

Polyacrylate emulsion containing IBOMA for removable pressure sensitive adhesives

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ABSTRACT: In the area of pressure sensitive adhesives (PSAs), corona treatment or other preprocessing on the carrier was commonly utilized for the enhancement of the interaction between the adhesive layer and the carrier, with complicated procedures and operations. To make it easy, one-pot emulsion polymerization using *n*-butyl acrylate, isobornyl methacrylate (IBOMA), 2-hydroxyethyl acrylate and acrylic acid was conducted in the presence of a reactive emulsifier SR-20 to obtain a reinforced adhesive tape. First, the emulsion had good stabilities, especially freeze-thaw stability, which was helpful to the fabrication of PSAs. Second, IBOMA increased the binding strength between adhesive layer and adherend. The properties of the final tapes well fulfilled the requirements of this areas, such as that the 180° peel strength of 125.9 N/m when coated on non-corona surfaces, the tack of 19 #, and the shear holding time of more than 100 h. Third, the tapes produced using the above emulsion could be removed many times, with no PSAs residue on the adherend even when coated on untreated PP, which showed the potential of recycling and environmental protection. Thus, these properties make the polyacrylate emulsion have great potential for practical applications. © 2015 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2016**, *133*, 42886.

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INTRODUCTION

Emulsion polymerization procedures are preferred due to the growing consciousness of safety and environmental protection, and the strict regulations regarding the removal of volatile organic compounds. Emulsion polymerization is a perfect candidate for preparing the polymers due to the emphasis on environmental-friendly.^{1–4} The kinetic stability of emulsions is quite important, hence, different emulsifiers are employed to enhance it.^{5–9} The traditional emulsifiers with low molecular weights were often utilized, with much negative effect on the stabilities of the emulsion, especially the freeze-thaw property. Furthermore, the traditional emulsifiers also have drawbacks when emulsion is coated and dried. The existence of emulsifier and the migration of emulsifier to interface between the coating and substrate make the properties worse. Therefore, reactive emulsifiers are good substitutes for traditional emulsifiers to reduce the detrimental effect.^{10–12} Some reactive emulsifiers are based on nonylphenol ethoxylates (NPEOs), which are restricted in many countries, because they are environmental endocrine disruptors. Hence, reactive emulsifiers with no nonylphenol groups should be employed.^{13–15}

Emulsion polymerization is a versatile industrial process, which is widely used for adhesives, paints, coatings, varnishes, etc.^{16–18} Especially, adhesives are now used in a broad range of scientific and industrial applications, such as tapes, labels, protective films, graphic films, medical dressing and other specialty applications.^{19–21} Pressure sensitive adhesives (PSAs) are particular adhesive materials that can firmly adhere to the various surfaces to form a strong bond under a light contact or no applied pressure without any activation.²⁰ They are aggressively and permanently tacky in a dry form at room temperature.^{21–24} The essential performance of PSAs is usually evaluated by tack, shear holding power and peel strength.^{25–27} High tack properties are needed to quickly adhere to adherend. Shear holding power should be high enough which can strengthen adhesion to avoid falling off when shear forces are applied. Peel strength of PSAs should be moderate in order to be easy removable for many times and to leave no residue on the adherend. For removable PSA tapes applications, the 180° peel strength should be lower than 160 N/m, while the tack should be more than 7 #, and the shear holding power should be more than 72 h. All the above parameters are the requirements in removable PSAs area.²⁸ Polyacrylate emulsions are often coated on carrier of polypropylene

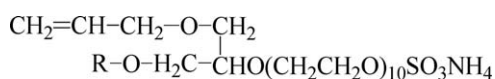
(PP) to obtain the PSA tapes, which has been commonly used in our daily life.^{29–34} While, some problems still exist. PP usually needs corona treatment or other preprocessing, because of its non-polarity and low surface energy. In adhesive area, corona treatment is most commonly applied to improve the polarity of PP carrier.^{35–40} However, the PP surface modified by corona discharge often undergoes severe degeneracy, especially in a damp environment.⁴¹ The binding strength between adhesive layer and carrier is not strong enough, which has drawbacks for PSAs. The adhesive layer may separate from the carrier during the adhesion process. Additionally, the cohesive failure occurs if the cohesive force is not strong enough. Hence, repeated adhesive ability was not good enough, in addition, left residue on the adherend.⁴² The ideal state is that there is only one separation between adhesive layer and adherend surface without any other failures when adhesives are peeled off from the adherend. According to above discussions, it's necessary to design and synthesize novel polyacrylate emulsion with high performances, to enhance the binding strength between adhesive layer and carrier of PP.

In this work, NPEO-free reactive emulsifier of SR-20 was adopted due to their environmental safety and good emulsification effectiveness. Additionally, IBOMA was introduced to directly increase the binding strength between adhesive layer and adherend. The peel strength of adhesives coated on untreated PP can reach 99.4% of that of corona PP. IBOMA also increases the cohesion force of the copolymer, which avoids the destruction of adhesive layer itself for its cohesive failure. There is no residue on the adherend after peeling off. The overall properties of the final product with the 180° peel strength of 125.9 N/m, the tack of 19 #, and the shear strength of more than 100 h are suitable to removable PSAs. The good adhesion properties and stabilities show that the resulting P(BA-IBOMA-2-HEA-AA) emulsion is suitable for practical applications.

EXPERIMENTAL

Materials

Reactive emulsifiers SR-10 and SR-20 were obtained from Adeka (Shanghai). The structure of SR-10 is as follows:



The structure of SR-20 is as follows:



Sodium dodecyl sulphate (SDS) was obtained from Shanghai Jingxi Chemical Technology. The monomers of n-butyl acrylate (BA) and acrylic acid (AA) were provided by Wuxi Jiani Chemical. 2-Hydroxyethyl acrylate (2-HEA) was purchased from Tokyo Chemical Industry. Isobornyl methacrylate (IBOMA) was obtained from Aladdin Industrial. Ammonium persulphate (APS) was purchased from Shanghai Lingfeng Chemical Reagent. NaHCO₃ was obtained from Shanghai Hongguang Chemical. Ammonia was provided by Shanghai Gaoxin Glassy Instrument. N,N-dimethylformamide (DMF) and NaCl were

obtained from Sinopharm Chemical Reagent. These materials were used as received. Deionized water (DW) was used throughout the experiment.

Emulsion Polymerization

The pre-emulsion, prepared at a stirring speed of 1200 rpm for 0.5 h, consisted of DW (30 g), reactive emulsifier of SR-10 or SR-20, or traditional emulsifier SDS (1.1 g), BA, IBOMA, AA (0.55 g) and 2-HEA (1.1 g). The monomers were total 55 g. Emulsion polymerizations were carried out in a flask which was dipped in a heated batch at 82°C. The stirring speed was 150 rpm for the following four procedures: 1, DW (25 g), APS (0.15) and NaHCO₃(0.11 g) were introduced into the flask and kept for 10 min. 2, The pre-emulsion and the initiator solution were added into the system by drop-wise addition which lasted for 2 h. The initiator solution consisted of DW (5 g) and APS (0.15 g). 3, The temperature was increased to 85°C and kept for 1 h to complete the polymerization. 4, The temperature was cooled to 40°C, and ammonia (0.1 g) was added into the flask.

Characterizations of Emulsion

Equipment for Characterizations. DSC test was carried out with a DSC 200 F3 apparatus (Netzsch, Germany) at a heating rate of 10 K/min in nitrogen atmosphere. The particle size of emulsion was measured using Mastersizer 2000 (Malvern Instruments, UK). Infrared spectrum was measured in the range from 4000 to 500 cm⁻¹ using a FT-IR spectrometer (Perkin Elmer, USA). Viscosity-average molecular weight of copolymer was determined by dilute solution viscometry using Ubbelohde viscometer (0.57 mm). NaCl solution was used for demulsification, and DMF was used as a solvent.

Stabilities. Stabilities of P(BA-IBOMA-2-HEA-AA) emulsion were tested under various conditions. The emulsion was mixed with 20 wt % of CaCl₂ solution, and then stored for 48h. The concentration of CaCl₂ solution was 5 wt %. The emulsion was also diluted to 3 wt % of solids content, and then stored for 72 h. Stability under mechanical stirring was tested at a stirring speed of 1500 rpm for 3 h. The test of resistance to freeze-thaw was realized at -10°C in a freezer for 24 h. And storage stability was tested by keeping properly at room temperature for 6 months.

Preparation of PSA Tapes

Emulsion was coated onto a 110 μm polypropylene (PP) film to get a 125-μm-dry-thickness PSA tape. The tapes were dried in an oven at 90°C for 2 min.

Characterizations of PSA Tapes

Tack Properties. A 100 mm × 100 mm PSA tape was cut for the test of tack. Steel balls with different sizes were released from the top and rolled onto the tape. The angle of the slope was 30°. The serial number of the biggest ball which stopped on the tape was recorded.

180° Peel Strength. A tape of 25 mm in width was cut and laminated onto the steel sheet using a rubber coated roller. After a dwelling time of 20 min, the 180° peel strength was tested at a tape distance of 75 mm by a tensile machine at a test speed of 300 mm/min.

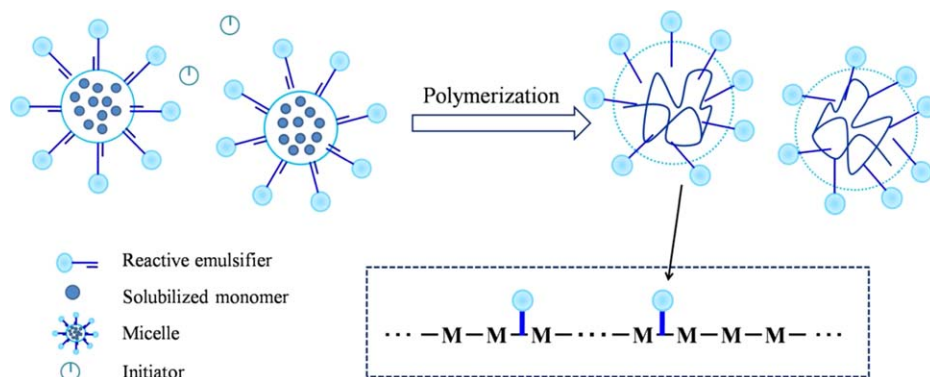


Figure 1. Scheme of emulsion polymerization in the presence of reactive emulsifier. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Shear Holding Power. A tape of 25 mm × 70 mm was prepared. An area of 25 mm × 25 mm was laminated onto the steel sheet using a rubber coated roller, and the rest for the other. A 1 kg load was placed at the end of the sample after a dwelling time of 1 h. The holding time between the start of loading and the complete separation was recorded automatically.

RESULTS AND DISCUSSION

Effect of Different Emulsifiers

Effect of Different Emulsifiers on the Stabilities of Emulsions. Pre-emulsion with traditional emulsifier SDS had obvious stratifications after standing at room temperature for 2 h, while pre-emulsion with reactive emulsifiers had no obvious variation. The stability of pre-emulsion using reactive emulsifier was much better than that of the pre-emulsion using SDS.

Stabilities of emulsions synthesized with different kinds of emulsifiers were tested under various conditions, including with calcium ions, dilution, mechanical stirring, freeze-thaw and storage. The obtained results show that emulsions polymerized with reactive emulsifiers had good stabilities, especially freeze-thaw stability. Emulsion with SDS froze after keeping in the freezer, and it cannot thaw, even heated at 60°C for 8 h. While the emulsion with SR-10 can thaw at the room temperature with no demulsification phenomenon, after keeping at −10°C for 24 h. The emulsion synthesized in the presence of SR-20 can still flow with no obvious variation after kept at −10°C for 24 h, showing that emulsion synthesized in the presence of SR-20 has better freeze-thaw stabilities than the emulsions with other emulsifiers.

There are mainly four reasons to explain the good stabilities of emulsion synthesized in the presence of SR-20. First, SR-20 has an allyl group, which can polymerize with acrylate monomers.²⁶ The scheme of emulsion polymerization in the presence of reactive emulsifier is shown in Figure 1. The reactive emulsifier has become one part of the polymer. Second, stern layer was formed on the surface of emulsion particles, due to the negative charges supplied by SR-20. Then, positive ions were absorbed around the stern layer because of the electrostatic attraction. Partial

ions of absorbed layer diffused into the surrounding medium, which made the charges generated. The formation of electric double layers is helpful for stabilities of emulsion due to the electrostatic repulsion. Third, the other reason is that the emulsion particles cannot get too close to each other due to the molecular volume of SR-20. Additionally, the stabilities of the emulsion can be enhanced by inhibiting the movement of segments due to the structure of SR-20. In a word, this stable emulsion can be utilized in many PSA applications.

The average particle size of P(BA-IBOMA-2-HEA-AA) emulsion was 198 nm with good monodispersity (Figure 2). The interactions between the EO segments of SR-20 with intermolecular hydrogen bonds made double electrode layer more stable. It also prohibited the particles from getting close to each other. In this work, the monomers were dispersed sufficiently at a high stirring speed of 1200 rpm when prepared the pre-emulsion. The drops immediately reacted during the drop-wise process, since part of APS was added into the flask previously. Thus, there was a continuous formation of new emulsion particles during the polymerization process. The polydispersity index (PDI) was 0.47. The size distribution with the average diameter of 198 nm was suitable for practical applications.

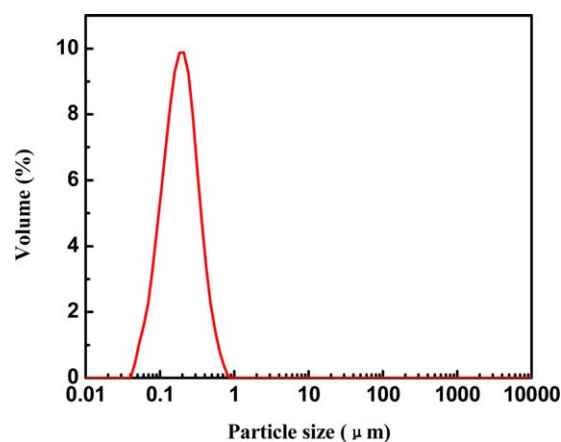


Figure 2. Size distribution by volume of P(BA-IBOMA-2-HEA-AA) emulsion. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table I. Properties of PSA Tapes with Different Emulsifier Types

Emulsifier type	Emulsifier	Tack (#)	180° peel strength (N/m)	Holding time (h)
Traditional emulsifier	SDS	3	66.4	2.4
Reactive emulsifier	SR-10	15	96.2	>100
Reactive emulsifier	SR-20	19	126.7	>100

Effect of Different Emulsifiers on Adhesion Properties of PSAs. Table I shows the adhesion properties of PSA tapes produced from P(BA-IBOMA-2-HEA-AA) emulsion with different emulsifiers. The properties of PSA tapes with reactive emulsifiers were much better than the PSA tapes with SDS. The holding time of emulsions was more than 100 h, which was much longer than that of traditional emulsion (6.6 h).

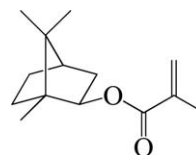
Figure 3 shows the schematic image of PSA tapes prepared from emulsion using different emulsifiers. Generally, traditional emulsifiers with low molecular weight are needed in normal polymerization method. The phenomenon of slipping will occur when there is the migration of redundant emulsifier from the PSAs to adhesive surface, which makes the emulsion exhibit worse adhesive properties and stabilities.^{10–12} SR-10 and SR-20 are anionic reactive emulsifiers, which can easily react with acrylate monomers and become a part of polymers without migration (Figure 3b). Therefore, the adhesion properties of emulsion PSAs using reactive emulsifier are much better than those of the other PSAs.

Hence, the tack and 180° peel strength of PSAs tapes using SR-20 are better than those of SR-10. There are more EO (CH₂CH₂O) units of SR-20 compared with SR-10, and the molecular volume of SR-20 is much larger, which curbs the motion of chain units, thus leading to the good adhesion properties. Therefore, the adhesion properties of PSA tapes prepared with SR-20 as a reactive emulsifier are much better than those of PSA tapes prepared with other emulsifiers.

Effect of IBOMA

Effect of Different Monomers. Table II shows the properties of PSAs coated on corona and untreated PP. For P(BA-2-HEA-AA), the holding time and tack properties decreased when coated on untreated PP. Although there was no palpable residue

adhesive when peeled off from the adherend, the cohesive of the adhesive, the binding strength between adhesive layer and PP surface were not strong enough. There was damage to the adhesive layer, which increased the record of peel strength. MMA and IBOMA can supply cohesive strength, which ensured that there was no damage to the adhesive layer when it was peeled off. Hence, the adhesion properties of P(BA-IBOMA-2-HEA-AA) were much better than those of P(BA-2-HEA-AA) and P(BA-MMA-2-HEA-AA). When PSAs were coated on untreated PP, the 180° peel strength was 125.9 N/m, which reached 99.4% when coated on corona PP (125.1 N/m). The tack was 19 #. And the shear holding time was more than 100 h. P(BA-IBOMA-2-HEA-AA) can supply good adhesion properties for practical applications even on the non-corona surface. The required removable adhesion properties can be realized. That is because IBOMA contains a large number of methylenes, which is structurally similar to PP carrier. The structure of IBOMA is as follows:



There is a pendant isobornyl group in the structure. The bulky cyclic can increase the space of the chains and weaken the applied force of the molecular chains, thus leading to the increase of chain flexibility. The isobornyl groups of the adhesive layer move to PP carrier. Therefore, the molecule was mutually entangled in the interface layer, which made a positive impact in the bonding process. It gives a strong interaction between the adhesive layer and the carrier, due to existence of IBOMA.

On one hand, the bond between the adhesive layer and carrier layer depends on Van der Waals force. The thermodynamic conditions for adhesive permeation were fulfilled by the thermodynamic movement of polymer molecular or chains.³⁴ And the molecule movement was reinforced under the heating condition during the drying process of PSA tapes, thus leading to the kinetic conditions for molecule diffusion. Moreover, there was a migration of nonpolar main chains going into the PP carrier caused by the molecule diffusion, while the branched chains of adhesive molecule moved to the coating surface.³⁸ Additionally, IBOMA increased the interaction between the adhesive and the PP carrier. On the other hand, chemical bond between adhesive and the carrier did not exist.³⁹ Besides, the mechanical bonding force could be ignored, since the carrier surface was not pretreated before coating.⁴⁰ Therefore, the bonding effect between

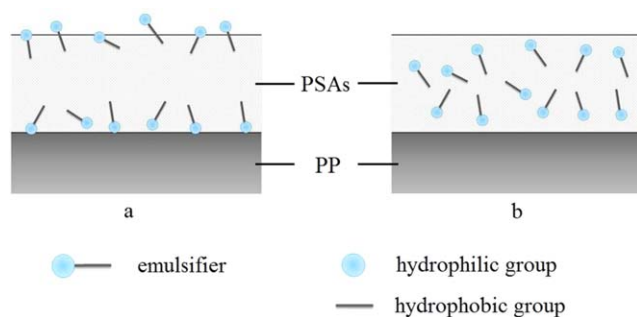


Figure 3. Scheme of PSA tapes of different types (a. PSA tapes prepared using traditional emulsifier; b. PSA tapes prepared using reactive emulsifier). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table II. Properties of PSAs Coated on Corona Treated and Untreated PP

PSAs	PP surface	Tack (#)	180° peel strength (N/m)	Holding time (h)
P(BA-2-HEA-AA)	Corona treated	19	92.1	>100
	Untreated	18	106.5	39.7
P(BA-MMA-2-HEA-AA)	Corona treated	19	109.4	>100
	Untreated	19	91.0	46.2
P(BA-IBOMA-2-HEA-AA)	Corona treated	19	126.7	>100
	Untreated	19	125.9	>100

the adhesive and the PP carrier included intermolecular van der Waals force, entanglement force after diffusion and infiltration.

Effect of the Amount of IBOMA. The glass transition temperatures (T_g) of pure P(BA-2-HEA-AA) is -47.6°C , shown in Figure 4. When 2 wt % and 4 wt % of IBOMA was added into the system, The T_g of P(BA-IBOMA-2-HEA-AA) increased to -44.8°C and -42.6°C , respectively. We can see that the T_g value

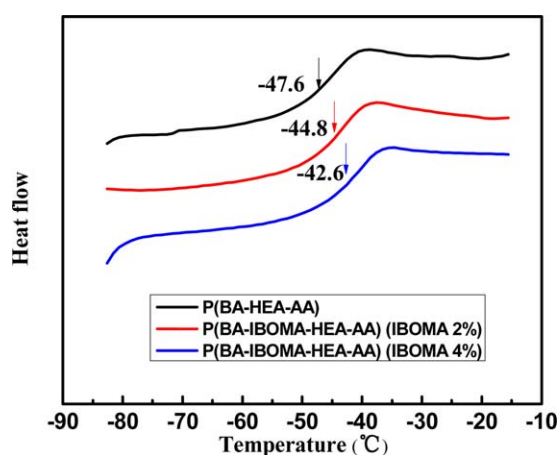


Figure 4. DSC thermograms of polyacrylates. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

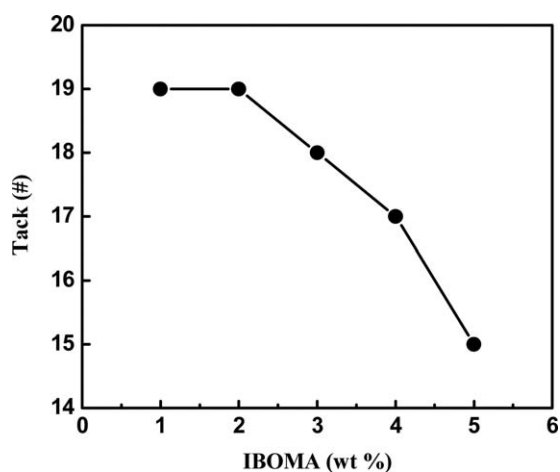


Figure 5. Effect of the amount of IBOMA on tack of PSA tapes.

of P(BA-IBOMA-2-HEA-AA) increased with the growth of the percentage of IBOMA ($T_g = 155^\circ\text{C}$) added into the system. For PSAs applications, the T_g of the polymers should be low, ranging from -60 to -20°C . In this work, the T_g of the polymers were pretty suitable for practical applications. The properties of PSAs depended on adhesion and cohesion. The cohesion could be enhanced by IBOMA, since the T_g value of P(BA-IBOMA-2-HEA-AA) was higher than that of P(BA-2-HEA-AA). Hence, good adhesion properties can be obtained with a small amount of IBOMA.

As is shown in Figure 5, the PSA tapes had excellent tack properties. The tack of PSAs coated on non-corona PP was the same as that when they were coated on corona PP using the same amount of IBOMA. PSA tapes with the high tack properties can quickly adhere to adherend. The tack was 19 # when the amount of IBOMA was less than 2 wt %. After that, the tack properties decreased gradually with the addition of IBOMA. And it was 15 # with IBOMA of 5 wt % in monomers. Too much IBOMA reduced the adhesion of PSAs.

Adhesion and cohesion are two significant factors to judge the properties of PSAs. IBOMA can enhance the cohesion of PSAs, which can be proved by the data of the 180° peel strength (Figure 6). The 180° peel strength increased with the growth of IBOMA amount when the amount of IBOMA was less than 2%.

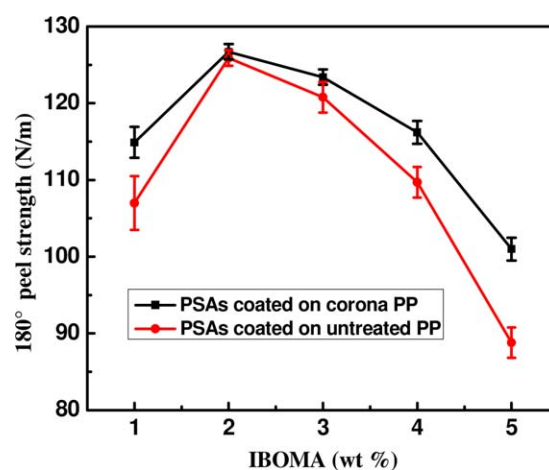


Figure 6. Effect of the amount of IBOMA on 180° peel strength of PSA tapes. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

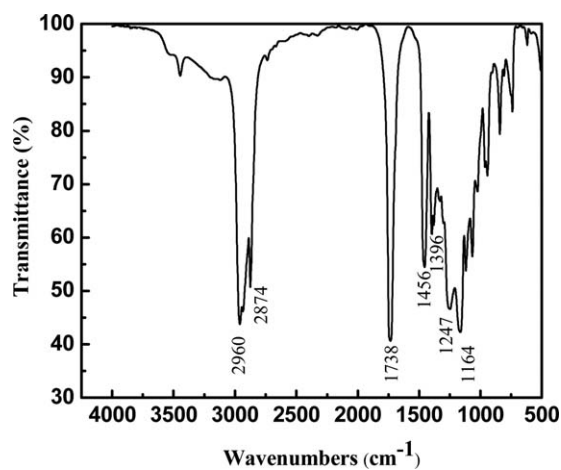


Figure 7. FT-IR spectrum of P(BA-IBOMA-2-HEA-AA).

The maximum value reached 126.7 N/m, when the amount of IBOMA was 2 wt %. Similarly, there was a peak value of 125.9 N/m of 180° peel strength, when PSAs were coated on untreated PP. After that, the 180° peel strength declined with the increase of IBOMA amount. IBOMA can supply the sufficient cohesive strength, which ensures that there is no damage to the adhesive layer when it is peeled off. The cohesion was enhanced with the growth of the amount of IBOMA. However, too much IBOMA weakened the adhesion of PSAs. It had a bulky cyclic functional group, which can curb the motion of chain segments. The rigid polymer chains reduced the adhesion properties.

The shear holding time of PSA tapes were more than 100 h. There was no slip phenomenon, since no traditional emulsifier with low molecular weight was used. The holding time of PSA tapes was also more than 100 h when PSAs were coated on untreated PP surface, since IBOMA enhanced the cohesion of PSAs. The sufficient cohesion strength helped to prevent PSAs from falling off when shear forces were applied.

Based on the above results, the optimal amount of IBOMA is 2 wt %. The PSA tapes produced from the resulting P(BA-IBOMA-2-HEA-AA) emulsion can be recycled many times, and there is no PSAs residue on the adherend. Hence, the requirements of practical applications can be well fulfilled by the P(BA-IBOMA-2-HEA-AA) emulsion with the good adhesion properties.²⁸

Viscosity-Average Molecular Weight

Viscosity was measured by Ubbelohde viscometer. The relative viscosity η_r can be defined as:

$$\eta_r = t/t_0$$

where, t is the efflux time of polymer solution, and t_0 is the efflux time of pure solvent.

The specific viscosity $[\eta]$ can be obtained from $\eta_{sp}/c \sim c$. η_{sp} can be expressed as

$$\eta_{sp} = \eta_r - 1$$

In this work, $[\eta] = 39.9 \text{ cm}^3/\text{g}$. The viscosity-average molecular weight M_η can be calculated by the following equation:

$$M_\eta = \sqrt[\alpha]{\frac{[\eta]}{K}}$$

where, $K = 7.4 \times 10^{-3} \text{ cm}^3/\text{g}$, and $\alpha = 0.73$.⁸

The viscosity-average molecular weight of P(BA-IBOMA-2-HEA-AA) was 1.3×10^5 .

Analysis of FTIR

The significant peak at 1738 cm^{-1} suggested the stretch vibration of C=O, in Figure 7. Peaks at 1247 cm^{-1} and 1164 cm^{-1} corresponded to C—O group. While, peaks at 2960 cm^{-1} and 2874 cm^{-1} were related with the stretch vibration of C—H group, and peaks at 1456 cm^{-1} and 1396 cm^{-1} were attributed to the bend vibration of C—H group. In addition, the characteristic C=C adsorption near $1650 - 1580 \text{ cm}^{-1}$ disappeared, indicating that C=C contained in SR-20 and other monomers was reacted and consumed.

CONCLUSIONS

In this work, a simple method for the synthesis of P(BA-IBOMA-2-HEA-AA) emulsion was successfully utilized for the removable PSAs applications. It was easy to obtain emulsion with good stabilities under different conditions, such as calcium ions, dilution, mechanical stirring, low-temperature resistance and storage, compared with emulsions of P(BA-2-HEA-AA) or P(BA-MMA-2-HEA-AA). The introduction of IBOMA directly improved the binding strength between the adhesives and the adherend. Additionally, all the properties of the tapes, such as the 180° peel strength, the shear holding time, and the tack, can fulfill the requirements of removable PSAs very well. Thus, the PSA tapes produced from the P(BA-IBOMA-2-HEA-AA) emulsion have excellent potential for the practical applications.

REFERENCES

1. Yu, C. L.; Zhang, F. A.; Zhu, X. H. *Polym. Adv. Technol.* **2009**, *20*, 347.
2. Jahanzad, F. *J Appl. Polym. Sci.* **2010**, *117*, 84.
3. Dobie, C. G.; Boodhoo, K. V. K. *Chem. Eng. Process.* **2010**, *49*, 901.
4. Blom, H. P.; Gauthier, M.; Li, K.; Nielsen, K. E. *J. Appl. Polym. Sci.* **1998**, *70*, 227.
5. Wang, J.; Zeng, X. R.; Li, H. Q. *J. Coat. Technol. Res.* **2010**, *7*, 469.
6. Unzué, M. J.; Schoonbrood, H. A. S.; Asua, J. M.; Goni, A. M.; Sherrington, D. C.; Stähler, K.; Goebel, K. H.; Tauer, K.; Sjöberg, M.; Holmberg, K. *J. Appl. Polym. Sci.* **1997**, *66*, 1803.
7. Xu, Z. S.; Yi, C. F.; Lu, G. H.; Zhang, J. Z.; Cheng, S. Y. *Polym. Int.* **1997**, *44*, 149.
8. Zhang, L.; Cao, Y. J.; Wang, S.; Crech, Z.; Shao, L.; Bai, Y. P. *Int. J. Adhes. Adhes.* **2013**, *47*, 69.
9. Zhang, J. Z.; Zou, Q. C.; Li, X. Q.; Cheng, S. Y. *J. Appl. Polym. Sci.* **2003**, *89*, 2791.
10. Abele, S.; Graillat, C.; Zicmanis, A.; Guyot, A. *Polym. Adv. Technol.* **1999**, *10*, 301.

11. Hu, A. X.; Yao, Z. G.; Yu, X. *J. Appl. Polym. Sci.* **2009**, *113*, 2202.
12. Zhu, M. Y.; Qiao, W. H.; Liu, H. Z.; Sun, Y. L. *J. Appl. Polym. Sci.* **2008**, *107*, 624.
13. Fernandez, A. M.; Held, U.; Willing, A.; Breuer, W. H. *Prog. Org. Coat.* **2005**, *53*, 246.
14. Betton-Perez, C.; Li, Y. Z.; Ruiz, J. P. Proceedings of the Thirty Sixth Annual International Waterborne, High-Solids, and Powder Coatings Symposium, New Orleans, Louisiana, February 18–20, **2009**.
15. Hosaka, M.; Shirai, H.; Tsukuhare, N. *Paint Coat. Ind.* **2013**, *9*, 72.
16. Kohut-Svelko, N.; Pirri, R.; Asua, J. M.; Leiza, J. R. *J. Polym. Sci. Part A: Polym. Chem.* **2009**, *47*, 2917.
17. Liu, Z. G.; Han, Y.; Zhou, C.; Zhang, M. Y.; Li, W. M.; Zhang, H. X.; Liu, F. Q.; Liu, W. J. *Ind. Eng. Chem. Res.* **2010**, *49*, 7152.
18. Agirre, A.; Nase, J.; Degrandi, E.; Creton, C.; Asua, J. M. *Macromolecules* **2010**, *43*, 8924.
19. Shen, H. Z.; Zhang, J. Y.; Liu, S. J.; Liu, G. D.; Zhang, L. Q.; Qu, X. W. *J. Appl. Polym. Sci.* **2008**, *107*, 1793.
20. Sakdapipanich, J.; Thananusont, N.; Pukkate, N. *J. Appl. Polym. Sci.* **2006**, *100*, 413.
21. Pocius, A. V. in *Polymer Science: A Comprehensive Reference*; Matyjaszewski, K.; Möller, M.; Elsevier: Waltham, **2012**; Vol. 8, p 305.
22. Lutz, H.; Weitzel, H.-P.; Huster, W. In *Polymer Science: A Comprehensive Reference*; Matyjaszewski, K.; Möller, M., Eds.; Elsevier: Waltham, **2012**; Vol. 10, p 479.
23. Zhu, J.; Zhang, X. Y.; Gao, F.; Chen, M.; Huang, S. Q. *J. Polym. Res.* **2011**, *18*, 1991.
24. Fonseca, G. E.; Mckenna, T. F.; Dubé, M. A. *Chem. Eng. Sci.* **2010**, *65*, 2797.
25. Qie, L. L.; Dubé, M. A. *Int. J. Adhes. Adhes.* **2010**, *30*, 654.
26. Zhang, L.; Cao, Y. J.; Wang, L.; Shao, L.; Bai, Y. P. *RSC Adv.* **2014**, *4*, 47708.
27. Benedek, I. In *Pressure-Sensitive Adhesives and Applications*; Marcel Dekker: New York, **2004**; ch.1, p 1.
28. Zhang, L.; Cao, Y. J.; Wang, L.; Shao, L.; Bai, Y. P. *RSC Adv.* **2015**, *5*, 36288.
29. Marcais, A.; Papon, E.; Villenave, J. J.; Tordjeman, P.; Pirri, R.; Gerard, P. *Macromol. Symp.* **2000**, *151*, 497.
30. Plessis, B.; Arzamendi, G.; Alberdi, J. M.; Agnely, M.; Leiza, J. R.; Asua, J. M. *Macromolecules* **2001**, *34*, 6138.
31. Roberge, S.; Dubé, M. A. *Polymer* **2006**, *47*, 799.
32. Li, A. L.; Li, K. C. *RSC Adv.* **2014**, *4*, 21521.
33. Moghbeli, M. R.; Zamir, S. M.; Molaei, B. *J. Appl. Polym. Sci.* **2008**, *108*, 606.
34. Xu, K.; Selby, J. C.; Shannon, M. A.; Economy, J. *J. Appl. Polym. Sci.* **2004**, *92*, 3843.
35. Sun, C.; Zhang, D.; Wadsworth, L. C. *Adv. Polym. Technol.* **1999**, *18*, 171.
36. Guimond, S.; Radu, I.; Czeremuskin, G.; Carlsson, D. J. *Plasmas. Polym.* **2002**, *7*, 71.
37. Novak, I.; Pollak, V.; Chodak, I. *Plasma Process. Polym.* **2006**, *3*, 355.
38. Malkapuram, R.; Kumar, V.; Negi, Y. S. *J. Reinf. Plast. Compos.* **2009**, *28*, 1169.
39. Tan, H. S.; Li, L.; Chen, Z. N.; Song, Y. H.; Zheng, Q. *Polymer* **2005**, *46*, 3522.
40. Manchado, M. A. L.; Arroyo, M.; Biagiotti, J.; Kenny, J. M. *J. Appl. Polym. Sci.* **2003**, *90*, 2170.
41. Akbari-Moghanjoughi, M.; Esfandyari-Kalejahi, A. *J. Plasma Phys.* **2013**, *79*, 1081.
42. Plessis, C.; Arzamendi, G.; Agnely, M.; Leiza, J. R.; Asua, J. M. *J. Polym. Sci. Part A: Polym. Chem.* **2002**, *40*, 2878.