

# Low Dielectric Thermoset. I. Synthesis and Properties of Novel 2,6-Dimethyl phenol-dicyclopentadiene Epoxy

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**ABSTRACT:** A 2,6-dimethyl phenol-dicyclopentadiene novolac was synthesized from dicyclopentadiene and 2,6-dimethyl phenol, and the resultant 2,6-dimethyl phenol-dicyclopentadiene novolac was epoxidized to 2,6-dimethyl phenol-dicyclopentadiene epoxy. The structures of novolac and epoxy were confirmed by Fourier transform infrared spectroscopy (FTIR), elemental analysis, mass spectroscopy (MS), nuclear magnetic resonance spectroscopy (NMR), and epoxy equivalent weight titration. The synthesized 2,6-dimethyl phenol-dicyclopentadiene epoxy was then cured with 4,4-diaminodiphenyl methane (DDM), phenol novolac (PN), 4,4-diaminodiphenyl sulfone (DDS), and 4,4-diaminodiphenyl ether (DDE). Thermal properties of cured epoxy resins were studied by differential scanning calorimetry (DSC), dynamic mechanical analysis (DMA), dielectric anal-

ysis (DEA), and thermal gravimetric analysis (TGA). These data were compared with those of the commercial bisphenol A epoxy system. Compared with the bisphenol A epoxy system, the cured 2,6-dimethyl phenol-dicyclopentadiene epoxy resins exhibited lower dielectric constants ( $\sim 3.0$  at 1 MHz and 2.8 at 1 GHz), dissipation factors ( $\sim 0.007$  at 1 MHz and 0.004 at 1 GHz), glass transition temperatures (140–188°C), thermal stability (5% degradation temperature at 382–404°C), thermal expansion coefficients [50–60 ppm/°C before glass-transition temperature ( $T_g$ )], and moisture absorption (0.9–1.1%), but higher modulus ( $\sim 2$  Gpa at 60°C). © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 88: 2607–2613, 2003

**Key words:** dielectric properties; thermal properties; thermosets

## INTRODUCTION

As the width of integrated circuits decreases, the future encapsulant must meet the following properties: (1) high purity, to exhibit good reliability; (2) low internal stress, to reduce the internal stress; (3) high thermal stability, for better solder resistance; (4) non-halogen flame retardancy, to avoid dioxin and toxicity; and (5) low dielectric constant and dissipation factor. The signal propagating speed ( $V_p$ ) in an integrated circuit is inversely proportional to square root of dielectric constant ( $D_k$ ):

$$V_p = C / \sqrt{D_k} \quad (1)$$

where  $C$  is the light speed. Furthermore, the signal propagation loss ( $L$ ) is proportional to the square root of dielectric constant ( $\sqrt{D_k}$ ) and dissipation factor ( $D_f$ ) as shown in eq. 2:

$$L = K \times (f/C) \times D_f \times \sqrt{D_k} \quad (2)$$

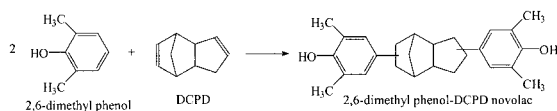
where  $K$  is a constant and  $f$  is frequency. According to the relationships described in eqs. 1 and 2, a material

with low dielectric constant and low dissipation factor will enhance the signal propagating speed and reduce the signal propagating loss.

Nelson et al.,<sup>1</sup> Hiroaki et al.,<sup>2</sup> Wang et al.,<sup>3, 5</sup> and Lin<sup>5</sup> synthesized hydrocarbon-containing epoxy resins, which were derived from the novolac of phenol and hydrocarbon in the presence of a Lewis acid, by epoxidizing the resulting hydrocarbon novolacs to epoxy resins. Among these hydrocarbons, dicyclopentadiene (DCPD) and higher oligomers of cyclopentadiene are the most commonly used starting materials because of their availability (DCPD is a byproduct of  $C_5$  streams in oil refineries), reactivity, and low cost. In addition to the epoxy, high purity (95%) DCPD can also be applied as a comonomer in ethylene propylene diene monomer (EDAM),<sup>6</sup> unsaturated polyester,<sup>7</sup> and olefin metathesis.<sup>8–10</sup>

Recently, Wang et al.<sup>11,12</sup> reported that the moisture absorption of triphenol and tetraphenol epoxy resins can be reduced by introducing hydrophobic methyl groups. In this article, tetra-methyl was introduced into the DCPD-based novolac (2,6-dimethyl phenol-DCPD novolac) by reacting DCPD and 2,6-dimethyl phenol to reduce the moisture absorption of the epoxy. Furthermore, a 2,6-dimethyl phenol-DCPD epoxy was synthesized from the resulting 2,6-dimethyl phenol-DCPD novolac with epichlorohydrin. The structures of the 2,6-dimethyl phenol-DCPD novolac and

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**Scheme 1** The reaction equation of 2,6-dimethyl phenol-DCPD novolac.

epoxy were characterized by infrared spectroscopy (IR), mass spectroscopy (MS), nuclear magnetic resonance spectroscopy (NMR), epoxy equivalent weight (EEW) titrations, and elemental analyses. The resultant epoxy resins were cured and their properties were evaluated by differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), dynamic mechanical analysis (DMA), and dielectric analyzer (DEA). These test results were compared with those for the bisphenol A epoxy system.

## EXPERIMENTAL

### Materials

Dicyclopentadiene (DCPD), imidazole, 2,6-dimethylphenol, 4,4-diaminodiphenyl methane (DDM), and epichlorohydrin were purchased from Acros. Phenol novolac (PN, PF-1120HK) with an OH equivalent weight of 105 g/eq and bisphenol A epoxy with EEW of 188 were kindly supplied by Chang Chun Petrochemical (ROC). 4,4-Diaminodiphenyl sulfone (DDS) and 4,4-diaminodiphenyl ether (DDE) were purchased from Chriskev. All solvents were commercial products (LC grade) and used without further purification.

### Characterization

DSC scans were obtained from samples of  $\sim 6$  mg in a nitrogen atmosphere at a heating rate of  $20^\circ\text{C}/\text{min}$  with a Perkin-Elmer DSC 7. TGA was performed with a Perkin-Elmer TGA 7 at a heating rate of  $20^\circ\text{C}/\text{min}$  under nitrogen or air from 60 to  $800^\circ\text{C}$ . DMA was carried out with a Perkin-Elmer DMA 7e. The storage modulus  $E'$  and  $\tan \delta$  were determined in the temperature scan mode at a programmed heating rate of  $10^\circ\text{C}/\text{min}$  from ambient to  $300^\circ\text{C}$  at a frequency of 1 Hz and an amplitude of  $6 \mu\text{m}$ . A sample 15 mm in length, 10 mm in width, and  $\sim 1.5$  mm in thickness was used. The test method was performed in the three-point bending mode with a tension ratio at 110%. The EEWs of the epoxy resins were determined by the  $\text{HClO}_4$ /potentiometric titration method.<sup>13</sup> Dielectric measurements were performed with an Agilent 4291B measurement system at  $30^\circ\text{C}$  in a nitrogen atmosphere at a flow rate of  $500 \text{ cm}^3/\text{min}$  by the two parallel plate mode. The covered range of frequencies was from 1 MHz to 1 GHz. The applied voltage was 1 V. Before testing, samples ( $1 \times 1 \text{ cm}^2$  and 0.3 cm thickness) were dried under vacuum at  $100^\circ\text{C}$  for 8 h.

Moisture absorption was tested as follows: Samples, with  $1 \times 1 \text{ cm}^2$  and 0.1 cm thickness, were dried under vacuum at  $120^\circ\text{C}$  until moisture had been expelled. After cooling to room temperature, the samples were weighed and then placed in  $100^\circ\text{C}$  water for 24 h and weighed again. The moisture absorption was calculated as percent weight gain =  $(W/W_0 - 1) \times 100$ , where  $W$  = weight of sample after placing in  $100^\circ\text{C}$  water for 24 h, and  $W_0$  = weight of sample before placing in water.

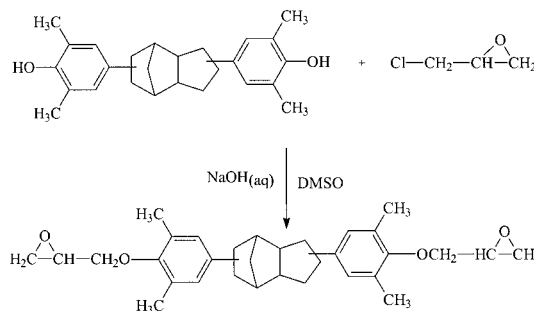
### Synthesis of 2,6-dimethyl phenol-DCPD novolac

2,6-Dimethyl phenol (0.7143 mol) and  $\text{AlCl}_3$  (0.015 mol) were added to a four-necked round-bottomed flask equipped with a nitrogen inlet, heating mantle, stirrer, thermocouple, and temperature controller. The reaction mixture was gradually heated to  $120^\circ\text{C}$ , 0.1 mol of DCPD was added gradually over a period of 2 h, and the mixture was maintained at  $120^\circ\text{C}$  for another 4 h. After the reaction was completed, 0.06 mol of 5 wt%  $\text{NaOH}_{(\text{aq})}$  was added, and the mixture was stirred for 1 h. The reaction mixture was filtered, and the filtrate was washed three times with water. Next, the organic phase was separated and distilled in a rotary evaporator to remove excess 2,6-dimethyl phenol. The crude products was dissolved in toluene and extracted with water for several times. The organic phase was distilled to remove toluene and water, and a deep-brown 2,6-dimethyl phenol-DCPD novolac was obtained in almost quantitative yield. The reaction equation is shown in Scheme 1.

ANAL. Calcd for  $\text{C}_{26}\text{H}_{32}\text{O}_2$ : C, 82.90%; H, 8.60%. Found: C, 82.59%; H, 8.63%.

### Synthesis of 2,6-dimethyl phenol-DCPD epoxy

DCPD novolac (0.1 mol), epichlorohydrin (2 mol), and dimethyl sulfoxide (DMSO, 20 wt% epichlorohydrin) were added to a four-necked round-bottomed flask equipped with a nitrogen inlet, heating mantle, stirrer, thermocouple, and temperature controller. The reaction mixture was gradually heated to  $100^\circ\text{C}$ , and then



**Scheme 2** The reaction equation of 2,6-dimethyl phenol-DCPD epoxy.

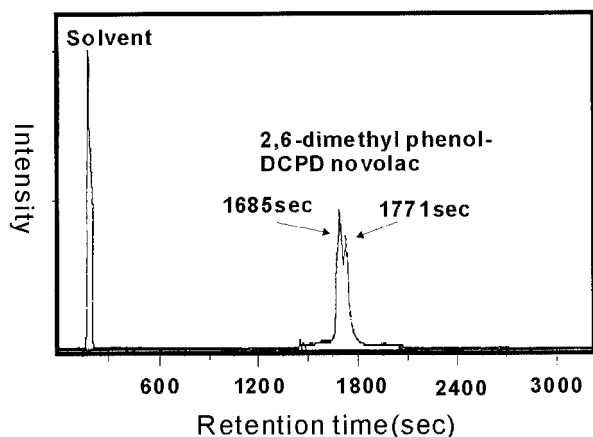


Figure 1 GCMS spectrum of the 2,6-dimethyl phenol-DCPD novolac.

0.2 mol of 20 wt%  $\text{NaOH}_{(\text{aq})}$  was added gradually and the mixture was maintained at  $100^\circ\text{C}$  for 2 h. The organic phase was separated. To the organic phase was added an additional 0.1 mol of 20 wt%  $\text{NaOH}_{(\text{aq})}$ , and the mixture was stirred for another 0.5 h. The

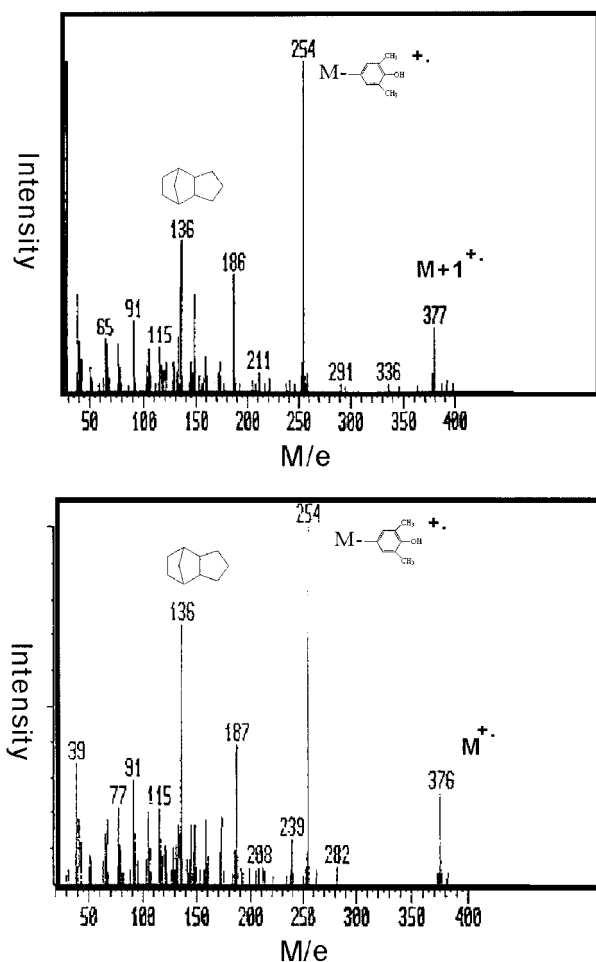
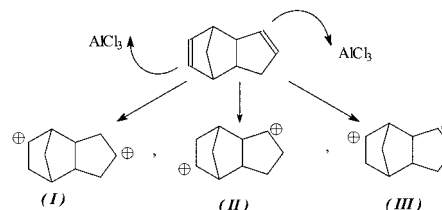


Figure 2 Mass spectra of 2,6-dimethyl phenol-DCPD novolac for retention times of (a) 1685 s and (b) 1771 s.



Scheme 3 Three possible carbocations after DCPD reacted with  $\text{AlCl}_3$  [(I), (II), and (III)].

reaction solution was filtered, and the organic phase was washed three times with water. The organic phase was separated, and the excess epichlorohydrin and DMSO were distilled in a rotary evaporator. The crude product was dissolved in a methyl ethyl ketone/methyl isobutyl ketone (1/1) mixture and washed with three times with water. The organic phase was separated and distilled to remove solvents. A brown 2,6-dimethyl phenol-DCPD epoxy was obtained in almost quantitative yield (based on novolac). The reaction equation is shown in Scheme 2.

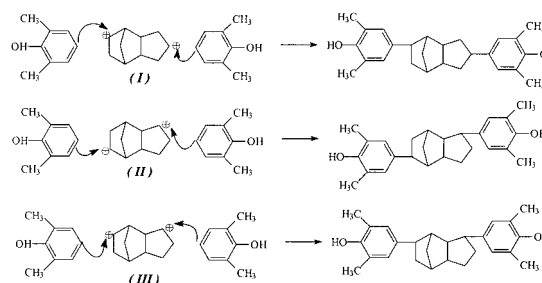
### Curing procedure

DCPD epoxy was cured with DDM, DDE, DDS, and PN. The reactant compositions were mixed in a 1:1 equivalent ratio. The mixture was crushed into fine powder and then heated on a hot plate at  $\sim 150^\circ\text{C}$  with continuous stirring until a homogeneous solution was obtained. DDM, DDE, and DDS curing systems did not require a curing accelerator but, for the PN curing system, 0.2 wt% imidazole was added as an accelerator and stirred by hand until a homogeneous solution was obtained. The mixtures were cured at  $160^\circ\text{C}$  for 1 h,  $180^\circ\text{C}$  for 2 h, and  $200^\circ\text{C}$  for 2 r. Next, samples were allowed to cool slowly to room temperature to prevent cracking.

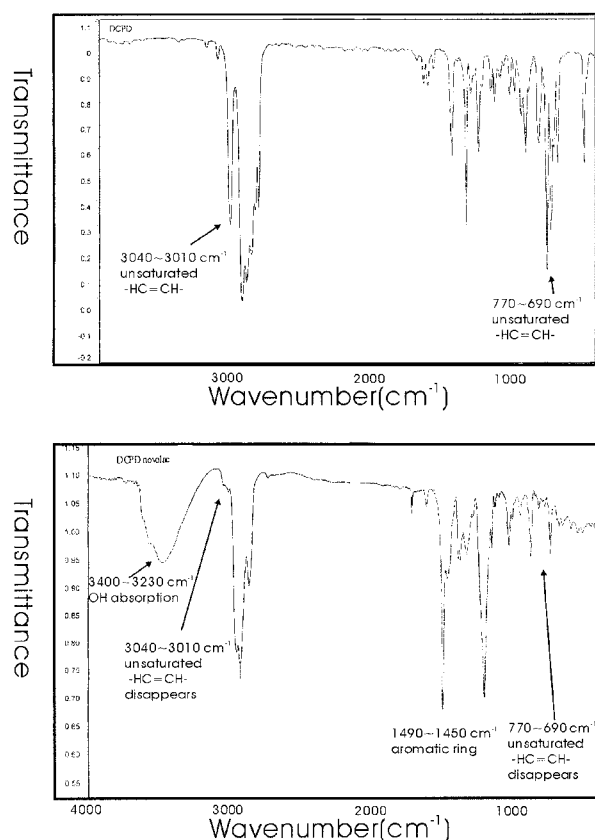
## RESULT AND DISCUSSION

### Characterization of 2,6-dimethyl phenol-DCPD novolac

As shown in Figure 1, when the 2,6-dimethyl phenol-DCPD novolac was analyzed by gas chromatography–



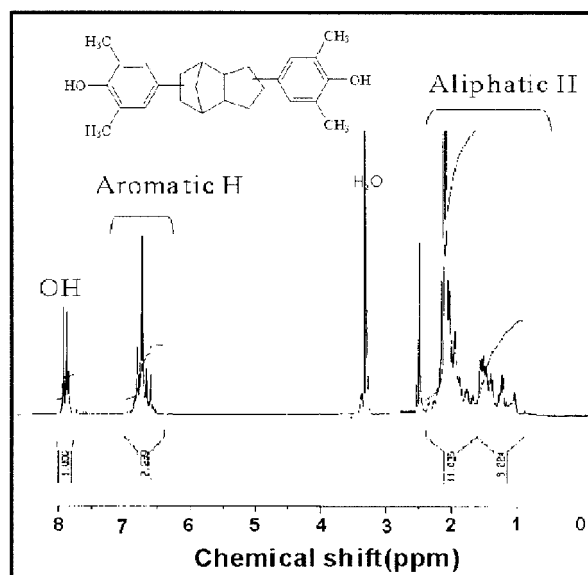
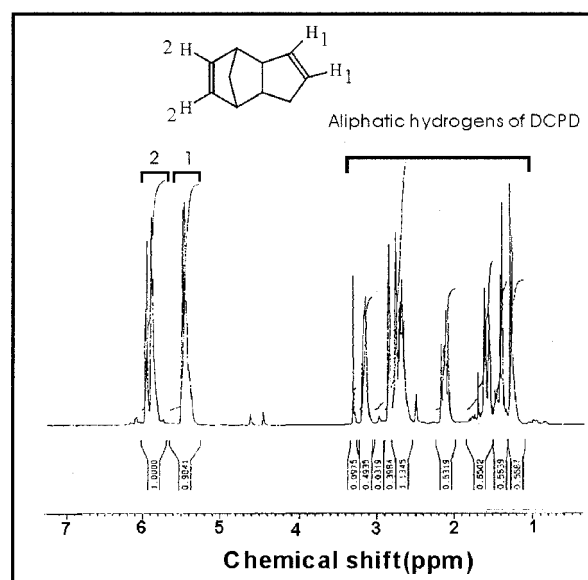
Scheme 4 The reaction of three possible carbocations [(I), (II), and (III)] with 2,6-dimethyl phenol.



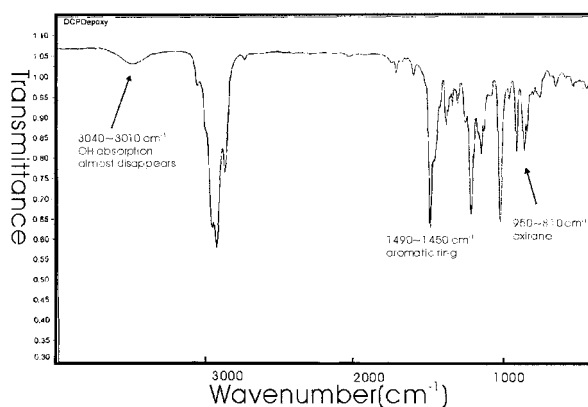
**Figure 3** IR spectra of (a) DCPD and (b) 2,6-dimethyl phenol-DCPD novolac.

mass spectroscopy (GCMS), two peaks at 1685 and 1771 s were observed. The mass spectra of the peaks at 1685 and 1771 s are shown in Figures 2a and 2b, respectively. The spectra exhibit similar patterns and have the same molecular weight ( $m/z$ : 377). This result implies there are at least two constitutional isomers in the 2,6-dimethyl phenol-DCPD novolac. The reaction mechanism may be a two-step process. In the first step, DCPD reacted with  $\text{AlCl}_3$  to form carbocations with three possible structures (Scheme 3). The three possible carbocations (I), (II), and (III), then react with 2,6-dimethyl phenol in the second step (Scheme 4). Which reaction occurs more easily and which product is predominant are difficult to determine and will not be discussed in this article.

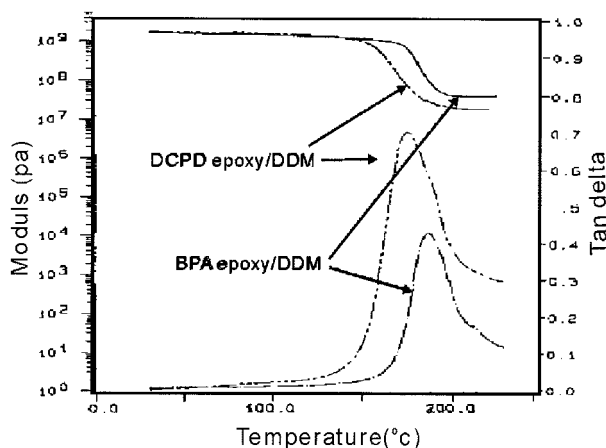
The IR spectra of DCPD and 2,6-dimethyl phenol DCPD novolac are shown in Figures 3a and 3b, respectively. The characteristic absorption peaks of  $-\text{HC}=\text{CH}-$  at 770–690 and 3040–3010  $\text{cm}^{-1}$  are evident in Figure 3a. However, these characteristic absorption peaks disappear in Figure 3b, implying all  $-\text{HC}=\text{CH}-$  functional groups had reacted with 2,6-dimethyl phenol. The characteristic absorption peaks of the aromatic ring at 1490–1450  $\text{cm}^{-1}$  and the OH absorption peaks of novolac at 3400–3230  $\text{cm}^{-1}$  are observed in Figure 3b.



**Figure 4** NMR spectra of (a) DCPD and (b) 2,6-dimethyl phenol-DCPD novolac.



**Figure 5** IR spectrum of 2,6-dimethyl phenol-DCPD epoxy.



**Figure 6** DMA scans of bisphenol A epoxy and 2,6-dimethyl phenol-DCPD epoxy cured with DDM.

The NMR spectra of DCPD and 2,6-dimethyl phenol-DCPD novolac are shown in Figures 4a and 4b, respectively. The signals of aliphatic hydrogens in DCPD in the range 1.2–3.2 ppm are shown in Figure 4a. The signals at 5.4 and 5.9 ppm are from  $\text{—HC=CH—}$  groups. However, the signals at 5.4 and 5.9 ppm disappear in Figure 4b, implying that all  $\text{—HC=CH—}$  functional groups had reacted with 2,6-dimethyl phenol. The characteristic peaks of methyl group at 0.7 ppm, aromatic ring at  $\sim 7.3$  ppm, and the OH absorption peaks at 7.76 ppm are all observed in Figure 4b.

#### Characterization of 2,6-dimethyl phenol-DCPD epoxy

The IR spectrum of 2,6-dimethyl phenol-DCPD epoxy is shown in Figure 5. The absorption at  $950\text{--}810\text{ cm}^{-1}$  is the characteristic absorption of oxirane. The characteristic absorption peak of the aromatic ring at  $1490\text{--}1450\text{ cm}^{-1}$  remains but the OH absorption peak at  $3400\text{--}3230\text{ cm}^{-1}$  decreases because of the reaction of the OH group with epichlorohydrin. According to the results of the EEW titration, the EEW of synthesized

2,6-dimethyl phenol-DCPD epoxy is 262 g/eq, which is close to the theoretical EEW of 244 g/eq.

#### Thermal properties of cured epoxy resins

##### DMA measurement

The DMA scans of bisphenol A epoxy and 2,6-dimethyl phenol-DCPD epoxy cured with DDM are shown in Figure 6. The  $T_g$  values (peak temperature of loss tangent) of these two epoxies are 184.5 and  $176.7^\circ\text{C}$ , respectively; the heights of loss tangent are 0.43 and 0.71, respectively; and the storage moduli at  $60^\circ\text{C}$  are 2.44 and  $2.7 \times 10^9$  Pa; and at  $120^\circ\text{C}$  are 2.03 and  $2.25 \times 10^9$  Pa, respectively. The results in Figure 6 indicate that the cured 2,6-dimethyl phenol-DCPD epoxy has a lower  $T_g$  and higher tangent loss than the bisphenol A system. This result may be attributed to the nonplanar DCPD structure that has a higher free volume than that of the bisphenol A system. Similar DMA scans were observed in other systems, and the results are shown in Table I. Among these curing agents, the DDS-cured system has highest  $T_g$  due to the intermolecular interaction of the sulfone group.

##### Moisture absorption

Moisture absorption will reduce the  $T_g$  of an encapsulant and may cause “popcorn phenomena” when the encapsulant is in contact with solder at high temperature. Moisture absorption will also ionize the ionic impurities (e.g.,  $\text{Cl}^-$ ) and thus corrode the integrated circuits. Furthermore, moisture absorption will increase the dielectric constant of the encapsulant because of the high dielectric constant of water. Thus, lower moisture absorption is necessary for the encapsulant. As seen in Table II, the moisture absorption of cured DCPD epoxy is in the range 0.9–1.1%, which is much lower than that of bisphenol A epoxy (1.8–2.3%). The low moisture absorption may be attributed to the hydrophobic nature of aliphatic DCPD structure and the tetramethyl group.

**TABLE I**  
DMA Analysis of Cured DCPD and BPA Epoxy

Epoxy	Curing agent	Storage modulus (Gpa)			Tangent delta ( $T_g$ ) ( $^\circ\text{C}$ )
		$E'(60^\circ\text{C})$	$E'(120^\circ\text{C})$	$E'_{\text{avg}}$	
BPA	DDM	2.44	2.03	2.24	184.5
DCPD	DDM	2.70	2.25	2.48	176.7
BPA	DDE	1.86	1.61	1.74	183.0
DCPD	DDE	1.93	1.64	1.79	175.6
BPA	DDS	2.31	1.98	2.15	194.9
DCPD	DDS	2.26	2.02	2.14	188.3
BPA	PN	1.57	0.83	1.20	147.6
DCPD	PN	2.05	0.97	1.51	138.9

TABLE II  
Moisture Absorption and Dielectric Properties of Cured DCPD Epoxy

Epoxy	Curing agent	Moisture absorption, %	1 MHz		1 GHz	
			$D_k$	$D_f * 10^3$	$D_k$	$D_f * 10^3$
BPA	DDM	1.98	3.86	139.7	3.28	94.67
DCPD	DDM	1.05	3.06	74.96	2.88	38.26
BPA	DDE	2.19	3.88	141.5	3.29	92.33
DCPD	DDE	1.09	3.15	76.70	2.89	39.02
BPA	DDS	2.25	3.59	101.3	3.20	52.55
DCPD	DDS	1.02	3.01	72.63	2.87	31.16
BPA	PN	1.87	3.51	94.42	3.04	61.31
DCPD	PN	0.99	3.03	58.25	2.86	42.11

#### Dielectric constant and dissipation factor

The dielectric constants of cured 2,6-dimethyl phenol-DCPD epoxy resins at 1 MHz and 1 GHz are  $\sim 3.0$  and  $\sim 2.8$ , respectively, whereas those of the bisphenol A system are  $\sim 3.5$  and  $\sim 3.1$ , respectively (Table II). No matter which curing agent was used, the dielectric constant and dissipation factor of the DCPD system were lower than those of the bisphenol A system. Hougham et al.<sup>14</sup> reported that the dielectric constant could be reduced by increasing the hydrophobicity and free volume and by decreasing polarization of the molecule. The nonplanar structure and four bulky methyl groups led to more spacing between polymer molecules, resulting in less efficient chain packaging and an increase in the free volume of the polymer, bringing the dielectric constant closer to the value of air ( $\sim 1.0$ ). Furthermore, the presence of low polar aliphatic dicyclopentadiene linkages may reduce intermolecular electronic interaction and increase the hydrophobicity, thus decreasing the dielectric constant and dissipation factor.

#### TMA analysis

There are two major reasons causing failure of electronic encapsulation. One is water absorption and the other is internal stress.<sup>15</sup> Thus, to increase the reliability of encapsulation, moisture absorption and internal stress must be reduced. The internal stress may result from the difference in thermal expansion coefficient between silicon and encapsulation material.<sup>14</sup> According to the results shown in Table III, the CTE (before  $T_g$ ) of the DCPD system was in the range 50–60 ppm/ $^{\circ}$ C, which is slightly lower than that of the bisphenol A system (53–62 ppm/ $^{\circ}$ C). Although the DCPD system has a higher free volume than does the bisphenol A system, the rigidity of dicyclopentadiene may compensate for the effect of higher free volume; thus, the DCPD has a similar (even lower) CTE than does the bisphenol A system.

#### TGA analysis

TGA traces of cured epoxy provided information regarding their thermal stability and thermal degrada-

tion behavior. TGA results of cured DCPD and bisphenol A system under nitrogen are shown in Table IV. These results indicate that the 5 and 10% degradation temperatures of the DCPD system are in the ranges 380–400 and 395–420 $^{\circ}$ C, respectively. These values are slightly lower than those for the bisphenol A system. These differences may be attributed to the aliphatic structure of dicyclopentadiene and the methyl group, which reduce the thermostability. Furthermore, the lower molecular interaction, due to higher free volume of the DCPD system, may also reduce the thermostability of the DCPD system.

## CONCLUSIONS

The 2,6-dimethyl phenol-DCPD epoxy was successfully synthesized from 2,6-dimethyl phenol and dicyclopentadiene by two steps. Because of the hydrophobic effect of the aliphatic DCPD structure and the methyl groups, the cured DCPD-containing epoxy resins exhibited low moisture absorption. Because of higher free volume and low polarity of aliphatic DCPD linkages, cured DCPD-containing epoxy exhibited a low dielectric constant and dissipation factor. DMA measurements show that the cured DCPD epoxy resins have lower  $T_g$  and higher tangent loss than does the bisphenol A system, which may be attributed to the nonplanar DCPD structure that has higher free volume than that of the bisphenol A system. TGA measurements show the 5 and 10% degradation temperatures of DCPD system are in the ranges 380–400 and 395–420 $^{\circ}$ C, respectively, which are also

TABLE III  
TMA Analysis of Cured DCPD and BPA Epoxy

Epoxy	Curing agent	CTE (ppm/ $^{\circ}$ C)	
		Before $T_g$	After $T_g$
BPA epoxy	DDM	56.0	192.3
DCPD epoxy	DDM	53.4	180.3
BPA epoxy	DDE	61.6	184.1
DCPD epoxy	DDE	59.0	171.7
BPA epoxy	DDS	50.7	182.3
DCPD epoxy	DDS	50.2	171.5
BPA epoxy	PN	62.3	176.6
DCPD epoxy	PN	59.4	160.5

TABLE IV  
TGA Analysis of Cured DCPD and BPA Epoxy under N<sub>2</sub> Atmosphere

Epoxy	Curing agent	Degradation temperature (°C) (N <sub>2</sub> )		Char yield (%) (N <sub>2</sub> )	
		T <sub>d</sub> 5%	T <sub>d</sub> 10%	600 °C	700 °C
BPA	DDM	408.5	420.3	22.8	20.7
DCPD	DDM	383.6	398.4	18.7	15.0
BPA	DDE	405.5	415.7	19.2	17.8
DCPD	DDE	382.6	394.2	10.9	6.51
BPA	DDS	414.9	430.4	22.5	18.4
DCPD	DDS	390.1	414.1	18.9	13.5
BPA	PN	431.6	442.9	30.1	26.9
DCPD	PN	404.1	428.0	18.5	16.9

lower than those of the bisphenol A system because the aliphatic structure of DCPD and the methyl groups reduce the thermostability. The combination of low dielectric constant, low dissipation factor, and outstanding resistance to moisture absorption make 2,6-dimethyl phenol-DCPD epoxy an attractive candidate for electrical printed wiring board and electronic encapsulation applications.

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