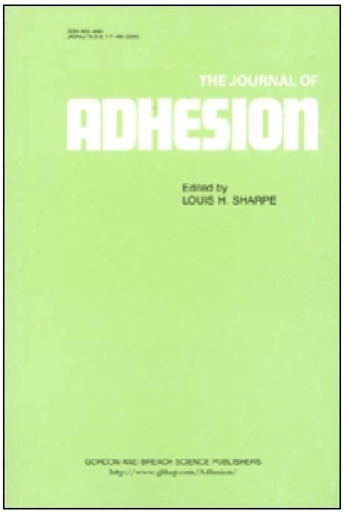

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Tack and Shear Strength of Adhesives Prepared from Styrene-Butadiene Rubber (SBR) Using Gum Rosin and Petro Resin as Tackifiers

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Tack and shear strength of styrene-butadiene rubber (SBR)-based pressure-sensitive adhesive were studied using gum rosin and petro resin as the tackifiers. The concentration of the tackifying resin was varied from 0 to 100 parts per hundred parts of rubber (phr). Toluene was used as the solvent throughout the experiment. The rolling ball technique was used to measure the tack of the adhesive, whereas, shear strength was determined by a TA-HDi Texture Analyser. Results show that the tack of the adhesive increases with increasing tackifier loadings for both tackifier systems. However, shear strength indicates the reverse behavior with increasing resin content, an observation which is attributed to the decrease in cohesive strength as the tackifier concentration is increased. Both tack and shear strength of the adhesives increases with molecular weight of SBR. Adhesive containing petro resin consistently exhibits higher values than the gum rosin system due to better wettability and compatibility in the former system.

Keywords: Adhesive; Rubber; Shear strength; Tack

INTRODUCTION

Several researchers have studied the effect of tackifying resin on the tack behavior of rubber-based adhesives. Kamagata *et al.* [1] correlated the internal structure of a natural rubber-based adhesive with tack. Kraus *et al.* studied the tack and viscoelasticity of a block copolymer [2]. They found that successful pressure-sensitive adhesives result wherever the tackifier is compatible with the rubber phase, which forms the continuum. The effect of molecular weight on the tack and shear properties of natural rubber and epoxidized natural rubber

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based adhesives were investigated by Poh and Yong [3–5]. Kraus and Rollmann [6] also have studied the effect of the entanglement plateau on the adhesive behavior of pressure-sensitive adhesives prepared from styrene-diene (butadiene or isoprene) block copolymers. It was found that compatibility between polymer and tackifier is essential for the development of tack in the adhesive formulations. They also investigated the structural changes in melts of butadiene-styrene and isoprene-styrene block polymer-based adhesives [7]. Recently, we have carried out studies on the adhesion properties of styrene-butadiene rubber (SBR)/standard Malaysian rubber (SMR L)-based adhesives in the presence of phenol formaldehyde and coumarone-indene resins [8,9]. Results show that viscosity of the adhesives increases with resin concentration, whereas, loop tack passes through a maximum value at 20% SBR for all resin loadings. On the other hand, peel strength indicates a maximum value at 40 and 60% SBR for coumarone-indene and phenol formaldehyde resins, respectively. With respect to the effect of gum rosin and petro resin on the rolling ball tack and shear strength of a SBR-based pressure-sensitive adhesive, no study has been reported so far. In view of the absence of data in this field of interest, we have carried out a systematic study to understand the effect of gum rosin and petro resin on the adhesion property of a SBR-based adhesive.

EXPERIMENT

Materials

Buna Hüls 1502 grade SBR was supplied by Bayer Company (Penang, Malaysia). It has a 33.5% by weight target of bound styrene. The density, ash content, Mooney viscosity, and volatile matter of the rubber are 0.9 g/cc, 0.5%, 50, and 0.2%, respectively. Gum rosin (ww grade) and petro resin (Nisseki 120) were supplied by EuroChemo-Pharma Company (Penang, Malaysia). The respective softening temperatures are 76 and 100°C. Gum rosin is a mixture of rosin acids which consists of abietic acid and pimaric acid. Petro resin is the product of polymerization of a C-5 petroleum fraction, mainly *cis*- and *trans*-piperylene and some amount of isoprene. Laboratory grade toluene was used as the solvent throughout the experiments.

Adhesive Preparation

SBR was masticated on a 2-roll mill for 10 min. For each adhesive formulation, 5 g of rubber and 20 mL of toluene was used. The rubber was dissolved in toluene and the rubber solution was left in a conditioned room for 24 h before adding gum rosin or petro resin. Five different weights, *i.e.*, 1, 2, 3, 4, and 5 g corresponding to 20, 40, 60, 80, and 100 parts per hundred parts of rubber (phr) of tackifying resin were then mixed with the rubber solution to prepare the adhesives. One control sample—without tackifier—was also prepared for comparison purposes. Constant stirring with a glass rod was carried out to ensure the formation of a homogeneous adhesive. The resulting adhesive was left for 2 h before testing.

Molecular Weight Determination

Different molecular weights of SBR were obtained by masticating the rubber on a two-roll mill for 5, 10, 15, and 20 min. The molecular weights of masticated and unmasticated rubber were measured using a viscometric method. Five different concentrations (C) of dilute rubber solutions were prepared in toluene for each rubber sample. An Ubbelohde viscometer was used to determine the flow time of toluene (t_o) and each rubber solution (t). The specific viscosity of the rubber solution (η_{sp}) is given in Eq. (1) since the densities of solvent and dilute rubber solution are similar:

$$\eta_{sp} \simeq (t - t_o)/t_o. \tag{1}$$

From the plot of reduced viscosity (η_{sp}/C) *versus* C, the intrinsic viscosity, $[\eta]$, was measured from the intercept at $C = 0$ by extrapolation. The viscosity-average molecular weight (M_v) of each rubber sample was then computed using the Mark-Houwink-Sakurada Eq. (2) below [10,11]:

$$[\eta] = kM_v^a, \tag{2}$$

where $k = 3.79 \times 10^{-2}$ mL/g and $a = 0.71$ in toluene.

Table 1 shows the intrinsic viscosity and viscosity-average molecular weight of SBR used in this study.

TABLE 1 Intrinsic Viscosity and Molecular Weight of SBR

Mastication time (minutes)	Intrinsic viscosity (mL/g)	Viscosity-average molecular weight
0	118	8.32×10^4
5	98	6.41×10^4
10	86	5.34×10^4
15	76	4.48×10^4
20	58	3.06×10^4

Testing

Rolling Ball Tack

Rolling ball tack was determined using a Rolling Ball Tack Tester Model TT-100 (Cheminstruments, Fairfield, CT, USA) according to ASTM D 3121-94. The inclined trough and steel ball (11.1-mm diameter) were cleaned with acetone before and after testing. The adhesive was coated on a polyethylene terephthalate (PET) film of 0.07-mm thickness; (38 cm long × 5 cm wide) at a dry coating thickness of 30, 60, 90, and 120 μm by a SHEEN hand coater (Teddington, Middlesex, UK). It was then placed horizontally—with adhesive side up—in line with the inclined trough at the base of the inclined trough. The end of the specimen opposite the incline was held to the table with a tape. Then, a steel ball was released at the top of the incline using dry tongs and allowed to accelerate down the incline and roll on to a horizontal PET film surface covered with the adhesive. The distance between the end of the incline and the point where the ball stopped was measured. The distance is inversely proportional to the tackiness of the adhesive. Three replicates were tested for each adhesive formulation and the average reading was taken as the tack value of the adhesive. The percentage error was estimated to be 5%.

Shear Strength

A shear test was carried out in order to determine the cohesive strength of the SBR-based adhesive. The dimensions of the release paper substrate (base stock) for the shear test were 2.5 cm (width) × 15 cm (length). A SHEEN Hand Coater was used to coat the adhesive with a dry coating area of 2.5 × 5 cm from the end of release paper. Another release paper—with the same dimensions as that of the base stock—was used as the face stock. The end of the face stock with dimensions 2.5 × 5 cm was gently laid on the coated area of the base stock. For comparison purposes, two coating thicknesses, *i.e.*, 60 and 120 μm, were used. The coated sample was then conditioned at room temperature for 24 h before testing on a TA-HDi Texture Analyzer (Model-Stable Micro System) (Surrey, UK) operating at a testing speed of 1 mm/s up to 50 sec. The testing distance was 5 cm which corresponded to the length of coated area. The shear force was determined from the peak force from a plot of force *versus* time. Shear strength was expressed as the shear force per unit area of testing.

RESULTS AND DISCUSSION

The effects of gum rosin and petro resin loading on the tack and shear strength of SBR-based pressure-sensitive adhesives are discussed below.

Rolling Ball Tack

Tack is the ability of two substrates to hold against separation after contact for a short duration of time. For rolling ball tack testing, two major retarding forces are applied by the adhesive to the ball, *i.e.*, the adhesion between the adhesive and the ball which is called "grab" and the energy required to push the adhesive out of the ball's path, the "plowing effect." Figure 1 shows that the distance travelled by the rolling ball decreases with increasing gum rosin loading for all coating thicknesses. The distance travelled is inversely proportional to the tackiness of the adhesive. In other words, tack increases with increasing gum rosin concentration. This observation is attributed to the increase in grab and plowing effect. A similar result is also obtained when petro resin is used as the tackifier as shown in Fig. 2. The rolling distance decreases with increasing petro resin concentration for all coating thicknesses investigated in this study. The continuous increase of tack with tackifier content indicates that maximum tack has not been reached, *i.e.*, phase inversion does not occur.

Figure 3 shows the effect of coating thickness on the rolling distance at 60 phr of resin loading. The plot indicates that the rolling distance decreases with increasing thickness for both tackifying resins. This observation is attributed to the higher amount of adhesive as the coating thickness is increased. This rolling ball tack method quantifies the

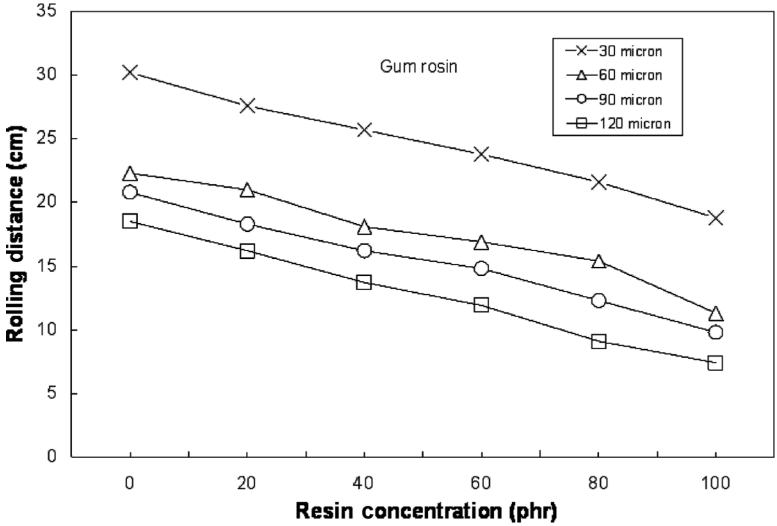

FIGURE 1 Variation of distance travelled with gum rosin concentration for various coating thicknesses.

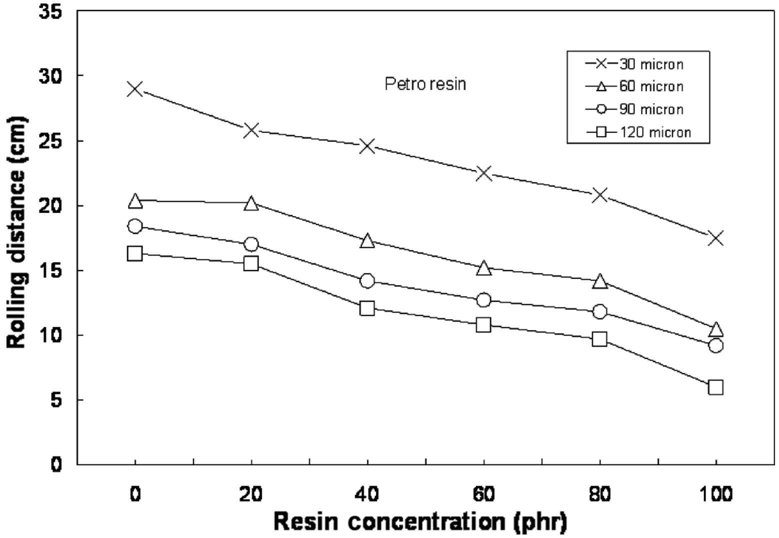

FIGURE 2 Variation of distance travelled with petro resin concentration for various coating thicknesses.

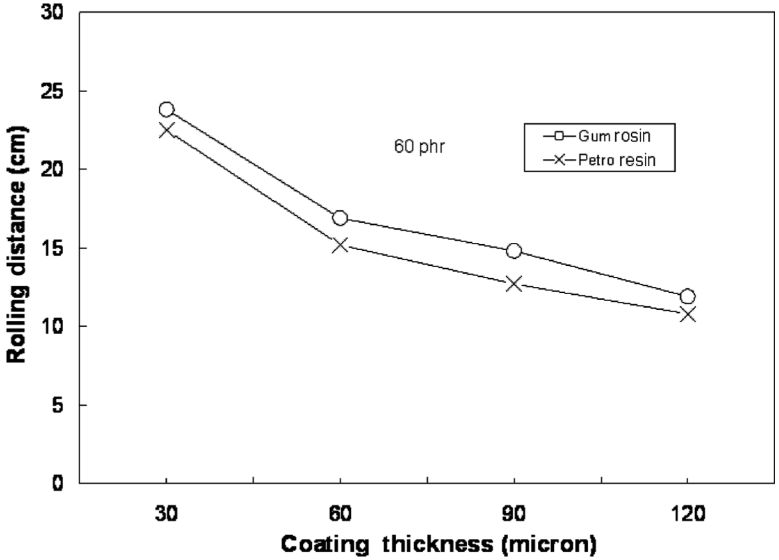

FIGURE 3 Variation of distance travelled with coating thickness at 60 phr of tackifier concentration.

ability of an adhesive to adhere quickly to another surface. The increase in peel force with increasing coating thickness has also been reported by previous researchers [12–15]. For a fixed coating thickness, adhesive containing petro resin—aliphatic hydrocarbon resin—consistently exhibits lower rolling distance compared with that of the gum rosin system. This means that the petro resin-based adhesive has a higher tack compared with that of gum rosin, a phenomenon which is ascribed to better wettability and compatibility between petro resin and SBR, both of which are non-polar in nature. On the other hand, gum rosin is a naturally occurring material obtained as oleoresin from living trees. It consists of abietic acid and pimaric acid which have cyclic structures. Lower wettability exists between gum rosin and SBR as reflected in the lower tack as shown in Fig. 3. The respective DSC thermographs indicating compatibility are shown in Figs. 4 and 5 for petro resin and gum rosin systems. Figure 4 shows the occurrence of compatibility between SBR and petro resin tackifier as indicated by a single glass transition at about 80°C. On the other hand, a single glass transition at about 60°C is exhibited by the SBR-gum rosin system as illustrated in Fig. 5.

Figure 6 shows the dependence of rolling distance on molecular weight of SBR. For both coating thicknesses, the rolling distance decreases with increasing molecular weight of rubber, *i.e.*, tack increases with increasing molecular weight of the rubber. This observation is attributed to the increasing wettability and compatibility of adhesive on the substrate. Our previous investigation on

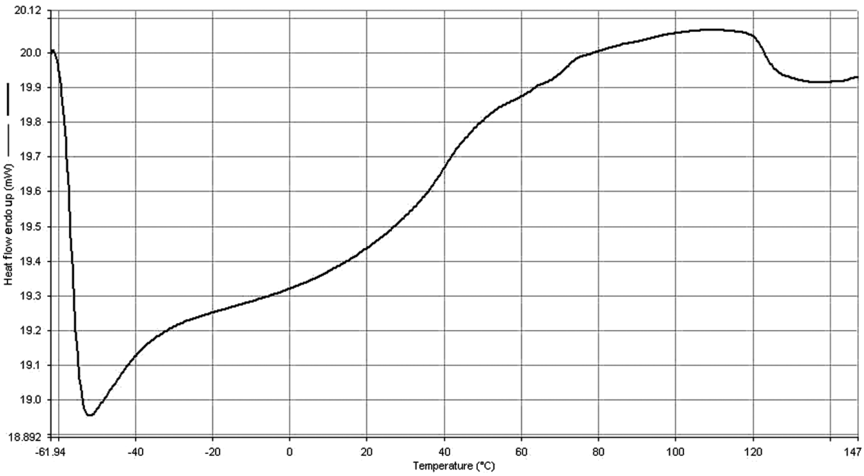

FIGURE 4 A DSC thermograph of adhesive containing 60 phr of petro resin.

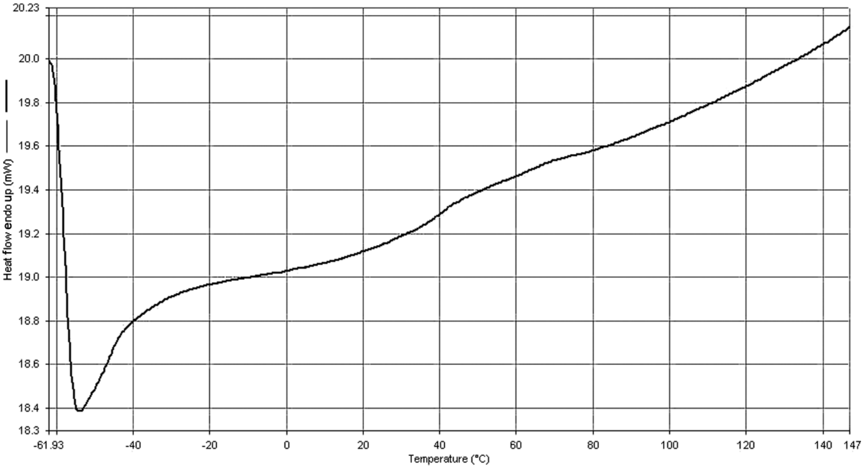

FIGURE 5 A DSC thermograph of adhesive containing 60 phr of gum rosin.

the adhesion property of natural rubber (NR) and epoxidized natural rubber (ENR)—both are crystallizable rubbers—shows a maximum value at an optimum molecular weight of rubber [16–19]. However, in this study, no optimum molecular weight of SBR on rolling ball tack is obtained. This observation may be attributed to the noncrystallizability of SBR [20], *i.e.*, strain-induced crystallization does not occur

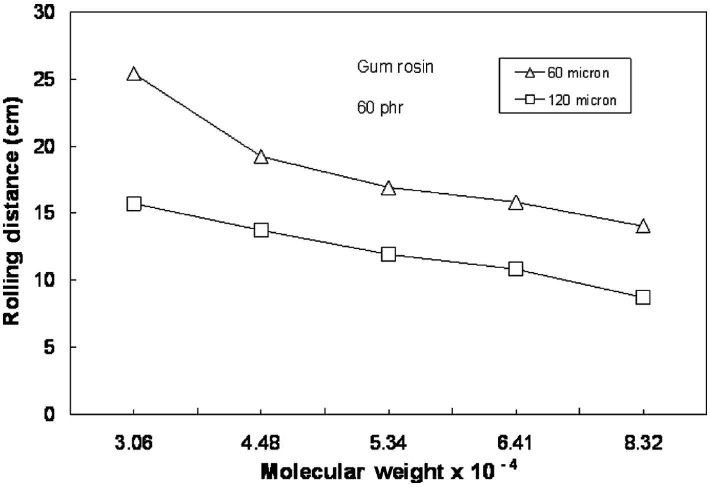

FIGURE 6 Dependence of distance travelled on molecular weight of SBR for adhesive containing 60 phr gum rosin.

in the SBR-based adhesive. For a fixed molecular weight of SBR, the 120-μm thick coated sample consistently indicates a higher tack value than the 60-μm coating thickness. This finding is ascribed to the higher amount of adhesive present which allows sufficient viscoelastic response to form adequate tack properties at the higher coating thickness [21].

Shear Strength

Figure 7 shows the dependence of shear strength on the gum rosin concentration at 60- and 120-μm coating thicknesses. For both coating thicknesses, shear strength decreases gradually with increase in resin loading. This observation is attributed to the decrease in the cohesive strength of the adhesive due to the decreasing SBR content which acts as the binder in the adhesive system. As the resin loading is increased, the dilution effect of resin becomes increasingly significant as reflected by the steady drop in the shear strengh of the adhesive. This finding is consistent with our previous study on adhesives prepared from natural rubber and epoxidized natural rubber [22–24]. A similar result is also obtained from the petro resin-based SBR adhesive as shown in Fig. 8. Again, shear strength decreases with increase in petro resin

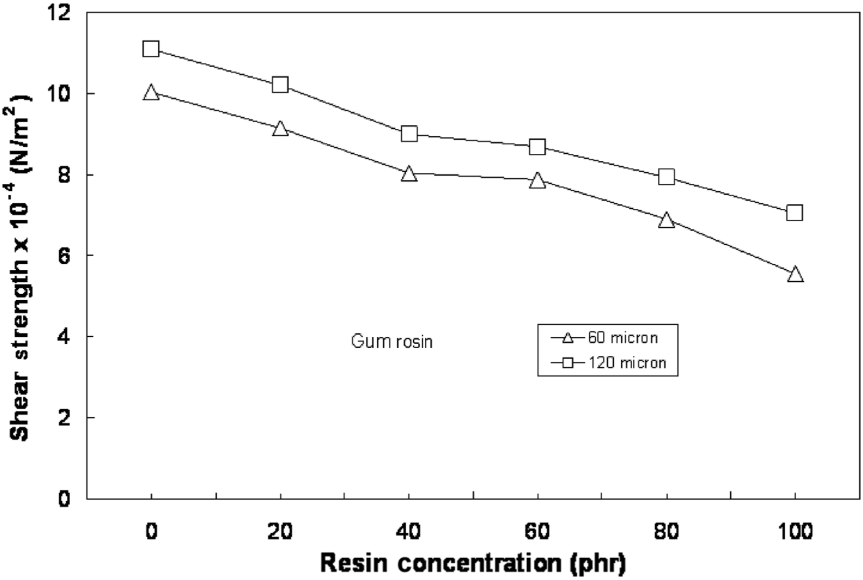

FIGURE 7 Variation of shear strength with gum rosin concentration at 60- and 120-μm coating thicknesses.

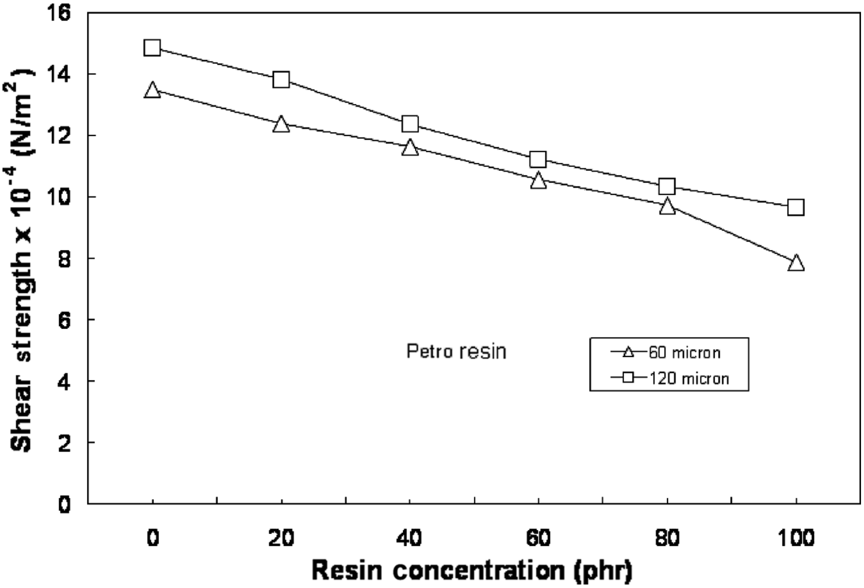

FIGURE 8 Variation of shear strength with petro resin concentration at 60- and 120-μm coating thicknesses.

loading for both coating thicknesses investigated. The dilution effect of the petro resin causes the decrease in cohesive strength of the adhesive and, hence, lower shear strength is observed. Figures 7 and 8 also indicate that the shear strength of the 120-μm coating is much higher than that of 60-μm coating. As coating thickness is increased, the amount of adhesive present in the coating layer is increased.

The average rate of decrease of shear strength with resin loading is shown in Table 2. For a fixed coating thickness, the rate of decrease for the gum rosin-based adhesive is lower than that for the petro resin. This observation is attributed to the difference in softening point of the tackifier where petro resin shows a higher softening point

TABLE 2 Average Rate of Decrease of Shear Strength

Coating thickness (μm)	Rate of decrease (N/m^2 per phr gum rosin)	Rate of decrease (N/m^2 per phr petro resin)
60	449	561
120	405	519

(100°C) than that of gum rosin (76°C). Besides, Table 2 also suggests that the dilution effect of gum rosin is less compared with that of petro resin. Most probably, petro resin being a polymerized material exerts a more pronounced plasticizing effect due to better compatibility in the polymer-resin system, hence, causing a greater drop in cohesive strength of the adhesive as resin loading is increased. For both tackifying systems, 60-µm coated samples consistently exhibit a higher rate of decrease of shear strength compared with the 120-µm coated substrate. This result indicates that the dilution effect is more significant in thinner coating. This may be attributed to the smaller amount of adhesive present in the thinner coated sample.

Figure 9 compares the shear strength between gum rosin and petro resin systems at 60 phr resin concentration for both coating thicknesses. It is obvious that the shear strength of petro resin-based adhesive is higher than that of the gum rosin system. This observation is attributed to the higher softening temperature of petro resin compared with that of gum rosin.

Figure 10 shows the effect of molecular weight of SBR on the shear strength of adhesives. The plot indicates that shear strength increases with molecular weight of rubber for both coating thickness, an observation which is attributed to the increase in cohesive strength as the

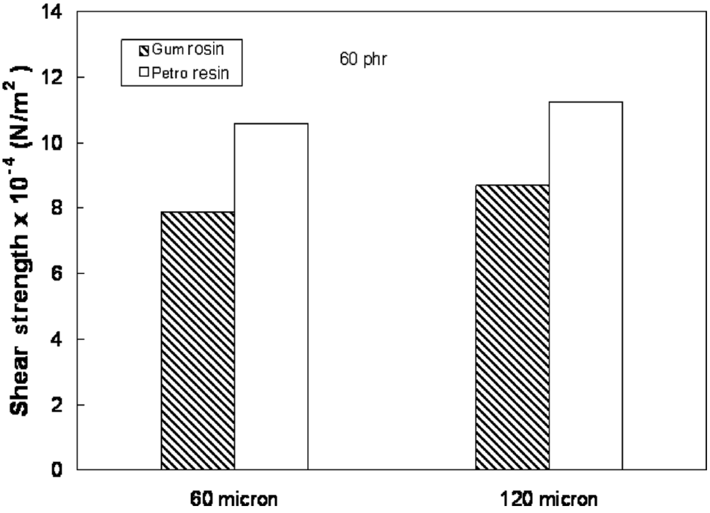

FIGURE 9 Comparison of shear strength between adhesives prepared from gum rosin and petro resin at 60 phr resin concentration for 60- and 120-µm coating thicknesses.

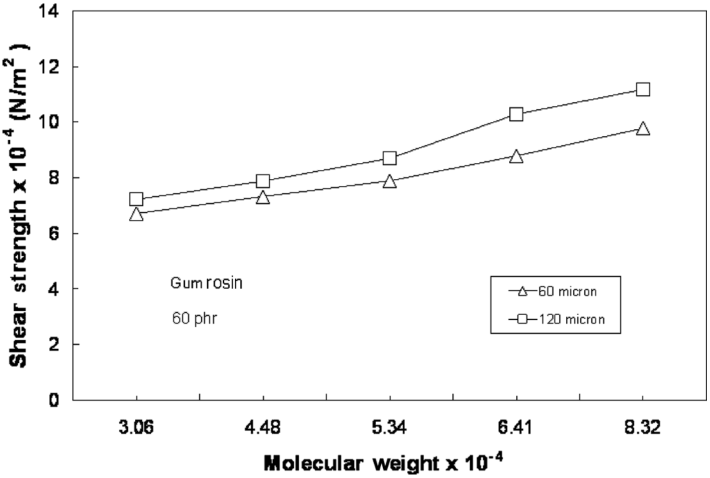

FIGURE 10 Dependence of shear strength on molecular weight of SBR for adhesive containing 60 phr gum rosin.

molecular weight of the rubber is increased. As in the case of tack, no maximum value of shear strength is observed. This phenomenon may be ascribed to the noncrystallizability of SBR [18]. At lower molecular weight, cohesive failure [25] due to shorter chain length of the rubber molecules occurs. However, as molecular weight of the rubber is increased, cohesive strength of the adhesive also increases correspondingly as reflected by the higher shear strength observed at higher molecular weight of SBR.

CONCLUSIONS

The following conclusions can be drawn from this study.

1. Rolling ball tack increases with increasing gum rosin and petro resin loading, an observation which is attributed to the increase in the adhesion between the adhesive and the ball, and the energy required to push the adhesive out of the ball's path. The tack also increases with coating thickness and molecular weight of rubber. The petro resin-based adhesive exhibits a higher tack compared with the gum rosin adhesive system resulting from better wettability and compatibility between petro resin and SBR.
2. Shear strength of adhesive decreases with gum rosin and petro resin loading. This observation is associated with the decrease in the cohesive strength of the adhesive resulting from the dilution

effect of resin, *i.e.*, decreasing amount of SBR content which acts as the binder in the adhesive system. It increases with coating thickness and molecular weight of rubber. Shear strength of petro resin-based adhesive is higher than that of the gum rosin system due to better wettability and compatibility in the former system.

REFERENCES

[1] Kamagata, K., Kosaka, H., Hina, K., and Toyama, M., *J. Appl. Polym. Sci.* **15**, 483–500 (1971).
[2] Kraus, G., Rollmann, K. W., and Gray, R. A., *J. Adhesion* **10**, 221–236 (1979).
[3] Poh, B. T. and Yong, A. T., *J. Macromol. Sci. Part A* **46**, 97–103 (2009).
[4] Poh, B. T. and Yong, A. T., *J. Appl. Polym. Sci.* **114**, 3976–3979 (2009).
[5] Poh, B. T. and Yong, A. T., *J. Appl. Polym. Sci.* **115**, 1120–1124 (2010).
[6] Kraus, G. and Rollmann, K. W., *J. Appl. Polym. Sci.* **21**, 3311–3318 (1977).
[7] Kraus, G. and Hashimoto, T., *J. Appl. Polym. Sci.* **27**, 1745–1757 (1982).
[8] Poh, B. T. and Ong, L. N., *eXPRESS Polym. Lett.* **1**, 654–659 (2007).
[9] Poh, B. T. and Heng, S. S., *Polym-Plast. Technol. Eng.* **47**, 325–329 (2008).
[10] Billmeyer, F. W., Jr., *Textbook of Polymer Science*, 3rd ed., (Wiley, New York, 1984), Ch. 8, p. 208.
[11] Bahadur, P. and Sastry, N. V., *Principles of Polymer Science*, 2nd ed., (Alpha Science International, Oxford, 2005), Ch. 3, p. 124.
[12] Aubrey, D. W., Welding, G. N., and Wong, T., *J. Appl. Polym. Sci.* **13**, 2193–2207 (1969).
[13] Gardon, J. L., *J. Appl. Polym. Sci.* **7**, 625–641 (1963).
[14] Johnston, J., *J. Adhesives Age.* **11**, 20–26 (1968).
[15] Chan, H. K. and Howard, G. J., *J. Adhesion* **9**, 279–304 (1978).
[16] Poh, B. T. and Yong, A. T., *J. Adhesion* **85**, 435–446 (2009).
[17] Poh, B. T. and Yong, A. T., *J. Appl. Polym. Sci.* **114**, 3976–3979 (2009).
[18] Poh, B. T. and Yong, A. T., *J. Macromol. Sci. Part A* **46**, 97–103 (2009).
[19] Poh, B. T. and Yong, A. T., *J. Appl. Polym. Sci.* **115**, 1120–1124 (2010).
[20] Mark, J. E., Erman, B., and Eirich, F. R. (Eds.), *Science and Technology of Rubber*, (Academic Press, New York, 1994), Ch. 2, p. 43.
[21] Poh, B. T. and Issam, A. M., *Plast Rubber Compos.* **38**, 453–456 (2009).
[22] Poh, B. T. and Kwo, H. K., *Polym-Plast. Technol. Eng.* **46**, 1021–1024 (2007).
[23] Poh, B. T., Yee, K. W., and Lim, H. B., *J. Appl. Polym. Sci.* **110**, 4079–4083 (2008).
[24] Poh, B. T. and Kwo, H. K., *J. Appl. Polym. Sci.* **105**, 680–684 (2007).
[25] Satas, D. (Ed.), *Handbook of Pressure-Sensitive Adhesive Technology*, (Van Nostrand Reinhold, New York, 1982), Ch. 4, p. 54.