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Viscoelastic properties and peel strength of water-borne acrylic PSAs for labels

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Abstract—The use of acrylic emulsion pressure-sensitive adhesives (PSAs) in labels continues to increase due to their environmental friendliness, good aging resistance and excellent processability during coating process. This study has investigated the acrylic emulsion/tackifier blend systems by examining their thermal properties as indicated by the glass transition temperature (T_g), by conducting optical microscopy to determine their structure, by exploring their viscoelastic properties with the advanced rheometric expansion system (ARES), and by measuring their peel strength with the 180° peel test. The acrylic emulsion/tackifier blends with low softening point and low molecular weight exhibited only a single T_g . However, the acrylic emulsion/tackifier blends with high softening point and high molecular weight showed two T_g values. Microscopic observation of blends with miscible tackifier did not show any evidence of tackifier traces, whereas the microscopic structures of blends with immiscible tackifier showed dispersed tackifier particles. As the tackifier content increased, the rubbery plateau region decreased for the miscible blends but increased for the immiscible blends. The 180° peel strength of the miscible blends was dependent on the viscoelastic properties and was influenced by incorporation of tackifiers in the PSA systems. However, the peel strength of the immiscible systems did not depend on the viscoelastic properties at low tackifier content, but it decreased as the tackifier content increased in accordance with the increase of the storage modulus G' .

Keywords: Water-borne acrylic PSA; viscoelastic property; miscibility; peel strength.

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1. INTRODUCTION

In the 1980s label manufacturers started converting from solvent to water-borne systems because of strict environmental protection regulations, resulting in water-borne acrylic emulsions becoming the most important group of pressure-sensitive adhesive (PSA) raw materials. This conversion continued in the 1990s, so that now the use of water-borne acrylic emulsions in PSA products is widespread and well developed [1]. The production of acrylic emulsion PSAs for labels will continue to increase due to their environmental friendliness, good aging resistance and excellent processability during coating process [2]. As the production and use of water-borne acrylic emulsion PSA labels have increased, various preparation methods for acrylic emulsion PSAs have been presented. Initially, simple emulsion polymerized acrylic PSAs were introduced, then water-borne PSAs for various applications and then highly functional PSAs were patented. In addition, the effect of tackifier for water-borne acrylic emulsion PSAs and their formulations with PSAs were investigated [3–9].

This study has investigated the miscibility and viscoelastic properties of water-borne acrylic PSAs tackified with various resin ester dispersions considering the softening point of the tackifier, type of esterified rosin, and tackifier content using differential scanning calorimetry (DSC), optical microscopy, and mechanical spectroscopy. The 180° peel strength was measured as a function of temperature and peel rate for varying softening point, molecular weight and type, and content of tackifier using a universal tensile machine (UTM). Furthermore, the master curves of peel strength *versus* $\log V a_T$ are analyzed to investigate the correlation between peel strength and miscibility, or peel strength and viscoelastic properties.

2. EXPERIMENTAL

2.1. Materials

The water-borne acrylic emulsion selected for this study was PR-60 (Hanwha Chemical, South Korea). Some of its properties are given in Table 1. The tackifiers selected for this study were Superesters E-650, E-720 and E-730-55 (rosin ester dispersions, Arakawa Chemical, Japan). The properties of the tackifiers are shown in Table 2. The wetting agent selected for this study was Aerosol OT-75 (Cytec).

2.2. Preparation of PSAs

Acrylic emulsion/tackifier dispersions were blended in the following blend ratios: 90:10, 80:20, 70:30 and 60:40 wt%. Wetting agent (0.9 wt%) was added in the direct driven stirrer (Tops Misung, South Korea) with an impeller at 500 rpm and the blends were left for 24 h at room temperature. The PSA specimens were prepared by coating the blends of acrylic emulsion and tackifier dispersion onto a release liner, using an automatic film applicator (Kee-Pae Trading, South Korea) at room

Table 1.

Some properties of water-borne acrylic PSA

Solid content (wt%)	T_g^a (°C)	pH	M_n^b	M_w^b	MWD ^b	Product name
58.0	-49.3	6.0	8150	55 300	6.8	PR-60

^a Measured by DSC.^b Measured by GPC.**Table 2.**

Some properties of rosin ester dispersions

Product name	E-720	E-730-55	E-650
Solid content (wt%)	50.4	55.0	51.0
Softening point (°C)	100	125	160
T_g (°C) ^a	55.4	78.3	103.1
M_n^b	710	1220	900
M_w^b	800	2780	2230
MWD ^b	1.1	2.3	2.5
Main component	Glycerol ester of stabilized resin	Pentaerythritol ester of stabilized resin	Pentaerythritol ester of polymerized resin

^a Measured by DSC.^b Measured by GPC.

temperature. A bar coater No. 22 (wet thickness 50.3 μm) was used. The PSA label was dried at 100°C for 5 min, and then laminated to the corona-treated poly(ethylene terephthalate) (PET) film of 50 μm average thickness.

2.3. Molecular weight and molecular weight distribution

The weight average molecular weight (M_w), number average molecular weight (M_n) and molecular weight distribution (M_w/M_n , MWD) of the PSAs were determined by gel-permeation chromatography (GPC, Waters). GPC was used to obtain narrow molecular weight fractions and separation was accomplished on a column of a highly porous material that separates the polymer molecules according to size.

2.4. Thermal properties

The glass transition temperature (T_g) was measured using DSC (TA Instruments, Q-1000). The samples were cooled to -85°C, heated to 150°C at a heating rate of 10°C/min in the first scan, immediately quenched to -85°C and then kept at this temperature for 15 min. The samples were then reheated to 150°C at a heating rate of 10°C/min in the second scan. The T_g in this study was obtained from the second scan to assure reproducible thermograms free from thermal history effects.

2.5. Viscoelastic properties

The viscoelastic properties (storage modulus, loss tangent and complex viscosity) of the blends were determined using ARES (at NICEM, South Korea) in the

8-mm parallel plate mode. A typical scan covered the range from -70 – 100°C . The frequency was 0.5 Hz and the cooling and heating rates were $5^{\circ}\text{C}/\text{min}$.

2.6. Microscopic observation

Some blends were observed under an optical microscope (Zeiss) equipped with a camera. The optical micrographs of blends were obtained after heating at 100°C for 10 min followed by cooling to room temperature.

2.7. Peel strength

Before measuring peel strength, PSA coated labels were pressed onto the stainless steel substrate by rolling a 2-kg rubber-roller onto the substrate for 5 s. The stainless steel was cleaned with acetone before attaching PSA labels. After keeping the specimen at room temperature for 1 h, peel strength was measured using a Universal testing machine (Zwick, Z101). The 180° peel strength of the PSA specimens was measured in tension mode at an angle of 180° between the substrate and PSA specimen with a crosshead speed of 75, 150, 300 and 600 mm/min and temperature of 25, 50 and 75°C .

3. RESULTS AND DISCUSSION

3.1. Thermal analysis

The T_g of a polymer is determined by the amorphous region of a partially crystalline polymer when a viscous or rubbery state is transformed into a hard, brittle, glass-like state. The T_g evaluated by DSC has been generally used in the determination of blend miscibility. The phase structure of a blend is assessed by the number of T_g values observed in the thermogram. Two transitions are a clear indication of phase separation, while a T_g located at a temperature intermediate between those of the pure components indicates miscibility [10].

Figures 1–3 show the T_g curves for acrylic emulsion/tackifier blend PSAs with three types of rosin ester dispersions. Figure 1 shows that the glycerol stabilized rosin ester is miscible with acrylic emulsion over all blend ratios. Some blends show partial miscibilities with acrylic emulsion. In Fig. 2, water-borne acrylic emulsion/Superester E-730-55 blend shows a single T_g at 40 wt%, but two T_g values are present over 40 wt%. In Figure 3, water-borne acrylic emulsion/Superester E-650 blend PSA has two T_g values from 70 to 90 wt%, but other blends have only a single T_g .

Water-borne acrylic emulsion/pentaerythritol stabilized rosin ester blend PSAs and water-borne acrylic emulsion/pentaerythritol polymerized rosin ester blend PSAs showed two T_g values as for natural rubber/pentaerythritol ester of stabilized rosin blend in the study of Sherriff *et al.* [11].

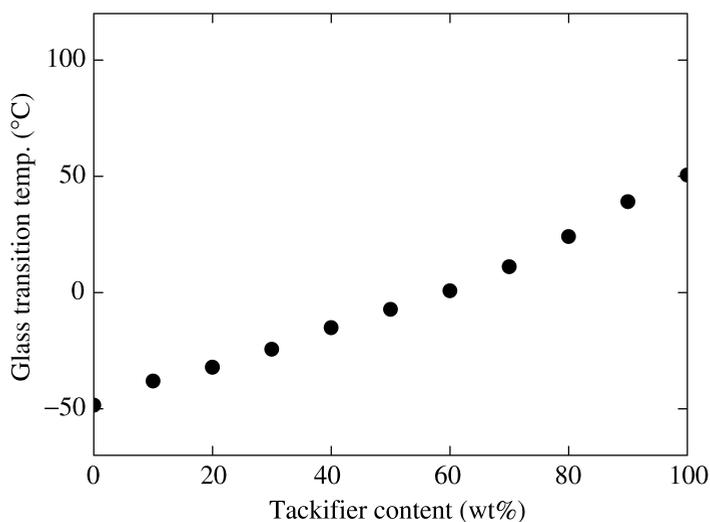


Figure 1. T_g versus tackifier content of acrylic emulsion/Superester E-720 blend PSAs.

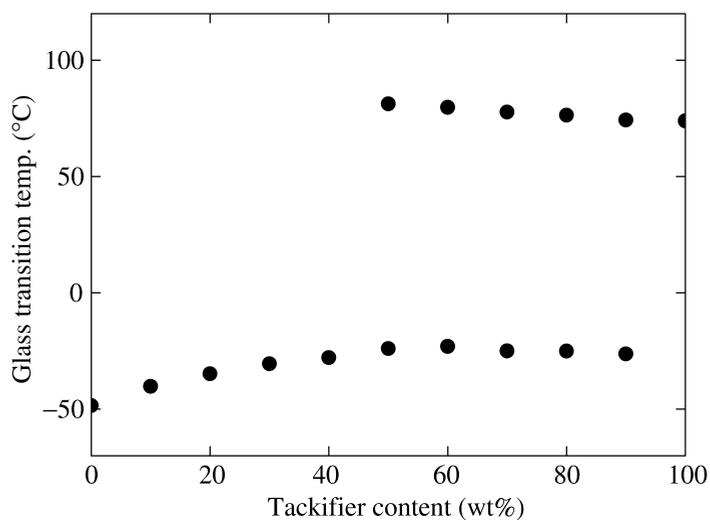


Figure 2. T_g versus tackifier content of acrylic emulsion/Superester E-730-55 blend PSAs.

3.2. Microscopic observation

The optical micrographs of acrylic emulsion/Superester E-720 blend PSA are shown in Fig. 4a and 4b with Superester E-720 contents of 20 and 40 wt%, respectively. All of these blend systems have only a single T_g , which suggests that these blends are miscible systems. The blends of acrylic emulsion/Superester E-650 are also shown in Fig. 5a and 5b with Superester E-650 contents of 20 and 40 wt%, respectively.

Observing the blends of 20 wt% tackifier content, a dot-like phase is seen. As the tackifier content increases, the dot-like phase becomes more apparent. The dot-like

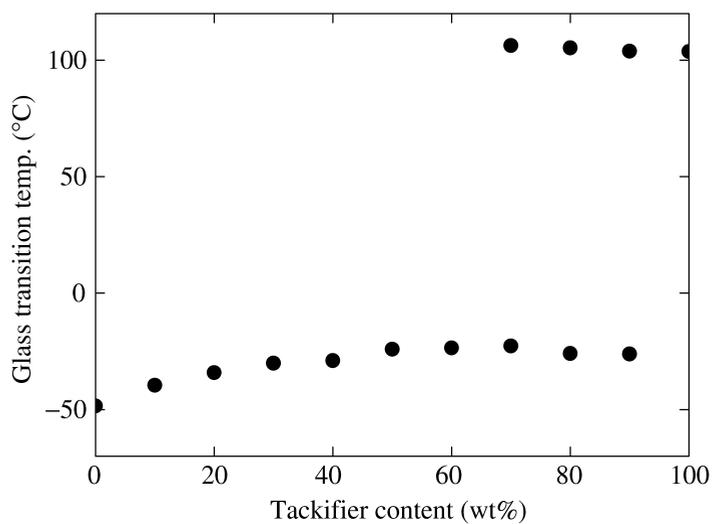


Figure 3. T_g versus tackifier content of acrylic emulsion/Superester E-650 blend PSAs.

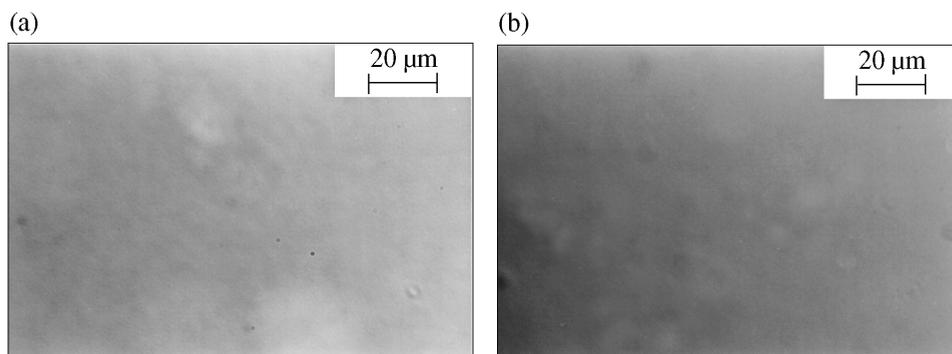


Figure 4. Acrylic emulsion/Superester E-720 blend PSA ($\times 1000$). (a) 20 wt% tackifier content: miscible system (b) 40 wt% tackifier content: miscible system.

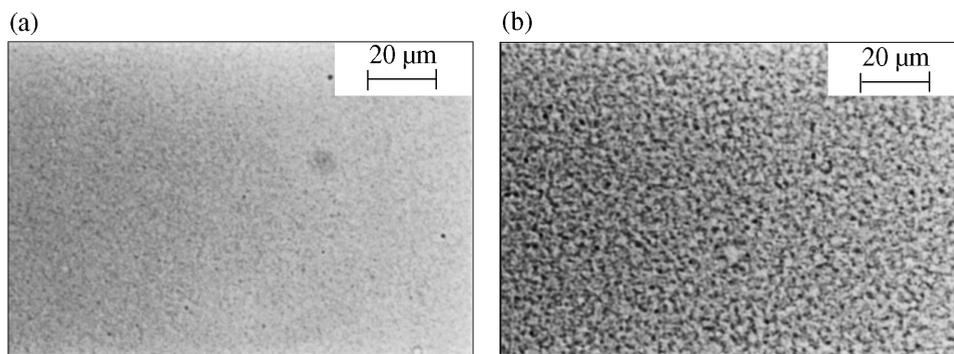


Figure 5. Acrylic emulsion/Superester E-650 blend PSA ($\times 1000$). (a) 20 wt% tackifier content: immiscible system (b) 40 wt% tackifier content: immiscible system.

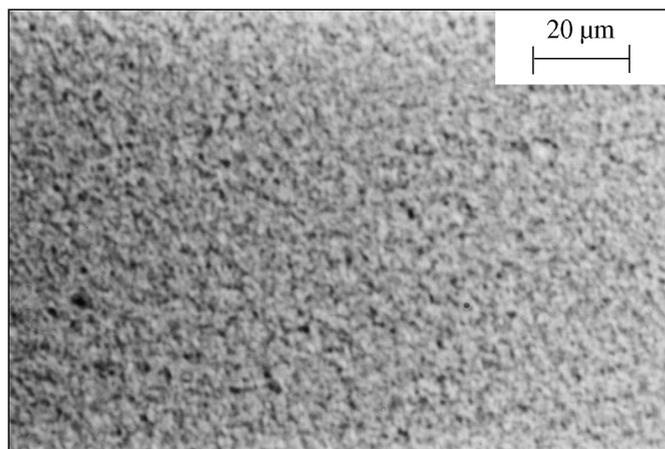


Figure 6. Acrylic emulsion/Superester E-730-55 blend PSA (40 wt% tackifier content) ($\times 1000$): immiscible system.

pattern and the dispersed pattern in these blend systems are thought to be a spreading particle or coagulation of the tackifier. Though these blends have a single T_g below 70 wt% tackifier content (by DSC data) and two T_g values over 70 wt%, the amount of tackifier particles was expected to be minimal and was not measured. Therefore, these blend systems are thought to be immiscible. Figure 6 shows the phase of acrylic emulsion/Superester E-730-55 blend PSA, a 40 wt% tackifier content blend system, in which the dispersed pattern structures were found. This Superester E-730-55 blend system also had a single T_g (up to 50 wt% tackifier content) but two T_g values (over 50 wt%) in the former DSC thermogram. Nevertheless, Fujita noted that these blend systems were somewhat immiscible due to the dispersed structure of this 40 wt% tackifier content blend [12].

3.3. Viscoelastic properties

The performance of a PSA is related to the viscoelastic properties of the bulk adhesives. Bonding is a relatively slow process accompanied by a slight deformation that occurs when the PSA is brought into contact with a surface, while debonding in a tack or peel test is a fast process, depending on the thickness of the adhesive. Thus, the viscoelastic behavior of an adhesive controls its response in label adhesive testing and is an important factor in controlling its performance [13].

In particular, acrylic PSAs show different viscoelastic properties and adhesive performance due to the compatibility between PSA and tackifiers [14].

3.3.1. Miscible blends. Figure 7 shows the plot of storage modulus G' versus temperature for a series of blends (as a function of tackifier content) of acrylic emulsion PSAs with glycerol ester of stabilized rosin (Superester E-720). The glass transition region of storage modulus G' shifted toward higher temperatures, while

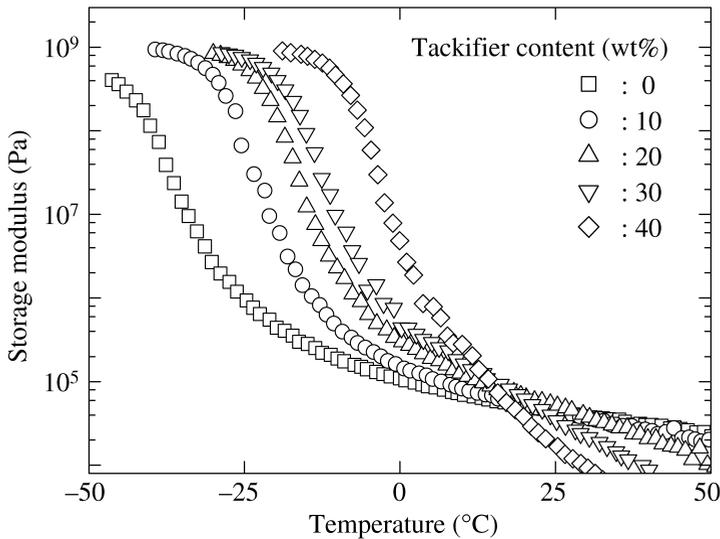


Figure 7. Storage modulus G' of acrylic emulsion/Superester E-720 blend PSAs with different tackifier contents.

the rubbery plateau region of storage modulus G' moved to a lower position, as the tackifier content in the blend increased. This indicates that the tackifier resin, Superester E-720, is miscible with acrylic emulsion. This was also confirmed by DSC (as shown in Fig. 1).

Aubrey and Sherriff [15] reported that the addition of a compatible resin to rubber shifted the entry to the transition zone to a lower frequency, and also reduced the modulus in the rubbery plateau. Kim [14] showed that in acrylic solution/glycerol and diethyleneglycol mixed with ester rosin blend, the storage modulus G' of the blend decreased as the tackifier content increased. It was further reported that glycerol and diethyleneglycol mixed with ester rosin was miscible with acrylic solution. This situation is also shown in case of water-borne acrylic emulsion tackified with glycerol ester of stabilized rosin (Superester E-720).

3.3.2. Immiscible blends. The storage moduli G' of acrylic emulsion blends with pentaerythritol ester of stabilized rosin (Superester E-730-55) and pentaerythritol ester of polymerized rosin (Superester E-650) are shown in Figs 8 and 9, respectively. The glass transition region of storage modulus G' of acrylic emulsion/Superester E-730-55 blend PSAs showed a shift toward higher temperatures, and also the rubbery plateau region of storage modulus G' moved to a higher position as the tackifier content in the blend increased. This definitely shows that the tackifier resin, Superester E-730-55, is immiscible with acrylic emulsion, as was also confirmed by DSC (as shown in Fig. 2). The glass transition region of storage modulus G' of acrylic emulsion/Superester E-650 blend PSAs showed a shift toward higher temperatures, and also the rubbery plateau region of storage modulus G' shifted to higher modulus domain as the tackifier content in the blend increased.

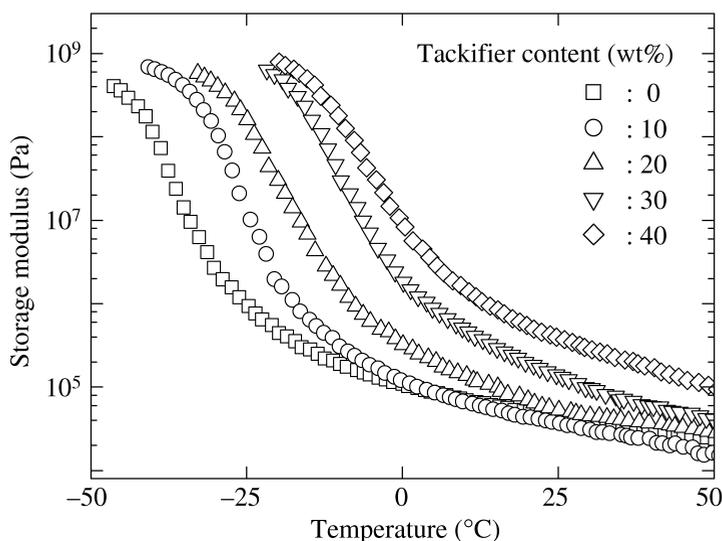


Figure 8. Storage modulus G' of acrylic emulsion/Superester E-730-55 blend PSAs with different tackifier contents.

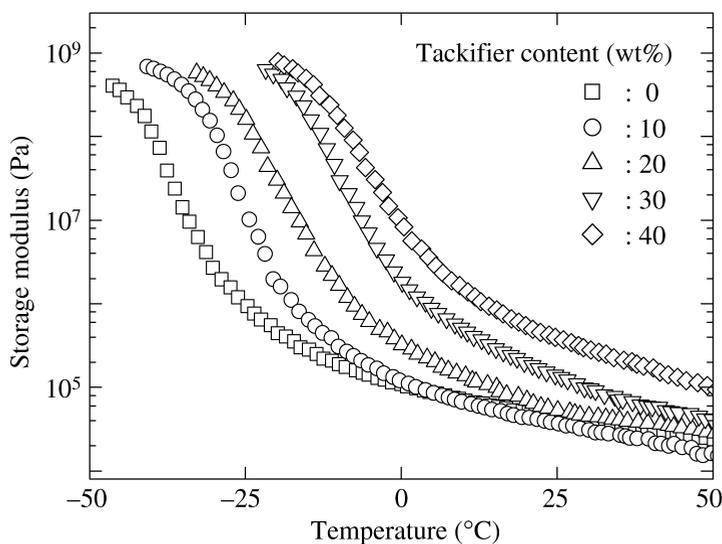


Figure 9. Storage modulus G' of acrylic emulsion/Superester E-650 blend PSAs with different tackifier contents.

This definitely shows that Superester E-650 is also immiscible with acrylic emulsion, as was further confirmed by DSC data (as shown in Fig. 3). As shown in Figs 8 and 9, the plots of storage modulus G' of both acrylic emulsion/Superester E-730-55 and acrylic emulsion/Superester E-650 blend systems show a similar pattern in all regions, but a careful investigation reveals difference between the two blends. The plots of storage modulus G' of acrylic emulsion/Superester E-650 blend sys-

tems shifted toward higher temperatures than those for acrylic emulsion/Superester E-730-55 blends because of the differences between the T_g values of Superester E-730-55 and Superester E-650.

Class and Chu [16] reported that in rubber-resin blends the immiscibility was observed in blends with a weight average molecular weight of more than 1000. Sherriff *et al.* [1] reported that natural rubber/pentaerythritol ester of stabilized rosin blends showed increased storage modulus G' as the amount of tackifier was increased, because the tackifier was immiscible with natural rubber. In these blend systems (acrylic emulsion/Superester E-730-55 and acrylic emulsion/Superester E-650) the average molecular weight of the tackifier resin is over 1000 (as shown in Table 2) and the tackifiers used in these blend systems are immiscible with acrylic emulsion (by DSC), which caused the increase of storage modulus G' of the above blends.

3.4. Peel strength

Peel strength is one of the recognizable parameters of PSA performance [17]. Peel strength depends on the temperature and the peel rate. At a given temperature, the peel strength increases with increasing peel rate. At low peel rates the viscous properties are dominant, polymer molecules have time to slide past one another, to disentangle and dissipate energy. At high peel rates the elastic properties of the polymer network predominate, the polymer molecules are not able to disentangle, and so the polymer modulus or “stiffness” increases. Since the mobility of polymer chains increases with temperature, the peel strength will also decrease at a constant peel rate [11].

3.4.1. Peel strength dependence on tackifier type. In Fig. 10 the peel strength of acrylic emulsion/Superester E-720 blends is increased as the content of Superester E-720 is increased, whereas the peel strength of acrylic emulsion/Superester E-730-55 and acrylic emulsion/Superester E-650 blends reaches a maximum at 30 and 20 wt%, respectively, and then decreases, being below that of the untackified blend system at 40 wt%. On the basis of the earlier DSC data, these two tackifiers (Superester E-730-55 and E-650) are immiscible with water-borne acrylic emulsion (PR-60), because both acrylic emulsion/Superester E-730-55 and acrylic emulsion/Superester E-650 blend systems show a second T_g over 50 wt% of tackifier content, although some differences between the two blends exist (Figs 2 and 3). In these peel strength tests it is thought that the decrease of peel strength at high tackifier content (at 40 wt%) is due to the immiscibility of both blend systems. Furthermore, the high peel strengths observed at low tackifier content (below 30 wt%) are thought to be a result of the agglomeration and dispersed structure of the tackifier in these blends. The reasons why the tackifier content of these two blends influences the maximum peel strength value of each should be studied further. Among the various blends studied here, the acrylic emulsion blends with glycerol esters of stabilized rosin tackifier showed better peel performance than

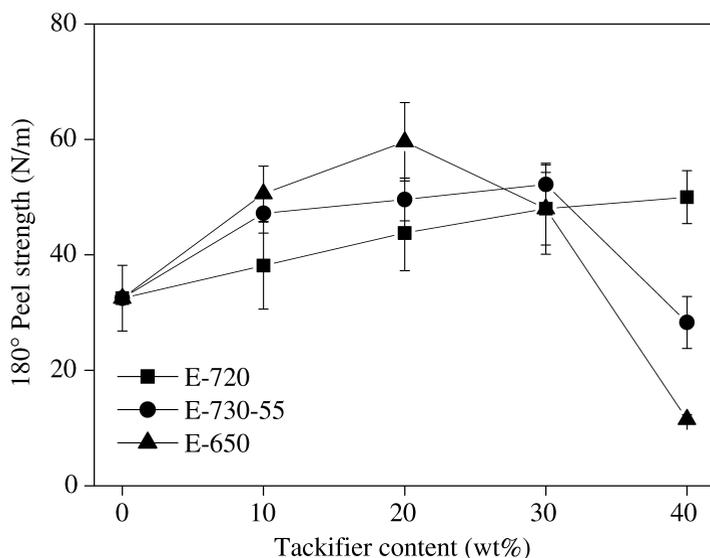


Figure 10. Peel strength *versus* tackifier content for three acrylic emulsion PSAs: Superesters E-720, E-730-55 and E-650 blend PSAs (at 25°C, cross-head speed 300 mm/min).

the other tackifier blend systems, but the other tackifier blend systems with high softening points showed greater peel strengths only when the conditions were limited to low tackifier content.

3.4.2. Time–temperature superposition. For polymers, the effect of varying the temperature or time is identical and a great amount of knowledge about the molecular basis for polymer properties has come from the experimental and theoretical studies of time–temperature superposition [17]. Relaxation and creep occur by diffusive molecular motions which become more rapid as the temperature increases. Temperature is a measure of molecular motion. At higher temperature, the molecules move faster. The WLF equation expresses a logarithmic relationship between time and temperature [18] as:

$$\log a_T = \frac{-C_1(T - T_s)}{C_2 + T - T_s}.$$

Here, T is the temperature when the experiment is done, T_s is the standard temperature (293.15 K), C_1 and C_2 are constants, and a_T is a shift factor. In a somewhat better approximation, fixed values of $C_1 = 8.86$ and $C_2 = 101.6$ were used in conjunction with a reference temperature T_s , which was allowed to be an adjustable parameter but generally fell about 50°C above the T_g , as follows:

$$\log a_T = \frac{-8.86(T - T_s)}{101.6 + T - T_s}.$$

Alternatively, if the standard temperature is chosen to be T_g , then

$$\log a_T = \frac{-17.5(T - T_g)}{51.6 + T - T_g}.$$

The existence of such apparently useful universal equations for the shift factor has formed the basis for many experiments and theories regarding the viscoelasticity of polymers. Building on these ideas, the time–temperature superposition principle states that with viscoelastic materials, time and temperature are equivalent to the extent that data at one temperature can be superimposed onto data at another temperature by shifting the curves along the time axis. The curve obtained after the shifts have been applied is called a master curve [19].

In any 180° peel test, one typically varies either the temperature or the peel rate over a specific range and measures the peel strength of PSAs. The range of peel rates over which one can do experiments easily is limited, since extremely low or high peel rates are hard to maintain and are difficult to measure. If we conduct the 180° peel test for a small range of easily accessible rates and measure the strength over a wide range of measurable temperatures (which are more easily variable, measurable and maintainable), we can obtain the family of curves shown in Fig. 11a. The temperatures are in the order $T_1 > T_2 > T_3 > T_4 > T_5 > T_6$. Examination of these generic data demonstrates that shifting the curves for temperatures T_1 through T_5 to the left from the standard temperature T_6 by some amount will cause all of the curves to form a smooth curve. This is found to be the case for most PSAs. The result is known as a master curve and the amount by which a segment is shifted is known as a shift factor. Thus, a master curve is a plot of the peel strength as a function of a reduced peel rate. The reduced variable is obtained by multiplying the peel rate by the shift factor. Thus, a set of curves shown in Fig. 11a can be reduced to a single curve shown in Fig. 11b [17].

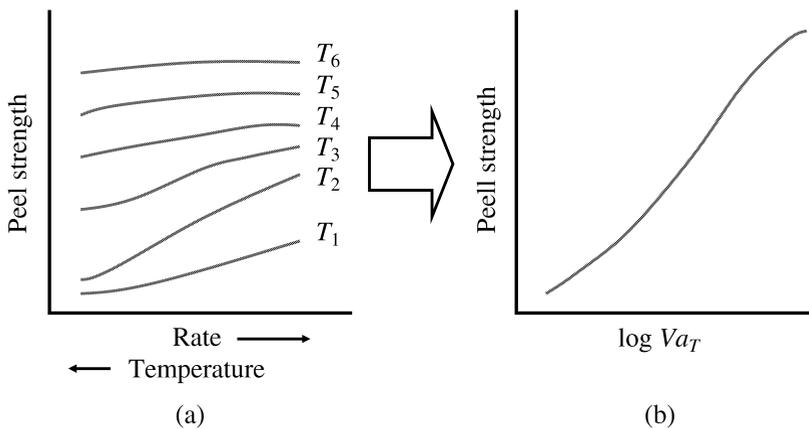


Figure 11. The making of a master curve [14].

The master curves of peel strength for a series of acrylic emulsion/Superester E-720 blends are shown in Fig. 12. As the tackifier content of the blends is increased, the master curves are shifted to upper peel strength values. Figure 13 shows the same patterns for acrylic emulsion PSA/Superester E-720 blend systems.

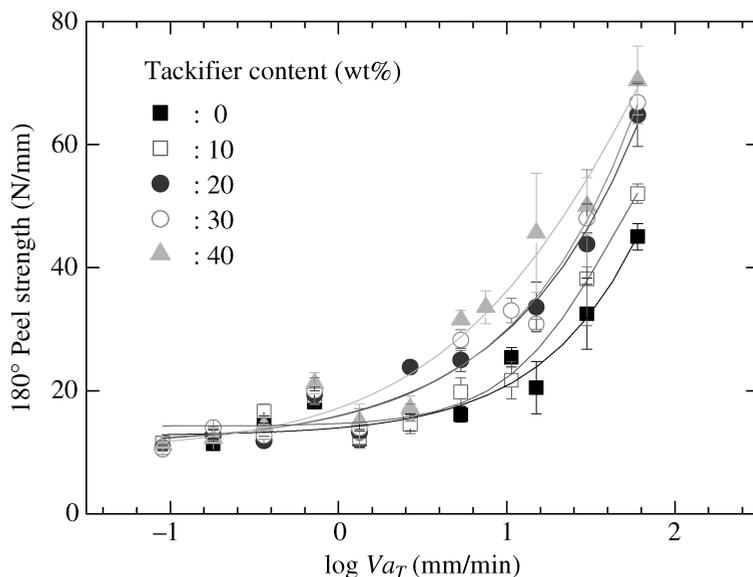


Figure 12. Master curves of a series of water-borne acrylic emulsion/Superester E-720 blend PSAs.

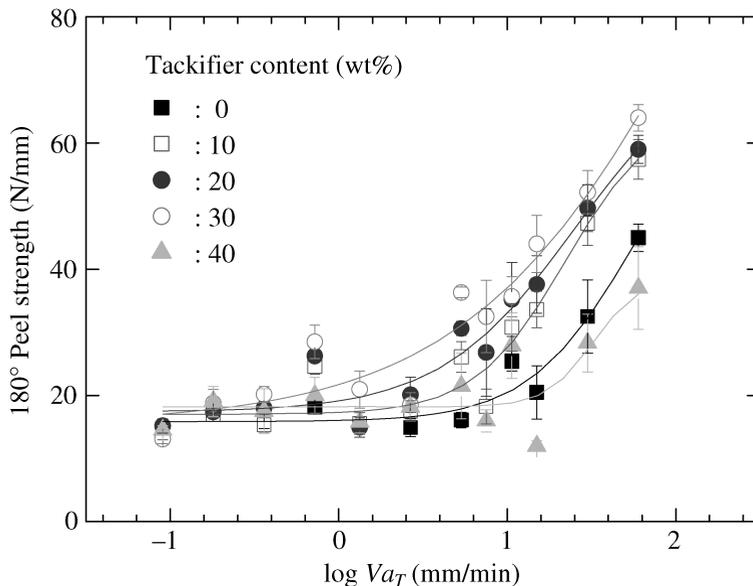


Figure 13. Master curves of a series of water-borne acrylic emulsion/Superester E-730-55 blend PSAs.

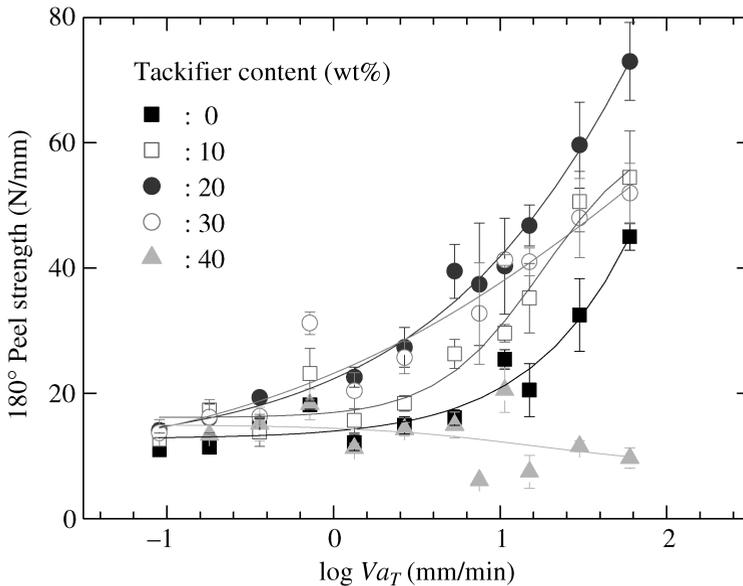


Figure 14. Master curves of a series of water-borne acrylic emulsion/Superester E-650 blend PSAs.

However, as the peel rate is increased at the tackifier content of 40 wt%, the peel strength values became lower than those of the other blends. Figure 14 shows the decline of peel strength at 40 wt% tackifier content. A decrease of peel strength is shown at 30 wt%.

4. CONCLUSIONS

Miscibility between water-borne acrylic emulsion and resin ester tackifiers was investigated using DSC and optical microscopy. The blend systems were sorted into two types: miscible and immiscible. The miscible blend systems had only a single T_g , whereas the immiscible blends showed two T_g values. Microscopic observation of the blends with miscible tackifier did not show the characteristic of tackifier. However, the microscopic structures of the blends with immiscible tackifier showed dispersed tackifier particles. Molecular weight was measured by GPC and was about 800 in miscible blends and over 2000 in immiscible blends.

The viscoelastic properties of these blend systems were determined as a function of temperature. The modulus of rubbery plateau of miscible blends decreased with increasing tackifier content, but that of immiscible blends increased.

The peel strength was measured as a function of tackifier content, peel rate and temperature. At 25°C, the peel strength of acrylic emulsion/tackifier blends peaked and then decreased as the temperature was raised.

The peel strength of miscible blends was dependent on the viscoelastic properties of the materials and was also influenced by the incorporation of tackifiers into the PSA systems. However, the peel strength of immiscible systems showed one

different feature at low tackifier content in that the peel strength of the immiscible blends was decreased as the amount of tackifier was increased, possibly due to the increase of its storage modulus G' at the rubbery plateau.

In both miscible and immiscible blends the master curves of peel strength versus log shift factor showed shifts as the tackifier content of the blends increased. However, in immiscible blends the peel strength was lower than that of the miscible blends at high tackifier contents as the peel rate increased. These decreases of peel strength were considered as a result of the difference in miscibility.

Considering the relationship between viscoelastic properties and peel strength, the increase of peel strength with decreasing storage modulus (G') in the rubbery plateau in miscible blends was observed, but there was no correlation in immiscible blends with tackifier content below 40 wt%.

Acknowledgements

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REFERENCES

1. I. Benedek, in: *Development and Manufacture of Pressure-Sensitive Products*, p. 682. Marcel Dekker, New York, NY (1999).
2. D. Urban and K. Takamura, in: *Polymer Dispersions and Their Industrial Applications*, p. 408. Wiley-VCH, Weinheim (2002).
3. R. Czerepinski, G. Gunderman and R. E. Midland, US Patent 762,083 (1985).
4. M. Bernard and S. Berman, US Patent 686,317 (1991).
5. J. Samonides, Sr., US Patent 6,075,053 (1996).
6. A. Mayer, P. Keller and P. Kreuzlingen, US Patent 819,114 (1992).
7. S. M. Hoff, L. Ternorutsky and L. MacChester, US Patent 827,857 (1997).
8. L. Varela and T. Sanborn, in: *PSTC Annual Technical Seminar Proceedings*, Orlando, FL, p. 125 (2001).
9. N. Hemmings, *Adhesives Age* **41**, 40–46 (1998).
10. T. Hatakeyama and F. X. Quinn, in: *Thermal Analysis Fundamentals and Applications to Polymer Science*, p. 180. Wiley, New York, NY (1999).
11. M. Sherriff, R. W. Knibbs and P. G. Langley, *J. Appl. Polym. Sci.* **17**, 3423–3438 (1973).
12. M. Fujita, *Miscibility and Performance of Natural Rubber Based Pressure Sensitive Adhesives*, Ph. D. Thesis. University of Tokyo, Tokyo (1998).
13. D. Satas, in: *Handbook of Pressure Sensitive Adhesive Technology and Applications*, p. 170. Satas & Associates, Warwick, RI (2002).
14. H.-J. Kim, *Miscibility and Performance of Acrylic Pressure Sensitive Adhesives*, Ph. D. Thesis, University of Tokyo, Tokyo (1995).
15. D. W. Aubrey and M. Sherriff, *J. Polym. Sci.: Polym. Chem.* **16**, 2631–2643 (1978).
16. J. B. Class and S. G. Chu, *J. Appl. Polym. Sci.* **30**, 815–824 (1985).
17. A. V. Pocius, in: *Adhesion and Adhesives Technology*, p. 319. Hanser, Munich (1997).
18. J. D. Ferry, in: *Viscoelastic Properties of Polymers*, p. 641. Wiley, New York, NY (1980).
19. L. H. Sperling, *Introduction to Physical Polymer Science*. Wiley, New York, NY (2001).