

Peel and Shear Strength of Pressure-Sensitive Adhesives Prepared From Epoxidized Natural Rubber

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Received 21 September 2006; accepted 20 December 2006

DOI 10.1002/app.26072

Published online 29 March 2007 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Peel and shear strength of two grades of epoxidized natural rubber (ENR 25 and ENR 50)-based pressure-sensitive adhesive was studied. Coumarone-indene resin was used as the tackifier, whereas toluene was chosen as the solvent throughout the experiment. The tackifier loading was varied from 0 to 80 parts per hundred parts of rubber (phr). A SHEEN hand coater was used to coat the adhesive on substrate to give a coating thickness of 30, 60, 90, and 120 μm . Peel strength and shear strength of the adhesive were determined by using a Lloyd adhesion tester and Texture analyzer, respectively. Results show that maximum peel strength occurs at 40 phr

of coumarone-indene resin for both ENRs studied an observation, which is attributed to the maximum wettability of the substrate. However, the shear strength shows a gradual decrease with increasing tackifier loading because of the decrease in cohesive strength of adhesive. ENR 25 consistently indicates higher peel strength and shear strength than ENR 50. Generally, peel and shear strength increases with coating thickness. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 105: 680–684, 2007

Key words: adhesion; peel strength; shear strength; epoxidized natural rubber; tackifier

INTRODUCTION

The curing characteristics and mechanical properties of epoxidized natural rubber (ENR) have been widely studied.^{1–10} All these studies involve the bulk property of ENR. The solution property of ENR, especially its use in pressure-sensitive adhesive (PSA) was not reported. On the other hand, some scientific studies on the physical properties of natural rubber-based PSA were investigated. Leong et al.¹¹ has reported the viscoelastic properties of natural rubber PSA using acrylic resin as a tackifier. It is found that a good natural rubber PSA exhibits higher loss tangent at higher frequencies. Florian and Novak¹² have carried out a study on the adhesive properties of atactic polypropylene (aPP) and its mixtures with styrene-2-ethylhexyl acrylate (S-EHA) copolymer in the presence of oligomer additives, viz. propylene oil and butylene oil of different molecular weights. They found that the mechanical work of adhesion of the mixture containing aPP and oligomer increases with molecular weight of the oligomer. For ternary mixtures of aPP-S-EHA/oligomer, maximum adhesion occurs at about 30 mass-% of S-EHA. Minimum adhesion is observed in the absence of oligomers, an observation which is attributed to the incompatibil-

ity of the remaining components. Recently, we have carried out a systematic study on the adhesion behavior of SMR 10 and SMR 20-based PSA.^{13–14} Results show that peel strength of the adhesive generally increases with an increase in tackifier resin. Also, the 10 min masticated sample indicates the highest peel strength as compared to other masticated samples. With regard to ENR, however, no previous study has been published in this field of interest. It is thus the aim of this article to report some of our findings on the peel and shear properties of the ENR-based PSA.

EXPERIMENTAL

Materials

ENR 25 and ENR 50 having 25 and 50 mol % of epoxidation were used as the elastomer for the preparation of PSA. The rubbers were supplied by Guthrie Company of Malaysia. Technical specification of the rubbers is given in Table I.

Coumarone-indene resin that has a molecular weight of 1000–3000 and specific gravity of 1.07 was obtained from EuroChemo-Pharma Company (Malaysia). Toluene and polyethylene terephthalate (PET) film were of commercial grades.

Preparation of PSA

ENR was mechanically masticated on a two-roll mill for 5 min prior to use. For each preparation of adhe-

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Contract grant sponsor: Universiti Sains Malaysia; contract grant number: 304/PTEKIND/636095.

TABLE I
Technical Specification of ENR

	ENR 25	ENR 50
Glass transition temperature (°C)	-45	-20
Specific gravity	0.97	1.03
Mooney viscosity, M_L , 1+4 (100°C)	110	140

sive, 10 g of masticated rubber was used. The milled rubber sheet was cut into smaller strips and dissolved in 50 mL of toluene. The rubber solution was prepared without mixing. It was tightly closed to prevent evaporation of solvent and kept in a conditioned room at 28°C for 24 h to ensure complete dissolution. Four different weights, i.e., 2, 4, 6, and 8 g of pulverized coumarone-indene resin corresponding to 20, 40, 60, and 80 phr of resin were then added separately to the rubber solution with manual stirring until a homogeneous PSA was produced for testing.

Methods

Peel Test

The substrate chosen for this test is PET film. It was coated using a SHEEN Hand Coater at 30, 60, 90, and 120 μm coating thickness. Three testing modes were carried out, viz. T-Peel Test, 90° Peel Test, and 180° Peel Test. For the T- and 90° Peel Tests, the dimension of substrate was 25 \times 100 mm², whereas for the 180° Peel Test it was 25 \times 150 mm². However, the coating area for all the testing modes was 50 \times 25 mm² at the end of substrate. The sample was then conditioned at room temperature for 24 h before testing. A Lloyd Adhesion Tester operating at a testing rate of 500 mm/min was used to determine the peeling force of the sample. 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).

Shear Test

The dimension of the shear test sample was 25 \times 150 mm². Coating was carried at the center of the substrate with 25 \times 50 mm² dimension. The coated sample was conditioned at room temperature for 24 h before testing. A TA-HDi texture analyzer (Model-Stable Micro System) was used to determine the shear strength of the sample. The testing speed was 1 mm/s up to 50 s and the testing distance was 50 mm corresponding to the length of coated area. Shear strength was expressed as the force per unit area of testing.

RESULTS AND DISCUSSION

The effects of coumarone-indene resin, coating thickness, and degree of epoxidation on the peel and

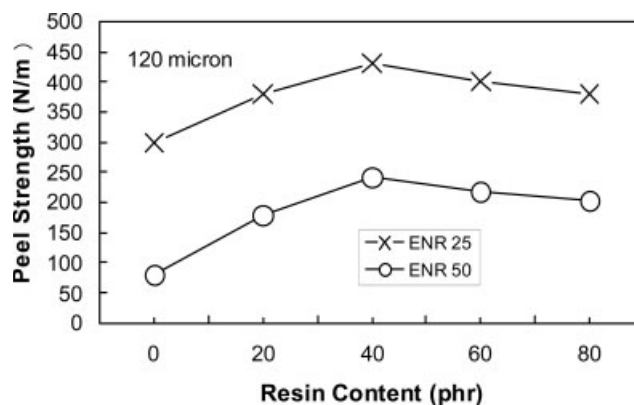


Figure 1 T-peel strength versus resin content.

shear strength of ENR-based PSA are discussed later.

Peel Strength

The peel strength between PET/PET films using a T-Peel test is shown in Figure 1. For both ENRs studied, the peel strength increases rapidly to a maximum value at 40 phr of tackifier resin and after, which it drops gradually with further loading of resin. Similar observation was reported by Leong et al.¹¹ who obtained a peak value at 40% acrylic resin in the SMR 5/acrylic resin system. The initial rapid increase of peel strength with increasing resin content is attributed to the increasing wettability of the adhesive on the substrate, resulting in mechanical interlocking and anchorage of the adhesive in pores and irregularities in the adherent.¹⁵⁻¹⁶ This increase in wettability and mechanical interlocking reaches a peak value at 40 phr resin content. Further addition of resin will decrease the wettability slowly because of the dilution effect of resin, which dominates the peel adhesion behavior, hence lower peel strength is observed.

Similar results are obtained for the 90° and 180° Peel Tests as shown in Figures 2 and 3, respectively.

For the three peel tests, ENR 25 consistently indicates higher peel strength compared to ENR 50. Figure 4 compares the peel strength between the two rubbers at 40 phr resin and 120 μm coating thickness.

The greater flexibility of ENR 25 is due to its lower T_g (-45°C) compared to ENR 50 (-20°C) as a result of higher epoxidation in the latter. Therefore, ENR 25 exhibits better viscoelastic property and enhances better wettability on the testing substrate. In addition, the rubber chains also undergo more strain-induced crystallization^{7,17-20} to increase its resistance to rupture under an applied force, i.e., the adhesive layer itself cannot easily be ruptured.²¹ Figure 4 also shows 90° Peel Test gives higher peel strength because of the mode of testing.

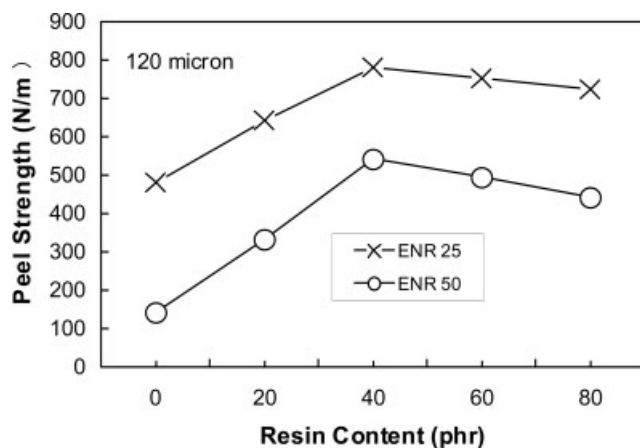


Figure 2 Peel strength (90°) versus resin content.

The dependence of peel strength on coating thickness for both rubbers is shown in Figure 5 for the 90° Peel Test at 20 phr of resin content.

The results obviously show that increasing thickness of coating will increase the peel strength of the ENR-based adhesive. This observation is consistent to the general belief that peel force increases with increasing adhesive thickness up to a certain limit.²² In this study, no maximum is observed indicating that the maximum coating thickness has not been reached. Increasing adhesive thickness causes the shift from cohesive to adhesive failure.¹⁴ Figure 5 also shows that peel strength of ENR 25-based PSA is always higher than that of ENR 50 for all the coating thickness studied. The difference in peel strength (Z) between the two rubbers is shown in Figure 6.

Z has a higher value at higher coating thickness suggesting that the rate of increase in peel strength with coating thickness is greater in ENR 25-based PSA because of the greater flexibility of the rubber to enhance better wettability as discussed earlier.

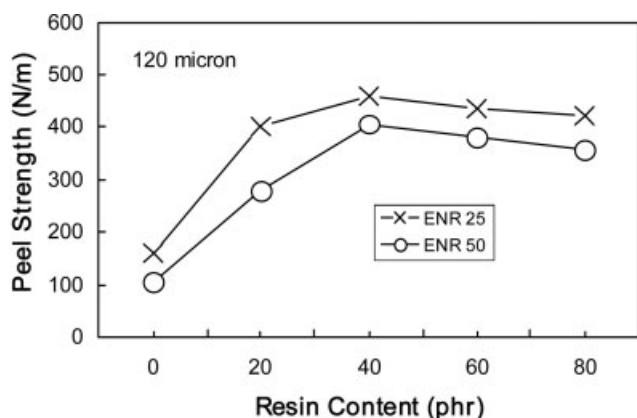


Figure 3 Peel strength (180°) versus resin content.

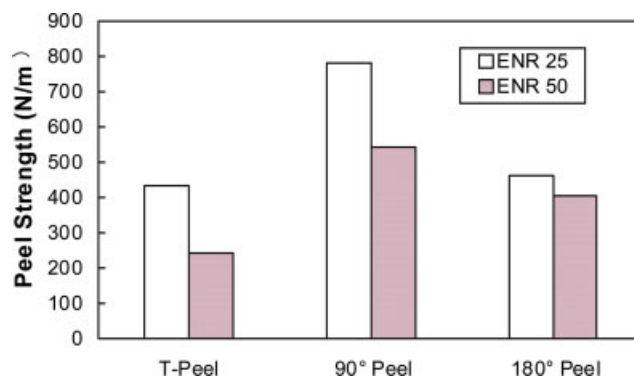


Figure 4 Comparison of peel strength between ENR 25 and ENR 50. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

Shear Strength

Figures 7 and 8 shows the dependence of shear strength on coumarone-indene resin content for ENR 25 and ENR 50-based PSA, respectively. It can be seen that shear strength decreases with increasing resin loading for all the coating thickness studied. The rate of decrease of shear strength is greatest for the thicker coating, i.e., 120 μm . This decrease in shear strength—a measure of holding power—of a PSA is attributed to the decrease in cohesive strength because of the decreasing amount of rubber content, which acts as the binder in the adhesive system. Increasing the resin loading will increase its dilution effect, which in turn weakens the adhesive to resist flow during shearing action. This weakening effect is very significant at higher coating thickness as shown in Figures 7 and 8. However, for thinner coating thickness, shear strength is affected slightly by the increase in resin content as reflected by the gradual decrease in shear strength with resin loading. Figure 8 also shows that ENR 50-based PSA indicates a faster decrease in shear strength compared to that of ENR 25. This observation may be attributed to

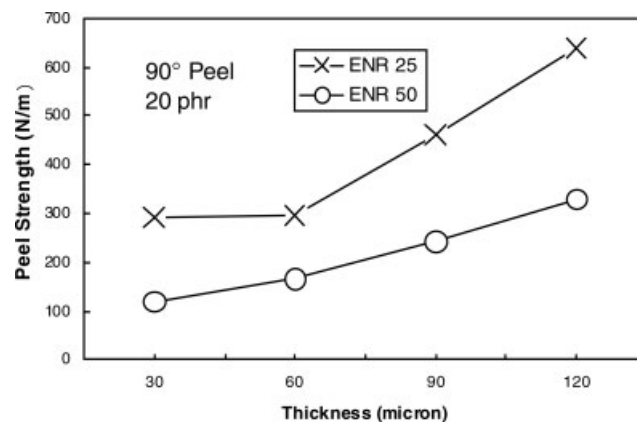


Figure 5 Peel strength versus coating thickness.

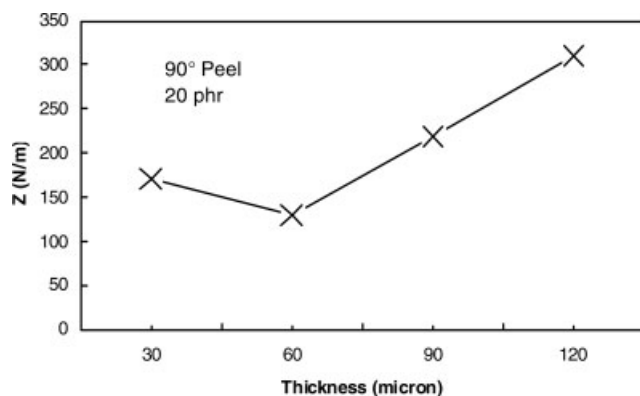


Figure 6 Dependence of peel strength difference (Z) between ENR 25 and ENR 50 on coating thickness.

poorer flexibility of the former to resist shearing forces, especially at higher resin loading. For all the coating thickness studied, ENR 25 consistently exhibits higher shear strength than ENR 50 as shown in Figure 9 at 40 phr of resin loading.

At 30 μm coating thickness, the shear strength of ENR 25-based PSA is about 4.5 times higher than that of ENR 50, whereas at 120 μm thickness, it is about 5 times higher. Higher shear strength of ENR 25-based PSA is ascribed to the flexibility and better crystallizability of the rubber when subjected to shearing action. Also, better compatibility between ENR 25 and coumarone-indene resin may contribute to the higher shear strength as shown in Figure 9. The rubber can resist shear deformation better than ENR 50 especially at higher coating thickness where the effect of ENR 25 becomes more pronounced.

CONCLUSIONS

The following conclusions can be drawn from this study:

1. For both ENRs studied, peel strength increases with increasing coumarone-indene resin content

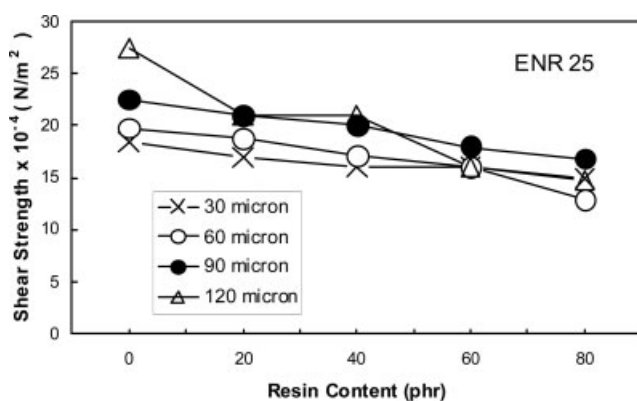


Figure 7 Shear strength versus resin content for ENR 25.

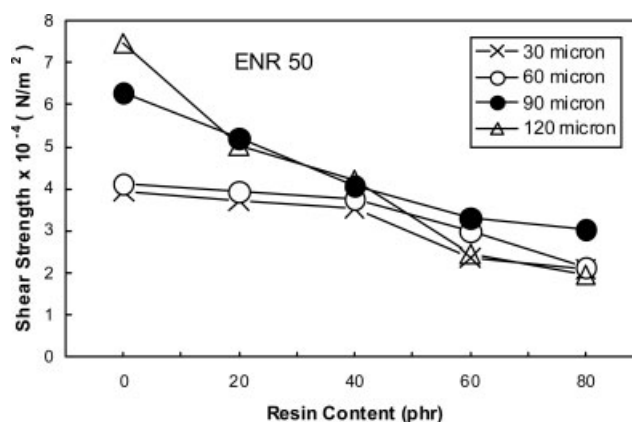


Figure 8 Shear strength versus resin content for ENR 50.

up to 40 phr of resin, after which it drops gradually with further loading of resin. This observation is attributed to the increasing wettability of the adhesive on the substrate, resulting in mechanical interlocking and anchorage of the adhesive in pores and irregularities in the adherent. ENR 25 consistently shows higher peel strength compared to ENR 50 because of the greater flexibility and strain-induced crystallization of the former. Increasing coating thickness will increase the peel strength of the ENR-based adhesive.

2. Shear strength decreases with increasing resin loading for all the coating thickness studied. The rate of decrease is greatest for the thicker coating sample. This observation is associated with the decrease in cohesive strength because of the dilution effect of resin, which weakens the adhesive to resist flow during shearing action. Similarly, ENR 25 exhibits higher shear strength than ENR 50 especially at higher coating thickness because of the greater flexibility and compatibility with resin in the former.

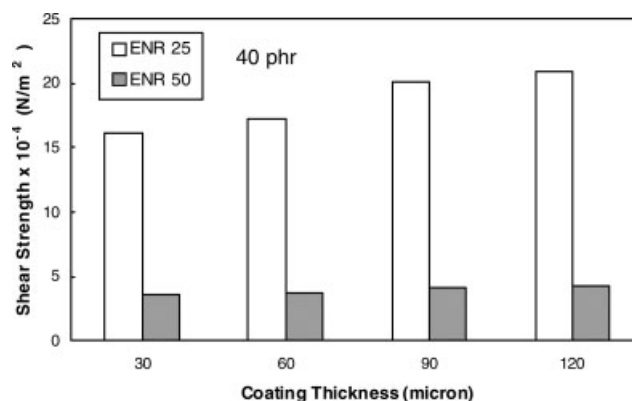


Figure 9 Comparison of shear strength between ENR 25 and ENR 50 for various coating thickness.

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