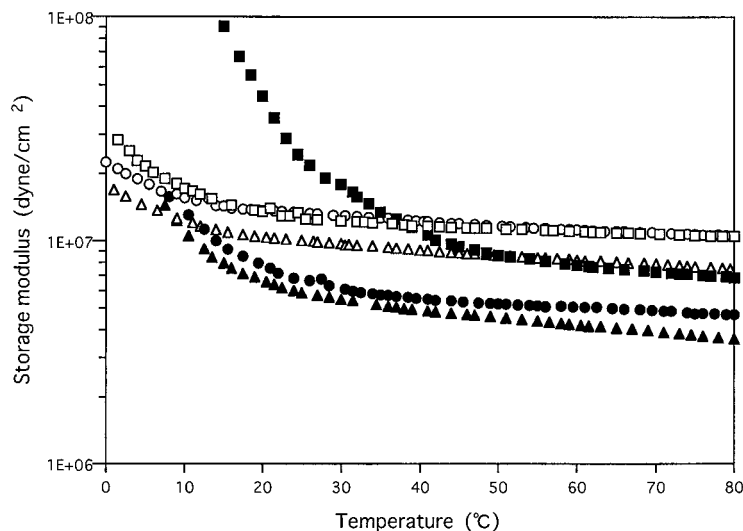


Figure 11 Shear storage moduli of miscible PSAs. □, ESCOREZ 5320 40%; ■, ESCOREZ 5320 60%; ○, Clearon K-4090 40%; ●, Clearon K-4090 60%; △, Superester A-75 40%; ▲, Superester A-75 60%.

by addition of a tackifier that is miscible with elastomer block, though it increases by addition of a tackifier miscible with styrene block.⁷ In short, the holding power increases as the plateau modulus of the PSA increases.

Shear Creep Resistance of Immiscible PSAs

NR/Kristalex 1120 system is completely immiscible (immiscible at any temperature and blend ratio of experiment) as shown in Figure 12. Holding powers of NR/Kristalex 1120 blends are listed in Table II. The holding time of this PSA system is very long (over 10^5 second) at 30°C under the load of 160–480 gf/cm^2 , and some of the PSA tapes did not slip at all in 20 days. Therefore,

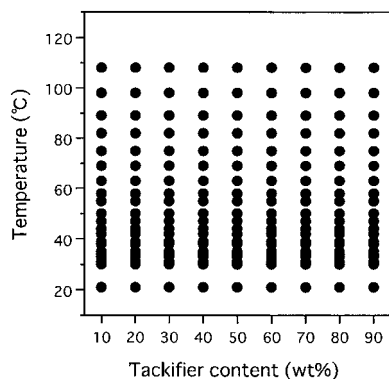


Figure 12 Phase diagram of NR/Kristalex 1120 system. ●, opaque.

most of the measurements of holding time of the system were performed at elevated temperatures (40°C, 50°C) under large shear load (480 gf/cm^2 or more). The result that the holding time of NR/Kristalex 1120 blends is very long was unexpected from peel strength and probe tack of this system.^{8,9} The storage moduli of NR/Kristalex 1120 blends are shown in Figure 13. The plateau moduli of the blends are higher than that of NR itself, and they increase as the tackifier content increases. This can be an explanation of the result that NR/Kristalex 1120 blends generally give longer holding time than NR itself under the same condition. Thus, holding time of typical immiscible PSAs such as NR/Kristalex 1120 blends is longer than that of NR itself. And holding time of immiscible PSAs should increase theoretically as the tackifier content increases, because of increase in plateau modulus. But the holding time does not always increase with increasing tackifier content (or plateau modulus) in NR/Kristalex 1120 system. At 40°C under the same load, the difference of the holding time by tackifier content is almost negligible when shear load is plotted against log (holding time), as shown in Figure 14. At 50°C under 480 gf/cm^2 , holding time of the system decreases as the tackifier content increases (Table II). The PSAs of this system often caused interfacial failure, especially at the tackifier content of 10 and 20%.

Table II Holding Time of NR/Kristalex 1120 Blends

Tackifier Content (%)	Shear Load	30°C	40°C	50°C
0	160 gf/cm ²	$> 6.26 \times 10^5$	8.64×10^5	4.60×10^5
	480 gf/cm ²	3.24×10^4	2.70×10^4	8.70×10^3
	800 gf/cm ²	92.5	—	—
10	160 gf/cm ²	$> 1.74 \times 10^6$	$> 1.10 \times 10^6$	$> 1.03 \times 10^5$
	480 gf/cm ²	$> 8.75 \times 10^5$	9.10×10^4	1.77×10^4
	800 gf/cm ²	1.10×10^4	4.41×10^3	—
20	160 gf/cm ²	$> 1.74 \times 10^6$	$> 1.10 \times 10^6$	$> 1.03 \times 10^5$
	480 gf/cm ²	—	8.58×10^4	1.55×10^4
	800 gf/cm ²	7.02×10^3	6.06×10^3	—
30	480 gf/cm ²	—	6.14×10^4	1.39×10^4
	800 gf/cm ²	1.01×10^4	1.24×10^3	—
40	160 gf/cm ²	$> 1.13 \times 10^6$	4.99×10^5	—
	480 gf/cm ²	2.46×10^5	1.26×10^5	1.00×10^4
50	480 gf/cm ²	—	6.04×10^4	9.90×10^3
60	480 gf/cm ²	—	2.69×10^5	3.07×10^3

The holding time and storage moduli of NR/Estergum AAG system are shown in Figures 15 and 16, respectively. Holding time of this system scarcely changes irrespective of tackifier content up to 60%, and the holding time of the blends is as long as that of NR itself. But the holding time of the blend including 70% of Estergum AAG is much longer than that of the blends containing 10–60% of the tackifier. Holding time of the blend

(Estergum AAG 70%) was 3.14×10^4 sec at 40°C under 320 gf/cm². Most of the samples of NR/Estergum AAG system caused cohesive failure. By examining at Figure 15 carefully it can be seen that the holding time is the second longest at the tackifier content of 20%, and that it is shortest at 40–50%. The plateau modulus of the system increases with increasing tackifier content from 40 to 70%, however, the change of the plateau mod-

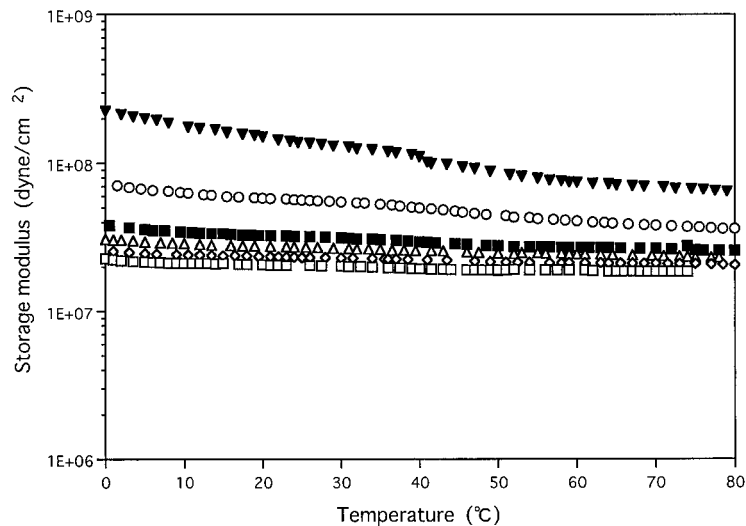


Figure 13 Shear storage moduli of NR/Kristalex 1120 blends. Tackifier content: □, 0%; ◇, 10%; △, 30%; ■, 40%; ○, 50%; ▼, 60%.

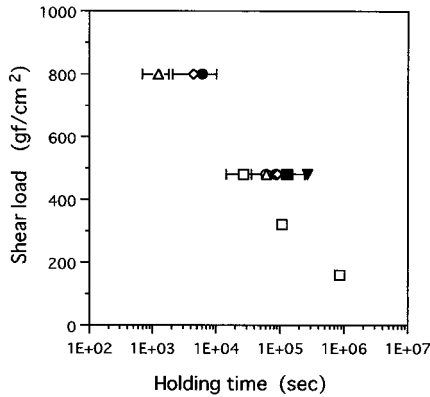


Figure 14 Holding power of NR/Kristalex 1120 blends at 40°C. Tackifier content: □, 0%; ◇, 10%; ●, 20%; △, 30%; ■, 40%; ○, 50%; ▼, 60%.

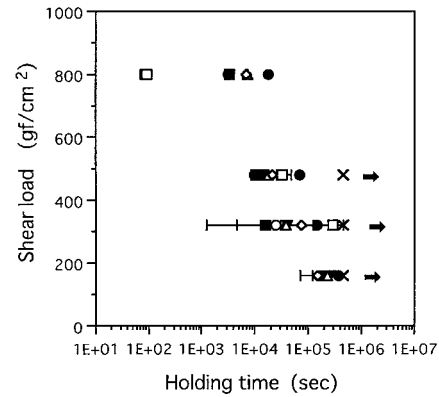


Figure 15 Holding power of NR/Estergum AAG blends at 30°C. Tackifier content: □, 0%; ◇, 10%; ●, 20%; △, 30%; ■, 40%; ○, 50%; ▼, 60%; ×, 70%; →, holding time of the blend containing 70% of the tackifier was longer than the data point of “×”.

ulus is smaller than that of NR/Kristalex 1120 system. As the holding time generally increases with increasing plateau modulus, the holding time of NR/Estergum AAG system should be in the order of tackifier 0–40% < 50% < 60% < 70%. It is certain that both holding power and plateau modulus are the highest at the tackifier content of 70%. However, this theoretical prediction does not always agree with the experimental data of this system. The reason is not clear, but we think as follows: as the plateau modulus of a PSA increases, the cohesion force (resistance to shear force) increases, however, the wetting to the adherend (adhesion force) decreases and the slip will be easy to take place at the interface between the adhesive and adherend.

The holding time of NR/Polypale blends tends to increase as the tackifier content increases up to 50%, but it is shorter than that of NR itself, as shown in Figure 17. The holding time of the PSA containing 60% of Polypale is a little shorter than that of the blend containing 50% of Polypale. Of course, the holding time tends to decrease as the shear load increases, but the data show rather large scattering and inversion. The storage moduli of NR/Polypale blends are shown in Figure 18. The plateau modulus of this system tends to decrease with increasing tackifier content as if this blend system were miscible. All of the samples of this system caused cohesive failure.

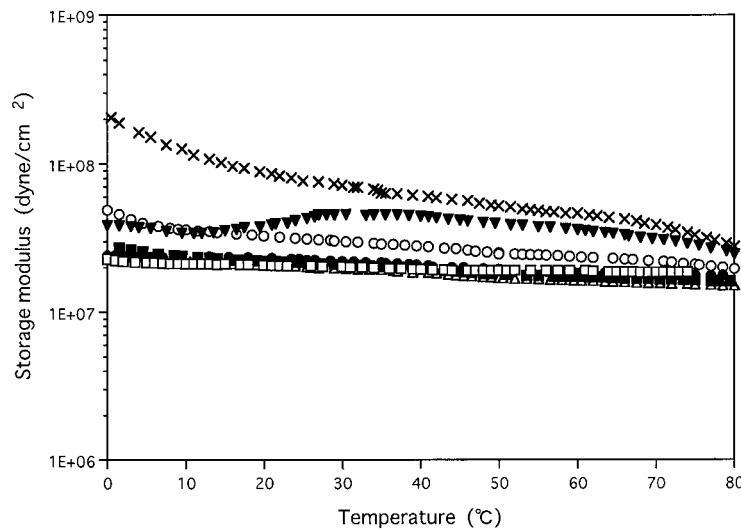


Figure 16 Shear storage moduli of NR/Estergum AAG blends. Tackifier content: □, 0%; ●, 20%; △, 30%; ■, 40%; ○, 50%; ▼, 60%; ×, 70%.

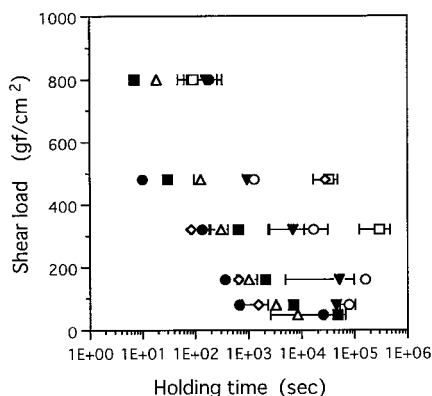


Figure 17 Holding power of NR/Polypale blends at 30°C. Tackifier content: □, 0%; ◇, 10%; ●, 20%; △, 30%; ■, 40%; ○, 50%; ▼, 60%.

Why is the effect of tackifier content on holding time different from tackifier to tackifier in immiscible PSAs? One of the possible reasons may be the difference in extent of phase separation. The micrographs of NR/Polypale, NR/Kristalex 1120, NR/Estergum AAG blends are shown in Figures 19–21, respectively. As shown in Figure 19, in NR/Polypale blends, very small dot-like particles of dispersed phase are seen under the magnification of 400-fold. Dispersed phase of NR/Kristalex 1120 blends is a little larger than that of NR/Polypale blends (Figure 20). On the contrary, the dispersed phase of NR/Estergum AAG blends is much larger, as shown in Figure 21. From these photos, it may be considered that the extent of

phase separation is in this order: NR/Polypale < NR/Kristalex 1120 < NR/Estergum AAG blends. NR/Polypale blends are not so phase-separated and may be miscible at the interface of the two phases. Consequently, the plateau modulus decreases with increasing tackifier content and holding time of the blends is shorter than that of NR itself. NR/Kristalex 1120 blends are fairly phase-separated and the tackifier particles act as a packing agent that go into between NR molecule chains and increase friction and entanglement between NR molecules; as a result, they increase plateau modulus and holding time.¹⁰ Because NR/Estergum AAG blends may be seriously (semi-macro) phase-separated and the dispersed phase is too large, tackifier phase of the blends acts only as a filler and does not change the mechanical properties of the matrix phase so much; almost the same holding time irrespective of tackifier content (0–60%). These discussions are just our opinion on the basis of experiment.

The extent of phase separation is also evaluated from plateau modulus. According to Kraus and Rollman,¹¹ quasiequilibrium shear modulus of an entanglement network is given by application of the theory of rubber elasticity as

$$G_{eN}^0 = (\rho/M_e)RT \quad (1)$$

where ρ is the density, M_e is the entanglement spacing molecular weight, R is the gas constant, and T is the temperature. If an immiscible filler is

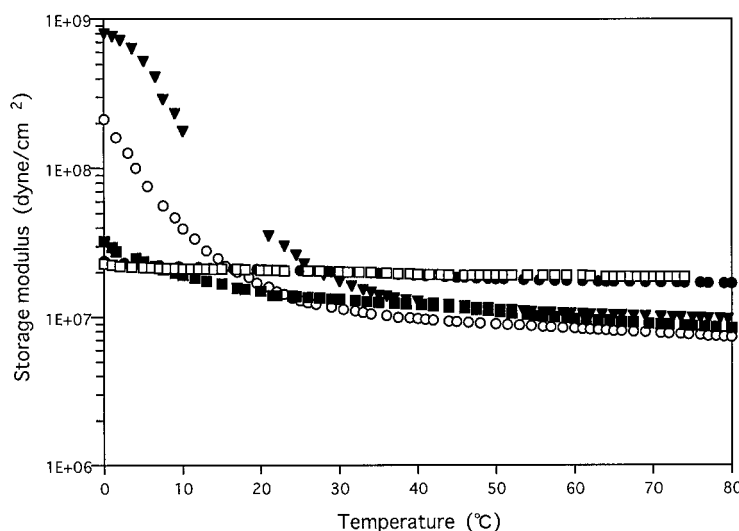


Figure 18 Shear storage moduli of NR/Polypale blends. Tackifier content: □, 0%; ●, 20%; ■, 40%; ○, 50%; ▼, 60%.

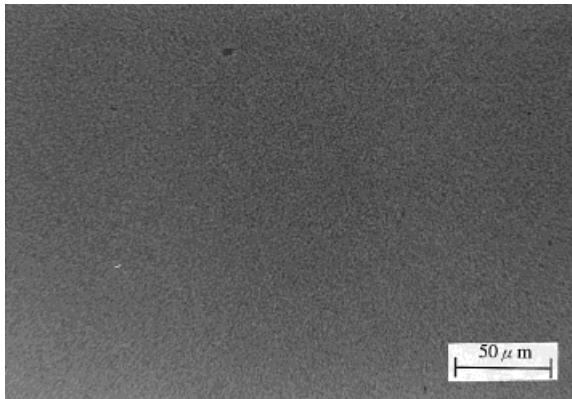


Figure 19 Micrograph of NR/Polypale 5 : 5 blend at room temperature ($\times 400$).

added to the elastomer (for example, polystyrene domains in SIS block polymer), shear modulus of the mixture is expressed as follows by Guth and Gold equation^{12,13}:

$$G_{eN}^0 = (\rho/M_e)RT(1 + 2.5c + 14.1c^2) \doteq G'(\tan \delta_{\min}) \quad (2)$$

where c is the filler volume fraction and T is the temperature at the minimum in $\tan \delta$. $G'(\tan \delta_{\min})$ is the shear storage modulus at the temperature of minimum in $\tan \delta$, and this value is treated as “plateau modulus” in Kraus and Rollmann.¹¹ If a miscible diluent (tackifier) is further blended with the elastomer/filler mixture, its effect on the plateau modulus is described as

$$G'(\tan \delta_{\min}) = v_2^2(\rho/M_e)RT(1 + 2.5c + 14.1c^2) \quad (3)$$

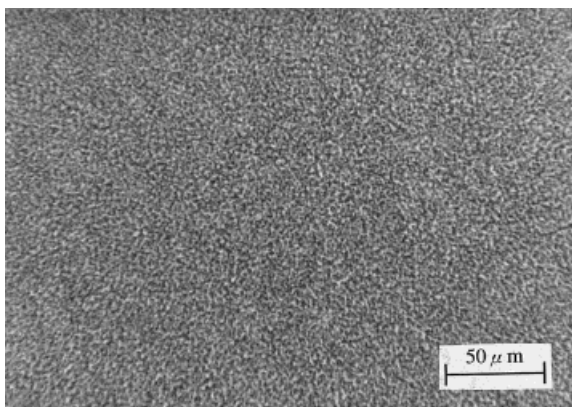


Figure 20 Micrograph of NR/Kristalex 1120 6 : 4 blend at room temperature ($\times 400$).

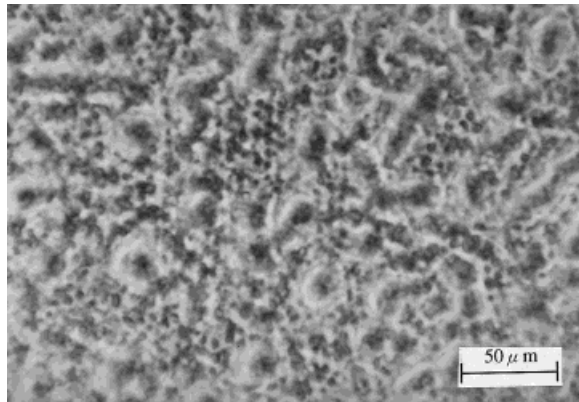


Figure 21 Micrograph of NR/Estergum AAG 6 : 4 blend at room temperature ($\times 400$).

where v_2 is the volume fraction of polymer in the rubbery phase alone, whereas c is the volume fraction of the filler in the entire composition. Also, ρ/M_e is the entanglement density of the unmixed (pure) elastomer. When this equation is applied to miscible NR/tackifier blends, $c = 0$ in eq. (3).

$$G'(\tan \delta_{\min}) = v_2^2(\rho/M_e)RT \quad (4)$$

In the case of completely immiscible NR/tackifier blends, eq. (2) is available. As the shear modulus of NR calculated from eq. (1) did not match with experimental value, we use the experimental modulus of NR (2.15×10^7 dyne/cm²) as $(\rho/M_e)RT$ to evaluate miscibility here.

Weight fraction of NR can be used instead of volume fraction, because the density of NR coated on a PET film was 1.1478 g/cm³ and that of any

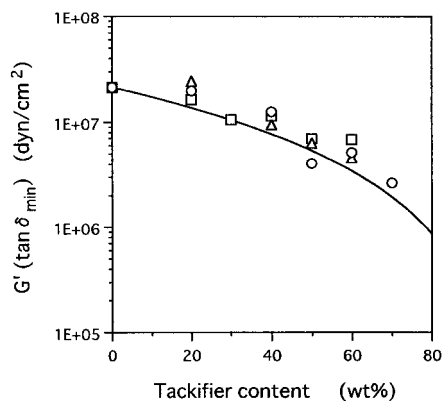


Figure 22 $G'(\tan \delta_{\min})$ of miscible NR-based PSAs. ○, NR/Clearon K-4090 blends; △, NR/Superester A-75 blends; □, NR/ESCOREZ 5320 blends; —, calculated values from eq. (4).

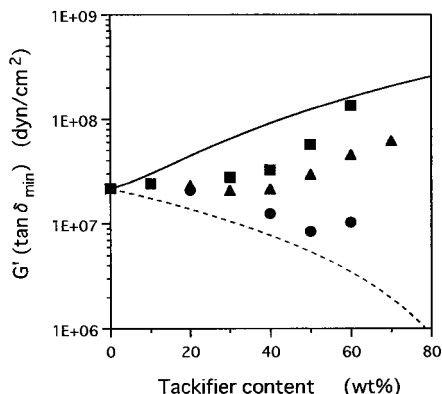


Figure 23 $G'(\tan \delta_{\min})$ of immiscible NR-based PSAs. ■, NR/Kristalex 1120 blends; ▲, NR/Estergum AAG blends; ●, NR/Polypale blends; —, calculated values from eq. (2) as completely immiscible; ---, calculated values from eq. (4) as miscible blends.

tackifier was 0.9–1.1 g/cm³. The experimental values of $G'(\tan \delta_{\min})$ of miscible NR/tackifier blends are plotted in Figure 22, comparing with the value calculated from eq. (4). The comparison of experimental and calculated $G'(\tan \delta_{\min})$ of immiscible NR/tackifier blends are shown in Figure 23. In the case of miscible blends, the experimental modulus agrees well with the calculated one. Thus, good miscibility is reconfirmed. $G'(\tan \delta_{\min})$ of immiscible blends does not agree with either the calculated value from eq. (2) nor that from eq. (4). These blends are not “absolutely immiscible” and of course, not miscible. However, experimental values of NR/Kristalex 1120 blends are the nearest from the calculated values as “absolutely immiscible” blends (eq. (2)), and experimental values of NR/Polypale blends are the nearest from the calculated values as miscible blends (eq. (4)). Therefore, from the viewpoint of viscoelasticity, it can be considered that the extent of phase separation is in this order: NR/Kristalex 1120 > NR/Estergum AAG > NR/Polypale blends. Though this order is different from the order based on microscopic observation, NR/Polypale blends have the highest miscibility of the three immiscible blend systems. Dispersed phase of NR/Estergum AAG blends is very large, but a small amount of NR molecules may be in the phase. We should study the extent of phase separation further.

As for PSAs that cause interfacial failure, the properties of the interface of PSA and adherend might have influence on holding power.

CONCLUSIONS

1. Holding time of miscible NR-based PSAs tended to decrease as the tackifier content increased. It was because plateau modulus of the PSA decreased with increasing tackifier content.
2. Holding time of typical immiscible PSAs was longer than that of NR itself. And holding time of immiscible PSAs should increase theoretically as the tackifier content increases, because of increase in plateau modulus. But in fact, the effect of tackifier content on holding time was different from tackifier to tackifier. This may be attributable to difference in extent of phase separation.

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