

Resultant Synergism in the Shear Resistance of Acrylic Pressure-Sensitive Adhesives Prepared by Emulsion Polymerization of *n*-Butyl Acrylate/2-Ethyl Hexyl Acrylate/Acrylic Acid

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ABSTRACT: Acrylic emulsion pressure-sensitive adhesives (PSAs) were synthesized by the copolymerization of *n*-butyl acrylate with various levels of 2-ethyl hexyl acrylate (2EHA) and a small constant amount of acrylic acid. The effect of varying the *n*-butyl acrylate/2EHA monomer composition on the kinetic behavior of the polymerization and the characteristics of the copolymers prepared in a batch process were investigated. The results showed that increasing the amount of 2EHA in the monomer caused the polymerization rate and the glass-transition temperature of the acrylic copolymers to decrease. Increasing the amount of 2EHA caused the gel content of the copolymers to decrease, reaching a minimum at 50 wt %; thereafter, the gel content increased at higher 2EHA levels. For the acrylic emulsion, the peel-fracture energy of the PSAs decreased as the amount of 2EHA in the monomer was increased up to 50 wt %. At higher 2EHA levels, the peel-fracture energy

was relatively constant. Interestingly, a synergistic effect of increased shear resistance at 25 wt % 2EHA was observed without a significant trade-off in terms of the peel and tack properties. This behavior was attributed to a good interconnection between the microgels and the free polymer chains inside the contacting particles in the adhesive film. Cooperation between various levels of 2EHA in the copolymer structure simultaneously changed the crosslink molecular weight (M_c) of the microgels and the entanglement molecular weight (M_e) of the free chains in the adhesive network morphology. The adhesive performance of the PSAs was found to be correlated with their M_c/M_e values as the 2EHA proportion was varied. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 108: 606–613, 2008

Key words: pressure-sensitive adhesives (PSAs); adhesion; emulsion copolymerization

INTRODUCTION

Pressure-sensitive adhesives (PSAs) have been widely used in many scientific and industrial applications over the past few decades.^{1–3} Recently, strict regulations regarding the removal of volatile organic contents have caused the PSAs prepared by emulsion polymerization to be more commonly used than their solvent-borne counterparts. However, the emulsion PSAs have other advantages, including low cost, inflammability, high molecular weight, high solid content, and ease of application, in addition to their environmental safety.^{4,5} Among the various rubbers available, the acrylic ones, such as poly(*n*-butyl acrylate) and poly(2-ethyl hexyl acrylate), are widely used to produce the emulsion PSAs because of their low plateau modulus and good weatherability.^{3,6} Nevertheless, the solvent-borne acrylic PSAs

have been marginally preferred over the emulsion PSAs in high-performance applications because of their superior adhesive performance at high temperatures.⁷ The former have a much higher shear resistance, together with higher peel and tack properties, than the latter for the aforementioned applications. This is due to the continuous network morphology formed by the chain entanglements in the solvent-borne adhesive when the solvent has evaporated. By contrast, the microgels in the latex particles of the water-borne adhesive retain their discrete morphology in the adhesive film.⁸ This morphology decreases the shear resistance of the emulsion adhesive because of the poor interconnections between the microgels and the linear polymers in the film. Further investigations were thus conducted to increase the shear resistance of the emulsion adhesives by the copolymerization of *n*-butyl acrylate (nBA) or 2-ethyl hexyl acrylate (2EHA) with other monomers, such as methyl methacrylate and acrylic acid (AA), which are able to create hard segments in their copolymer structure.^{9–11} The reaction between functional groups of the polymer chains and the crosslinking reaction in the PSA films represent

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alternative approaches to enhancing the shear resistance. Nevertheless, the aforementioned approaches could increase the shear resistance of the emulsion adhesives at the expense of lower peel and tack adhesive properties.^{5,11,12} Recently, Tobing and Klein¹³ prepared acrylic emulsion PSAs with different functional groups, using isobutoxy methyl acrylamide and AA comonomers. Their results showed that crosslinking reactions between reactive groups in the adhesive film at a higher temperature resulted in a high shear resistance without a significant loss in the peel and tack properties. In another investigation, Tobing et al.¹⁴ prepared latex blends of high-gel-containing and gel-free 2EHA/AA copolymers. They showed that for the latex blends in which the crosslink molecular weight (M_c) of the microgels was greater than the entanglement molecular weight (M_e) of the linear polymers and the weight-average molecular weight (M_w) of the linear polymer was greater than $2M_e$, a synergistic effect in increasing the shear resistance of the PSA films was observed. Nevertheless, their results showed that the tack and peel of the emulsion blend adhesives decreased substantially when the highest shear resistance was obtained.

The substantial difference in the M_e values of the nBA/AA and 2EHA/AA copolymers prompted this work, in which nBA was copolymerized with various levels of 2EHA in the presence of a small constant amount of AA. In other words, the nBA/2EHA copolymerization was able to adjust the molecular parameters of the adhesive network and to influence the shear resistance in conjunction with the tack and peel properties. The effect of varying the nBA/2EHA monomer composition on the polymerization kinetics of the acrylic adhesives was also investigated.

EXPERIMENTAL

Materials

All reagents were purchased from Merck Co. (Darmstadt, Germany) unless otherwise stated. nBA, 2EHA, and AA were distilled *in vacuo* and stored at 0–5°C before use. Potassium persulfate and sodium dodecyl sulfate were used without any further purification. The solvents, high performance liquid chromatography (HPLC)-grade tetrahydrofuran (THF) and toluene, were also used directly. The deionized distilled water was prepared in our laboratory.

Emulsion polymerization

The synthesis was carried out in a 100-mL glass reactor equipped with a reflux condenser, a nitrogen inlet, and a septum through which kinetic samples were taken. The polymerization took place in a

batch emulsion process that was thermostatically controlled and continued for 180 min. The reaction mixture was held at 80°C and stirred magnetically at 300 rpm under a nitrogen flow. Emulsion polymerization was carried out for nBA/2EHA/AA monomer feeds in which the AA content of all recipes was 2.5 wt % based on the total monomer. Without consideration of the AA content in the monomer composition, the 2EHA levels in the monomer mixtures were 0, 25, 50, 75, and 100 wt %.

Kinetic study

Samples (1 mL) were taken from the reaction mixture at different polymerization times and poured into the aluminum dishes cooled in an ice–water mixture. The dishes were placed in a vacuum oven at 25°C for 24 h. The evolution of the weight conversion versus time was measured gravimetrically.

Gel content and M_c determination

The gel content and molecular characteristics of the PSAs were determined after the drying of the adhesive latices under the following conditions, which were similar to those used to prepare the adhesive test samples. The gel content of the acrylic PSA copolymers was measured via the solvent-extraction method. Three samples (0.25 g) of dried film were added to 25 mL of THF and stirred at 300 rpm and room temperature for 48 h. The samples were then centrifuged at 6000 rpm for 1 h. The sediment gel phase was separated from the sol phase and dried in a vacuum oven at 25°C for 24 h. The gel fraction was determined gravimetrically.

For the determination of M_c , 2.5 g of PSA film was added to 25 mL of THF and stirred at 350 rpm and room temperature for 48 h. Then, the sample was centrifuged at 6000 rpm for 2 h at 4°C. The sediment gel phase was separated from the sol phase and dried in a vacuum oven at 35°C for 24 h, and the sol phase was saved for gel permeation chromatography (GPC) experimentation and molecular weight determination. Approximately 100 mg of adhesive dried gel was pressed as a 20-mm disk between two paper microfilters. The pouch was heat sealed and placed in a glass vial with a plastic seal. Thirty milliliters of toluene was added to the vial. After 36 h, the pouch was removed and immersed in liquid N₂. By completion of the surface evaporation of liquid N₂, the swollen gel sample was weighted. Finally, the weight fraction of the polymer in the swollen gel was determined gravimetrically, and the volume fraction of the polymer (ϕ_p) was calculated. The Flory–Rehner equation was used to calculate M_c .^{7,15}

$$M_c = \frac{V_1 \rho_p [\phi_p^{1/3} - \phi_p/2]}{-[\ln(1 - \phi_p) + \phi_p + \chi \phi_p^2]} \quad (1)$$

where V_1 is the molar volume of toluene; ρ_p is the density of the polymer; and χ is the polymer–solvent interaction parameter, which can be estimated with the solubility parameter of the polymer and solvent.¹⁶

Molecular weight determination

The molecular weight of the acrylic PSA copolymers was measured with a GPC apparatus (Shimadzu, Tokyo, Japan) equipped with a refractive-index detector. The THF-soluble fraction from the aforementioned adhesive extraction experiment was injected into the GPC column (Waters Corp., Milford, MA; 10^4 Å). The temperature of the column was 40°C.

Particle size and size distribution

The particle size and size distribution of the PSA latices were measured via a dynamic laser light scattering apparatus with a wavelength of 632.8 nm and a laser light source of He and Ne gas. Light scattering measurements were performed with a Sematech SEM-633 (Nice, France) stepper-motor-driven goniometer.

Viscosity of the PSA latex

The viscosity of the PSA copolymer latex products was evaluated with Visco Star Plus (Lucerne, Switzerland) a rotational viscometer. The viscometer operates by the principle of rotation of a cylinder or disc (spindle L1), which is submerged in the latex to be analyzed, measuring the resistance of the substance at a preset speed. The resulting resistance or torque is the measurement of the flow viscosity. The measurements were carried out at room temperature and 200 rpm.

Determination of the glass-transition temperature (T_g)

The T_g values of the acrylate copolymers were determined with a Mettler–Toledo (Schwerzenbach, Switzerland) DSC 822 system differential scanning calorimeter. The temperature scanning range was -150 to 150°C at a heating rate of $-10^\circ\text{C}/\text{min}$ under a nitrogen atmosphere.

Adhesive-performance testing

The solid contents of the PSA latices were increased to 50 wt % via heating at 50°C before they were cast onto a substrate. The high-solid latices were used to coat 50- μm polypropylene films with a wire-rod die to give a 30- μm dry-film thickness. The coated films

were dried at room temperature for 24 h and then at 50°C for 3 h. PSA testing was done at 25°C and 45% relative humidity. The test samples were left to relax under these conditions at least overnight before testing.

For the 180° peel test, strips (15×40 mm²) of the adhesive-coated films were peeled off stainless steel substrates at a 2 mm/min tensile rate under the aforementioned test conditions. The adhesive strips were bonded to the stainless steel substrate by the application of a 13.4 kPa static contact stress over the bonded area for 30 s. For each prepared acrylic PSA, the peel test was carried out five times, and the average of the results was reported.

The shear resistances of the PSAs were measured on stainless steel with strips (15×40 mm) of the aforementioned adhesive-coated films. The steel substrates were washed with acetone and dried at 50°C to remove impurities. One end of each adhesive strip was bonded to the stainless steel with a contact area of 15×15 mm², whereas the other end was connected to a 9.5 N hanging weight. Before testing, a static contact stress of 13.4 kPa was applied across the bonded area for 30 s. In this case, the time to failure of the adhesive bond was recorded as a criterion for the shear resistance.

A loop tack test was carried out to evaluate the tack adhesive performance of the acrylic emulsion PSAs. In this case, part of the adhesive loop was bonded to the steel surface under a static contact stress of 1 kPa for a contact time of 10 s. The average debonding fracture energy was measured and reported as the tack-fracture energy. The pulling rate was 50 mm/min, and the measurements were done under the aforementioned test conditions.

RESULTS AND DISCUSSION

The acrylic emulsion PSAs used in this study were based on nBA, 2EHA, and AA, which were synthesized with a batch polymerization process. Table I shows the various monomer compositions used to synthesize the acrylic emulsion PSAs with different molecular compositions. The AA content used in all of the recipes was 2.5 wt %. The 2EHA proportions in the monomer compositions were 0, 25, 50, 75, and 100 wt % based on the 2EHA/nBA total monomer. Table II shows the properties of the prepared PSAs. T_g of the acrylic emulsion polymers decreased as the monomer composition was varied from entirely nBA to predominantly 2EHA. As expected, increasing the soft 2EHA units in the emulsion copolymers decreased T_g . The mean particle size of the adhesive latices was found to increase as the number of 2EHA units in the acrylic polymers increased (Fig. 1 and Table II).

TABLE I
Acrylate Copolymer Compositions and Sample Designations

Formulation ^a	Monomer charged per total monomer (wt %) ^b		
	AA	nBA	2EHA
K1	2.50 (0.042)	97.50 (0.958)	0.0 (0.0)
K2	2.50 (0.045)	73.13 (0.776)	24.37 (0.179)
K3	2.50 (0.049)	48.75 (0.561)	48.75 (0.390)
K4	2.50 (0.053)	24.37 (0.306)	73.13 (0.641)
K5	2.50 (0.059)	0.0 (0.0)	97.50 (0.941)

^a The water/total monomer weight ratio was 3, and the solid content of the latex products was 24.9 wt %.

^b The amounts in parentheses are the molar ratios.

The gel content of the acrylic PSAs was decreased by an increase in the amount of 2EHA up to 50 wt %, although higher 2EHA levels led to higher gel contents (Table III). This behavior could be attributed to the distribution state of the AA units in the polymer chains as the 2EHA content increased. More uniform AA units in the adhesive polymer chains resulted in a greater crosslink density and a higher gel content.

M_e of acrylic PSAs

The method used to predict the entanglement molecular weight of the acrylic-gel-containing polydisperse PSAs ($M_{e,\text{polydisperse}}$) was similar to that reported by Tobing and Klein.⁷ Using the plasticizer model, they predicted M_e values for polydisperse nBA/AA and 2EHA/AA copolymers based on M_e values for monodisperse nBA and 2EHA homopolymers:¹⁷

$$M_{e,\text{polydisperse}} = \frac{M_{e,\text{monodisperse}}}{\phi^{2.3}} \quad (2)$$

where $M_{e,\text{monodisperse}}$ is the entanglement molecular weight value for the monodisperse sample and ϕ is the weight fraction of the polymer species having a molecular weight greater than M_e . Nevertheless, for a gel-containing polydisperse sample having $M_c \geq M_e$, the gel fraction must be included as a high-molecular-weight polymer added together with the high-molecular-weight fraction of the sol polymer.

TABLE II
Properties of the Acrylic PSA Latices

Sample	Diameter (nm)	Polydispersity index	T_g (°C)	Viscosity (μ) (mPa s)
K1	63.7	1.15	-45.4	9
K2	58.4	1.17	-49.8	12
K3	59.9	1.20	-54.9	11
K4	61.4	1.23	-68.4	8
K5	75.9	1.11	-61.3	10

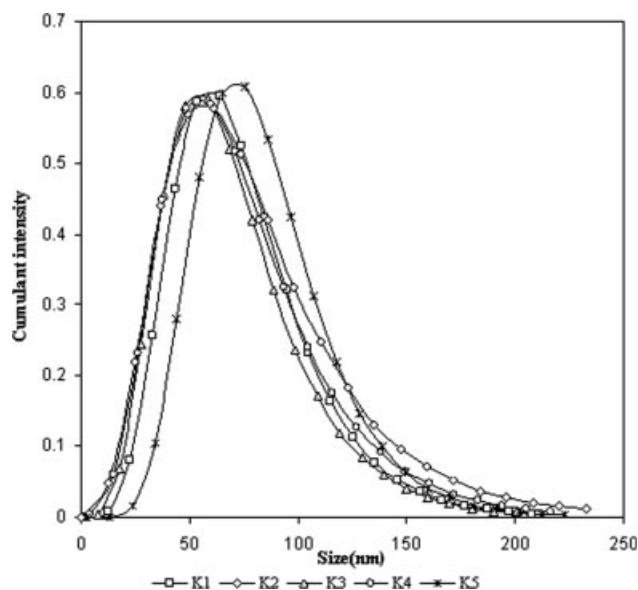


Figure 1 Particle size and size distribution curves of acrylic PSA latices with various 2EHA/nBA weight ratios.

Otherwise, for a sample having $M_c < M_e$, the gel fraction need not be included in calculating ϕ . Unlike the acrylic emulsion PSA having 100 wt % 2EHA, other acrylic PSA samples contained gel of the former type (Table III). The M_e values reported for monodisperse nBA and 2EHA polymers obtained from DMA measurements were 20,773 and 37,432 g/mol, respectively.⁷ The M_e values for monodisperse 2EHA/nBA copolymers having 25, 50, and 75 wt % 2EHA, estimated by the mixing rule, were 24,937, 29,102, and 33,267 g/mol, respectively. On the other hand, the values of ϕ were obtained from the GPC results and gel contents of the polydisperse PSA copolymers (Fig. 2). The calculated $M_{e,\text{monodisperse}}$ values for the acrylic PSAs are shown in Table III.

As shown, the variation of M_c with the 2EHA level for the acrylic latices showed the highest value at 50 wt % 2EHA, whereas M_e was increased monotonically by an increase in the 2EHA proportion. Nevertheless, the M_c/M_e values decreased from 2 for the nBA/AA copolymer to 0.45 for the 2EHA/AA copolymer. Varying the monomer composition followed by the acrylic copolymer composition changed the molecular parameters and, subsequently, the network morphology of the adhesive films.

The M_w values of the free chains in all of the PSA samples were much higher than their M_e 's ($M_w > M_e$; Table III). Therefore, the free copolymer chains of the contacting particles could become entangled with one another and interconnect the microgels. Nevertheless, the PSAs with a low M_c/M_e value had a tight network, which complicated the chain diffusion between the particles in the film.

TABLE III
Gel Content and Molecular Characteristics of the Synthesized Acrylic PSAs (M_w and M_n Applied to the THF-Soluble Fractions)

Sample	Gel (%)	M_c (kg/mol)	M_w (kg/mol)	M_n (kg/mol)	ϕ^a	M_e (kg/mol) ^b	M_e (kg/mol) ^c	M_c/M_e^c
K1	75.4	44	278	46	0.98	21	22	2.00
K2	73.1	44	363	65	0.96	25	28	1.57
K3	60.3	50	153	38	0.93	29	34	1.47
K4	68.2	38	309	62	0.94	33	38	1.00
K5	79.0	30	199	60	0.78	37	66	0.45

M_n = number-average molecular weight.

^a The value of ϕ was determined from GPC and the gel content.

^b M_e values of monodisperse acrylic polymers without AA.

^c M_e values of gel-containing polydisperse PSA samples estimated with the plasticizer model.¹⁷

Kinetic study

In comparison with the 97.5/2.5 nBA/AA copolymer with a high gel content (Table III), a low gel content was observed for the nBA homopolymer with no AA. This finding revealed the crucial role of AA units in the copolymer structure. The AA functional

groups were able to create crosslinks in the latex particles during polymerization and especially after the drying of the latex at higher temperatures. It was observed that adding a low level of AA to the monomer composition changed the kinetic behavior of the emulsion copolymerization. Figure 3 indicates the

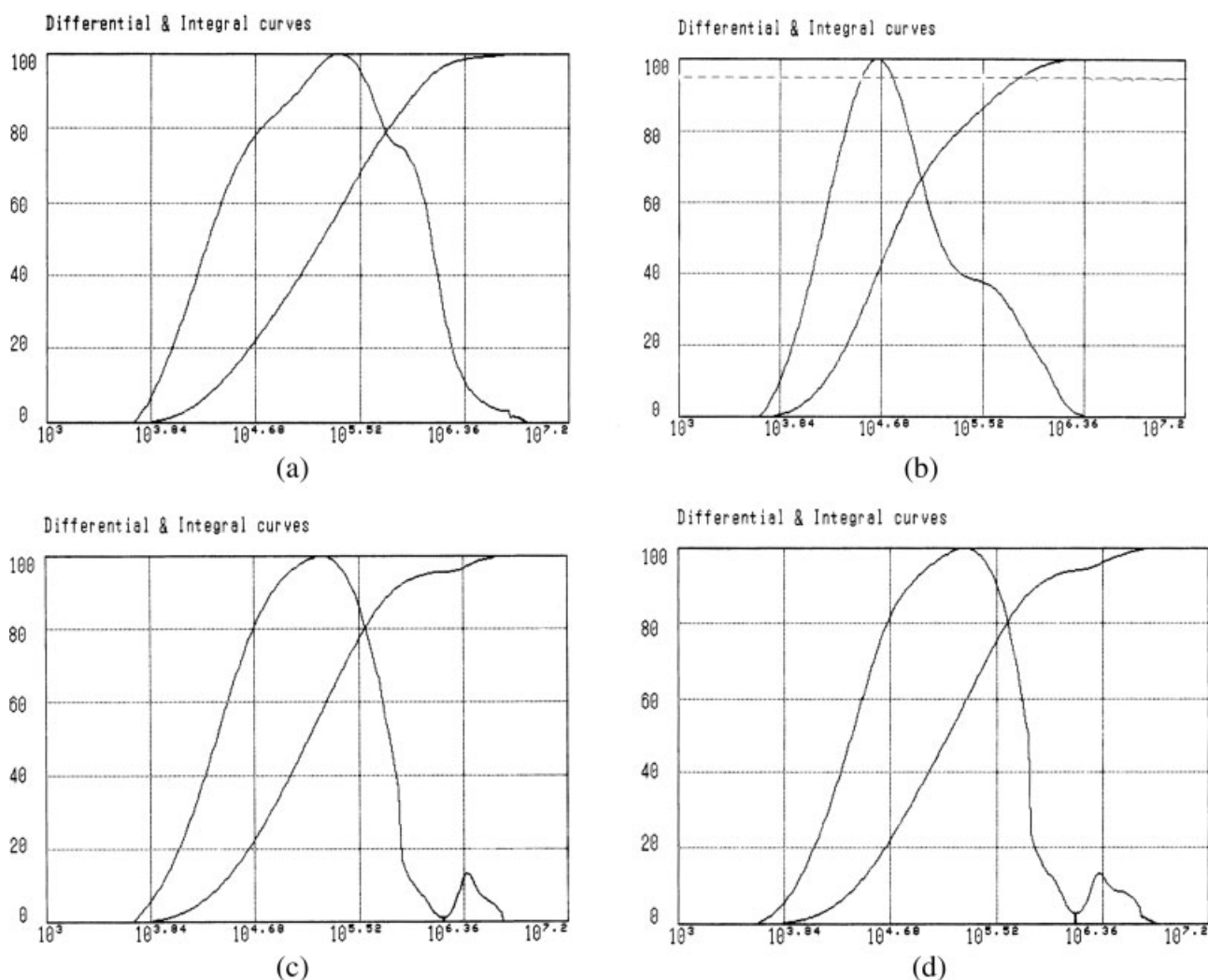


Figure 2 GPC molecular weight distributions and cumulative curves of polydisperse PSAs with various 2EHA/nBA weight ratios: (a) K2 (25/75), (b) K3 (50/50), (c) K4 (75/25), and (d) K5 (100/0).

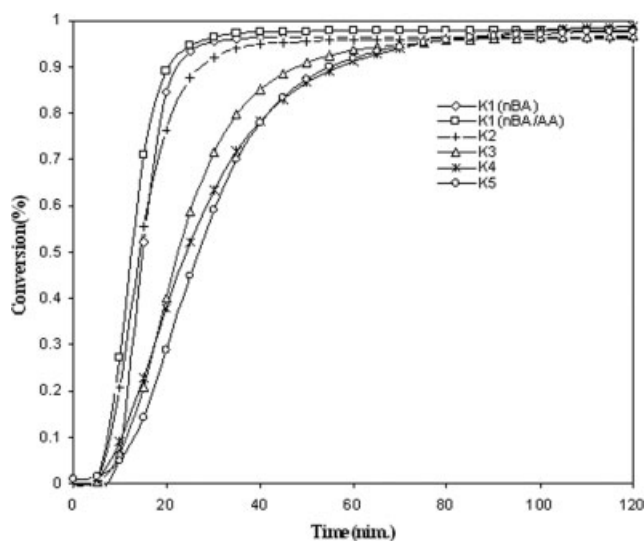


Figure 3 Dependence of the typical conversion–time curves in the emulsion polymerization of nBA/2EHA/AA on the percentage of 2EHA units in the initial feed.

evolution of the weight conversion versus time for the typical nBA homopolymerization and nBA/AA copolymerization. As shown, both curves are S-shaped, although the latter is shifted to the left on the time axis. As shown, the rate of nBA/AA copolymerization is predominantly higher than that of the nBA homopolymerization. This could be connected to the gel-point effect occurring over a shorter time period during the copolymerization.

The kinetic behavior of nBA copolymerized with various levels of 2EHA along with a small constant amount of AA was investigated. Figure 3 shows the variation of the conversion versus time for the acrylic copolymers according to the recipes listed in Table I. The polymerization rate clearly depended substantially on the monomer composition. The results indicated that the polymerization rate was increased whereas the 2EHA level decreased in the composition. For instance, the conversions of the nBA/AA copolymer without 2EHA and 2EHA/AA with the highest 2EHA proportion reached 90% after 21 and 58 min, respectively. By contrast, the gel contents of the PSA latices were decreased as the 2EHA concentration increased up to 50 wt %, after which further 2EHA increases led to higher gel contents (Table III).

Adhesive performance

Figure 4 shows the peel-fracture energy and the M_c/M_e values for the acrylic emulsion PSAs with various 2EHA proportions. As shown, a decrease in the peel-fracture energy was observed as the 2EHA level was increased to 50 wt %. When the 2EHA content was increased to more than 50 wt %, the peel-frac-

ture energy was retained at a constant level. This behavior could be attributed to the viscoelastic dissipation energy produced during adhesive debonding, which depended substantially on the molecular network parameters of the acrylic adhesives. The results showed that both M_e and the M_c of the PSAs depended on the adhesive composition (Table III). With an increase in the 2EHA content in the adhesives, M_c of the adhesive copolymer showed a peak at 50 wt %, whereas M_e increased uniformly. Nonetheless, the M_c/M_e value decreased only as the 2EHA level increased (Fig. 4). This finding showed that the fracture energy during adhesive debonding was lowered as M_c/M_e decreased. In other words, by an increase in the 2EHA level to 50 wt % and an increase in M_c , the bulk-fracture energy of the acrylic PSAs was decreased. It can be postulated that below 50 wt % 2EHA, the bulk-fracture energy depended predominantly on the M_c/M_e values. At higher M_c/M_e values, the microfibrils needed higher energy to deform and fracture during the peeling process. By an increase in the ethyl hexyl acrylate level above 50 wt %, no significant change in peel was observed, although the M_c/M_e value of the PSAs decreased. This behavior could be attributed to other parameters that affect the peel-fracture energy according to the adhesion rheological model presented by Gent and Schultz.¹⁸ Adhesives with higher soft segments or lower T_g values showed higher thermodynamic work of adhesion values. Above 50 wt % 2EHA, a sufficient increase in the thermodynamic work of adhesion compensated for the peel reduction as a result of the lowering of the M_c/M_e values.

Shear resistance is known to depend on both the gel content and the entanglement density, which

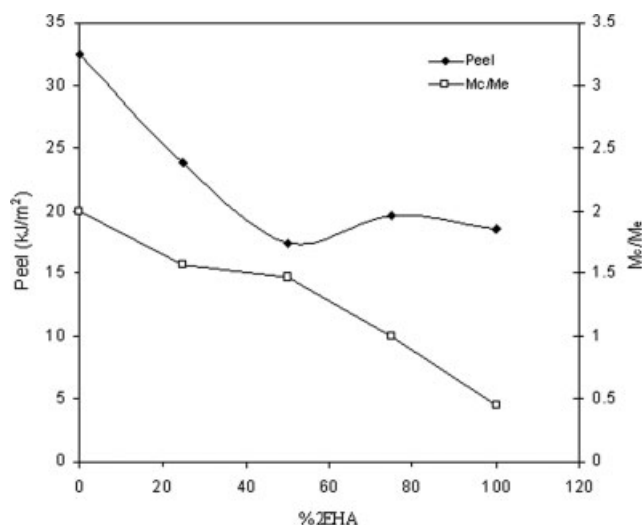


Figure 4 Dependence of the peel and M_c/M_e values of the acrylic PSA copolymers on the percentage of 2EHA units in the initial monomer feed.

influence the network morphology and the zero shear viscosity of the adhesive film. Nevertheless, for emulsion PSAs, the interconnection between the microgels and the free chains of the contacting particles was a crucial parameter that could improve the shear resistance. Figure 5 shows a plot of the shear holding power off stainless steel versus the 2EHA proportion. Interestingly, a synergistic effect in increasing the shear resistance was observed at 25 wt % 2EHA. This behavior could be attributed to a good interconnection between the microgels and the free chains of the contacting particles in the PSA film. It seemed that the interconnection was dependent on the M_c/M_e values, which varied according to the 2EHA level in the emulsion PSAs. The emulsion adhesive with 25 wt % 2EHA showed an optimum M_c/M_e value close to 1.57 and exhibited the highest shear resistance. Above 25 wt % 2EHA in the monomer feed, the shear resistance started to decrease monotonically. This behavior was in accordance with that reported by Tobing et al.,¹³ who showed that acrylic PSAs with $M_c > M_e$ and $M_w \geq 2M_e$ achieved a high shear resistance without a significant trade-off in peel and tack. This work revealed that acrylic emulsion adhesives with $M_w \geq 2M_e$ had an optimum M_c/M_e value close to 1.57, which resulted in the highest shear resistance without a significant decrease in the peel and tack properties (Figs. 4–6).

Figure 6 illustrates the tack energy of the emulsion acrylic PSAs versus the 2EHA proportion in the monomer composition. Increasing the 2EHA level in the polymerization reaction increased the tack energy. The tack adhesive performance was mainly affected by the polymer chain mobility of the adhe-

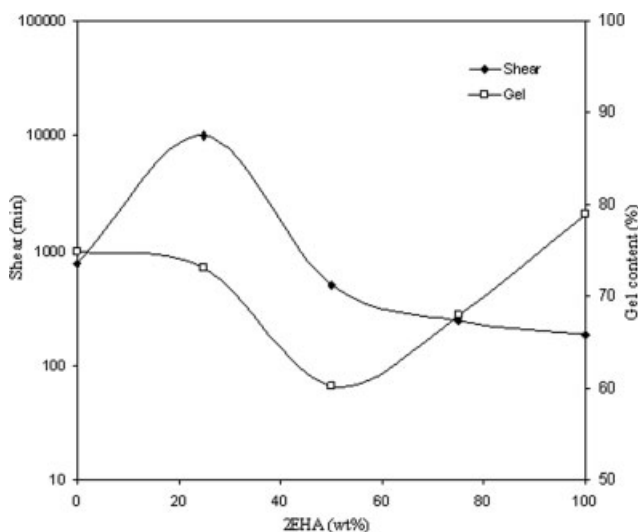


Figure 5 Shear resistance and gel content versus the 2EHA level for the synthesized acrylic emulsion PSAs with various 2EHA units in the initial monomer feed.

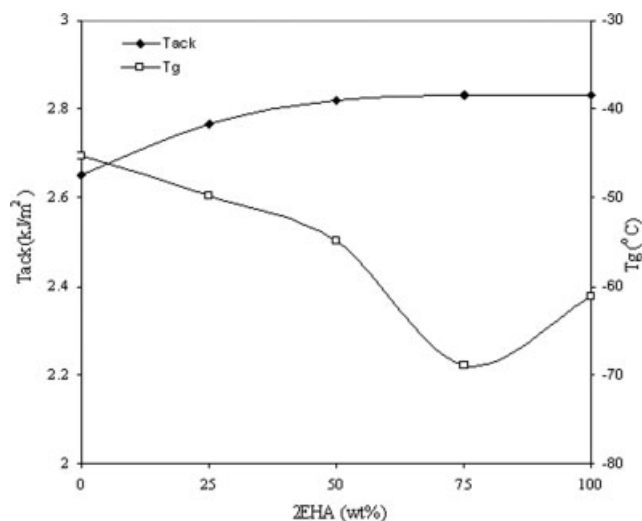


Figure 6 Tack and T_g of the acrylic copolymers versus the 2EHA level for the synthesized acrylic emulsion PSAs with various 2EHA units in the initial monomer feed.

sive surface rather than the adhesive bulk property. The tack enhancement for the acrylic emulsion PSAs was a result of the increased mobility of the surface segments arising from the decreased T_g of the PSAs with more 2EHA units (Fig. 6). Nevertheless, the variation of tack was not remarkable because the acrylic adhesives consisted of both soft nBA and 2EHA units with sufficiently low T_g values to wet the substrates sufficiently.

CONCLUSIONS

Emulsion acrylic PSAs with various entanglement densities and gel contents were prepared by the copolymerization of nBA and 2EHA along with a small amount of AA. The results showed that the M_c and M_e values of the acrylic PSAs were substantially influenced by the 2EHA proportion in the monomer composition. A synergistic effect in increasing the shear resistance was observed at a gel content greater than 70% and M_c/M_e values around 1.5 without a trade-off in the tack and peel properties. This behavior could be attributed to the optimum chain network morphology, which led to a good interconnection between the microgels in the emulsion adhesive film.

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