

Synthesis and Characterization of Pyridine-Containing Poly(imide-siloxane)s and Their Adhesion to Copper Foil

Chun-Kang Ku, Chang-Hong Ho, Ting-Shun Chen, Yu-Der Lee

Department of Chemical Engineering, National Tsing Hua University, Hsinchu, Taiwan 30013, Republic of China

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ABSTRACT: A series of pyridine-containing poly(imide-siloxane) (PIS) copolymers with different amounts of PDMS with various segmental lengths were synthesized from 2,6-diaminopyridine (DAP), α,ω' -aminopropylpoly(dimethylsiloxane) (PDMS), 1,3-bis(4-aminophenoxy)benzene (APB), and 4,4'-oxydiphthalic dianhydride (ODPA). A modified synthetic approach was applied instead of approaches commonly reported in the literature, to ensure the incorporation of DAP and PDMS. The effects of the content and the segmental length of PDMS on the thermal glass transition temperature (T_g), dielectric constant, and surface electrical resistivity of the copolymer are investigated. The copolymers were attached to copper foil by hot-pressing, and changes in

wettability caused the peel strength of the laminates to increase with the PDMS content, but to decrease as the DAP content increased. Furthermore, X-ray photoelectron spectroscopy was employed to determine the loci of failures (LOF) of the laminates and to monitor the movement of LOF, which varies with the PDMS content. For those laminates with good peel strengths, the LOF occur in the interior of PIS layer, indicating that the adhesion is cohesive rather than adhesive. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 104: 2561–2568, 2007

Key words: adhesion; copolymerization; polyimides; poly-siloxanes; thermoplastics

INTRODUCTION

Polyimide-based flexible copper clad laminates (FCCL) play an important role in high-density packages and microelectronic circuitry; unfortunately, the adhesion of polyimide to copper is usually poor. Available methods for improving the adhesion between polyimide and copper can be classified into two groups—those that involve surface modification of conventional polyimide films^{1–3} and those that involve the synthesis of new polyimides.^{4–6} Excellent adhesive properties, good gas permeability, low dielectric constants, and favorable overall thermal and mechanical properties have led to poly(imide-siloxane) (PIS) copolymers being a noticeable and potential branch of novel polyimides and widely studied in the applications of photosensitive films, gas permeable membranes, and adhesives to alloy 42, but their adhesions to copper are rarely reported.^{7–12} Lee et al.¹³ cast triazole-terminated polyisoimide on a copper foil before thermal dehydration, which cause the triazole to form a complex with copper. Wang et al.¹⁴ and Ku

et al.¹⁵ proposed that pyridine also forms a complex with copper. Both of the two concepts mentioned above were applied herein to improve the adhesion between polyimide and copper foil. To our knowledge, the detailed discussion of the adhesion between such copolymers and copper foil has not been reported.

In most of the literature, PIS was synthesized by adding the diamine and PDMS into a reactor that contained the desired solvent and then adding dianhydride.^{8–12} Arnold et al.¹⁶ and Furukawa et al.¹⁷ synthesized the PIS copolymers via a two-stage reaction to ensure the incorporation of the PDMS. In this work, a series of pyridine-containing PIS copolymers, which exhibit good adhesion with copper foil, were synthesized using a modified approach that was derived from the reported two-stage method. They structurally analyzed, and their thermal and electrical properties were characterized. Their adhesive properties with copper foil and the loci of failures of the FCCLs are also discussed in detail.

EXPERIMENTAL

Materials

4,4'-Oxydiphthalic dianhydride (ODPA) and 1,3-bis(4-aminophenoxy)benzene (APB) were purchased from Chriskev (Leawood, KS) and Mitsui Chemicals (Tokyo, Japan), respectively. 2,6-Diaminopyridine (DAP) was purchased from Lancaster (Morecambe, Lancashire,

Correspondence to: Y.-D. Lee (ydlee@che.nthu.edu.tw).

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England). α,ω' -Aminopropylpoly(dimethyl siloxane) (PDMS) with various segmental lengths ($M_n = 900, 1680, \text{ and } 4600$) was purchased from Shinetsu Chemicals (Tokyo, Japan). ODPa was recrystallized from acetic anhydride. APB was recrystallized from methanol. DAP was purified by sublimation. PDMS was dehydrated using a molecular sieve (type 4A, Aldrich, St. Louis, MO). *N,N*-dimethylacetamide (DMAc) and tetrahydrofuran (THF) were both purchased from Tedia (Fairfield, OH). DMAc was dehydrated by mixing it with calcium hydride for 12 h in nitrogen, followed by vacuum distillation. THF was dehydrated by mixing it with sodium metal under nitrogen until the indicator, benzophenone, became dark blue and then the THF was distilled. The dehydrated PDMS, DMAc, and THF were stored in vials that were filled with dry nitrogen before use.

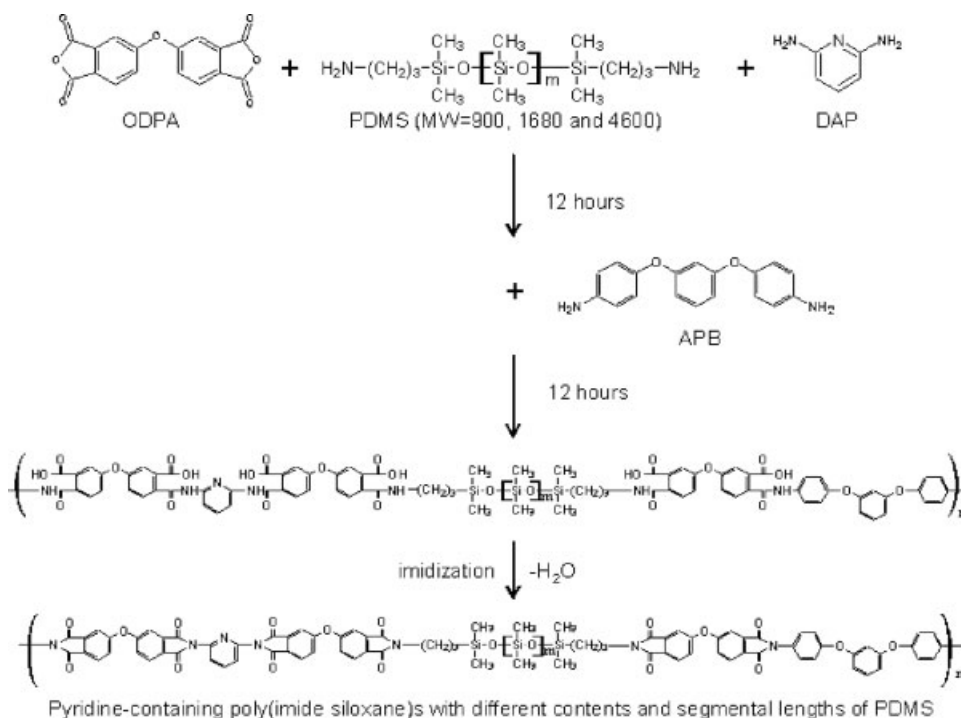
Synthesis of poly(imide-siloxane) copolymers

Scheme 1 presents the synthetic route. All polymerizations were carried out in an atmosphere of nitrogen, and equimolar amounts of diamine and dianhydride monomers were used in all cases. ODPa was applied as the dianhydride monomer and the diamine monomers were APB, DAP, and PDMS. DMAc and THF were used as the mixed solvent in a weight ratio of 1 : 1. A typical polymerization procedure is described as follows. A 100 mL three-neck flask was equipped with a nitrogen inlet and outlet, a rubber septum, and a

feeding tube. The flask was initially charged with a suitable amount of ODPa, and then an appropriate amount of DMAc was added using a disposable syringe by piercing through the rubber septum. The mixture was stirred using a magnetic stirrer at room temperature for 5 min, and then the solution of PDMS and DAP in THF was added dropwise via the feeding tube for 10 min. The solution was stirred at room temperature for 12 h before the APB was added; the reaction proceeded for another 12 h to yield poly(amic acid) (PAA) solution with a solid content of 20 wt %. The PAA solution was cast onto a polyethylene terephthalate (PET) film and then thermally dehydrated at 170°C for 30 min in nitrogen. Free-standing PIS gel films were obtained by peeling them off from the PET films. The gel films were fixed on glass plates and then thermally cyclodehydrated at 350°C for 2 h in nitrogen. The PIS films with a thickness of around 20 μm were obtained by releasing them from the glass plates. A series of PIS copolymers of various compositions were synthesized, as shown in Table I.

Manufacture of a copper foil laminate

A PAA solution on 3,3',4,4'-pyromellitic dianhydride (PMDA) and 4,4'-oxydianiline (ODA) obtained from Taimide Technology (Hsinchu, Taiwan) was cast on a glass plate with a thickness of 500 μm , onto which a PIS PAA solution with a thickness of 20 μm was then cast. The whole was cyclodehydrated at 350°C for 2 h in



Scheme 1 Synthetic route of PIS copolymers.

TABLE I
Thermal Properties of the PIS Copolymers
with Different Contents and Segmental Lengths
of PDMS and Their Adhesion to Copper Foil

Sample	MW of PDMS (g mol ⁻¹)	PDMS content in diamine (wt %)	T_g (°C)	T_{d5} (°C)
1	900	5	213	519
2	900	10	211	521
3	900	15	210	518
4	1680	5	218	531
5	1680	10	219	527
6	1680	15	220	531
7	4600	5	221	529
8	4600	10	223	526
9	4600	15	224	523

nitrogen by stepwise heating. Following thermal cyclodehydration, a two-layer film was peeled off from the glass plate. Then, an electrodeposited copper foil (3ECTHE with a thickness of 1 oz, Mitsui Kinzoku, Japan) was placed on the top of the film (PIS side), and the foil and film were pressed subsequently at 350°C for 1 min at a pressure of 50 kgf cm⁻².

Measurements

¹H nuclear magnetic resonance spectroscopy

Before the ¹H nuclear magnetic resonance (¹H NMR) measurements were made, the PAAs were precipitated and then washed with methanol : water of 1 : 1 (volume-to-volume) using a Soxhlet extractor for 24 h. They were dried at 90°C *in vacuo* for 12 h. ¹H NMR spectra were obtained using a Varian Unitynova-500 spectrometer by dissolving the PAAs in TMS free dimethyl sulfoxide-d₆ (DMSO-d₆).

Attenuated total reflectance infrared spectroscopy

Infrared spectra of the PIS copolymers were recorded on a Perkin–Elmer Spectrum One spectrometer and 50 scans were recorded.

Measurement of electrical resistivity

The films were washed for 3 min with ethanol by ultrasonic cleaning, which was repeated three times. The films were then rinsed with deionized water and placed in a circulation oven at 200°C for 1 h before measurements were made using an Agilent high resistance meter, 4339B, at 23°C, according to ASTM D257.

Measurement of dielectric constant

The films were cleaned with ethanol followed by deionized water. They were then dried at 200°C for 1 h.

Gold electrodes with a diameter of 1 cm were vacuum-deposited on both surfaces of the dried films. The dielectric constants of the PIS films were evaluated using an Agilent dielectric analyzer, 4294A, at a frequency of 1 MHz. The measurements were made at 23°C. The test method followed the specifications of ASTM D-150.

Thermogravimetric analysis

The PIS copolymers were thermogravimetrically analyzed using a Perkin–Elmer TGA7, at a heating rate of 10°C/min from 30 to 800°C in nitrogen. The sample weight in all measurements was 5–10 mg. The thermal degradation temperature, associated with a 5 wt % loss, was recorded.

Measurement of glass transition temperature

Before the glass transition temperature was measured, the samples were heated to 300°C at a rate of 20°C/min and then naturally cooled to room temperature. The glass transition temperatures were measured using a differential scanning calorimeter (DSC), TA DSC2920, at a heating rate of 10°C/min from 50 to 300°C in nitrogen. The sample weight in the measurements was around 10 mg.

Measurement of peel strength

Peel strength was measured according to the specifications of the Institute for Interconnecting and Packaging Electronic Circuits (IPC). The copper on the laminate was chemically etched into strips with dimensions of 3 mm × 100 mm. The 90° peel test was conducted using an Instron H10KS at a peeling rate of 50.8 mm min⁻¹. The reported values were averages of three measurements.

Soldering test

The laminates with chemically-etched copper strips were fixed on the surfaces of steel plates. Before the laminates were immersed in the soldering bath at 288°C for 30 s, they were dipped into silicon oil to avoid the attachment of tin to the surface of the copper. Following the soldering test, the laminates were cleaned with acetone and then dried at 70°C for 2 h *in vacuo*.

X-ray photoelectron spectroscopy analysis

The surface compositions of the laminates were recorded on a Perkin–Elmer PHI 1600 X-ray photoelectron spectrometer with a scanning monochromated Mg anode X-ray source (K_{α} excitation = 1253.6 eV).

The electron take-off angle was 54° unless specified elsewhere. The surface of the laminate was sputter-etched using an Ar ion gun with a voltage of 5 kV.

RESULTS AND DISCUSSION

Synthesis of poly(imide-siloxane)

Unlike in the previously reported two-stage method,^{16,17} PDMS and DAP were dissolved in THF, before being added to the DMAc solution of ODPA. The excellent compatibility of THF and DMAc enables PDMS to be dissolved much easily to initiate the polymerization. Additionally, the THF solution of PDMS and DAP was added dropwise such that the PDMS and DAP immediately reacted with ODPA. The IR and ^1H NMR spectra demonstrate that the PDMS and DAP were confirmed to be incorporated into the mainchain of the copolymers.

Figure 1 displays the FTIR spectra of PIS copolymers.^{11,18–20} The IR spectrum of the ODPA-APB polyimide exhibits the absorptions at 1776 and 1712 cm^{-1} , which are related to the asymmetric and symmetric stretchings, respectively, of the imide carbonyl group. The absorption at 3058 cm^{-1} is associated with the stretching of aromatic C—H. The band at 1367 cm^{-1} is attributed to C—N—C stretching and the band at 1109 cm^{-1} is associated with the vibration of the imide ring. The skeletal vibration of aromatic C=C resulted in the absorptions at 1587 and 1474 cm^{-1} . The PIS copolymers also exhibit the following absorption bands. The absorption at 2957 cm^{-1} is associated with the asymmetric stretching of aliphatic C—H. The absorptions at 1257 and 794 cm^{-1} are attributed to the stretching and bending of Si—CH₃, respectively. The band at 1342 cm^{-1} is due to the stretching of Si—CH₂.

The absorption band at 1012 cm^{-1} is attributed to the stretching of Si—O—Si linkage. Figure 2 depicts the ^1H NMR spectra of corresponding poly(amic acid).^{18,21} The aromatic hydrogen of APB reveals that absorptions between $\delta = 6.4$ ppm and $\delta = 7.0$ ppm and those in the range of $\delta = 7$ –8.2 ppm are associated with the aromatic hydrogen of ODPA. The absorption near $\delta = 0$ ppm is attributed to the methyl hydrogen attached to the silicon of the PDMS. The aromatic hydrogen in the meta position of DAP has a characteristic absorption at $\delta = 5.8$ ppm.

Surface electrical resistivity

Electrical resistivity is a measurement indicating how strongly a material opposes the flow of electric current. Figure 3 demonstrates that the surface electrical resistivities of the films decline as the PDMS segmental length increase. Mahoney and Gardella⁹ reported that the PDMS segregation to surface is independent of the PDMS content, but a longer PDMS segment results in greater PDMS segregation. Since the electrical conductivity of PDMS is higher than that of polyimide²² and the segregation of PDMS-4600 to the surface of the film is greater, the surface resistivities of PIS copolymers with PDMS-4600 are much lower than those with PDMS-900 or PDMS-1680. Moreover, the increase in the PDMS content causes the number of PDMS on the surface to increase, and thus the surface electrical resistivity falls slightly.

Dielectric constant

The dielectric constants of PIS copolymers are in the range of 2.8–3.3, and depend directly on the chemical structure and composition of the copolymers. The

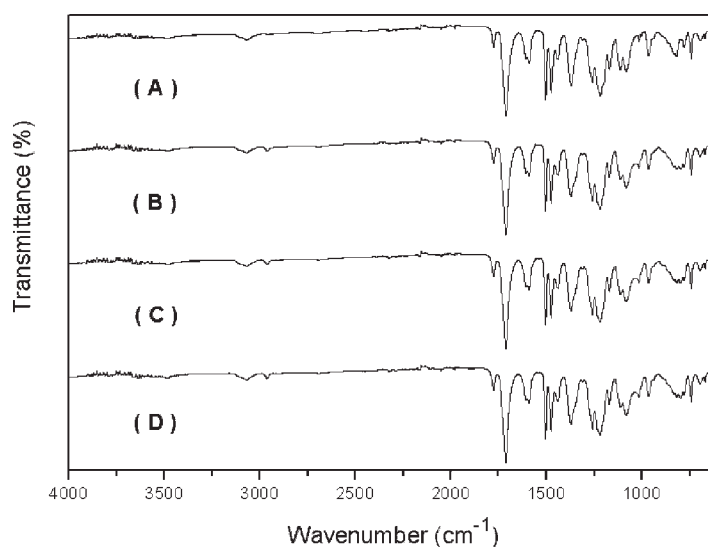


Figure 1 Representative IR spectra of ODPA-APB polyimide and the derived PIS copolymers. (A) ODPA-APB polyimide; (B) PIS with PDMS-900 = 15 wt %; (C) PIS with PDMS-1680 = 15 wt %; (D) PIS with PDMS-4600 = 15 wt %.

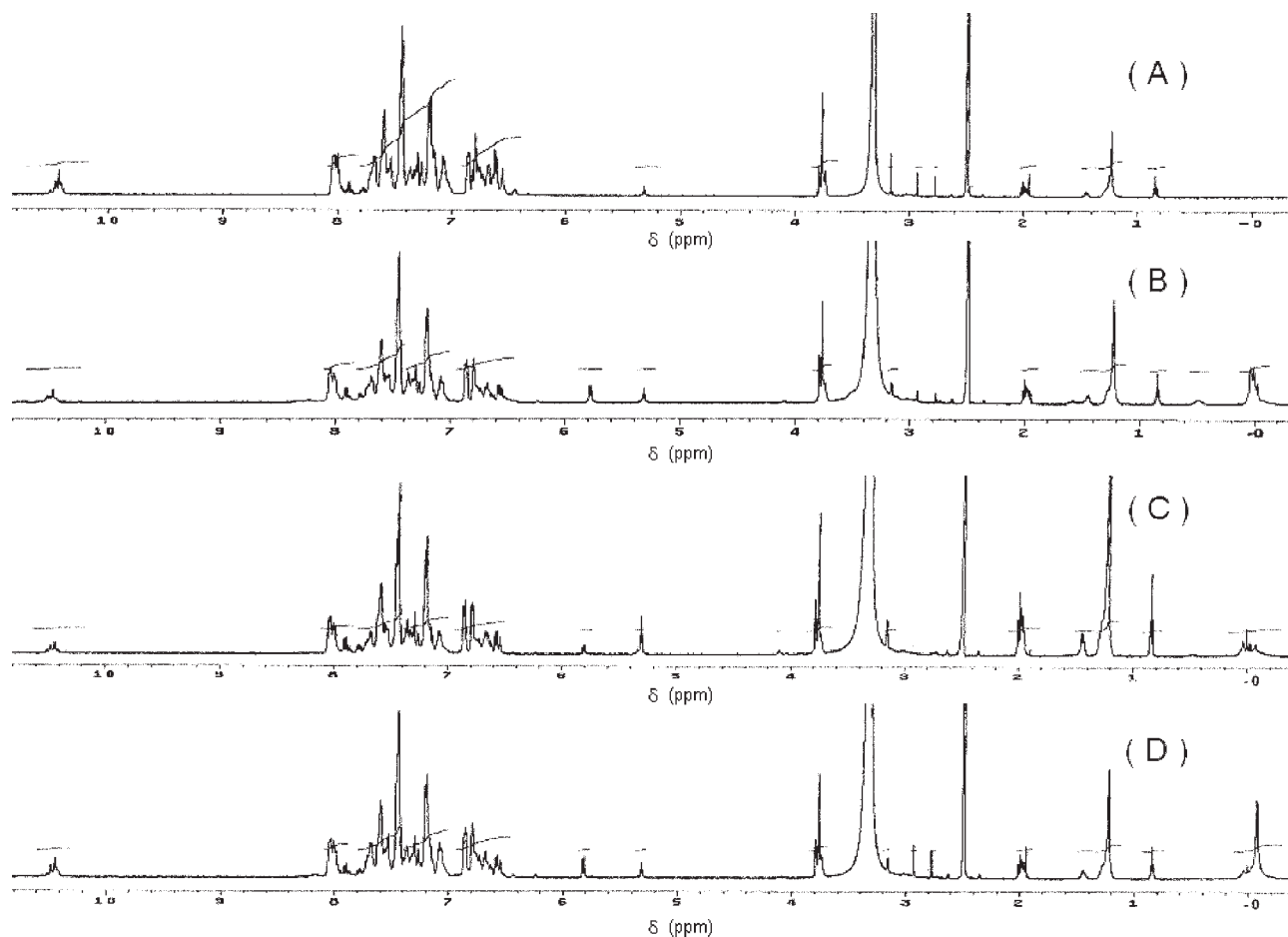


Figure 2 Representative NMR spectra of the corresponding poly(amic acid). (A) ODPA-APB polyimide; (B) PIS with PDMS-900 = 15 wt %; (C) PIS with PDMS-1680 = 15 wt %; (D) PIS with PDMS-4600 = 15 wt %.

dielectric behavior of a polymer is determined primarily by its polarizability and free volume. A polymer with a higher polarizability or a lower free volume has a higher dielectric constant.^{23,24} The polarizability

of a polymer is reduced by incorporating a symmetric monomer, such as PDMS, into the backbone of the polymer chain. As shown in Figure 4, the dielectric constants of PIS copolymers decrease as the segmental

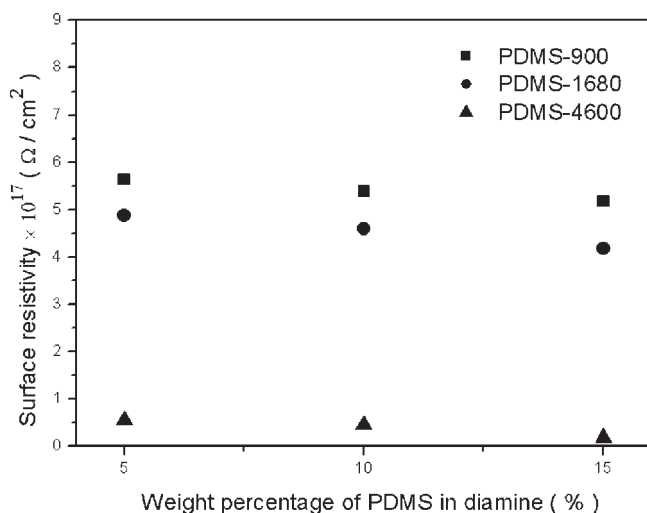


Figure 3 The dependence of surface resistivity on the content and segmental length of PDMS.

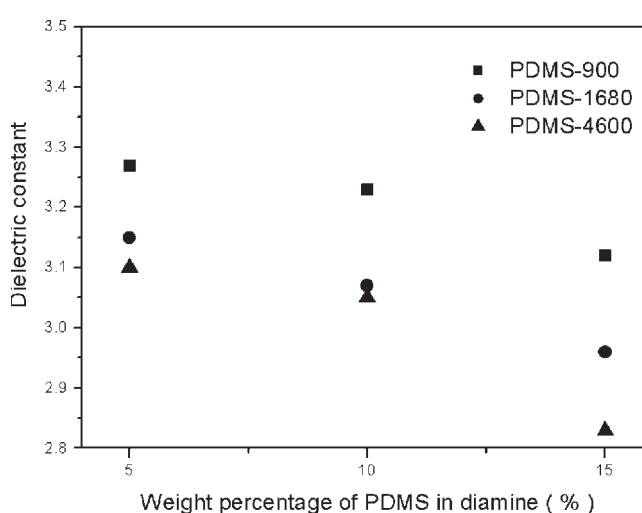


Figure 4 The dependence of dielectric constant on the content and segmental length of PDMS.

length and the amount of PDMS moiety increase. Chen et al.⁷ proposed that the introduction of the PDMS moiety into polyimide disrupts the original packing of polyimide, increasing its free volume. Consequently, the decrease in dielectric constants of the PIS copolymers, as the segmental length of PDMS increases, is attributed not only to the decrease in polarizability but also to the increase in free volume.

Thermal stability

Table I presents the thermal degradation temperatures that correspond to 5 wt % loss (T_{d5}) of PIS copolymers. The T_{d5} s of the copolymers all exceed 500°C, indicating that they possess excellent thermal stability. Park et al.²⁵ reported that the thermal degradation of PIS begins at the aliphatic propyl segments that are linked to the dimethylsiloxane segment. In this study, the weight loss caused by the thermal decomposition of aliphatic propyl segment is not clear because the amount of PDMS in the copolymer is small. Thus, the T_{d5} of the copolymers does not vary much as the amount and the segmental length of PDMS increase.

Glass transition temperature

Table I presents the glass transition temperatures (T_g s) of the copolymers. The ODPa-APB copolymer without DAP and PDMS exhibits a T_g of 220°C, but the T_g of the ODPa-APB-DAP copolymer (the molar fraction of DAP in the diamine was 1/7) increases to 243°C, revealing that the incorporation of DAP into the main-chain of the copolymer makes it more structurally rigid. Incorporation of PDMS into the mainchain clearly reduces the T_g of PIS copolymers. This effect is caused by the increase in the chain flexibility and the free volume of the copolymer. Mahoney and Gardella⁹ demonstrated that the T_g of PIS copolymer increases with the segmental length of PDMS but declines as the PDMS content increases. In this investigation, a similar trend was observed. However, because of the small amount of PDMS in the copolymer, the T_g s of the copolymers do not vary markedly with the PDMS content.

Peel strength

Figure 5 plots the dependence of the adhesion between the PIS and copper foil on the composition of the PIS copolymer. In all cases studied herein, the adhesive strength increases with the PDMS content. Kang et al.⁸ proposed that the adhesive strength between PIS and alloy-42 increases with the PDMS content. Since the surface energy of PDMS is much lower than that of imide segment, the PDMS tends to migrate to the airside surface during thermal cyclodehydration. Accordingly, the wettability of PIS with

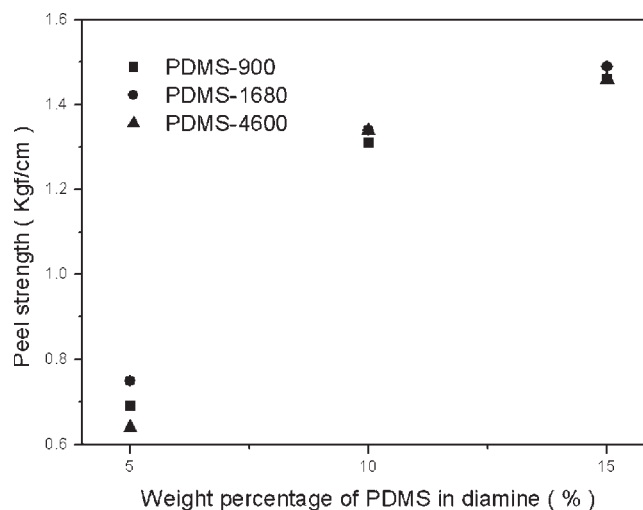


Figure 5 Peel strengths of the copper clad laminates with varying composition of PIS copolymers.

PDMS-rich surface is improved, enhancing the adhesive strength by the flexible nature of PDMS, while too much segregation of PDMS weakens the adhesion by the formation of a weak boundary layer. Mahoney and Gardella⁹ offered a similar explanation. Hergenrother et al.²⁶ demonstrated that the introduction of a-BPDA into s-BPDA series polyimides improves their thermoplasticities without lowering T_g s. Therefore, in this study, the increase in adhesion with the PDMS content might have attributed to the increases in both the thermoplasticity of PIS and the appropriate extent of PDMS segregation, even though the T_g of the PIS copolymer does not change considerably. The PIS copolymers adhere strongly with copper foil with a peel strength of above 1.4 kgf cm⁻¹ when the PDMS content in diamine is 15 wt %.

A series of PIS copolymers with various DAP contents were synthesized to explore the effect of DAP content on the peel strengths of their corresponding laminates. Table II presents their T_g s and corresponding peel strengths. The results demonstrate that the peel strength decreases as the DAP content increases; this relationship is attributable directly to the lowering in the wettability of the copolymers. Since the structure of DAP is rigid, T_g increases with the DAP content and, consequently, the wettability of copolymers decreases. On the other hand, the results imply that the DAP does not contribute to the peel strength substantially by forming a complex with copper, and this may be attributed to the small amount of DAP. However, the PIS copolymers cannot be films when the molar fraction of DAP in diamine exceeds 1/7. After the soldering test, the peel strength of the laminates falls, whereas the adhesive strengths of DAP-containing PIS copolymers do not decline so much. During the soldering test, the PIS copolymer, which is the middle layer of the laminate, is softened,

TABLE II
Properties of PIS Copolymers with Different Contents of DAP

Sample	MW of PDMS (g mol ⁻¹)	PDMS content in diamine (wt %)	DAP molar fraction in diamine (mol/mol)	T _g (°C)	Original peel strength (kgf cm ⁻¹)	Peel strength after soldering test (kgf cm ⁻¹)
10	900	15	1/5	217	1.1	0.68
2	900	15	1/7	210	1.32	0.64
11	900	15	1/9	202	1.54	0.55
12	900	15	1/11	195	1.71	0.41

destroying its original good contact with the copper surface. Consequently, the peel strength of the laminates is lower after the soldering test. However, as the DAP content increases, the T_g of the PIS copoly-

mer increases, along with its resistance to softening. Figure 6 indicates that the degree of decline in peel strength, defined as follows, is inversely proportional to the DAP content.

$$\text{Degree of decline in peel strengths} = \frac{\text{Original peel strength} - \text{Peel strength after soldering test}}{\text{Original peel strength}} \times 100\%$$

X-ray photoelectron spectroscopy

The elemental analysis of the surface of laminates was conducted by X-ray photoelectron spectroscopy (XPS). After peeling the copper strips off from the laminates, both delaminated copper and polyimide surfaces were examined to determine the locus of failure. The represented survey spectra of the delaminated copper surfaces, shown in Figure 7, depict peaks that correspond to the elements C, N, O, and Si, suggesting that the delaminated copper surfaces are covered with a layer of PIS copolymers. This fact indicates that the loci of failures of the laminates occur in the inner region of the PIS layer.²⁷

For laminates with PDMS-900, the delaminated copper surfaces were sputter-etched with an Ar ion gun

for different time periods and then surveyed by XPS. After a particular etching time, a certain thickness of the PIS layer on the delaminated copper strip was removed, exposing a fresh surface for detection. Since the surface of the copper foil is rough, a PIS-copper inter-region forms after hot-pressing lamination. As shown in Figure 8, at the beginning stage of etching, no copper is detected in the PIS region, while the concentration of copper increases with the etching time in the PIS-copper inter-region. The etching times for detecting a specified concentration of copper on the delaminated copper surfaces of laminates 2 and 3 exceed that of laminate 1, revealing that the PIS layers of formers are much thicker. Restated, the loci of failures of lami-

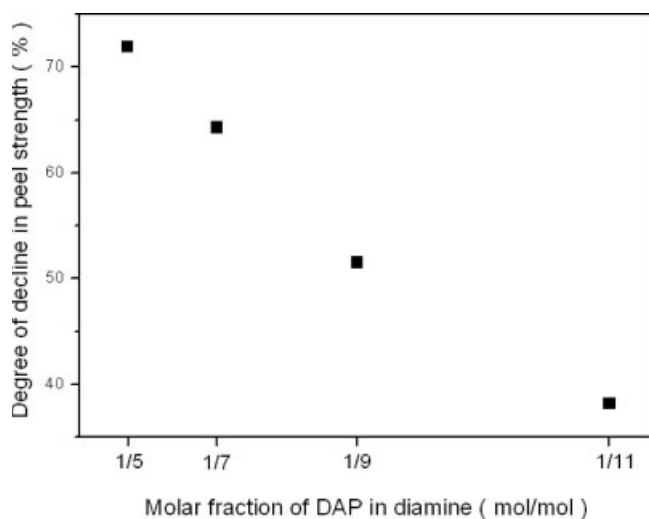


Figure 6 The relationship between the degree of decline in peel strength and the content of DAP.

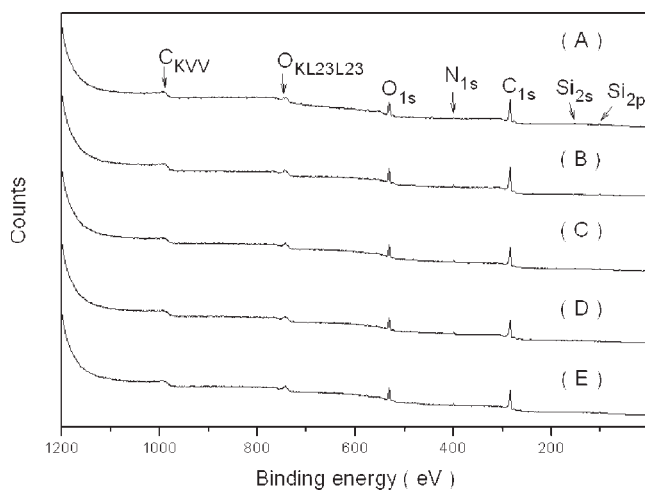


Figure 7 XPS survey spectra of the delaminated copper surface. (A) PDMS-900 = 5 wt %; (B) PDMS-900 = 10 wt %; (C) PDMS-900 = 15 wt %; (D) PDMS-1680 = 15 wt %; (E) PDMS-4600 = 15 wt %.

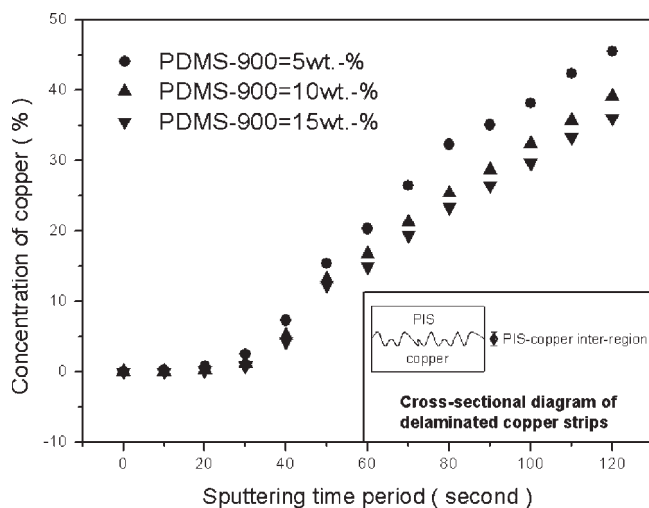


Figure 8 The copper concentration on the surface of delaminated copper surface after sputtering for various time periods.

nates 2 and 3 are much farther from the PIS/copper interface. During the hot-pressing lamination at 350°C, the PIS copolymer with better wettability is softened to come into better contact with the copper surface. Therefore, the anchoring effect of the laminates is improved. The locus of failure of the laminate with poor anchoring is at the PIS/copper interface or in the PIS layers close to the interface. In summary, the loci of failures of the laminates with better anchoring effect nearer to the interior of the PIS layer and the corresponding failures are cohesive rather than adhesive, such that stronger peel strengths are measured.

CONCLUSIONS

A series of pyridine-containing PIS copolymers that possess excellent thermal and dielectric properties and adhere strongly to copper foil were synthesized via a modified two-stage approach. The loci of failures of their copper clad laminates were verified by XPS to occur in the PIS layers. As the PDMS content increases, the locus of failure moves toward the interior of the PIS layer, indicating that the failure is cohesive rather than adhesive. Because of the increase in wettability of PIS copolymer, the peel strengths of the copper clad laminates increase with the PDMS content. The structural rigidity of DAP causes the wettability of PIS copolymer to decrease as the DAP content increases. Hence, the copolymers with more DAP are more resistant to softening during the soldering test.

A comparison with conventional aromatic polyimides demonstrates that the poly(imide-siloxane) has

some attractive characteristics, such as strong adhesion to copper, low moisture absorption, and low dielectric constant, making it preferable for applications in FCCL.

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