

# Development of an Anisotropic Conductive Adhesive Film (ACAF) from Epoxy Resins

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## SYNOPSIS

A thermoset type anisotropic conductive adhesive film (ACAF), which electrically connects an ITO glass and a flex circuit both having conducting patterns of less than 100  $\mu\text{m}$  in pitch, has been developed. In this development, bisphenol A and bisphenol F epoxy resins were tested to formulate an ACAF with easy-to-handle tackiness, flexibility, and strength. A curing agent that gives fast cure and long shelf-life properties to the epoxy resin was also selected. The tensile stress-strain responses of obtained adhesive films were successfully used for determining the formulation. Good aftercure physical properties (electrical and mechanical) were obtained. Durability against high temperature and high humidity were also tested to confirm long-term stability of the conduction of this ACAF. © 1995 John Wiley & Sons, Inc.

## INTRODUCTION

The requirement for connecting many closely spaced electrodes at a time is increasing with the development of recent fine-pitch digital circuitries and matrix drive displays. Along with the massive production of LCDs, anisotropic conductive adhesive films (ACAFs) are being widely used by flat-panel display manufacturers to electrically connect and mechanically bond flex circuits to glass displays.

In earlier times, the ACAF was made from a thermoplastic elastomer, as the main component of the adhesive, and carbon fibers as the conductive filler.<sup>1</sup> The conductive filler has been replaced first by solder balls and then Ni balls.<sup>2</sup> This type of ACAF is still in use in various applications. The thermoplastic resin adhesive is easy to repair, but has poor stability at high temperatures. The adhesion is not strong enough to hold the conductive particles in position, and the contact resistance increases after thermal shocks. Moreover, a phenomenon called "spring back" increases the contact resistance while the adhesive layer recovers from the stress caused by the

pressing of the ACAF onto the electrode during bonding. This phenomenon, originating from the creep of thermoplastic elastomer, occurs more than a few weeks after the film has been heated to connect circuits. In the spring-back phenomenon, the contact resistance sometimes increases to more than three times the original resistance. To overcome the disadvantage of the thermoplastic elastomer type ACAFs, thermoset type epoxy ACAFs have been developed.<sup>2-4</sup>

The thermoset epoxy adhesive is stable at high temperature and, more importantly, gives low contact resistance. The compressive force on the conductive particles is locked in by the epoxy adhesive after cure and, more importantly, the additional shrink force caused by the curing reaction of epoxy accomplishes the low contact resistance with long-time stability.

The reduction of contact resistance was further improved by the replacement of Ni balls with Au-plated Ni balls, and these balls have been replaced with Au/Ni-plated polystyrene (PS) beads cross-linked with divinyl-benzene.<sup>5,6</sup> These tiny metal-plated PS beads deform when heated/pressed above 100°C to give wider contact area between electroterminals. A further trial to obtain fine pitch connection has been made, although the process has not been popular until today, in which the above-

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**Table I** A Typical (Our Final) Formulation of Adhesive

Materials		Weight (%)	Remarks
Epoxy resin	Bisphenol A diglycidyl ether (epoxy equivalent 3880 g/mol)	22	Epotohto YD-020 (Tohto Kasei)
	Bisphenol F diglycidyl ether (epoxy equivalent 169 g/mol)	28	Epotohto YDF-170 (Tohto Kasei)
Phenoxy resin	$M_w$ 46,200 $T_g$ 94.0°C	22	Phenotohto YP-50S (Tohto Kasei)
Curing agent	Microcapsulated imidazole	22	Novacure HX-3941HP (Asahi Chemical Industry)
Coupling agent	Epoxy silane coupling agent	0.5	A-187 (Nippon Unicar)
Conducting filler	Au/Ni plated styrene beads <sup>a</sup>	5.5	Micropearl Au-206
		(3.0 vol%)	(Sekisui Fine Chemical)

<sup>a</sup> Crosslinked with divinylbenzene, plated thickness 428 Å.

mentioned metal-plated PS beads were again coated with an insulating resin; the insulating resin layer is broken only under pressure to expose the conducting layer on the PS sphere.<sup>7,8</sup>

Because the thermal expansion coefficient of metal-coated PS beads is very close to that of thermoset adhesive, the combination of epoxy resin and metal-plated PS beads results in a large improvement in thermal stability.

In the development of the thermoset type ACAF, flexibility and tack of uncured adhesive films are important for handling and setting of the films at a preset position. Tackiness is easily attained in thermoplastic type adhesive films, but not easy to achieve in thermoset type epoxy films because of the low molecular weight of the epoxy resin.

The long shelf life and the fast cure are also very important properties in thermoset type adhesive films.

Today, the ACAF technology, together with anisotropic conductive adhesive paste, is widely used in surface-mount device (SMD) assemblies.<sup>9</sup>

In this article we demonstrate the development of a thermoset type epoxy ACAF to meet the recent requirement for narrow pitch conduction of less than 100  $\mu\text{m}$ .

In this development, various kinds of epoxy resins were formulated to produce a flexible film with enough tack and strength. A curing agent that gives fast-cure and long shelf-life stability to the epoxy system was also selected.

The tensile stress-strain (S-S) responses of the adhesive films were used successfully to characterize the formulation.

The long-time stability of the electrical contacts at high temperature and high humidity was also examined.

## EXPERIMENT

### Fabrication of ACAFs

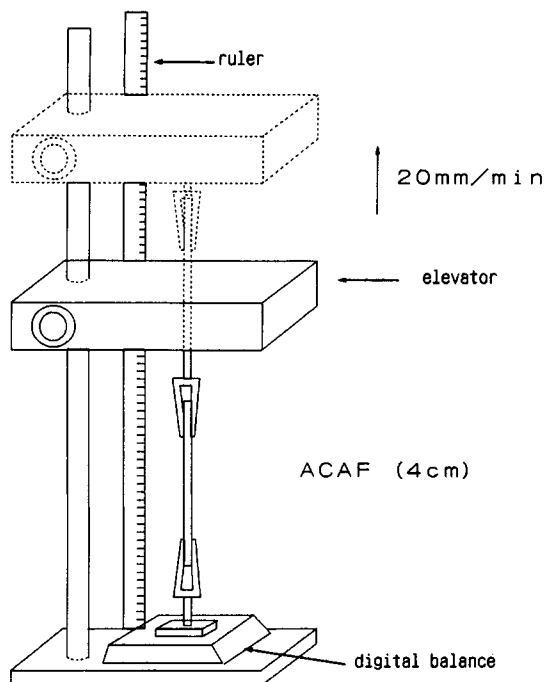
Bisphenol A, bisphenol F, phenoxy-resin and silane-coupling agent were dissolved in a toluene/MEK (3 : 1) mixture at 80°C over 2 h; the solution was cooled to room temperature and to this solution a curing agent was added. Then Au/Ni-plated styrene beads crosslinked with divinyl-benzene were added to make a final 45 wt % adhesive solution. A typical (our final) formulation is listed in Table I.

The solution was doctor bladed onto a silicone-treated polyethyleneterephthalate (PET) film 50  $\mu\text{m}$  in thickness (Toyobo E7001). Scotch tapes were layered at the both sides of the PET film to obtain a desired thickness of the solution. The PET film acts as a liner or separator for the resulting ACAF. An ACAF was obtained after solvent evaporation at 60°C over 20 min; the adhesive layer was about 25  $\mu\text{m}$  in thickness.

For large-scale production, a solvent cast-type automatic coating machine (Multi-Coater M-200, Hirano-Tecseed K.K.) was utilized. The speed of production was 1 m/min, with a 24.5 cm width PET film. The gap of the doctor blade was 67  $\mu\text{m}$  to obtain an adhesive layer of 25  $\mu\text{m}$ . For drying the film, 80°C air was blown at a speed of 15 m/s and the mean resident time of the film in dry zone was 1 min.

### Tensile S-S Response of ACAFs

An ACAF was peeled off from the liner in a prescribed width/length (3 mm in width and a length that is long enough to be held between two chucks 40 mm in span). The tensile S-S response was



**Figure 1** Apparatus used for tensile stress-strain response measurement of adhesive films. Two small paper clips were used as chucks. One was positioned on the balance and the other at the elevator. A preliminary bias weight was determined on the balance to compensate for the value of optimum stress. The weight reduces as the stress increases.

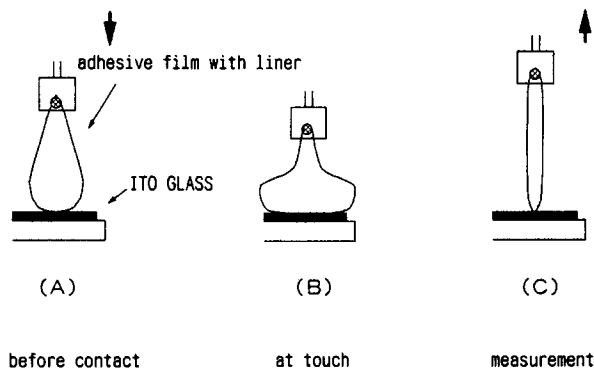
measured with a digital balance and an elevator, as shown in Figure 1. The elevator speed was 20 mm/min.

When the film was too tacky to handle, calcium-carbonate was dusted on the adhesive film.

#### Adhesion of a Flex Circuit onto an ITO Glass

A thermo-compression bonder equipped with a heat-pulsing thermode (TCW-115A Japan Avionics Co. Ltd), a sliding table, and a video monitor camera were used to permit precise positioning of the terminal electrode pattern with rapid heating and cooling under pressure.

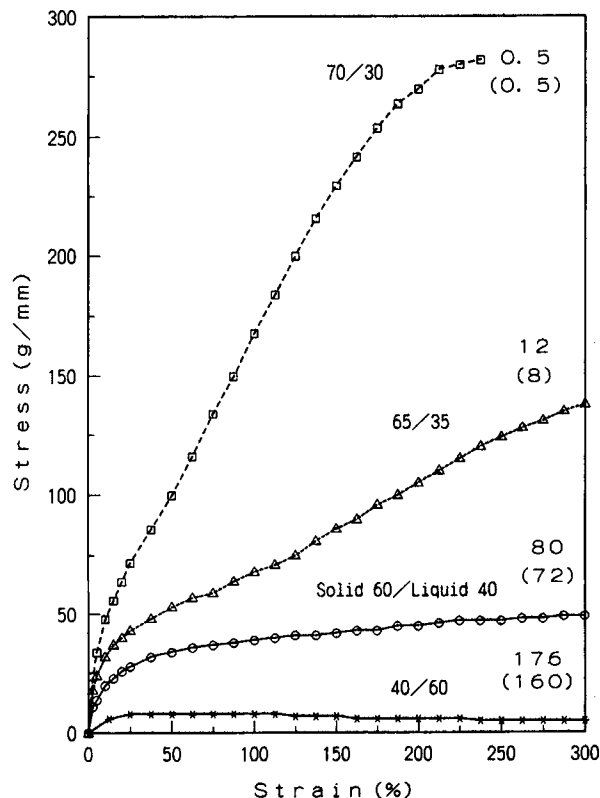
In this adhesion, an ACAF was pretacked onto the surface of an ITO glass at room temperature. The glass was 1.1 mm in thickness and the ITO patterns ( $50\ \mu \times 3\ \text{mm}$  with  $100\ \mu\text{m}$  pitch) were  $2300\ \text{\AA}$  in thickness coated on a  $1000\ \text{\AA}$   $\text{SiO}_2$  layer on the glass. The ITO glass was further heated/pressed with the ACAF at  $80^\circ\text{C}$ ,  $5\ \text{kg/cm}^2$ , for 5 s to initially set the ACAF on the ITO. After this, the liner or separator tape was peeled off from the adhesive. The ITO was then viewed through the video monitor to



**Figure 2** Process for loop tack measurement.

adjust the position of the terminal patterns to meet with the  $18\ \mu\text{m}$  thick copper patterns ( $50\ \mu\text{m} \times 3\ \text{mm}$  with  $100\ \mu\text{m}$  pitch) of polyimide flex circuit, on a table with vacuum holes to retain the preset position.

After completion of the adjustment, the table was moved under the heat-pulsing thermode again to



**Figure 3** Tensile stress-strain responses of the ACAFs obtained from the mixture of solid bisphenol A and liquid bisphenol F. The fractions represent the ratio of solid bisphenol A/liquid bisphenol F; the other numbers represent the  $90^\circ$  peel strength and the loop tack strength (in parentheses) of uncured films.

**Table II Catalysts Tested for the Epoxy Adhesives**

No.	Curing Agent	Weight (g) <sup>a</sup>	Gel Time (s)		Stability <sup>b</sup> at 150°C
			at 150°C	at 180°C	
1)	DICY <sup>1</sup>	10	—	330	—
2)	ADH <sup>2</sup>	20	700	95	—
3)	2MZ <sup>3</sup>	5	16	6	0
4)	Capsulated 2MZ <sup>4</sup>	100	14	7	14
5)	Capsulated 2MZ <sup>5</sup>	100	15	8	16
6)	DICY + 5)	12.5 + 37.5	40	15	—
7)	ADH + 5)	25 + 37.5	25	7	17
8)	Modified amine <sup>6</sup>	20	30	27	18
9)	DICY + 8)	8 + 5	110	39	—
10)	ADH + 8)	23 + 5	50	17	—
11)	DICY + Guanazine	8 + 5	48	22	—
12)	BF <sub>3</sub> Amine adduct <sup>7</sup>	3	270	65	—
13)	DDS <sup>8</sup> + 12)	29 + 1	320	140	—
14)	Sulphonium salt <sup>9</sup>	9	30	5	18
15)	Sulphonium salt <sup>10</sup>	1	24	—	30

<sup>a</sup> This amount was added to 100 g of liquid bisphenol A epoxy resin.<sup>b</sup> Gel time after storage at 40°C for 30 days.<sup>1</sup> Dicyanodiamide.<sup>2</sup> Adipic dihydrazide.<sup>3</sup> 2-Methylimidazole.<sup>4</sup> Novacure HX-3941HP dispersion in liquid bisphenol A epoxy (Asahi Chemical Industry).<sup>5</sup> Novacure HX-3748 dispersion in liquid bisphenol A epoxy (Asahi Chemical Industry).<sup>6</sup> Amicure PN-23 (Ajinomoto Co., Inc.).<sup>7</sup> Monoethylamine.<sup>8</sup> Diaminodiphenylsulphone.<sup>9</sup> Mixture with epoxy resin Opton KT-980 (Adeka Co., Ltd.).<sup>10</sup> *p*-hydroxyphenyl benzyl methyl sulphonium antimony hexafluoride (San'aid SI-L85, Sanshin Chemical Industry Co., Ltd.).

pretack the laminate in position at 80°C, 5 kg/cm<sup>2</sup> for 2 s. After confirming the positioning of each terminal electrode pattern, the laminate was finally pressed and cured at 170°C, 30 kg/cm<sup>2</sup> for 30 s. The laminate was then cooled to release from the table below 100°C.

### Loop Tack

Figure 2 shows the process for loop tack measurement.

An ACAF, 3 mm in width and 175 mm in length, with its liner was looped so that it looked like the leaf of a clover and pushed on an ITO glass with 0.3 g load to make tack contact 40 mm in length. The loop tack was then measured by pulling the loop at a speed of 20 mm/min. A conventional circular loop sometimes failed in tacking onto the ITO glass by flattening its shape before the load reached 0.3 g.

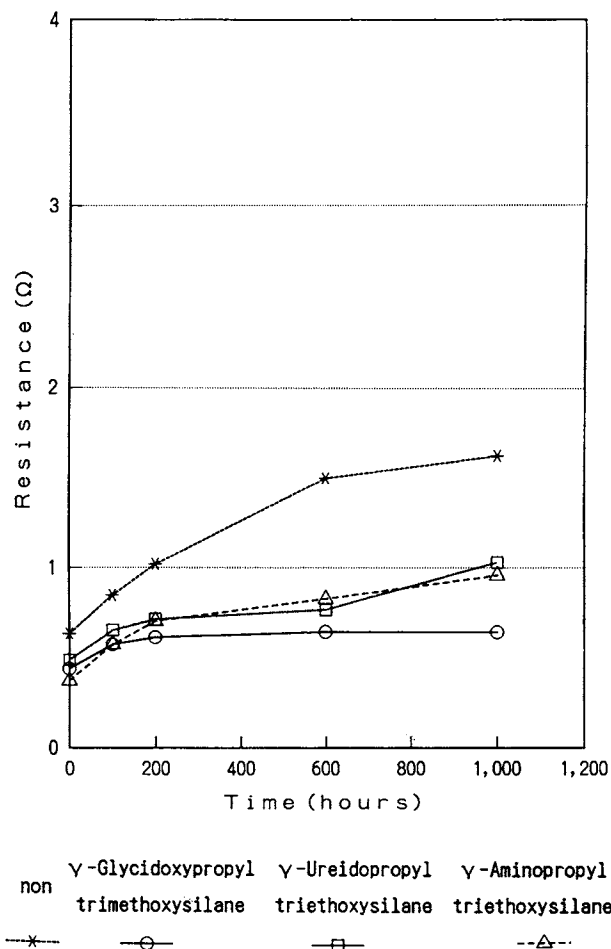
## RESULTS AND DISCUSSION

### Formulation to Give Tackiness to Epoxy Adhesives

In the application of an ACAF, the film is first placed between two conductors and pressed at room temperature or a little higher, to initially hold them in place. If the position is wrong, the conductors can be easily repositioned by removing the preliminary adhesion. Therefore, the film must have tackiness to hold the two conductors and at the same time be easily peeled off.

The tensile S-S responses of adhesives were measured to evaluate the various formulations of adhesives and to examine commercially available ACAFs. This "tensile S-S method" has been applied in our patent publications to examine pressure-sensitive adhesives, and the method is useful to obtain information from the adhesives in addition to information obtained by the other methods.<sup>10,11</sup>

The method is very convenient to analyze pressure-sensitive adhesives as well as to examine the



**Figure 4** Contact resistances of the films with various coupling agents measured at room temperature after leaving at 100°C for up to 1,000 h. The contact resistance of the film was measured between an ITO glass and a polyimide film both having array conductors 100  $\mu\text{m}$  in pitch. The contact resistance was taken as an average value of 30 contacts; the contact area of each conductor was 50  $\mu\text{m}$   $\times$  3 mm.

tackiness of ACAFs. The tensile S-S method allows the evaluation of a pressure-sensitive adhesive film with only a tiny portion of the film or with less than 10 mg of the adhesive.

Although the method is not ideal because it is not analyzing the compressive deformation of the adhesive at the tack, it is still useful to study it within the ideal elastic deformation region. The method describes the creep of the adhesive measured at different elongating speeds. The tensile S-S response also reveals the temperature dependence of a pressure-sensitive adhesive if it is measured at different temperatures. We can estimate the low temperature at which the film loses tack, showing a yielding point on the tensile S-S response. We can also tell the

high temperature at which the film loses its holding power by losing its modulus.

The modulus at low elongation relates to the tack of the adhesive. The modulus at a certain elongation suggests the content of tackifier in a high molecular weight polymer. Thus, the tensile S-S method is useful to understand the nature of ACAFs and for predicting the resin flow/creep under a particular temperature/pressure. Therefore, the method was applied to evaluate commercially available ACAFs and to decide the formulation of our film.

Conventional tackifiers, like terpene rosin, are commonly used in thermo-plastic-type pressure-sensitive adhesives; block copolymers can be modified with this.

For thermoset resin, the selection is different. We selected many kinds of epoxies and additives; finally, we selected a mixture of solid (high molecular weight) bisphenol A and liquid (low molecular weight) bisphenol F epoxies to give pressure-sensitive tack to the adhesive. This gave a flexible adhesive-film with fairly high green strength. Figure 3 shows the tensile S-S responses of adhesive films made from solid bisphenol A/liquid bisphenol F at various mixing ratios with the values of 90° peel strength (before cure) and loop tack (in parentheses).

The mixture shows higher peel strength with increasing amount of bisphenol F in the system. Excessive bisphenol F results in the excessive low modulus of adhesive film and "dirty tack" of the film on the surface of adherent that wets the surface with adhesive when peeled off. We finally found the best ratio (44 : 56) for bisphenol A/bisphenol F mixture.

In order to increase the strength/flexibility of the film, we further added phenoxy resin to the mixture. The same effect may be obtained with butyral or another polymer that has a solubility parameter close to epoxy resins and a high molecular weight to make a certain moiety hold onto the matrix.

### Long Shelf Life and Fast Cure

The selection of curing agent was done after considering long shelf life and fast-cure properties.

Table II shows the curing agents that were tested for cure time and shelf life.

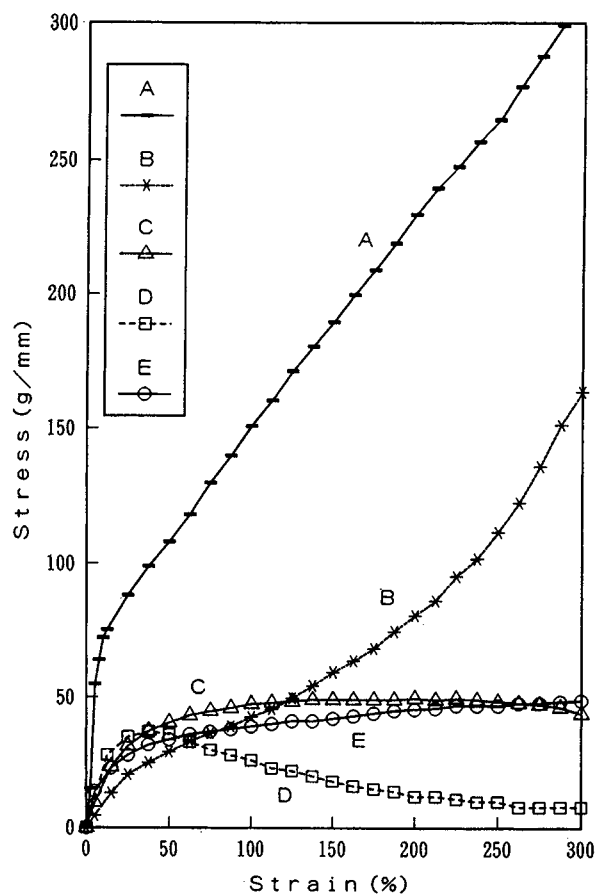
The prescribed amount of curing agent was added to each 100 g of bisphenol A, and the gel time was measured at 150 and 180°C, respectively. The mixture was further stored at 40°C for 30 days to test the shelf life.

2-Methylimidazole showed very fast gel time but the shelf life was poor. *p*-Hydroxyphenyl benzyl

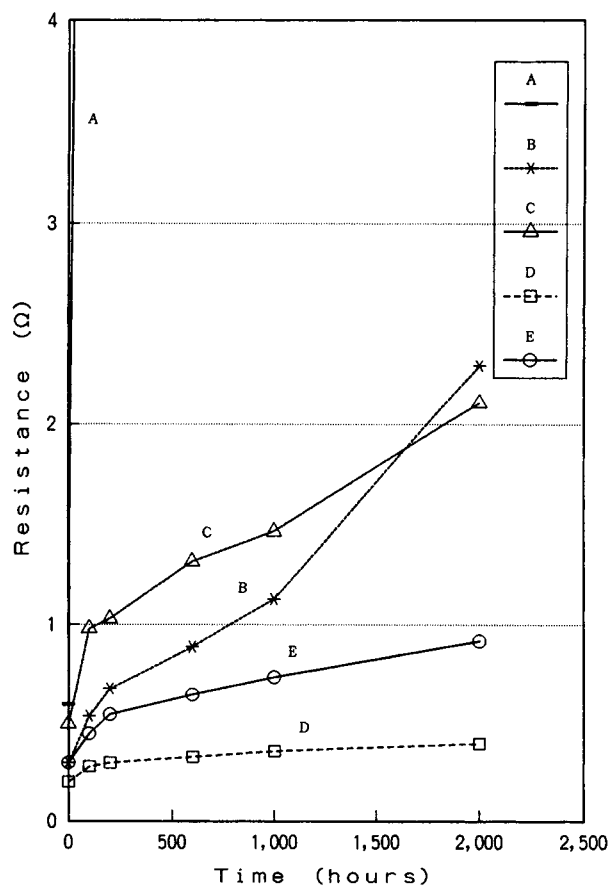
**Table III** Physical Properties of the ACAFs

Specifications	A	B	C	D	E
Geltime at 170°C (s)					
Initial	—	20	19	19	21
After storage (40°C 11 days)	—	9	14	20	19
Adhesion strength <sup>a</sup> (g/cm)					
Peel (after cure)	650	1500	900	400	1200
Peel (before cure)	60	88	52	2	96
Looptack (before cure)	0.3	64	96	2	80
Electrical property					
Contact resistance ( $\Omega$ )	0.6	0.3	0.5	0.2	0.3
Insulation resistance ( $\Omega$ )	$> 1 \times 10^{12}$	<sup>b</sup>	$> 1 \times 10^{12}$	$> 1 \times 10^{12}$	$> 1 \times 10^{12}$
Glass transition temperature <sup>c</sup> (after cure, °C)	—	85	90	103	100

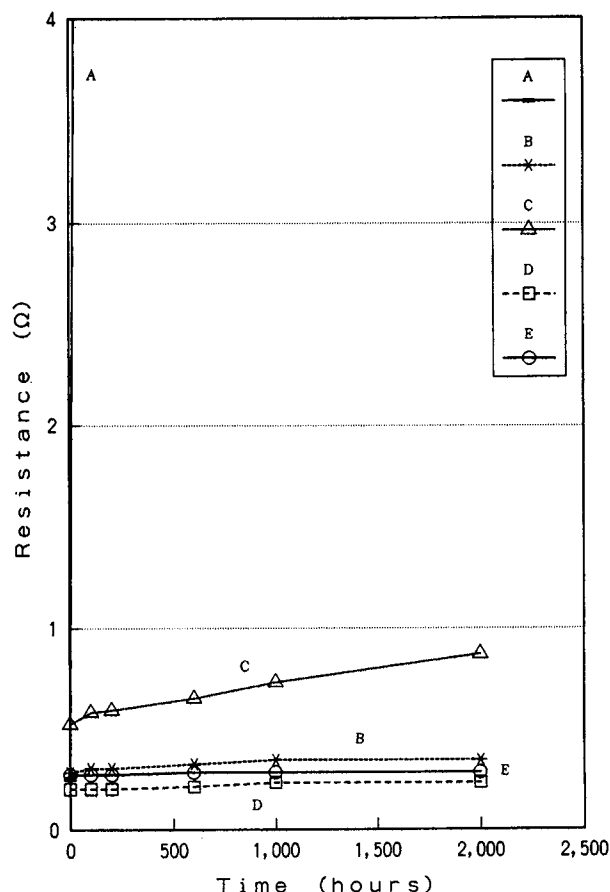
<sup>a</sup> The peel strength was obtained by the 90° peel strength measurement; a patterned polyimide film with an array conductor of 100  $\mu\text{m}$  in pitch was peeled off from an ITO glass. <sup>b</sup> 30% of short circuits with neighbor electrocodes. <sup>c</sup> The glass transition temperature of the film was measured with a differential scanning calorimeter (DSC).



**Figure 5** Tensile stress-strain responses of ACAFs: A–D are commercially available and E is our developed film. The thickness of all films is 25  $\mu\text{m}$ , as reported by the producers.



**Figure 6** Contact resistances of ACAFs at room temperature after leaving at 85°C/85% RH for up to 2,000 h.



**Figure 7** Contact resistances of ACAFs at room temperature after leaving at 100°C for up to 2,000 h.

methyl sulphonium antimony hexafluoride gave a fairly fast gel time and showed good shelf life; in addition, only a small amount was required. The problem with this curing agent is loss of the curing capability after ageing that extends the apparent gel time. The salt structure of this curing agent also discourages its use for electronic fields for fear that it would lead to terminal pattern corrosion at high humidity. Microcapsuled 2-methylimidazole showed the best performance here. In this curing agent, it is said that 2-methyl-imidazole is adducted with bisphenol-A first and modified with isocyanate to give a urethane shell. The amount of this curing agent required is almost the same as the bisphenol-A in our formulation, but the resulting film showed sufficient strength after cure.

The problem with this curing agent is the susceptibility of the microcapsules to polar solvents like alcohols or ketones. Because we've adopted the film casting process, the curing agent must be stable in the solvent. We selected a toluene/MEK (3 : 1) mixture. Pure toluene is good for protecting microcap-

sules, but it has insufficient polarity to dissolve the epoxies, giving turbidity to the solution. A hot melt process may solve the above problem.

### Silane Coupling Agent

A silane coupling agent was also added to the mixture to improve the durability of conduction. The durability of ACAFs obtained with various silane coupling agents was examined at high temperature and high humidity. Figure 4 shows the contact resistances of the films measured at room temperature after leaving at 85°C/85% RH up to 1,000 h.

Almost all the silane coupling agents improved the durability; we selected  $\gamma$ -glycidoxypropyl trimethoxysilane, which showed the best performance.

Our final formulation (Table I) provided a flexible and pressure sensitive ACAF with fast-cure and long shelf-life properties.

### Evaluation of ACAFs

The physical properties and the tensile stress-strain response of four commercially available ACAF samples, obtained independently from four different companies, and our final product were tested. The physical properties of these films are in Table III.

The reproducibility of the peel strengths of all samples was within  $\pm 15\%$ . The peel strengths after cure for all samples seem sufficient for practical use. The tensile S-S responses measured at room temperature are in Figure 5.

"A" is made from thermoplastic resin, and the resin used in the formulation was analyzed to be a thermo-plastic elastomer (styrene-*b*-ethylene-butylene-*b*-styrene block copolymer). The conductive material was Ni balls. The initial contact resistance was 0.6 ohm. The value was more than doubled after 1 week because of the spring-back phenomenon. To our surprise the modulus of the film was rather high, and there was little tackiness at room temperature. The lack of tackiness at room temperature seems to be due to the effort to obtain better temperature stability. The necessary amount of tackifier to give good tackiness leads to resulting adhesive creep. This film had enough tackiness when tacked at 80°C.

The other three are thermoset types made from epoxy base resins.

"B" contains Pb-In alloy conducting balls 10–30  $\mu\text{m}$  in diameter. The melting point of the alloy is 170–180°C. This film was designed to connect patterns of more than 200  $\mu\text{m}$  in pitch. The modulus of the adhesive is quite high at 300% elon-

gation, indicating a high volume content of high molecular weight polymer. However, the modulus at low elongation is low enough to show good tackiness at room temperature. The high modulus at elongation above 150% seems to relate unintentionally with the viscosity at curing temperature, which restricts the resin from spreading during the early stage of curing reaction. This is necessary when the diameter of the conducting ball is large, because the wider gap made by the conducting balls permits easy flow of a low viscosity adhesive. Although the curing temperature is very close to the melting point of the Pb-In alloy, the conducting balls did not melt/spread, but deformed. About 30% of terminal electrodes 100  $\mu\text{m}$  in pitch were shorted with neighboring electrodes.

"C" is designed for the connection of 140  $\mu\text{m}$  pitched patterns and contains Au/Ni plated styrene balls 10  $\mu\text{m}$  in diameter as the conductive filler.

"D" is designed to conduct 100  $\mu\text{m}$  pitched patterns and contains 5  $\mu\text{m}$  Au/Ni plated styrene beads as conductive filler. The modulus above 100% elongation was very low due to the creep of the adhesive. This suggests that the adhesive consists of only low molecular weight epoxies. The extremely low values of the peel strength (before cure) and the loop tack as well as the high modulus at low elongation suggest a lesser concern over the dirty tack of the uncured film during peel off.

Our ACAF ("E") is also aimed to conduct 100  $\mu\text{m}$  pitched patterns with 6  $\mu\text{m}$  Au/Ni-plated styrene beads as conductive filler.

The moduli at 150–200% elongation of those uncured films were well coordinated with the size of conducting balls. The reason may relate, as mentioned above, to resin flow at high temperature before they are cured. Although there are no measured viscosity values at high temperature (because there was no practical method to measure the viscosities with such small amount of adhesive samples), the moduli at this elongation seem to correspond with the viscosities at the curing temperature.

The strength of the adhesion of our ACAF after cure was enough to sustain good conduction between an ITO glass and a copper-patterned polyimide film. The gel time and the shelf life of the film were considered satisfactory.

The contact resistance of the film was as low as that of other films tested, and the insulation between conductors was also good.

The durability test of the film was performed with

the other commercially available ACAF samples. Figure 6 shows the contact resistances of the samples measured at room temperature after leaving at 85°C/85% RH up to 2,000 h.

Figure 7 shows the result of the same test after leaving at 100°C for up to 2,000 h.

Both of the durability tests did not give any significant change of contact resistance, although some of the other samples showed a considerable increase of contact resistance after leaving at 85°C/85% RH. Sample "A" from thermo-elastomer failed in both of the above tests.

## CONCLUSION

An ACAF designed for connecting terminals with patterns of less than 100  $\mu\text{m}$  in pitch was developed. Various epoxy resins and curing agents were studied to obtain a fast cure and long shelf-life epoxy adhesive film with sufficient tack. In this development, tensile S-S measurement was successfully used to determine a final formulation. An ACAF for conducting patterns of less than 100  $\mu\text{m}$  in pitch was finally obtained from a mixture of solid bisphenol A and liquid bisphenol F, and microcapsuled imidazole curing agent. The durability tests at 85°C/85% RH and at 100°C showed excellent performance of this ACAF.

The commercially available ACAFs were also tested to compare performance. The tensile S-S responses of these films were different from each other, depending on the intended pitch of the terminal electrode and the size of the conducting balls.

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