### ORIGINAL PAPER

# Material Properties and Statistical Analysis of Natural Rubber-Based Adhesives

Imran Khan · B. T. Poh · Khai Ern Lee

Published online: 15 December 2012

© Springer Science+Business Media New York 2012

**Abstract** Material properties of epoxidized natural rubber (ENR 50)-based adhesive were determined using DSC, TGA and FTIR analysis which was prepared using gum rosin and toluene as tackifier and solvent respectively. Quadratic regression was fitted to model the effect of peel strength and shear strength of the adhesives. Result shows that peel and shear strength increases up to an optimum molecular weight of  $4.2 \times 10^4$  g/mol of ENR 50. Peel strength and shear strength also increases with increase in rate of testing, an observation which is associated to the viscoelastic response of the adhesive. Statistical analysis was carried out to further investigate the effect of molecular weight and testing rate on peel and shear strength.

**Keywords** Polymer · Coating · Adhesion · Thermal properties · FTIR · Statistical analysis

#### **Abbreviations**

ASTM American Society for Testing and Materials

ENR Epoxidized natural rubber

NR Natural rubber

PET Polyethylene terephthalate PVP Poly (vinylpyrrolidone) PSA Pressure-sensitive adhesive

I. Khan  $(\boxtimes)$  · B. T. Poh

Division of Bio-Resource, Paper and Coatings Technology, School of Industrial Technology, Universiti Sains Malaysia, 11800 Penang, Malaysia

e-mail: imran.pchr@gmail.com

K. E. Lee

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

#### Introduction

Natural Rubber (NR) alone is not sufficient to provide the required adhesion and tack of an adhesive. It is necessary to blend tackifier resins with NR in order to improve its wettability to the adherend and to achieve rapid and effective bonding. At specific blend ratios, these rubber- resin blends become pressure-sensitive adhesives (PSAs), while at other concentrations, they do not provide adequate performances as PSAs. PSAs performances, such as peel adhesion, tack, and shear strength, depend strongly on viscoelastic properties of the PSAs [1-3]. Moreover, miscibility between the components is one of the key factors in proper selection because it has a great influence on practical performances of PSAs [4–6]. Therefore, it is very important to clarify the relationship between miscibility, viscoelastic properties, and performances of PSAs systematically. PSA is defined as a viscoelastic material, which in a solvent free state remains permanently tacky at room temperature [7]. Allen [8] reviewed on different theories of adhesion and revealed electrostatic interactions across an interface have been more deeply explored and their significance recognized and expounded. Detail review [9] about natural rubber based adhesive and adhesion property is summarized by our group recently. These are characterized by instantaneous adhesion upon application of light pressure [10]. The most common products that utilize PSAs are tapes, labels, and protective films. The PSA sector is among the fastest growing in the adhesive market, making the search for new pressure-sensitive products (PSP) and applications highly competitive. When PSAs were made of NR and resins, their use was restricted to low technology application such as adhesive plasters and packaging or covering tapes. To describe mechanisms of adhesion several theory such as mechanical interlocking, electrostatic, diffusion, wetting, chemical bonding



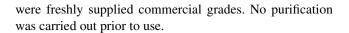
and adsorption/surface reaction theories have been postulated to understand. There are several failures to describe the mechanism of adhesion i.e. from adhesive to cohesive failure. If the bond failure occurs between the adhesive layer and one of the adherends, it is called adhesive failure and a failure in which the separation occurs in such a manner that both adherend surfaces remain covered with the adhesive is called cohesive failure in the adhesive layer. Kim and Mizumachi founded [11] miscibility and peel strength of acrylic pressure-sensitive adhesives of acrylic copolymer-tackifier resin systems. The miscible systems exhibit higher peel strength and tack value compared with immiscible ones. Miscibility in the rubber/resin system depends on the solvent, the overall bulk resin concentration, the thickness of the film and the equilibration time after the film has dried [12]. Pal et al. [13] investigated effect of fillers on morphological properties and wear characteristics of carboxylated acrylonitrile butadiene rubber (XNBR)/NR blends in the presence of different types of carbon black. Saito et al. [14] characterized the epoxidized natural rubber was by 1D- and 2D NMR spectroscopy.

The aim of the present study is to investigate the effect of molecular weight and testing rate on adhesion properties of epoxidized natural rubber-based adhesives using statistical analysis. ENR is prepared by peroxyacetic acid epoxidation of natural rubber (NR) latex. It was chosen as the rubber in order to study the effect of epoxidation of NR on adhesion properties. Unmodified natural rubber-based adhesives have also been investigated by various researchers [15–22].

#### **Experimental**

## Materials

ENR-50 having 50 mol % of epoxidation was used as the elastomer for the preparation of the PSAs. It was supplied by Rubber Research Institute of Malaysia. Gum rosin—the tackifier—is a chemically inert hydrocarbon resin and shows low reactivity property and resistant to alkalis, acids, and moisture. It was freshly supplied by Euro Chemo-Pharma Company (Malaysia). Toluene and polyethylene terephthalate (PET) film were chosen as the solvent and substrate respectively. Toluene was supplied by Brightchem Company, Malaysia and PET was supplied by DSB, OHB, Transparency film, Malaysia. Coating was done using SHEEN Hand Coater made by SHEEN Instruments Ltd. Teddington, MIDDX, England and PET was supplied by DSB, OHB, Transparency film, Malaysia. The effect of molecular weight and testing rate on peel and shear strength of epoxidized natural rubber (ENR 50)-based adhesive was evaluated using Llyod Adhesion Tester operating at different rates of testing. All the materials used in this experiment



## Preparation of Adhesive

2 g each of unmasticated and masticated rubber was shredded into small pieces and dissolved in 15 mL of toluene. The rubber solution was then left overnight to ensure complete dissolution. With constant stirring, 0.2, 0.6 and 1.0 g of pulverized gum rosin that corresponded to 10, 30, 50 parts per hundred parts of rubber (phr) respectively of tackifying resin was slowly put into the rubber solution. The gum rosin was pounded into a fine powder to facilitate easy dissolution in the rubber solution.

## Determination of Molecular Weight

A viscometric method was used to determine the molecular weight of each masticated sample. The intrinsic viscosity ( $[\eta]$ ) was measured according to the method described by Billmeyer [23]. Five rubber samples were obtained by mastication with a two-roll mill. In a two-roll mill, the back roll generally moves faster than the front roll with a speed ratio of 1.1–1.5: 1. The length and diameter of two-roll mill is 15.5 and 7.5 cm respectively. The viscosity-average molecular weight ( $M_v$ ) of the rubber was computed using the Mark–Houwink equation below [24]. Huggins has shown that the reduced viscosity ( $\eta_{sp}/c$ ) is related to the intrinsic viscosity [ $\eta$ ] and concentration as

$$\eta_{\rm sp}/c = [\eta] + k[\eta]^2 c \tag{1}$$

where k is a constant for a given polymer, solvent and temperature. By extrapolating  $\eta_{sp}/c$  to zero concentration,  $[\eta]$  can be obtained. Empirically,  $[\eta]$  is related to the molecular weight of polymer (M) as

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

where k and a are constants. This is called Mark-Houwink equation. In the said system k is  $5.00 \times 10^{-4}$  dL/g and a is 0.67 in toluene [25]. Molecular weight determination was done at 30 °C.

## Measurement

# Peel Strength

The dimensions of PET substrates (base stock and face stock) for the T- and  $90^{\circ}$  Peel Tests, were  $20 \text{ cm} \times 4 \text{ cm}$ . The adhesive was coated from the end of the PET film at a coating area of  $10 \text{ cm} \times 4 \text{ cm}$  for various coating thickness using a SHEEN Hand Coater. The face stock was then placed on the coated PET film (base stock). The sample



was then conditioned at room temperature for 24 h before testing on a Lloyd Adhesion Tester operating at different rates (cm/min). The average peeling force was determined from the three highest peaks recorded from the load-propagation graph. Peel strength is defined as the average load per width of the bond line required to separate progressively a flexible member from a rigid member or another flexible member (ASTM D 907-03, 2004) [26].

### Shear Strength

The dimension of the PET substrate was  $15 \text{ cm} \times 2.5 \text{ cm}$ . A SHEEN Hand Coater was used to coat the adhesive from the end of the substrate with  $5 \text{ cm} \times 2.5 \text{ cm}$  dimension. It was then conditioned at room temperature for 24 h before testing on a Lloyd Adhesion Tester operating at different rates (cm/min). The testing distance was 5 cm which corresponded to the length of the coated area. Shear strength was expressed as the shear force per unit area of testing  $(N/m^2)$ .

# Differential Scanning Calorimetry

The differential scanning calorimetric measurements were measured using a Perkin-Elmer-Pyris DSC-6 differential scanning calorimeter supported by a Perkin-Elmer computer for data acquisition. The samples ( $\leq 5$  mg), sealed under aluminum pans were scanned in the temperature range of  $-40-100^{\circ}$  C. The heating rate was  $10^{\circ}$  C min<sup>-1</sup> under the nitrogen atmosphere with a flow rate of 20 ml min<sup>-1</sup>.

## Thermal Gravimetric Analysis (TGA)

Thermal Gravimetric Analysis (TGA) was carried out using Thermal Gravimetric Analyzer (Perkin Elmer, Pyris 1). The weight of an empty platinum pan was measured

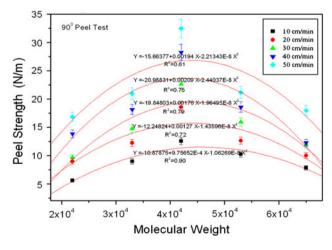


Fig. 1 Variation of peel strength (90° Test) with molecular weight for various testing rate for 10 phr of gum rosin at the coating thickness of 120  $\mu$ m

prior to any measurements to obtain the zero weight of the samples. The samples were loaded in the platinum pan. The reaction was initiated by immersing the sample-loaded pan in a heating furnace and heat was supplied to the furnace at a prescribed rate of heating. The samples ( $\leq 5$  mg) were scanned in the temperature range of  $30\text{--}800^\circ$  C. The heating rate was  $10^\circ$  C min<sup>-1</sup> under the nitrogen atmosphere with a flow rate of 20 ml min<sup>-1</sup>.

### Infrared Spectroscopy (FT-IR)

FTIR spectra were obtained by direct transmittance by means of the KBr pellet technique using a Nicolet Impact 400 FTIR spectrometer equipped with a DTGS detector. The respective adhesive was coated on the KBr pellet and then a spectrum was recorded. Spectra were measured at a spectral resolution of 4 cm<sup>-1</sup>. Good quality FTIR spectra were obtained based on

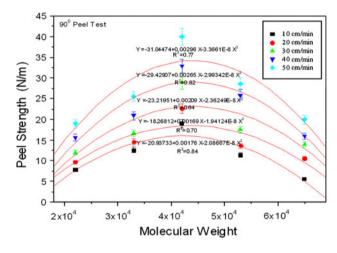


Fig. 2 Variation of peel strength (90° Test) with molecular weight for various testing rate for 30 phr of gum rosin at the coating thickness of 120  $\mu m$ 

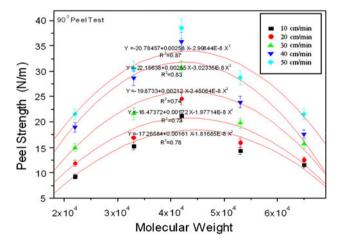


Fig. 3 Variation of peel strength (90° Test) with molecular weight for various testing rate for 50 phr of gum rosin at the coating thickness of 120  $\mu$ m



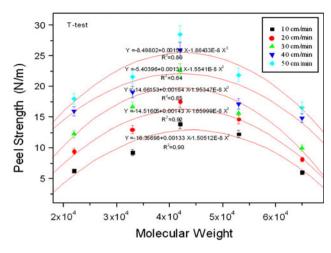


Fig. 4 Variation of peel strength (T Test) with molecular weight for various testing rate for 10 phr of gum rosin at the coating thickness of 120  $\mu$ m

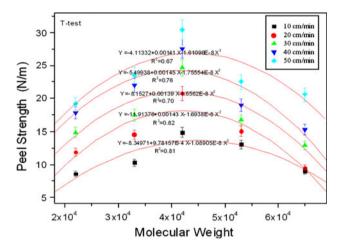


Fig. 5 Variation of peel strength (T Test) with molecular weight for various testing rate for 30 phr of gum rosin at the coating thickness of 120  $\mu$ m

the smoothness of the baseline and resolution. Peak heights of spectra were measured using OMNIC software.

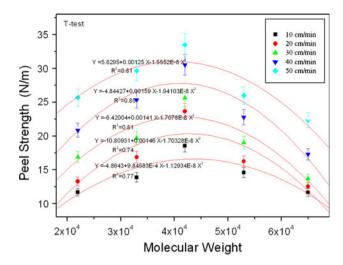
#### **Results and Discussion**

#### Peel Strength and Shear Strength

The dependence of peel and shear strength of adhesive on the molecular weight of rubber and testing rate for various gum rosin contents is discussed below.

# Peel Analysis

Peel analysis was carried out using  $90^{\circ}$  and T test. The effect of molecular weight and testing rate (10, 20, 30, 40 and 50 cm/min) for various loadings of gum rosin at the



**Fig. 6** Variation of peel strength (T Test) with molecular weight for various testing rate for 50 phr of gum rosin at the coating thickness of 120  $\mu$ m

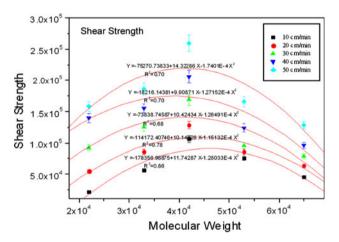


Fig. 7 Variation of shear strength with molecular weight for various testing rate for 10 phr of gum rosin at the coating thickness of 120  $\mu$ m

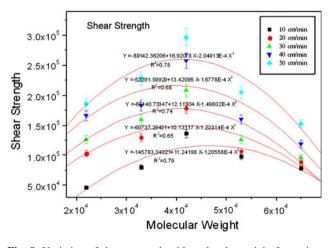


Fig. 8 Variation of shear strength with molecular weight for various testing rate for 30 phr of gum rosin at the coating thickness of 120  $\mu$ m



coating thickness of 120 um are shown in Figs. 1, 2, 3, 4, 5, 6. The measurements of peel strength were done in two, T test and  $90^{\circ}$  peel tests to confirm the variation of peel strength with the various parameters studied. The figures represent the regression model for the shear strength at different testing rate. The R-squared values showed good fitness of the peel strength data to the second-order regression model. The good regression model was obtained as it is evident from the value of R<sup>2</sup> which is in the range of 0.8 and above. Generally, the R-squared values reduced with the increase of the testing rate and this could be due to the reduction of the stability as the testing rate increase. Figures 1, 2, 3, 4, 5, 6 show that the peel strength (T test and 90°) increases with molecular weight of rubber up to  $4.2 \times 10^4$  g/mol for ENR 50, and drops with further increase in molecular weight of rubber for all testing rates. This observation is attributed to the increasing wettability of adhesive up to a maximum value  $4.2 \times 10^4$  g/mol for ENR 50, where maximum wettability is observed. At this adhesive formulation, mechanical interlocking anchorage of the adhesive in pores and irregularities in the adherent are formed [27, 28]. The separation front is blunted, the stresses are minimized and separation is prevented [29], when the adhesive is able to flow like a vielding solid. The adhesive at the optimum molecular weight most probably hardens at high strain levels to become a tough solid; the adhesive layer itself cannot easily be ruptured and hence showing maximum peel strength. By saying optimum molecular weight of  $4.2 \times 10^4$  g/mol means is the molecular weight where peel and shear strength has been observed highest strength. Higher molecular weight sample lowers the wettability of adhesive, probably due to the effect of entanglement as the entangled rubber chain does not flow effectively to produce good wettability on the substrate.

The effect of testing rate on the peel strength (T test and 90°) of adhesives at 120 µm coating thickness are shown in Figs. 1, 2, 3, 4, 5, 6. The plot indicates that peel strength increases with increase in the peeling rate. This observation is attributed to cohesive failure at the low peeling due to the predominantly viscous response. Cohesive fracture is obtained when the crack propagates in the bulk polymer which constitutes the adhesive. Cohesive failure is defined as the inability of an adhesive to resist internal separation. During cohesive failure, the adhesive sticks to both surfaces, but cannot hold them together. However, as the peeling rate is increased, the predominantly elastic response results in adhesive failure which is reflected by the higher peel strength. Adhesive failure occurs at the interface between the adhesive and the substrate. The transition from cohesive to adhesive failure in a peel test is observed in the region of maximum peel force versus molecular weight [30]. Adhesive failure is the failure of the adhesive to stick or bond with the material to be adhered (also known as the substrate or adherent). Cohesive failure is structural failure of the adhesive.

## Shear Analysis

Shear strength is defined as the shear force per unit area of testing. The dependence of shear strength of ENR 50 - based adhesives on molecular weight at 120  $\mu$ m coating thickness is shown in Figs. 7, 8, 9 which represents the regression model for the shear strength at different testing rate. In this paper an attempt is made to obtain a regression equation model at different testing rate showing deviations between the observed values and the estimated values. The R-squared values also showed the good fitness of the shear strength data to the second-order regression model. The good regression model was obtained as it is evident from the value of  $R^2$  which is in the range of 0.8 and above.

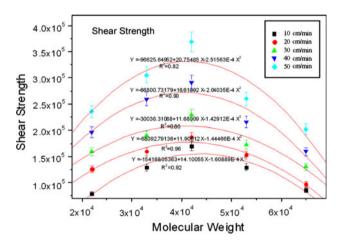


Fig. 9 Variation of shear strength with molecular weight for various testing rate for 50 phr of gum rosin at the coating thickness of 120  $\mu$ m

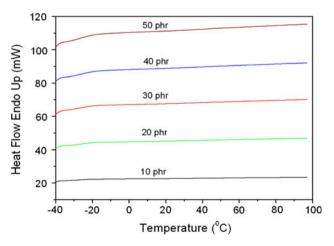


Fig. 10 DSC scan of ENR 50/gum rosin blends at the optimum molecular weight for various resin concentration (phr)



**Table 1** Composition of rubber-resin, values of glass transition temperatures of blend, measured experimentally, and calculated theoretically

Rubber-resin ratio (phr)	Transition temp	erature (°C)
	Experimental	Calculated according to Fox Equation
90:10	-13.20	-13.36
80:20	-7.50	-7.69
70:30	1.88	-2.69
60:40	4.10	1.73
50:50	8.25	5.69

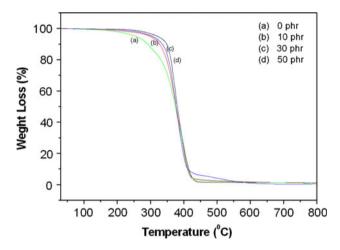


Fig. 11 TGA thermograms of ENR 50/gum rosin blends at optimum molecular weight

Generally, the R-squared values reduced with the increase of the testing rate and this could be attributed to the reduction of the stability as the testing rate increase. Figures 7, 8, 9 also show that shear strength increases with molecular weight up to  $4.2 \times 10^4$  g/mol and drops with higher molecular weight of rubber for coating thicknesses of 120 µm. This observation is attributed to the optimum molecular weight that is necessary to provide for the maximum cohesive and adhesive strength during the shearing action. For lower molecular weight, cohesive failure is observed [27] due to shorter chain length of the rubber molecules. On the contrary, for high molecular weight, poor adhesive strength is observed as a result of poor wettability. Thus, at the optimum molecular weight of  $4.2 \times 10^4$  g/mol, the optimum combination of cohesive and adhesive strength is achieved which contributes to the highest shear strength in the study. From the results, it is obvious that shear strength of adhesive increases with coating thickness. This observation is attributed to increasing amount of rubber component present in the coating layer which enhances the shear resistance of the adhesive.

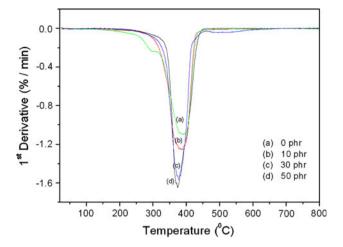


Fig. 12 DTG thermograms of ENR 50/gum rosin blends at optimum molecular weight

**Table 2** Degradation temperatures and ash contents of rubber-resin blend at optimum molecular weight

Rubber-resin (phr)	T <sub>i</sub> (°C)	$T_f$ (°C)	Ash/700 (°C)
0	257.94	419.22	0.21456
10	300.96	424.50	0.60245
30	318.24	430.45	0.94581
50	338.79	431.35	1.27849

As in the case of peel strength, shear strength also increases with increase in the rate of testing. This observation is attributed to the increasing effect of elastic component of the adhesive as the rate of testing is increased. The response of pressure-sensitive adhesive to the shear stress is of a viscoelastic nature. Adhesive hardens at high strain levels to become a tough solid; the adhesive layer itself cannot easily be ruptured [29].

# **Material Properties**

Differential Scanning Calorimetry (DSC)

While dealing with blended materials it is necessary to examine the miscibility between components which affects the physical properties and also on the performance of the materials. The properties of the PSAs are known to be strongly dependent on the glass transition temperature of the adhesive. Figure 10 represent the DSC thermograms at the optimum molecular weight of the ENR 50. The glass transition temperature T<sub>g</sub>, which marks the characteristic transition of the amorphous region of the blend from glassy state to a rubbery state, is the most convenient and popular way of investigating the miscibility or immiscibility of the of polymers and tackifier [31]. Hayashi et al. [32] have



reported that the dynamic mechanical properties and performance of the PSA. However, in the immiscible systems, the mechanical properties of the PSA are not modified [33]. The most widely used criterion for the judgment of the miscibility behavior of the blends is the existence of a single Tg. The presence of one single glass transition temperature in Fig. 10 indicates the miscibility of polymer and tackifier hence the good compatibility of the blend [34, 35]. The presence of only one single peak indicates very clearly that this blend system is a compatible blend. For binary miscible systems, the dependence of T<sub>g</sub> on composition can be presented by well-known Fox equation [36]. The thermal study of pure ENR-50 has been previously reported [37] and is in close agreement with our results. The  $T_g$  of ENR 50 is found to be  $-20~^{\circ}\text{C}$  and taking softening point of gum rosin (76 °C) as glass transition temperature we have calculated the  $T_{\sigma}$  values from Fox equation. The softening point of gum rosin has been previously reported [38] as 76 °C which is agreement with our result. The Fox equation is quite applicable for predicting the Tg of a miscible blend with certain Tg and weight fractions of component polymers.

Fox equation is given in Eq. (3) below:

$$\frac{1}{T_g} = \frac{w_1}{T_{g1}} + \frac{w_2}{T_{g2}} \tag{3}$$

where  $T_{g1}$ ,  $T_{g2}$  and  $T_{g}$  is the glass transition of the rubber, tackifier and mixture and  $w_{1}$  and  $w_{2}$  is the weight fraction of rubber and tackifier respectively. The transition temperatures determined by DSC measurements and

**Fig. 13** FTIR spectra of ENR 50/gum rosin blend at optimum molecular weight

theoretical calculated values according to Fox equation is shown in Table 1. The experimental values and theoretical values by Fox equation are in close agreement for 10, 20, 30, 40 and 50 phr tackifier. This agreement, in turn, proves the validity of the Fox equation for these blends and the fact of complete miscibility in the ENR 50 and gum rosin systems.

## Differential and Thermogravimetric Analysis

Figure 11 shows the thermogravimetric curves of ENR 50/gum rosin blend at optimum molecular weight. It reveals that all blends show similar thermal stabilities up to the region ≈200 °C. In general, major weight losses are observed in the range of  $\approx 200-430$  °C for different blend samples at the optimum molecular weight which may due to the structural decomposition of the natural rubber based adhesive or may associate to high thermal stability of the blend in this region. With the increase in gum rosin concentration at optimum molecular weight, the thermal stability is increased, as indicated by the increase in the onset and offset temperatures. The differential thermal gravimetric (DTG) curves of ENR 50/gum rosin blend at optimum molecular weight is shown in Fig. 12. From the plot, it is evident that the thermograms involve single step degradation. The initial and final degradation step in the case of ENR 50 (0 phr) occurred at 257.94 and 419.22 °C, while the blend with 10, 30 and 50 phr at optimum molecular weight showed higher initial and final degradation temperature respectively, as shown in Table 2.

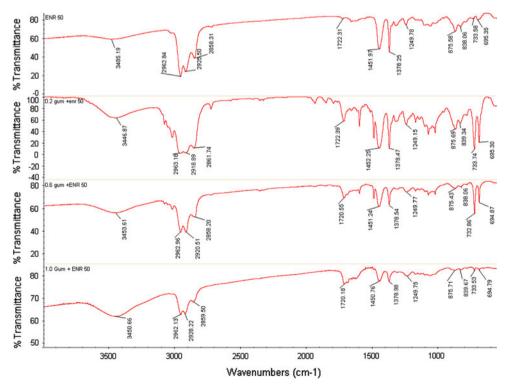




Table 3 Levene statistic of peel (90° test & T test) and shear Strength with molecular weight for various testing rate for 10, 30 and 50 phr of gum rosin at the coating thickness of 120  $\mu$ m

Testing rate (cm/min)	10 phr				30 phr				50 phr			
	Levene statistic	df1	df2	Sig.	Levene statistic	df1	df2	Sig.	Levene statistic	df1	df2	Sig.
Peel strength (90° test)												
10	0.629	4	10	0.65	0.192	4	10	0.98	6.772	4	10	0.01
20	0.928	4	10	0.48	1.759	4	10	0.21	2.048	4	10	0.16
30	0.937	4	10	0.48	3.238	4	10	0.06	12.519	4	10	0.01
40	2.246	4	10	0.13	1.979	4	10	0.17	0.890	4	10	0.50
50	0.988	4	10	0.45	1.621	4	10	0.24	4.124	4	10	0.03
Peel strength (T test)												
10	0.874	4	10	0.51	0.780	4	10	0.56	0.780	4	10	0.56
20	1.653	4	10	0.23	0.351	4	10	0.84	0.351	4	10	0.84
30	2.345	4	10	0.12	1.411	4	10	0.30	1.411	4	10	0.30
40	3.292	4	10	0.05	3.872	4	10	0.04	3.872	4	10	0.04
50	1.259	4	10	0.35	1.347	4	10	0.32	1.347	4	10	0.32
Shear strength												
10	0.821	4	10	0.54	2.187	4	10	0.14	2.063	4	10	0.16
20	1.077	4	10	0.42	1.192	4	10	0.37	4.061	4	10	0.03
30	1.372	4	10	0.31	2.376	4	10	0.12	1.515	4	10	0.27
40	1.986	4	10	0.17	0.836	4	10	0.53	2.018	4	10	0.17
50	2.091	4	10	0.16	1.827	4	10	0.20	1.482	4	10	0.28

#### FTIR Analysis

Figure 13 shows the FTIR spectra of ENR 50/gum rosin blend at optimum molecular weight. At 10, 30 and 50 phr of tackifier contents, epoxy group of ENR 50 opens up and undergoes esterfication reaction as is evident from FTIR spectroscopy while for 0 phr tackifier loading, the esterfication is not prominent. This clearly shows that the presence of carbonyl group peak frequency at 1,720.18–1,722.39 cm<sup>-1</sup> implies that the sample has undergone esterfication reaction. Broad peak in the region 3,400–3,200 cm<sup>-1</sup> is associated with the presence of hydroxyl groups. The hydroxyl group of the rubber shows red shift in its peak frequency with the increase in the gum rosin concentration, and this can be attributed to the physical interaction between the hydroxyl group and epoxy group of the resin. Peak at 2,963.18-2,962.13 and 2,928.22-2,918.89 cm<sup>-1</sup> are due to asymmetric stretching vibration of methyl (-CH<sub>3</sub>) and methylene (=CH<sub>2</sub>) group respectively. Peak at 2,861.74–2,858.31 cm<sup>-1</sup> are attributed to symmetric stretching vibration of methylene (-CH<sub>2</sub>) group. Aromatic combination band appeared at 1,660–2,200 cm<sup>-1</sup>. Absorption bands at 1,452.25–1,450.76 cm<sup>-1</sup> and around 1,378.25 cm<sup>-1</sup> are characteristic peaks of -C-H bending vibrations of methylene (-CH<sub>2</sub>) group. The C-O-C asymmetric stretching of epoxides showed strong absorption cross section in the region of 838.67 cm<sup>-1</sup>. Peaks at 1,615–1,600 cm<sup>-1</sup> clearly indicates aromatic ring pattern. Absorption peak around 875 cm<sup>-1</sup> and at 732.86-733.74 cm<sup>-1</sup> clearly showed meta substituted benzene and ortho substituted benzene respectively. Peaks at  $\sim 1,250 \text{ cm}^{-1}$  again confirm the presence of epoxy moiety.

#### Statistical Analysis

A statistical approach was used to compare the effect of molecular weight at different testing rate on the peel strength and shear strength of ENR 50 -based adhesives. One-way ANOVA was carried out to produce a one-way analysis of variance at respective testing rate to determine the influence of molecular weight on the peel and shear strength. Appendix Tables 4, 5, 6 show the corresponding ANOVA tables for peel test, t test and shear test as discussed in Figs. 1, 2, 3, 4, 5, 6, 7, 8, 9, respectively. Results show that different molecular weight of ENR 50-based adhesives have significance effect (Sig. < 0.05) on peel strength and shear strength at different testing rate of peel test, t test and shear test whereby the peel strength and shear strength change significantly with the molecular weight. This is in agreement with our previous results and others [39, 40].

Levene statistic is carried out for each testing rate of peel test, *t* test and shear test to determine the homogeneity of group variance as given in Table 3. The significance value obtained that is greater than 0.05 indicates equal group variance is assumed for peel test, *t* test and shear test. Post hoc multiple comparisons provide a comparison matrix that is showing the statistically comparison between group means at 95 % of confidence level. Post hoc test is selected based on



the Levene statistic whereby the test is selected based on the homogeneity of group variance. Tukey test is used for the post hoc multiple comparisons to determine which means differ as the Levene's test shows equal group variance (Sig. > 0.05) whereas Tamhane's T2 multiple comparisons is used for the group with significant difference (Sig. < 0.05) in group variance. Appendix Tables 7, 8, 9, 10, 11, 12, 13, 14, 15 show the post hoc multiple comparisons for peel-test, T test and shear test in terms of molecular weight at different testing rate. Generally, molecular weights show significant difference in terms of peel strength and shear strength among them at different testing rate. Therefore, it can be concluded that the properties of the ENR 50 -based adhesives change significantly with the molecular weight.

#### **Conclusions**

From this study, the following conclusions can be drawn.

 Both peel strength and shear strength increases with molecular weight of rubber up to an optimum molecular weight of 4.2 × 10<sup>4</sup> g/mol of ENR 50. For peel strength, this observation is attributed to the combined effects of wettability and mechanical strength of rubber at the optimum molecular weight. However, in the case of

- shear strength, it is ascribed to the optimum cohesive and adhesive strength during the shearing action.
- 2. Peel strength of ENR 50-based adhesives increases with increasing rate of testing, an observation which is attributed to the viscoelastic response of the adhesive. At low rates of testing, the response is predominantly viscous and cohesive failure occurs whereas at higher rates of testing, the response becomes predominantly elastic which results in adhesive failure. Shear strength of the rubber-based adhesive also increases with increasing rate of testing which is attributed to the increasing effect of elastic component of the adhesive as the rate of testing is increased.
- 3. Thermal study, FTIR study confirms the miscibility of tackifier and ENR 50 in the adhesive system.
- 4. Statistical analysis that use ANOVA and post hoc multiple comparisons show that molecular weights have significant effect on the peel strength and shear strength of ENR 50 at different testing rate.

**Acknowledgments** The authors acknowledge the research grant (FRGS) provided by Universiti Sains Malaysia that has resulted in this article.

## **Appendix**

See Tables 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15.

Table 4 ANOVA table for peel strength ( $90^{\circ}$  Test) with molecular weight for various testing rate for 10, 30 and 50 phr of gum rosin at the coating thickness of 120  $\mu$ m

Testing rate	10 phr					30 phr					50 phr				
(cm/min)	Sum of squares	df	Mean square	F	Sig	Sum of squares	df	Mean squares	F	Sig	Sum of squares	df	Mean square	F	Sig
10															
Between groups	74.52	4	18.63	142.68	.00	315.14	4	78.79	777.3	.00	296.86	4	74.21	179.27	.00
Within groups	1.30	10	0.13			1.01	10	0.10			4.14	10	0.41		
Total	75.83	14				316.15	14				301.00	14			
20															
Between groups	173.35	4	43.34	881.82	.00	318.39	4	79.60	156.5	.00	314.87	4	78.71	1,420.9	.00
Within groups	0.49	10	0.05			5.09	10	0.51			.55	10	0.05		
Total	173.84	14				323.48	14				315.42	14			
30															
Between groups	279.71	4	69.93	331.41	.00	527.97	4	131.9	372.5	.00	532.28	4	133.07	68.50	.00
Within groups	2.11	10	0.21			3.54	10	0.354			19.42	10	1.94		
Total	281.83	14				531.52	14				551.70	14			
40															
Between groups	453.28	4	113.32	826.43	.00	569.07	4	142.26	233.7	.00	650.15	4	162.53	279.97	.00
Within groups	1.37	10	0.137			6.087	10	0.61			5.80	10	0.58		
Total	454.6	14				575.15	14				655.96	14			
50															
Between groups	478.24	4	119.56	1,733.5	.00	856.67	4	214.16	700.4	.00	647.40	4	161.85	208.93	.00
Within groups	0.69	10	.069			3.058	10	0.30			7.74	10	0.77		
Total	478.9	14				859.7	14				655.14	14			



Table 5 ANOVA table for peel strength (T Test) with molecular weight for various testing rate for 10, 30 and 50 phr of gum rosin at the coating thickness of 120 µm

Testing rate	10 phr					30 phr					50 phr				
(cm/min)	Sum of squares	df	Mean square	F	Sig	Sum of squares	df	Mean squares	ц	Sig	Sum of squares	df	Mean square	H	Sig
10															
Between groups	127.39	4	31.85	132.00	8.	82.62	4	20.65	226.614	90.	116.32	4	29.08	110.09	9.
Within groups	2.41	10	0.24			0.91	10	60.0			2.64	10	0.26		
Total	129.81	14				83.53	14				118.96	14			
20															
Between groups	156.00	4	39.00	168.35	8.	211.49	4	52.87	57.95	90.	240.33	4	80.09	131.77	9.
Within groups	2.31	10	0.23			9.12	10	.91			4.56	10	0.45		
Total	158.32	14				220.61	14				244.89	14			
30															
Between groups	285.93	4	71.48	240.19	00.	286.28	4	71.57	362.261	00.	239.49	4	59.87	98.37	00.
Within groups	2.97	10	0.29			1.97	10	0.19			6.087	10	0.60		
Total	288.91	14				288.25	14				245.58	14			
40															
Between groups	219.06	4	54.76	206.82	00.	294.66	4	73.66	158.053	00.	281.54	4	70.38	74.20	00.
Within groups	2.64	10	0.26			4.66	10	0.46			9.486	10	0.94		
Total	221.70	14				299.32	14				291.03	14			
50															
Between groups	224.53	4	56.13	118.70	00.	216.35	4	54.08	145.554	00.	279.34	4	69.83	68.24	00.
Within groups	4.72	10	0.47			3.71	10	0.37			10.23	10	1.02		
Total						220.06	14				289.58	14			



Table 6 ANOVA table for shear strength with molecular weight for various testing rate for 10, 30 and 50 phr of gum rosin at the coating thickness of 120 µm

Testing rate(cm/min) 10 phr	10 phr					30 phr					50 phr				
	Sum of squares	df	Mean square	ഥ	Sig	Sum of squares	df	Mean squares	Ħ	Sig	Sum of squares	df	Mean square	묘	Sig
01															
Between groups	$1.20 \times 10^{10}$	4	$3.01 \times 10^9$	573.07	00:	$1.30\times10^{11}$	4	$3.25 \times 10^9$	159.32	00.	$1.62\times10^{10}$	4	$4.06 \times 10^9$	313.0	00:
Within groups	$5.25 \times 10^7$	10	$5.25 \times 10^6$			$2.04 \times 10^8$	10	$2.04 \times 10^7$			$1.29 \times 10^{8}$	10	$1.29 \times 10^7$		
Total	$1.20 \times 10^{10}$	14				$1.32\times10^{10}$	14				$1.63 \times 10^{10}$	14			
20															
Between groups	$1.06 \times 10^{10}$	4	$2.66 \times 10^{9}$	158.11	00:	$1.42 \times 10^{9}$	4	$3.56\times10^9$	139.83	00.	$1.41 \times 10^{10}$	4	$3.52 \times 10^9$	362.3	00:
Within groups	$1.68 \times 10^{9}$	10	$1.68 \times 10^7$			$2.55 \times 10^8$	10	$2.55 \times 10^7$			$9.73 \times 10^7$	10	$9.73 \times 10^{6}$		
Total	$1.08 \times 10^{10}$	14				$1.45 \times 10^{10}$	14				$1.42 \times 10^{10}$	14			
30															
Between groups	$1.47 \times 10^{10}$	4	$3.68 \times 10^9$	185.51	00:	$2.42\times10^{10}$	4	$6.06\times10^{9}$	461.90	00.	$1.31\times10^{10}$	4	$3.28 \times 10^9$	74.97	00:
Within groups	$1.98 \times 10^{9}$	10	$1.98 \times 10^7$			$1.31\times10^8$	10	$1.31 \times 10^7$			$4.37\times10^8$	10	$4.37 \times 10^7$		
Total	$1.49 \times 10^{10}$	14				$2.44 \times 10^{10}$	14				$1.35 \times 10^{10}$	14			
40															
Between groups	$1.81 \times 10^{10}$	4	$4.54 \times 10^9$	224.57	00.	$3.54 \times 10^{10}$	4	$8.86 \times 10^9$	240.93	00.	$3.45 \times 10^{10}$	4	$8.62 \times 10^9$	403.3	00.
Within groups	$2.02 \times 10^9$	10	$2.02 \times 10^7$			$3.68 \times 10^8$	10	$3.68 \times 10^7$			$2.13\times10^8$	10	$2.13 \times 10^7$		
Total	$1.83 \times 10^{10}$	14				$3.58\times10^{10}$	14				$3.47 \times 10^{8}$	14			
50															
Between groups	$2.56\times10^{10}$	4	$6.41 \times 10^9$	183.28	00:	$3.57\times10^{10}$	4	$8.93 \times 10^9$	598.10	00.	$4.99 \times 10^{10}$	4	$1.24\times10^{10}$	262.7	00.
Within groups	$3.49 \times 10^{8}$	10	$3.49 \times 10^{7}$			$1.49 \times 10^{8}$	10	$1.49 \times 10^{7}$			$4.75 \times 10^{10}$	10	$4.75 \times 10^7$		
Total	$2.60 \times 10^{10}$	14				$3.59\times10^{10}$	14				$5.04\times10^{10}$	14			



Table 7 Tukey multiple comparisons of peel strength (90° Test) with molecular weight for various testing rate for 10 phr of gum rosin at the coating thickness of 120  $\mu$ m

Peel (10	cm/min)		Peel (20	cm/min)		Peel (30	cm/min)		Peel (40	cm/min)		Peel (50	cm/min)	
MW(I)	MW(J)	Sig.												
22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.000		53,000	.000		53,000	.000		53,000	.000
	65,000	.000		65,000	.031		65,000	.000		65,000	.008		65,000	.001
33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.006		53,000	.395		53,000	.065		53,000	.939		53,000	.022
	65,000	.012		65,000	.000		65,000	.001		65,000	.000		65,000	.000
42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000
	33,000	.000		33,000	.000		33,000	.000		33,000	.000		33,000	.000
	53,000	.004		53,000	.000		53,000	.000		53,000	.000		53,000	.000
	65,000	.000		65,000	.000		65,000	.000		65,000	.000		65,000	.000
53,000	22,000	.000	53,000	22,000	.000	53,000	22,000	.000	53,000	22,000	.000	53,000	22,000	.000
	33,000	.006		33,000	.395		33,000	.065		33,000	.939		33,000	.022
	42,000	.004		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	65,000	.000		65,000	.000		65,000	.000		65,000	.000		65,000	.000
65,000	22,000	.000	65,000	22,000	.031	65,000	22,000	.000	65,000	22,000	.008	65,000	22,000	.001
	33,000	.012		33,000	.000		33,000	.001		33,000	.000		33,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.000		53,000	.000		53,000	.000		53,000	.000

**Table 8** Tukey multiple comparisons of peel strength (90° Test) with molecular weight for various testing rate for 30 phr of gum rosin at the coating thickness of 120  $\mu$ m

Peel (10	cm/min)		Peel (20	cm/min)		Peel (30	cm/min)		Peel (40	cm/min)		Peel (50	cm/min)	
MW(I)	MW(J)	Sig.												
22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.000		53,000	.000		53,000	.000		53,000	.000
	65,000	.000		65,000	.286		65,000	.001		65,000	.640		65,000	.876
33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.013		53,000	.945		53,000	.031		53,000	.000		53,000	.000
	65,000	.000		65,000	.001		65,000	.020		65,000	.000		65,000	.000
42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000
	33,000	.000		33,000	.000		33,000	.000		33,000	.000		33,000	.000
	53,000	.000		53,000	.000		53,000	.000		53,000	.000		53,000	.000
	65,000	.000		65,000	.000		65,000	.000		65,000	.000		65,000	.000
53,000	22,000	.000	53,000	22,000	.000	53,000	22,000	.000	53,000	22,000	.000	53,000	22,000	.000
	33,000	.013		33,000	.945		33,000	.031		33,000	.000		33,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	65,000	.000		65,000	.002		65,000	.000		65,000	.000		65,000	.000
65,000	22,000	.000	65,000	22,000	.286	65,000	22,000	.001	65,000	22,000	.640	65,000	22,000	.876
	33,000	.000		33,000	.001		33,000	.020		33,000	.000		33,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.002		53,000	.000		53,000	.000		53,000	.000



Table 9 Tukey test (20 and 40 cm/min) and Tamhane's T2 (10, 30 and 50 cm/min) multiple comparisons of peel strength (90° Test) with molecular weight for various testing rate for 30 phr of gum rosin at the coating thickness of 120  $\mu$ m

Peel (10	cm/min)		Peel (20	cm/min)		Peel (30	cm/min)		Peel (40	cm/min)		Peel (50	cm/min)	
MW(I)	MW(J)	Sig.												
22,000	33,000	.016	22,000	33,000	.000	22,000	33,000	.317	22,000	33,000	.000	22,000	33,000	.002
	42,000	.008		42,000	.000		42,000	.003		42,000	.000		42,000	.001
	53,000	.004		53,000	.000		53,000	.000		53,000	.000		53,000	.122
	65,000	.052		65,000	.002		65,000	.093		65,000	.740		65,000	.998
33,000	22,000	.016	33,000	22,000	.000	33,000	22,000	.317	33,000	22,000	.000	33,000	22,000	.002
	42,000	.104		42,000	.000		42,000	.338		42,000	.000		42,000	.031
	53,000	.344		53,000	.002		53,000	.849		53,000	.000		53,000	.480
	65,000	.002		65,000	.000		65,000	.383		65,000	.000		65,000	.001
42,000	22,000	.008	42,000	22,000	.000	42,000	22,000	.003	42,000	22,000	.000	42,000	22,000	.001
	33,000	.104		33,000	.000		33,000	.338		33,000	.000		33,000	.031
	53,000	.063		53,000	.000		53,000	.011		53,000	.000		53,000	.078
	65,000	.038		65,000	.000		65,000	.007		65,000	.000		65,000	.003
53,000	22,000	.004	53,000	22,000	.000	53,000	22,000	.000	53,000	22,000	.000	53,000	22,000	.122
	33,000	.344		33,000	.002		33,000	.849		33,000	.000		33,000	.480
	42,000	.063		42,000	.000		42,000	.011		42,000	.000		42,000	.078
	65,000	.004		65,000	.000		65,000	.000		65,000	.000		65,000	.105
65,000	22,000	.052	65,000	22,000	.002	65,000	22,000	.093	65,000	22,000	.740	65,000	22,000	.998
	33,000	.002		33,000	.000		33,000	.383		33,000	.000		33,000	.001
	42,000	.038		42,000	.000		42,000	.007		42,000	.000		42,000	.003
	53,000	.004		53,000	.000		53,000	.000		53,000	.000		53,000	.105

**Table 10** Tukey multiple comparison of peel strength (T Test) with molecular weight for various testing rate for 10 phr of gum rosin at the coating thickness of 120  $\mu$ m

T test (1	0 cm/min)		T test (2	0 cm/min)		T test (3	0 cm/min)		T test (4	0 cm/min)		T test (5	0 cm/min)	
MW(I)	MW(J)	Sig.												
22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.006
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.000		53,000	.000		53,000	.012		53,000	.000
	65,000	.996		65,000	.253		65,000	.007		65,000	.661		65,000	.034
33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.006
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.010		53,000	.574		53,000	.019		53,000	.206
	65,000	.000		65,000	.000		65,000	.000		65,000	.000		65,000	.000
42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000
	33,000	.000		33,000	.000		33,000	.000		33,000	.000		33,000	.000
	53,000	.708		53,000	.000		53,000	.000		53,000	.000		53,000	.000
	65,000	.000		65,000	.000		65,000	.000		65,000	.000		65,000	.000
53,000	22,000	.000	53,000	22,000	.000	53,000	22,000	.000	53,000	22,000	.012	53,000	22,000	.000
	33,000	.000		33,000	.010		33,000	.574		33,000	.019		33,000	.206
	42,000	.708		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	65,000	.000		65,000	.000		65,000	.000		65,000	.002		65,000	.000
65,000	22,000	.996	65,000	22,000	.253	65,000	22,000	.007	65,000	22,000	.661	65,000	22,000	.034
	33,000	.000		33,000	.000		33,000	.000		33,000	.000		33,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.000		53,000	.000		53,000	.002		53,000	.000



**Table 11** Tukey (10, 20, 30 and 50) and Tamhane's T2 (40) multiple comparisons of peel strength (T Test) with molecular weight for various testing rate for 30 phr of gum rosin at the coating thickness of 120  $\mu$ m

T test (1	0 cm/min)		T test (2	0 cm/min)		T test (3	0 cm/min)		T test (4	0 cm/min)		T test (5	0 cm/min)	
MW(I)	MW(J)	Sig.												
22,000	33,000	.000	22,000	33,000	.005	22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.028		42,000	.000
	53,000	.000		53,000	.011		53,000	.001		53,000	.812		53,000	.002
	65,000	.367		65,000	.056		65,000	.001		65,000	.733		65,000	.043
33,000	22,000	.000	33,000	22,000	.005	33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.000
	42,000	.000		42,000	.001		42,000	.000		42,000	.063		42,000	.000
	53,000	.000		53,000	.987		53,000	.948		53,000	.032		53,000	.008
	65,000	.001		65,000	.000		65,000	.000		65,000	.078		65,000	.001
42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.028	42,000	22,000	.000
	33,000	.000		33,000	.001		33,000	.000		33,000	.063		33,000	.000
	53,000	.001		53,000	.000		53,000	.000		53,000	.008		53,000	.000
	65,000	.000		65,000	.000		65,000	.000		65,000	.001		65,000	.000
53,000	22,000	.000	53,000	22,000	.011	53,000	22,000	.001	53,000	22,000	.812	53,000	22,000	.002
	33,000	.000		33,000	.987		33,000	.948		33,000	.032		33,000	.008
	42,000	.001		42,000	.000		42,000	.000		42,000	.008		42,000	.000
	65,000	.000		65,000	.000		65,000	.000		65,000	.376		65,000	.360
65,000	22,000	.367	65,000	22,000	.056	65,000	22,000	.001	65,000	22,000	.733	65,000	22,000	.043
	33,000	.001		33,000	.000		33,000	.000		33,000	.078		33,000	.001
	42,000	.000		42,000	.000		42,000	.000		42,000	.001		42,000	.000
	53,000	.000		53,000	.000		53,000	.000		53,000	.376		53,000	.360

**Table 12** Tukey multiple comparisons of peel strength (T Test) with molecular weight for various testing rate for 50 phr of gum rosin at the coating thickness of 120  $\mu$ m

<i>T</i> test (1	0 cm/min)		T test (2	0 cm/min)		T test (3	0 cm/min)		T test (4	0 cm/min)		T test (5	0 cm/min)	
MW(I)	MW(J)	Sig.	MW(I)	MW(J)	Sig.	MW(I)	MW(J)	Sig.	MW(I)	MW(J)	Sig.	MW(I)	MW(J)	Sig.
22,000	33,000	.004	22,000	33,000	.001	44,000	22,000	.001	22,000	33,000	.000	22,000	33,000	.004
	42,000	.000		42,000	.000			.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.001			.164		53,000	.521		53,000	1.000
	65,000	.919		65,000	.263			.022		65,000	.171		65,000	.001
33,000	22,000	.004	33,000	22,000	.001	54,000	33,000	.001	33,000	22,000	.000	33,000	22,000	.004
	42,000	.000		42,000	.000			.000		42,000	.003		42,000	.008
	53,000	.254		53,000	.999			.022		53,000	.001		53,000	.004
	65,000	.001		65,000	.000			.000		65,000	.000		65,000	.000
42,000	22,000	.000	42,000	22,000	.000	65,000	42,000	.000	42,000	22,000	.000	42,000	22,000	.000
	33,000	.000		33,000	.000			.000		33,000	.003		33,000	.008
	53,000	.000		53,000	.000			.000		53,000	.000		53,000	.000
	65,000	.000		65,000	.000			.000		65,000	.000		65,000	.000
53,000	22,000	.000	53,000	22,000	.001	76,000	53,000	.164	53,000	22,000	.521	53,000	22,000	1.000
	33,000	.254		33,000	.999			.022		33,000	.001		33,000	.004
	42,000	.000		42,000	.000			.000		42,000	.000		42,000	.000
	65,000	.000		65,000	.000			.001		65,000	.014		65,000	.001
65,000	22,000	.919	65,000	22,000	.263	84,000	65,000	.022	65,000	22,000	.171	65,000	22,000	.001
	33,000	.001		33,000	.000			.000		33,000	.000		33,000	.000
	42,000	.000		42,000	.000			.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.000			.001		53,000	.014		53,000	.001



Table 13 Tukey multiple comparisons of shear strength with molecular weight for various testing rate for 10 phr of gum rosin at the coating thickness of 120  $\mu$ m

T test (10 cm/min)			T test (20 cm/min)			T test (30 cm/min)			T test (40 cm/min)			T test (50 cm/min)		
MW(I)	MW(J)	Sig.												
22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.000	22,000	33,000	.003	22,000	33,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.000		53,000	.997		53,000	.052		53,000	.024
	65,000	.000		65,000	.389		65,000	.036		65,000	.000		65,000	.001
33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.000	33,000	22,000	.003	33,000	22,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.611		53,000	.000		53,000	.000		53,000	.105
	65,000	.001		65,000	.001		65,000	.000		65,000	.000		65,000	.000
42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000
	33,000	.000		33,000	.000		33,000	.000		33,000	.000		33,000	.000
	53,000	.000		53,000	.000		53,000	.000		53,000	.000		53,000	.000
	65,000	.000		65,000	.000		65,000	.000		65,000	.000		65,000	.000
53,000	22,000	.000	53,000	22,000	.000	53,000	22,000	.997	53,000	22,000	.052	53,000	22,000	.024
	33,000	.000		33,000	.611		33,000	.000		33,000	.000		33,000	.105
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	65,000	.000		65,000	.000		65,000	.060		65,000	.000		65,000	.000
65,000	22,000	.000	65,000	22,000	.389	65,000	22,000	.036	65,000	22,000	.000	65,000	22,000	.001
	33,000	.001		33,000	.001		33,000	.000		33,000	.000		33,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.000		53,000	.060		53,000	.000		53,000	.000

Table 14 Tukey multiple comparisons of shear strength with molecular weight for various testing rate for 30 phr of gum rosin at the coating thickness of  $120~\mu m$ 

T test (10 cm/min)			T test (20 cm/min)			T test (30 cm/min)			T test (40 cm/min)			T test (50 cm/min)		
MW(I)	MW(J)	Sig.												
22,000	33,000	.000	22,000	33,000	.005	22,000	33,000	.000	22,000	33,000	.010	22,000	33,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.380		53,000	.964		53,000	.432		53,000	.000
	65,000	.000		65,000	.009		65,000	.000		65,000	.000		65,000	.000
33,000	22,000	.000	33,000	22,000	.005	33,000	22,000	.000	33,000	22,000	.010	33,000	22,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.002		53,000	.089		53,000	.000		53,000	.001		53,000	.002
	65,000	.836		65,000	.000		65,000	.000		65,000	.000		65,000	.000
42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000	42,000	22,000	.000
	33,000	.000		33,000	.000		33,000	.000		33,000	.000		33,000	.000
	53,000	.000		53,000	.000		53,000	.000		53,000	.000		53,000	.000
	65,000	.000		65,000	.000		65,000	.000		65,000	.000		65,000	.000
53,000	22,000	.000	53,000	22,000	.380	53,000	22,000	.964	53,000	22,000	.432	53,000	22,000	.000
	33,000	.002		33,000	.089		33,000	.000		33,000	.001		33,000	.002
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	65,000	.000		65,000	.001		65,000	.000		65,000	.000		65,000	.000
65,000	22,000	.000	65,000	22,000	.009	65,000	22,000	.000	65,000	22,000	.000	65,000	22,000	.000
	33,000	.836		33,000	.000		33,000	.000		33,000	.000		33,000	.000
	42,000	.000		42,000	.000		42,000	.000		42,000	.000		42,000	.000
	53,000	.000		53,000	.001		53,000	.000		53,000	.000		53,000	.000



T test (10 cm/min) T test (20 cm/min) T test (30 cm/min) T test (40 cm/min) T test (50 cm/min) MW(J) MW(I) MW(I) MW(J) MW(I) MW(J) MW(I) MW(J) MW(I) MW(J) Sig. Sig. Sig. Sig. Sig. 22,000 33,000 .000 22,000 33,000 .001 22,000 33,000 .001 22,000 33,000 .000 22,000 .000 33,000 42,000 .000 42,000 .013 42,000 .000 42,000 .000 42,000 .000 53,000 .000 53,000 .001 53,000 .024 53,000 .002 53,000 .195 65,000 .021 65,000 .006 65,000 .004 65,000 .000 65,000 .001 33,000 22,000 33,000 22,000 .001 33,000 22,000 .001 33,000 22,000 33,000 22,000 .000 .000 .00042,000 .000 42,000 .108 42,000 .001 42,000 .000 42,000 .000 53,000 53,000 .039 53,000 .373 53,000 53,000 .000 .912 .000 .000 .000 .000 65,000 65,000 .004 .000 65,000 65,000 65,000 42,000 22,000 .000 42,000 22,000 .013 42,000 22,000 .000 42,000 22,000 000. 42,000 22,000 000 33,000 .000 33,000 .108 33,000 .001 33,000 .000 33,000 .000 53,000 53,000 53,000 53,000 .000 .058 .000 53,000 000..000 65,000 .000 65,000 .001 65,000 .000 65,000 .000 65,000 .000 53,000 22,000 22,000 .001 53,000 .024 53,000 22,000 .002 .000 53,000 22,000 53,000 22,000 .195 33,000 .912 33,000 .039 33,000 .373 33,000 .000 33,000 .000 .058 .000 42,000 .000 42,000 42,000 .000 42,000 42,000 .000 .000 65,000 65,000 .003 65,000 65,000 .000 65,000 .000 .000 65,000 22,000 .021 65,000 22,000 .006 65,000 22,000 .004 65,000 22,000 .000 65,000 22,000 .001 33,000 33,000 .004 33,000 .000 33,000 .000 33,000 .000 .000 42,000 .000 42,000 .001 42,000 .000 42,000 .000 42,000 .000 .000 53,000 .000 53,000 .000 53,000 .003 53,000 53,000 .000

Table 15 Tukey (10, 30, 40 and 50 cm/min) and Tamhane's T2 (20 cm/min) multiple comparisons of shear strength with molecular weight for various testing rate for 30 phr of gum rosin at the coating thickness of 120 μm

# References

- Aubrey DW, Sherriff M (1980) Peel adhesion and viscoelasticity of rubber–resin blends. J Polym Sci Pol Chem 18:2597–2608
- Sherriff M, Knibbs RW, Langley PG (1973) Mechanism for the action of tackifying resins in pressure-sensitive adhesives. J Appl Polym Sci 17:3423–3438
- Hata T, Tsukatani T, Mizumachi H (1994) Holding power (tb) and sliding friction coefficient of pressure sensitive adhesives. J Adhes Soc Jpn 30:307–312
- Khan I, Poh BT (2012) Material properties and influence of molecular weight and testing rate on adhesion properties of epoxidized natural rubber-based adhesives. J Polym Environ 20:132–141
- Khan I, Poh BT (2011) Effect of molecular weight and testing rate on peel and shear strength of epoxidized natural rubber (ENR 50)-based adhesives. J Appl Polym Sci 120:2641–2647
- Khan I, Poh BT (2011) Effect of molecular weight and testing rate on adhesion property of pressure-sensitive adhesives prepared from epoxidized natural rubber. Mater Des 32:2513–2519
- Lanrock A (1985) Adhesive technology hand book. Noyes Publishing, Park Ridge
- Allen KW (1987) A review of contemporary views of theories of adhesion. J Adhes 21:261–277
- Khan I, Poh BT (2011) Natural rubber-based pressure-sensitive adhesives: a review. J Polym Environ 19:793–811
- Dahlquist CA (1989) Pressure sensitive adhesive technology, vol Chapter 20, 2nd edn. Van Nostrand Reinhold, New York
- Kim H-J, Mizumachi H (1995) Miscibility and peel strength of acrylic pressure-sensitive adhesives: acrylic copolymer–tackifier resin systems. J Appl Polym Sci 56:201–209

- Whitehouse RS, Counsell PJC, Lewis G (1976) Composition of rubber/resin adhesive films: 1. Surface composition as determined by ATR spectroscopy. Polymer 17:699–704
- Pal K, Pal SK, Das CK, Kim JK (2011) Effect of fillers on morphological properties and wear characteristics of XNBR/NR blends. J Appl Polym Sci 120:710–718
- Saito T, Klinklai W, Kawahara S (2007) Characterization of epoxidized natural rubber by 2D NMR spectroscopy. Polymer 48:750–757
- Pal K, Rajasekar R, Kang DJ, Zhang ZX, Pal SK, Das CK, Kim JK (2010) Effect of fillers on natural rubber/high styrene rubber blends with nano silica: morphology and wear. Mater Des 31:677–686
- Vatansever N, Polat S (2010) Effect of zinc oxide type on ageing properties of styrene butadiene rubber compounds. Mater Des 31:1533–1539
- 17. Pal K, Rajasekar R, Kang DJ, Zhang ZX, Pal SK, Das CK, Kim JK (2010) Influence of carbon blacks on butadiene rubber/high styrene rubber/natural rubber with nanosilica: morphology and wear. Mater Des 31:1156–1164
- Bitinis N, Verdejo R, Cassagnau P, Lopez-Manchado MA (2011) Structure and properties of polylactide/natural rubber blends. Mater Chem Phys 129:823–831
- Vidal A, Haidar B (2007) Filled elastomers: characteristics and properties of interfaces and interphases, and their role in reinforcement processes. Soft Mater 5:155–167
- Khan I, Poh BT (2010) Effect of silica on viscosity, tack, and shear strength of epoxidized natural rubber-based pressure-sensitive adhesives in the presence of coumarone-indene resin. J Appl Polym Sci 118:3439–3444
- Khan I, Poh BT, Badriah CM (2011) Effect of sodium sulfate on the viscosity, tack, and adhesion properties of SMR 10-based pressure-sensitive adhesive. J Elastom Plast 43:85–95



- Khan I, Poh BT (2010) The effect of silica on the peel adhesion of epoxidized natural rubber-based adhesive containing coumarone- indene resin. Polym Plast Technol 49:1356–1360
- Billmeyer FW (1984) Textbook of polymer science, 3rd edn. Wiley, New York, p 208
- 24. Bateman L (1963) The chemistry and physics of rubber-like substances. Maclaren Ed, London, p 149
- Brandrup J, EHI, Grulke EA (2005) Soultion properties, section VII. In: Polymer handbook, 4th edn. Wiley, New York
- ASTM D907-03 (2004) Terminology of adhesives. Annual book of ASTM standards, vol 15.06. American Society for Testing and Materials, Philadelphia
- Satas D (1982) Handbook of pressure sensitive adhesive technology. Van Nostrand Reinhold, New York, p 54
- Lee LH (1991) Adhesive bonding, vol 19. Plenum Press, New York
- Skeist I (1990) Handbook of adhesives, 3rd edn. Van Nostrand Reinhold, New York
- 30. Satas D (1989) Handbook of pressure sensitive adhesive technology. In: Satas I (ed) Van Nostrand Reinhold, New York, p 61
- Somanathan N, Sanjay A, Arumugam V (2002) Compatibility studies on polystyrene and poly-n-butyl methacrylate. J Appl Polym Sci 83:2322–2330
- Hayashi S, Kim HJ, Kajiyama M, Ono H, Mizumachi H, Zufu Z (1999) Miscibility and pressure-sensitive adhesive performances of acrylic copolymer and hydrogenated rosin systems. J Appl Polym Sci 71:651–663

- Gordon M, Taylor JS (1952) Ideal copolymers and the secondorder transitions of synthetic rubbers. i. non-crystalline copolymers. J Appl Chem 2:493–500
- Zerjal B, Jelcic Z, Malavasic T (1996) Miscibility in thermoplastic polyurethane elastomer/poly(styrene-co-acrylonitrile) blends. Eur Polym J 32:1351–1354
- 35. Song M, Hammiche A, Pollock HM, Hourston DJ, Reading M (1995) Modulated differential scanning calorimetry: 1. A study of the glass transition behaviour of blends of poly(methyl methacrylate) and poly(styrene-co-acrylonitrile). Polymer 36:3313–3316
- Fox TG (1956) Influence of diluent and of copolymer composition on the glass temperature of a polymer system. Bull Am Phys Soc 1:123
- 37. Noriman NZ, Ismail H, Rashid AA (2010) Characterization of styrene butadiene rubber/recycled acrylonitrile-butadiene rubber (SBR/NBRr) blends: the effects of epoxidized natural rubber (ENR-50) as a compatibilizer. Polym Tes 29:200–208
- 38. Poh BT, Giam YF, Yeong FPA (2010) Tack and shear strength of adhesives prepared from styrene-butadiene rubber (SBR) using gum rosin and petro resin as tackifiers. J Adhes 86:846–858
- Gardon JL (1963) Peel adhesion. II. A theoretical analysis. J Appl Polym Sci 7:643–665
- Gardon JL (1963) Peel adhesion. I. Some phenomenological aspects of the test. J Appl Polym Sci 7:625–641

