



Crosslinked acrylic pressure sensitive adhesives. 3. Effect of adherend on film formation

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Abstract

The reaction of isocyanate in pressure sensitive adhesive (PSA) films adhered on to various adherends having different surface tensions was monitored by depth profiling using attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy.

In the latter stages of crosslinking reaction, unreacted isocyanate and its derivatives exist more in the bulk of the PSA than in the interfaces between PSA and adherends which are Teflon sheet and PE film having relatively lower surface tensions. In the case of using stainless steel having relatively higher surface tension as adherend, opposite segregation was observed compared to Teflon and PE.

From X-ray photoelectron spectroscopy (XPS), it was revealed that N atoms exist more in surface than in bulk when stainless steel is used as adherend. We conclude that polyisocyanates migrate in the PSA film in order to minimize the magnitude of interfacial free energy between the PSA and the adherend, which leads to the change of surface tension of PSA film.

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1. Introduction

In our previous papers [1,2], the effect of crosslinking reaction on peel strength and that of humidity on crosslinking reaction for crosslinked acrylic pressure sensitive adhesives (PSAs) were explored. It was known that crosslinking reaction affected to a great extent on peel strength, which is considered by the wettability of the PSAs [1]. It was also known that the urea and the urethane formations are affected by the humidity of the crosslinking atmosphere [2].

It has been revealed that the surface structure of multicomponent polymer systems is different from that in the bulk, that is, a component having lower surface free energy is generally enriched in the surface region in order to minimize the interfacial free energy between air and the polymer. This phenomenon is called surface segregation or

interface segregation, which are reviewed elsewhere [3–7]. In the crosslinked PSA system, where PSA is curing on the surface of various adherends, the interface segregation of PSA component occurs to minimize the interfacial energy, which is considered to affect the adhesive performances.

The objective of this research is to elucidate the effect of adherend on the crosslinking reaction in PSA film. Poly(*co*-ethyl acrylate-2-ethylhexyl acrylate-2-hydroxyethyl methacrylate) and polyisocyanate was chosen as a base resin and a crosslinker, respectively. We monitored the change in the concentration gradient of unreacted isocyanate groups and its derivatives in the PSA films by depth profiling using attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy. The surface properties of the PSA films finely obtained was determined by surface tension measurements. Additionally, the surface composition of the PSA films finely obtained was revealed by X-ray photoelectron spectroscopy (XPS).

In this study, we will focus on reactions of isocyanate groups in the PSA films affected by adherends having various surface tensions during the process of crosslink.

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2. Experimental

2.1. Materials

Hydroxy-substituted polyacrylic resin prepared by copolymerization of ethyl acrylate, 2-ethylhexyl acrylate and 2-hydroxyethyl methacrylate with a ratio of 60/35/5 wt% was furnished by Hitachi Chemical Co., Ltd. (Ibaraki, Japan) in form of ethyl acetate/toluene solution (35/65 wt%). The glass transition temperature (T_g) of the polymer was -28.1°C by differential scanning calorimetry (DSC) measurement. Polyisocyanate crosslinker CORONATE L, trimethylpropane-1-methyl-3-isocyno-4-carbamate (Nippon Polyurethane Ind. Co., Ltd. Tokyo, Japan) was used as a crosslinker in ethyl acetate/toluene solution (20/80 wt%). These two materials were used without further purification. Other chemicals used were reagent grade in accordance with Japanese Industrial Standard.

2.2. Preparation of PSA films

PSA films used in this study were prepared by the following procedure. The polyacrylic solution was diluted to 5 wt% solids contents with toluene. Three phr of CORONATE L was added to the solution, which gave a ratio of $\text{NCO/OH} = 1/3$. The mixture was stirred for 20 min, and then cast onto polyethylene (PE) films of 60 μm thickness, which were a backing of PSA films. The PSA films were dried at 80°C for 2 min, and then wound into a roll. The thickness of the PSA layer thus obtained was 3 μm . It was confirmed by gas chromatography that there was no residual solvent in the PSA films under this drying condition. The PSA layer was transparent over the experiment, suggesting the acrylic resin and the crosslinker are miscible at the formulation above. For the control experiments, polyacrylic PSA films without crosslinker were also prepared similarly. The rolls of PSA films were left under ambient air for 2 days, of which procedure was same as commercial PSAs'. Time zero in these experiments thus represents 2 days after the end of preparation of PSA films.

2.3. Sample handling

PSA film was adhered to a stainless steel (SUS-430BA) and a Teflon sheet by means of a rubber roll with a pressure of 6 kg/cm and a press rate of 2 m/min. And the roll of PSA film was assumed that PE film was an adherend, because the surface of PSA layer contacted with backside of PE backing film during storage period. These specimens were stored at 65°C .

On the other hand, the rolls of PSA films were stored in two glass jars to simulate dry and humid atmospheres. One jar contained a saturated ammonium chloride solution and was maintained at 20°C , generating 79.2% relative

humidity. The other jar was desiccated with phosphorus pentoxide.

2.4. FTIR measurements

Transmission and ATR-FTIR spectra were collected on a Nicolet Model MAGNA-IR 860 Spectrometer (Thermo Nicolet Japan Co., Ltd. Kanagawa, Japan) equipped with a liquid nitrogen cooled mercury cadmium telluride (MCT) detector. An attenuated total reflectance accessory (Spectra-Tech, Inc., Shelton, CT) was utilized. In order to vary the depth of penetration into the PSA films, three kinds of internal reflection elements (IRE) were used, i.e. ZnSe (Zinc Selenium) crystals for 45° and 60° angles of incidence and a Ge (Germanium) crystal for 60° angle of incidence. Spectral features of overlapping bands were decomposed using a peak solving software, PEAKSOLVE (Thermo Galactic, Salem, NH).

2.5. Contact angle measurements

Surface tensions of the PSA films and the adherends used were obtained from experimental determinations of contact angle for sessile drops. The static contact angles of the PSA films and the adherends used were measured with the CA-A contact angle meter (Kyowa Interface Science Co., Ltd. Tokyo, Japan) at 20°C . In order to measure the contact angle of the PSA films, they were fixed to a sample stage with a double-faced tape. Surface tensions of pure water, formamide, diiodomethane, α -bromonaphthalene and ethyl malonate are 72.5, 58.2, 50.8, 42.7 and 31.8 (mN/m), respectively. Zisman plot was utilized to obtain a critical surface tension (γ_c) of the PSA films and the adherends.

2.6. Refractive index measurements

The refractive indices of the PSA films were measured with the 1-T Abbe Refractometer (Atago Co., Ltd. Tokyo, Japan) at 20°C .

2.7. XPS analysis

The XPS spectra were collected on Electron Spectrometer ESCA-850 (Shimadzu co., Kyoto, Japan). The XPS measurement was performed with Mg K_{α} X-ray source at 7 kV and 30 mA. The emission angle was 30° .

3. Results and discussion

3.1. FTIR spectrum

Fig. 1 shows FTIR spectra of the polyacrylic resin, CORONATE L and PSA film where the crosslinker was formulated. Trace A and B in Fig. 1 show transmission FTIR spectra of the polyacrylic resin and CORONATE L,

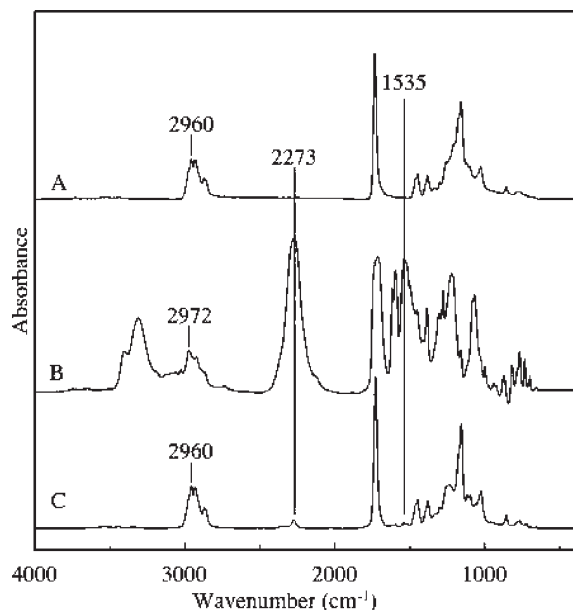


Fig. 1. Transmission FTIR spectra of acrylic resin (A) and CORONATE L (B) and ATR-FTIR spectra of the PSA film after pretreatment at 80 °C for 2 min (C).

respectively. The band at 2960 cm^{-1} in trace A is due to CH_2 stretching mode. Trace B illustrates a pronounced band at 2273 cm^{-1} due to NCO asymmetric stretching mode. The band at 1535 cm^{-1} is due to amide II band which is a mixed mode containing contributions from the N–H in-plane bending, the C–N stretching and the C–C stretching vibrations [8]. And the band at 2982 cm^{-1} is attributed to the CH stretching mode. Trace C in Fig. 1 represents ATR-FTIR spectrum of PSA film where the crosslinker was formulated. Of particular interest is the band due to NCO groups at 2273 cm^{-1} because this band decreased as crosslinking reaction proceeded [1]. Thus this band was used for the measurements of relative amount of unreacted NCO groups. The band at 1535 cm^{-1} is due to amide II band of urethane and urea linkages contained in CORONATE L and the products obtained from crosslinking reaction. This band increased as crosslinking reaction proceeds [2]. Thus this band was used to measure relative amount of urethane and urea linkages and decomposed into individual absorption bands with a peak resolving method. Fig. 2 shows the results of the peak resolving in amide II region from 1590 to 1490 cm^{-1} for the PSA film after pretreatment at 80 °C for 2 min and aging at 23 °C for 2 days. The spectrum in this region is resolved into four bands at 1554, 1536, 1520 and 1505 cm^{-1} . Assignments of these four bands were reported in our previous paper [2]. In an effort to minimize the effects of sample crystal coverage on the band intensity, all ATR-FTIR spectra were normalized to the CH_2 stretching vibrations at 2960 cm^{-1} in the polyacrylic resin used [9–11], because the intensity of the CH stretching band at 2972 cm^{-1} in CORONATE L which have been charged only 3 phr is considered to be negligible

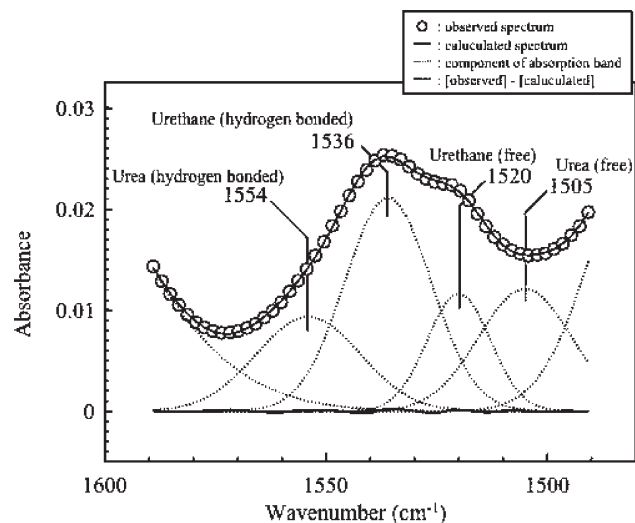


Fig. 2. Curve-fitting results in amide II region of the PSA films after pretreatment at 80 °C for 2 min and aging at room temperature for 2 days.

and because the methylene groups in the resin are stable during the crosslinking reaction.

3.2. The effect of adherends on distribution of functional groups in the PSA film

Crosslinking reaction between the acrylic base polymer and polyisocyanate crosslinker in the PSA film on the various adherends was investigated by ATR-FTIR measurements. Refractive indices of ZnSe, Ge and the PSA films used are 2.4, 4.0 and 1.5, respectively. Refractive indices both of the acrylic PSA film and the crosslinked PSA film are equal to 1.5. Thus the depths of penetration (d_p), as defined by Harrick [12], were estimated to be about 0.88 and 0.49 μm with ZnSe crystals for 45 and 60° angles of incidence, respectively, and about 0.22 μm with a Ge crystal for 60° angle of incidence for the isocyanate band at 2273 cm^{-1} . And the d_p for the amide II band at 1535 cm^{-1} were estimated to be about 1.31 and 0.72 μm with ZnSe crystals for 45 and 60° angles of incidence, respectively, and about 0.33 μm with a Ge crystal for 60° angle of incidence.

The relative amount of isocyanate groups ($A(\text{NCO})/A(\text{CH})$ ratio) of the crosslinked PSA films used Teflon sheets as the adherends is plotted as a function of the contact time in Fig. 3. $A(\text{NCO})/A(\text{CH})$ ratio decreases exponentially with an increase of the contact time at every d_p investigated. On the other hand $A(\text{NCO})/A(\text{CH})$ ratio increases with an increase of the d_p throughout our observation period. The contact time when NCO band at 2273 cm^{-1} diminishes is around 25 days at every d_p investigated. $A(\text{NCO})/A(\text{CH})$ ratio of the PSA films which used the PE films as the adherends showed almost the same pattern to those used Teflon sheets as the adherends.

On the other hand, the relative amount of isocyanate groups ($A(\text{NCO})/A(\text{CH})$ ratio) of the crosslinked PSA films used the stainless steels as the adherends is plotted as a

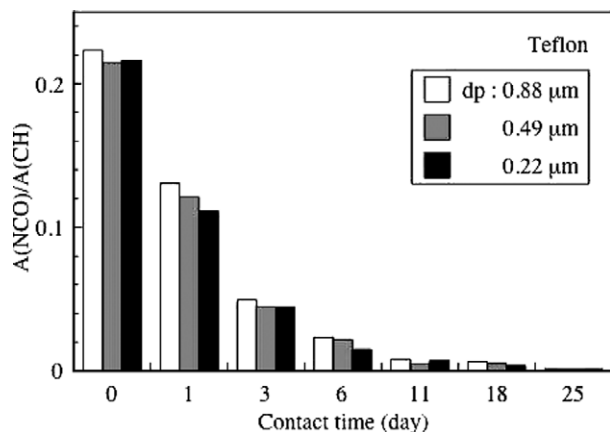


Fig. 3. A(NCO)/A(CH) ratio of the crosslinked PSA film as a function of the contact time used Teflon sheet as the adherend.

function of the contact time in Fig. 4. A(NCO)/A(CH) ratio increases with an increase of the d_p in the early stages of crosslinking reaction. Then A(NCO)/A(CH) ratio decreases exponentially with the increase of the contact time at every d_p investigated, which decreases slower than when using Teflon as adherend. And A(NCO)/A(CH) ratio decreases with an increase of the d_p in the latter stages of crosslinking reaction, of which ratio is too small that might be within the experimental error.

These results may possibly indicate that there are more isocyanate groups at the bulks of the PSA layers than at the interfaces between the PSA layers and the Teflon sheet or the PE film, while there are more isocyanate groups at the interface between the PSA layer and the stainless steel than at the bulk of the PSA in the latter stages.

In order to clarify the effect of adherends on the crosslinking reaction in more detail, formations of urethane and urea linkages during crosslinking reaction were monitored. Fig. 5 shows plots of the relative amount of the hydrogen-bonded urethane band (A(H-Urethane)/A(CH) ratio) at 1536 cm^{-1} as a function of the contact time for the PSA film adhered to a Teflon sheet. A(H-Urethane)/A(CH) ratio increases with an increase of the

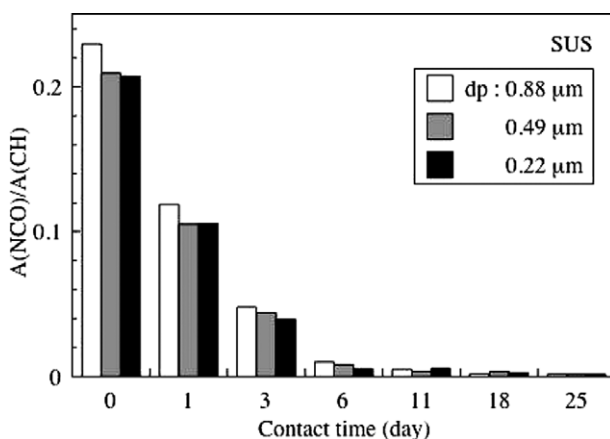


Fig. 4. A(NCO)/A(CH) ratio of the crosslinked PSA film as a function of the contact time used the stainless steel as the adherend.

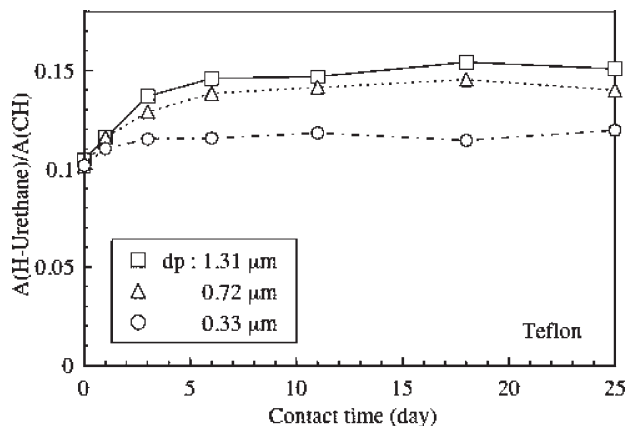


Fig. 5. A(H-Urethane)/A(CH) ratio of the crosslinked PSA film as a function of the contact time used Teflon sheet as the adherend and stored at $65\text{ }^{\circ}\text{C}$.

contact time at every d_p investigated. The increment in A(H-Urethane)/A(CH) ratio depends on the d_p . As the d_p increases, the increment in A(H-Urethane)/A(CH) ratio increases. A(H-Urethane)/A(CH) ratio of the PSA film thus obtained increases with an increase of the distance from the interface between the PSA layer and a Teflon sheet. A(H-Urethane)/A(CH) ratio of the PSA film adhered to the PE film showed much the same pattern shown in Fig. 5.

Similarly, A(H-Urethane)/A(CH) ratio of the PSA film adhered to a stainless steel is plotted as a function of the contact time shown in Fig. 6. A(H-Urethane)/A(CH) ratio increases with an increase of the contact time at every d_p investigated. The increment in A(H-Urethane)/A(CH) ratio is dependent on the d_p . As the d_p increases, the increment in A(H-Urethane)/A(CH) ratio decreases. A(H-Urethane)/A(CH) ratio of the PSA film thus obtained decreases with an increase of the distance from the interface between the PSA layer and a stainless steel.

These results suggest that the amounts of the hydrogen-bonded urethane linkage depend on the distances from the interfaces between the PSA layers and the adherends.

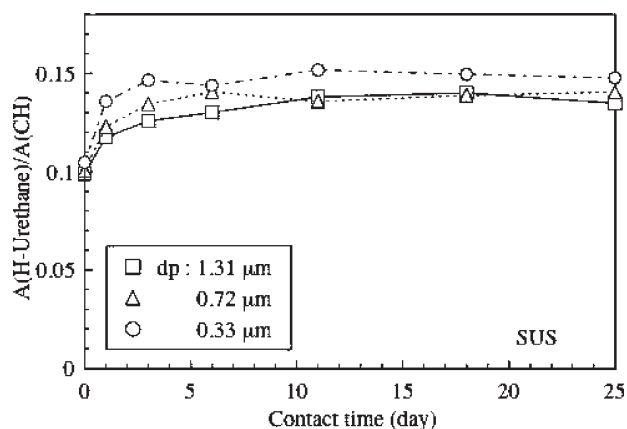


Fig. 6. A(H-Urethane)/A(CH) ratio of the crosslinked PSA film as a function of the contact time used the stainless steel as the adherend and stored at $65\text{ }^{\circ}\text{C}$.

For more information, A(H-Urethane)/A(CH) ratio of the fully crosslinked PSA film is plotted as a function of the d_p in Fig. 7. A(H-Urethane)/A(CH) ratio of the PSA adhered to a stainless steel decreases as the d_p increases. On the other hand, for the PSA films adhered to a Teflon sheet and a PE film, A(H-Urethane)/A(CH) ratio increases as the d_p increases. It is obvious that there are more hydrogen-bonded urethane linkages at the interface between the PSA layer and a stainless steel than the bulk of the PSA layer. In contrast, for the PSA film adhered to a Teflon sheet and a PE film, there are more hydrogen-bonded urethane linkages at the bulk of the PSA layer than at the interface between the PSA layer and the adherends. The tendency for the distribution of the hydrogen-bonded urethane linkages in the PSA film is similar to that of the unreacted isocyanate groups in the PSA film shown in Figs. 3 and 4. And similar tendencies were also observed for the distributions of the free urethane, the hydrogen-bonded urea and the free urea linkages in the PSA film. Therefore, it is considered that the polyisocyanate crosslinker and its derivatives migrate in the PSA layer. Moreover, the tendency for the polyisocyanate and its derivatives to migrate would differ with the adherends used.

3.3. The effect of adherends on the surface tension of the PSA film

Critical surface tensions (γ_c) of the acrylic and the fully crosslinked PSA films adhered to various adherends were obtained from the static contact angles for sessile drops of various testing liquids. γ_c of the adherends used in this report are 40.2, 30.3 and 18.0 (mN/m) for SUS430BA, Polyethylene and Teflon, respectively.

Fig. 8 shows a plot of γ_c of the acrylic and the fully crosslinked PSA film as a function of γ_c of the adherends used. γ_c of the acrylic PSA film shows no significant changes even though those of the adherends used changes from 18 to 40 mN/m. On the other hand, γ_c of the fully

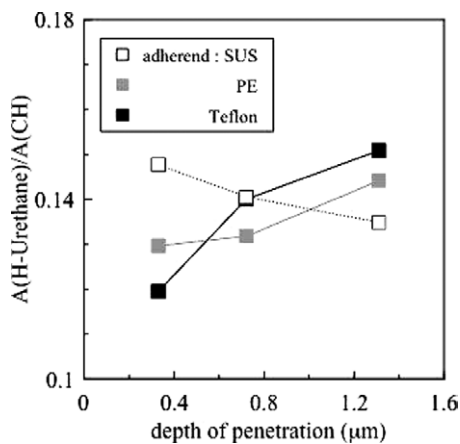


Fig. 7. Depth profile of A(H-Urethane)/A(CH) ratio for the crosslinked PSA film stored at 65 °C for 25 days.

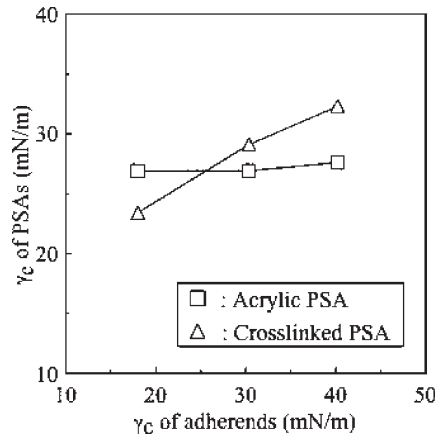


Fig. 8. Critical surface tension of the acrylic and the crosslinked PSA film stored at 65 °C for 25 days as a function of critical surface tension of the adherends used.

crosslinked PSA film increases with an increase of those of the adherends used.

As mentioned above, it is suggested that the polyisocyanate crosslinker and its derivatives segregate preferentially to the interface between the PSA film and a stainless steel having a higher surface free energy than a PE film or a Teflon sheet. The crosslinker and its derivatives would segregate to the bulk of the PSA film when the PSA film was adhered to a PE film or a Teflon sheet, which had lower surface energies compared with the stainless steel. Therefore, it seems that the segregations of the crosslinker and its derivatives are related to the variation of γ_c of the fully crosslinked PSA film.

In an effort to examine an effect of a surface structure on a surface tension, the relative amounts of urethane and urea linkages were observed as a function of the d_p for the rolls of the crosslinked PSA film stored under the dry and the humid conditions. Fig. 9 shows plots of A(H-Urethane)/A(CH) and A(H-Urea)/A(CH) ratios of the fully crosslinked PSA films as a function of the d_p . A(H-Urethane)/A(CH) ratio of the

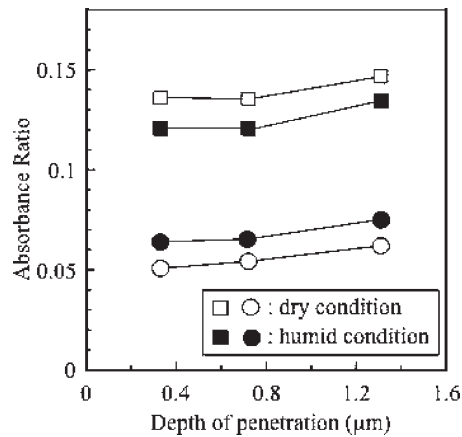


Fig. 9. Depth profile of A(H-Urea)/A(CH) ratio (circle) and A(H-Urethane)/A(CH) ratio (square) for the crosslinked PSA films stored at 23 °C for 180 days under the dry and the humid condition used PE film as the adherend.

PSA film stored under the dry condition is higher than that of stored under the humid condition at every d_p observed. Under both conditions, A(H-Urethane)/A(CH) ratio increases with an increase of the d_p . Moreover, tendency observed for the free urethane linkages was similar to the hydrogen-bonded urethane shown in Fig. 9.

In contrast to urethane linkages, A(H-Urea)/A(CH) ratio of the PSA film stored under the humid condition is higher than that stored under the dry condition at every d_p observed. Under both conditions, A(H-Urea)/A(CH) ratio increases with an increase of the d_p , and the relative amount of the free urea linkages showed similar tendency to that of the hydrogen-bonded urea linkages shown in Fig. 9.

These results apparently indicate that there are more urea linkages on the surface of the PSA stored under the humid condition than that stored under the dry condition. In contrast, there are more urethane linkages on the surface of the PSA stored under the dry condition than that stored under the humid condition.

In order to make out the effect of a difference in the amount of urethane and urea linkages at surface of the fully crosslinked PSA film on the surface properties of these PSA films, surface tensions of these PSA films are obtained. Fig. 10 shows a Zisman plot of the fully crosslinked PSA films stored under the dry and the humid conditions for 180 days. It is obvious that a Zisman plot of the fully crosslinked PSA film stored under the dry condition is in good agreement with that of the fully crosslinked PSA film stored under the humid condition. And γ_c of these PSA films are approximately 28 mN/m, which is agreed with γ_c of the fully crosslinked PSA film adhered the PE film shown in Fig. 8. This finding suggests that γ_c of the PSA film is independent of a difference in the amount of urethane and urea linkages at the interface between the PSA film and the PE film. Moreover, the γ_c of the PSA film seems to be attributed to migrations of the polyisocyanate crosslinker and its derivatives.

Okamoto et al. indicated that the surface free energy of a dimethoxysilyl-terminated polypropylene oxide (DMSi-

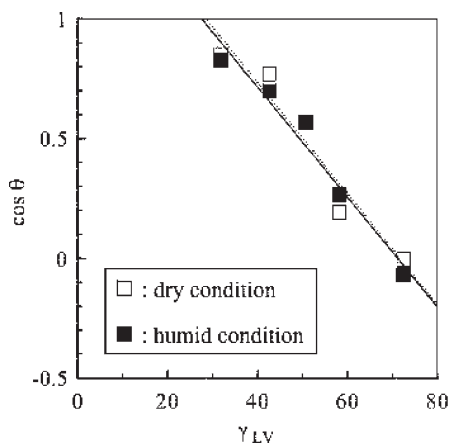


Fig. 10. Zisman plot for the crosslinked PSA films stored at 23 °C for 180 days under the dry and the humid conditions used PE film as the adherend.

PPO)/diglycidylether of bisphenol A (DGEBA) blend had varied with the surface free energies of several polymeric substrates to minimize the blend-substrate interfacial free energy, as a result of segregation of DGEBA to the interface on high surface free energy substrates and that of DMSi-PPO to the interface on low surface free energy substrates [13,14].

Our results suggest that the polyisocyanate and its derivatives segregate to the interface on a higher γ_c adherend such as the stainless steel. Based on the parachor defined by Sugden [15], γ_c of the crosslinker is calculated to be 81 mN/m, which is much higher than γ_c of the acrylic base resin of 27 mN/m as shown in Fig. 8.

The above facts suggested that the more polyisocyanate having higher γ_c exists in the interface between PSA and stainless steel having relatively higher γ_c , which forms PSA surface of higher γ_c .

Then, the chemical structures of PSA surface were determined by XPS analysis. Fig. 11 shows C1s spectra of PSA surface stored at 65 °C for 25 days adhered to three substrate having different surface tensions. In C1s spectrum of PSA surface adhered to stainless steel (a), two peaks are observed at 288.5 and 285 eV, which are assigned to carbon atom of O–C=O and C–C, respectively. The shoulder around 286.5 eV is assigned as C atom of C–O. The above C atoms are derived from acrylic base polymer. Similar spectrum was observed in C1s (b) using PE as adherend. However, new peak of 292 eV is detected in C1s (c) using Teflon as adherend, which is assigned as C atom of F–C–F. The above fact indicates that the component of Teflon was transferred to the PSA surface. But, this phenomenon does not affect the surface properties of PSA, because the surface

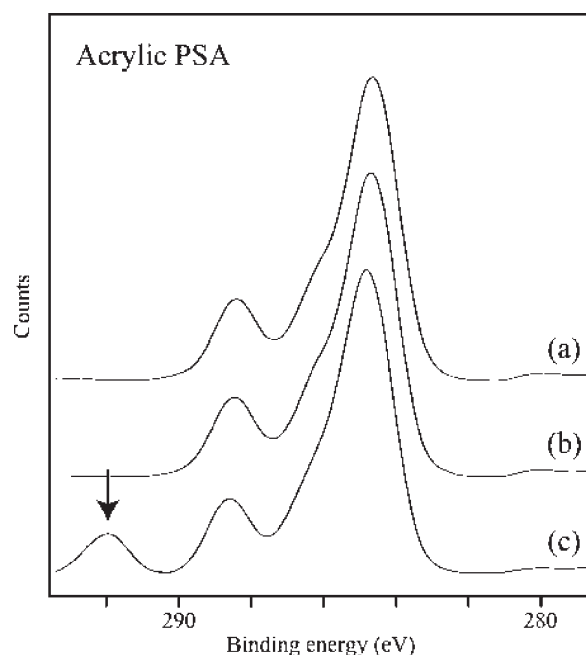


Fig. 11. XPS C1s spectra of the acrylic PSA film adhered to the stainless steel (a), the PE film (b) and Teflon sheet (c).

tensions of non-crosslinked PSA were same against various adherends as shown in Fig. 8. Similar tendency was observed in crosslinked PSA surface shown in Fig. 12, where the transfer of CF₂ makes a little affect on the surface properties of PSA. The distinct reason why the surface tension for PSA unchanged is not known. But it can be thought that Teflon is not transferred to the PSA surface thoroughly but dispersedly, which did not change the surface tensions of PSAs.

Figs. 13 and 14 show the N1s spectra of PSA and crosslinked PSA surfaces, respectively. In the PSA surface, no peak derived from N atom was observed, which is reasonable that there is no N atom in the linear PSA. In the case of crosslinked PSA, a sharp peak around 400 eV derived from N atom is observed when using stainless steel as adherent. The formulation of the above PSA is polyacrylate with 3 phr of CORONATE L as a crosslinker, of which calculated C/N ratio is 100/0.6, and the magnitude is less than the detection limit of XPS. Considering the above fact, the more N atom exists in the interface of PSA and stainless steel than calculated N atom. As a consequence, it is shown that CORONATE L or its derivatives segregate to the interface of stainless steel and crosslinked PSA.

4. Conclusions

The reaction of isocyanate in PSA films adhered on to a various adherends having different surface tensions was monitored using ATR-FTIR analysis.

In the latter stages of crosslinking reaction, unreacted

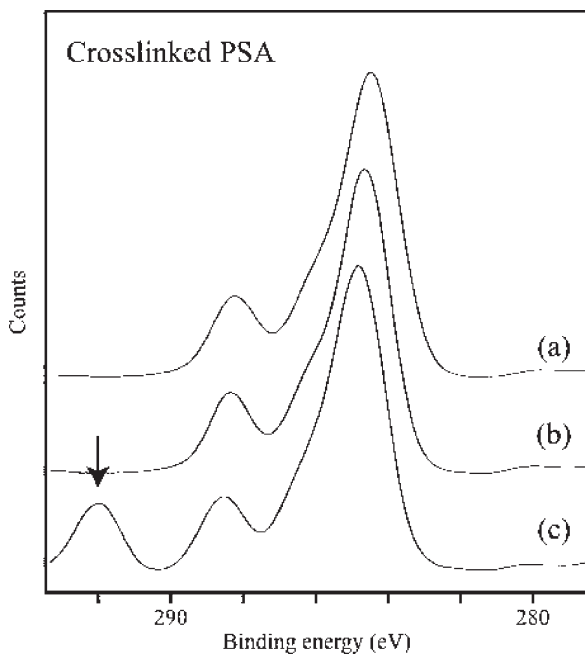


Fig. 12. XPS C1s spectra of the crosslinked PSA film adhered to the stainless steel (a), the PE film (b) and Teflon sheet (c).

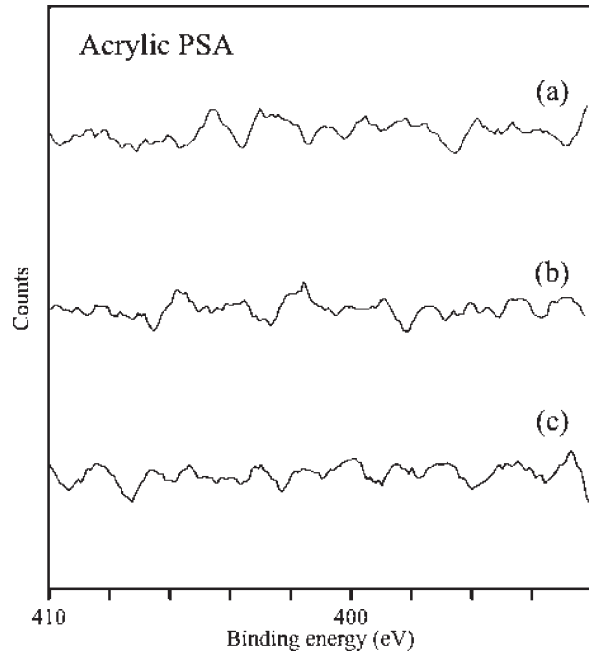


Fig. 13. XPS N1s spectra of the acrylic PSA film adhered to the stainless steel (a), the PE film (b) and Teflon sheet (c).

isocyanate and its derivatives exist more in the bulk of the PSA than in the interfaces between PSA and adherends that are Teflon sheet and PE film having relatively lower surface tensions. In the case of using stainless steel having relatively higher surface tension as adherend, opposite segregation was observed compared to Teflon and PE.

From the XPS analysis, N atoms segregate more in surface than in bulk when stainless steel is used as adherend. We conclude that polyisocyanates migrate in the PSA film

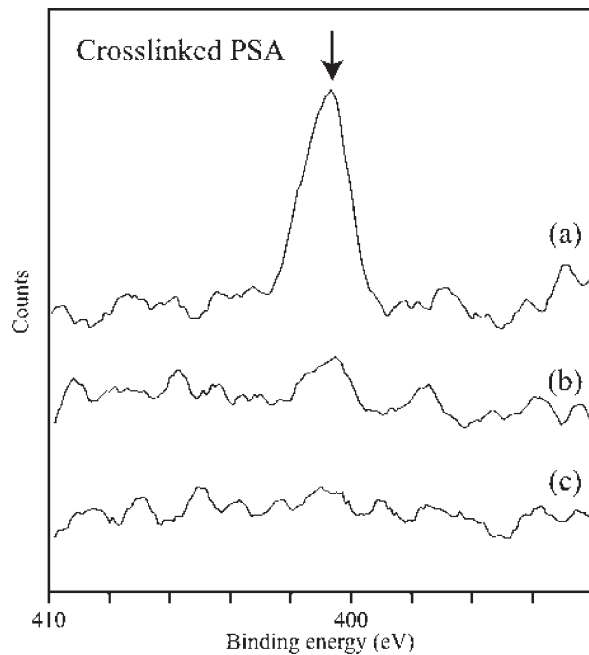


Fig. 14. XPS N1s spectra of the crosslinked PSA film adhered to the stainless steel (a), the PE film (b) and Teflon sheet (c).

in order to minimize the magnitude of interfacial free energy between the PSA and the adherend, which leads to the change of surface tension of PSA film.

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References

- [1] Asahara J, Hori N, Takemura A, Ono H. *J Appl Polym Sci* 2003;87:1493–9.
- [2] Asahara J, Sano A, Hori N, Takemura A, Ono H. *J Appl Polym Sci* 2003;89:3039–45.
- [3] Tanaka K, Takahara A, Kajiyama T. *Macromolecules* 1996;29:3232–9.
- [4] Brant P, Karim A, Douglas JF, Bates FS. *Macromolecules* 1996;29:5628–34.
- [5] Tanaka K, Takahara A, Kajiyama T. *Macromolecules* 1998;31:863–9.
- [6] Tanaka K, Takahara A, Kajiyama T. *Macromolecules* 1998;31:3746–9.
- [7] Kasemura T. *J Adh Soc Jpn* 1993;29:273.
- [8] Skrovanek DJ, Howe SE, Painter PC, Coleman MM. *Macromolecules* 1985;18:1676–83.
- [9] Kaminski AM, Urban MW. *J Coat Technol* 1997;69(872):55–66.
- [10] Kaminski AM, Urban MW. *J Coat Technol* 1997;69(873):113–21.
- [11] Cole KC, Gheluwe PV, Hebrard MJ, Leroux J. *J Appl Polym Sci* 1987;34:395–407.
- [12] Harrick NJ. *J Opt Soc Am* 1965;55:851–7.
- [13] Okamatsu T, Yasuda Y, Ochi M. *J Appl Polym Sci* 2001;80:1920–30.
- [14] Okamatsu T. *J Adh Soc Jpn* 2001;37:358–64.
- [15] Van Krevelen DW, Hoftyzer PJ. *Properties of polymers, their estimation and correlation with chemical structure*. Amsterdam: Elsevier; 1976.