

Development of Flexible Polymer Blend Films from Advanced COPNA Resin and Nylon 6

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SYNOPSIS

The application of advanced polycondensed fused *polynuclear aromatic* (COPNA) resin to a material for flexible printed wiring boards was studied in this work. To give flexibility to the advanced COPNA resin, a polymer blend technology was introduced. Nylon 6 was selected from the nominated blend polymers with advanced COPNA resin because of sufficient compatibility between the advanced COPNA resin and the amino groups. The polymer blend film, consisting of advanced COPNA resin and nylon 6 prepared at a blend fraction of 50/50 (COPNA/nylon 6), exhibited sufficient flexibility for printed wiring boards, and exhibited attractive properties of T_g at 158°C and a dielectric constant at 3.6. Morphological analysis was also carried out by transmission electron microscopy to the blends. Phase-separated structures were observed in entire systems of fully cured films because of crystallinity of the nylon 6, but sufficient compatibility for practical uses was observed in both the 30/70 and 50/50 systems. The compatibility became poor with an increase of the advanced COPNA resin fraction. © 1996 John Wiley & Sons, Inc.

INTRODUCTION

Because large-scale integrated (LSI) process technology to expand the memory size of LSI chips has been highly advanced, 1 GB scale technology was developed and presented in technical conferences.^{1,2} The microprocessors for personal computers have been improved to increase the processing speed and to exhibit more typical functions. Therefore, the numbers of terminals of LSI chips have increased, and the width of the lines and spaces of terminals of lead frames have become as narrow as 300 μm .^{3,4} The chip size of electrical resistance and electrical capacitance has become as small as 1 mm. With the spread of recent technological trends, the density of electronic packages has become dramatically higher; moreover, novel types of electronic packaging technology have been invented to realize ultrahigh density as breakthrough technologies. In the field of conventional rigid printed wiring boards, there are now more than 10 internal layers and the thickness of prepregs and laminates has become less than 50

μm to meet recent technological demands.⁵ Flexible printed wiring boards and multichip modules were invented as breakthrough technologies to realize ultrahigh packaging density. The flexible printed wiring boards, invented to save space, have been generally used in compact cameras and liquid crystal displays. The flexible printed wiring boards have been produced by attaching copper foils to flexible polymer films.^{3,4} Multichip modules, invented to prevent transmission delay, have been used as a switching system of asynchronous transfer mode technology.^{6,7} The multichip modules have been produced to build up electrical insulating thin films and electrical conductors alternately onto silicon wafers.^{6,7}

Polycondensed fused *polynuclear aromatic* (COPNA) resin was suggested in 1986 as a novel type of thermosetting and heat-resistant resin.⁸ The COPNA resin was synthesized by the dehydrating reaction of fused aromatic rings and 1,4-benzenedimethanol. The reaction catalyst and condition were improved to avoid the defects of the initial COPNA resin, and advanced COPNA resin was invented at Sumikin Chemicals.⁹⁻¹¹ The chemical reaction and chemical structure of the advanced

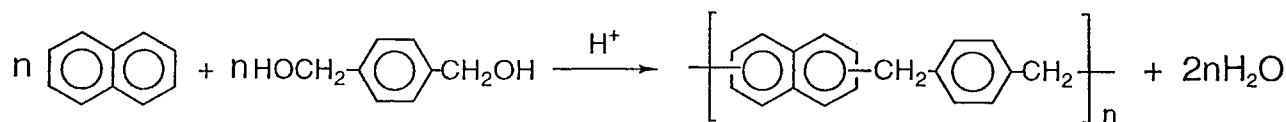


Figure 1 Chemical reaction of advanced COPNA resin.

COPNA resin are shown in Figure 1. The fully cured advanced COPNA resin exhibits a 3-dimensional network structure, as shown in Figure 2. The resin exhibits a characteristic structure: aromatic rings and naphthalene rings are connected by methylene groups, and the resin does not include any functional groups, therefore, it exhibits strong heat resistance (T_g 250°C), a low dielectric constant (ϵ 3.1), and a low water absorption (0.37 wt %) as shown in Table I. The new resin was considered to be suitable for an electrical insulator, so application of the resin to electrical insulating materials was begun. Three targets were selected for development: prepregs and laminates for rigid printed wiring boards, flexible films for flexible printed wiring boards, and internal insulating layers for multichip modules. The application of the advanced COPNA resin to rigid printed wiring boards¹²⁻¹⁴ and multichip modules was previously reported.¹⁵⁻¹⁷ The fabrication and properties of prepregs and double-sided copper-clad laminates, using advanced COPNA resin as an electrical insulating material, were shown in the initial application. The laminates, reinforced by E-glass fiber woven fabrics, exhibited a glass transition temperature (T_g) of 255°C, a dielectric constant of 4.0 at 1-MHz frequency, and a linear thermal expansion coefficient of 5-7 ppm (xy axis) and 29 ppm (z axis). In the later application, synthesis of impurity-free

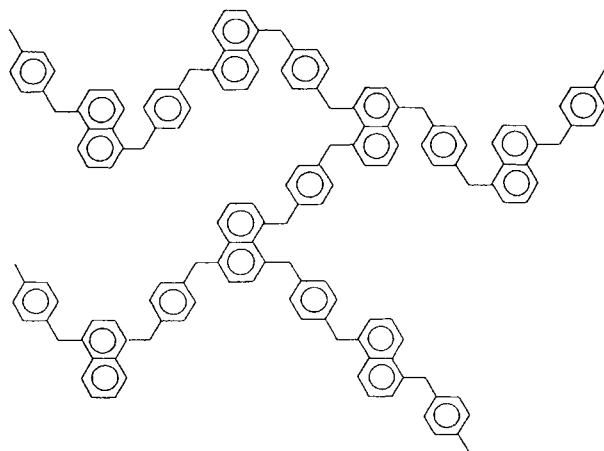


Figure 2 Network structure of fully cured advanced COPNA resin.

resin, a spin-coating process onto silicon wafers, properties of spin-coated thin films obtained, and properties of dry etching of the thin films were reported.

Flexible printed wiring boards are generally produced by laminating of copper foils and flexible polymer films. Characteristic properties have been required of the polymer films because the packaging density has been highly advanced, and the cost of the materials has been highly competitive. The polymer films are required to exhibit flexibility, heat resistance, a low dielectric constant, and cost performance. The properties of the polymer films, polyimide films, and polyester films, generally used as materials for flexible printed wiring boards, are shown in Table II.¹⁸ The properties of epoxy resin used for FR-4 graded rigid printed wiring boards are also shown for comparison.^{19,20} The polyimide films exhibit the outstanding properties of a low dielectric constant and strong heat resistance. However, they have two weak points: high water absorption and high cost. The polyester films exhibit attractive cost performance, but have weak heat resistance owing to a property peculiar to thermoplastic resins. Therefore, the application of the advanced COPNA resin, exhibiting high T_g , a low dielectric constant, and low water absorption, to the materials of flexible printed wiring boards was studied.

The fully cured advanced COPNA resin, however, exhibits poor flexibility because of its 3-dimensional network structure as shown in Figure 2. For example, the advanced COPNA resin films obtained from the solvent casting method or hot-press fabricating

Table I Properties of Advanced COPNA Resin

Properties	Values
Glass transition temperature (°C)	250
Thermal expansion coefficient (ppm)	56
Tensile strength (kgf/mm ²)	4.7
Volume resistivity (Ω cm)	5×10^{15}
Dielectric constant (at 1 MHz)	3.05
Dissipation factor (at 1 MHz)	0.0028
Water absorption (wt %)	0.37
Specific gravity	1.20

Table II Properties of Polyimide Film, Polyester Film, and Epoxy Resin

Properties	Polyimide ^a	Polyester ^a	Epoxy ^b
Glass transition temperature (°C)	—	—	125–135
Melting point (°C)	—	263	—
Thermal expansion coefficient (ppm)	20	30	60–70
10% weight loss temperature (°C)	520	—	350
Tensile strength (kgf/mm ²)	25	25	7–10
Young's modulus (kgf/mm ²)	302	400	350–400
Elongation (%)	80	130	—
Dielectric constant (at 1 MHz)	3.5	3.2	4.0–4.5
Dissipation factor (at 1 MHz)	0.003	0.01	0.03–0.05
Volume resistivity (Ω cm)	10 ¹⁸	10 ¹⁸	10 ¹⁵ –10 ¹⁶
Water absorption (wt %)	2.9	0.4	0.08–0.15

^a Data from *Guide Book for Plastic Films '93*.¹⁸^b Data from Sakurai.¹⁹

method, exhibit poor flexibility similar to that of epoxy resin; they generate cracks in the folded region when they are folded up to 180°. Therefore, polymer blend technology was introduced to give the advanced COPNA resin sufficient flexibility for flexible printed wiring boards. Polyamide resin was selected from several kinds of polymers nominated for the polymer blend system with the advanced COPNA resin, because the polyamide resin did not prevent the dehydrating reaction of the advanced COPNA resin, and amino groups included in the polyamide resin exhibited sufficient compatibility with the advanced COPNA resin.²¹ In this article nylon 6 was selected as the polyamide resin, and the compatibility of the advanced COPNA resin with the nylon 6, fabrication of polymer blend films, properties of the obtained blend films, and their structure are reported.

EXPERIMENTAL

Materials

Advanced COPNA resin (SKR-NM) was from Sumikin Chemicals. B-staged advanced COPNA resin exhibited M_n 2.5×10^2 , M_w 9.1×10^2 , and M_w/M_n 3.70. Viscosity was 500 P at room temperature. Synthesis, details of characteristics, and a background of the advanced COPNA resin were described in previous papers.^{12–17} Nylon 6 was from Ube Chemicals (1013B). Molecular weights of the nylon 6 were M_n 2.82×10^4 , M_w 5.59×10^4 , and M_w/M_n 1.98. Properties of the resin are shown in Table III.²² Methylene chloride and 1,1,1,3,3,3-hexafluoro-2-

propanol were from Kishida Chemicals. The copper paste was from Kyoto Elex.

Apparatus

The impedance analyzer and high resistance meter were Hewlett-Packard HP4194A and HP4329A, respectively. The tensile strength measuring machine was a Shimadzu AGS-50ND. The transmission electron microscope was a Hitachi H-4100FA. The differential scanning calorimeter, thermal mechanical analyzer, and thermogravimeter was Mac Science DSC 3200, TMA 4000, and TG 2000, respectively.

Fabrication of B-Staged Polymer Blend Films

The advanced COPNA resin solution (methylene chloride as solvent) was prepared at the concentration of 9.1 wt %. The nylon 6 solution (1,1,1,3,3,3-hexafluoro-2-propanol as solvent) was prepared at the concentration of 5.4 wt %. These two prepared

Table III Properties of Nylon 6

Properties	Values
Glass transition temperature (°C)	60–70
Melting point (°C)	215–225
Thermal expansion coefficient (ppm)	80
Tensile strength (kgf/mm ²)	4.0
Elongation (%)	200
Dielectric constant (at 1 MHz)	4.5
Dissipation factor (at 1 MHz)	0.12
Volume resistivity (Ω cm)	1×10^{15}
Water absorption (wt %)	1.8

solutions were mixed at the ratio of 30/70, 50/50, and 70/30. (Note: In this work, blend fraction means weight percent of advanced COPNA resin and nylon 6 to the total weight, respectively.) The blend solutions were cast into petri dishes maintained at room temperature to remove solvents. The semidried blend films removed from the support had been dried in a vacuum desiccator for more than 48 h to remove solvents perfectly. Fully cured blend films were obtained by the method of hot-press fabrication of the B-staged films under the predefined condition.

Morphological Analysis by Transmission Electron Microscopy (TEM) of Blend Films

Morphological analysis of the blend films was carried out by TEM analysis at 100-kV voltage. Ultrathin film samples were prepared and dyed by tungstophosphoric acid.

Measurement of Properties of Blend Films

Thermal properties of the blend films were measured by differential scanning calorimetry, thermal mechanical analysis, and thermogravimetry. Their mechanical properties were measured according to ASTM D-882. Their electrical properties were measured according to ASTM D-150 and IPC-S-100.

RESULTS AND DISCUSSION

Thermal Analysis of B-Staged Polymer Blend Films

To analyze the thermal properties of B-staged polymer blend films consisting of three different fractions, differential scanning calorimetry was carried out. Figure 3 shows the thermal behavior of the blend films. The thermal behavior of nylon 6 and B-staged advanced COPNA resin are also given in Figure 3 for comparison. In this figure, characteristic phenomena can be observed: the peak temperature of endothermic behavior shifted, and the total calories calculated from exothermic calories and endothermic calories changed. In the case of the homopolymer, typical endothermic behavior with a peak temperature at 221°C from the melt of nylon 6 was observed, and typical exothermic behavior with a peak temperature at 176°C from the dehydrating reaction of advanced COPNA resin was observed. The endothermic and exothermic calories were 19.0 and 33.9 cal/g, respectively. In the case of the poly-

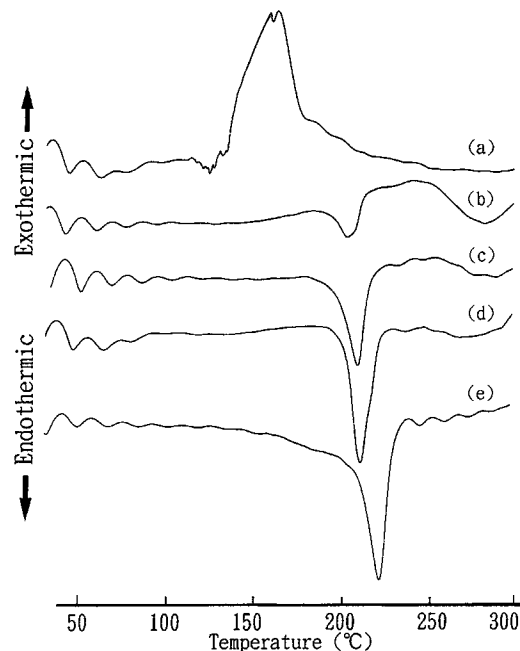


Figure 3 DSC curves of B-staged polymer blend films whose fractions are (a) 100/0, (b) 70/30, (c) 50/50, (d) 30/70, and (e) 0/100 (COPNA/nylon 6). Raising speed of temperature is 10°C/min.

mer blend system, the peak temperature from the melting behavior of nylon 6 shifted to a lower temperature range and total calories of endothermic behavior decreased, according to the increase of the advanced COPNA resin fraction. However, typical exothermic behavior from the dehydrating reaction of the advanced COPNA resin was not observed in the temperature range from 130 to 170°C. According to increase of the advanced COPNA resin fraction, slow exothermic behavior was newly observed in the temperature region from 170 to 270°C, with a peak temperature of 240°C. Table IV shows peak temperatures and total calories of the thermal behavior. Figure 4 shows dependence of total calories on the fraction. In this figure, calculated total calories were plotted for comparison when the dehydrating reaction of the advanced COPNA resin and the melt behavior of the nylon 6 were considered to be independent of each other. In this comparison between experimental values and calculated values, this proved that experimental total calories were smaller than calculated ones. The difference of the total calories might depend on an interaction at the interface between the advanced COPNA resin and the nylon 6. This phenomenon will be discussed in the next article.

Table IV Thermal Properties of B-Staged Polymer Blend Films

System ^a	Exothermic Behavior		Endothermic Behavior		Total Calorie ^b (cal/g)
	Peak Temp. (°C)	Exothermic Cal. (cal/g)	Peak Temp. (°C)	Endothermic Cal. (cal/g)	
100/0	176	33.9	—	—	33.9
70/30	230–240	15.0	203	3.4	11.6
50/50	230–240	5.7	212	6.8	-1.1
30/70	—	—	215	11.1	-11.1
0/100	—	—	221	19.0	-19.0

^a Weight percent of advanced COPNA resin/nylon 6.

^b Total calories were calculated from the equation of "exothermic calories - endothermic calories."

Morphological Analysis of B-Staged Blend Films

Nylon 6 generally exists in a partially crystallized state at room temperature, but the advanced COPNA resin exists in an amorphous state at room temperature. Generally, the compatibility between a polymer with crystallinity and a polymer without crystallinity is poor under the melting point of the crystalline polymer; therefore, the blend polymer exhibits a phase-separated state in entire fractions under the melting point. In the case of the blend polymer between nylon 6 and advanced COPNA resin, the same phenomena as observed in the phase-separated state were observed. Figures 5 and 6 show TEM images of the blend systems obtained from mixing nylon 6 and B-staged advanced COPNA resin. In the tungtrophosphoric acid dyeing system, the nylon 6 is in a black needle state and the ad-

vanced COPNA resin is in a white amorphous state. The blend systems between B-staged advanced COPNA resin and nylon 6 exhibited phase-separated structures, but their structures were very similar to each other. At the magnification of 10^4 (Fig. 5), the spherulites of the crystallized nylon 6 were observed in the matrix of amorphous advanced COPNA resin in entire systems. The diameters of the spherulites at the fractions of both 30/70 and 50/50 (COPNA/nylon 6) were approximately $10\ \mu\text{m}$, and that of the 70/30 system was $2\text{--}3\ \mu\text{m}$. Small aggregations of the advanced COPNA resin, whose size was approximately $1\ \mu\text{m}$, were also observed in all systems. At such a high magnification as 10^5 (Fig. 6), lamella structures from the spherulite state were observed for all systems. The width of nylon 6 and advanced COPNA resin were independent of fractions, and they were approximately $100\ \text{\AA}$.

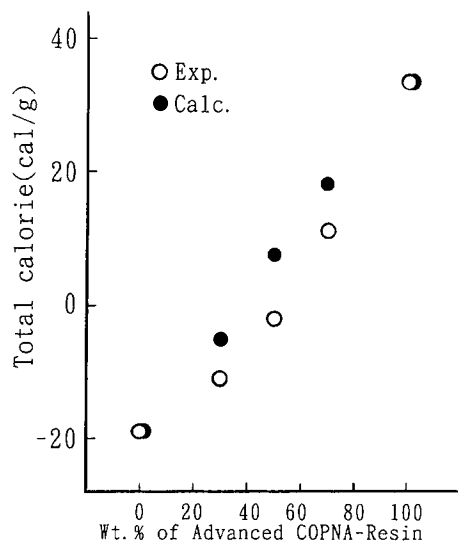


Figure 4 Dependence of total calorie on advanced COPNA resin fraction.

Hot Press Fabrication of B-Staged Polymer Blend Films

Based on the information obtained from differential scanning calorimetry of B-staged advanced COPNA resin, hot-press fabrication was carried out to obtain fully cured polymer blend films. At first, the B-staged films were cured at the temperature of 140°C for 5 min in air. After the curing process, they were hot pressed under a condition of $180^\circ\text{C}/30\ \text{min} + 230^\circ\text{C}/60\ \text{min}$ at the pressure of $25\ \text{kgf/cm}^2$ in air. The thickness of the obtained fully cured films was $50\text{--}70\ \mu\text{m}$.

Thermal Analysis of Fully Cured Blend Films

Thermal properties of the fully cured blend films were measured by the same method of differential scanning calorimetry as used for the B-staged films

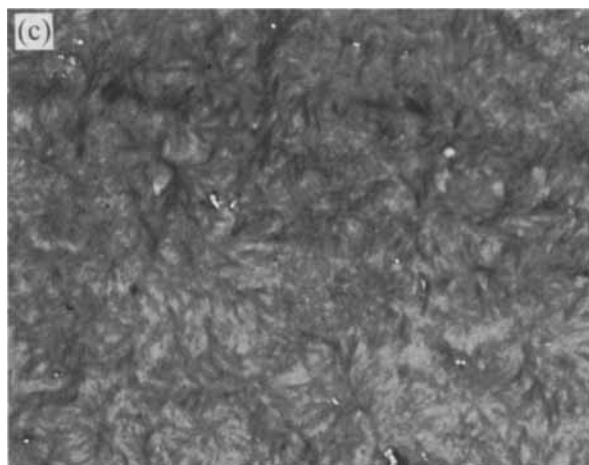
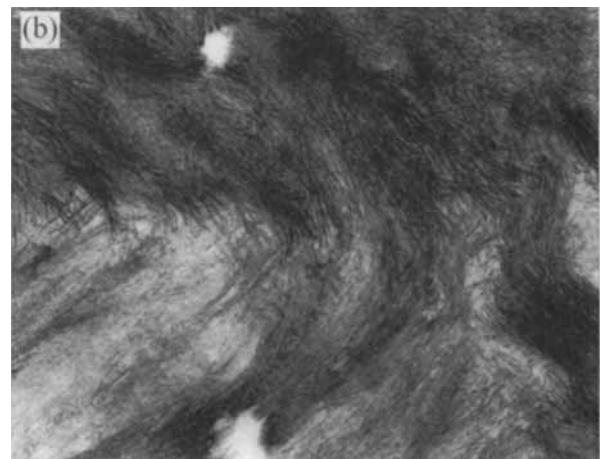
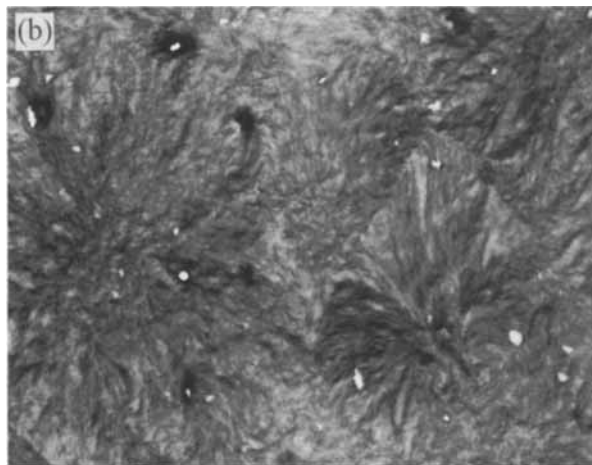
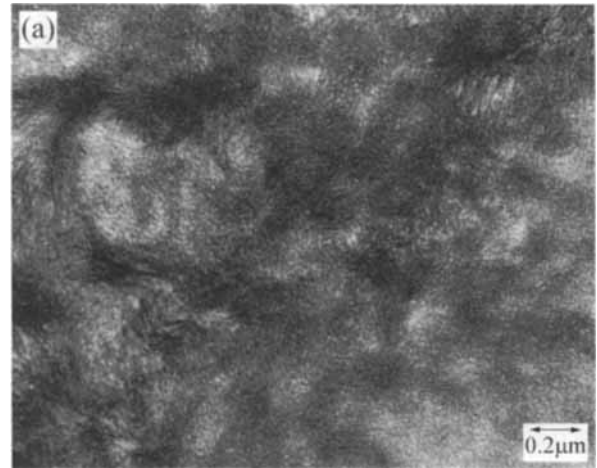
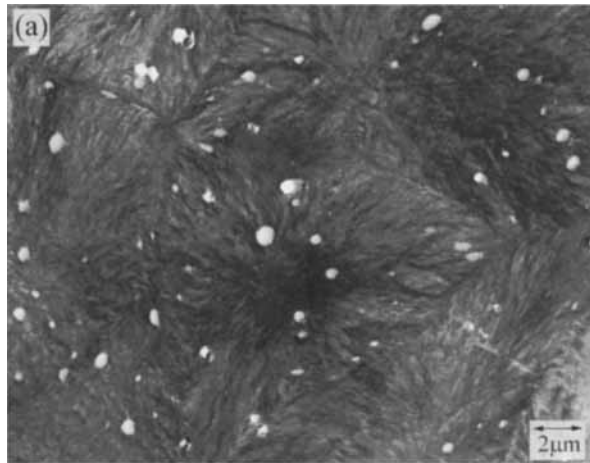


Figure 5 TEM images of B-staged polymer blend films ($\times 10^4$): (a) 30/70, (b) 50/50, and (c) 70/30 (COPNA/nylon 6).

Figure 6 TEM images of B-staged polymer blend films ($\times 10^5$): (a) 30/70, (b) 50/50, and (c) 70/30 (COPNA/nylon 6).

(Fig. 7). Characteristic behavior was not observed in all systems, apart from a weak endothermic region close to the temperature of 100°C observed in the 70/30 system.

Morphological Analysis of Blend Systems

The morphology of the fully cured blend films was observed by the same method of TEM as used for the B-staged films. The morphology was entirely different from that of the B-staged films at the same fraction; moreover, the morphology of the fully cured films whose fractions were 70/30, 50/50, and 30/70 (COPNA/nylon 6) were entirely different from each other. Figures 8 and 9 show TEM images of fully cured polymer blend films. In the 30/70 system, crystallized nylon 6 was comparatively randomly dispersed in the matrix of the advanced COPNA resin [Fig. 8(a)]. The needle crystals of nylon 6 were arranged in a definite direction within the region of approximately 5 μm . At the magnification of 4×10^4 [Fig. 9(a)], the nylon 6 crystals exhibited a network structure. In the 50/50 system, a typical phase-separated structure, called "sea-island structure," was observed [Fig. 8(b)]. The aggregated advanced COPNA resin, with diameter 1–3 μm , was observed as islands in the sea of needle crystals of the nylon 6. The needle crystals of nylon 6 were arranged in a definite direction within the same region of 5 μm as that of the 30/70 system. In the 70/30 system, the morphology exhibited a definitely phase-separated and incompatible structure [Fig. 8(c)]. Aggregated nylon 6 crystals were observed in the matrix of the advanced COPNA resin, and they were also observed to be a partially formed lamella

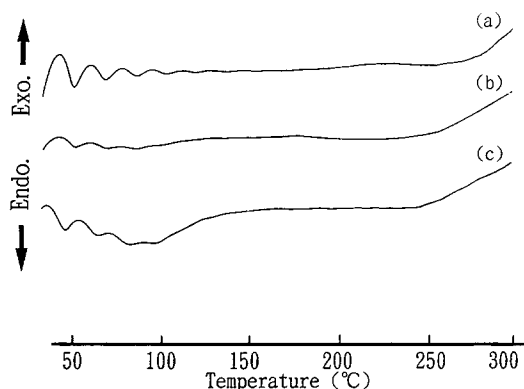


Figure 7 DSC curves of fully cured polymer blend films whose fractions are (a) 30/70, (b) 50/50, and (c) 70/30 (COPNA/nylon 6). Raising speed of temperature is 10°C/min.

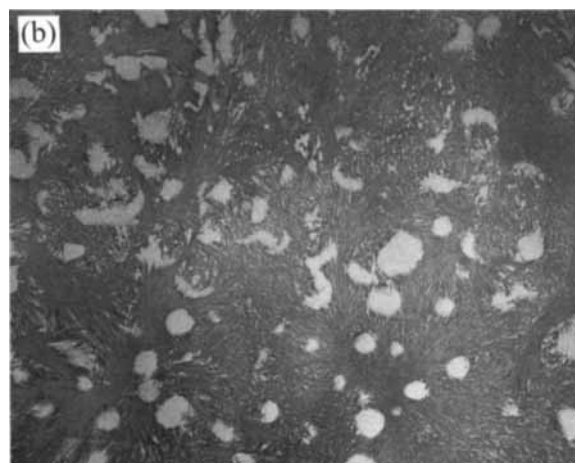
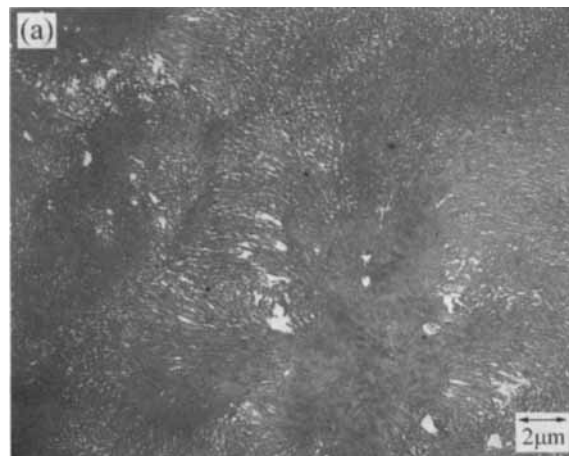


Figure 8 TEM images of fully cured polymer blend films ($\times 10^4$) (a) 30/70, (b) 50/50, and (c) 70/30 (COPNA/nylon 6).

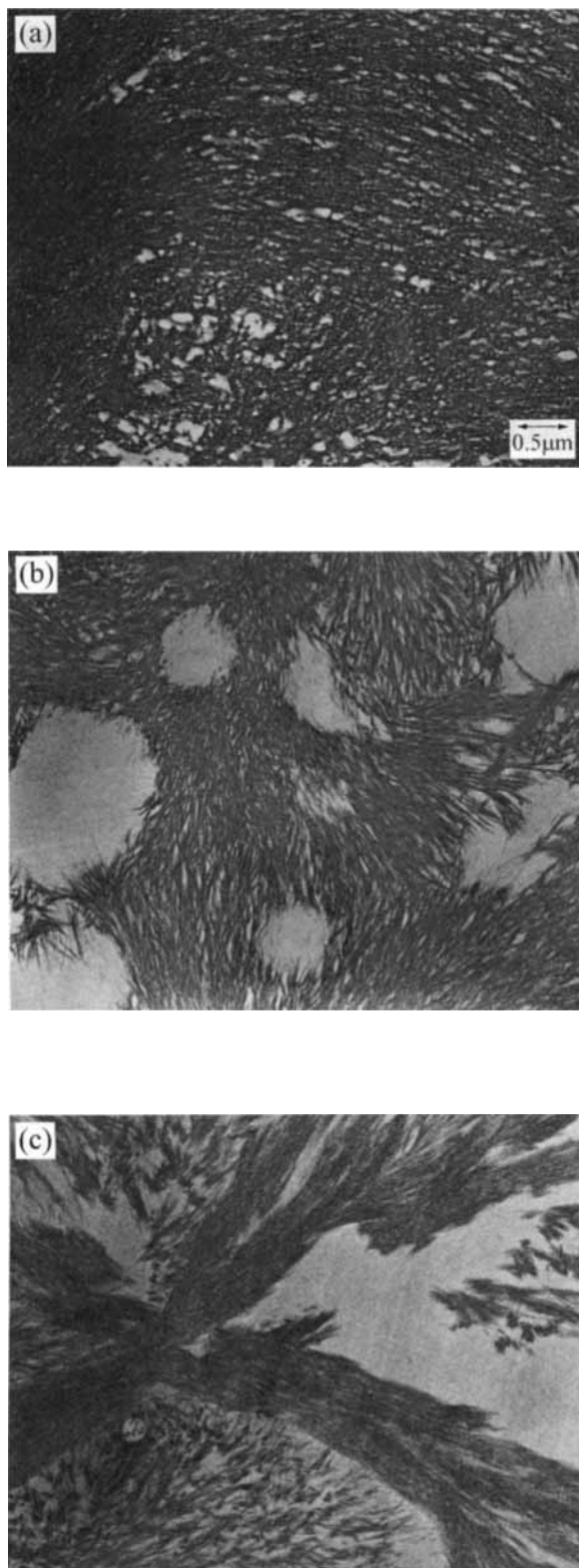


Figure 9 TEM images of fully cured polymer blend films (4×10^4) (a) 30/70, (b) 50/50, and (c) 70/30 (COPNA/nylon 6).

structure where the width of the nylon 6 was approximately 100 \AA at the magnification of 4×10^4 [Fig. 9(c)]. The results obtained from TEM observation (an entirely different structure from the B-staged state) were due to the following reasons: when advanced COPNA resin had such a small molecular weight of approximately 250 as M_n in the B stage, the resin exhibited better compatibility with nylon 6. According to the increase of the molecular weight of the advanced COPNA resin by a dehydrating reaction, the compatibility of the resin with nylon 6 decreased, and the typical phase-separated structure shown in Figures 8 and 9 appeared. Furthermore, the morphological analysis also proved that the compatibility decreased in accordance with the increase of advanced COPNA resin fraction.

Evaluation of Flexibility of Polymer Blend Films

Flexibility of the polymer blend films prepared by the method described in the Experimental section was evaluated by a folding test up to 180° . The results are shown in Table V. The systems whose fraction were 30/70 and 50/50 (COPNA/nylon 6) did not exhibit any cracks after the folding tests were repeated for 100 cycles; therefore, the systems exhibited sufficient flexibility to be used for the flexible printed wiring boards. It was proved that the initial plan to give flexibility to advanced COPNA resin succeeded. The obtained result was that the 30/70 and 50/50 systems exhibited flexibility, and the 70/30 system exhibited brittleness. This is supported by the morphological analysis of TEM images shown in Figures 8 and 9. Both the 30/70 system and the 50/50 system exhibited compatibility in a comparatively wide region of $5 \mu\text{m}$, while the 70/30 system exhibited poor compatibility in its structure.

Properties of Obtained Blend Films

The properties of the polymer blend films are shown in Table VI. All systems exhibited glass transition temperatures in the temperature region from 153 to

Table V Flexibility of Blends

	System (COPNA/Nylon 6)		
	30/70	50/50	70/30
Flexibility ^a	No cracks	No cracks	Cracks

^a Evaluated by folding tests up to 180° repeated for 100 cycles.

Table VI Properties of Fully Cured Polymer Blend Films

	System (COPNA/Nylon 6)		
	30/70	50/50	70/30
Film thickness (μm)	65	60	57
Glass transition temperature ($^{\circ}\text{C}$)	153	158	168
Thermal expansion coefficient (ppm)	148	98	69
10% weight loss temperature ($^{\circ}\text{C}$)	378	360	380
Tensile strength (kgf/mm^2)	3.1	3.2	2.8
Young's modulus (kgf/mm^2)	74	79	83
Elongation (%)	23	23	29
Dielectric constant (at 1 MHz)	3.7	3.6	3.5
Dissipation factor (at 1 MHz)	0.01	0.01	0.01
Volume resistivity ($\Omega\text{ cm}$)	5×10^{15}	3×10^{15}	5×10^{15}
Water absorption (wt %)	3.3	1.4	0.71

168°C. Those values are higher by 20–30° than that of FR-4 graded epoxy resin, which is generally used as a composite material for a rigid printed wiring board. The material with glass transition temperature of 153–168°C exhibited sufficient reliability to be used in a soldering process. The blend systems exhibited a linear thermal expansion coefficient of 69–148 ppm. The coefficients depended on the fraction of the advanced COPNA resin, and decreased according to the increase of the advanced COPNA resin fraction. The linear thermal expansion coefficients of all systems were larger than those of the polyimide film and the polyester film, but the coefficient of the 50/50 (COPNA/nylon 6) system was slightly larger than that of the epoxy resin, and that of the 30/70 system was twice as large as that of the epoxy resin. The blend systems exhibited a dielectric constant, which directly affected transmission properties, in the region of 3.5–3.7 for 1-MHz frequency. Those values are attractive for use in high-density electronic packages. The dielectric constant became smaller when the fraction of the advanced COPNA resin was increased, because the dielectric constant of the advanced COPNA resin was smaller than that of nylon 6. The systems exhibited a dissipation factor at 0.01, which was approximately the same as the polyester film. The polymer blend films exhibited a sufficient volume resistivity of more than $10^{15}\ \Omega\text{ cm}$ for use in flexible printed wiring boards in entire systems. In the case of water absorption, the 30/70 (COPNA/nylon 6) system exhibited greater value than that of the polyimide film, but it decreased according to the increase of advanced COPNA resin fraction. The 70/30 system exhibited 0.71 wt %. The price of the advanced COPNA resin

and nylon 6 has been estimated to be much lower than that of polyimide resin; therefore, the price of the polymer blend systems consisting of the advanced COPNA resin and nylon 6 would be much lower than that of the polyimide resin.

From the discussion in this section and the previous section, it has been proved that the 50/50 system was the best choice and the 30/70 system was the second-best choice as the material for flexible films in flexible printed wiring boards.

CONCLUSION

To study the application of the advanced COPNA resin as a flexible film material for a flexible printed wiring board, polymer blend technology was introduced to give flexibility to the resin. The blend system consisting of advanced COPNA resin and nylon 6 exhibited sufficient flexibility at the fraction of 30/70 and 50/50 (COPNA/nylon 6). Those two systems exhibited glass transition temperatures of 153–158°C and a dielectric constant of 3.6 to 3.7. These values are attractive in the flexible films for high-density electronic packages. From the morphological analysis, the fully cured polymer blend systems exhibited phase-separated structure, and the degree of phase separation increased according to the increase of the advanced COPNA resin fraction.

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