Improvement in mechanical properties and thermal stability of solvent-based pressure-sensitive adhesives based on triazine heterocyclic monomer

Jun Hua Chen,[1,2] Qiang Zhao,[1,2] Ying Wang,[1,2,3] Fubin Luo,[1,2] Lu Shen,[1,2,3] Kun Wu,[1] Li Yan Liang,[1] ManGeng Lu[1]

[1]Key Laboratory of Cellulose and Lignocellulosics Chemistry, Guangzhou Institute of Chemistry, Chinese Academy of Sciences, Guangzhou 510650, People's Republic of China
[2]University of Chinese Academy of Sciences, Beijing 100039, People's Republic of China
[3]Key Laboratory of Polymer Materials for Electronics, Guangzhou Institute of Chemistry, Chinese Academy of Sciences, Guangzhou 510650, People's Republic of China
Correspondence to: M. Lu (E-mail: mglu@gic.ac.cn)

ABSTRACT: A heat-resistance monomer denoted as triazine heterocyclic compound (TGIC-AA) was synthesized and applied into improving the thermal stability of solvent-based acrylic pressure sensitive adhesives (PSAs) through copolymerization. The modified acrylic PSAs tapes possessed longer holding time at temperature up to 150°C and no large areas of residues could be seen when peeled off on the substrate while the temperature of test was cooled down to room temperature. The thermal stability could be significantly enhanced in PSAs as the content of triazine heterocyclic compounds increased due to the extensive crosslinking networks. This indicated a worthy method to prepared heat resistant acrylic PSAs. An obvious reduction in peel adhesion occurred at the content of crosslinkers range 5 wt %-7 wt %, while beyond 7 wt % adhesion failure occurred. The influences of crosslinking density on the molecular weight, glass transition temperature and viscosity, etc. for PSAs were also studied. © 2015 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 2016, 133, 43281.

INTRODUCTION

Pressure sensitive adhesives (PSAs) are viscoelastic materials that can bond to solid substrates under a light pressure load and removed easily without any visible residues.[1–3] The acrylic PSAs are widely used in mounting tapes, labels, double-side tapes, medical pads and decorates films.[4] Most conventional acrylic PSAs are applied at room temperature.[5,6] However, single-components acrylic PSAs tapes with excellent adhesive properties and shear holding power at elevated temperature have developed rapidly in the recent years.[7,8]

The acrylic PSAs are semisolid phase materials, the tack, peel and shear strengths can be impacted by the properties of polymer, such as viscosity, molecular weight, copolymer composition as well as glass transition temperature (T_g).[9] As for selection of the chemical composition, the rheological behaviors of the acrylic PSAs can be controlled and tailored to meet the needs of particular application.[9] So the solvent-borne pressure sensitive adhesives are composed by multiblock linear polymers,

which have a restriction in mechanical and thermal properties.[10] To improve thermal stability, the mobility of molecular chains on acrylic PSAs need to be controlled. Generally, the most effective method to restrict the molecular mobility is to modulate the crosslinking density for improving adhesive properties, especially thermal stability. Three ways are usually used to improve the thermally stable properties of polymers by introducing polar group to the molecular chain, incorporating heteroaromatic ring and curing for crosslinking. The introductions of heteroaromatic ring and crosslinking for single-component adhesive are the most widely used because of various advantages compared to conventional multicomponent adhesives, such as well-adapted, high stability.[11–16] The acrylic PSAs are prepared by various monomers with unsaturated carbon double bonds which can be further crosslinked by a heating treatment temperature.[17–21]

Triglycidyl isocyanurate (TGIC) is the triazine-based epoxy compound, which can be used as a hardening agent for
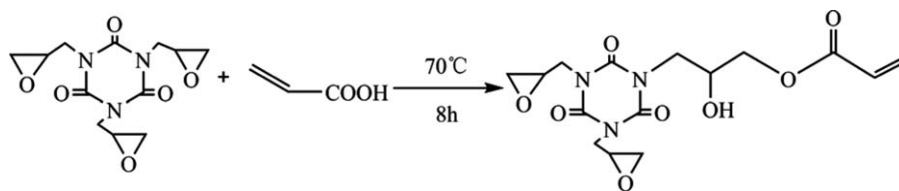
Figure 1. Schematic diagram of synthesis of the triazine heterocyclic compound (TGIC-AA).

laminates, coatings and adhesives.[22–24] It is generally known to have a high glass transition temperature (T_g) and thermal stability. It is reported that, one of the most effective method that can improve the thermal resistance by introducing heteroaromatic ring and triazine heterocyclic to the polymeric chains.[25–27] The modified acrylic PSAs with multi-dimensional network structure comprise acrylic polymer links and rigid rings. The flexibility of acrylic chains is correlated closely the adhesion properties. Moreover, the variation of molecular structure would obtain an improvement by the chains with rigid rings. Therefore, it is a good method to achieve thermally stable structure of acrylic PSAs by introducing the isocyanurate groups to the main chains. However, there were very few reports about isocyanurate groups that were used for improving the thermal resistance of acrylic PSAs.

The key to obtain excellent thermo-resistance in pressure sensitive adhesives is to generate superior crosslinkers. In this study, a heat-resistance monomer denoted as triazine heterocyclic compound (TGIC-AA) was synthesized and applied into improving the thermal stability of solvent-based acrylic pressure sensitive adhesives (PSAs) through copolymerization with acrylic precopolymer. The modified acrylic PSAs have unsaturated double bonds for further crosslinking.

EXPERIMENTAL

Materials
N-butyl acrylate (BA, Tianjin Kemiou Chemical Reagent), acrylic acid (AA, Shanghai Aladdin Reagents), and vinyl acetate (VAc, Shanghai Aladdin Reagents) were employed as monomers to prepare acrylic copolymer by radical polymerization. Benzoyl peroxide (BPO, Shanghai Aladdin Reagents) and ethyl acetate (EAC, Guangzhou Chemical Reagent Factory) were used as radical initiator and solvent. Triglycidyl isocyanurate (TGIC, epoxy equivalent: ≤103, total chlorine: ≤0.08) was supplied by Kunshan Xinkui Polymer New Materials. *N,N*-Dimethylformamide (DMF, Shanghai Titan Scientific) was used as a solvent when synthesizing the special monomer. N,N-Dimethylaniline (Tianjin Fuchen Chemical Reagents Factory) and 2,6-Di-tert-butyl-*p*-cresol (Tianjin Fuchen Chemical Reagents Factory) were applied as a catalyst and a polymerization inhibitor for the reaction between triglycidyl isocyanurate and acrylic acid.

Synthesis of Acrylic Copolymer
The acrylic copolymer was synthesized in ethyl acetate at temperature of 80°C with 60 wt % polymer content by using the fixed composition of *n*-butyl acrylate (BA), vinyl acetate (VAc), and acrylic acid (AA) at presence of 0.5 wt % radical starter benzoyl peroxide (BPO). The solvent-based acrylic copolymer

was synthesized with solid content on 35 wt %. The dosage time was 3 h.

Synthesis of Heat-Resistance Monomer
The triazine heterocyclic compound (TGIC-AA) was synthesized as shown in Figure 1. The compound was synthesized by the following procedure. A 100-ml single-neck round-bottomed flask equipped with a magnetic stirrer was charged with triglycidyl isocyanurate (TGIC), acrylic acid (AA), *N,N*-Dimethylaniline (1 wt % to TGIC), 2,6-di-tert-butyl-*p*-cresol (1.5 wt % to TGIC) and a solvent of DMF. To synthesize the heat-resistance monomer, the mole ratio of TGIC to AA was about 1:1. The reaction was carried out at 70°C until the acid value was less than 5 mg KOH/g to obtain the heat-resistance monomer.

Preparation of Modified Acrylic PSA Film
Before the end of synthesis of acrylic copolymer, the triazine heterocyclic compound (0–10 phr to acrylic copolymer) was dropped in acrylic copolymer for 2 h to prepare modified acrylic PSAs. The mixture obtained was casted on to polyethylene terephthalate (PET) films of about 60 μm in thickness. The adhesive films were dried at 120°C for 5 min to remove the solvent. Eventually, the thickness of the acrylic PSA layer was about 25 ± 3 μm.

Characterization
Composition Analysis. Fourier-transform infrared (FT-IR) spectra of the triazine heterocyclic compound (TGIC-AA) and the reaction products were performed by Analects RFX-65 spectrometer over the frequency range of 4000–400 cm^{-1} with KBr pellets. ^{1}H-NMR of the heat-resistance monomer (TGIC-AA) was recorded on a Bruker AVANCE AV400 (400 Hz) spectrometer using CDCl$_3$ and TMS as a solvent and the internal reference at room temperature. Determining the acid value of the compound was according to GB/T 601-2002. The epoxy value was measured according to GB/T 1677-2008.

Measurement of Viscosity. The viscosities of pressure-sensitive adhesives were measured with a rotary viscometer (NDJ-1 Shanghai) at room temperature.

Molecular Weight (GPC). Molecular weights of the adhesive solutions were determined by gel permeation chromatography (GPCmax VE 2001 Malvern). A small amount of dry adhesive was dissolved in tetrahydrofuran (THF) and shaken up for 24 h. The solution was filtered using 0.2 μm polytetrafluoroethylene filter. The mobile phase was at the rate of 1 ml/min and the injection volume was 0.1 ml.

Glass Transition Temperature (T_g). The glass transition temperatures (T_g) was measured by a DSC 200 F3 (Netzsch, Germany). The dry samples of 5 mg were sealed in aluminum
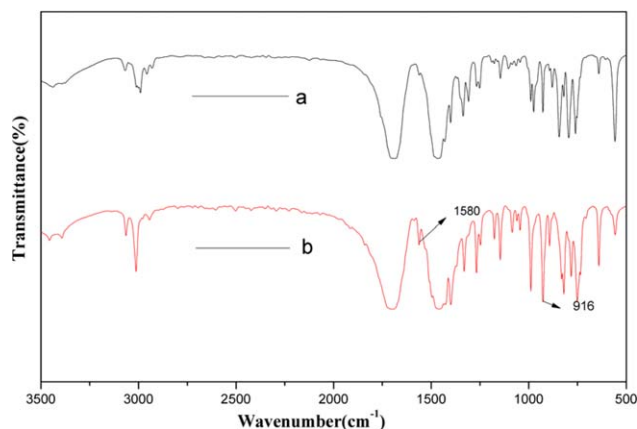
Figure 2. FTIR spectra of (a) TGIC and (b) heat-resistance monomer. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Figure 3. The acid value of the compound.

sample plans. The temperature of samples was drop to $-70^\circ C$ over 3 min and then heated to $110^\circ C$ at rate of $10^\circ C/min$.

Tack Properties. The balls of different size passed through a slope below the 100 mm × 100 mm PSA tape sample and the number of biggest ball which stopped on it would be recorded.

180° Peel Strength. The tapes with a width of 25 mm were posted in the surface of stainless steel plates which had been cleaned by tissue. The standard roller rolled on tapes back and forth three times. 30 min later, 180° peel strength was tested by an intelligent electronic tensile testing machine at a peeling speed of 300 mm/min.

Shear Holding Power. The prepared standard tapes of 25 mm × 70 mm were attach to the surface of stainless steel sheets. A standard roller moved back and forth three times to improve the contact between the PSA tapes and the substrate. One hour later, the samples hanged vertically in the test rack with a 2 kg load. The time of tamp which fell off from the steel sheets was recorded as shear holding power.

Shear Strength at High Temperature. The method of test was the same as the regular shear strength test described. The big difference was that the test temperature shifted from room temperature to high temperature (150°C). The test samples were placed in an oven.

Thermo Resistance. The degrease cotton with ethanol cleaned stainless steels which was used as a substrate. The tape samples that attached to surface of steel sheets were pressed three times with a 2 kg roller. Then, the tape samples were placed in an oven at 150°C for 4 h. When the temperature of the test was cooled down to room temperature, the tapes were peeled off slowly to observe the surface conditions of the stainless steel planes about amount of the residues.

Thermal Stability Analysis (TGA). The thermal stability of PSA samples were carried out in a Perkin-Elmer Pyris-1 TG analyzer at a heating rate 10°C/min from 40 to 550°C under a nitrogen atmosphere.
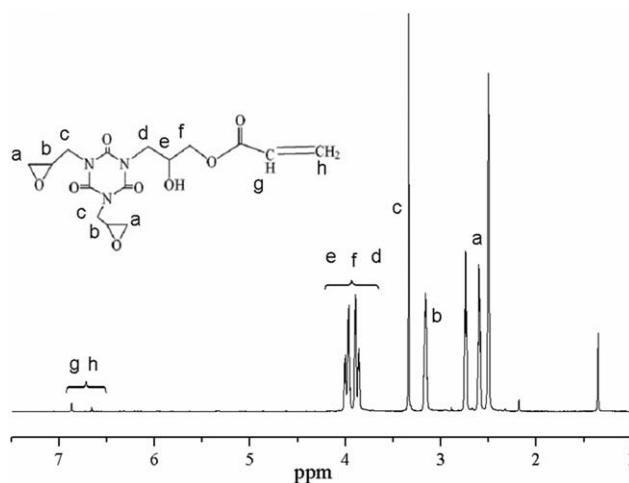
RESULTS AND DISCUSSION

Characterization of Synthesized Heat-Resistance Monomer

Figure 2 showed the typical FTIR spectra of TGIC (line a) and heat-resistance monomer (line b). The characteristic absorption band of C═C group was observed at 1585 cm^{-1} in the line b. Figure 3 showed that the acid value decreased to 5 mg KOH/g after 30 min. what's more, the epoxy value was calculated from 0.99 to 0.59 after 60 min. It meant that acrylic acid (AA) basically grafted on the main structure of TGIC. It could be seen from the curves, the peaks at 916 cm^{-1} which represented the epoxy groups of TGIC and TGIC-AA, as shown in the all lines of a and b, were still there.

The ^1H-NMR spectrum of heat-resistance monomer (TGIC-AA) was shown in Figure 4. It was clear to observe two peaks with the chemical shift about 6.7 ppm in the spectrum because of the influence of C═O band, which was the typical chemical shift of double bond and indicated that TGIC reacted with AA. So, it was concluded that the heat-resistance monomer (TGIC) had been synthesized.
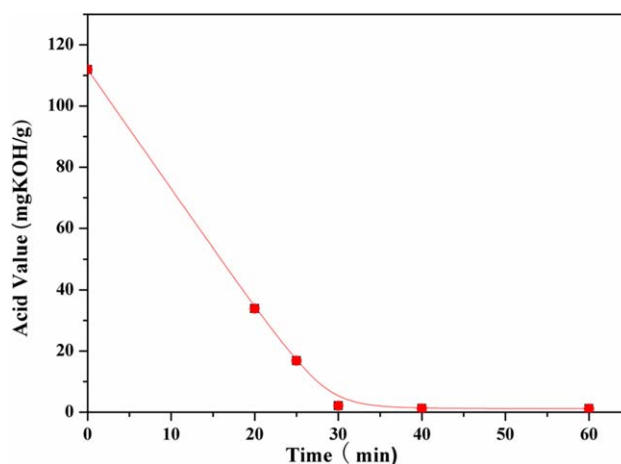
Figure 4. ^1H-NMR of spectrum of the heat-resistance monomer. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
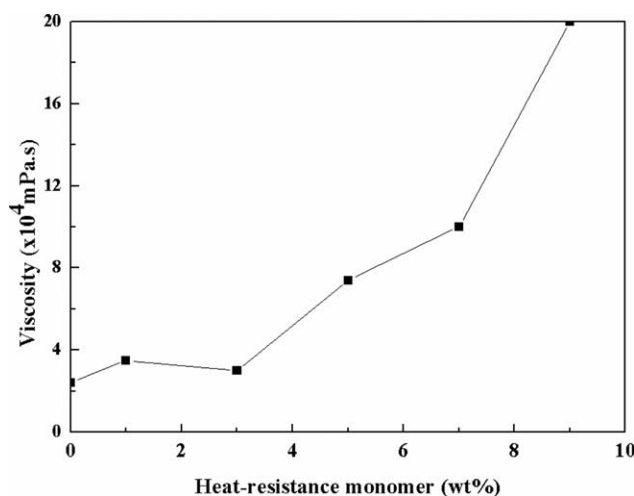
Figure 5. Effect of the heat-resistance monomer on viscosity of radical polymerization.

Figure 6. Effect of heat-resistance monomer content on 180° peel strength of PSA tapes (monomer ration: BA:VAc:AA = 81:15:4 wt %; reaction temperature: 80°C; reaction time: 8 h; peeling speed: 300 mm/min). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

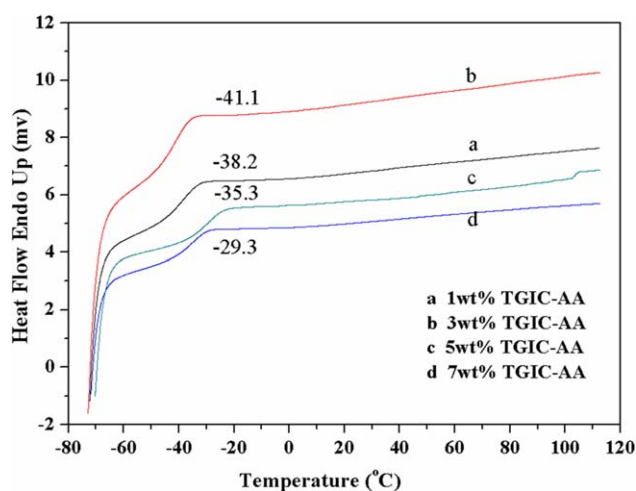### Viscosity of Properties of the Acrylic PSAs

The viscosity values of the acrylic PSAs were shown in Figure 5. The value of viscosity strongly depended on content of the heat-resistance monomer. In general, the viscosity increased with the increasing of crosslinker concentration.[28] The adhesion performance would be lost after sizing when the viscosity was high.[8] During polymerization, with the crosslinking points increased, the specific surface area increased as well, which may result in a crowded population in limited area. Besides, the molecular weight of the acrylic PSAs would gradually increase, thus the viscosity was elevated.

Molecular Weight of Acrylic PSA

As shown in Table I, all mentioned molecular weights and molecular weight distribution called polydispersity (P_d) were regulated using heat-resistance monomer concentrations. The weight average molecular ($\overline{M_w}$) and the number molecular weight ($\overline{M_n}$) increased with the increasing of content of the heat-resistance monomer. Besides, polydispersity (P_d) was improved by as much as 40%. The reason was that the main chains of acrylic PSAs formed crowded molecular. The highest $\overline{M_w}$ was reached with the use of 9 wt % heat-resistance monomer as crosslinker. When the molecular weight was above 1.5×10^6 g/mol, an absolute interface adhesion occurred when the tapes were peeled off as shown in Figure 6. It could be explained that heteroaromatic ring was easy to produce

Table I. Molecular Weight and Molecular Distribution of Synthesized Acrylic PSAs

Samples	$\overline{M_w}$	$\overline{M_n}$	$P_d = \frac{\overline{M_w}}{\overline{M_n}}$
PSA-0wt %	1013000	825000	1.23
PSA-1wt %	1043000	833000	1.25
PSA-3wt %	1178000	842000	1.40
PSA-5wt %	1195000	854000	1.40
PSA-7wt %	1510000	992000	1.52
PSA-9wt %	2100000	1210000	1.73

gelatinizing for acrylic PSAs. There were more crosslinks available during polymerization, longer polymer chains were formed and so the viscosity increased. This effect governed the trend in relatively wide polydispersity, which rapidly increased with increase in the concentration of the crosslinkers. The increases in the polydispersities of the acrylic polymer were due to the remarkable increases in the proportions of molecular fractions with higher molecular weights. There were more crosslinking points and longer chain lengths in those polymer fractions.

DSC Analysis

Glass transition temperature (T_g) of uncrosslinked acrylic PSAs calculated by the Fox equation was close to $-41°C$. The actual measured value was as much as $-38°C$. The reason might be that triazine groups were in polymer chain and the rest of epoxy groups reacted with the carboxyl groups, which contributed to physical crosslinking. As shown in Figure 7, the flexible
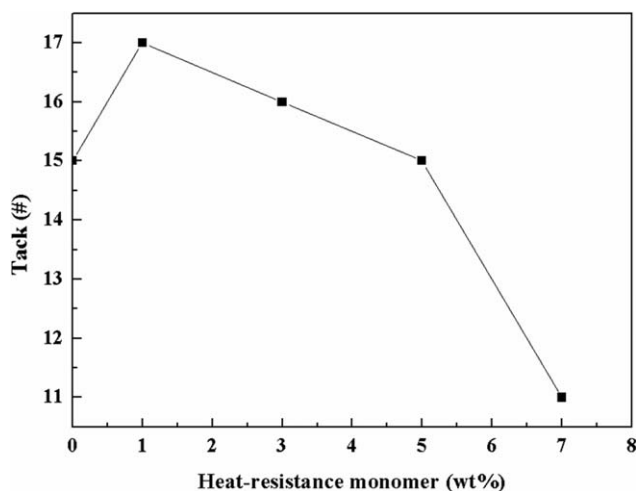
Figure 7. Effect of the heat-resistance monomer amount on glass transition temperature (T_g).
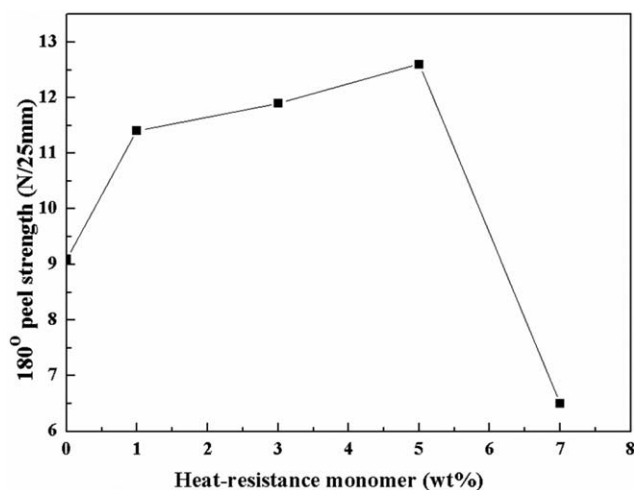
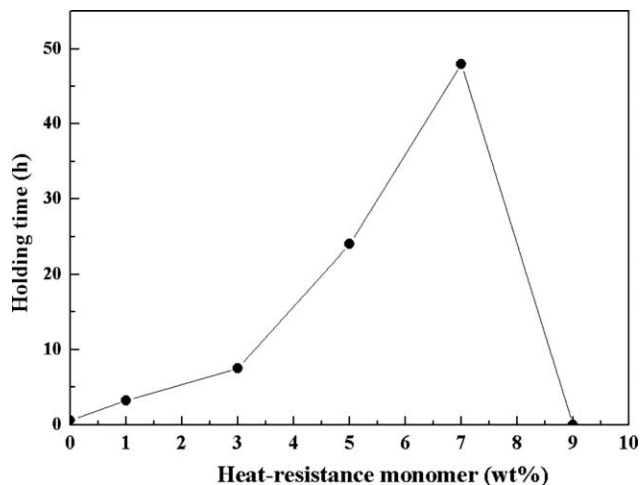**Figure 8.** Effect of heat-resistance monomer content on tack properties of PSA tapes. (monomer ration: BA:VAc:AA = 81:15:4 wt %; reaction temperature: 80°C; reaction time: 8 h).

Figure 9. Effect of heat-resistance monomer content on shear holding power of PSAs tapes at room temperature (monomer ration: BA:VAc:AA = 81:15:4 wt %; reaction temperature: 80°C; reaction time: 8 h).

side groups and symmetrical structure in heteroaromatic rings may lead to the decreasing of the glass transition temperature when the amount of heat-resistance monomer increased from 1 to 3 wt %. The value of T_g increased with the crosslinker content increasing to 5 wt %. It could be explained that the chemical crosslinking networks which formed via crosslinking reaction may hinder the movement of molecular chains. As a result, the T_g of the acrylic PSAs was elevated.

Adhesion Properties
As shown in Figure 8, the acrylic PSAs tapes with heat-resistance monomer had excellent tack properties. The tack increased from 15# to 17# when the amount of heat-resistance monomer increased from 0 to 1 wt %, then decreased at 3 wt %. And it was only 11# with heat-resistance monomer of 7 wt % in the polymer. However, a large amount of heat-resistance monomer would reduce the adhesion of PSAs tapes. The tack failure occurred when the amount of heat-resistance monomer exceeded 9 wt % and the steel ball was difficult to stop on the surface of PSAs tapes. It could be explained that the crosslinking points increased when the heat-resistance monomer content was tiny. And relative motion of segments was occurred. Therefore, the tack would firstly increase and then decrease with the heat-resistance monomer content increased. The excessive crosslinking points would appear in the system when the heat-resistance monomer content gradually increased. The results showed that the rigidity of the system and the glass transition temperature would increase with the elevation of the crosslinking density. So, the bonding properties of the system and tack would decrease.

As shown in Figure 6, the 180° peel strength increased firstly and then decreased when the amount of heat-resistance monomer increased. The heat-resistance monomer could enhance the cohesive of polymer. The maximum value reached 12.5 N/25 mm with heat-resistance monomer of 5 wt % in monomers. But the excessive crosslinking points would reduce the adhesives

of PSAs when the content of crosslinker monomer further increased.

The shear strength of crosslinked acrylic PSAs containing the heat-resistance monomer as crosslinkers measured at room temperature (25°C) were presented in Figure 9. As indicated the shear strength of the acrylic PSA after crosslinking is directly proportional to the concentration of the heat-resistance monomer employed. With the amount of heat-resistance monomer increasing, the holding time distinctly increased from 0.6 to 48 h. After that the holding time rapidly declined to zero. It could be explained that, with a small amount of heat-resistance monomer, the linear molecular chains began to form polymeric networks or crosslinked systems consisting of interconnected macromolecules.[13] As the amount of crosslinker monomer increased, the crosslinking density and the cohesion of acrylic PSAs would increase. When heat-resistance monomer content reached 7 wt %, the three dimensional crosslinking networks completely formed. However, a large amount of crosslinking points can produce excessively crosslinked and affect the holding adhesive of pressure-sensitive adhesives when the amount of heat-resistance monomer was over 7 wt %.

Table II. Effect of Heat-Resistance Monomer (TGIC-AA) Content on Holding Time at Different Temperature

Samples	Holding time (h)	
	100°C	150°C
PSA-0 wt %	0.03	0.01
PSA-1 wt %	0.05	0.04
PSA-3 wt %	24	0.23
PSA-5 wt %	40	25
PSA-7 wt %	>72	>72
PSA-9 wt %	–	–

Figure 10. Thermoresistance of PSA was tested at 150 and 25°C. (a)–(e) PSA tapes were crosslinked by 0, 1, 3, 5, 7 wt % heat-resistance monomer, respectively at 25°C; (g)–(k) PSA tapes were crosslinked by 0, 1, 3, 5, 7 wt % heat-resistance monomer, respectively at 150°C (monomer ration: BA:VAc:AA = 81:15:4 wt %; reaction temperature: 80°C; reaction time: 8 h; heating rate: 10°C/min; repeated test: three times. Peeling speed: 300 mm/min). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Effect of Crosslinking on Adhesive Properties

As shown in Table II, shear strength also increased with an increase in the heat-resistance monomer amount for PSAs at high temperature (150°C). The holding time increased owing to the formation of crosslinking network. The holding time of the acrylic PSAs would obtain minor changes at room temperature in Figure 6 when the amount of heat-resistance monomer was under 3 wt %. When the addition amount of heat-resistance monomer was more than 9 wt %, the large number of crosslinking points with unsaturated double bonds would result in cohesive failure under high temperature tests. When the amount of heat-resistance monomer was higher than 5 wt %, the shear strength maintained over 24 h at 150°C, which got the similar level of thermoresistance PSAs.[29] The cohesive strength could also be influenced by the average molecular weight about crosslinking points (M_c), which was calculated according to this eq. (1) below[30]:

$$M_c = \frac{V_1 \rho \left[\varphi^{1/3} - \varphi/2 \right]}{-\left[\ln(1-\varphi) + \varphi + \chi \varphi^2 \right]} \quad (1)$$

V_1: molar volume of toluene, ρ: density of polymer, φ: volume fraction of polymer in toluene, χ: polymer–solvent interaction parameter.

The liner PSAs can form a large crosslinking network. When the $M_c \geq M_e$.[31] A decrease in M_c caused by the increase of crosslinker amount, which also resulted in improvement of cohesive strength and crosslinking density of acrylic PSAs.

Figure 10 indicated the effect of the crosslinker on the thermoresistance property. The crosslinked PSAs had excellent peeling property that the tapes did not have any residues left at room temperature while they might have some obvious residues when they were peeled at 150°C. It could be explained that crosslinking by the heat-resistance monomer was insufficient and cohesive strength of acrylic PSAs was not enough to easily peel off from surface of steel sheets. What's more, the

Table III. Effect of the Commercial Crosslinker Content on Appearance of Steel Sheets at 24 h/150°C (the Amount of Addition: 7 wt %)

Samples	Appearance
Polyfunctional aziridine	Large residue
Isocyanate	Small residue
GMA	Large residue
MQ silicone resin	No residue
TGIC-AA	No residue

viscosity of adhesives was low and liquid that can easily leave some obvious residues at elevated temperatures. With the introduction of chemical bonds during chemical crosslinking, an increase in cohesive strength of acrylic PSAs would be accomplished during the drying of the synthesized PSA coating in the drying step.[32] Figure 10(h) and (i) revealed that the peeling properties of crosslinked PSAs were markedly improved at 150°C. Table II also showed that the shear strength maintained over 72 h at 150°C when the heat-resistance monomer content was 7 wt %. Compare with the commercial crosslinker, the appearance on surface of steel sheets can still remain excellent thermal and mechanical properties as shown in Table III. The thermoresistance test
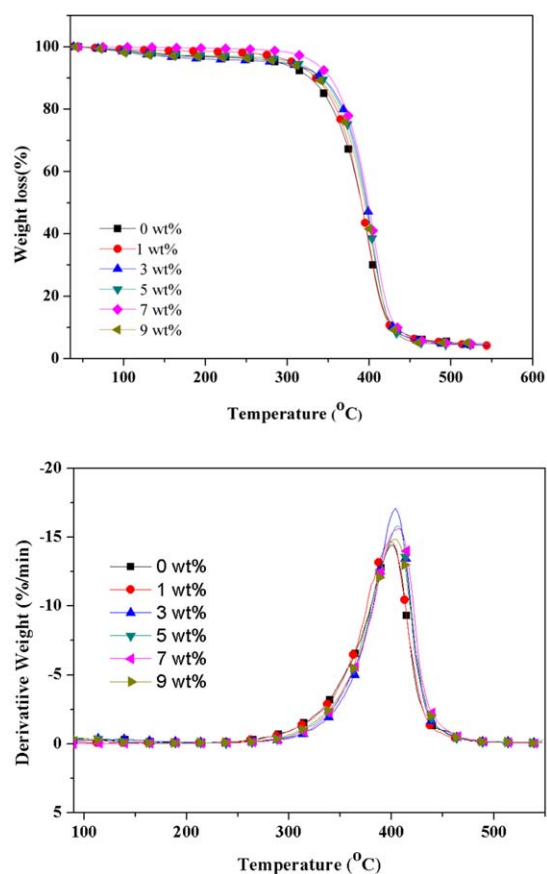
Figure 11. TG and DTG curves of acrylic PSAs with heat-resistance monomer in nitrogen atmosphere. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table IV. Effect of Heat-Resistance Monomer Content on Residue Char Yields

Samples	Char residue at 550°C (wt %)
PSA-0 wt %	3.82
PSA-1 wt %	4.11
PSA-3 wt %	4.19
PSA-5 wt %	4.42
PSA-7 wt %	4.51
PSA-9 wt %	4.86

indicated that the thermal stability of the crosslinked acrylic PSAs was maintained at elevated temperatures. It can be explained that the heat-resistance monomer may contribute to the stable configurations of acrylic PSAs.

Thermal Stability of Acrylic PSAs

Thermogravimetric analysis was used to verify the effect of heat-resistance monomer on thermal stability of acrylic PSAs. TG and DTG curves of acrylic PSAs with heat-resistance monomer in nitrogen atmosphere were shown in Figure 11, all these acrylic PSAs exhibited a similar trend of degradation. The beginning degradation temperature of samples, which was evaluated by the temperature of 10% weight loss, and the solid residue at 550°C, were obtained from the TG curves. The temperature of the maximum weight loss rate of samples was got from the DTG curves.

The thermal degradation of samples took place in a single main process for the acrylic PSAs in the temperature range 300–450°C with a weight loss of 94.3%. This might be due to the fracture of the acrylic main chains and its thermal dissociation to small molecules. After the decomposition of 550°C, the acrylic PSAs left few residue as shown in Table IV. Because of stable heteroaromatic rings of heat-resistance monomer, the residue char yields increased with the increasing of heat-resistance monomer content.

Compared to uncrosslinked acrylic PSAs, the degradation temperature of the crosslinked acrylic PSA samples increased significantly with the content of heat-resistance monomer. It was observed from Figure 11 that the thermal degradation temperature of the acrylic PSAs maximally increases by 80° when incorporated with 7 and 5 wt % of heat-resistance monomer, while the uncrosslinked acrylic PSAs underwent a slow degradation up to 250°C with 5% mass loss. This might be due to some weak covalent bonds in chains. The peak decomposition temperature (T_{max}) of the crosslinked acrylic PSAs sample shifted to higher temperature with increasing heat-resistance monomer content. The results indicated that the thermal stability of the acrylic PSAs containing heteroaromatic ring was improved. It could be explained that the formation of three-dimensional networks of acrylic PSAs systems when added vinyl-TGIC, which may also decrease the flexibility of the main chains, restricted the formation of oligomers, and delayed the temperature of the thermal degradation. In short, the higher amount of heat-resistance monomer could increase the crosslinking density of the acrylic PSAs, thus the temperature of thermal dissociation increased.

CONCLUSIONS

In this work, a heat-resistance monomer denoted as triazine heterocyclic compound (TGIC-AA) was synthesized and applied into improving the thermal stability of solvent-based acrylic pressure sensitive adhesives (PSAs) through copolymerization with acrylic precopolymer. The changes in thermally stability and adhesion properties of the acrylic PSAs, mainly associated with the content of the special monomer with rigid structure. The molecular weight of acrylic PSAs increased with the increasing of the heat-resistance monomer content, which also caused a change in viscosity for extensive crosslinking points. The glass transition temperature was significantly improved for acrylic PSAs due to the strong and substantial crosslinking networks formed in the acrylic polymer chains. Both the tack and 180° peel strength increased by the increasing of the heat-resistance monomer content, then decreased because of restriction in mobility of molecular chains. The holding time was affected by the cohesive strength, so the 5 wt % crosslinkers were chosen properly. The thermal stability of the acrylic PSAs was improved obviously by adding the crosslinker (TGIC-AA) due to the triazine heterocyclic structure and extensive crosslinking points. The adhesive properties at high temperature were excellent owing to the formation of crosslinking network. The crosslinked acrylic PSAs with heat-resistance monomer kept effective at 150°C. The acrylic PSAs can be placed in condition of high temperature and there was no residue on the surface, which would have various promising practical application. Some disadvantages cannot be ignored that the storage time was about 6 days because the isocyanurate groups of the acrylic PSAs may easily lead to the increase of viscosity and gel. The further study was needed.

REFERENCES

1. Sowa, D.; Czech, Z.; Byczynski, L. *Int. J. Adhes. Adhes.* **2014**, *49*, 38.

2. Zhang, L.; Cao, Y.; Wang, L.; Shao, L.; Bai, Y. *RSC Adv.* **2014**, *4*, 47708.

3. Lu, X.; Cao, G.; Niu, Z.; Pan, Q. *J. Appl. Polym. Sci.* **2014**, *131*, 40086.

4. Czech, Z. *Int. J. Adhes. Adhes.* **2006**, *26*, 414.

5. Vendamme, R.; Schuewer, N.; Eevers, W. *J. Appl. Polym. Sci.* **2014**, *131*, 40669.

6. Jovanovic, R.; Dube, M. A. *J. Macromol. Sci. Polym. Rev.* **2004**, *C44*, 1.

7. Sun, S.; Li, M.; Liu, A. *Int. J. Adhes. Adhes.* **2013**, *41*, 98.

8. Falsafi, A.; Tirrell, M.; Pocius, A. V. *Langmuir* **2000**, *16*, 1816.

9. Chung, H. S.; Park, G. H.; Kim, T.; Ahn, H.; Kim, D. H.; Chung, I. *Mol. Cryst. Liq. Cryst.* **2013**, *583*, 43.

10. Wang, J.; Lin, M.; Wang, C.; Chu, F. X. *J. Appl. Polym. Sci.* **2009**, *113*, 3757.

11. Yamaguchi, T.; Morita, H.; Doi, M. *Eur. Phys. J. E* **2006**, *20*, 7.

12. Yang, M. S.; Ko, S. W.; Choi, H. J. *J. Macromol. Sci. A: Pure Appl. Chem.* **2009**, *46*, 1142.

13. Czech, Z. *Polym. Int.* **2003**, *52*, 347.

14. Czech, Z. *Polimery* **2007**, *52*, 56.

15. Czech, Z. *Polym. Adv. Technol.* **2004**, *15*, 539.

16. Lee, S. W.; Lee, T. H.; Park, J. W.; Park, C. H.; Kim, H. J.; Song, J. Y.; Lee, J. H. *J. Electron. Mater.* **2014**, *43*, 4246.

17. Pang, B.; Ryu, C. M.; Kim, H. I. *J. Indus. Eng. Chem.* **2014**, *20*, 3195.

18. Baek, S. S.; Jang, S. J.; Lee, J. H.; Kho, D. H.; Lee, S. L.; Hwang, S. H. *Polym. Kor.* **2014**, *38*, 199.

19. Zhang, J. D.; Yang, M. J.; Zhu, Y. R.; Yang, H. *Polym. Int.* **2006**, *55*, 951.

20. Nakamura, Y.; Sakai, Y.; Imamura, K.; Ito, K.; Fujii, S.; Urahama, Y. *J. Appl. Polym. Sci.* **2012**, *123*, 2883.

21. Asada, K.; Fukano, K.; Yamashita, K.; Nakanishi, E. *J. Appl. Polym. Sci.* **2015**, *132*, 41444.

22. Anees, W.; Moore, V. C.; Croft, J. S.; Robertson, A. S.; Burge, P. S. *Med. Oxf.* **2011**, *61*, 65.

23. Sastre, J.; Carnes, J.; Garcia del Potro, M.; Manso, L.; Aguado, E.; Fernandez-Nieto, M. *Int. Arch. Occup. Environ. Health* **2011**, *84*, 547.

24. Gagne, S.; Carrier, M.; Aubin, S. *Rapid Commun. Mass Spectrom.* **2015**, *29*, 913.

25. Piirila, P.; Estlander, T.; Keskinen, H.; Jolanki, R.; Laakkonen, A.; Pfaffli, P.; Tupasela, O.; Tuppurainen, M.; Nordman, H. *Clin. Exp. Allergy* **1997**, *27*, 510.

26. Erikstam, U.; Bruze, M.; Goossens, A. *Contact Derm.* **2001**, *44*, 13.

27. Beyer, H.; Sajo, I.; Szegedi, A.; Vargha, V. *Eur. Polym. J.* **2007**, *43*, 2350.

28. Czech, Z.; Butwin, A.; Hefczyc, B.; Zawadiak, J. *Polimery* **2009**, *54*, 283.

29. Hariharan, D. U.S. Patent 5886071 (**1999**).

30. Sperling, L. H. In: Introduction to Physical Polymer; Wiley: New York, **1992**.

31. Tobing, S. D.; Klein, A. *J. Appl. Polym. Sci.* **2001**, *79*, 2230.

32. Czech, Z.; Wojciechowicz, M. *Eur. Polym. J.* **2006**, *42*, 2153.