Adhesion Properties of Pressure-Sensitive Adhesives Prepared from SMR 10/ENR 25, SMR 10/ENR 50, and ENR 25/ENR 50 Blends

B. T. Poh, A. L. Lim

School of Industrial Technology, Universiti Sains Malaysia, 11800 Penang, Malaysia

Received 9 May 2007; accepted 10 October 2007
DOI 10.1002/app.27489
Published online 20 March 2008 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The adhesion properties, i.e. viscosity, tack, and peel strength of pressure-sensitive adhesives prepared from natural rubber/epoxidized natural rubber blends were investigated using coumarone-indene resin and toluene as the tackifier and solvent respectively. One grade of natural rubber (SMR 10) and two grades of epoxidized natural rubbers (ENR 25 and ENR 50) were used to prepare the rubber blends with blend ratio ranging from 0 to 100%. Coumarone-indene resin content was fixed at 40 parts per hundred parts of rubber (phr) in the adhesive formulation. The viscosity of adhesive was measured by a HAAKE Rotary Viscometer whereas loop tack and peel strength was determined using a Lloyd Adhesion Tester operating at 30 cm/min. Results show that the viscosity of the adhesive passes through a minimum value at 20% blend ratio. For loop tack and peel strength, it indicates a maximum at 60% blend ratio for SMR 10/ENR 25 and SMR 10/ENR 50 systems. However, for ENR 25/ENR 50 blend, maximum value is observed at 80% blend ratio. SMR 10/ENR 25 blend consistently exhibits the best adhesion property in this study, an observation which is attributed to the optimum compatibility between rubbers and wettability of adhesive on the substrate. J Appl Polym Sci 109: 115–119, 2008

Key words: adhesive; viscosity; tack; peel strength; rubber

INTRODUCTION

Rubber-based pressure-sensitive adhesives (PSA) have been studied by several researchers.[1–3] However, systematic study of the adhesion property of natural rubber seems scarce. Recently, we have reported the viscosity, tack, peel, and shear strength of PSA prepared from natural rubber (SMR L, SMR 10, and SMR 20 grades).[4–6] From the study, it is observed that viscosity, tack, and peel strength of the adhesives shows an increasing trend with coumarone-indene resin tackifier concentration. The shear strength, on the contrary, indicates a downward behavior with increase in the resin loading. With respect to epoxidized natural rubber (ENR)—a chemically modified natural rubber—results show that peel strength passes through a maximum value at 40 phr of coumarone-indene resin,[7] an observation which is associated to the maximum wettability of adhesive on the substrate. On the other hand, the shear strength of ENR-based PSA exhibits similar behavior as the unmodified natural rubber, i.e. it decreases gradually with increasing tackifier loading due to the decrease in cohesive strength of adhesive. Generally, peel and shear strength increases with coating thickness. However, the effect of blend ratio of natural rubber/ENR on the adhesion property of PSA is so far not studied. Owing to the scarcity of research in this field of interest, we have carried out a systematic investigation of the effect of blend ratio on the viscosity, tack, and peel strength of the rubber blends.

Correspondence to: B. T. Poh (btpoh@usm.my).
Contract grant sponsor: Universiti Sains Malaysia.

EXPERIMENT

Materials

Standard Malaysian rubber (SMR 10 grade) and ENR 25 and ENR 50 grades having 25 and 50 mol % of epoxidation, respectively were used as the elastomers in this study. The respective technical specifications[8,9] of the rubbers are shown in Table I. The rubbers were supplied by Rubber Research Institute of Malaysia (RRIM). The rubbers were masticated using a two-roll mill for 10 min to facilitate easy dissolution of the rubbers in toluene. The viscosity-average molecular weights of SMR 10, ENR 25, and ENR 50 were 3×10^5, 2×10^5, and 1.5×10^5, respectively.

Coumarone-indene resin supplied by EuroChemo-Pharma Company (Malaysia) was chosen as the tackifier. Commercial grade toluene was used as the

TABLE I
Technical Specification of SMR 10 and ENR

	SMR 10	ENR 25	ENR 50
Glass transition temperature (°C)	−72	−45	−20
Specific gravity	0.92	0.97	1.03
Mooney viscosity, $M_{L,\ 1+4}$ (100°C)	78	110	140

solvent to prepare the PSA. All the chemicals were used as supplied and no purification was carried out in this experiment.

Adhesive preparation

Three rubber blends, i.e. SMR 10/ENR 25, SMR 10/ENR 50, and ENR 25/ENR 50 were used with blend ratios ranging from 0 to 100%. The total weight of the rubber blend was 5 g. The rubber was then dissolved in 20 mL of toluene and the resulting rubber solution was tightly closed. It was conditioned at room temperature (30°C) for 24 h prior to the addition of 2 g of pulverized coumarone-indene resin which corresponded to 40 parts per hundred parts of rubber (phr). The PSA thus prepared was constantly stirred before testing.

Testing

Viscosity

Viscosity of the adhesive was determined by a HAAKE Rotary Viscometer (Model PK 100) with spindle head (PK1;1°). The platform of the viscometer were cleaned with acetone and then raised up to touch the spindle head. The gap between spindle head and platform was adjusted to zero. A few drops of adhesive were placed at the middle of platform which was then raised to squeeze the adhesive. Acetone was used to wipe-off excessive adhesive around the spindle head. Testing of viscosity was ended after 1 min or 10 rounds of spinning. The average viscosity of adhesive was calculated from at least five readings recorded.

Tack

Loop tack test was used to determine the tack property of the PSA. A polyethylene terephthalate (PET) film with dimension of 4 cm × 25 cm was coated at the centre (4 cm × 4 cm) using a SHEEN Hand Coater at a coating thickness of 60 μm. The film was then formed into a loop and the adhesive area was gently brought into contact with a glass panel. The debonding force of the adhesive from the glass panel was measured by a Lloyd Adhesion Tester (Model LRXPlus with NEXYGEN software) operating at a testing rate of 30 cm/min. The three highest peaks detected were used to compute the average debonding force. The loop tack value was expressed as the average debonding force per area of contact (N/m^2).

Peel strength

The substrates used for peel tests were PET film (base stock) and release paper (face stock). Three testing modes of peel tests, i.e. T-peel, 90-° and 180°-peel tests were employed in this study. The dimensions of the substrates were 20 cm × 4 cm for the T- and 90°-peel tests. However, for the 180°-peel test, the dimensions of PET film and paper substrates were 25 cm × 4 cm and 12 cm × 6 cm, respectively. A SHEEN Hand Coater was used to coat the PET film at 60 μm coating thickness and at a coating area of 10 cm × 4 cm from the end of the film. The face stock (release paper) was then placed on the coated PET film to form the testing sample which was conditioned at room temperature for 24 h. A Lloyd adhesion tester operating at 30 cm/min was used to measure the peeling force of the adhesive. The average peeling force was determined from the three highest peaks of a load-propagation graph. Peel strength is defined as the average load per width of the bondline required to separate progressively a flexible member from a rigid member or another flexible member (ASTM D 907).

RESULTS AND DISCUSSION

The results of this study is discussed with respect to the effect of blend ratio of rubbers on the viscosity, tack, and peel strength of the adhesives prepared from various rubber blends.

Viscosity of adhesive

The dependence of viscosity of adhesive on blend ratio for SMR 10/ENR 25-, SMR 10/ENR 50-, and ENR 25/ENR 50-based PSA is shown in Figure 1. From the plot, it indicates that viscosity of adhesives prepared from the blended rubbers decreases with blend ratio until a minimum value is obtained at 20% blend ratio for all the systems studied. This lowering of viscosity is attributed to the initial ''plasticizing'' effect of the first rubber component. The introduction of less than 20% of the new rubber into the blend may disrupt the morphology of the existing rubber. This means that additional ''free volume'' is created which enhances the flow behavior, i.e. lower viscosity of blend is observed initially. However, after 20% blend ratio, the ''plasticizing'' effect decreases gradually with increasing content of

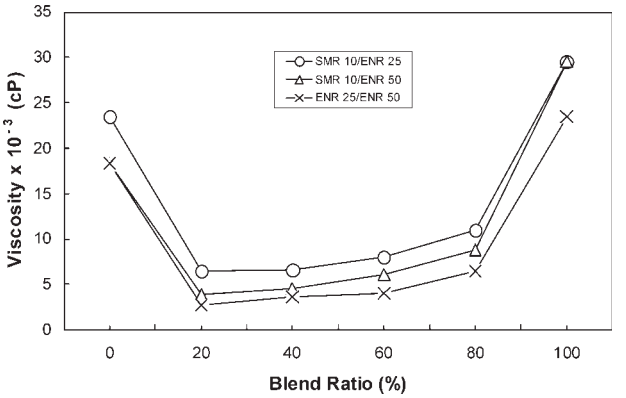

Figure 1 Variation of viscosity with blend ratios of rubbers.

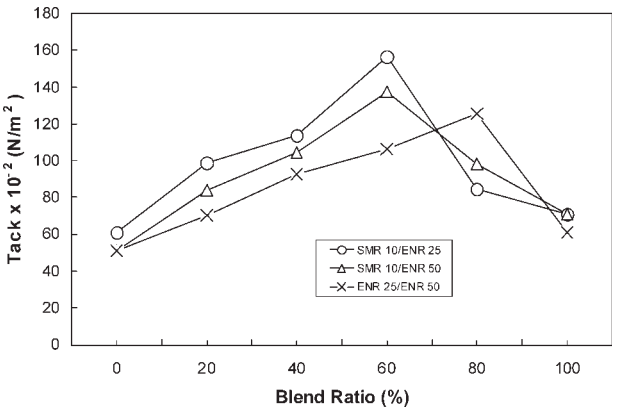

Figure 2 Variation of loop tack with blend ratio of rubbers.

the first rubber component as reflected by the gentle increase of viscosity from 20 to 80% blend ratio. A marked increase in viscosity is observed after 80% blend ratio suggesting that the viscosity is primarily dominated by the first rubber component. Figure 1 also shows that for single component rubber, the viscosity of SMR 10-based adhesive is higher than that of ENR 25 and followed by ENR 50. This observation is ascribed to the higher molecular weight of SMR 10, followed by ENR 25 and ENR 50. As shown by our previous study,[4] viscosity of natural rubber-based adhesive increases with molecular weight of the rubber.

Loop tack

Figure 2 shows the dependence of loop tack on the blend ratio for the three rubber blend systems. For the SMR 10/ENR 25 and SMR 10/ENR 50 systems, tack increases with blend ratio up to 60% SMR 10 component and drops with further increment of SMR 10. This observation is attributed to the increasing compatibility and wettability of blending system where maximum compatibility and wettability is achieved at 60% blend ratio, after which compatibility between the rubbers decreases with further addition of SMR 10. At 60% SMR 10, the adhesive conforms to the irregularities of the substrate, i.e. low surface energy condition is observed[10] to give the maximum tack as shown in Figure 2. At this composition, the rubber blend and resin components achieves optimum elastic and viscous property that is necessary for the maximum tack in a PSA. Further increase of blend ratio will lower the compatibility between the rubbers as the viscosity of SMR 10-dominated adhesives increases as discussed earlier. However, for the ENR 25/ENR 50 system, the peak value occurs at 80% blend ratio suggesting that the blend system is more compatible than SMR 10/ENR systems. Also, the peak tack value for the ENR 25/ENR 50 system is lower than that of SMR 10/ENR 25 and SMR 10/ENR 50 systems. In fact, the highest peak value is exhibited by SMR 10/ENR 25 system followed by SMR 10/ENR 50 and ENR 25/ENR 50 systems. For the SMR 10/ENR 25 blend, the maximum tack value is 1.6×10^4 N/m^2 which is slightly lower than the commercially acceptable adhesive tack value of 2×10^4 N/m^2. Peak value increases with decreasing T_g of rubber. SMR 10 which has the lowest T_g—as shown in Table I exhibits the highest peak value followed by ENR 25 and ENR 50. The higher the T_g, the less flexible is the rubber which affects the rheological property of the adhesive and hence lower the wettability of the adhesive on the substrate.

Peel strength

Figure 3 shows the peel strength of the adhesive using paper/PET film as the substrates for the T-peel test experiment. Maximum value is observed at 60% blend ratio for the SMR 10/ENR 25 and SMR 10/ENR 50 systems whereas for the ENR 25/ENR 50 system, peak value occurs at 80% blend ratio. The

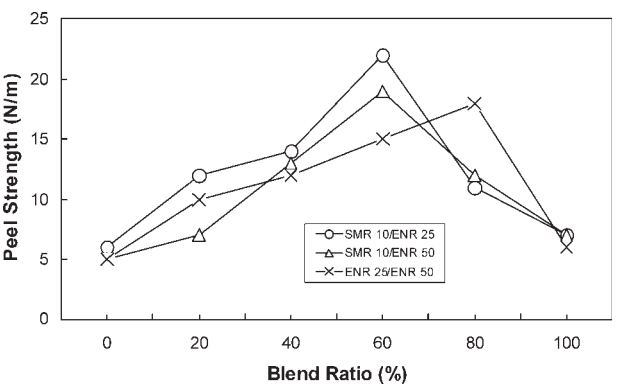

Figure 3 Peel strength versus blend ratio of rubbers for T-peel test.

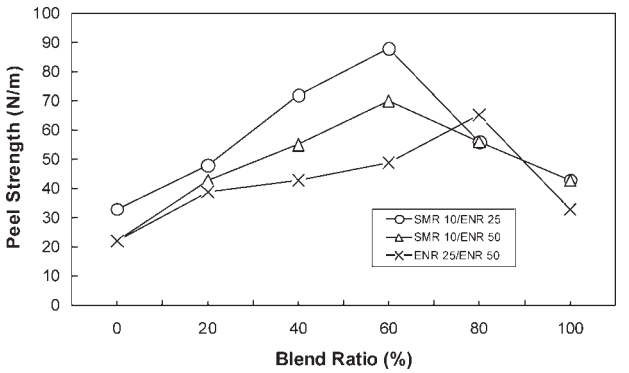

Figure 4 Peel strength versus blend ratio of rubbers for 90°-peel test.

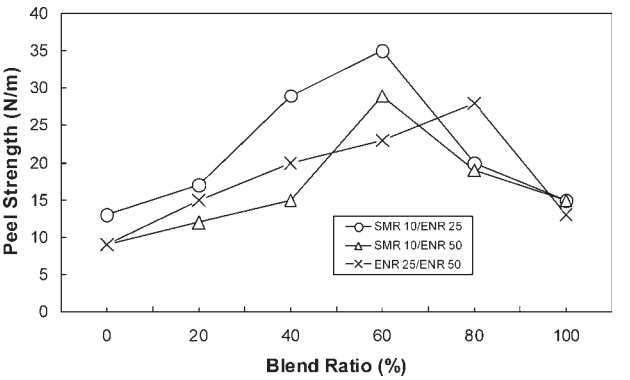

Figure 5 Peel strength versus blend ratio of rubbers for 180°-peel test.

increase of peel strength up to the maximum value is attributed to the increasing wettability of the adhesive which enhances the peel adhesion property of the adhesive. Further increase in the blend ratio will decrease the compatibility of rubbers as reflected by the lower peel strength as shown in Figure 3. As in the case of tack, the absolute value of peak peel strength decreases from SMR 10/ENR 25, followed by SMR 10/ENR 50 and ENR 25/ENR 50 blends. Again, this observation is associated with the greatest flexibility of SMR 10, followed by ENR 25 and ENR 50 due to the effect of T_g as discussed earlier. The low T_g value of SMR 10 means greater flexibility of the rubber which enhances the rheological property of the adhesive. This is manifested by the improved wettability in the SMR 10/ENR 25 system as indicated by the highest peak value in the peel adhesion study. Similar observation is also obtained for the 90° and 180° peel tests as shown in Figures 4 and 5 respectively, thus confirming the dependence of peel strength on blend ratio for the three rubber blend systems investigated in this study. Figure 6 compares the peak values of peel strength between the three blend systems for each mode of peel test. It is obvious that for each peel test, SMR 10/ENR 25 blend exhibits the highest peel value, followed by SMR 10/ENR 25 and ENR 25/ENR 50. Figure 6 also shows that for the three blending systems, 90° peel test consistently gives the highest peel strength, followed by 180° peel test and T-peel test. This observation may be ascribed to the angle of testing which suggests that 90° test requires higher peeling force to separate the mechanical interlocking and anchorage of the adhesive in pores and irregularities in the substrate.[11,12] The maximum peel strength obtained from the SMR 10/ENR 25 system using 90° peel test is 88 N/m compared with 200 N/m from the commercially acceptable adhesive. This difference between the two peel values is attributed to the testing substrates. Commercial adhesive is tested using plastic films—base stock and face stock—as the substrates whereas PET film/release paper are used as substrates in our study.

CONCLUSIONS

The following conclusions can be drawn from this study.

1. Viscosity of adhesives prepared from the blended rubbers decreases with blend ratio until a minimum value is reached at 20% blend ratio for all the systems studied, an observation which is attributed to the initial ''plasticizing'' effect of the first rubber component. The viscosity of SMR 10-based adhesive is higher than that of ENR due to the higher molecular weight of the former.
2. For the SMR 10/ENR 25 and SMR 10/ENR 50 systems, loop tack passes through a maximum at 60% blend ratio after which it decreases with further increase in SMR 10 component. The observation is attributed to the varying degree of wettability and compatibility with blend ratio. However, for the ENR 25/ENR 50

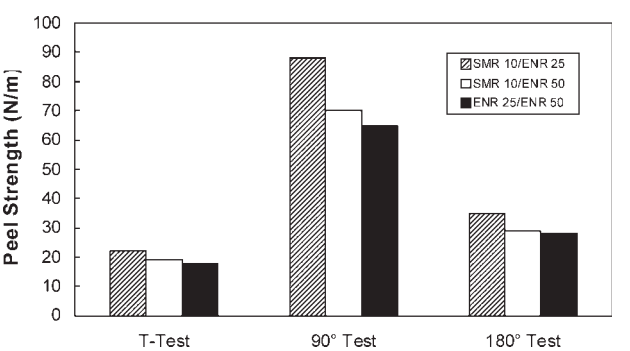

Figure 6 Comparison of peak values of peel strength between various blend systems at three modes of peel tests.

system, the peak value occurs at 80% blend ratio.

3. Maximum value of peel strength is also observed at 60% blend ratio for the SMR 10/ENR 25 and SMR 10/ENR 50 systems whereas for the ENR 25/ENR 50 system, the peak value is obtained at 80% blend ratio. Increasing wettability of the adhesive and compatibility between rubbers with blend ratio enhances the peel adhesion property of the adhesive up to a certain value. The 90° peel test consistently shows the highest peel strength, followed by 180° peel test and T-peel test, an observation indicating that 90° test requires higher peeling force than the others.

References

1. Kraus, G.; Rollmann, K. W. J Appl Polym Sci 1977, 21, 3311.
2. Kraus, G.; Hashimoto, T. J Appl Polym Sci 1982, 27, 1745.
3. Leong, Y. C.; Lee, L. M. S.; Gan, S. N. J Appl Polym Sci 2003, 88, 2118.
4. Poh, B. T.; Chang, Y. Y. Polym Plast Technol Eng 2006, 45, 1251.
5. Poh, B. T.; Chee, C. L. Int J Polym Mater 2007, 56, 247.
6. Poh, B. T.; Kwo, H. K. Polym Plast Technol Eng 2007, 46, 1021.
7. Poh, B. T.; Kwo, H. K. J Appl Polym Sci 2007, 105, 680.
8. Poh, B. T.; Tan, B. K. J Appl Polym Sci 1991, 42, 1407.
9. Poh, B. T.; Lee, K. S. Eur Polym J 1994, 30, 17.
10. Satas, D., Ed. Handbook of Pressure-Sensitive Adhesive Technology; Van Nostrand Reinhold: New York, 1982; p 41.
11. Gierenz, G.; Karmann, W., Eds. Adhesives and Adhesive Tapes; Wiley-VCH: New York, 2001; p 4.
12. Lee, L. H., Ed. Adhesive Bonding; Plenum: New York, 1991; p 19.