

HIGH PERFORMANCE WATERBORNE ACRYLIC PSAS FOR BUILDING AND CONSTRUCTION APPLICATIONS

Keith Andes, Senior Research Chemist, Arkema Coating Resins, Cary NC
Keith Olesen, Field Marketing Manager, Arkema Coating Resins, Cary NC
Dr. Wenjun Wu, Senior Principal Scientist, Arkema Coating Resins, Cary, NC

Abstract

Acrylic polymers have long been used to manufacture pressure sensitive adhesives (PSAs) for a variety of applications including tapes, labels, decorative and protective films. In the last decade, acrylic PSA tapes have found increased utility in building and construction for joining, sealing and/or thermal protection.

Acrylic adhesives can be either solvent- or water-based, and are prepared by reacting monomers with the desired properties, which are then typically crosslinked and/or tackified for specific end-use applications. Waterborne PSAs have become more attractive than solvent-based acrylics due to advantages such as health and environmental benefits, high solids content, and ease of handling. However, waterborne adhesives have struggled to meet the high performance demands of some specialty tape markets. The adhesion and cohesion balance is critically important as most PSA applications require high shear strength together with good peel and tack properties. Solvent-borne PSAs can be formulated to have higher cohesive strength than acrylic emulsion counterparts owing to their continuous film morphology. It is thus an even greater challenge for waterborne acrylic PSAs to achieve a high Shear Adhesion Failure Temperature (SAFT) for high-temperature applications where PSA tapes are subject to extreme temperature and harsh environmental conditions.

Many factors affect the three performance criteria of a waterborne acrylic PSA: tack, peel and shear strength. This paper will show that desired physical and rheological properties can be achieved by careful selection of the chemical components and control of polymer molecular weight. The resulting acrylic emulsion polymer features balanced adhesion and cohesion properties and is suitable for high-temperature applications such as HVAC duct tapes and house wrap.

Introduction

Pressure-sensitive adhesives (PSAs) are viscoelastic materials. They are permanently tacky and will adhere to a substrate under light pressure. Acrylic polymers have long been used to manufacture PSAs for a variety of applications including tapes, labels, decorative and protective films. In the last decade, acrylic PSA tapes have found increased utility in building and construction for joining, sealing and/or thermal protection.

Acrylic PSAs can be either solvent- or water-based, and are prepared by reacting monomers with the desired properties, which are then typically crosslinked and/or tackified for specific end-use applications. PSA performance has been shown to depend on bulk properties such as mechanical properties and viscoelastic properties.¹ These bulk properties are affected by the adhesives' glass transition temperature (T_g), morphology, and microstructure, such as molecular weight, degree of branching or cross-linking and gel content.²⁻⁷

To achieve desired performance, acrylic PSAs are produced using a combination monomers. The monomer mixture will typically include; (i) a "soft" monomer such as butyl acrylate (BA) or 2-ethyl hexyl acrylate (2-EHA), to impart good deformability and flowability to the PSA; (ii) a "hard" monomer such as methyl methacrylate (MMA), styrene (S) or vinyl acetate (VAM), to endow the PSA with sufficient cohesive strength and; (iii) a set of "functional" monomers such as methacrylic acid (MAA) or acrylic acid (AA) and 2-hydroxy ethyl methacrylate (HEMA), to improve adhesion characteristics to certain substrates.² The term "soft" monomer is meant to describe monomers whose corresponding homopolymer would exhibit a glass transition temperature less than -20°C. Conversely, "hard" is meant to describe monomers whose corresponding homopolymer would exhibit a glass transition temperature greater than +20°C.

Due to the importance of polymer microstructure, much work has been done to manipulate it by varying polymerization conditions such as polymerization temperature and monomer addition polices, or including a chain transfer agent (CTA) and/or cross-linkers.⁸⁻¹² The use of chain transfer agents and/or cross-linker are the most effective in altering polymer molecular weight and gel content.

Waterborne PSAs have become more attractive than solvent-based acrylics due to advantages such as health and environmental benefits, high solids content, and ease of handling. However, waterborne adhesives have struggled to meet the high performance demands of some specialty tape markets. The adhesion and cohesion balance is critically important as most PSA applications require high shear strength together with good peel and tack properties. Solvent-borne PSAs can be formulated to have higher cohesive strength than their acrylic emulsion counterparts owing to their continuous film morphology.¹³ It is thus an even greater challenge for waterborne acrylic PSAs to achieve a high Shear Adhesion Failure Temperature (SAFT) for high-temperature applications where PSA tapes are subject to extreme temperature and harsh environmental conditions.

Many factors affect the three performance criteria of a waterborne acrylic PSA: tack, peel and shear strength. This paper will show that desired physical and rheological properties can be achieved by careful selection of the chemical components and control of polymer molecular weight. The resulting acrylic emulsion polymer features balanced adhesion and cohesion properties and is suitable for high-temperature applications such as HVAC duct tapes and house wrap.

Experimental

Materials

The experimental adhesives were prepared via conventional semi-batch emulsion polymerization in three-liter, jacketed glass reactors equipped with dual impellers, reflux condensers and stainless steel feed lines. The feed rates and temperature profiles were controlled manually. The polymerization temperature was controlled $\pm 1^{\circ}\text{C}$ utilizing a temperature controlled water bath. All raw materials were used as received from the supplier.

Commercial tapes were purchased from Home Centers.

Methods

Molecular weight and Gel Content Determination

The dried latex sample was submerged in tetrahydrofuran (THF) for a minimum of 48 hours. The percentage of insoluble fraction was determined gravimetrically. The molecular weight and molecular weight distribution of the soluble fraction were analyzed using gel permeation chromatography (GPC). Mono-dispersed polystyrene standards, with molecular weight ranging from 580 to 7,500,000 Dalton, were used to generate the calibration curve.

Complex Modulus Measurement

The PSA samples were prepared by drying approximately 27g of the adhesive in 10 cm x 10 cm silicone release paper dishes. The adhesive was dried in a 50°C oven for 7 days. A 1 cm x 1 cm square of the adhesive was then cut from the dried film.

The viscoelastic properties of the adhesives were measured using a Discovery Hybrid Rheometer dynamic mechanical analyzer from TA Instruments. Measurements were conducted at 25°C using 8 mm stainless steel parallel plates with a gap setting of $1.5 \pm 0.2\text{mm}$. The frequency sweep measurements were performed over a frequency range of about 0.002–60 Hz, under a compression mode and with a strain of 1%.

PSA Sample Preparation

All samples were prepared by direct coating the emulsion onto 2-mil polyester, and covering the sample with a release liner. Unless otherwise specified, the dried sample had a coat weight of $1.5 \pm 0.1\text{ g}/100\text{in}^2$. All data reported are the average of three individual tests.

180° Peel Adhesion

Peel tests were performed following PSTC-101 Test Method A in which a strip of tape is applied to a standard test panel with controlled pressure. The tape is peeled from the panel at 180° angle at a specified rate, during which time the force required to effect peel is measured. Two different dwell times were employed; 30 min (initial) and 24 hrs, on both stainless steel, aluminum and HDPE substrates.

Loop Tack

Loop tack was measured using the PSTC-16 Test Method B which involves allowing a loop of pressure sensitive adhesive with its backing to be brought into controlled contact with a 24 mm x 24 mm (one square inch) surface of stainless steel, with the only force applied being the weight of the pressure sensitive article itself. The pressure sensitive article is then removed from the substrate, with the force to remove the pressure sensitive article from the adherend measured by a recording instrument.

Shear Adhesion

Shear adhesion test were conducted following PSTC-107 in which a strip of tape is applied to a standard steel panel under controlled roll down. The panel is mounted vertically, a standard mass is attached to the free end of the tape and the time to failure is determined. Instead of the standard 23°C and 50% R.H., the testing was conducted at 65°C.

Shear Adhesion Failure Temperature (SAFT)

SAFT adhesion tests were performed following PSTC-17 in which a strip of tape is applied to a standard steel panel under controlled roll down. The panel is mounted vertically in a programmable convection oven, a standard mass is attached to the free end of the tape, the oven temperature is increased at a constant, controlled rate, and the time to failure is determined. The shear adhesion failure temperature is the temperature at which the bond area fails in shear when a sample is subjected to a standard load.

Results and Discussion

Several commercial tapes were purchased to benchmark their adhesive characteristics. Products were selected from UL181 type HVAC and weather barrier house wrap seam sealing categories. Dramatic differences were observed in the adhesive characteristics of these two disparate classes of tape (see Table 1). The HVAC type tape displayed much higher cohesive strength as manifest by higher shear adhesion performance. Both the elevated temperature status shear test and the SAFT values were significantly greater than those exhibited by the weather barrier house wrap tape. Conversely, the peel and loop tack adhesion of the weather barrier house wrap tape were significantly higher than the HVAC type on all substrates.

The differences in the adhesive characteristics of these two classes of tape are perhaps not unexpected given the different conditions under which they are expected to perform. The lower peel and loop tack adhesion of the HVAC tape would allow for repositioning by the installer. The higher SAFT performance would ensure that the tape remained in place even with heating of the duct work due to high temperature air flow. The higher peel adhesion of the weather barrier house wrap tapes would resist debonding during wind gusts, and the lower SAFT performance may simply be the consequence of designing a pressure sensitive adhesive which may be applied at lower temperatures.

Table 1. Adhesive characteristics of commercial tapes

Sample ID	Coat Weight (g/100 in ²)	180° Peel Adhesion (lbs/in)			Loop Tack (lbs/in ²)			Shear Adhesion	
		SS	AI	HDPE	SS	AI	HDPE	@ 65°C	SAFT
HVAC 1	2.6	2.6	3.3	3.2	2.4	2.4	2.0	>48 hrs	121+/-7°C
HVAC 2	3.8	4.5	3.8	3.7	3.6	2.1	4.1	>48 hrs	184+/-1°C
HVAC 3	5.0	4.4	4.1	3.9	4.0	3.3	3.0	>48 hrs	134+/-2°C
House Wrap 1	2.5	7.0	6.4	6.4	3.9	3.1	3.6	<0.1 hrs	<40°C
House Wrap 2	2.7	3.7	6.8	6.3	3.6	3.2	3.2	2.4+/-0.6 hrs	92+/-5°C

To explore the impact of polymer design on the balance of adhesive and cohesive characteristics two experimental pressure sensitive adhesives were prepared. A mixture of “hard” and “soft” monomers were employed to achieve a polymer with a glass transition temperature of -44°C. The molecular weight and gel content were varied through the use of an appropriate chain transfer agent and/or cross-linker. The design variables of the two experimental polymers are outlined in Table 2.

Table 2. Composition of experimental polymers

Sample ID	CTA	Cross-linker
Polymer 1	✓	
Polymer 2		✓

The gel content and molecular weight of the soluble fraction of the two experimental polymers are presented in Table 3. The elimination of chain transfer agent and the inclusion of cross-linker in the recipe increased the THF insoluble fraction. The number average molecular weight of the soluble fraction was similar for the two samples. A decrease in the polydispersity (M_w/M_n) was observed in the sample utilizing cross-linker.

Table 3. Molecular weight and Gel fraction of experimental polymers

Sample ID	Sol Fraction Molecular Weight		Gel Fraction
	Mn	Mw	
Polymer 1	28,193+/-6	103,354+/-2592	52+/-2
Polymer 2	28,764+/-19	69,844+/-1794	69+/-0

Pressure sensitive adhesives and other polymeric materials (because of their viscoelastic nature) exhibit temperature and time dependent behavior during deformation and flow. Table 4 shows a summary of the rheological properties that indicate specific adhesive behavior using dynamic mechanical properties.¹⁴

Table 4. Viscoelastic Properties Related to PSA Characteristics

Mechanical test	Rheological behavior	Performances
TACK	Low $\text{Tan } \delta$ peak and low G' Low cross-links ($G'' > G'$) at approx. 1 Hz	HIGH TACK
SHEAR resistance	High G' at low frequencies (< 0.1 Hz) High viscosity at low shear rates	HIGH SHEAR
PEEL strength	High G'' at high frequencies (100 Hz)	HIGH PEEL
Cohesive strength	High G' and low $\text{Tan } \delta$	HIGH cohesive strength (Bulk properties)
Adhesive strength	High G'' and high $\text{Tan } \delta$	HIGH adhesion strength with surface

The frequency sweep curves were obtained using data at 25°C. The tested range was from 0.002 - 60 Hz. PSA performance is typically related to the viscoelastic properties in this frequency range. Shear resistance has been found to correlate to the elastic modulus at low frequency (G' at < 0.1 Hz). Figure 1 is a plot of the elastic modulus for the experimental samples as a function of oscillation frequency. At frequency rates between 0.1 and 0.01 Hz, Polymer 2 displayed a much higher elastic modulus than Polymer 1. This can be attributed to higher crosslink density and gel fraction associated with this sample and would be expected to yield an adhesive with higher cohesive strength.

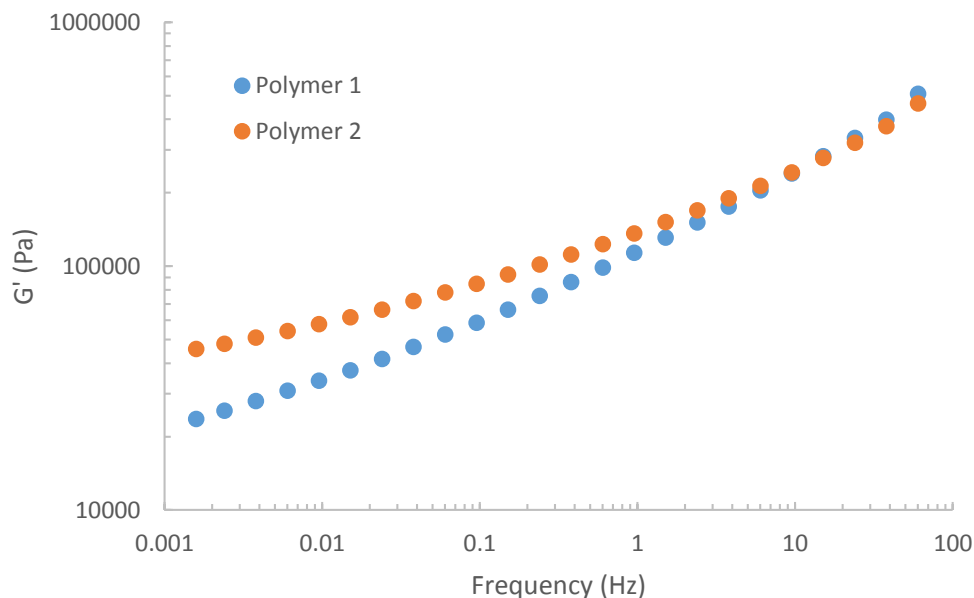


Figure 1. Plot of Storage Modulus of Experimental Polymers

A plot of the loss modulus (G'') of the PSA samples is presented in Figure 2. Peel resistance is dependent on both bond formation and bond deformation. Decreased values of G' at the bonding frequency (0.1 Hz) indicate good wet out. Increased values of G'' at the debonding frequency (100 Hz) indicates high peel strength. Polymer 1 has the lower G' value at 0.1 Hz and the higher G'' values at the

debonding frequency which should correlate to the higher peel strength. The higher elastic modulus of Polymer 2 at low frequency suggests that the wet out and bond formation was low compared to Polymer 1, which can be expected to result in lower 180° peel and loop tack adhesion.

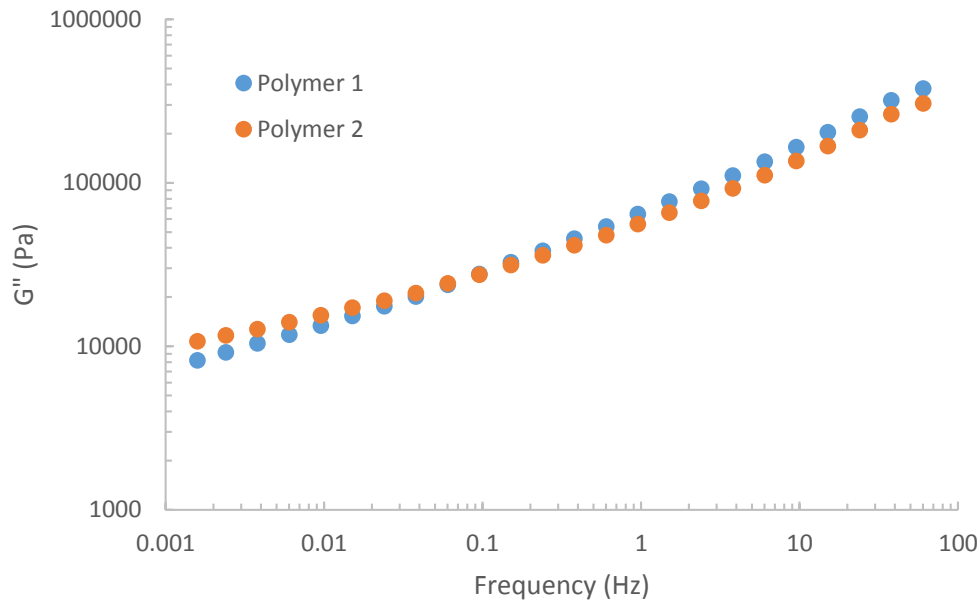


Figure 2. Plot of Loss Modulus of Experimental Polymers

The values of $\tan \delta$ (G''/G') over the frequency range evaluated provide information on the relative tack of the PSAs. For increased tack, G'' is greater than G' at 1 Hz, indicating that the polymer dissipates energy through its own deformation. This allows the material to adhere and easily form good contact with the substrate. Figure 3 clearly shows the higher $\tan \delta$ for Polymer 1 at 1 Hz and thus a higher loop tack would be expected.

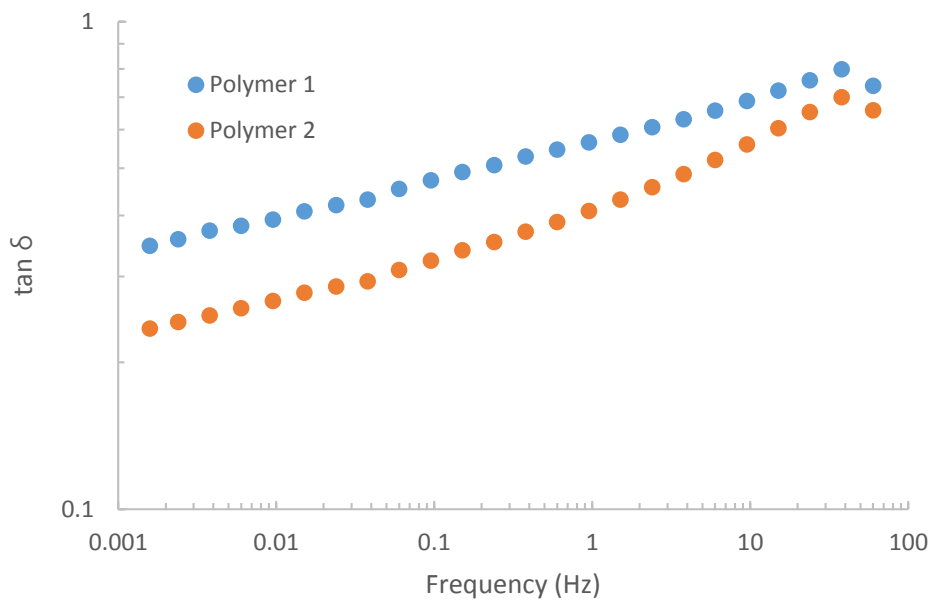


Figure 3. Plot of Tan Delta of Polymers

The adhesive performance of the two experimental polymers are presented in Table 5. The elimination of chain transfer agent and replacement with cross-linker had a dramatic impact on the adhesive properties which were consistent with the predictions from the dynamic mechanical analysis. The cohesive strength of Polymer 2 was significantly higher than that of Polymer 1. High temperature shear adhesion was increased from ~ 1hr to greater than 48 hrs. The SAFT value increased from 66 to 306°C. The improvement in shear adhesion was, however, accompanied by a reduction in both peel adhesion and loop tack.

Table 5. Adhesive performance of experimental samples

Sample ID	Coat Weight (g/100 in ²)	180° Peel Adhesion (lbs/in)			Loop Tack (lbs/in ²)			Shear Adhesion	
		SS	Al	HDPE	SS	Al	HDPE	@ 65°C	SAFT
Polymer 1	1.5	4.5+/-0.7	2.9+/-0.5	2.6+/-0.7	3.2+/-0.5	2.1+/-0.3	2.3+/-0.5	1.2+/-1.1hrs	66+/-9°C
Polymer 2	1.5	2.4	1.6	2.0	2.2	1.8	2.1	>48 hrs	306+/-2°C

In the initial round of adhesive testing the adhesive performance of the experimental polymers was conducted at a coat weight of 1.5 g/100in². This is significantly lower than that employed in any of the commercial tapes, and so additional testing was conducted to establish the impact of coat weight on the adhesive properties. Table 6 summarizes the effect of coat weight on the adhesive properties of Polymer 1. In general, high coat weight resulted in higher peel adhesion and loop tack. No impact was observed on either the high temperature shear adhesion or SAFT. Similar trends were observed with Polymer 2. These findings are consistent with those reported in the literature.¹⁵

Table 6. Impact of coat weight on adhesive performance

Sample ID	Coat Weight (g/100 in ²)	180° Peel Adhesion (lbs/in)			Loop Tack (lbs/in ²)			Shear Adhesion	
		SS	Al	HDPE	SS	Al	HDPE	@ 65°C	SAFT
Polymer 1	1.5	4.5+/-0.7	2.9+/-0.5	2.6+/-0.7	3.2+/-0.5	2.1+/-0.3	2.3+/-0.5	1.2+/-1.1hrs	66+/-9°C
Polymer 1	2.9	5.9	5.6	5.3	5.2	3.4	3.8	0.3+/-0.1hrs	68+/-1°C
Polymer 1	5.1	3.5	3.3	6.4	5.2	4.4	5.2	0.4+/-0.1hrs	71+/-6°C

Blending of polymers is sometimes employed by formulators as a means of obtaining characteristics intermediate of those of the blend constituents. Polymer compatibility can, however, impact the attributes of the mixture.¹⁶ Blends of Polymer 1 and 2 were prepared to assess the impact on adhesive performance. Due to the compositional similarity of the two component polymers a miscible mixture was predicted which would yield a linear relationship between the adhesive performance and the polymer blend ratio.¹⁶

Figure 4 illustrates the impact of polymer blend ratio on the 180° peel adhesion from stainless steel and HDPE substrates. The peel adhesion varies linearly with the polymer blend ratio.

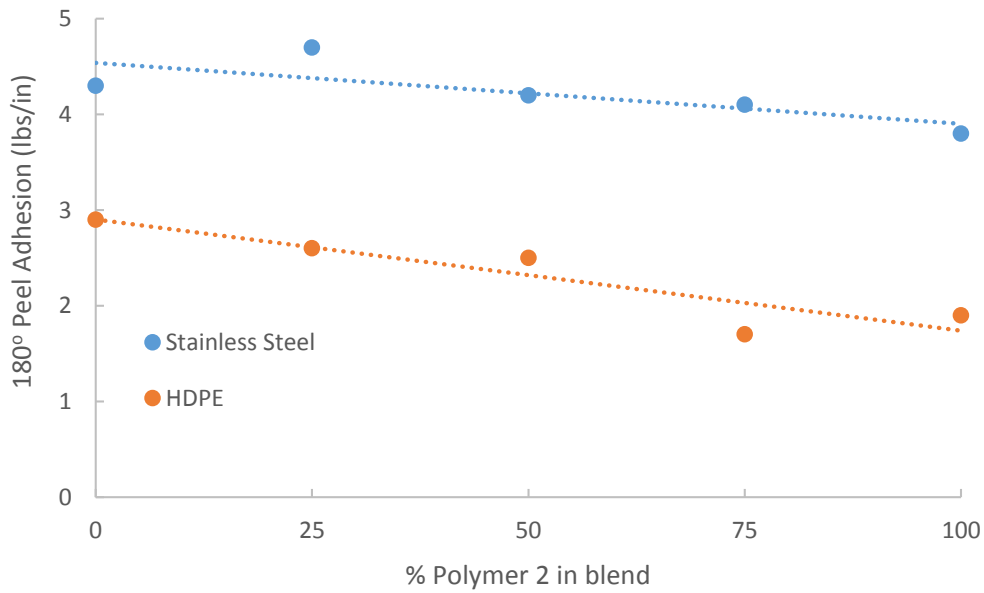


Figure 4. Plot of 180° peel adhesion as a function of Polymer 1 and 2 blend ratio

Similarly, Figure 5 illustrates that the loop tack were found to be in between those of the blend constituents.

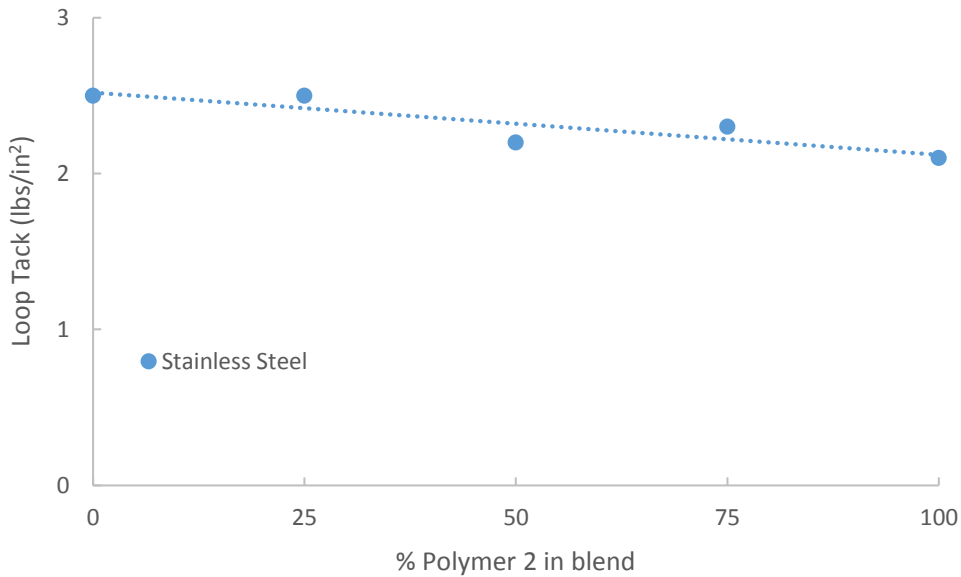


Figure 5. Plot of loop tack as a function of Polymer 1 and 2 blend ratio

Finally, the SAFT of the polymer blend are presented in Figure 6. As with the peel adhesion and loop tack, SAFT values of the polymer blend were intermediate of those of Polymer 1 and Polymer 2.

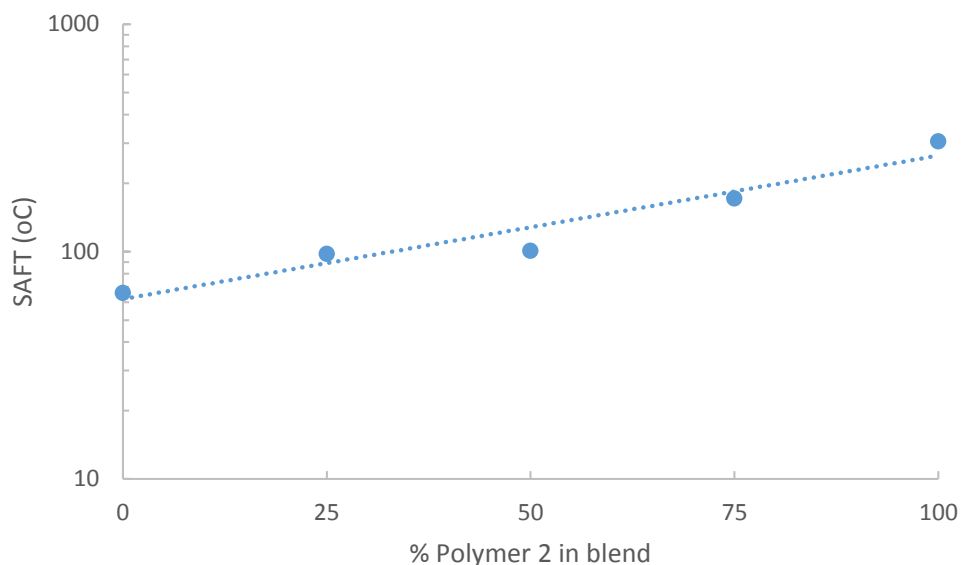


Figure 6. Plot of SAFT as a function of Polymer 1 and 2 blend ratio

Conclusions

Many factors affect the three performance criteria of a waterborne acrylic PSA: tack, peel and shear strength. In this study, improvements in peel adhesion and loop tack, through the manipulation of the adhesive composition, were generally found to come at the expense of cohesive strength. This was true whether the molecular weight was manipulated through the replacement of chain transfer agent with cross-linker, or, through polymer blending. However, other aspects of tape design, such as coated weight, were found to be important in achieving the desired balance of adhesive performance.

Achieving the desired physical and rheological properties requires careful selection of the chemical components and control of polymer molecular weight. Guided by structure-property understanding of PSA performance in relation to polymer's viscoelastic characteristics, Acrylic emulsion polymers, featuring a balance of adhesive and cohesive properties suitable for high-temperature applications, such as HVAC duct tapes and house wrap, were prepared.

Acknowledgements

The authors would like to recognize the contributions of Dr. Robert Fu and Sonia Chalmers.

References

1. I. Benedek, *Pressure-Sensitive Adhesives and Applications*, 2nd edition, Marcel Dekker, Inc., New York, 2004 pp. 89-138
2. D. Satas, *Handbook of Pressure Sensitive Adhesive Technology*, 2nd edition, Van Nostrand Reinhold, Ltd., New York 1989, p.396-456
3. Druschke, W., *Adhesion and Tack of Pressure Sensitive Adhesives*, presented at the AFERA Meeting, Edinburgh, Scotland (October 1-4, 1986)
4. Midgley, A., *Adhes. Age*, (9) 17 (1986)
5. Kendall, J., Foley, F. and Chu, S.G., *Adhes. Age*, (9) 26 (1986)
6. Sobieski, L.A. and Tangney, T.J., *Adhes. Age*, (12) 23 (1988)
7. J. D. Ferry, *Viscoelastic Properties of Polymers*, 3rd edition, John Wiley & Sons, Inc., New York 1980, p. 42.
8. Chauvet, J., Asua, J.A. and Leiza, J.R., *Polymer* 2005, 46, 9555.
9. Shen, H.Z., Zhang, J.Y. and Liu, S.J., *J. Appl. Polym. Sci.* 2008, 107, 1793.
10. Plessis, C., Arzamendi, G. and Leiza, J.R., *Macromolecules* 2000, 33, 5041.
11. Gower, M.D. and Shanks, R.A., *J. Appl. Polym. Sci.* 2004, 93, 2909.
12. Bouvier-Fontes, L., Pirri, R. and Asua, J.M., *J. Polym. Sci. Polym. Chem.* 2005, 43, 4684.
13. Tobing, S.D. and Klein, A., *Synthesis and Structure Property Studies in Acrylic Pressure Sensitive Adhesives*, Proc. 24th Annual Meeting of Adh. Soc., Feb 25, 2001, Williamsburg, VA p. 59
14. Mazzeo F.A. (2002), "Characterization of Pressure Sensitive Adhesives by Rheology", TA Instruments report RH082, 1-8.
15. I. Benedek, *Pressure-Sensitive Adhesives and Applications*, 2nd edition, Marcel Dekker, Inc., New York, 2004 pp. 235-352
16. Brooks, T.W., Kell, R.M., Boss, L.G., and Nordhaus, D.E., *Analysis of Factors Important in Emulsion Acrylic Pressure Sensitive Adhesive Design*, in Proc. 1984 TAPPI Polymers, Lamination and Coatings Conference pp. 469-477. Boston, September 24-26