

# Flame Retardant Epoxy Resins Based on Diglycidyl Ether of Isobutyl Bis(hydroxypropyl)phosphine Oxide

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**ABSTRACT:** Phosphorous-containing epoxy resins were prepared from diglycidyl ether of isobutyl bis(hydroxypropyl)phosphine oxide (IHPOGly) and diglycidyl ether of bisphenol A (DGEBA) by crosslinking with 2,4-diaminotoluene. Several IHPOGly/DGEBA molar ratios were used to obtain materials with different phosphorous content. Thermal, dynamomechanical, and flame retardant properties were evaluated and related with the phosphorous content. The weight loss rate of phosphorous-containing resins is lower than that of the phosphorous-free resin for the thermoxidative degradation. Char yields under nitrogen do not show significant differences among the phosphorous-con-

taining resins and the phosphorous-free resin, while under air char yields increase with the phosphorous content. The presence of phosphorous increases the limiting oxygen index (LOI) values even when the phosphorous content is low, and no significant differences with the phosphorous content are observed. V-0 materials were obtained when the resins were tested for ignition resistance with the UL-94 test. © 2005 Wiley Periodicals, Inc. *J Appl Polym Sci* 99: 1367–1373, 2006

**Key words:** heteroatom-containing polymers; flame retardance; degradation; curing of polymers; crosslinking

## INTRODUCTION

Epoxy resins have many attractive properties: they are easy to cure and process, they are resistant to moisture, solvents and chemicals, and they have good mechanical and electrical properties, and adhesive strength. However, their flammability is a major limitation. The main fields that require the fire-retardancy of epoxy resins are electronics (printed wiring boards and semiconductor encapsulation) and transportation (automobiles and aircraft), in composite, structural, and furnishing elements.<sup>1</sup> Several approaches have been used to enhance flame retardancy in epoxies. The epoxy backbone has been modified with monomers containing flame retardant elements, and flame retardant additives have been used to achieve the desired level of flame retardancy.<sup>2</sup> The most effective way of improving fire retardancy in epoxy resins is the reactive approach. Additive type fire retardants are mostly used in coating or encapsulation, whereas reactive flame retardants are preferable in printed circuit

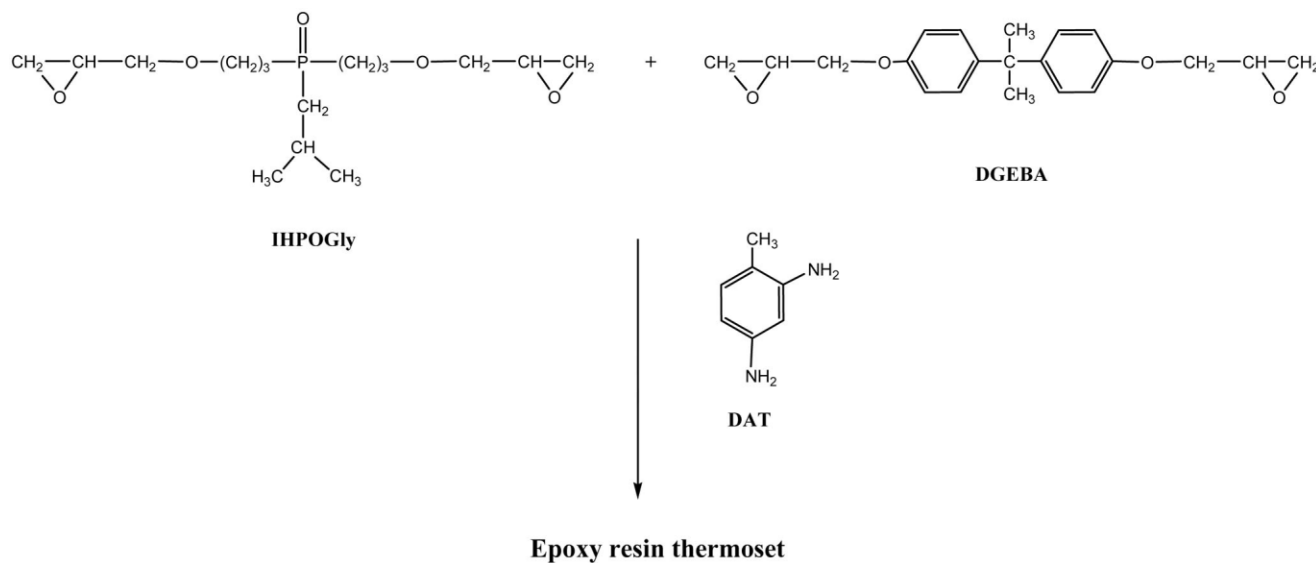
boards and composites to prevent the physical properties from deteriorating.

Halogen-containing monomers, such as diglycidylether of tetrabromo bisphenol A, have been used along with diglycidyl ether of bisphenol A to make commercial laminates more flame retardant. Under the effects of fire, they produce very dense smoke and toxic decomposition products. Phosphorous-containing monomers or hardeners can be used to make the flame retardancy of epoxy resins very efficient. They have been reported to exert both condensed-phase and vapor-phase flame retardant action.<sup>3</sup> Series of diglycidyl phosphates, diglycidyl phosphonates, and glycidyl phosphinates have been prepared. A large number of publications discuss the use of 9,10-dihydro-9-oxa-10-phosphaphenanthrene 10-oxide in the formulation of epoxy resins.<sup>2</sup> Moreover, phosphine oxide structures are often used to impart flame retardancy to epoxy resins, specially through curing agents because phosphine oxides are thermally and hydrolytically very stable. Indeed, the P—C bond in a phosphonate is more stable to hydrolysis than the P—O—C bond in a phosphate, and the P—C bond is broken just before the C—C bond because of its lower bond energy.<sup>4</sup> Moreover, the polarity of the phosphoryl group increases the hydrogen bonding ability of the resulting epoxy, thus improving network adhesion to various substrates.<sup>5</sup> However, these phosphine oxide structures are not frequent in epoxy monomers.

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The commercial phosphorous-containing diol isobutyl bis(hydroxypropyl)phosphine oxide (IHPO) has been used in halogen-free flame retardant formulations of a dual-cured thermosetting system consisting of an epoxy resin blended with an unsaturated polyether.<sup>6</sup> In a previous paper, we described how we obtained advanced phosphorous-containing epoxy resins by reacting IHPO with DGEBA and how they were cured with primary amines to give V-0 materials.<sup>7</sup> We also synthesized isobutyl bis(glycidylpropylether)phosphine oxide (IHPOGly) from IHPO and epichlorohydrin and investigated its thermal behavior. We studied the reactivity of this novel diglycidyl compound using initiators and hardeners.<sup>8</sup>

In this paper, we describe how epoxy systems containing different amounts of this new diglycidylether are prepared from a phosphine oxide (IHPOGly) and DGEBA by curing with a primary amine (Scheme 1). We also discuss their thermal, mechanical, and flame-retardant properties.

## EXPERIMENTAL

### Materials

Commercial isobutyl bis(hydroxypropyl)phosphine oxide (IHPO) was a gift from Cytec Canada Inc. (trade name Cyagard RF1243). Epichlorohydrin (EPC) (Fluka), diglycidyl ether of bisphenol A (DGEBA) Eq. Epoxy ( $E.E = 192$  g/eq) (Aldrich), and tetrabutylammonium hydrogen sulfate (TBAH) (Aldrich) were used as received. All solvents were purified by standard procedures.

The curing agent 2,4-diaminotoluene (DAT) was purchased from Aldrich and used without any further purification. Isobutyl(glycidylpropylether) phosphine oxide was synthesized as previously described.<sup>8</sup>

### Crosslinking reaction

Diglycidyl compounds and amine hardener were mixed in stoichiometric amounts by dissolving the

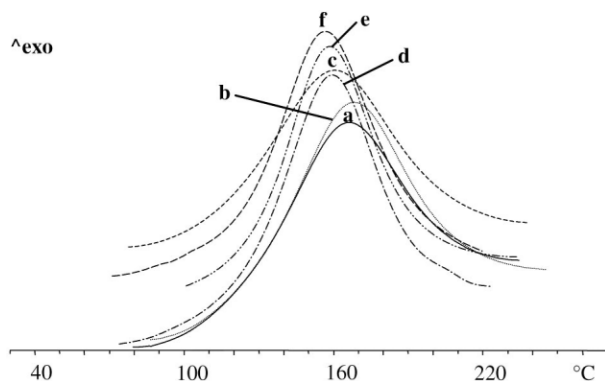
**TABLE I**  
Crosslinking Data of IHPOGly, DGEBA, and IHPOGly/DGEBA using DAT as Hardener

|        | Initial mixture | Molar ratio | $T_g$ (°C) | $T_{onset}$ (°C) <sup>a</sup> | $T_{max}$ (°C) <sup>b</sup> | $\Delta H$ (KJ/mol epoxide) <sup>c</sup> |
|--------|-----------------|-------------|------------|-------------------------------|-----------------------------|--|
| IT-7.8 | IHPOGly         | —           | 72         | 88                            | 165                         | 131                                      |
| IDT-7  | IHPOGly/DGEBA   | 0.90/0.10   | 86         | 85                            | 166                         | 170                                      |
| IDT-5  | IHPOGly/DGEBA   | 0.65/0.35   | 105        | 78                            | 160                         | 146                                      |
| IDT-3  | IHPOGly/DGEBA   | 0.40/0.60   | 122        | 79                            | 158                         | 157                                      |
| IDT-1  | IHPOGly/DGEBA   | 0.125/0.875 | 128        | 78                            | 158                         | 161                                      |
| DT-0   | DGEBA           | —           | 159        | 85                            | 156                         | 101                                      |

<sup>a</sup> Initial temperature of the crosslinking exotherm.

<sup>b</sup> Temperature of the maximum heat release rate.

<sup>c</sup> Reaction enthalpy.



**Figure 1** DSC plots (10°C/min) of curing of IHPOGly, DGEBA, and IHPOGly/DGEBA with DAT (a) IT-7.8, (b) IDT-7, (c) IDT-5, (d) IDT-3, (e) IDT-1, and (f) DT-0.

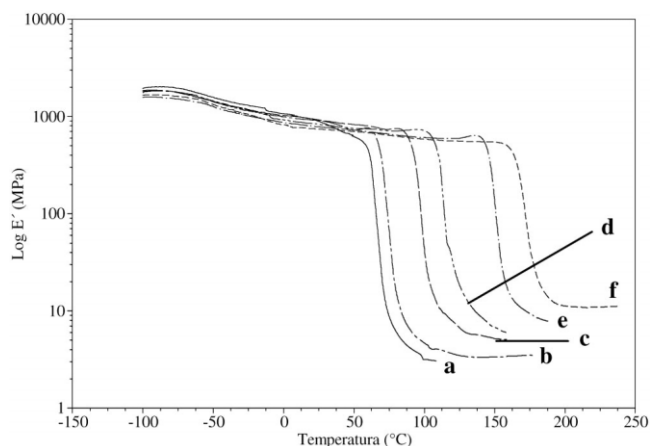
components in  $\text{CH}_2\text{Cl}_2$  and subsequently evaporating the solvent at room temperature. Sample bars for dynamomechanical analysis, thermogravimetric analysis, and burn tests were obtained by placing in a mold and cured at 140°C for 5 h and postcured at 180°C for 4 h.

### Instrumentation

$^1\text{H}$  300 MHz,  $^{13}\text{C}$  75.4 MHz, and  $^{31}\text{P}$  161.9 MHz NMR spectra were obtained using a Varian Gemini 300 spectrometer with Fourier transform,  $\text{CDCl}_3$  as solvent, and tetramethyl silane (TMS) or phosphoric acid as internal standards. Calorimetric studies were carried out on a Mettler DSC821e thermal analyzer using  $\text{N}_2$  as a purge gas (20 mL/min) at scan rates between 5 and 20°C/min. Samples were prepared by dissolving IHPOGly in  $\text{CH}_2\text{Cl}_2$  and mixing the resulting solution with the required amount of DGEBA. This solution was evaporated at room temperature under vacuum. About 5 mg of a known weight of the mixture was put into the aluminum pan and polymerization was monitored in a DSC experiment. Thermal stability studies were carried out on a Mettler TGA/SDTA851e/LF/1100 with  $\text{N}_2$  as a purge gas at scan rates of 10°C/min. Mechanical properties were measured using a dynamic mechanical thermal analysis (DMTA) apparatus (TA DMA 2928). Specimens ( $0.5 \times 10 \times 40 \text{ mm}^3$ , depth  $\times$  width  $\times$  length) were tested in a single cantilever configuration. The thermal transitions were studied in the  $-100$  to  $250^\circ\text{C}$  range at a heating rate of 2 or 5°C/min and at a fixed frequency of 1 Hz.

### General procedure for modified UL-94 burn test

Five sample bars ( $60 \times 40 \times 0.5 \text{ mm}^3$ ) were used for this test. The height of the Bunsen burner flame was 25 mm, and the height from the top of the Bunsen burner



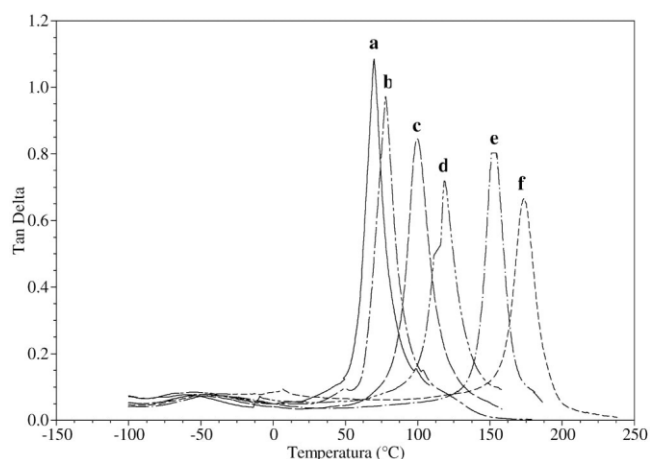
**Figure 2** Storage modulus of crosslinked polymers (a) IT-7.8 (—), (b) IDT-7 (---), (c) IDT-5 (—), (d) IDT-3 (---), (e) IDT-1 (- -), and (f) DT-0 (—).

to the bottom of the test bar was 10 mm. All test bars underwent two trials and each trial consisted of ignition for 10 s, after which the flame was removed and the time for self-extinguishing and dripping characteristics were recorded.

The limiting oxygen index (LOI) is the minimum concentration of oxygen determined in a flowing mixture of oxygen and nitrogen that will just support the flaming combustion of materials. LOI values were measured on a Stanton Redcroft, provided with an Oxygen Analyser, on plaques of the polymers  $100 \times 6 \times 4 \text{ mm}^3$  prepared by molding.

## RESULTS AND DISCUSSION

Epoxy resins can be fire retarded by using phosphorous-containing monomers. To preserve the functionality of the epoxy resins, isobutyl bis(hydroxypropy-



**Figure 3** Loss factor of crosslinked polymers (a) IT-7.8 (—), (b) IDT-7 (---), (c) IDT-5 (—), (d) IDT-3 (---), (e) IDT-1 (- -), and (f) DT-0 (—).

**TABLE II**  
Thermal Transitions of the Crosslinked Materials

|        | Molar ratio <sup>a</sup> | $T_{\beta}$ (°C) | $T_g$ (°C) |             |                     |
|--------|--------------------------|------------------|------------|-------------|---------------------|
|        |                          |                  | DSC        | $E''_{max}$ | $\tan \delta_{max}$ |
| IT-7.8 | —                        | -59              | 72         | 62          | 70                  |
| IDT-7  | 1.8/0.2                  | -53              | 86         | 68          | 78                  |
| IDT-5  | 1.3/0.7                  | -51              | 105        | 92          | 100                 |
| IDT-3  | 0.8/1.2                  | -50              | 122        | 109         | 118                 |
| IDT-1  | 0.25/1.75                | -40              | 128        | 146         | 152                 |
| DT-0   | —                        | -41              | 159        | 165         | 174                 |

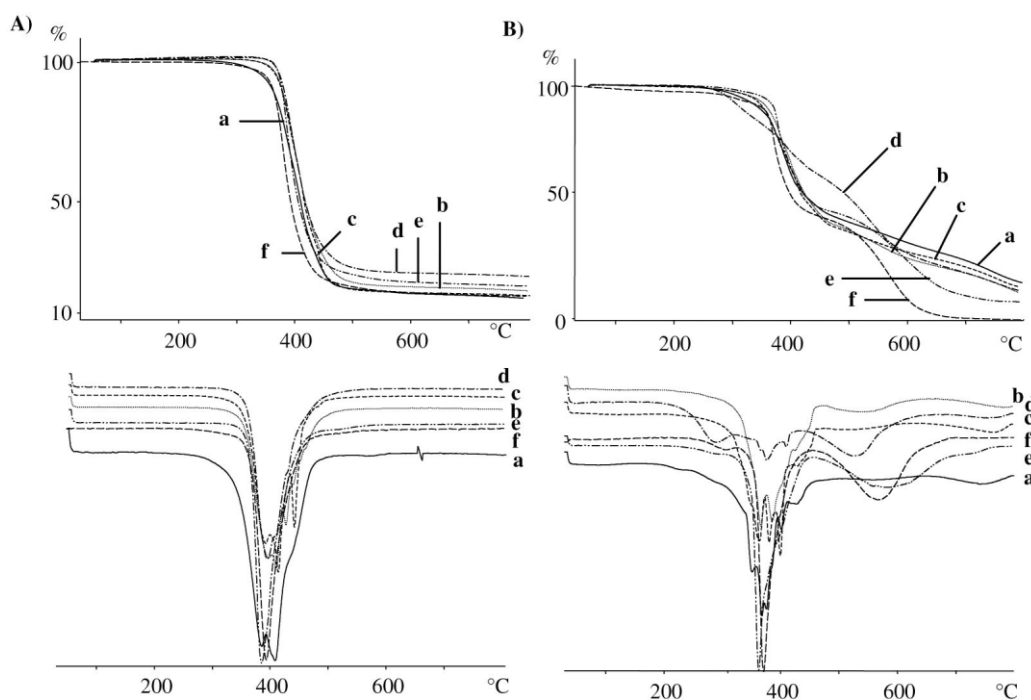
<sup>a</sup> Molar ratio of IHPOGly/DGEBA.

l)phosphine oxide (IHPO) was used as a reactive flame retardant in combination with bisphenol A epoxy resins, to obtain advanced prepolymers that were crosslinked with primary amines.<sup>7</sup> Alternatively, isobutyl bis(hydroxypropyl)phosphine oxide (IHPO) can be reacted with epichlorhydrin (EPC), which will lead to a phosphorous-containing monomer, IHPOGly used for preparing the flame retarding epoxy resins as was previously described.<sup>8</sup>

In this work, the IHPOGly was mixed with regular DGEBA and crosslinked with DAT to obtain materials with different contents of phosphorous. Mixtures of IHPOGly and DGEBA were prepared in suitable molar ratios to obtain final materials with a phosphorous content of 1, 3, 5, and 7% and crosslinked with DAT (IDT-1, IDT-3, IDT-5, and IDT-7). Table I summarizes the sample compositions, crosslinking data and  $T_g$ 's of

the final materials. To compare, the crosslinking data of IHPOGly with DAT (IT-7.8) and DGEBA with DAT (DT-0) were also collected in the same table. Figure 1 shows the DSC plots of these mixtures. As can be seen from the crosslinking exotherms, the reaction starts at about 80°C and the temperature of the maximum rate of heat release is around 160°C. Few differences can be found among the mixtures, or between the mixtures and the pure diglycidyl compounds. However, the  $T_g$  values increase with the DGEBA content. Since similar reactivity can be inferred from DSC data for both the diglycidyl compounds, IHPOGly and DGEBA, crosslinking density must be similar, and differences in  $T_g$  values must be related to the chemical structure of DGEBA with aromatic moieties that causes restriction in the segmental mobility. It should be pointed out that samples with lower amounts of IHPOGly have higher  $T_g$  values, even when this monomer contains a strong polar P=O group. This shows that aromatic rings have a major influence on  $T_g$  values. The reaction enthalpy values cannot be correlated with the mixture compositions, but are slightly higher for the mixtures than for the pure diglycidyls.

According to DSC data, the networks were prepared by heating at 140°C for 5 h and at 180°C for 4 h. The dynamic mechanical behavior of the phosphorous-containing epoxy resins was obtained as a function of the temperature beginning in the glassy state of each composition to the rubbery plateau of each ma-



**Figure 4** TGA plots (10°C/min) and first derivative curves of crosslinked polymers (a) IT-7.8 (—), (b) IDT-7 (---), (c) IDT-5 (—), (d) IDT-3 (- - -), (e) IDT-1 (- · - · -), (f) DT-0 (—); (A) in N<sub>2</sub> and (B) in air.

TABLE III  
Thermogravimetric Data of Crosslinked Materials

|        | Molar ratio <sup>a</sup> | P (%) | Nitrogen                     |                              |                                      | Air                          |                              |                                      |
|--------|--------------------------|-------|------------------------------|------------------------------|--------------------------------------|------------------------------|------------------------------|--------------------------------------|
|        |                          |       | $T_{10\%}$ (°C) <sup>b</sup> | $T_{\max}$ (°C) <sup>c</sup> | $R_{700^\circ\text{C}}$ <sup>d</sup> | $T_{10\%}$ (°C) <sup>b</sup> | $T_{\max}$ (°C) <sup>c</sup> | $R_{700^\circ\text{C}}$ <sup>d</sup> |
| IT-7.8 | 1/0                      | 7.8   | 357                          | 400                          | 15                                   | 344                          | 385                          | 18                                   |
| IDT-7  | 0.900/0.100              | 7.0   | 372                          | 414                          | 17                                   | 349                          | 413                          | 17                                   |
| IDT-5  | 0.650/0.350              | 5.0   | 374                          | 411                          | 20                                   | 354                          | 400                          | 13                                   |
| IDT-3  | 0.400/0.600              | 3.0   | 375                          | 394                          | 24                                   | 309                          | 390,533                      | 13                                   |
| IDT-1  | 0.125/0.875              | 1.0   | 372                          | 384                          | 20                                   | 367                          | 378,583                      | 4                                    |
| DT-0   | 0/1                      | 0     | 363                          | 379                          | 16                                   | 349                          | 370,568                      | 0                                    |

<sup>a</sup> Molar ratio of IHPOGly/DGEBA.

<sup>b</sup> Temperature of 10% of weight loss.

<sup>c</sup> Temperature of the maximum weight loss rate.

<sup>d</sup> Char yield at 700°C.

terial (Fig. 2). The crosslink density of a polymer can be estimated from the plateau of the elastic modulus in the rubbery state.<sup>9</sup> However, this theory is strictly valid only for lightly crosslinked materials, and was therefore used only to make qualitative comparisons of the level of crosslinking among the various polymers. As can be seen, crosslink density increases as the IHPOGly content decreases. As far as the chemical structures of these networks are concerned, this behavior is to be expected since aromatic structures should lead to tighter networks.

Figure 3 shows the plots of loss factor ( $\tan \delta$ ) versus temperature. As can be seen,  $\beta$  relaxations occur around  $-50^\circ\text{C}$ , which may be due to the motions of the  $[-\text{CH}_2-\text{CH}(\text{OH})-\text{CH}_2-\text{O}-]$  aliphatic segments from the ring opening of the epoxide<sup>10</sup> (Table II). The  $T_g$ 's of the crosslinked materials can be detected as the maximum of the loss modulus ( $E''$ ) or as the  $\alpha$  relaxation peak of the loss factor. Table II shows the  $T_g$  values of both measurements. From a practical point of view, the maximum  $E''$  is the most appropriate value, since it corresponds to the highest temperature of use. As can be seen,  $T_g'$  values from DMTA follow the trend mentioned earlier.  $T_g'$ 's increase as the content of DGEBA increases. Moreover, the analysis of the height and width of the  $\alpha$  relaxation peak shows trends in the crosslinking densities and network homogeneities, as the composition of the material changes. The height of the  $\tan \delta$  peak, which is associated with the crosslink density, decreases as the IHPOGly content increases. Because  $\tan \delta$  is the ratio of viscous components to elastic components, it can be assumed that the decreasing height is associated with lower segmental mobility and fewer relaxing species and is therefore indicative that the networks for the DGEBA-rich samples are tighter.

The peak width at half-height broadens as the number of branching modes increases, which produces a wider distribution of structures. The range of temperatures at which the different network segments gain mobility therefore increases. There were no significant

differences among the IHPOGly/DGEBA samples, thus showing similar branching distribution for all samples.

To examine the effect of phosphorus content on thermal stability and the decomposition behavior, TGA data under nitrogen and air atmospheres were determined and analyzed. Figure 4 shows the weight loss with the temperature for the epoxy compositions as well as the derivative curves. Table III summarizes the thermogravimetric data. As can be seen, the temperature of 10% weight loss ( $T_{10\%}$ ) does not change significantly with phosphorous content. The shape of the derivative curves in nitrogen show that the degradation takes place in a broad temperature range and the phosphorous content again seems to have no influence on the degradation. In air, we observe that the first step takes place at temperatures similar to those in nitrogen and the second step takes place at temperatures above  $500^\circ\text{C}$ , which is when the thermoxidative degradation takes place. It can be seen that the weight loss rate of phosphorous-containing resins is significantly lower than that of the phosphorous-free resins for the thermoxidative degradation. This behavior is in accordance with the mechanism of improved fire performance via phosphorous modification. In this retarded-degradation phenomenon, the phosphorous groups form an insulating protective layer, which prevents the combustible gases from transferring to the surface of the materials, increases the thermal stability at higher temperatures, and improves the fire resistance. To show the different behavior of phosphorous-containing resins, a plot of weight loss rate versus weight loss in air has been made (Fig. 5). As can be seen, the phosphorous-free DT-0 resin shows maxima around 30 and 90% weight loss. For the phosphorous-containing resin IDT-1, a similar process takes place with a maximum around 20% and a similar weight loss rate. For the phosphorous-containing resin IDT-7.8, which contains only IHPOGly, the maximum around 35% shows a shoulder at 20%, and the maximum weight loss rate is lower than that of the afore-

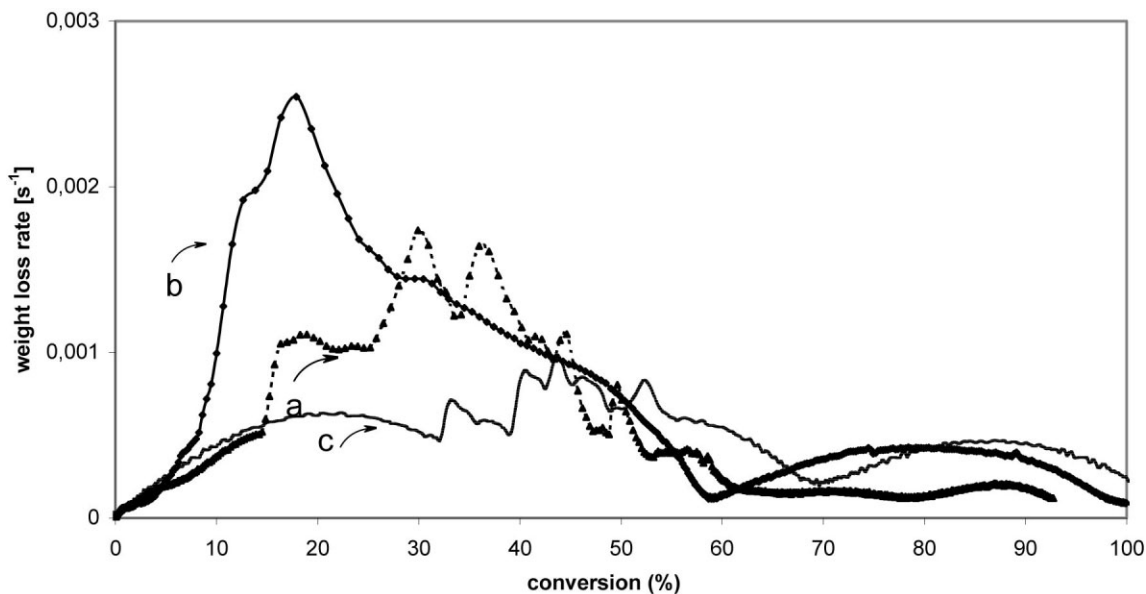


Figure 5 Weight loss rate versus decomposition degree in air for (a) IT-7.8, (b) IDT-1, and (c) DT-0.

mentioned resins. The step that takes place at higher temperatures must be due to thermoxidative degradation. It can be seen that the weight loss rate of phosphorous-containing resins is lower than that of phosphorous-free resins for weight losses higher than 60%.

Char yield under nitrogen is correlated with the polymer's flame retardancy<sup>11</sup> but it should be pointed out that, in our case, the experimental char yields of the phosphorous-containing resins and the phosphorous-free resins are not significantly different under nitrogen. Under air, the char yield is low and increases with the phosphorous content.

We tested the flame retardancy of the thermosets with the UL-94 flame test, in which the sample was suspended above cotton.<sup>12</sup> The sample was subjected to two 10-s ignitions, with a calibrated methane-fuelled flame in a controlled-size unit that was free of passing air currents. After the first ignition, the flame was removed and the time the polymer took to self-extinguish and the time/dripping characteristics were recorded. The same sample was then subjected to a second ignition and the self-extinguishing time/drip-

ping characteristics were recorded. If the sample self-extinguished in less than 10 s for first and second ignitions with no dripping, we considered it to be a V-0 material, which is an industry standard for flame retardancy. The results of the burn tests for the materials crosslinked with DAT are V-0 for all phosphorous-containing materials, while the phosphorous-free resins were V-1, as expected.

The LOI values, which can be taken as an indicator to evaluate the polymer's flame retardancy of the phosphorous-containing resins, were also measured and shown in Table IV. As can be seen, the presence of phosphorous increases the LOI values even when the phosphorous content is low, and no significant differences with the phosphorous content are observed.

## CONCLUSIONS

Phosphorous-containing epoxy resins with different phosphorous content were obtained from IHPOGly and DGEBA by crosslinking with DAT. The crosslinking behavior does not show significant differences

TABLE IV  
UL-94 and LOI Flame Tests Results for IHPOGly, DGEBA, and IHPOGly/DGEBA Cured with DAT

|       | $R_{700^{\circ}\text{C}}$<br>(nitrogen) | $R_{700^{\circ}\text{C}}$<br>(air) | 1 <sup>a</sup> ignition <sup>a</sup> (s) | 2 <sup>a</sup> ignition <sup>a</sup> (s) | Test UL-94 | LOI (% O <sub>2</sub> , v/v) |
|-------|---|------------------------------------|--|--|------------|------------------------------|
| IDT-7 | 17                                      | 17                                 | 1, 1, 0, 1, 0                            | 0, 2, 5, 2, 3                            | V-0        | 29.2                         |
| IDT-5 | 20                                      | 13                                 | 1, 0, 0, 0, 1                            | 5, 3, 2, 2, 3                            | V-0        | 29.5                         |
| IDT-3 | 24                                      | 13                                 | 2, 0, 1, 0, 1                            | 1, 5, 2, 3, 2                            | V-0        | 30.6                         |
| IDT-1 | 20                                      | 4                                  | 2, 2, 3, 2, 3                            | 2, 5, 5, 4, 5                            | V-0        | 28.2                         |
| DT-0  | 16                                      | 0                                  | 12, 14, 13, 10, 12                       | 4, 5, 5, 4, 4                            | V-1        | 24.5                         |

<sup>a</sup> Time to self-extinguish after ignition. The five numbers are for five separate tests on separate rectangular samples.

among the different mixtures or between the mixtures and pure diglycidyl compounds.  $T_g$  values of thermosets increase with the DGEBA content, as well as crosslinking density. Char yields under nitrogen do not show significant differences among the phosphorous-containing resins and the phosphorous-free resin. The presence of phosphorous increases the LOI values even when the phosphorous content is low (1%), and no significant differences with the phosphorous content are observed. V-0 materials were obtained when the resins were tested for ignition resistance with the UL-94 test.

## References

1. Hamerton, I.; Lu, S.-Y. *Prog Polym Sci* 2002, 27, 1661.
2. Levchik, S. V.; Weil, E. D. *Polym Int* 2004, 53, 1901.
3. Jain, P.; Choudhary, V.; Varma, I. K. *J Macromol Sci Polym Rev* 2002, 42, 139.
4. Quittmann, U.; Lecamp, L.; Youssef, W.; Brunel, C. *Macromol Chem Phys* 2001, 202, 608.
5. Wang, S.; Zhuang, H.; Shobha, H. K.; Glass, T. E.; Shankarapandian, M.; Shultz, A. R.; McGrath, J. E. *Macromolecules* 2001, 34, 8051.
6. La Rosa, A. D.; Recca, A.; Carter, J. T.; McGrail, P. T. *Polymer* 1999, 40, 4093.
7. Alcón, M. J.; Ribera, G.; Galià, M.; Cádiz, V. *J Polym Sci Part A: Polym Chem* 2005, 43, 3510.
8. Alcón, M.; Ribera, G.; Galià, M.; Cádiz, V. *Polymer* 2003, 44, 7291.
9. Tobolsky, A. V.; Carlson, D. W.; Indictor, N. J. *J Polym Sci* 1961, 54, 175.
10. Yang, X.; Painter, P. C.; Coleman, M. M. *Macromolecules* 1992, 25, 2156.
11. Van Krevelen, D. W. *Polymer* 1975, 16, 615.
12. Underwriters Laboratories. UL94: Standard for Test for Flammability of Plastic Materials for Parts in Devices and Appliances, 4th ed.; Underwriters Laboratories: Research Triangle Park, NC, 1991.