Low transmission loss flexible substrates using low Dk/Df polyimide adhesives

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ABSTRACT

We developed solvent-soluble polyimides with good heat resistance and low dielectric constant (Dk) / dissipation factor (Df) characteristics by optimizing the composition ratio of the aliphatic, cycloaliphatic, and aromatic groups present in the polyimide back bone. We found that the adhesives prepared using our polyimides showed good adhesion to polyimide films and low profile copper foils and low Dk / Df. Furthermore, we developed a flexible copper clad laminate (FCCL) with our polyimde (PI) adhesives, low-pofile copper foils, and normal PI films. This FCCL showed a transmission loss similar to that of liquid crystal polymer (LCP) FCCLs at frequencies less than 15GHz. This result indicates that by using our PI adhesives, lower cost FCCLs can be prepared, which can then be used as high-frequency substrates.

Keywords: transmission loss, insertion loss, flexible printed circuit boards (FPCBs), lowDk/Df polyimides, liquid crystal polymers (LCPs)

1 INTRODUCTION

The field of wireless communication and broadband technology has progressed dramatically with a growth in the market for information technology gadgets such as mobile phones and tablets. To meet the ever-increasing requirements of transmission data, the transmission frequency of circuits is increased. However, the integrity of high-frequency signals can be damaged by transmission loss.

In high-frequency circuits, two factors are responsible for the transmission loss: conductor (circuits) loss and dielectric (insulating materials) loss. The former is related to the skin effect of circuits. The skin effect is the tendency for an alternating current to flow mainly near the surface of conductors. The general method for reducing the conductor loss is to make the surface of circuits smooth. However, smoothing the surface of copper circuits tends to weaken the adhesion between them and the insulating materials. Hence, it is difficult to fabricate copper circuits with a smooth surface and good adhesion to insulating materials.

The dielectric loss of dielectric materials depends on their current frequency, dielectric constant (Dk), and dissipation factor (Df). Consequently, the dielectric loss increases with an increase in the current frequency. The general method for reducing this loss is to use low Dk and Df materials.

Hence, low Dk and Df materials having good adhesion to the smooth copper surfaces are required for the fabrication of high-frequency printed circuit boards (PCBs).

We recently developed novel solvent-soluble polyimides. By using these materials, we obtained low Dk and Df adhesives with good heat resistance and adhesion to smooth copper surfaces such as rolled copper foils.

In this study, we investigated the properties of the polyimides developed by us previously along with those of the adhesives prepared using these polyimides. We also investigated the effect of these adhesives on the transmission loss of copper circuits.

2 POLYMER DESIGN

In PCBs, thermosetting insulating materials are used. For example, coverlays and flexible copper clad laminates(FCCLs) are used as raw materials for flexible printed circuit boards (FPCBs). These materials are mainly composed of cross-linking agents such as epoxy resins, fillers such as silica or organic phosphate to control the coefficient of thermal expansion (CTE) or to provide non-flammability, and polymers to provide flexibility and good adhesion.

Examples of polymers used in FPCBs include acrylic polymers, butadiene-acrylonitrile copolymers, and phenoxy resins. In general, the polymers used in PCBs should possess heat resistance (mainly for solder reflow process), laminating workability, good adhesion properties, and flexibility. As already discussed, low Dk and Df materials are required for PCBs. Hence, the polymers used in PCBs should also have low Dk and Df. However, it is difficult for polymers to achieve a good balance between low dielectric properties and the other properties mentioned above. In addition, good adhesion to smooth copper surfaces is necessary to minimize the conductor loss.To meet these requirements novel polymers should be developed. Therefore, we focused on solvent-soluble polyimides as polymers required for high frequency PCBs.

Polyimides possess excellent thermal, mechanical, and chemical properties and have been applied to various microelectronic applications such as IC passivation overcoats, FPCBs etc. Conventional polyimides are insoluble in organic solvents because of their strong cohesive and orientation forces, which result from their linear / stiff chains and imide groups having high polarity. Hence, for the fabrication of conventional polyimides, the preparation of a poly (amic acid) solution (precursor) is necessary. Poly (amic acid) is prepared by imidization of dianhydrides and diamines. The treatment of poly(amic acid) solution results in a decrease in its molecular weight and deteriorates its absorbent property. Hence, the treatment should be carefully monitored. Imidization is a difficult reaction as it requires high temperatures (300 °C or more) and causes dehydration. Hence, the applications of polyimides are limited as they are difficult to process.

In order to make the processing of polyimides easy, many researchers have developed solvent-soluble polyimides. The primary objective of improving the solvent solubility is to decrease the interaction among the polymer chains and stiffness of the polymer structure by introducing non-coplanar, flexible, and kinked units [1].

Much research has been done on decreasing the dielectric constant of polyimides. According to the Clausius-Mossotti equation, which gives the relation between the dielectric constant and molecular polarization, the dielectric constant of a material is proportional to the polarization rate and is inversely proportional to the molar free volume content [2]. Hence, the introduction of fluorine and cycloaliphatic groups [3], bulky groups [4], poly(silsesquioxane) [5], hyper branched structures, and silica [6] into polyimide structures has been carried out for improving their dielectric properties.

Moreover, the thermosetting insulating materials used in PCBs should be easy of heat treatment (drying, laminating, curing) at temperatures below 200 °C. Hence, the softening point of the polymers used in PCBs should be controlled. The softening point of materials reduces with a decrease in the number of functional groups and by the introduction of the flexible units used for improving the solvent solubility. In polymers, a reduction in the softening point makes their processing easy. However, it deteriorates their handling properties because of an increase in the stickiness and a decrease in the heat resistance and adhesion to films and metal foils.

Based on the above discussion, we tried to design optimal polyimides for adhesives; in other words, we tried to optimize the composition ratio of aliphatic, cycloaliphatic, and aromatic groups in the polyimide backbone. We developed novel solvent-soluble polyimides with low Dk and Df, high heat resistance, good adhesion, and stability against processing conditions. Figure 1 shows the schematic of the polyimide structure developed by us.

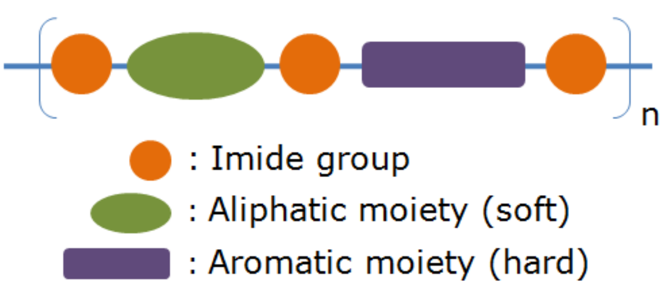


Figure 1: Schematic of the polyimide structure.

3 PROPERTIES OF POLYIMIDES

Figure 2 shows the photographs of the polyimide resin (in solution and solid states) developed by us. These polyimides show good solubility in solvents with relatively low boiling points (below 160 °C) such as toluene, cyclohexanone, butyl acetate, etc. These properties make these polyimides suitable for PCBs.

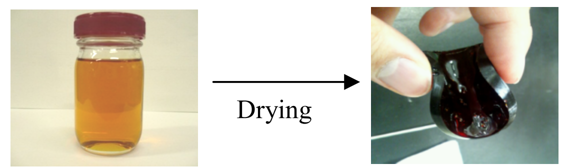


Figure 2: Photographs of the polyimide resin.

Table 1 lists the mechanical and electrical properties of the polyimides developed by us. The fracture elongation (100% or more) and elastic modulus (136 MPa) values indicate that the polyimides have good ductility and flexibility. The water absorption rate of the polyimides is less than 1%. This value is lower than the water absorption rate of conventional polyimides (2–3%). This low water-absorption improves the insulation properties of the polyimides. Note that the dielectric properties of these polyimides at 10 GHz (Dk = 2.5, Df = 0.0017) are lower than those of normal polyimides (Dk = 3.2~3.3, Df = 0.005~0.01) and as low as those of liquid crystal polymers (LCPs) (Dk = 2.9~3.2, Df = 0.002). The thermal decomposition temperature (1%) of the polyimides is greater than 300 °C. Thus, these polyimides are suitable for the solder reflow process carried out at a temperature of 288 °C. Hence, we think that these polyimides possess properties which made them a good choice for PCB polymers particularly for high-frequency applications.

Figure 3 shows the extent to which the properties of our polyimides could be controlled. The left vertical axis shows Df, while the right vertical axis shows Dk. The horizontal axis shows the Tg of the polyimides. Red square points show Dk. Blue rhombic points show Df. We could control the Df (less than 0.0035), Dk (less than 2.8) and Tg (from 80 to 180 °C) of these polyimides.

Test item		Unit	Values	Test Method
Maximum Stress		MPa	6.91	ASTM D882-97 IPC-TM-650 m2.4.19
Fracture Elongation		%GL	100 <	
Elastic modulus		GPa	0.136	
Water Absorption Rate		%	0.60	25 °C×24h
Dielectric Constant at 10GHz		–	2.50	ASTM D2520
Dissipation Factor at 10GHz		–	0.0017	
Weight Loss Temperature	1%	–	330	TG/DTA
	5%		440	
	10%		455	
Softening point		°C	80	Rheometer

Table 1: Properties of our polyimides.

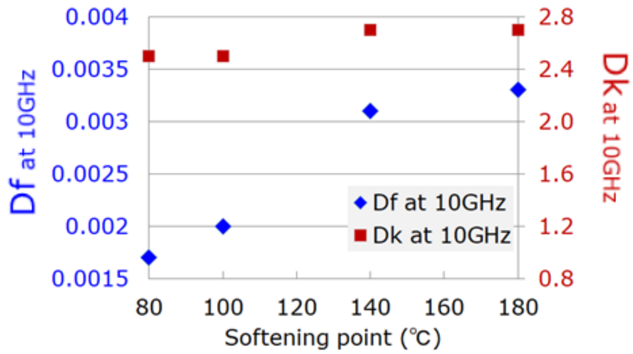


Figure 3: The extent to which the properties (Dk, Df, and softening point) of our polyimides could be controlled.

4 PROPERTIES OF THE ADHESIVE FOR A FCCL

As mentioned before, FCCLs are used as substrates for FPCs. Polyimides are conventionally used as insulating layers in FCCLs and FPCs. But they are unsuitable for high freqeuincy applications because of their high Dk and Df. On the other hand, there have been reports of using LCPs in substrates and adhesives for FCCL and FPCs because of their low Dk and Df values. However, special apparatus is required along with high temperature and pressure conditions to realize the lamination between LCPs. It is known that the adhesion of LCPs to copper circuits is not so good. Because of these problems, the applications of LCPs are limited. Therefore, we compared our polyimides with LCPs as raw materials for FCCLs to check whether our polyimides can overcome the problems of LCPs.

We prepared an adhesive solution for a FCCL, as shown in Figure 4 . We mixed two types of polyimides (high Mw-type and low Mw-type) developed by us for improving the adhesion, heat resistance, and compatibility with the cross-linking agents. We used multifunctional epoxy and cyanate ester as cross-linking agents.The concentration of the adhesive solution was made 30% by using toluene and cyclohexanone.

We prepared the FCCL samples (Figure5) according to the production flow diagram shown in Figure6. First, we coated low-profile rolled copper foils made in JX Nippon Mining & Metals Corporation, GHF5 with an Rz (ten point mean roughness) of 0.45 µm and a thickness of 12 µm with the above-mentioned adhesive solution. Next, the coated samples were baked at 150 °C for 3 min to obtain B-stage adhesive-backed copper foils with a 12–13 µm thick adhesive layer. The B-stage adhesive-backed copper foils were bonded to the top and bottom surfaces of a 25 µm thick polyimide film made in DU PONT-TORAY CO., LTD., Kapton EN at 170 °C and curing the adhesive at 180 °C for 4h to obtain the adhesive test samples.

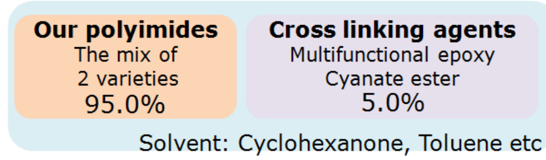


Figure 4: Composition of the adhesive solution for a FCCL.

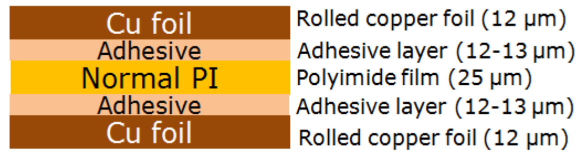
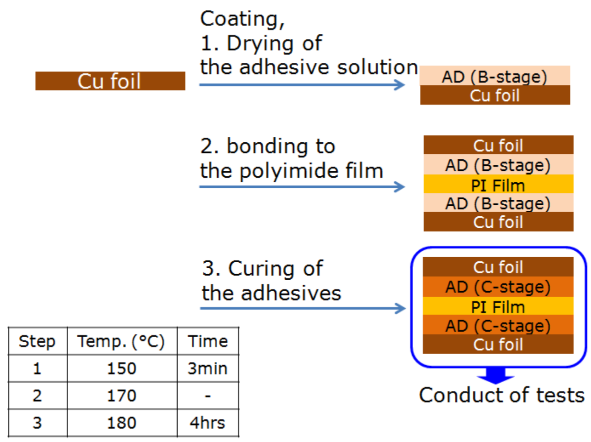


Figure 5: Structure of the adhesive test sample for a FCCL.



Step	Temp. (°C)	Time
1	150	3min
2	170	-
3	180	4hrs

Figure 6: The production flow diagram for the adhesive test sample for a FCCL.

We investigated the properties of these samples. Table 3 lists the test results of the test sample. The peeling strength (0.8 N/mm) indicates that this adhesive showed good adhesion to the polyimide film and low-profile copper foil. Moreover, the Dk (2.5) and Df (0.003) values for this adhesive layer were low as compared to normal polyimides (Dk = 3.2~3.3, Df = 0.005~0.01). On the basis of the test results listed in Table 3, we can conclude that this adhesive test sample is suitable for the FCCLs used in FPCs operating at high frequencies.

Items	Targets	Results
Peeling strength (90°, 50mm / min)	0.6 N / mm <	0.8 N / mm
Dk at 10GHz*	< 2.8	2.5
Df at 10GHz*	< 0.005	0.003
Solder heat resistance (288°C x 3min)	No foaming No delamination	No foaming No delamination

* We tested the adhesive alone.

Table 2: Test results of the FCCL using our polyimdes.

We also investigated the transmission loss of a micro-strip line fabricated by using the PI-type FCCL developed by us (represented as "Our PI").

At the same time, we also tested a normal PI-type FCCL(represented as "Normal PI"). and an LCP-type FCCL(represented as "LCP") as a reference. In these reference FCCLs, The specifications of the micro-strip line used in this test are shown in Figure 11, and the structure of the samples used is shown in Figure 12. Table 4 lists the dielectirc properties of each sample used in this test.

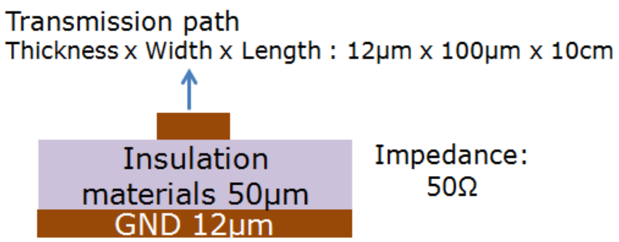


Figure 7: Test results of the FCCL using our polyimdes.

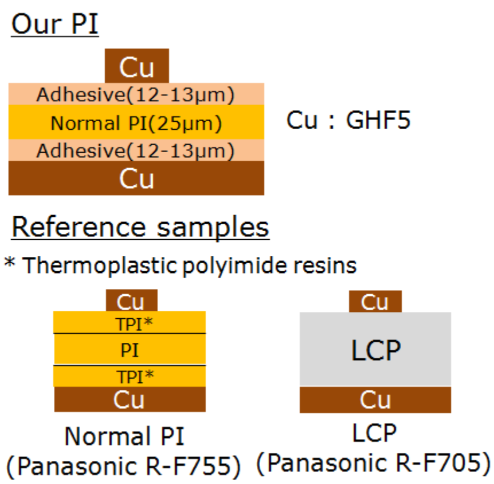


Figure 8: Structure of the samples used in the transmission loss test.

Sample type	Dk / Df@10GHz	
	Dk	Df
Our PI (adhesive only)	2.5	0.003
Normal PI	3.2	0.007
LCP	3.0	0.002

Table 3: Dk/Df of each sample used in the transmission loss test at 10 GHz.

Figure 9 shows the results of the transmission loss test (insertion loss, S21) carried out using a network analyzer. The S21 of our polyimide adhesive samples (represented as "Our PI" in the figure) were lower than that of the normal PI-type FCCL (represented as "Normal PI" in the figure) and Our PI was cl ose in value to that of the LCP-type FCCL (represented as "LCP" in the figure) below 15GHz.

This result suggests that FCCLs with low transmission loss like LCP FCCL can be manufactured even in the case of using normal PI as core substrates by using our PI adhesives and low profile copper foils. The FCCL obtained by using the PI adhesives developed by us and low profile copper foils was lower cost than the LCP-based FCCL, and thus can be used for the widespread commercialization of high-frequency FCCLs.

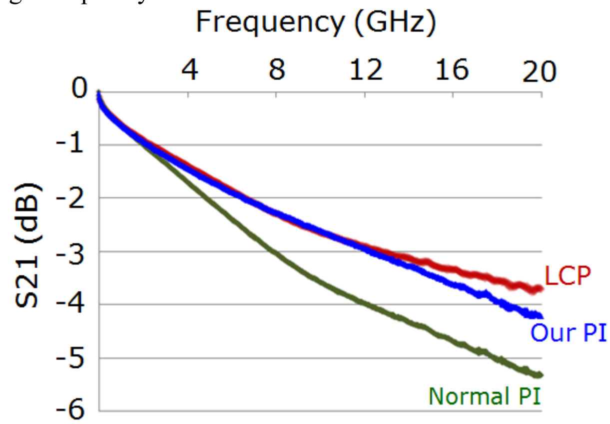


Figure 9: Transmission loss test results (insertion loss, S21).

5 CONCLUSIONS

We developed solvent-soluble polyimides with good heat resistance and low Dk/Df characteristics by optimizing the composition ratio of the aliphatic, cycloaliphatic, and aromatic groups present in the polyimide backbone. We found that the adhesives prepared using our polyimides showed good adhesion to polyimide films and copper foils, good heat resistance, and low Dk / Df. Furthermore, we developed a FCCL with our PI adhesives, low-profile copper foils, and normal PI films. This FCCL showed a transmission loss similar to that of LCP FCCLs at frequencies less than 15 GHz. This result indicates that by using our polyimide adhesives, lower cost FCCLs can be prepared, which can then be used as high-frequency substrates.

Hence, we believe that the polyimides developed by us are suitable to be used as raw materials for high-frequency PCB substrates.

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