

# Adhesion Promotion of the Polyimide–Copper Interface Using Silane-Modified Polyvinylimidazoles

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**ABSTRACT:** In order to promote the interfacial adhesion strength between polyimide (PI) and copper, the copper surface was treated with polyvinylimidazole (PVI) or silane-modified PVIs. They were prepared by the copolymerization of 1-vinylimidazole (VI) with the following silane coupling agents: 3-(*N*-styrylmethyl-2-amino-ethylamino)propyltrimethoxysilane (STS), vinyltrimethoxy silane (VTS), allyltrimethoxy silane (ATS), and  $\gamma$ -methacryloxypropyltrimethoxysilane ( $\gamma$ -MPS). The mole ratio of the silane coupling agent to VI was fixed at 1 : 1. The lap shear strengths between PI and copper were measured at the following different bonding temperatures: 290, 320, 350, and 380°C. In each case, the maximum adhesion strength was obtained at 350°C. VTS-modified PVI showed the best performance on adhesion promotion of the PI–copper interface. Fourier transform infrared spectroscopy was applied to investigate the thermo-oxidative degradation of PI and oxidation of copper. In addition, scanning electron microscopic analysis and contact angle measurements were performed for the investigation of the interaction between PI and silane-modified PVIs. © 1998 John Wiley & Sons, Inc. *J Appl Polym Sci* 68: 1343–1351, 1998

**Key words:** silane-modified polyvinylimidazole; interfacial adhesion strength of polyimide–copper; bonding temperature

## INTRODUCTION

Polyimide (PI)s are a class of polymers that have a high thermal quality, thermo-oxidative stability, and chemical resistance, coupled with good mechanical properties. These PIs have attracted an extensive attention as adhesives, fibers, films, moldings, composite matrices, coatings, and membranes.<sup>1</sup>

In microelectronics and aerospace industries, the adhesion of PI onto metal surface is a crucial factor for the performance improvement. Especially in copper–PI adhesion, PI near the copper surface degrades faster at a high temperature than PI near other metals such as aluminum and

chromium.<sup>2,3</sup> It has been attributed to the fact that copper compounds catalyze the thermo-oxidative degradation of polymer in air. It was reported that copper and/or cuprous ions could diffuse into a polymer and cause the decomposition of hydroperoxides formed by the oxidation of polymer film on copper at elevated temperatures.<sup>4–10</sup>

Thermo-oxidative degradation of PI on copper could be reduced by the insertion of barrier films between copper and PI. These barrier films prevent the diffusion of copper into PI. The barrier materials are divided into two types: inorganic and organic. Chromium has often been used as an inorganic barrier. However, chromium must be vapor-deposited on copper surface, and it is time-consuming to lay down.<sup>11</sup> Organic materials, such as benzotriazole, benzium, indazole, and imidazole, have been also used for the barrier applications. These small molecules did not suppress

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copper corrosion at elevated temperature due to their poor thermal stabilities.<sup>12–14</sup> Therefore, thermally stable polymers have been demanded for copper corrosion protection.

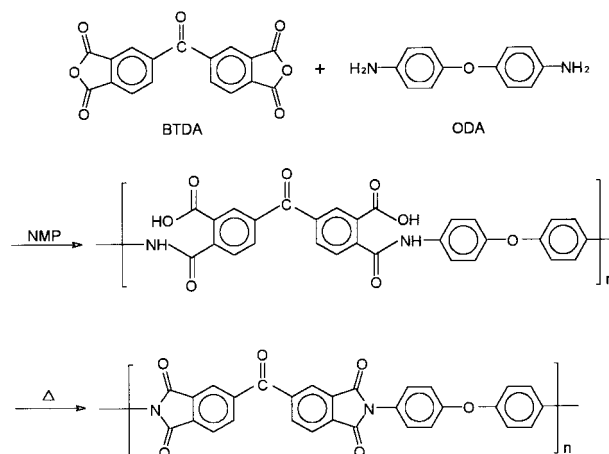
Polyvinylimidazole (PVI) is one of the anticorrosion agents for copper surface at elevated temperature. The imidazole ring of PVI forms complex with copper to suppress the oxidation of copper.<sup>15–18</sup> Recently, the copolymer of silane coupling agent and 1-vinylimidazole (VI) was reported to improve the anticorrosion performance for copper surface.<sup>19,20</sup> Ishida and Kelley reported that the copolymer of methacryloxy silane and VI suppressed thermal degradation of pyromellitic dianhydride-oxydianiline (PMDA-ODA) polyimide on the copper surface.<sup>12</sup>

In this study, in order to improve the interfacial adhesion strength between copper and 3,3',4,4'-benzophenonetetracarboxylic dianhydride-oxydianiline (BTDA-ODA) polyimide, PVI and four silane-modified PVIs were applied as primers. The BTDA-ODA polyimide has a more flexible structure than the PMDA-ODA polyimide. The effect of bonding temperature on the interfacial adhesion strengths between copper and PI was studied by the measurement of lap shear strength. Degradation mechanism of the BTDA-ODA polyimide on copper and the oxidation of copper were investigated with Fourier transform infrared reflection-absorption spectroscopy (FTIR-RAS).

## EXPERIMENTAL

### Materials

*N*-Methyl-2-pyrrolidinone (NMP, Aldrich Chemical Co., Milwaukee, WI, USA) was dehydrated with phosphorous pentoxide and distilled under reduced pressure. 3,3',4,4'-benzophenonetetracarboxylic dianhydride (BTDA, Aldrich Chemical Co.) and 4,4'-diaminophenylether (ODA, Aldrich Chemical Co.) were recrystallized in acetic anhydride and ethanol, respectively. VI (Aldrich Chemical Co.), VTS (Aldrich Chemical Co.), ATS (Petrach Systems, Inc.), and MPS (Petrach Systems Inc.) were distilled *in vacuo* to yield pure and colorless liquids. STS (Petrach Systems, Inc.) was used without distillation. Azobisisobutyronitrile (AIBN, Wako Pure Chemical Industries, Ltd.) was recrystallized in methanol. Copper plates (1.2 mm thick, Poongsan Co., Korea) were mechanically polished and cleaned with hexane and ethanol in an ultrasonic bath.



**Figure 1** The synthetic scheme of the BTDA-ODA polyimide.

### Synthesis

ODA was dissolved in NMP and BTDA was added into the ODA-NMP solution at room temperature. The total solid content was 15%. The reactor was stirred in an ice water bath for 12 h. Viscous polyamic acid (PAA) solution was obtained and refrigerated until use. Figure 1 shows the synthetic scheme of BTDA-ODA polyimide.

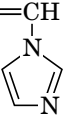
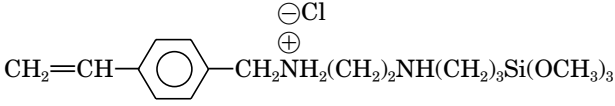
PVI and four silane-modified PVIs were synthesized by free radical polymerization using AIBN as an initiator. VI was homopolymerized and copolymerized with four silane coupling agents for 24 h in benzene at 68°C with stirring in an argon atmosphere. Total monomer concentration was fixed at 1M, and the initiator concentration was  $1 \times 10^{-3}M$ . The mole ratio of the two monomers in feed was 1 : 1. The chemical structures of VI and four silane coupling agents are represented in Table I, and the synthetic scheme of silane-modified PVI is shown in Figure 2.

### Lap Shear Test

Copper plates were coated with PVI or silane-modified PVI and then cast with diluted polyamic acid solution (7 wt %). Preimidization was performed at 100°C for 30 min, at 150°C for 30 min, and 175°C for 30 min in NMP saturated condition.

Adhesive tape was prepared for lap shear test specimen. The polyamic acid solution was coated onto E-glass fabric with  $\gamma$ -aminopropyltriethoxy silane ( $\gamma$ -APS) finish. After coating, E-glass fabrics were heat-treated to drive off the solvent and other volatiles. Heat treatment process is as follows: 1 h at 100°C, 30 min at 150°C, and 30 min at

**Table I. The Chemical Structures of Vinylimidazole and Various Silane Coupling Agents**

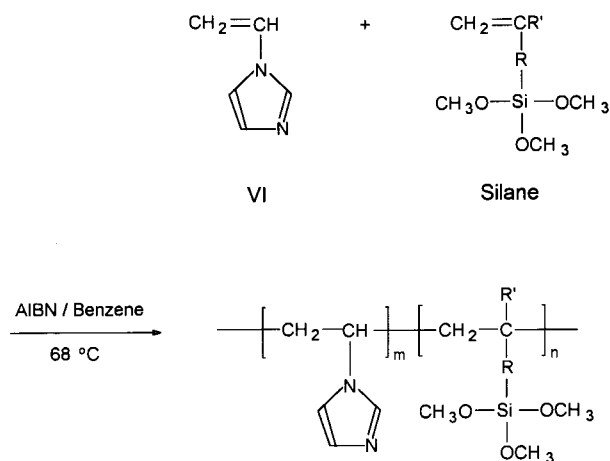
Monomers	Chemical Structures
VI	$\text{CH}_2=\text{CH}$ 
VTS	$\text{CH}_2=\text{CHSi}(\text{OCH}_3)_3$
ATS	$\text{CH}_2=\text{CHCH}_2\text{Si}(\text{OCH}_3)_3$
$\gamma$ -MPS	$\text{CH}_2=\text{C}(\text{CH}_3)\text{COO}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$
STS	$\text{CH}_2=\text{CH}$ - 

175°C. The coating thickness was approximately 0.25 mm.

Specimens for lap shear test were fabricated by hot pressing. The adhesive tape was placed between the two copper plates. The cure temperature was raised from room temperature to the bonding temperature at a heating rate of 5°C/min and held for 1 h at the bonding temperature. The bonding temperatures were 290, 320, 350, and 380°C. 500 psi of pressure was applied during the cure process. A lap shear test was performed with an Instron 4465 according to ASTM D1002.

#### Instrumental Analysis

A Bomem MB-100 FTIR spectrophotometer was used to investigate the interface between copper



**Figure 2** The synthetic scheme of silane-modified PVI.

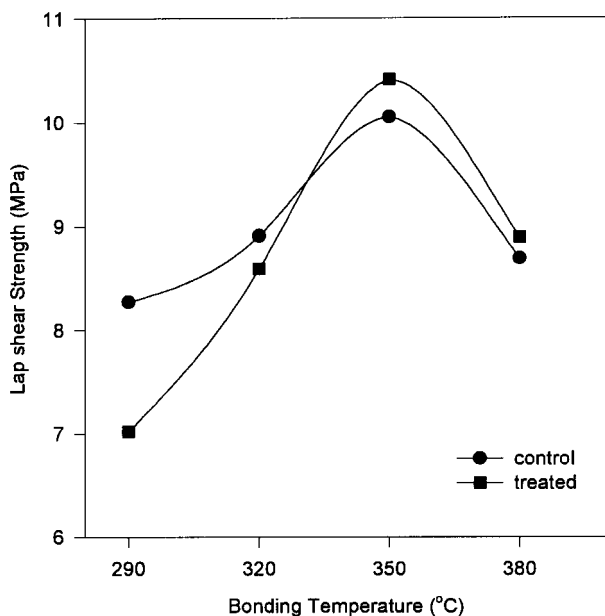
and PI. PAA solution was cast onto bare copper surfaces or copper plates coated with PVI or silane-modified PVIs. The copper plates were heat-treated by the same methods used in the fabrication of lap shear test specimens. IR spectra were obtained by the reflection and absorption (R-A) technique. 32 scans were collected with a spectral resolution of 4  $\text{cm}^{-1}$ .

Contact angles of diluted PAA solutions (7 wt %) on the primer-coated copper surface were measured to investigate the initial wettabilities of PAA solutions on each primer. The measurement was performed by a Ramé–Hart NRL contact angle goniometer.

Fractured surfaces of PI–PVI and PI–silane-modified PVI systems were investigated with a scanning electron microscope, JEOL JSM-35. PI and primer were solution-blended and imidized at 250°C. The imidized samples were fractured in liquid  $\text{N}_2$  to observe the fractured surface.

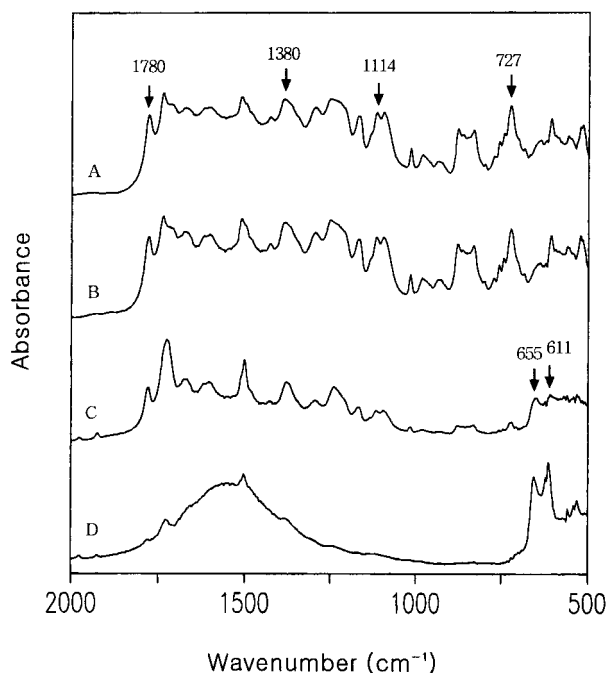
#### RESULTS AND DISCUSSION

The interfacial adhesion strength of PI–PVI-coated copper was compared with that of PI–bare copper. Figure 3 shows the lap shear strengths of PI–Cu and PI–PVI–Cu as a function of bonding temperature. The maximum lap shear strengths are observed at 350°C in both cases. As the bonding temperature of PI–Cu and PI–PVI–Cu systems increases, the mechanical properties of PI are improved due to the enhanced molecular packing of PI with increasing bonding temperature.<sup>2</sup> At 380°C, however, the thermal degradation of PI

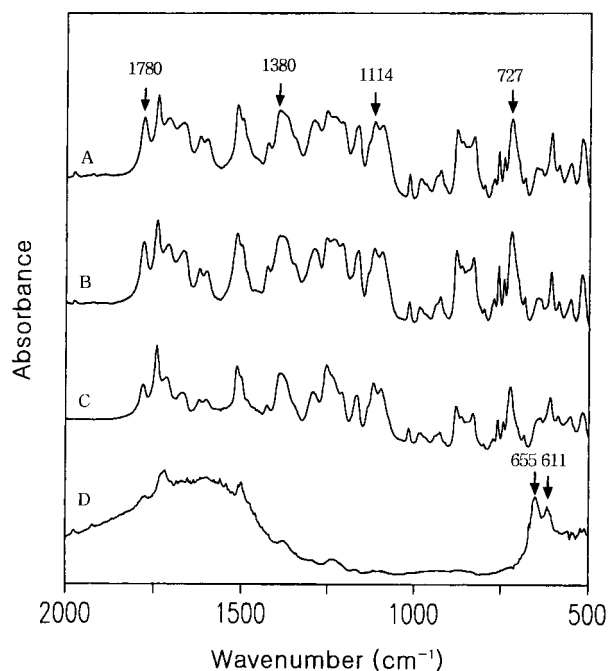


**Figure 3** The lap shear strength of PI-PVI-Cu as a function of the bonding temperature.

and PVI lowers the interfacial adhesion strengths in both cases. The PI-Cu system shows higher lap shear strengths than the PI-PVI-Cu system at lower bonding temperatures (290 and 320°C),



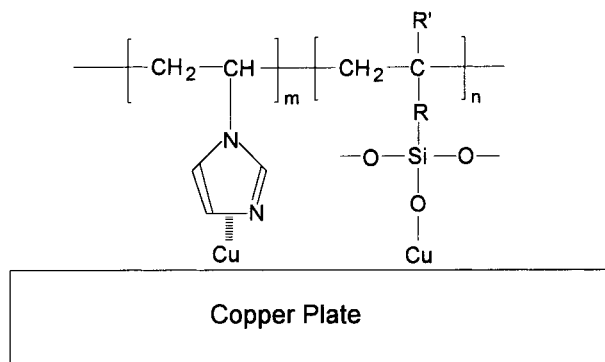
**Figure 4** R-A spectra of PI on copper with the different bonding temperatures: (A) 290, (B) 320, (C) 350, and (D) 380°C.



**Figure 5** R-A spectra of PI on PVI-coated copper with the different bonding temperatures: (A) 290, (B) 320, (C) 350, and (D) 380°C.

and lower lap shear strengths at higher bonding temperatures (350 and 380°C). The former result is due to the strong ionic bond between copper and PI in the PI-Cu system.<sup>2,10</sup> However, in the case of the PI-PVI-Cu, imidazole rings of PVI lead to the formation of coordination complexes with copper, and so there is no distinct chemical bond between PVI and PI. The higher lap shear strengths of PI-PVI-Cu system than PI-Cu at 350 and 380°C represent less degradation of PI films on PVI-coated coppers. From this fact, it can be inferred that PVI film delays or suppresses the diffusion of copper into the PI phase.

Figures 4 and 5 show the spectral changes of PI films on bare coppers and on PVI-coated coppers, respectively. Samples were heated for 1 h at each bonding temperature. The characteristic peaks of PI appear at 1780  $\text{cm}^{-1}$  (the symmetric C=O stretching), 1380  $\text{cm}^{-1}$  (C-N stretching), 1114  $\text{cm}^{-1}$  (aromatic C-H in-plane bending), and 727  $\text{cm}^{-1}$  (C=O bending).<sup>21</sup> The distinct decreases of PI peak intensities are observed in Figure 4(C), but there is less decrease in Figure 5(C). In addition, the bands at 655 and 611  $\text{cm}^{-1}$  assigned to cuprous oxide appeared in Figure 4(C)<sup>22</sup> but not in Figure 5(C). This means that PVI on copper retards or inhibits the thermal degradation of PI. At 380°C, however, PI films are severely de-

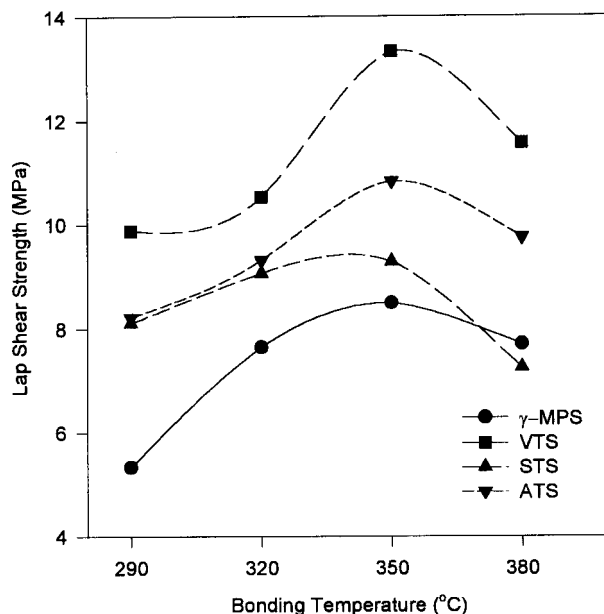


**Figure 6** The schematic diagram of the interaction between silane-modified PVI and copper.

graded, and cuprous oxide peaks are observed at 655 and 611  $\text{cm}^{-1}$  in both systems. The extent of thermal degradation and copper oxidation is more remarkable in the PI-Cu system.

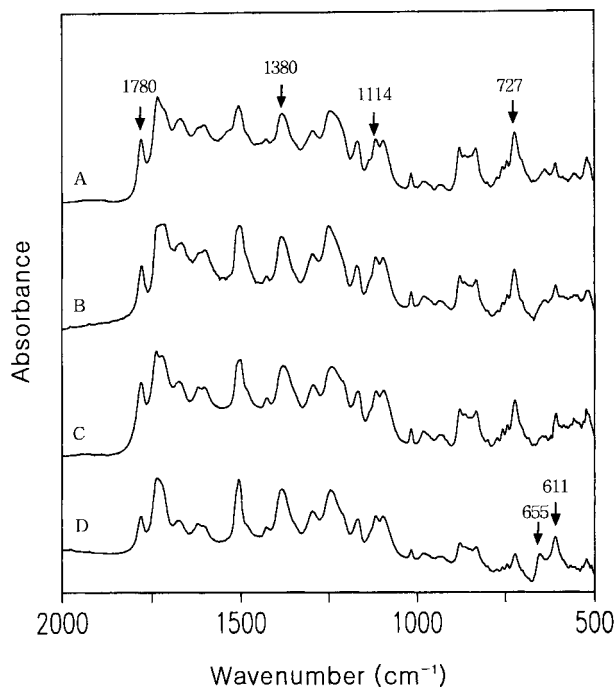
Figure 6 represents the interaction between silane-modified PVI and copper surface. Silane unit and VI unit in the copolymer can form the covalent bond and the coordinated bond with copper surface, respectively. Since the covalent bond of silane is stronger than the coordinated bond of PVI, silane-modified PVI has a stronger interaction with copper surface than PVI.<sup>23</sup> Generally, the silane unit of silane-modified PVI is thermally more stable than the VI unit at elevated temperature. In addition, silane unit prevents the diffusion of copper into polyimide effectively.<sup>23</sup> The migration of copper in the silane-modified PVI film is suppressed because the silanol group of silane unit interacts strongly with copper.

Figure 7 shows the lap shear strengths of PI-silane-modified PVI-Cu as a function of bonding temperature. In all cases, the lap shear strengths increase up to 350°C and then decrease. VTS-modified PVI and ATS-modified PVI enhance the lap shear strengths between copper and PI. However, STS-modified PVI and  $\gamma$ -MPS-modified PVI are not effective in improving the lap shear strengths. Especially, VTS-modified PVI shows the remarkable improvement of the lap shear strengths in all bonding temperatures. The lap shear strengths in these systems can be affected by several factors, as follows: the interfacial strength between copper surface and primer, the interfacial strength between primer and polyimide, the degree of copper diffusion into PI, and the thermal stability of primer. To investigate the effects of these factors on lap shear strength, FTIR analysis, contact angle measurement, and scanning electron microscopic (SEM) analysis were carried out.

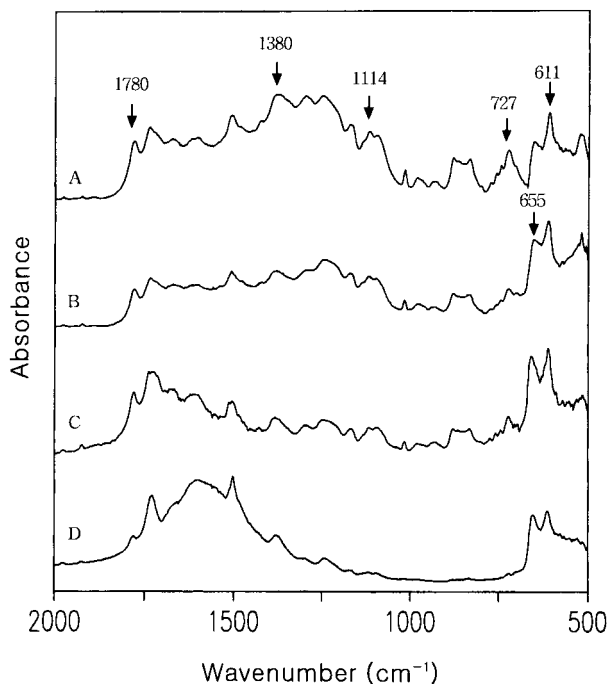


**Figure 7** The lap shear strength of the PI-silane-modified PVI-copper system as a function of bonding temperature.

In Figures 8 and 9, the thermal degradation behaviors of PI on copper treated with silane-modified PVIs are shown. The extent of PI degradation



**Figure 8** R-A spectra of PI-silane-modified PVI-copper heated at 350°C: (A) poly(VI-co-VTS), (B) poly(ATS-co-VI), (C) poly( $\gamma$ -MPS-co-VI), and (D) poly(STS-co-VI).



**Figure 9** R-A spectra of PI-silane-modified PVI-copper heated at 380°C: (A) poly(VI-co-VTS), (B) poly(ATS-co-VI), (C) poly( $\gamma$ -MPS-co-VI), and (D) poly(STS-co-VI).

at 350 and 380°C depended on the sorts of the primers. In the case of STS-modified PVI, the reductions of the peak intensities at 1780, 1380, 1114, and 727  $\text{cm}^{-1}$  are observed at 350°C, and cuprous oxide peaks at 655 and 611  $\text{cm}^{-1}$  begin to appear. This phenomenon is more outstanding at 380°C. These results indicate that STS-modified PVI has the worst thermal stability on copper surface.

In contrast to STS-modified PVI, the spectrum of PI-VTS-modified PVI-copper shows a little reduction of PI peak intensities, and cuprous oxide peaks are not observed at 350°C. Even at 380°C, the reduction of PI peaks and the peak intensities of cuprous oxide are less than the other spectra. This means that VTS-modified PVI has the excellent thermal stability on copper surface and that it inhibits the diffusion of copper into PI most effectively.

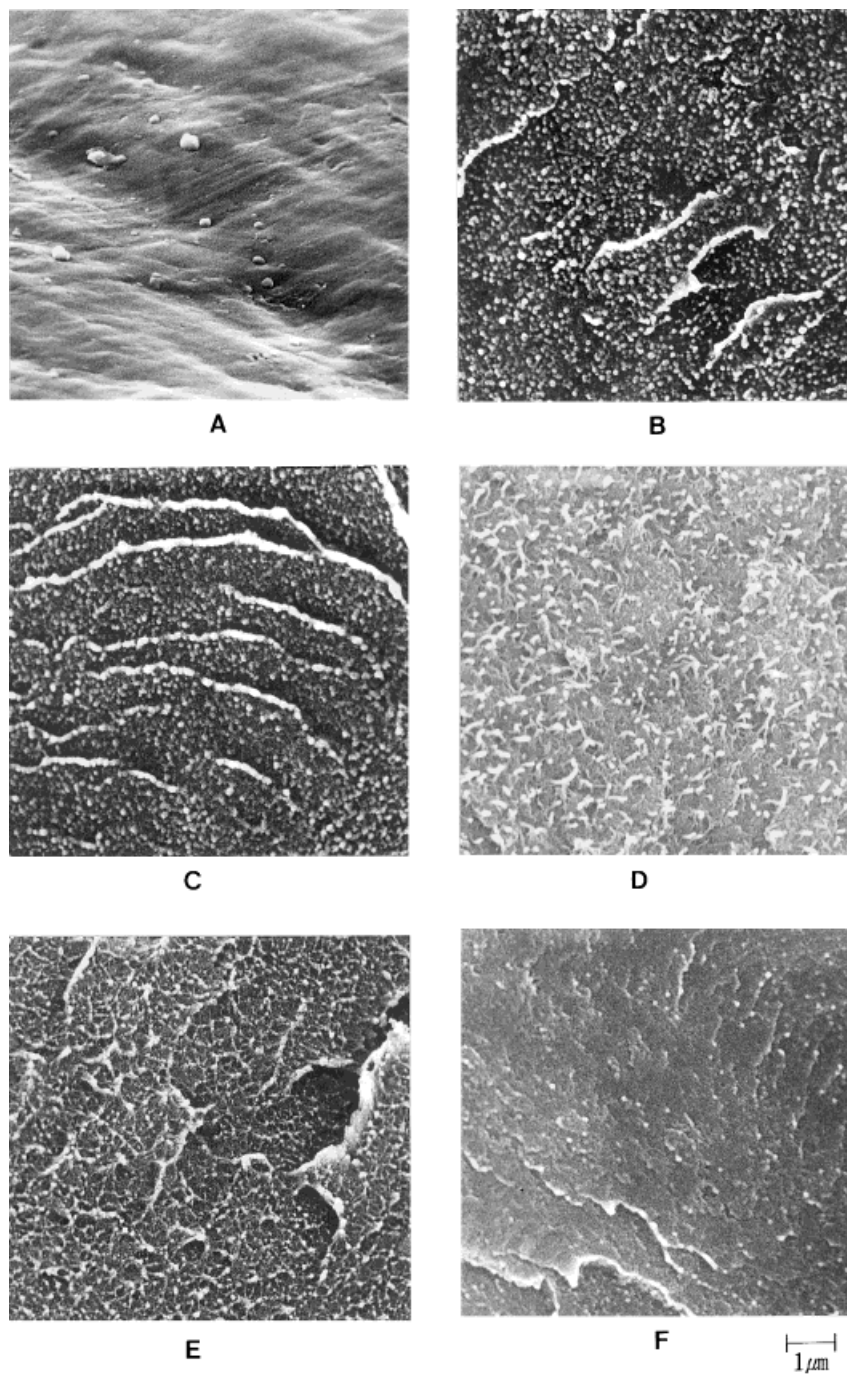
The spectra of PI on ATS-modified PVI and PI on  $\gamma$ -MPS-modified PVI show the intermediate trends between PI on STS-modified PVI and PI on VTS-modified PVI. The PI peaks are not reduced at 350°C but conspicuously reduced at 380°C. Cuprous oxide peaks are hardly observed at 350°C. On the other hand, at 380°C, PI on  $\gamma$ -

MPS-modified PVI shows a strong peak at 655  $\text{cm}^{-1}$  and weak PI peaks in comparison with PI on ATS-modified PVI. Therefore, ATS-modified PVI has better thermal stability and is more effective to suppress the diffusion of copper into PI than the  $\gamma$ -MPS-modified PVI.

From these spectra, the thermal stabilities of silane-modified PVIs decreased in the order of VTS, ATS,  $\gamma$ -MPS, and STS. The thermal degradation of PI by copper diffusion was also suppressed in the same order. Kim et al. reported that as the number of siloxane linkages per weight increases in the silane coupling agent, silane-modified PVI has better thermal stability and inhibits copper diffusion into PI more effectively.<sup>23,24</sup>

In order to investigate the initial wettabilities of PAA solution on various primers, contact angles were measured by dropping the diluted PAA solution onto the copper surfaces coated with PVI or silane-modified PVIs. The observed contact angles are represented in Table II. The contact angle of PAA solution on PVI is very small. This implies that PVI can form a good interface with PAA solution easily in the initial stage of coating, whereas the contact angles on copper surfaces coated with silane-modified PVIs are large in comparison with that on PVI. This means that the introduction of silanes into PVI has a negative effect on formation of good interfaces between primers and PAA solution. The contact angle on  $\gamma$ -MPS-modified PVI, especially, shows the largest.

To investigate the miscibilities between PI and primers, the fractured surfaces of PI-primer blend films were observed by SEM. The fractured surfaces of PI and PI blended with primers are shown in Figure 10. Pure PI shows a smooth fracture surface. In the case of  $\gamma$ -MPS-modified PVI, a lot of small spheres are seen. These small spheres are thought to be formed by phase separation between PI and  $\gamma$ -MPS-modified PVI. This suggests that  $\gamma$ -MPS-modified PVI has the poor miscibility with PI. STS-modified PVI shows a similar trend to  $\gamma$ -MPS-modified PVI. In PI-ATS-modified PVI system, many fibrils are observed. These fibrils are considered to be formed as a result of the interaction between PI and primer. Accordingly, it can be inferred that ATS-modified PVI is more miscible with PI than the previous two silane-modified PVIs. In the fractured surface of PI-VTS-modified PVI blend film, the trace of phase separation is hardly seen. This suggests that VTS-modified PVI has better miscibility than the other silane-modified PVIs. There is no sepa-



**Figure 10** Scanning electron micrographs of the fractured surfaces of pure PI and PI blended with primer: (A) pure PI, (B) poly( $\gamma$ -MPS-co-VI), (C) poly(STS-co-VI), (D) poly(ATS-co-VI), (E) poly(VI-co-VTS), and (F) PVI.

rated particle in the PI–PVI system. Judging from these results, it can be concluded that four silane-modified PVIs show the different compatibilities with PI. Specifically, VTS-modified PVI has an adequate compatibility with PI. Therefore, the interdiffusion of polymer chains between

primer and PI can be facilitated at bonding temperatures, and a strong interface can be formed. On the other hand,  $\gamma$ -MPS-modified PVI shows very poor compatibility with PI. This poor compatibility of  $\gamma$ -MPS-modified PVI weakened the interfacial adhesion strength between primer and

**Table II The Contact Angles of PAA Solution on Primer-Treated Copper**

Primer	Contact Angle (°)
PVI	14.5
Poly(VI-co-VTS)	41
Poly(ATS-co-VI)	49
Poly( $\gamma$ -MPS-co-VI)	60
Poly(STS-co-VI)	53

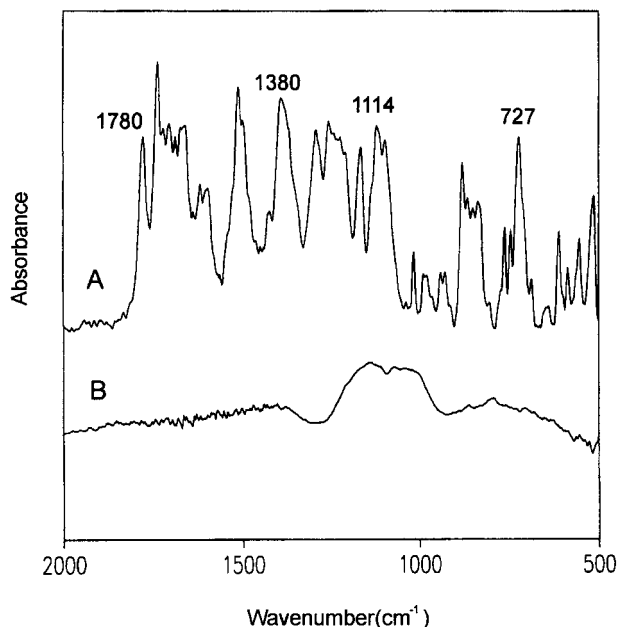
PI and contributes to the poor lap shear strength. These different miscibilities between primers and PI can be confirmed with R-A spectra of copper surface after the lap shear test. R-A spectra of VTS-modified PVI and  $\gamma$ -MPS-modified PVI tested at 350°C are shown in Figure 11. In the case of VTS-modified PVI, PI characteristic peaks are observed as a result of the cohesive failure of PI layer. This supports the existence of a strong interface between PI and VTS-modified PVI, whereas,  $\gamma$ -MPS-modified PVI shows no PI peaks and the peaks from siloxane linkages are observed in the 1250–950  $\text{cm}^{-1}$  region. This suggests that the adhesive failure occurred at the interface between PI and  $\gamma$ -MPS-modified PVI.

From these IR, SEM, and contact angle results, some conclusions can be derived as follows. In Figure 7, the lap shear strength decreases in the order of VTS, ATS,  $\gamma$ -MPS, and STS-modified PVI at 380°C. This trend is well matched with the order of the thermal stabilities of PIs on the different silane-modified PVIs. Therefore, the lap shear strength of PI–silane-modified PVI–copper system at 380°C is thought to be mainly influenced by the thermal stability of PI on silane-modified PVI. On the other hand, the lap shear strengths below 350°C are affected by several factors, as mentioned previously. In all systems, the lap shear strength increases up to 350°C, a trend that can be explained by the effect of the molecular packing and the interdiffusion between primer and PI. However, STS-modified PVI did not show the distinct maximum value at 350°C, which is attributed to the thermal degradation of PI on the silane-modified PVI and the copper–primer interface deteriorated by copper corrosion (Fig. 8). VTS-modified PVI system also showed the highest lap shear strength below 350°C. This is due to a good miscibility of PI and VTS-modified PVI as well as a strong copper–silane-modified PVI interface. At 350°C, the high thermal stability of PI on VTS-modified PVI also contributes to the improvement of lap shear strength (Fig. 8). The

trend of ATS-modified PVI can be explained in the same way as VTS-modified PVI. However, its miscibility with PI and the thermal stability of PI on it are worse than VTS-modified PVI. Therefore, its lap shear strength is lower than VTS-modified PVI in all bonding temperatures.  $\gamma$ -MPS-modified PVI system showed the lowest lap shear strengths below 350°C. This is because  $\gamma$ -MPS-modified PVI has the poorest miscibility with PI.

## CONCLUSION

The lap shear strengths of the PI–primer–copper systems were measured to investigate the effects of primer on the interfacial adhesion strength between copper and PI. VTS-modified PVI and ATS-modified PVI improved the lap shear strength of PI–copper, but PVI, STS-modified PVI, and  $\gamma$ -MPS-modified PVI were not effective. The lap shear strengths of PI–primer–copper systems were affected by the following four factors: the interfacial adhesion strength between copper surface and primer, the interfacial strength between primer and polyimide, the capability of primer to suppress copper diffusion into PI, and the thermal stability of primer. PVI showed the best compatibility with PI. The capability of silane-modified PVI to suppress copper diffusion into PI and the



**Figure 11** R-A spectra of the lap-sheared copper surfaces of the samples prepared at 350°C: (A) poly(VI-co-VTS) and (B) poly( $\gamma$ -MPS-co-VI).



thermal stability of PI on the silane-modified PVI decreased in the order of VTS, ATS,  $\gamma$ -MPS, and STS-modified PVI. PI-VTS-modified PVI-copper system showed the highest lap shear strength at all bonding temperatures and the maximum value at 350°C.

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