

Improvement in the Adhesion of Polyimide/Epoxy Joints Using Various Curing Agents

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ABSTRACT: An improvement in the adhesion strength of polyimide/epoxy joints was obtained by (1) introducing a functional group on the polyimide surface, (2) improving the mechanical properties of the epoxy adhesive, (3) increasing the curing temperature, and (4) using polyamic acid as an adhesion-promoting layer. The functional group on polyimide was introduced via treatment with aqueous KOH. An adhesion-promoting layer was formed by spin coating polyamic acid onto a modified polyimide surface. The maximum adhesion strength of the polyimide/epoxy joint was obtained using polyamic acid as both the adhesion-promoting

layer and as the curing agent. The surface energy of the modified polyimide was examined using contact angle measurements and Fourier transform infrared spectroscopy, and the peel strength was determined by the T-peel method. The peeled surfaces were analyzed using scanning electron microscopy and X-ray photoelectron spectroscopy. © 2002 Wiley Periodicals, Inc. *J Appl Polym Sci* 86: 812–820, 2002

Key words: surface modification; polyimide; adhesion; epoxy adhesive; polyamic acid

INTRODUCTION

An improvement in the interfacial adhesion of polyimide (PI) thin films with epoxy adhesive layers is required for the fabrication of compact and light-weight electronic components, such as three-dimensional circuits and flexible printed circuits. PI has many advantages for the above purpose, such as excellent mechanical properties, high thermal stability, low dielectric constant, and low moisture absorption.¹ However, PI shows poor adhesion to epoxy adhesives and to copper. This arises from the fact that there are no reactive functional groups on the surface of a PI film. To improve its adhesion strength, a PI surface can be modified by dry processes such as plasma,² ion beam,³ or ammonia treatment^{4,5} and by wet processes such as treatment with potassium hydroxide,⁶ sodium hydroxide, or amine solutions.⁷ Wet processes are preferred commercially over the dry processes because of their simplicity and cost effectiveness. In particular, treatment with KOH has been well documented because of its simple and fast processing route. As shown in Figure 1, a PI such as pyromellitic dianhydride/oxydianiline (PMDA/ODA) reacts with KOH to give a polyamate (the potassium salt of polyamic

acid), which is subsequently protonated using hydrochloric acid to give the corresponding polyamic acid. The functional group, which in polyamic acid is the –OH group, is supposed to provide a better wettability and, thus, better adhesion of a PI/epoxy joint. For good adhesion between PI and an epoxy adhesive, it is necessary to improve the intermolecular wetting, the chemical reaction (curing), and the interlayer diffusion. The generally accepted mechanisms of adhesive/adherend adhesion include the mechanical interlocking theory and theories based on surface energies, wetting, adsorption, diffusion theory, and chemical bonding.⁸ Among these theories, wetting is the one concerned with the establishment of intimate molecular contact as a prerequisite for developing strong adhesion. Intermolecular forces, which are usually divided into polar and dispersion forces, can be achieved by creating intimate molecular contact between two materials to attain good adhesion. The degree of wetting can be quantitatively defined by the contact angle. Good adhesion strength can be established by forming a covalent bond at the interface because covalent chemical bonds are stronger than intermolecular forces. In addition, the diffusion of polymer molecules across the interface, allowing the polymeric chains to entangle, seems to be generally accepted as playing an important role in the polymer–polymer adhesion process. However, neither chemical bonding nor interdiffusion can be established in case

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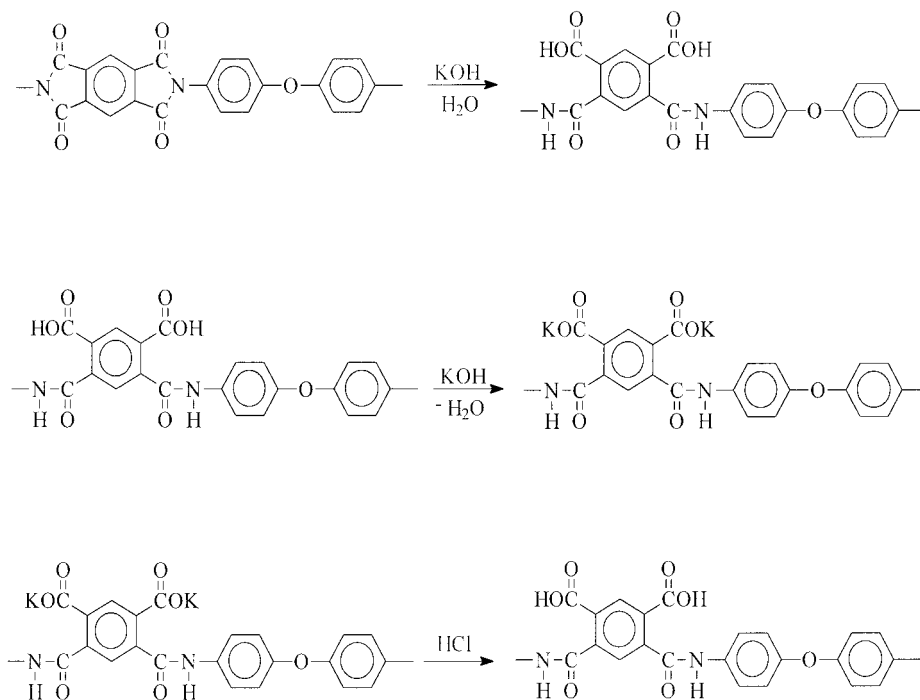


Figure 1 Schematic of the imide ring opening on Kapton H polyimide films by the KOH treatment at 22°C.

of poor wettability because intermolecular contact cannot be achieved. Therefore, good wettability is a prerequisite for achieving good adhesion.

Epoxy adhesives, which were used in this research, have been widely employed as an adhesive in the electronics industry because they have good electrical properties, thermal and chemical resistance, and adhesion. However, epoxy adhesives cannot absorb and distribute stress, because of their brittleness. It can be assumed that the toughness of an adhesive can affect its adhesion strength: The adhesion strength increases with increasing toughness of adhesive.⁸

In this research, four approaches were considered to increase adhesion strength. First, functional groups were introduced on the PI surface to achieve chemical bonding between the PI and epoxy adhesive. Second, the toughness of the epoxy adhesive was increased using polyamic acid as a curing agent to absorb and distribute stress. Third, the curing temperature was increased using high-temperature curing agents, and the effect of curing temperature was investigated. Fourth, a polyamic acid layer was used as an adhesion-promoting layer between the modified PI and the epoxy adhesive to facilitate intermolecular diffusion. The effects of the following parameters on adhesion strength were investigated: (1) the molar concentration of aqueous KOH, (2) the treatment time of PI, (3) the toughness of the epoxy adhesive, and (4) the concentration of polyamic acid (wt%) mixed with the epoxy adhesive. In addition, the effect of having a polyamic acid layer between the modified PI film and the epoxy adhesive was also investigated.

EXPERIMENTAL

Materials

The PI film used was Kapton H (pyromellitic dianhydride/4,4'-oxydianiline [PMDA/ODA]), produced by Dupont. PMDA/ODA polyamic acid was synthesized by mixing stoichiometric quantities of PMDA (TCI, Japan) and ODA (TCI, Japan) in the solvent *N*-methyl-2-pyrrolidone (NMP; Junsei, Japan). Synthesis of 6FDA/ODA polyamic acid was carried out using 4,4'-(hexafluoro-isopropylidene)diphthalic anhydride (Aldrich) and ODA according to the same process used for PMDA/ODA.⁹ All the reagents were used as received, without further purification. The epoxy resins used were diglycidyl ether of bisphenol A epoxy resin (DGEBA: YD-128[®], epoxy equivalent weight = 180–190 g/eq) and rubber-modified epoxy resin (R-1309[®], epoxy equivalent weight = 300 g/eq), manufactured by Kukdo Chemicals in Korea, and linear amide type (G-0331, Kukdo Chemical) and 3,3'-methylenedianiline (MDA; TCI, Japan) were used as the curing agents, respectively. An aqueous KOH (Junsei) solution was used for the surface treatment of the PI.

Chemical surface modification of the PI film

PMDA/ODA PI films were treated with 0.5, 1, and 2 M aqueous KOH solutions at room temperature for times between 3 and 60 min, followed by washing with distilled water. The optimum conditions were found using 1 M aqueous KOH solution. The optimum conditions were found using 1 M aqueous

TABLE I
Composition and Curing Condition of the Epoxy Adhesives.

Type	Epoxy resin	Curing agent	Solvent	Curing condition
Low temp.	DGEBA	Linear amide type, 40 phr ^a		80°C, 3 h
	Rubber modified epoxy	Linear amide type, 28 phr		80°C, 3 h
Medium temp.	DGEBA	MDA, 28 phr	THF	80°C, 2 h then 150°C, 2 h
	Rubber modified epoxy	MDA, 17.5 phr	THF	80°C, 2 h then 150°C, 2 h
High temp.	DGEBA	<i>Polyamic acid</i>	NMP	120°C, 2 h then 205°C, 5 h

^a part per hundred of resin

KOH solution. The samples were then dried at room temperature for 12 h. The resulting modified surface consisted of potassium polyamate, which was converted to polyamic acid by treatment with 0.2 M HCl solution for 6 min, followed by a rinse with water.

Preparation of PI/epoxy adhesive joints

For the low- and medium-temperature curing agents, the epoxy adhesives were mixed with the linear amide and the 3,3'-methylenedianiline and then poured onto a PI film, which was then cured at various temperatures in a hot press. For high-temperature curing agents, the synthesized PMDA/ODA polyamic acid was coated onto the modified PI films using a spin coater (forming a multilayer joint). The epoxy adhesives were mixed with polyamic acid and were then poured onto the coated PI films, which were then cured at 120°C for 2 h, followed by curing at 205°C for 5 h. The whole sandwich structure was cured in a hot press. In addition, modified PI film/epoxy adhesive joints were prepared without a polyamic acid inter-layer (a single-layer joint), and the effect of the inter-

layer was investigated. The compositions and curing conditions of the epoxy adhesives are listed in Table I.

Peel test and mechanical properties of the epoxy adhesives

The peel strength of various PI/epoxy adhesive joints was measured by a T-peel test using a Universal Testing Machine (Instron Model 4465) with a 254 mm/min peel rate according to the ASTM D1876 procedure. The mechanical properties of the epoxy adhesives were also measured at room temperature using the Instron. A specific gauge length of 25 mm and a cross-head speed of 50 mm/min were used according to the ASTM D638 procedure.

Surface analysis

The surface energy of the KOH-modified PI was determined by measuring the contact angle from the sessile drop method using a Contact Angle Analyzer (Model G-1, Erma Co.).¹⁰ The following equations⁸ were used to obtain the disperse (γ^d) and polar (γ^p) components of the surface energy of PI films. The

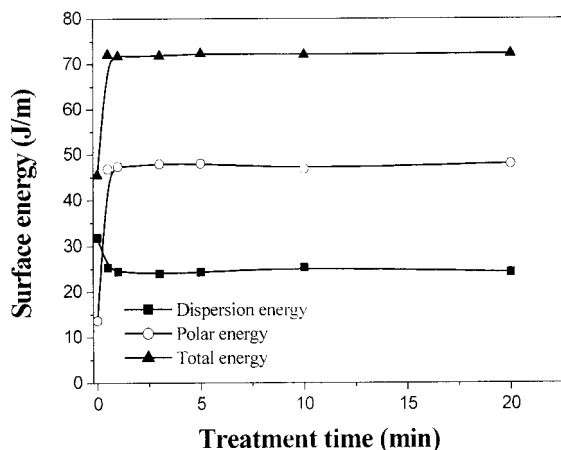


Figure 2 The surface energy of the pyromellitic dianhydrid-4,4'-oxydianiline (PMDA-ODA) films as a function of treatment time of PMDA/ODA in 1 M aqueous KOH solution.

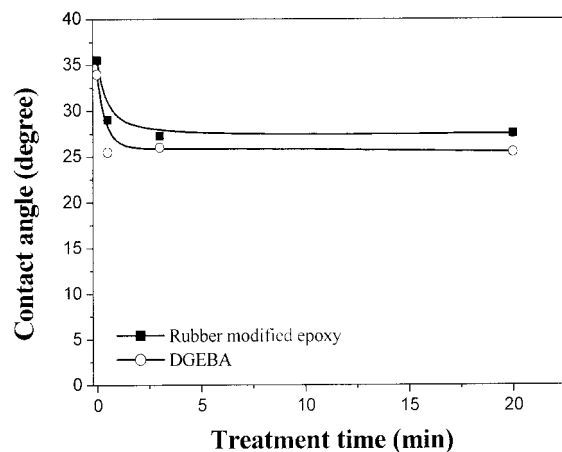


Figure 3 The contact angle of pyromellitic dianhydrid/4,4'-oxydianiline (PMDA/ODA)-epoxy resin as a function of treatment time of PMDA/ODA in 1 M aqueous KOH solution.

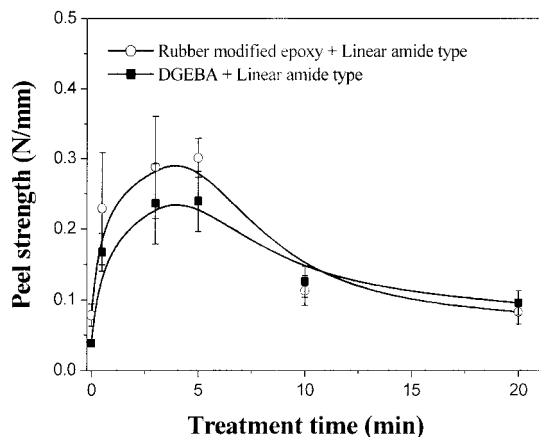


Figure 4 The peel strength of epoxy adhesive (diglycidyl ether of bisphenol A epoxy resin + linear amide type, rubber-modified epoxy + linear amide type)/polyimide film (pyromellitic dianhydrid/4,4'-oxydianiline [PMDA/ODA]) joints as a function of treatment time of PMDA/ODA in 1 M aqueous KOH solution.

contact angle of the modified PI films was determined by using two test liquids: water (subscript 1) and methylene iodide (subscript 2).

$$\gamma_1(\cos \theta) = 2[(\gamma_1^d)^{1/2}(\gamma^d)^{1/2}] + 2[(\gamma_1^p)^{1/2}(\gamma^p)^{1/2}];$$

$$\gamma_2(\cos \theta) = 2[(\gamma_2^d)^{1/2}(\gamma^d)^{1/2}] + 2[(\gamma_2^p)^{1/2}(\gamma^p)^{1/2}].$$

The surface energy γ , the disperse component γ^d , and the polar component γ^p values used were as reported in the literature.⁸ These were 72.2, 22.0, and 50.2 mJ/m² for water and 50.8, 48.5, and 2.3 mJ/m² for methylene iodide, respectively.

The Fourier transform infrared (FTIR) spectra of the modified PI films were obtained using a Nicolet Co. Magna-IR 550 FTIR spectrometer. To obtain an almost fully modified PI film, the PI films were immersed in 6 M aqueous KOH solution at room temperature for 6 h. Then the KOH-treated PI films were dried in a convection oven at various temperatures for 4 h. The surface morphologies of the peeled surface of high-temperature cured samples were analyzed using a JEOL JSM-6300 scanning electron microscope (SEM). The surface atomic composition of the high-temperature cured samples was detected using an SSI 2803-S X-ray photoelectron spectrometer (XPS). To examine the failure locus of the multilayer joints, an interlayer composed of 6FDA/ODA polyamic acid was used instead of PMDA/ODA polyamic acid.

RESULTS AND DISCUSSION

Surface energy and contact angle of modified PI films

Figure 2 shows the change over time of the surface energy of a PI film treated with 1 M aqueous KOH

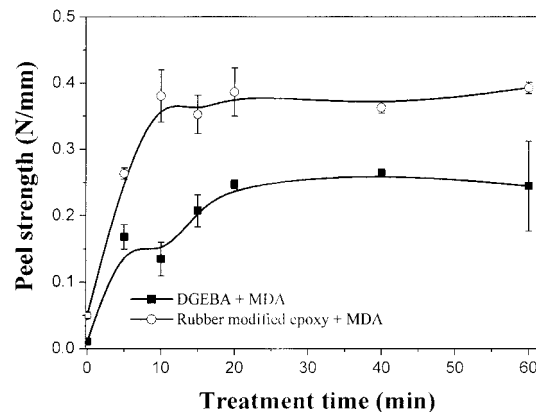


Figure 5 The peel strength of epoxy adhesive (diglycidyl ether of bisphenol A epoxy resin + 3,3'-methylendianiline [MDA], linear amide type + MDA)/pyromellitic dianhydrid/4,4'-oxydianiline (PMDA/ODA) joints as a function of treatment time of PMDA/ODA in 1 M aqueous KOH solution.

solution. The surface energy rapidly increased after 30 s, and the total surface energy and the polar surface energy were higher, and the disperse surface energy lower, than those of an unmodified PI film. These phenomena can be explained by the carboxylic acid (-COOH) group introduced on the PI surface by modifying the PI to polyamic acid, as illustrated in Figure 1. Figure 3 shows the contact angles of the modified PI surface as a function of the treatment time in 1 M aqueous KOH solution, using the epoxy resins that were used as adhesives in this research. Figure 3 shows the contact angles of the modified PI surface as a function of the treatment time in 1 M aqueous KOH solution, using the epoxy resins that were used as adhesives in this research. When compared with an

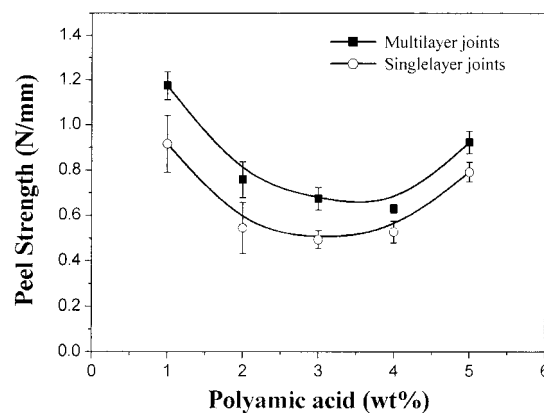


Figure 6 The peel strength of epoxy adhesive (diglycidyl ether of bisphenol A epoxy resin + polyamic acid)/polyimide film (pyromellitic dianhydrid/4,4'-oxydianiline) joints as a function of concentration of polyamic acid in epoxy adhesive. (Multilayer = modified polyimide + spin coated polyimide layer + epoxy adhesive. Single layer = modified polyimide + epoxy adhesive.)

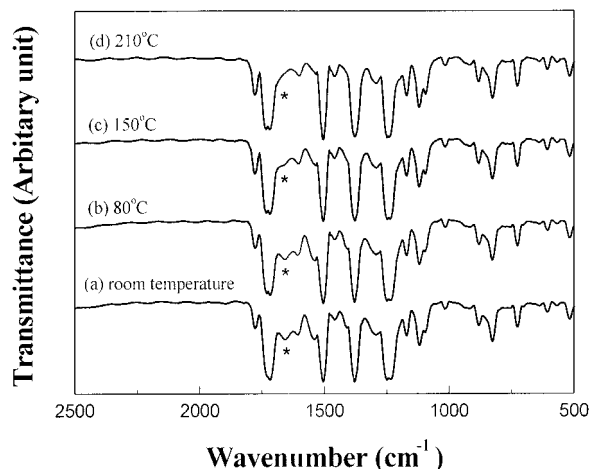


Figure 7 Fourier transform infrared spectra of the KOH-modified pyromellitic dianhydride/4,4'-oxydianiline films.

unmodified PI film, the contact angles decreased significantly with increasing treatment time.

Adhesion strength of the KOH-treated PI/epoxy adhesive joint

Curing temperature and KOH-treatment time

Low-temperature-cure epoxy adhesives. Figure 4 shows the peel strength of DGEBA and the rubber-modified epoxy resin with the linear amide curing agent as a function of treatment time in 1 M aqueous KOH solution. The curing was carried out at 80°C for 3 h. The peel strengths of these epoxy resins show a maximum at about 5 min KOH treatment time. The increase in peel strength arises from the reaction between the

epoxy adhesives and the carbonyl group ($-\text{COOH}$) of the KOH-modified PI. As the treatment time increased, the peel strength decreased because the KOH-treated layer acted as a weak point, as mentioned above. In addition, the adhesive was itself affected by the stress. Hence, the rubber-modified epoxy adhesive had a higher peel strength than the DGEBA epoxy adhesive because of the high toughness of the rubber-modified epoxy adhesive. We will discuss this further in the section on the mechanical properties of the epoxy adhesives.

Medium-temperature-cure epoxy adhesives. Figure 5 shows the peel strength of DGEBA and the rubber-modified epoxy resin with MDA as a function of treatment time in 1 M aqueous KOH solution. The curing was carried out at 80°C for 2 h and then at 150°C for 2 h. The maximum peel strength was obtained at a 20-min KOH treatment time. In contrast to the low-temperature curing agents, the peel strength exhibited a uniform value with increasing treatment time. This result is caused by the partial imidization of the polyamic acid layer, which is supposed to function as a weak boundary layer. This will be discussed in more detail in the section dealing with the FTIR results. In addition, the rubber-modified epoxy adhesive had a higher peel strength than the DGEBA, as was found for the low-temperature curing agents.

High-temperature-cure epoxy adhesive. To obtain higher peel strength than was found for the low- and medium-temperature curing agents, we decided to use polyamic acid as the curing agent. The curing was carried out at 120°C for 2 h, and then at 205°C for 5 h. The curing was carried out at 120°C for 2 h, and then at 205°C for 5 h. Figure 6 shows the peel strength

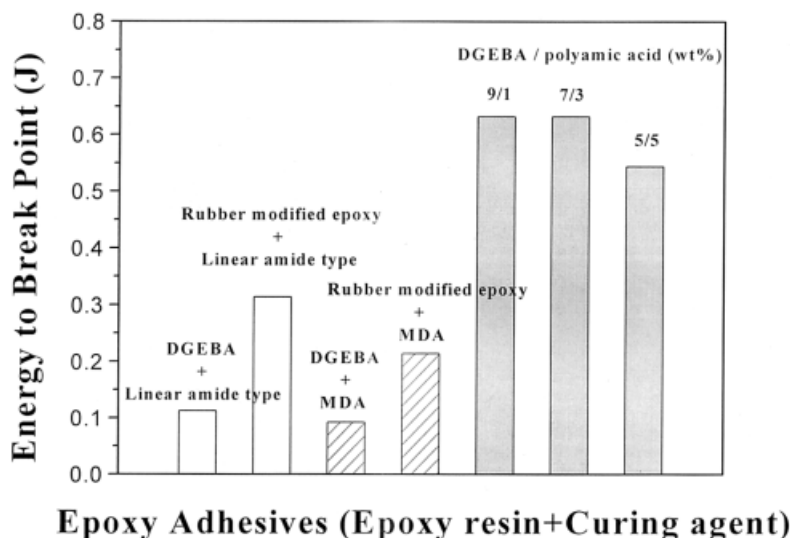
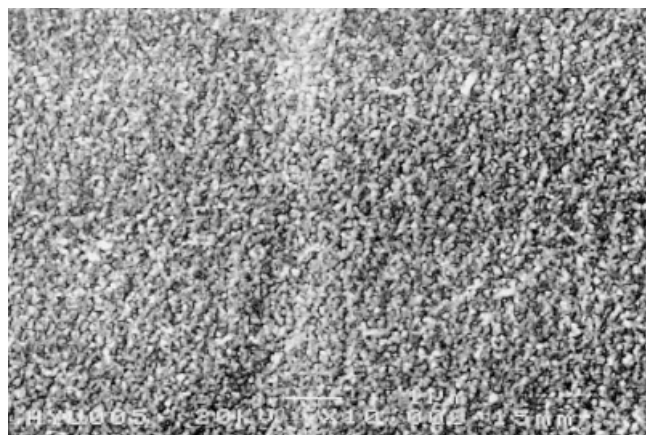
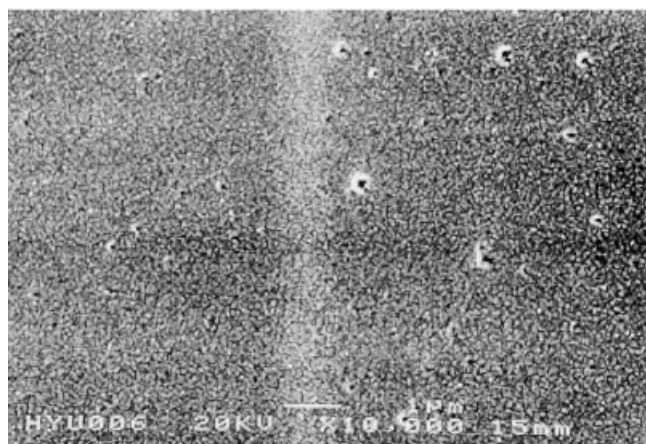


Figure 8 Energy at break point of the epoxy adhesives used together with various curing agents. Diglycidyl ether of bisphenol A epoxy resin (DGEBA), rubber-modified epoxy + linear amide type = cured at 80°C for 3 h, DGEBA, rubber-modified epoxy + 3,3'-methylenedianiline = cured at 80°C for 2 h then 150°C for 2 h, DGEBA + polyamic acid = cured at 120°C for 2 h then 210°C for 5 h.



(a)



(b)

Figure 9 The peeled surfaces of the modified polyimide/polyamic acid + diglycidyl ether of bisphenol A epoxy resin joints: (a) the polyimide surface, (b) the epoxy surface.

of the DGEBA/KOH-treated PI composite as a function of the concentration of polyamic acid (wt%). In the single-layer joint, the highest peel strength was achieved at a polyamic acid loading of 10 wt%, and the maximum peel strength was 0.9 N/mm. This value is much larger than that seen with the low- and medium-temperature curing agents. The polyamic acid layer on the KOH-treated PI surface was completely converted to PI using this curing process.

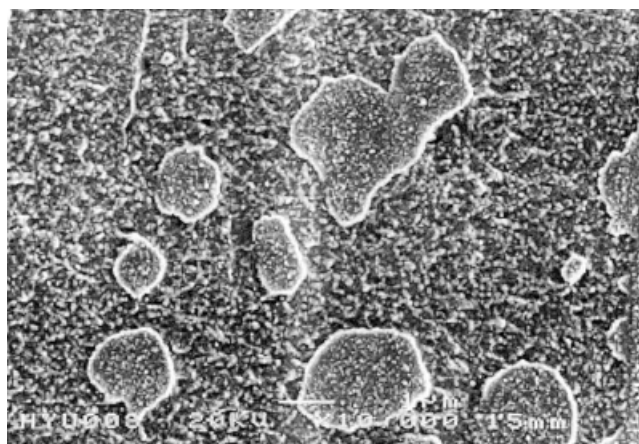
To investigate the effect of the interlayer used as an adhesion-promoting layer, we introduced a multilayer joint, which had two interfaces; the first being the interface between the KOH-treated PI and the spin-coated polyamic acid and the second being the interface between the spin-coated polyamic acid and the epoxy adhesive. If the epoxy adhesive molecules diffused into the spin-coated polyamic acid layer, then the adhesion strength would be larger than that of the single-layer joint. Lee et al.¹² reported that a synthe-

sized multilayer joint had good mechanical strength, with values over 0.85 N/mm. These results will be discussed in more detail in the section reporting on the peeled surface study.

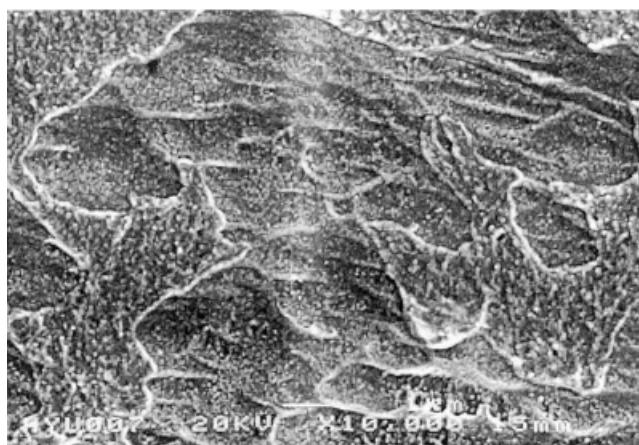
Investigation of the modified PI using FTIR

The thermal conversion from polyamic acid to PI was confirmed using FTIR spectroscopy. The KOH-treated PIs were dried at room temperature, 80°, 150°, and 205°C in a vacuum oven for 2 h, respectively. These temperatures were the curing temperatures of the epoxy/PI joints. Figure 7 shows typical FTIR spectra from each sample at the different drying temperatures.

When the sample was dried at room temperature and 80°C, the characteristic absorption of amide car-



(a)



(b)

Figure 10 The peeled surfaces of the modified polyimide/pyromellitic dianhydrid/4,4'-oxydianiline spin-coated layer/polyamic acid + diglycidyl ether of bisphenol A epoxy resin joints: (a) the polyimide surface, (b) the epoxy surface.

bonyl (C=O stretch) was exhibited at 1660 cm^{-1} . Therefore, we concluded that the imidization of the polyamic acid to PI had not occurred at these temperatures. This is the reason for the decrease in the peel strength in the low-temperature curing agents. However, the amide absorption band decreased at the drying temperature of 150°C and disappeared altogether at the drying temperature of 205°C . These results indicate that the imidization of the modified polyamic acid, which acts as a weak boundary layer, had partially occurred and then completely occurred at drying temperatures of 150° and 205°C , respectively.

In brief, the weak boundary layer from the imidization of the polyamic acid was enhanced and the adhesion strength gradually increased with increasing curing temperature.

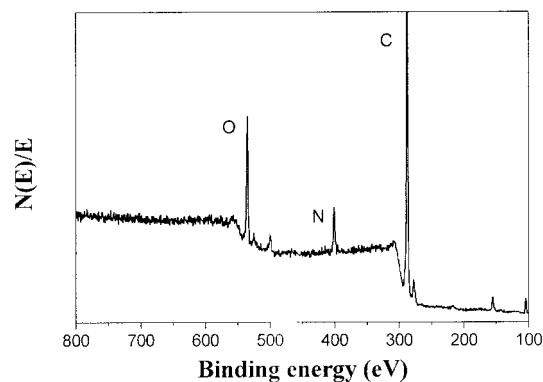
Toughness of the epoxy adhesive

As discussed earlier, the high-temperature-cure epoxy adhesive/PI joints had a higher adhesion strength than those with low- and medium-temperature curing agents. Hence, the rubber-modified epoxy adhesive showed a higher adhesion strength than the DGEBA. Figure 8 shows the toughness of epoxy adhesives themselves. The rubber-modified epoxy resin had a higher toughness than the DGEBA. In addition, the toughness of the epoxy resin dramatically increased when polyamic acid was used as the curing agent. These results show that the adhesion strength of the epoxy adhesive/PI joints increased with increases in the toughness of the epoxy adhesive.

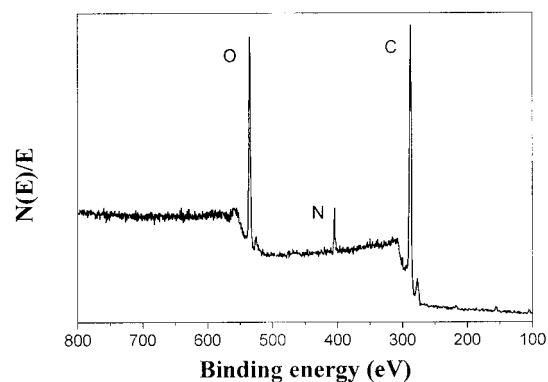
Investigation of the peeled surfaces using the high-temperature curing agents

To understand the interfacial behavior, surface analysis was carried out after the peel test to identify the locus of the failure. The peeled surfaces of the samples cured at high temperatures were analyzed using SEM and XPS. Figures 9 and 10 show the peeled surface morphologies of the single- and multilayer joints, respectively.

PMDA-ODA PI cured at 230°C has a disordered structure, and when it is treated at temperatures of 300°C or higher, it becomes more ordered, forming a relatively more crystalline state.¹¹ Lee et al.¹² reported that PI, formed from the modification of polyamic acid in aqueous KOH solution followed by curing at 230°C for 30 min, dissolved in NMP, because NMP diffused into the modified layer. This result indicates that the disordered PI is relatively amorphous. Thus, the locus of the failure of the single-layer system should be the amorphous PI layer, which was near to the PI surface. As illustrated in Figure 9, the failure did occur near the interfacial region. In the multilayer joint, the spin-coated layer was observed on both the PI and epoxy



(a)



(b)

Figure 11 X-ray photoelectron spectrometer spectra of (a) the pyromellitic dianhydride/4,4'-oxydianiline (PMDA/ODA) polyimide surface, and (b) the epoxy adhesive surface. (Diglycidyl ether of bisphenol A epoxy resin + PMDA-ODA polyamic acid [wt% 5/5].)

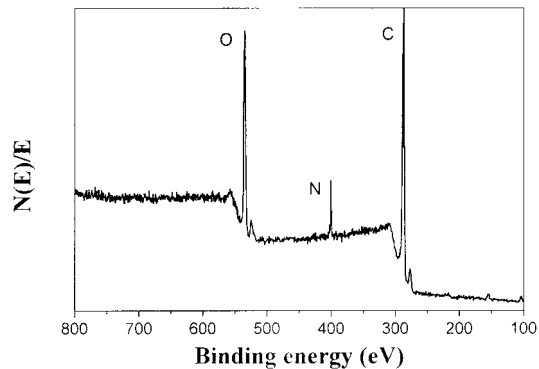
adhesive sides. Therefore, the failure occurred at the spin-coated PI layer.

In the case of PMDA/ODA to epoxy adhesives adhesion, both sides of the surfaces were identical after peeling, and they therefore had the same chemical composition. Thus, it is not possible to identify the exact locus of the failure in this system, as is shown in Figure 11. If one polymer contains an element that does not exist in the other polymer, it is possible to detect the different element using XPS. Because XPS can detect the fluorine atom, a fluorinated PI was synthesized to analyze the locus of the failure. In the single-layer joint, 6FDA-ODA polyamic acid was used instead of PMDA-ODA as the curing agent for the epoxy adhesive, and 6FDA-ODA was used as the spin-coated layer rather than PMDA-ODA in the multilayer joint. Figures 12 and 13 show the elemental analyses of the peeled surfaces of the single-layer and multilayer joints, respectively, as measured using XPS.

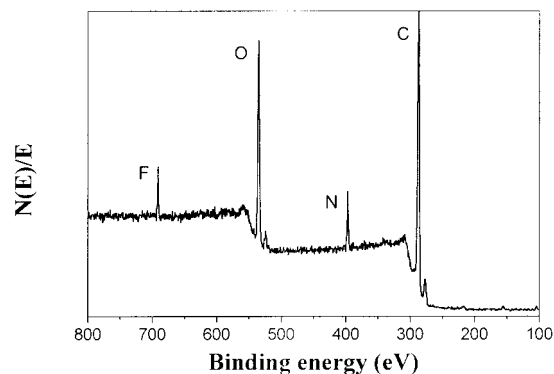
In Figure 12, the PI side contained no F atoms, whereas F atoms were detected in the epoxy adhesive surface. The presence of these atoms confirmed that exfoliation of the PI/epoxy joint occurred at the interface, or near to the PI interface region, as discussed earlier. In Figure 13, both the PI and epoxy adhesive sides contained significant numbers of F atoms. This result indicates that exfoliation of the PI/epoxy joint occurred at the spin-coated PI layer, which confirms that the epoxy molecules diffuse into the spin-coated PI layer. Therefore, multilayer joints have a higher adhesion strength than single-layer joints.

CONCLUSIONS

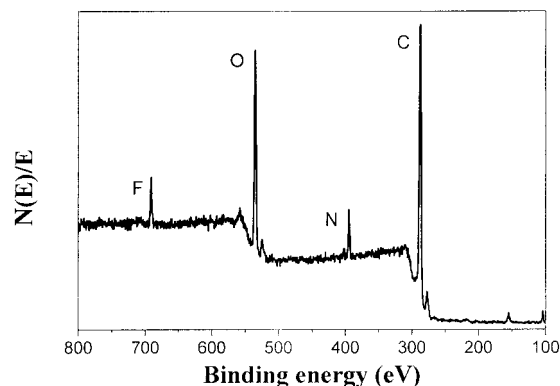
This research examined the effect of introducing functional groups onto the surface of PI to improve the



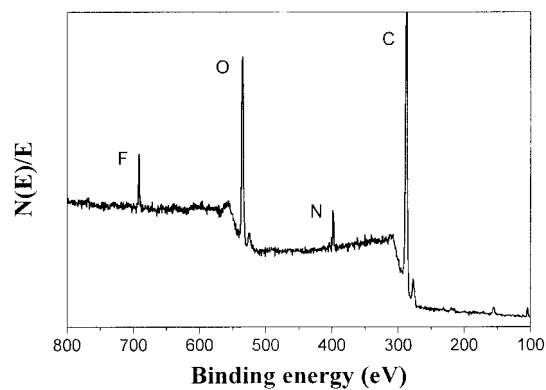
(a)



(a)



(b)



(b)

Figure 12 X-ray photoelectron spectrometer spectra of the peeled surfaces of the (modified polyimide/[6FDA-ODA polyamic acid + diglycidyl ether of bisphenol A epoxy resin]) joints: (a) the polyimide surface, and (b) the epoxy surface.

Figure 13 X-ray photoelectron spectrometer spectra of the peeled surfaces of the (modified polyimide/[6FDA-ODA spin coated layer]/(pyromellitic dianhydrid/4,4'-oxydianiline polyamic acid + diglycidyl ether of bisphenol A epoxy resin)) joints: (a) the polyimide surface, (b) the epoxy surface.

adhesion between PI and epoxy layers. Further improvement of the adhesion strength was investigated using factors such as an increase in the toughness of the epoxy adhesive, an increase in the curing temperature, and the use of polyamic acid as an adhesion promoting layer. All these approaches were effective in increasing the adhesion strength of the PI/epoxy adhesive joint.

Using low-temperature curing agents, the adhesion strength and wettability were improved by introduction of the functional group on the PI surface. The optimal KOH treatment was at 1 M concentration and 5 min treatment time. Rubber-modified epoxy adhesive had a higher adhesion strength than DGEBA because the toughness of the epoxy adhesive was increased. Severe treatment conditions produced a

thicker modified PAA layer, which acted as a weak boundary layer.

A higher adhesion strength was achieved using medium-temperature curing agents than that acquired using low-temperature agents because an increase in curing temperature partially enhanced the weak boundary layer.

Using high-temperature curing agents, the single-layer joint had an even higher adhesion strength because the increase in curing temperature increased the toughness of the epoxy adhesive. The multilayer joint had a higher adhesion strength than the single-layer joint because of the diffusion of epoxy molecules into the spin-coated PI layer.

From XPS and SEM analysis, the failure loci of high-temperature cure samples showed that for the single-layer joints, the locus of the failure was at the interface or near the PI interface region. However, the locus of the failure in the multilayer joints was at the spin-coated PI layer. Therefore, the highest adhesion strength was achieved by diffusion of the epoxy adhesive molecules, but all the other methods mentioned above also improved the adhesion.

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