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Effect of Coumarone-Indene Resin On Adhesion Property of SMR 20-Based Pressure-Sensitive Adhesives

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The viscosity, tack, and peel strength of a natural rubber (SMR 20)-based pressure-sensitive adhesive (PSA) was studied using coumarone-indene resin as the tackifier. The resin loading was varied from 0–80 parts per hundred parts of rubber (phr). Toluene was used as the solvent throughout the experiment. The viscosity of PSA was measured using a Haake Rotary Viscometer whereas loop tack and peel strength were determined using a Lloyd Adhesion Tester. PSA was coated onto the substrates using a SHEEN hand coater to give a coating thickness of 60 μm and 120 μm . Results show that the viscosity and tack of the adhesive increases with resin content due to the concentration effect of tackifier resin. However, for the peel strength, it increases up to 40 phr of resin for both coating thickness, an observation that is attributed to the wettability of substrates.

Keywords: adhesive, natural rubber, peel strength, tack, viscosity

INTRODUCTION

Natural rubber has been widely used as the elastomer in the preparation of pressure-sensitive adhesives (PSA). However, scientific reports on the physical properties of natural rubber-based PSA are scarce although other systems have been reported recently [1–2]. Leong et al. [3] investigated the viscoelastic properties of natural rubber PSA using acrylic resin as a tackifier. The blends were coated onto strips of paper and tested for shear and peel strengths. It was found that for a good PSA, the ratio of storage modulus at high frequencies

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to low frequencies should be high. A higher loss tangent at high frequencies was reported for a good natural rubber PSA. No previous study has been reported for Standard Malaysian Rubber grade 20 (SMR 20) as an elastomer in PSA so far. In view of the scarcity of data, the authors have thus carried out a systematic study on the effect of coumarone-indene resin tackifier on the viscosity, tack, and peel strength of SMR 20-based PSA. The effect of coating thickness on tack and peel strength is also investigated and discussed in this article.

EXPERIMENT

Materials

Standard Malaysian Rubber (SMR 20 grade) was used as the base elastomer for the preparation of PSA. Technical specification [4] of SMR 20 are given in Table 1.

Coumarone-indene resin, which was freshly supplied by Euro-Chemo-Pharma Company (Malaysia), was used as the tackifier in this study. Toluene was used as the solvent to prepare the PSA throughout the experiment. The rubber, resin, and toluene were used as received and no further purification was carried out.

Preparation of PSA

SMR 20 was mechanically masticated on a two-roll mill for 5 min prior to use. For each preparation of adhesive, 5 g of masticated rubber was used. It was cut into small pieces to facilitate easy dissolution. 20 ml of toluene were added to each rubber sample and the rubber solution was left in a conditioned room for 24 h to ensure complete dissolution. Different weights (1, 2, 3, and 4 g) of coumarone-indene resin—corresponding to 20, 40, 60, 80 phr—were then added slowly to the rubber solution to prepare the SMR 20-based PSA.

Measurements

Viscosity and Tack

Viscosity of the adhesives was determined by a Haake Rotary Viscometer (Model PK 100). For the loop tack test, which is the peel

TABLE 1 Technical Specifications of SMR 20

Dirt content (max. % wt)	0.20
Ash content (max. % wt)	1.00
Nitrogen (max. %wt)	0.60
Volatile matter (max. % wt)	0.80
Plasticity retention index (min.%)	40

test involving low contact pressure and short application time [5], the adhesive was coated on a strip of A4 paper (4 cm × 25 cm) using a SHEEN Hand Coater. The coated area was 4 cm × 4 cm at the center of the paper strip at a coating thickness of 60 μm and 120 μm. The sample was conditioned at room temperature for 24 h before testing. The paper strip was then formed into a loop and brought into contact with a glass plate until the coated area was in full contact with the glass plate. A Lloyd Adhesion Tester operating at a testing rate of 10 cm/min was used to determine the pulling force to detach the loop from the glass plate. The tack value was expressed as the detached force per area of contact.

Peel Strength

The peel adhesion test was carried out by using A4 paper (base stock) and polyethylene terephthalate (PET) film (face stock) as substrates. Three modes of peeling tests were conducted: T-Peel Test, 90° Peel Test, and 180° Peel Test.

T-peel and 90° Peel Tests. For the T- and 90° Peel Tests, the size of the sample was 20 cm × 4 cm. The adhesive was coated on the A4 paper to cover an area of 10 cm × 4 cm at coating thickness of 60 μm and 120 μm. Then, PET film was stuck on the A4 paper. The testing sample was conditioned for 24 h before testing on a Lloyd Adhesion Tester at a testing rate of 10 cm/min. Five testing specimens were used for each adhesive formulation.

180° Peel Test. For the 180° Peel Test, the size of A4 paper and PET film were 25 cm × 4 cm and 12 cm × 6 cm, respectively. The adhesive was coated on the A4 paper covering an area of 10 cm × 4 cm at a coating thickness of 60 μm and 120 μm. PET film was then stuck on the A4 paper and the sample was conditioned for 24 h before testing. Again, Lloyd Adhesion Tester operating at 10 cm/min was used to determine the peel force. Five specimens were used for each adhesive formulation. From the load-propagation graph, the three highest peaks were chosen to compute the average peeling force. 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 are discussed with respect to the effect of coumarone-indene resin content on the viscosity, tack, and peel adhesion strength of the SMR 20-based PSA. The effect of coating thickness on tack and peel strength is also discussed.

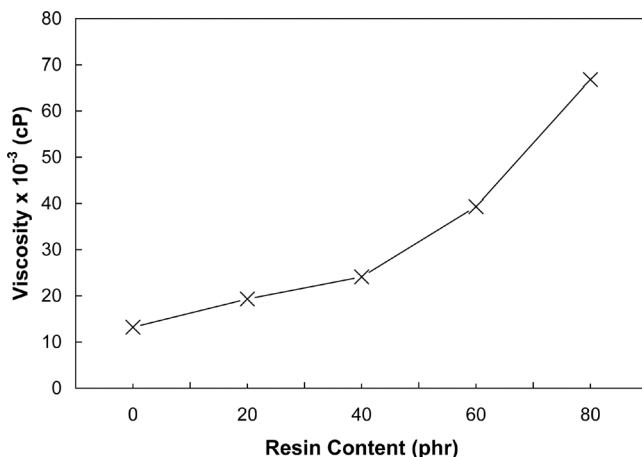


FIGURE 1 Variation of viscosity with coumarone-indene resin content for the SMR 20 based PSA.

Viscosity of Adhesive

Figure 1 shows the dependence of the viscosity of the adhesive on coumarone-indene resin content. The viscosity increases gradually up to 40 phr resin and after that, it increases rapidly. The initial gradual increase in viscosity is attributed to the concentration effect of the resin. However, at higher resin concentration, stronger rubber-resin interaction occurs as a result of increasing concentration of the rubber-resin solution. Besides, above 40 phr resin content, there is a possibility of chain entanglement between rubber chains and resin molecules [6] as the critical resin concentration for entanglement is exceeded.

Tack

Tack may be defined as the property of a material that enables it to form a bond of measurable strength immediately upon contact with another surface, usually with low applied pressure [7]. Tack gives an indication of how quickly an adhesive can wet and make intimate contact with a surface. In this study of SMR 20-based PSA, the adhesive tack obtained from the Loop Tack Test is shown in Figure 2 for 60 μm and 120 μm coating thickness. For both coating thicknesses, tack increases gradually up to 40 phr resin. After that critical concentration, a sudden increase in tack is observed. Tackifying resin increases tackiness by softening the natural rubber matrix. A key

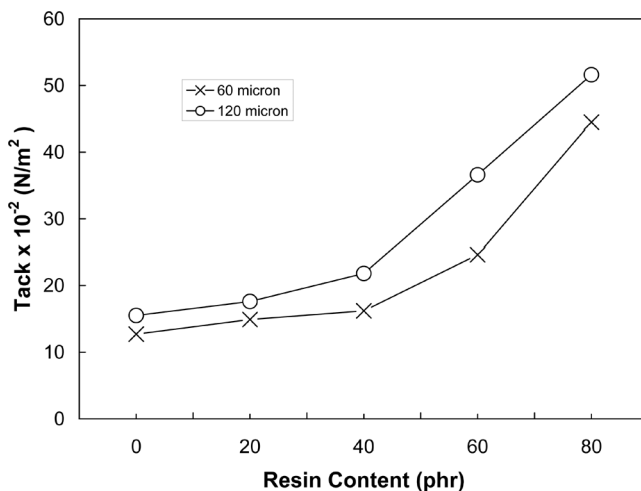


FIGURE 2 Variation of loop tack with coumarone-indene resin content at two different coating thickness.

property of this adhesive is that tack does not build significantly without tackifier. So, PSA will be able to form a bond of measurable strength immediately on contact with another surface. With respect to coating thickness, Figure 2 indicates that higher coating thickness shows a higher tack value. Increasing the coating thickness means to increase the mass of adhesive containing higher concentration of tackifying resin. The results obtained in this study are consistent with those reported for NR/Staybelite Ester where the probe tack shows a rapid increase after about 55 phr resin concentration [8]. The increase in adhesive coating thickness will enhance the wettability of the adhesive, thus higher tack is observed.

Peel Strength

T-Peel Test

The peel strength between paper/PET film using a T-Peel test is shown in Figure 3. In the peel test, the stress is confined to a very fine line at the edge of the joint. For both coating thickness, the peel strength increases to a maximum value at 40 phr resin content and drops with further addition of the resin. This result agrees well with that obtained by Leong et al. [3] who obtained a peak value at 40% acrylic resin in the SMR 5/acrylic resin system. The increase of peel strength up to 40 phr resin is attributed to the increasing wettability

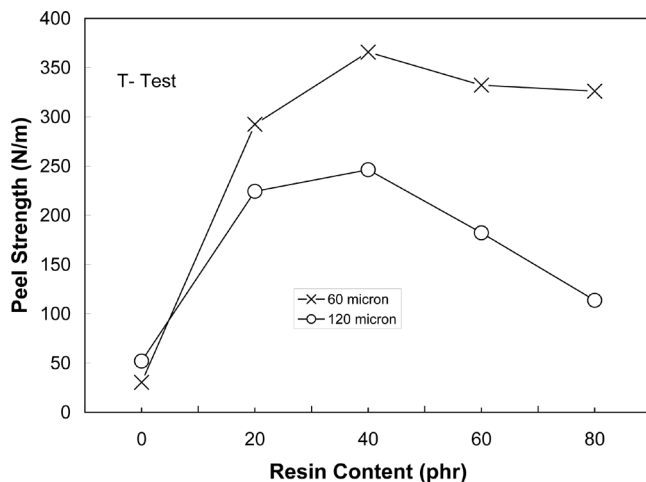


FIGURE 3 Dependence of peel strength (T-Test) on coumarone-indene resin content at two different coating thickness.

of the adhesive on the substrate, resulting in mechanical interlocking and anchorage of the adhesive in pores and irregularities in the adherent [5,9]. The adhesive at 40 phr resin content hardens at high strain levels to become a tough solid, it cannot easily be ruptured and hence the highest peel strength is obtained. Owing to the dilution effect with further addition of resin, peel strength decreases as shown in Figure 3. This is because the wettability of adhesive decreases beyond 40 phr resin and lesser interfacial interaction with the substrate occurs. With respect to coating thickness, it is generally accepted that peel force increases with increasing adhesive thickness up to a certain limit [8]. The present study of SMR 20/coumarone-indene resin based adhesive indicates that 60 μm thickness has a higher peel strength compared to 120 μm thickness, which means that the coating limit has been exceeded. Increasing adhesive thickness causes shift from cohesive to adhesive failure.

90° and 180° Peel Test

The effect of coumarone-indene resin content on the 90° and 180° Peel Tests is shown in Figure 4 and Figure 5, respectively. For both studies, the peel strength increases to a maximum at 40 phr resin and drops with further addition of resin. This observation is attributed to the increasing wettability up to a certain limit of resin concentration and after which dilution effect of resin dominates the peel adhesion behavior. This result is similar to that obtained for the

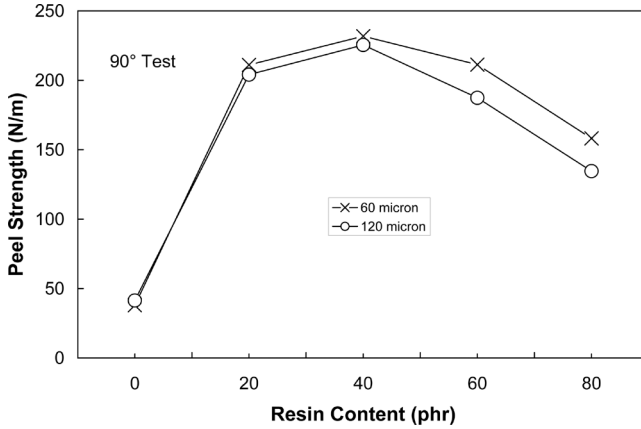


FIGURE 4 Dependence of peel strength (90° Peel Test) on coumarone-indene resin content at two different coating thickness.

T-Test. For the coating thickness, 60 μm sample again shows a higher peel strength compared to 120 μm coated thickness. However, for both 90° and 180° Peel Tests, the effect of coating thickness is not so significant compared to that of T-Test. This observation may be due to the mode of testing indicating that cohesive to adhesive failure transition is not so obvious in the two former tests.

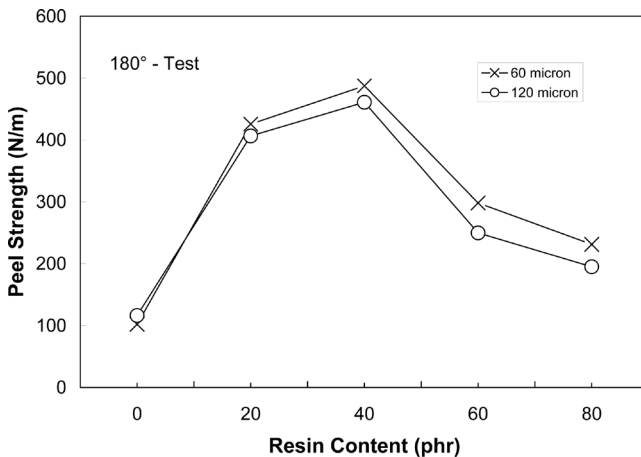


FIGURE 5 Dependence of peel strength (180° Peel Test) on coumarone-indene resin content at two different coating thickness.

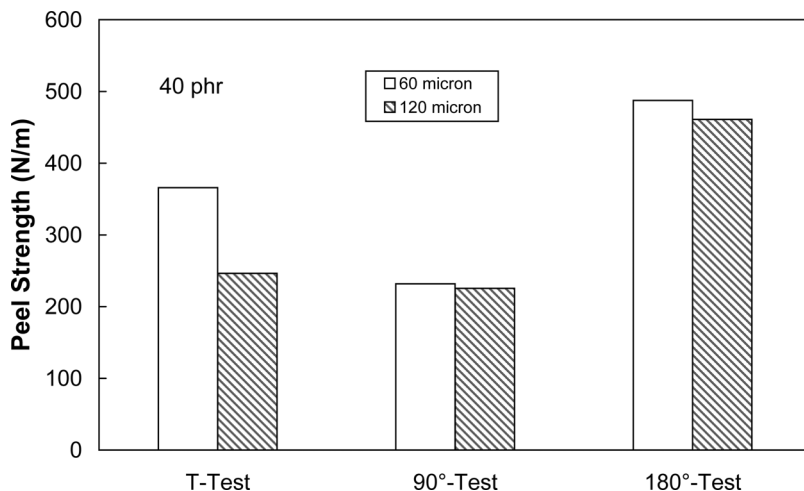


FIGURE 6 Comparison of peel strength between three modes of peel tests at two different coating thickness.

Figure 6 compares the effect of coating thickness for the three different modes of peel tests. The 60 μm thickness sample consistently shows a higher peel strength for the three types of peel tests as a result of better wettability. For a fixed coating thickness, 180°-Test exhibits the highest peel strength, followed by the T-Test and 90° Peel Test. This phenomenon is associated with the difference in the angle of testing, indicating that a higher peel force is needed in the case of 180° Peel Test. This means that for 180° Peel Test, the rubber chains undergo more strain-induced crystallization [10–14] than the other two peel tests. The adhesive hardens at high strain levels to become a tough solid and the adhesive layer itself cannot easily be ruptured [7] in the case of 180° Peel Test.

CONCLUSIONS

The following conclusions can be drawn from this study:

1. The viscosity of the SMR 20-based PSA increases gradually up to 40 phr resin and after which, it increases rapidly. This observation is attributed to the concentration effect of the resin and stronger rubber-resin interaction at higher resin content. The loop tack similarly shows a sudden increase in tack after 40 phr of resin concentration for both coating thickness. A higher tack value is obtained for higher coating thickness due to the increase of the mass of adhesive.

2. Regardless of the peel test modes, the peel strength increases to a maximum value at 40 phr resin content and drops with further loading of resin. This is ascribed to the increased wettability of the adhesive on the substrate, resulting in mechanical interlocking and anchorage of the adhesive in pores and irregularities in the substrate. However, after the peak value, dilution effect causes the peel strength to decline with further addition of resin. The 60 μm -coated sample consistently indicates a higher peel strength compared to the 120 μm sample, indicating a shift from cohesive to adhesive failure. Owing to the difference in the angle of testing, 180° Peel Test exhibits a higher peel strength than the corresponding T- and 90° Peel Tests.

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