

# Synthesis and Properties of Phosphorus-Containing Advanced Epoxy Resins. II

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**ABSTRACT:** Two phosphorus-containing diacids were synthesized from 9,10-dihydro-9-oxa-10-phosphaphenanthrene 10-oxide (DOPO) and either maleic acid or itaconic acid and then reacted with diglycidyl ether of bisphenol A (DGEBA) to form two series of advanced epoxy resins. Reaction conditions, such as reaction time, temperature and catalyst, are discussed in this article. After curing with 4,4'-diaminodiphenyl sulfone (DDS), thermal properties of cured epoxy resins were studied using dynamic mechanical analysis (DMA) and thermal gravimetric analysis (TGA). The flame retardancy of cured epoxy resins was evaluated using a UL-94 measurement. © 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 78: 228–235, 2000

**Key words:** DOPO; maleic acid; itaconic acid; epoxy; flame retardancy; UL-94

## INTRODUCTION

Epoxy resins possess a combination of desirable properties that have resulted in their widespread use for the fabrication of laminates. Among the epoxy resins, advanced epoxy resins containing halogen atoms are particularly useful in circuit boards or other electrical laminate applications and in encapsulation and other applications where flame-retardant properties are desired. However, halogen-containing epoxy resins generate toxic and corrosive fumes upon combustion; therefore, many efforts have centered on searching for a halogen fire retardant. It has recently been reported that organophosphorus compounds demonstrate good flame retardancy and generate less toxic gas and smoke than halogen-containing compounds.<sup>1–15</sup> Our previous article<sup>9</sup> discussed the development of a series of advanced epoxy resins with a reactive phosphorus containing diol,

DOPOBQ (Scheme 1) that have proven their thermal stability and flame retardancy. However, this type of epoxy resin exhibited low solubility in acetone, a disadvantage for the printed-circuit laminates industry. For this article, two series of acetone-soluble advanced epoxy resins were synthesized. The reaction conditions and the effect of epoxy equivalent weights (EEW) on epoxy properties are discussed in this article.

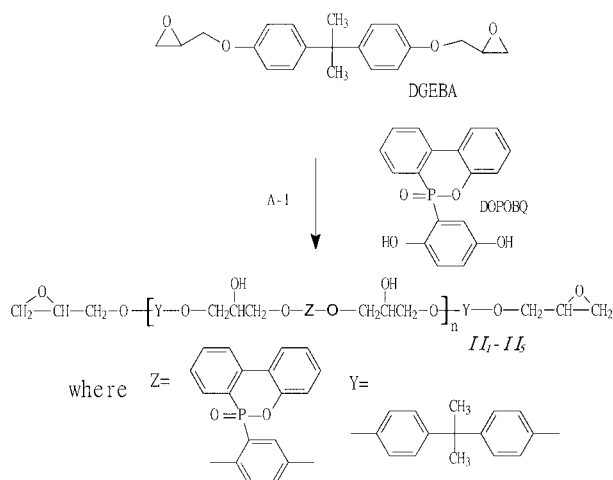
## EXPERIMENTAL

### Materials

The solvents 9,10-dihydro-9-oxa-10-phosphaphenanthrene 10-oxide (DOPO) and 2-ethyl 4-methyl imidazole (2E4MI) were purchased from TCI. Diglycidyl ether of bisphenol A (DGEBA) with an EEW of 187 g/eq was kindly supplied by CCP in the Republic of China. Purchased from Acros was 4,4'-Diaminodiphenyl sulfone (DDS). Ethyl triphenyl phosphonium acetate acetic acid complex (A-1) was bought from Morton Chemical.

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**Scheme 1** The reaction of DGEBA with DOPOBQ.

All solvents used were commercial products and used without further purification.

### Characterization

Thermogravimetric analysis (TGA) was carried out with a Perkin-Elmer TGA 7 at a heating rate of 20°C/min under nitrogen or air from 60°C to 800°C. Dynamic mechanical analyses (DMA) were made with a Perkin-Elmer DMA 7e. The storage modulus  $G'$  and  $\tan \delta$  were studied when the sample was subjected to temperature scan mode at a programmed heating rate of 10°C/min from ambient to 220°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 approximately 1.5 mm in thickness was used. The test method was performed by a three-point bending mode with a tension ratio at 110%. Epoxy equivalent weights (EEW) of advanced epoxy resins were determined by the  $\text{HClO}_4$ /potentiometric titration method. The UL-94 vertical test was performed according to the testing procedure of FMVSS 302/ZSO 3975, with a test specimen bar of 127 mm in length, 12.7 mm in width, and about 1.27 mm in thickness.

### Synthesis of Monomer

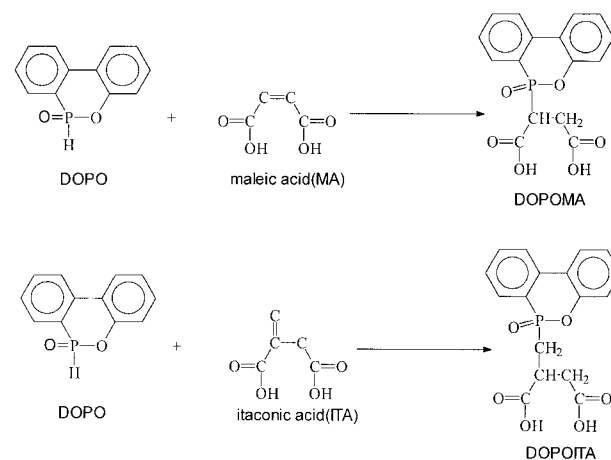
#### Synthesis of DOPOMA

Introduced into a round-bottom 1000-mL glass flask equipped with a nitrogen inlet, a condenser, and a mechanical stirrer were 216 g (1.0 mol) of 9,10-dihydro-9-oxa-10-phosphaphenanthrene 10-oxide (DOPO), 300 g of xylene, and 300 g of THF. The flask was heated to 80°C; then 116 g (1 mol)

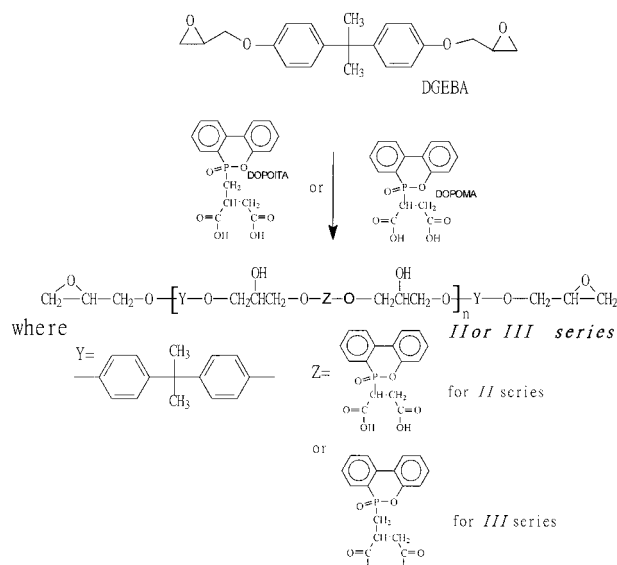
of maleic acid (MA) was added to the reactor for 2 h (Scheme 2). After the complete addition of maleic acid, the reaction mixture was maintained at that temperature for 20 h in order to ensure completion of the reaction. The reaction product was filtered after cooling to room temperature and washed with xylene/THF (1:1) several times, then dried in an oven at 120°C for 8 h. White powders (mp 221°C, DSC) of DOPOMA were obtained. Mass (FAB) ( $m/e$ , relative intensity): 332 ( $M^+$ ), 288 ( $M-\text{COOH}^+$ ), 243 ( $M-2\text{COOH}^+$ ), 215 ( $M-109^+$   $\alpha$  cleavage of  $\text{O}=\text{P}-\text{Ar}$ ). ELEM ANAL: Calcd: C, 57.84%; H, 3.94%; Found: C, 57.78%; H, 3.93%. IR: 1160–1260  $\text{cm}^{-1}$  ( $\text{P}-\text{O}-\text{Ar}$ ), 1190  $\text{cm}^{-1}$  ( $\text{P}=\text{O}$ ), 3230–3400  $\text{cm}^{-1}$  (OH), and 1735–1750  $\text{cm}^{-1}$  ( $\text{C}=\text{O}$ ). Absorption of the  $\text{P}-\text{H}$  of DOPO at 2275–2440  $\text{cm}^{-1}$  and of the  $\text{C}=\text{C}$  of maleic acid at 1600  $\text{cm}^{-1}$  disappeared completely.

### Synthesis of DOPOITA

Introduced into a round-bottom 1000-mL glass flask equipped with a nitrogen inlet, a condenser, and a mechanical stirrer were 216 g (1.0 mol) of 9,10-dihydro-9-oxa-10-phosphaphenanthrene 10-oxide (DOPO), 690 g of xylene, and 5 drops of catalyst 5%  $\text{H}_2\text{PtCl}_6$  (in isopropyl alcohol). The flask was heated to 125°C; then itaconic acid 130 g (1 mol) was added to the reactor for 2 h (Scheme 2). After the complete addition of itaconic acid, the reaction was maintained at that temperature for 20 h in order to ensure completion of the reaction. The reaction product was filtered after cooling to room temperature and was washed with acetone several times, then dried in an oven at 120°C for 8 h. White powders



**Scheme 2** The synthetic scheme of DOPOMA and DOPOITA.



**Scheme 3** The reactions of DGEBA with DOPOMA or DOPOITA.

(mp 188°C, DSC) of DOPOITA were obtained. Mass (FAB) (*m/e*, relative intensity): 346 ( $M^+$ ), 328 ( $M-H_2O^+$ ), 300 ( $M-COOH^+$ ), 255 ( $M-2COOH^+$ ), 215 ( $M-109^+$ )  $\alpha$  cleavage of  $O=P-Ar$ . ELEM ANAL: Calcd: C, 58.97%; H, 4.37%; Found: C, 58.88%; H, 4.32%. IR: 1160–1260  $cm^{-1}$  ( $P-O-Ar$ ), 1190  $cm^{-1}$  ( $P=O$ ), 3230–3400  $cm^{-1}$  (OH), and 1735–1750  $cm^{-1}$  ( $C=O$ ). Absorption of the  $P-H$  of DOPO at 2275–2440  $cm^{-1}$  and of the  $C=C$  of itaconic acid at 1600  $cm^{-1}$  disappeared completely.

### Synthesis of Advanced Epoxy

For the DGEBA–DOPOMA system, DGEBA (EEW of 187) was reacted with various moles ratio of DOPOMA at 130°C for 60 min in the presence of 0.1 wt % of A-1 catalyst (based on total weight). Thus, advanced epoxy resins with various phosphorus contents and EEWs were obtained (see Scheme 3 and Table I). A typical experimental procedure for **II**<sub>3</sub> (phosphorus content = 2 wt %) is shown below

To a four-neck round-bottom flask equipped with a heating mantle, stirrer, thermocouple and temperature controller, DGEBA 56g was added. The epoxy resin was heated to 120°C and then vigorously stirred and dehydrated for 2 h. Under a nitrogen atmosphere, DOPOMA 9.1 g of 0.1 wt % A-1 catalyst were added at 120°C. The reaction temperature gradually rose to 130°C because of its exothermic character. After the exotherm sub-

sided, the reaction mixture was then heated to 130°C and maintained at this temperature for 60 min.

For the DGEBA–DOPOITA system, the reaction condition was similar to DGEBA–DOPOMA except the reaction temperature was 160°C. Other advanced resins were synthesized in the same manner as described above, and the sample codes and EEWs are shown in Table I.

### Curing Procedure

The phosphorus-containing advanced epoxy (**II**<sub>1</sub>–**II**<sub>4</sub> or **III**<sub>1</sub>–**III**<sub>4</sub>) and the DGEBA (**II**<sub>0</sub>) were cured with DDS. The reactants were mixed in a 1:1 equivalent ratio. The mixture was heated on a hotplate at about 130–150°C with a continuous stirring until the DDS was completely dissolved in epoxy. Then the mixtures were cured at 150°C for 1 h and post-cure at 180°C for 4 h. After that, samples were allowed to cool slowly to room temperature in order to prevent cracking.

## RESULT AND DISCUSSION

### Monomer Synthesis

According to the results of our previous experiments, when DOPO reacts with a double-bond-containing compound, the structures of the double-bond-containing compounds have a significant influence on the reaction rate. For example, when DOPO reacted with benzoquinone<sup>9</sup> or maleic acid (discussed in this article), the reaction temperature was about 70–90°C. However, when DOPO reacted with itaconic acid (discussed in

**Table I** Sample Codes, Epoxy Equivalent Weight (EEW), and Phosphorus Content of Synthesized Advanced Epoxy

	Sample Code	EEW (g/eq)	P % before Curing (wt %)
DGEBA	<b>II</b> <sub>0</sub>	187	0
DGEBA/DOPOMA series	<b>II</b> <sub>1</sub>	250.8	1
	<b>II</b> <sub>2</sub>	301.4	1.5
	<b>II</b> <sub>3</sub>	356.8	2
	<b>II</b> <sub>4</sub>	458.7	2.5
DGEBA/DOPOITA series	<b>III</b> <sub>1</sub>	260.4	1
	<b>III</b> <sub>2</sub>	334.3	1.5
	<b>III</b> <sub>3</sub>	402.9	2
	<b>III</b> <sub>4</sub>	525.7	2.5

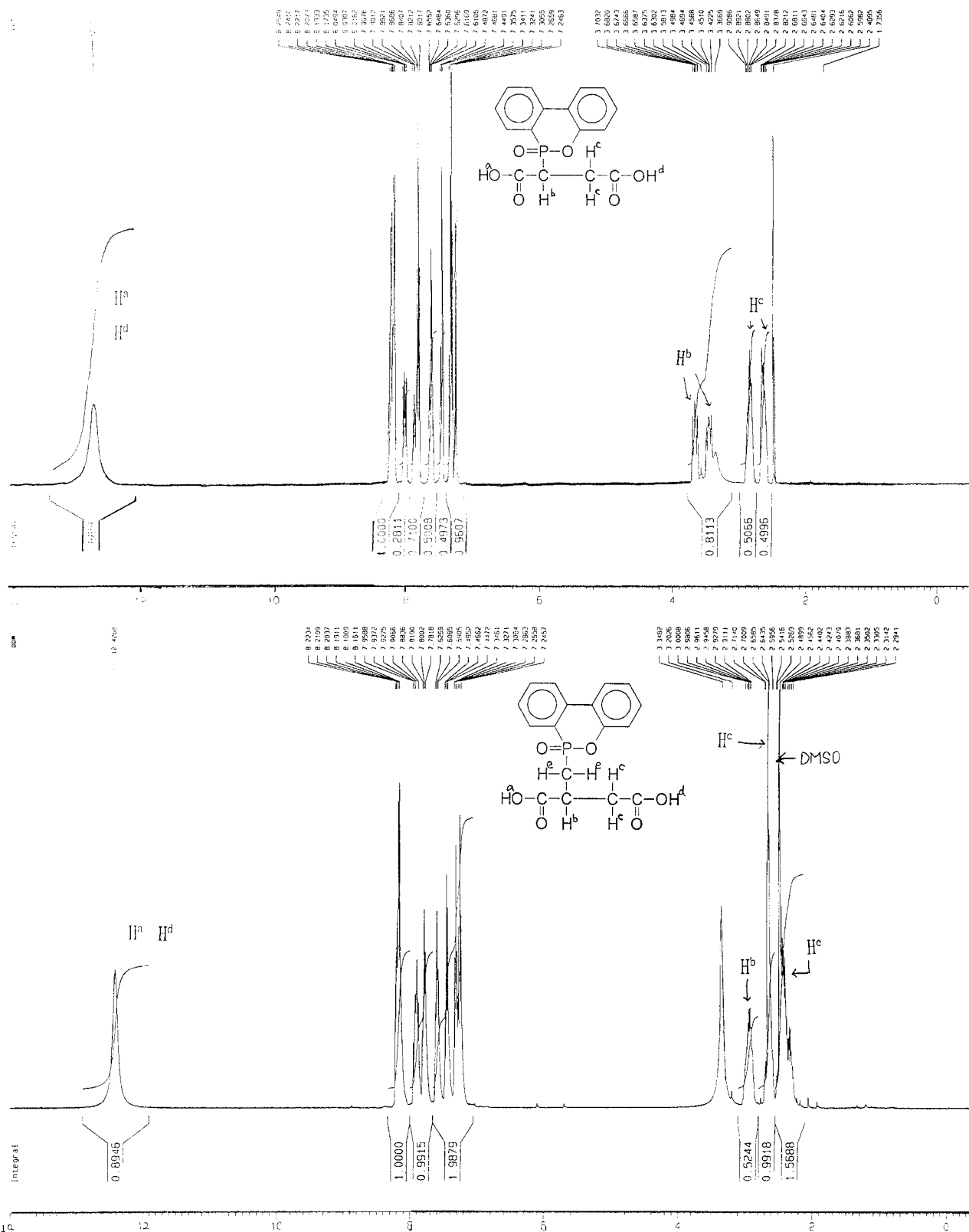
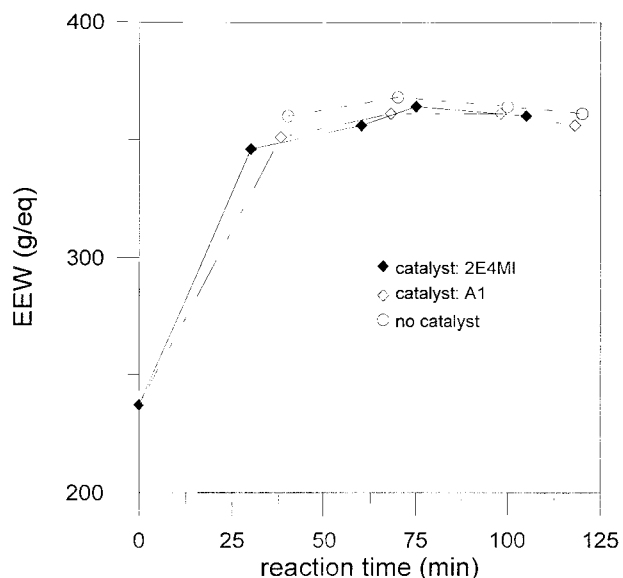


Figure 1  $^1\text{H-NMR}$  spectra of (a) DOPOMA and (b) DOPOITA.



**Figure 2** Effect of reaction time and catalyst on EEW.

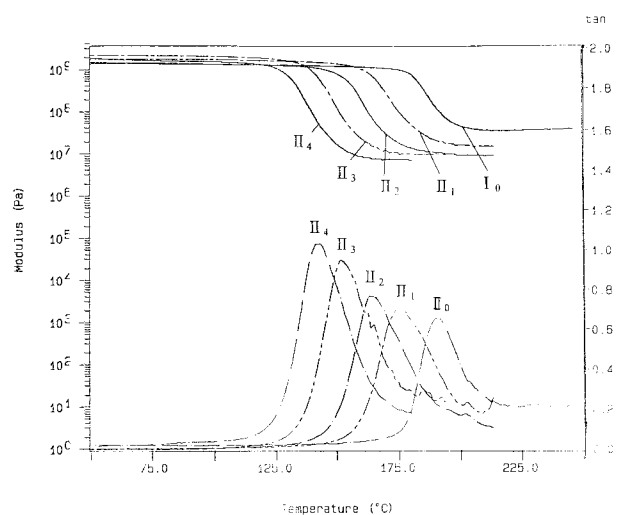
this article), the reaction was very slow, even at 140°C. Thus, the addition of the  $\text{H}_2\text{PtCl}_6$  catalyst was necessary to facilitate the reaction. The electron density of double bonds in the double-bond-containing compound is responsible for this. Benzoquinone and maleic acid, with two electron-withdrawing carbonyl groups next to the double bond could draw the electron of the double bond and consequently reduce the electron density of the double bond [negative inductive effect ( $-\text{I}$ ) of carbonyl group]. Furthermore, the electron of the double bond could resonate with two carbonyl groups [negative mesomeric effect ( $-\text{M}$ ) of carbonyl group]. In the case of itaconic acid, because one carbonyl group was not next to the double bond, the double bond could only resonate with one carbonyl group, and the inductive effect of carbonyl decreased because of the longer distance between the carbonyl group and the double bond. In this case, maleic acid, with a double bond conjugated with two carbonyl groups (the same conjugation structure as benzoquinone), has exhibited almost the same reactivity toward DOPO. Thus, DOPO-maleic acid was reacted at 80–90°C in a xylene solution (a similar reaction condition for DOPO-benzoquinone system<sup>9</sup>).

Figures 1(a) and 1(b) show the  $^1\text{H-NMR}$  spectra of DOPOMA and DOPOITA, respectively. For the DOPOMA system, chemical shifts at 7–8 ppm were the eight aromatic hydrogens of the biphenol section, the chemical shift at about 12.7 ppm was

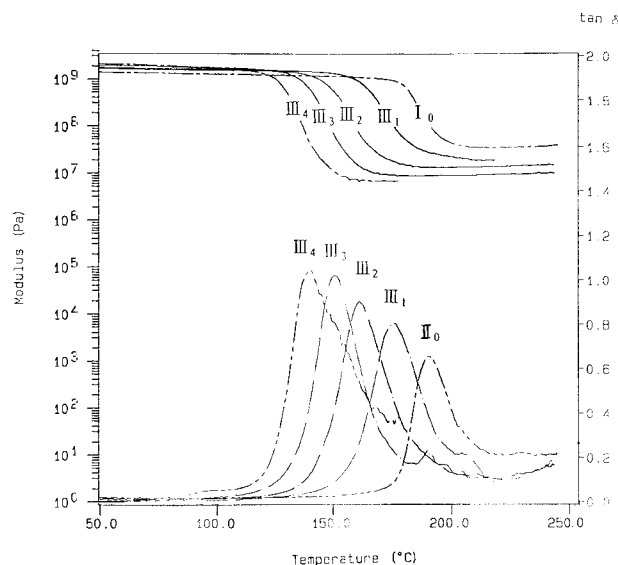
the two hydrogens of carboxylic acid (Ha and Hd); chemical shifts at around 2.6–2.9 ppm (Hc) and 3.5–3.7 ppm (Hb) for other hydrogens were also observed. The splits of Hb and Hc can be explained by (1) the P–H coupling— $^{31}\text{P}$  can couple with H (P–H, J1~J4 coupling), (b) the carbonyl b is a chiral center, and (c) the coupling of H–H. For the DOPOITA system chemical shifts at 7–8 ppm resulted from the eight aromatic hydrogens of the biphenol section; the chemical shift at about 12.4 ppm was from the two carboxylic acid hydrogens (Ha and Hd); and chemical shifts at around 2.9–3.0 ppm (Hb), 2.6–2.7 ppm (Hb), and 2.3–2.5 ppm (He) for other hydrogens were also observed. The splits of Hb, Hc, and He can be explained by reasons similar to that of DOPOMA. The assignments of  $^1\text{H-NMR}$  spectra shown in Figure 1 were consistent with the structure of the synthesized monomers.

#### Effect of Reaction Time and Catalyst on Advancement Reaction

Figure 2 displays the effect of reaction time on the EEW of the  $\text{II}_1\text{--II}_4$  system with a different catalyst such as 2E4MI or A1 and with no catalyst. The reaction was almost complete after the first 30 min. EEW increased with time and reached a constant value after 60 min. Thus they were reacted at 130°C for 60 min. For  $\text{III}_1\text{--III}_4$  the solubility of DOPOITA in DGEBA at 130°C was low, thus the reaction rate at 130°C was slow. However, if the temperature were raised to 160°C, the reaction could be completed in 60 min. Further-



**Figure 3** Dynamic mechanical analyses of  $\text{II/DDS}$  series epoxy resins.



**Figure 4** Dynamic mechanical analyses of **III/DDS** series epoxy resins.

more, unlike what was reported in our previous article, the reaction took place without any catalyst, which may be attributed to the solubility of DOPOMA and DOPOITA in DGEBA being higher than that of DOPOBQ in DGEBA.

### Thermal Properties of Cured Epoxy Resins

#### DMA Analysis

Figure 3 shows the dynamic mechanical analyses curves of the **II/DDS** series. All cured epoxy resins showed similar moduli by the dynamic mechanical analyses, and  $T_g$  (taken as the peak of relaxation, i.e., the peak of  $\tan \delta$ ) decreased with the increasing EEW of advanced epoxy. DGEBA–DDS has a smaller loss tangent and higher  $T_g$  than those of

**II<sub>1</sub>–II<sub>4</sub>/DDS**, from which it can be inferred that DGEBA–DDS was more rigid than the **II<sub>1</sub>–II<sub>5</sub>/DDS** systems. This may be attributed to the higher crosslink density of DGEBA–DDS than that of the advanced cured epoxy resins. Similar results were observed for the **III/DDS** system shown in Figure 4. Detailed DMA results, listed in Table II, show that at the same phosphorus content, the glass-transition temperature of **II/DDS** was higher than that of **III/DDS**. This may be explained by: (1) DOPOITA having a higher molecular weight than DOPOMA, thus at the same phosphorus content, the **III** series had a higher EEW than did the **II** series, which caused the crosslink density of **III/DDS** to be less than that of **II/DDS**, consequently reducing the glass-transition temperatures; and (2) the extra  $\text{CH}_2$  group in DOPOITA (compared with DOPOMA) increasing the flexibility (reduced rigidity) of polymer chains, thus lowering the glass temperature.

#### TGA Analysis

The TGA results of **II/DDS** and **III/DDS** under nitrogen and air are listed in Table III. The 5% degradation ( $T_{d,5\%}$ ) temperatures decreased with phosphorus content, while char yield increased with phosphorus content. Van Krevelen<sup>16</sup> has proposed that char residue on pyrolysis is linearly proportional to the oxygen index of halogen-free polymers. Increasing char formation limits the production of combustible gases, decreases the exothermicity of the pyrolysis reaction, and decreases the thermal conductivity of the burning materials, consequently limiting the flammability of the materials. The results shown in Table III suggest that the flame retardancy of epoxy resins was elevated via the phosphorus group. The results are consistent with

**Table II** DMA Results of Cured Epoxy Resins

Sample Code	$T_g$ °C (DMA)	Height of Loss Tangent	Modulus at 50°C, *10 <sup>9</sup> Pa	Modulus at $T_g + 30^\circ\text{C}$ , *10 <sup>9</sup> Pa
<b>II<sub>0</sub>/DDS</b>	190.3	0.655	1.880	0.044
<b>II<sub>1</sub>/DDS</b>	179.2	0.726	1.886	0.021
<b>II<sub>2</sub>/DDS</b>	167.7	0.794	1.693	0.022
<b>II<sub>3</sub>/DDS</b>	152.2	0.897	2.445	0.011
<b>II<sub>4</sub>/DDS</b>	142.7	1.029	2.349	0.009
<b>III<sub>1</sub>/DDS</b>	175.5	0.804	2.270	0.025
<b>III<sub>2</sub>/DDS</b>	160.9	0.904	2.380	0.017
<b>III<sub>3</sub>/DDS</b>	150.7	1.019	2.589	0.011
<b>III<sub>4</sub>/DDS</b>	139.9	1.043	2.845	0.009

**Table III TGA Results of Cured Epoxy Resins**

Sample Code	Air Atmosphere				N <sub>2</sub> Atmosphere		
	$T_{d,5\%}$	$T_{d,max}$		Char at 700°C (%)	$T_{d,\%}$	$T_{d,max}$	Char at 700°C (%)
		1st <sup>a</sup>	2nd <sup>b</sup>				
<b>II</b> <sub>0</sub> /DDS	439.2	472.4	641.1	6.0	441.7	475.1	16.8
<b>II</b> <sub>1</sub> /DDS	391.2	440.2	712.5	19.2	387.3	450.4	20.6
<b>II</b> <sub>2</sub> /DDS	389.7	431.9	660.7	20.5	385.6	425.2	19.9
<b>II</b> <sub>3</sub> /DDS	382.5	424.8	650.4	25.4	385.8	426.2	20.3
<b>II</b> <sub>4</sub> /DDS	384.5	435.6	648.0	27.5	391.8	456.3	21.6
<b>III</b> <sub>1</sub> /DDS	430.9	475.3	704.2	19.2	433.6	477.3	20.5
<b>III</b> <sub>2</sub> /DDS	420.2	460.4	648.1	21.2	426.2	460.6	19.6
<b>III</b> <sub>3</sub> /DDS	419.5	458.7	642.7	24.9	419.3	460.6	20.3
<b>III</b> <sub>4</sub> /DDS	416.6	460.6	ND <sup>c</sup>	27.8	425.9	472.4	20.2

<sup>a</sup> the first decomposition peak temperature<sup>b</sup> the second decomposition peak temperature<sup>c</sup> no second decomposition peak temperature was detected

the UL-94 measurement discussed below (Table IV). To our surprise, the thermal stability of **III**/DDS series (DGEBA-DOPOITA) was higher than that of the **II**/DDS (DGEBA-DOPOMA) series. According to organic chemistry, the P—C chain has the lowest chain energy, thus the initial degradation of polymer occurs at that chain. According to the results of our previous studies, the order of thermal stability of the phosphorus-containing epoxy was: DOPOBQ series > DOPOITA series > DOPOMA series (the structures of DOPOITA, DOPOMA, and DOPOBQ are shown in Scheme 4. Thermal stability can be explained by the effect of electron density. DOPOMA with two electron-withdrawing P=O and C=O groups reduced the electron density of the carbon; thus the carbon in P—C chain had the lowest electron

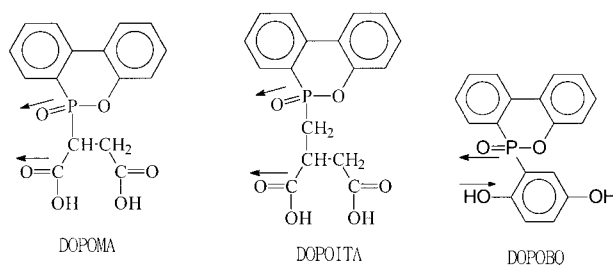
density. DOPOITA, with two electron-withdrawing P=O and C=O groups, had one CH<sub>2</sub> group between the C=O and P—C chains, which reduced the electron-withdrawing ability of C=O group and consequently caused the electron deficiency of the carbon in the P—C chain to less than that of DOPOMA. DOPOBQ has one electron-withdrawing P=O chain, while the OH can resonate with the carbon in the ortho position, which balances the electron-withdrawing effect of the C=O group. Thus, the order of thermal stability for the phosphorus-containing epoxy was: DOPOBQ series > DOPOITA series > DOPOMA.

#### UL-94 Measurement

UL-94 is another testing method for flame retardancy. During the test the polymer specimen was

**Table IV Phosphorus Content and UL-94 Results of Cured Epoxy Resins**

Sample Code	P% After Curing	Burning Time	Burning Time	UL-94 Rating
		After 1st Ignition (sec)	After 2nd Ignition (sec)	
<b>II</b> <sub>0</sub> /DDS	0%	>100	—	V-2
<b>II</b> <sub>1</sub> /DDS	0.80%	29	0	V-1
<b>II</b> <sub>2</sub> /DDS	1.24%	12	0	V-1
<b>II</b> <sub>3</sub> /DDS	1.70%	<1	1	V-0
<b>II</b> <sub>4</sub> /DDS	2.20%	<1	<1	V-0
<b>III</b> <sub>1</sub> /DDS	0.81%	14	1	V-1
<b>III</b> <sub>2</sub> /DDS	1.27%	9	1	V-1
<b>III</b> <sub>3</sub> /DDS	1.73%	7	1	V-0
<b>III</b> <sub>4</sub> /DDS	2.24%	4	<1	V-0



**Scheme 4** The structures of DOPOMA, DOPOITA, and DOPOBQ.

subjected to two 10-sec ignitions. After the first ignition, the flame was removed and the time for the polymer to self-extinguish ( $t_1$ ) recorded. Cotton ignition would be noted if polymer dripping occurred during the test. After cooling, a second ignition was performed on the same sample, and the self-extinguishing time ( $t_2$ ) and dripping characteristics were recorded. If  $t_1 + t_2$  were less than 10 sec with no dripping, it would be considered a V-0 material, an industrial standard for flame retardancy. As shown in Table IV, a V-1 grade for UL-94 can be achieved for **II**<sub>1</sub>-**II**<sub>2</sub>/DDS and **III**<sub>1</sub>-**III**<sub>2</sub>/DDS and V-0 grade can be achieved for **II**<sub>3</sub>-**II**<sub>4</sub>/DDS and **III**<sub>3</sub>-**III**<sub>4</sub>/DDS. Thus, incorporating 1.7% phosphorus into a cured epoxy chain makes these advanced epoxies into flame-retardant grade. From Table IV the effect of phosphorus contents on flame retardancy is obvious in the **II**/DDS series and in the **III**/DDS series. If the phosphorus content were less than 1.27%, the **III**/DDS series had a higher flame retardancy, while **II**/DDS series had a higher flame retardancy at higher phosphorus content.

## CONCLUSION

Two phosphorus-containing diacids, DOPOMA and DOPOITA, were synthesized from DOPO and maleic acid or itaconic acid, respectively. The structures of synthesized monomers were confirmed by EA, IR, mass, and NMR spectra. Two series of advanced epoxy resins were synthesized

and their thermal and flame-retardant properties examined. The reaction between DGEBA-DOPOMA and DGEBA-DOPOITA can be completed in about 60 min without any catalyst at 130°C and 160°C, respectively. The order of thermal stability of synthesized phosphorus-containing epoxy resins was: DOPOBQ series > DOPOITA series > DOPOMA, which can be explained by the effect of electron density. The glass-transition temperature decreased with the increasing EEW of advanced epoxy and can be explained by the effect of crosslink density. From UL-94 measurement, incorporating 0.8 wt % phosphorus into both cured epoxy chains made these epoxies meet V-1 specification grade, and incorporating 1.7 wt % phosphorus into both cured epoxy chain made those epoxies flame-retardant V-0 specification grade.

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