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Effect of Molecular Weight of Rubber on Tack and Peel Strength of SMR L-Based Pressure-Sensitive Adhesives using Gum Rosin and Petroresin as Tackifiers

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Loop tack and peel strength of natural rubber (SMR L)-based pressure-sensitive adhesive were studied using five different molecular weights of SMR L. Gum rosin and petroresin were used as tackifiers, whereas toluene was chosen as the solvent throughout the experiment. A SHEEN hand coater was used to coat the adhesive on a polyethylene terephthalate (PET) substrate at a coating thickness of 30, 60, 90 and 120 μm. Loop tack and peel strength were determined by a Llyod Adhesion Tester operating at 30 cm/min. Results show that maximum values of loop tack and peel strength were obtained at a molecular weight of 8.5×10^4, an observation which is attributed to maximum wettability of adhesive on the substrate. Loop tack and peel strength increases with coating thickness for all molecular weight of rubber and tackifiers studied.

Keywords: Adhesive, rubber, tackifier, molecular weight

1 Introduction

Pressure-sensitive adhesives are commonly prepared from natural rubber. In order to achieve the desired tack, peel adhesion and shear properties, tackifiers are added to the rubber (1). The rubber provides the elastic component whereas the tackifier imparts the viscous component. Systematic studies on the adhesion properties of rubber-based adhesives were scarcely reported. Kraus et al. (2) have reported the effect of entanglement plateau on the adhesive behavior of the styrene-diene-based pressure-sensitive adhesives. They (3) have also studied the structural changes in melts of butadiene-styrene and isoprene-styrene block polymer-based adhesives. Leong et al. (4) have studied the viscoelastic properties of natural rubber pressure-sensitive adhesive using acrylic resin as a tackifier. Fujita et al. (5), on the other hand, reported the effects of miscibility and viscoelasticity on shear creep resistance of natural rubber-based pressure-sensitive adhesives. Besides the rubber-based adhesives, other adhesive systems have also been investigated (6–7). Recently, we have reported several studies on the adhesion properties of Standard Malaysian Rubber (SMR L, SMR 10 and SMR 20 grades)-based pressure-sensitive adhesives (8–10). Results have shown that viscosity, peel strength and tack of SMR-based adhesives generally increases with an increase of tackifier loading, an observation which is attributed to the increased wettability and formation of mechanical interlocking, and anchorage of adhesive in pores and irregularities in the substrate. However, shear strength decreases gradually with increasing tackifier concentration due to the decreasing cohesive strength of adhesive as resin loading is increased. We have also carried out a systematic study on the adhesion properties of adhesives prepared from styrene-butadiene rubber (SBR)/Standard Malaysian Rubber (SMR L) blends (11–12). It is observed that the viscosity of the SBR/SMR L-based adhesive decreases with increasing % SBR. Loop tack of the rubber blend-based adhesive passes through a maximum value at 20% SBR composition for all resin loadings investigated. For the coumarone-indene resin adhesive, peel strength exhibits maximum value at 40% SBR whereas for the phenol-formaldehyde resin system, maximum peel strength occurs at 60% SBR composition. With regard to the effect of molecular weight of rubber on the adhesion property of pressure-sensitive adhesives prepared from SMR rubber, there is virtually no study published so far in this field of interest. Owing to the scarcity of data in this area of research, we have conducted a systematic investigation on the dependence of the loop tack and peel strength of SMR L-based adhesive on molecular weight of rubber using gum rosin and petroresin as the tackifying resins.

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Table 1. Technical specifications of SMR L

Dirt content (max. % wt.)	0.03
Ash content (max. % wt.)	0.50
Nitrogen (max. % wt.)	0.60
Volatile matter (max. % wt.)	0.80
Plasticity retention index (min. %)	60

2 Experimental

2.1 Materials

Natural rubber (SMR L grade) was used as the elastomer for the preparation of pressure-sensitive adhesive. Its technical specification is given in Table 1. Gum rosin and petroresin were used as tackifiers and freshly supplied by EuroChemo-Pharma Company (Malaysia). Commercial grade toluene was chosen as the solvent throughout the experiment.

2.2 Molecular Weight Determination

SMR L was masticated on a two-roll mill for 5, 10, 15 and 20 min in order to obtain different chain length of rubber for this study. The respective molecular weights of rubber (including the unmasticated sample) were determined by viscometry. The intrinsic viscosity $[\eta]$ was obtained according to the method described by Billmeyer (13). The viscosity-average molecular weight (M_v) of the rubber was calculated from the Mark-Houwink equation (14) as shown below:

$$[\eta] = kM_v^a$$

where $k = 5.00 \times 10^{-4}$ dl/g and $a = 0.67$ in toluene.

2.3 Adhesive Preparation

5 g of each rubber sample was cut into smaller strips and dissolved in 30 ml of toluene. The rubber solution was kept in a conditioned room for 24 h before adding 2g of each tackifier -i.e., gum rosin and petroresin- which corresponds to 40 parts per hundred parts of rubber (phr). The resulting rubber solution was constantly stirred and then left for at least 2 h to produce a homogeneous pressure-sensitive adhesive for testing.

2.4 Loop Tack Determination

Loop tack test is the peel test involving low contact pressure and short application time [15]. A polyethylene terephthalate (PET) with dimension of 25 cm × 4 cm was used as the substrate. It was coated at the center (4 cm × 4 cm) with the adhesive using a SHEEN Hand Coater at various coating thickness, i.e., 30, 60, 90 and 120 μm. The coated sample was conditioned at room temperature (30°C) for 24 h before testing. The PET strip was then formed into a loop where the coated adhesive area was gently brought into contact with a glass plate. A Lloyd Adhesion Tester (Model LRXPlus with NEXYGEN software) operating at 30 cm/min was used to determine the debonding force of adhesive from the glass plate at room temperature. The three highest peaks recorded were used to compute the average debonding force. The loop tack of the adhesive was expressed as the average debonding force per unit area of contact (N/m^2).

2.5 Peel Test

Three different modes of peel tests - i.e., T-Peel, 90° Peel and 180° Peel -were conducted on PET substrates. For the T-Peel and 90° Peel Tests, the dimensions of substrates were

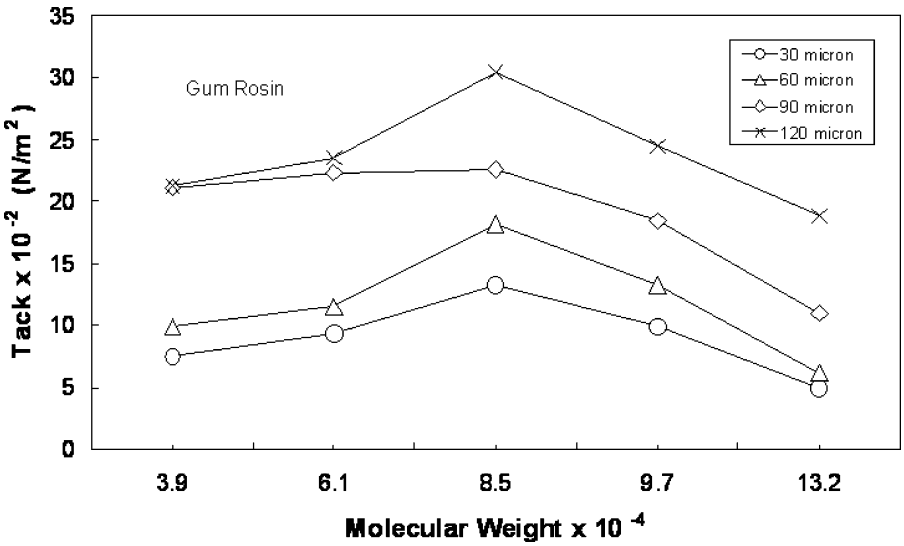

Fig. 1. Variation of loop tack of gum rosin-based adhesive with molecular weight of rubber for various coating thickness.

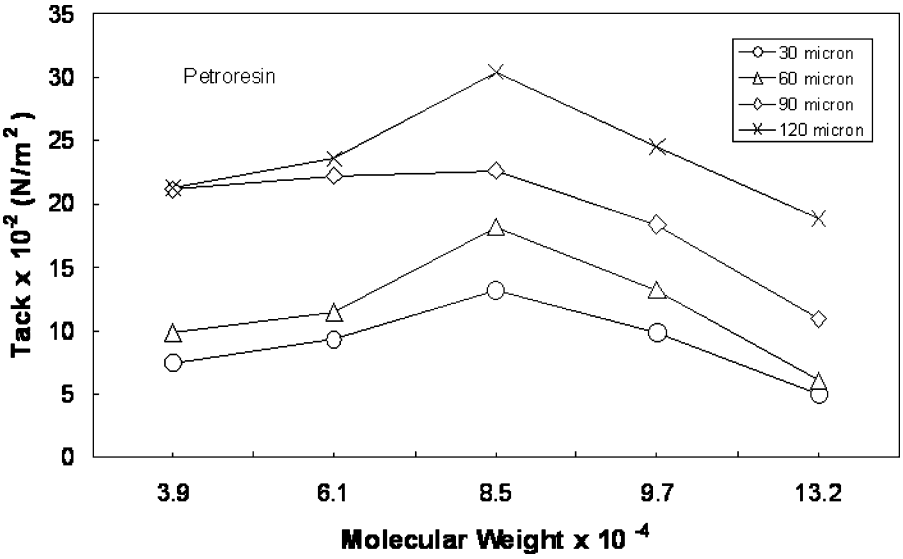

Fig. 2. Variation of loop tack of petroresin-based adhesive with molecular weight of rubber for various coating thickness.

20 cm × 4 cm, whereas for the 180° Peel Test, its dimension was 25 cm × 4 cm. A SHEEN Hand Coater was used to coat the adhesive from the end of the PET film at a coating area of 10 cm × 4 cm. The coating thickness used in this study was 30, 60, 90 and 120 μm. The coated sample was then conditioned for 24 h prior to testing on a Lloyd Adhesion Tester at 30 cm/min. The average peeling force was calculated from the three highest peaks obtained from the 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).

3 Results and Discussion

The dependence of loop tack and peel strength of adhesive on the molecular weight of rubber is discussed below.

3.1 Loop Tack

The effect of molecular weight of rubber on the loop tack of gum rosin-based adhesives is shown in Figure 1 for various coating thickness. From the graph, it indicates that loop tack increases with molecular weight of rubber up to 8.5×10^4, after which it decreases with increasing molecular

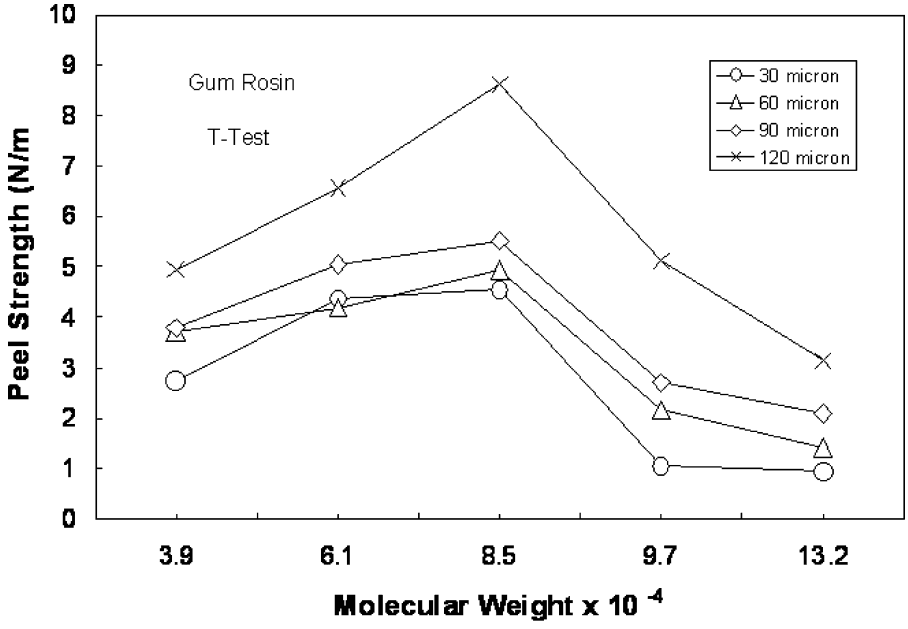

Fig. 3. Variation of peel strength (T-Test) of gum rosin-based adhesive with molecular weight of rubber for various coating thickness.

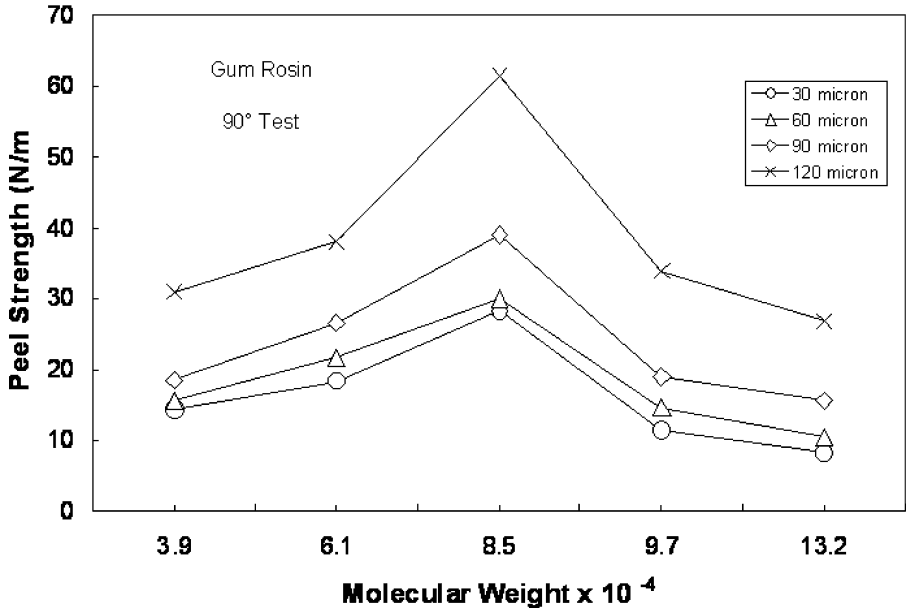

Fig. 4. Variation of peel strength (90° Test) of gum rosin-based adhesive with molecular weight of rubber for various coating thickness.

weight. This observation is attributed to the increasing wettability of adhesive up to a maximum value at 8.5×10^4 optimum molecular weight for maximum wettability for all the coating thickness. At this optimum molecular weight, the adhesive system indicates the optimum elastic and viscous property that is necessary for the maximum tack in a pressure-sensitive adhesive (4). Higher rubber molecular weight would lower the wettability of adhesive due to a poorer viscoelastic response, as reflected by the lower tack value after 8.5×10^4 molecular weight of rubber. Similar behavior is also exhibited by the petroersin-based adhesive as shown in Figure 2. Again, loop tack reaches a maximum value at the same molecular weight of 8.5×10^4 suggesting that optimum molecular weight of rubber to achieve maximum wettability is independent on the tackifier system used. For both tackifiers used, the 120 μm coated sample consistently exhibits the highest tack value, followed by the 90, 60 and 30 μm systems. The presence of higher amount of rubber component in the 120 μm coating thickness enhances viscoelastic response to form adequate tack properties, thus giving rise to higher tack value compared to thinner coating thickness as shown in Figures 1 and 2.

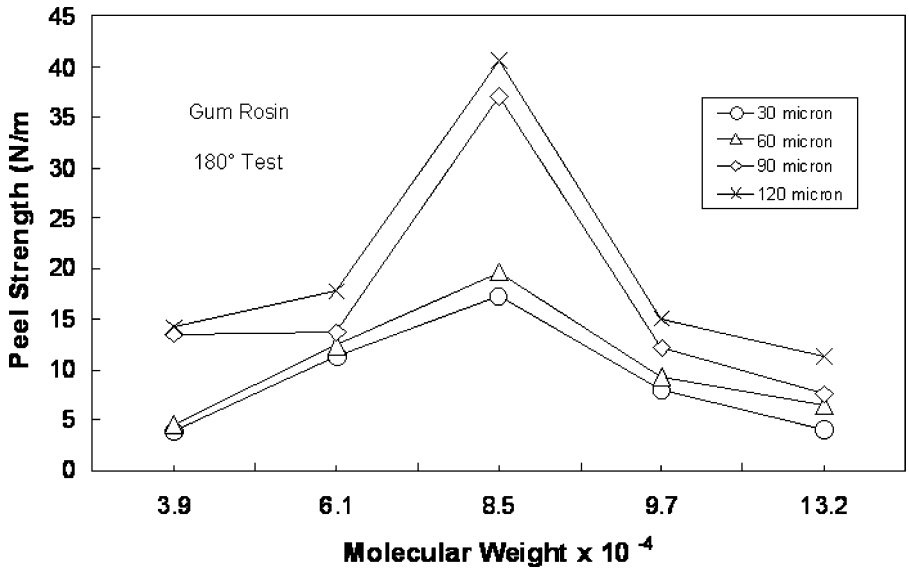

Fig. 5. Variation of peel strength (180° Test) of gum rosin-based adhesive with molecular weight of rubber for various coating thickness.

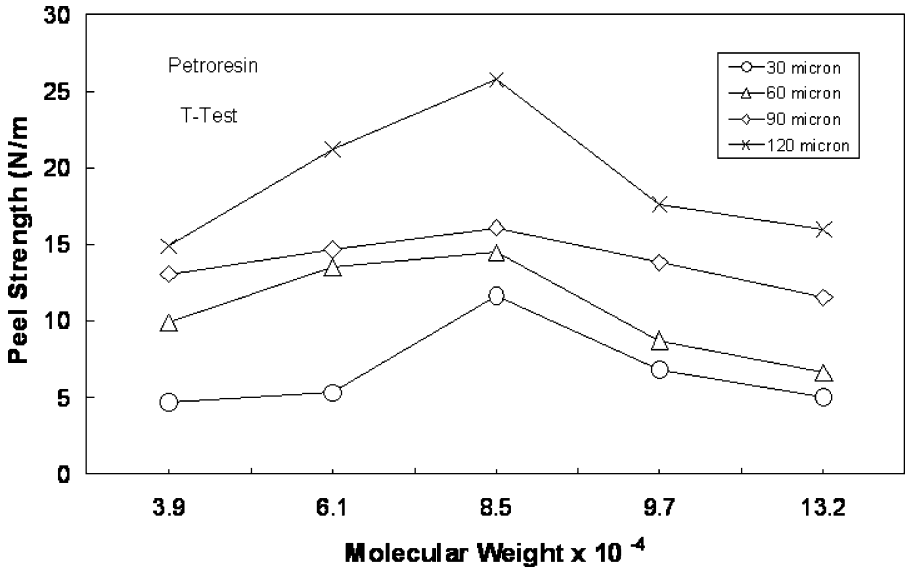

Fig. 6. Variation of peel strength (T-Test) of petroresin-based adhesive with molecular weight of rubber for various coating thickness.

3.2 Peel Strength

Figure 3 shows the dependence of peel strength (T- Test) of gum rosin-based adhesive on the molecular weights of rubber for various coating thickness. It can be seen from the graph that the effect of molecular weight of rubber on peel strength exhibits similar behavior as that reported for the loop tack as discussed earlier. Peel strength also indicates maximum value at 8.5×10^4 molecular weight of rubber, an observation which is attributed to maximum wettability of adhesive on substrate. Maximum mechanical interlocking and anchorage of the adhesive in pores and irregularities in the adherent occurs at the maximum peel strength (16). Peel strength decreases after the optimum molecular weight due to the drop of wettability as a result of increasing effect of chain entanglement as the molecular weight of rubber is increased. Figures 4 and 5 show the results obtained from the other two modes of peel tests, i.e., 90° and 180° tests on the gum rosin-based adhesives. Both graphs indicate similar behavior as that reported for the T-Test, thus confirming that 8.5×10^4 is the optimum molecular weight of rubber for maximum peel strength. For the petroresin-based system, the peel strength obtained

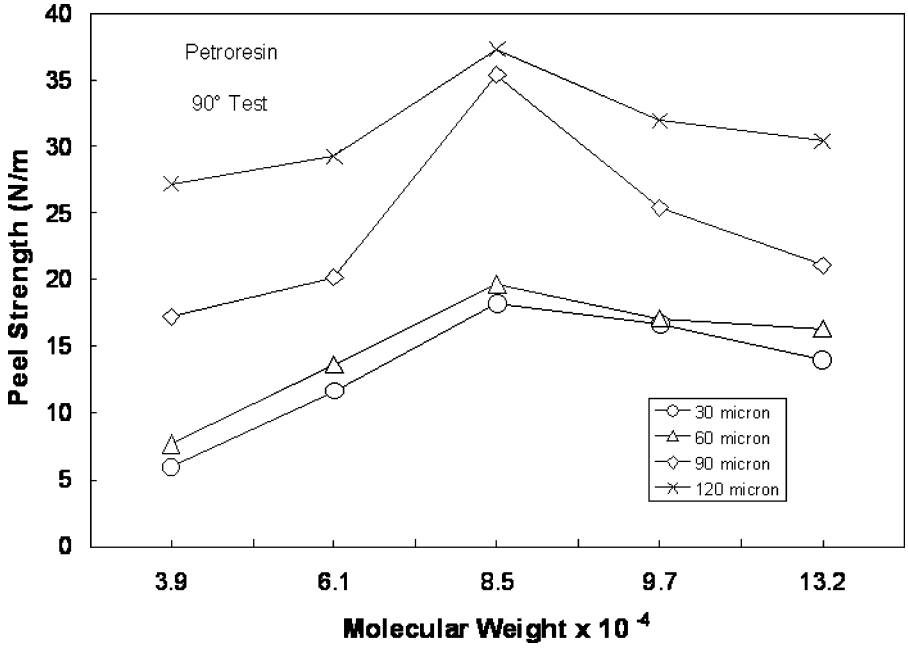

Fig. 7. Variation of peel strength (90° Test) of petroresin-based adhesive with molecular weight of rubber for various coating thickness.

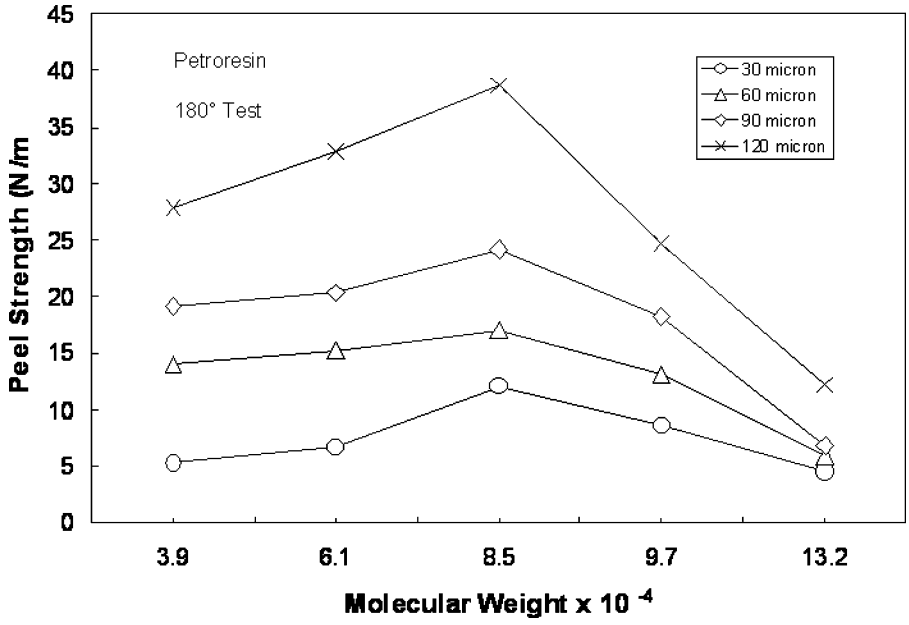

Fig. 8. Variation of peel strength (180° Test) of petroresin-based adhesive with molecular weight of rubber for various coating thickness.

for the T-, 90° and 180° tests are shown in Figures 6, 7 and 8, respectively. All the graphs show the same dependence of peel strength on molecular weight of rubber where maximum peel strength occurs at 8.5×10^4 molecular weight. The maximum peel strength observed is also attributed to the maximum wettability of petroresin-based adhesive on the substrate, thus giving rise to maximum peel strength. For the peel study, it can be seen that 120 μm coated samples consistently exhibits the highest peel strength than the other coating thickness. This observation is similar to that obtained for the loop tack as discussed previously. Thicker coating contains a higher amount of rubber component in the coating layer which will improve the peel resistance of the adhesive. Figure 9 compares the peel strength between gum rosin-based adhesive with that of petroresin. Except for the T-peel test, gum rosin-based adhesive shows higher peel strength than petroresin-based adhesive suggesting that better compatibility exists between the gum rosin and natural rubber, both of which are natural occurring materials. On the other hand, petroresin which is a synthetic resin derived from the polymerization of petroleum cracked products (1) indicates poorer compatibility between the resin and natural rubber. Figure 9 also shows that 90° peel test exhibits the highest peel strength than the other two modes of peel tests. This phenomenon is explained by the angle of testing where higher strain-induced crystallization (17–18) of natural rubber chains occurs at 90° peel test, hence the adhesive layer itself cannot easily be ruptured (19).

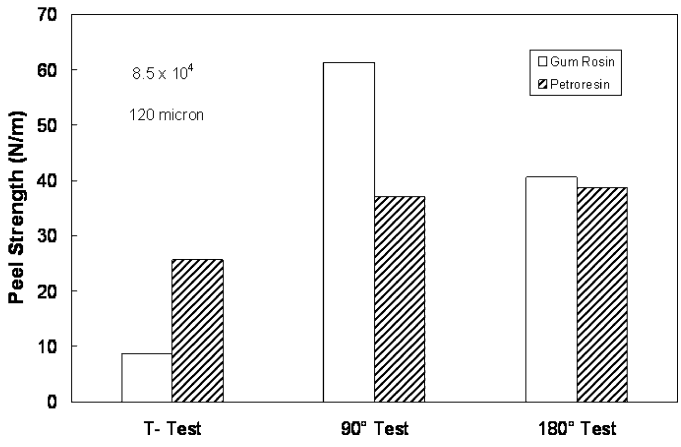

Fig. 9. Comparison of peel strength between gum rosin and petroresin-based adhesives for various modes of peel tests.

4 Conclusions

The following conclusions can be drawn from this study.

1. For both gum rosin and petroresin, loop tack increases with molecular weight of rubber up to 8.5×10^4, after which it decreases with further increase in molecular weight. This observation is attributed to the increasing wettability of adhesive with molecular weight up to 8.5×10^4 which is the optimum molecular weight for maximum wettability for all coating thickness. For a fixed molecular weight, tack value increases with coating thickness due to the presence of higher amount of rubber component in the thicker coating sample.

2. Maximum peel strength is also observed at a molecular weight of 8.5×10^4, an observation which is associated with the maximum wettability of adhesive on substrate at this optimum molecular weight of rubber. The drop in peel strength with increase in molecular weight of rubber is attributed to increasing effect of chain entanglement which lowers the wettability of the adhesive. Gum rosin-based adhesive generally shows higher peel strength than petroresin-based adhesive indicating that better compatibility exists in the former system. Higher strain-induced crystallization of natural rubber in 90° peel test accounts for the higher peel strength observed in this study.

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