



## Synthesis of phosphorus-containing polyol and its effects on impact resistance and flame retardancy of structural epoxy adhesives

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### ARTICLE INFO

#### Keywords:

Structural epoxy adhesive  
Impact resistance  
Flame retardant

### ABSTRACT

Structural epoxy adhesives are commonly used in many industries but are limited by their flammability and low impact resistance. In order to improve the flame retardancy of epoxy adhesives, phosphorus-containing species are mainly used as reactive flame retardants, but these generally cause a decrease in mechanical strength. In this study, an oligomeric phosphorus-containing polyol (P-polyol) was synthesized by step polymerization between phenylphosphonic dichloride and ethylene glycol. The curing behavior, viscoelastic properties, adhesion performance, impact resistance, and flame retardancy of an epoxy resin containing P-polyol were characterized. Especially, impact resistance was evaluated by the impact wedge-peel test, an industry-standard testing method. The flame retardancy of the structural epoxy adhesive improved steadily with the addition of *p*-polyol. Impact resistance also improved at moderate P-polyol contents (8–10 parts per 100 parts of resin (8–10 phr)) but drastically decreased at a high content (15 phr). Based on this study, by incorporating P-polyol, it is possible to simultaneously improve the impact resistance and flame retardancy of structural epoxy adhesives. We obtained both a high impact resistance (dynamic resistance to cleavage: 16.9 N/mm) and flame retardancy (heat release capacity: 255 J/(g·°C)) at an optimum P-polyol content (8 phr).

### 1. Introduction

Structural epoxy adhesives have good wetting properties, high mechanical strengths, and low shrinkage and creep, and thus they are widely used in many industries including automobiles, aircrafts, buildings, and so on [1–3]. For specific applications, such as for automobiles, the impact resistance of the structural epoxy adhesive should be maximized [4]. There are many ways to improve impact resistance, like reinforcement fillers [5–9] and chemical modification of the epoxy resin [10–14].

Methods for evaluating the impact resistance of structural epoxy adhesives are generally divided into two groups [4]: analyzing the intrinsic impact resistance of the epoxy resin or the impact resistance of

a complete adhesive joint. The former is characterized without any adherents, only with the epoxy resin itself, like in the Izod impact test. On the other hand, since the latter represents the impact behavior of a complete adhesive joint, like in the impact wedge-peel test, it can determine many factors such as surface properties, adherent properties, and joint geometry. For these reasons, the impact wedge-peel test is commonly used for many industrial applications [4].

The flammability of structural epoxy adhesives limits their industrial applications, especially in the automobile and aerospace industries [15, 16]. Non-flammable fillers including clay, graphene, and carbon nanotubes have previously been used to improve the flame retardancy of structural epoxy adhesives [17–21]. However, because of the resulting disadvantages such as reduced mechanical strength, poor compatibility,

*Abbreviations:* DMA, dynamic mechanical analysis; DOPO, 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide; DSC, differential scanning calorimetry; PCFC, pyrolysis combustion flow calorimetry; phr, parts per hundred parts of the resin; TGA, thermogravimetric analysis.

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<https://doi.org/10.1016/j.ijadhadh.2020.102601>

Received 15 September 2019; Accepted 19 March 2020

Available online 20 March 2020

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and leaching, reactive flame retardants have instead received increased attention [15,22–25]. Because phosphorus components promote the formation of char, which can act as a protective coating, phosphorus-containing species are mainly used as reactive flame retardants [26].

Generally, flame retardancy can be improved by the addition of flame retardants, but the mechanical strength, including the impact resistance, of the epoxy resin also decreases sharply [27]. Hu et al. investigated the mechanical strength and flame retardancy of epoxy resins containing derivatives of DOPO (9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide) as reactive flame retardants [22]. They evaluated the intrinsic impact resistance of the epoxy resins using a pendulum impact testing machine and showed that the DOPO derivatives could reduce the extent to which the impact resistance decreased. To the best of our knowledge, however, there have been few other studies on improving impact resistance with reactive flame retardants. Furthermore, the impact resistance of flame-retardant epoxy adhesives is typically only characterized with the epoxy resin itself rather than with a complete adhesive joint.

In this study, to simultaneously improve both the impact resistance and flame retardancy of epoxy adhesives, we synthesized an oligomeric phosphorus-containing polyol (P-polyol) as a reactive flame retardant by step polymerization of phenylphosphonic dichloride and ethylene glycol. With controlled amounts of the synthesized P-polyol, we characterized the curing behavior, viscoelastic properties, adhesion performance, impact resistance, and flame retardancy of the epoxy adhesives. Especially, impact wedge-peel testing was performed to characterize the impact resistance of a complete adhesive joint.

## 2. Materials and methods

### 2.1. Synthesis of the phosphorus-containing polyol (P-polyol)

Phenylphosphonic dichloride and ethylene glycol (Sigma-Aldrich, USA) were used as monomers. Dichloromethane and 4-dimethylaminopyridine (Sigma-Aldrich) were used as the solvent and catalyst, respectively, and triethylamine (Sigma-Aldrich) was used to trap the hydrogen chloride. Sodium sulfate (Samchun, Republic of Korea) was used as a water remover, and deuterated dimethyl sulfoxide (DMSO- $d_6$ , Sigma-Aldrich, USA) was used as a nuclear magnetic resonance (NMR) solvent.

Dichloromethane (80 g), 4-dimethylaminopyridine (4.9 g, 0.1 eq.), triethylamine (81.0 g, 2 eq.), and ethylene glycol (37.2 g, 1.5 eq.) were added to a three-neck flask. The reaction temperature was 45 °C, and the magnetic stirrer was set to 300 rpm. The system was refluxed and purged by  $N_2$  gas. Dichloromethane (80 g) and phenylphosphonic dichloride

(77.8 g, 1 eq.) were added to a dropping funnel, then dropped slowly into the solution. After 24 h, the product was filtered, salt was removed, and the filtrate was washed by distilled water three times. Water was removed by sodium sulfate, and the final product was obtained by vacuum drying. The reaction equation between phenylphosphonic dichloride and ethylene glycol is shown in Fig. 1 (a).

The chemical structure of the synthesized P-polyol was characterized by Fourier-transform infrared spectroscopy (FT-IR, Nicolet 6700, Thermo Scientific, USA) and  $^1H$  and  $^{31}P$  NMR spectroscopies (Avance DPX-400, Bruker, USA) with DMSO- $d_6$  as an NMR solvent. Titration (888 Titrand, Metrohm, Switzerland) was carried out to evaluate the OH value. The absolute molecular weight of the P-polyol was calculated from the OH value and  $^1H$  NMR results.

### 2.2. Preparation and curing of epoxy adhesives containing P-polyol

An epoxy adhesive was prepared by blending epoxy resin and curing agent. As shown in Table 1, two epoxy resins (bisphenol-A diglycidyl epoxy and urethane-modified epoxy) and two curing agents (dicyandiamide and substituted urea) were used. The equivalent ratio of epoxy to curing agent was set to 1.00. The synthesized P-polyol was added at 0, 2, 4, 8, 10, and 15 phr (per hundred resin) to the epoxy adhesive. After blending, all samples were cured at 170 °C for 40 min. The equation of the curing reaction is shown in Fig. 1 (b) and hydroxyl group of the P-polyol reacted with hydroxyl group of the epoxy resin.

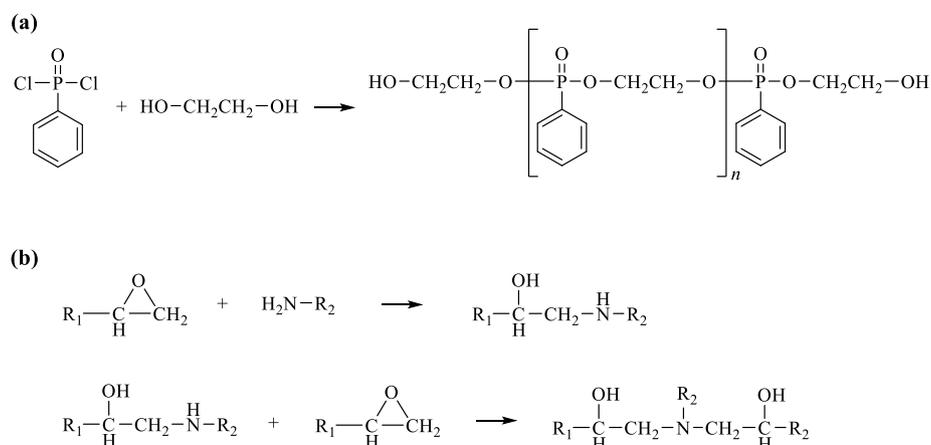
### 2.3. Differential scanning calorimetry (DSC)

The curing behaviors of the epoxy adhesives containing P-polyol were characterized using DSC (Q200, TA Instruments, USA). As the temperature was increased from 100 °C to 240 °C at a constant heating rate (5 °C/min), the heat flow of the curing reaction was measured under  $N_2$  purging conditions (50 ml/min).

**Table 1**

Composition of the epoxy adhesive.

	Product (Company)	Chemical	Equivalent weight (g/eq.)	Content (g)
Epoxy	YD-128 (Kukdo Chemical Co.)	Bisphenol-A diglycidyl epoxy	187	8.93
	UME (Kukdo Chemical Co.)	Urethane-modified epoxy	475	11.88
Curing agent	Dyhard 100S (Alzchem)	Dicyandiamide	21	0.955
	Dyhard UR500 (Alzchem)	Substituted urea	3	0.082



**Fig. 1.** (a) Synthesis of P-polyol and (b) curing equation of epoxy adhesives.

## 2.4. Dynamic mechanical analysis (DMA) & single lap shear test

DMA (Q800, TA Instruments, USA) was used to characterize the viscoelastic properties of the epoxy adhesives containing P-polyol. Flat-shaped specimens were prepared by curing the epoxy adhesives in a flat-shaped PTFE mold ( $l, w, t = 60 \text{ mm}, 12.8 \text{ mm}, 3 \text{ mm}$ ); the specimens were then fixed under a dual cantilever clamp. The temperature was increased from  $50 \text{ }^\circ\text{C}$  to  $200 \text{ }^\circ\text{C}$  at a constant heating rate ( $5 \text{ }^\circ\text{C}/\text{min}$ ), strain (0.1%), and frequency (1 Hz).

Single lap shear tests were performed to compare the adhesion properties of the epoxy adhesives containing P-polyol. The epoxy adhesives were applied to a CR340 steel plate ( $l, w, t = 100 \text{ mm}, 25 \text{ mm}, 1.6 \text{ mm}$ ) with a specific area ( $12.5 \text{ mm} \times 25 \text{ mm}$ ), then cured at  $170 \text{ }^\circ\text{C}$  for 40 min. The thickness of the epoxy adhesives was set to 0.2 mm. Stress-strain curves were obtained by using a universal testing machine (UTM 5982, Instron, UK) with a constant strain rate ( $1.2 \text{ mm}/\text{min}$ ), and lap shear strength was defined as the maximum stress (MPa) on the stress-strain curve. The single lap shear tests followed the ASTM D1002 standard.

## 2.5. Impact wedge-peel test

The impact resistance of the epoxy adhesives containing P-polyol was characterized by impact wedge-peel tests. Test specimens were prepared as shown in Fig. 2 (a); the epoxy adhesives were applied (area:  $20 \text{ mm} \times 20 \text{ mm}$ , thickness: 0.2 mm) to two bent CR340 steel plates ( $l, w, t = 90 \text{ mm}, 20 \text{ mm}, 1.6 \text{ mm}$ ) [5] and then cured at  $170 \text{ }^\circ\text{C}$  for 40 min. After preparation, as the specimens were cleaved by the wedge at a specific velocity ( $2.0 \text{ m/s}$ ), the force was measured by a drop weight tester (Dyntaup Model 9250HV, Instron, USA). The impact wedge-peel tests followed the ISO 11343 standard.

An epoxy adhesive with a high impact resistance represents stable crack growth, as shown in Fig. 2 (b), whereas an epoxy adhesive with a

low impact resistance represents unstable crack growth, as shown in Fig. 2 (c). Typically, the impact resistance is evaluated by calculating the average force. However, as it is difficult to compare average forces between stable and unstable crack growth, we instead evaluated impact resistance by calculating the energy for crack growth ( $E_C$ ) [5].

## 2.6. Thermogravimetric analysis (TGA)

TGA (Q500, TA instruments, USA) was carried out with a specimen size of 15 mg. The temperature was increased from  $50 \text{ }^\circ\text{C}$  to  $800 \text{ }^\circ\text{C}$  at a constant rate ( $10 \text{ }^\circ\text{C}/\text{min}$ ) in an air atmosphere (air purging,  $40.0 \text{ ml}/\text{min}$ ).

## 2.7. Pyrolysis combustion flow calorimetry (PCFC)

PCFC (FAA-PCFC, Fire Testing Technology, UK) was carried out with a specimen size of 5 mg. The specimens were heated to  $750 \text{ }^\circ\text{C}$  at a constant heating rate ( $1 \text{ }^\circ\text{C}/\text{min}$ ) in a stream of  $\text{N}_2$  ( $80 \text{ cm}^3/\text{min}$ ). The fuel gases were mixed with a stream of oxygen ( $20 \text{ cm}^3/\text{min}$ ), then entered a combustion furnace that was pre-set to  $900 \text{ }^\circ\text{C}$ .

## 3. Results and discussion

### 3.1. Synthesis of P-polyol

The chemical structure of the P-polyol synthesized by step polymerization between phenolphosphonic dichloride and ethylene glycol was characterized by FT-IR and NMR (Fig. 3). FT-IR:  $970\text{--}1100 \text{ (m, P-O)}$ ,  $1180\text{--}1300 \text{ (w, P=O)}$ ,  $1430\text{--}1500 \text{ (w, aromatic C=C)}$ ,  $2800\text{--}3100 \text{ (m, aromatic and aliphatic C-H)}$ ,  $3200\text{--}3600 \text{ (w, -CH}_2\text{OH)}$   $\text{cm}^{-1}$ .  $^1\text{H NMR}$  (300 MHz,  $\text{DMSO-}d_6$ ):  $\delta = 3.5\text{--}3.6 \text{ (2H, -CH}_2\text{OH)}$ ,  $3.9\text{--}4.0 \text{ (2H, -CH}_2\text{CH}_2\text{OH)}$ ,  $4.0\text{--}4.2 \text{ (4H, P-O-CH}_2\text{CH}_2\text{-O-P)}$ ,  $7.4\text{--}7.8 \text{ (5H, aromatic C-H)}$ .  $^{31}\text{P NMR}$  (300 MHz,  $\text{DMSO-}d_6$ ):  $\delta = 18.5$ .

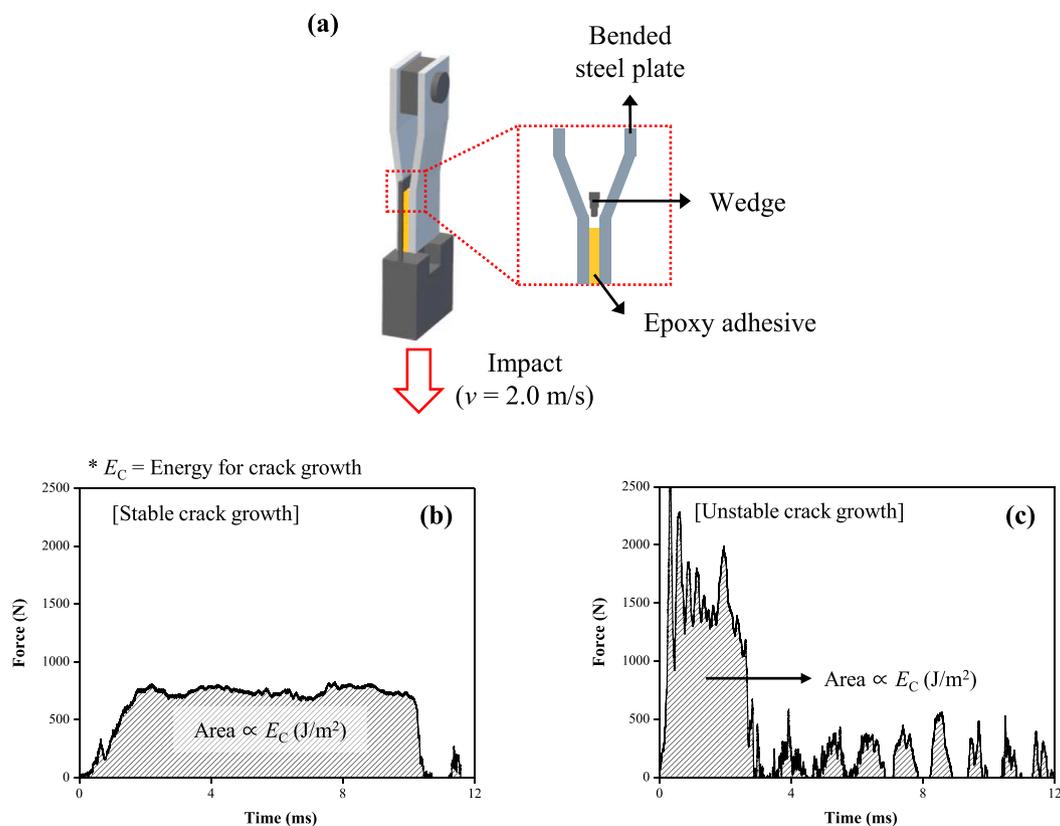


Fig. 2. (a) Test scheme and results of impact wedge-peel test: time-force curves for (b) stable crack growth and (c) unstable crack growth. (The figure was originally generated from Ref. [5]).

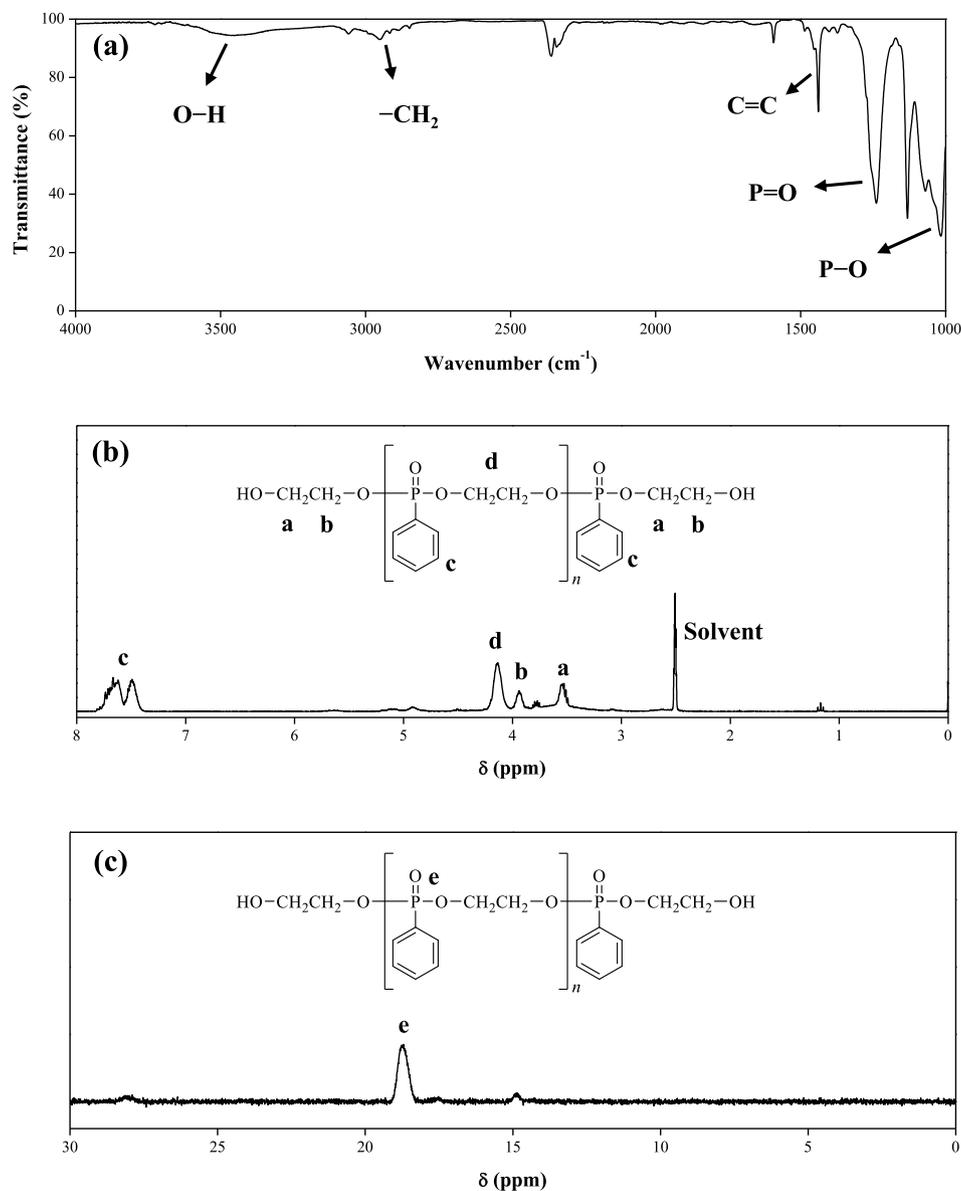


Fig. 3. (a) FT-IR, (b)  $^1\text{H}$  NMR, and (c)  $^{31}\text{P}$  NMR results of P-polyol.

From the  $^1\text{H}$  NMR results, the degree of polymerization ( $n$ ) and absolute molecular weight ( $Mn$ ) of P-polyol were calculated by the following equations:

$$n = \frac{8}{5} \frac{N_c}{N_a + N_b} - 1 = 3.44, \quad (1)$$

$$Mn \text{ (g/mol)} = 184 \times n + 246 = 880, \quad (2)$$

where  $N_a$  (= 1.00),  $N_b$  (= 1.02), and  $N_c$  (= 5.62) are the mean intensities of peaks a, b, and c, respectively (Fig. 3 (b)).

The OH value and  $Mn$  of the P-polyol were calculated by the following equations:

$$\text{OH value (mgKOH/g)} = \frac{(EP_2 - EP_1) \times N \times 56.1}{W} = 122.15, \quad (3)$$

$$Mn \text{ (g/mol)} = \frac{F \times 56100 \text{ (mgKOH/mol)}}{\text{OH value (mgKOH/g)}} = 919, \quad (4)$$

where  $N$  (= 0.1),  $EP_1$  (= 1.6807 ml),  $EP_2$  (= 3.7448 ml),  $W$  (= 0.0948 g), and  $F$  (= 2) are the normality of the titrant, titration volume at the first

peak of the 1st derivative, titration volume at the second peak of the 1st derivative, sample weight, and functionality of the sample, respectively.

The calculated  $Mn$  of P-polyol from the OH value (919 g/mol) was similar to that obtained from the  $^1\text{H}$  NMR results (880 g/mol).

### 3.2. Curing behavior

Because the curing of an epoxy adhesive is an exothermic reaction, exothermic peaks were obtained for all samples in the DSC results (Fig. 4 (a)). All samples were cured in a similar temperature range (170–180 °C), and the exothermic peak broadened upon incorporation of P-polyol. This indicated that the curing reaction between P-polyol and the epoxy system occurred in a broad temperature range. As shown in Fig. 4 (b), the reaction enthalpy decreased steadily with increasing amounts of incorporated P-polyol. The molecular weight of P-polyol is higher than that of the epoxy resin, and thus the reaction enthalpy per specific mass decreased with increasing addition of P-polyol.

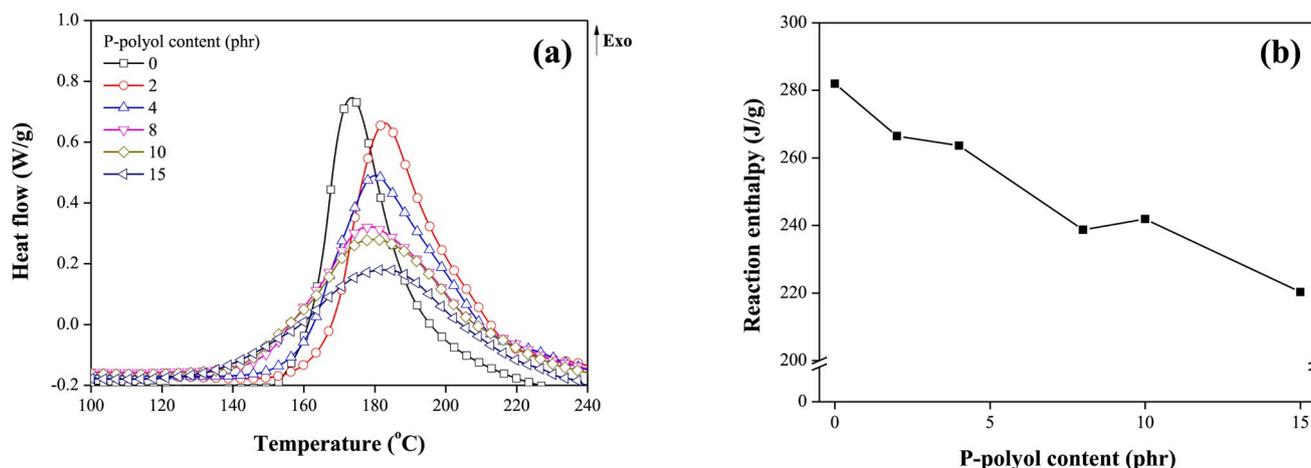


Fig. 4. (a) DSC curves and (b) reaction enthalpy of epoxy adhesives containing P-polyol.

### 3.3. Viscoelastic and adhesion properties of epoxy adhesive containing P-polyol

An epoxy adhesive is a thermosetting polymer material and thus has viscoelastic behavior. As shown in Fig. 5, the viscoelastic properties (storage modulus and  $\tan \delta$ ) of the epoxy adhesives containing P-polyol were characterized by DMA. At the same temperature, the storage modulus decreased with increasing addition of P-polyol (Fig. 5 (a)). Compared with the epoxy resin, which incorporates many aromatic rings, P-polyol has a relatively flexible chain, which lowered the storage modulus of the epoxy adhesive.

The  $\tan \delta$  peak shifted to lower temperatures with increasing P-polyol addition, but there was no significant change in peak height (Fig. 5 (b)). This indicated that the addition of P-polyol had little effect on the damping of the epoxy adhesive.

The glass transition temperature ( $T_g$ ) of the epoxy adhesive decreased with increasing addition of P-polyol (Fig. 6 (a)). Whereas the epoxy adhesive had a relatively high  $T_g$  due to the incorporation of numerous aromatic rings, P-polyol lowered the  $T_g$  of the epoxy adhesive due to its flexible chain.

The crosslink density of a polymer can be estimated by the following equations:

$$M_c = \frac{\rho RT}{G_r} \tag{5}$$

$$E_r = 2G_r(1 + \nu) \tag{6}$$

where  $\rho$  ( $\text{g}/\text{cm}^3$ ),  $G_r$  (MPa),  $R$  ( $\text{J}/(\text{K}\cdot\text{mol})$ ),  $T$  (K),  $E_r$  (MPa), and  $\nu$  are the density of the polymer ( $1.1 \text{ g}/\text{cm}^3$ , assumed to be equal for all samples), shear storage modulus at a rubbery state, real gas constant ( $8.314 \text{ J}/(\text{K}\cdot\text{mol})$ ), temperature, flexural storage modulus at the plateau region, and Poisson's ratio (0.5, assumed to be same as that of rubber), respectively [28].

The value of  $M_c$  represents the molecular weight between two crosslink points, and thus an increase in  $M_c$  indicates a decrease in crosslink density. As the P-polyol content increased,  $M_c$  of the epoxy adhesive increased, which showed that the crosslink density decreased by the addition of P-polyol (Fig. 6 (b)). The molecular weight of P-polyol ( $\sim 900 \text{ g}/\text{mol}$ ) is higher than that of the bisphenol-A diglycidyl epoxy resin ( $\sim 400 \text{ g}/\text{mol}$ ), and thus incorporating P-polyol lowered the crosslink density of the epoxy adhesive.

The lap shear strength and elongation of the epoxy adhesives were improved by low contents of P-polyol but deteriorated at a high content (15 phr) (Fig. 7(a) and (b)). Components with a high polarity, such as phosphorus, could improve the interfacial adhesion [29–32]. As shown in Fig. 8, the fracture behavior changed from interfacial failure (0–4 phr) to cohesive failure (10–15 phr) with increasing P-Polyol content, indicating an improvement to interfacial adhesion via the incorporation of P-polyol. However, excess P-polyol (15 phr) led to the formation of many bubbles inside the adhesive layer, which indicated that hydroxyl group of P-polyol reacted with hydroxyl group of the resin so that gaseous  $\text{H}_2\text{O}$  was generated. These bubbles lowered the interfacial adhesion and cohesion of the epoxy adhesive, thereby decreasing the lap

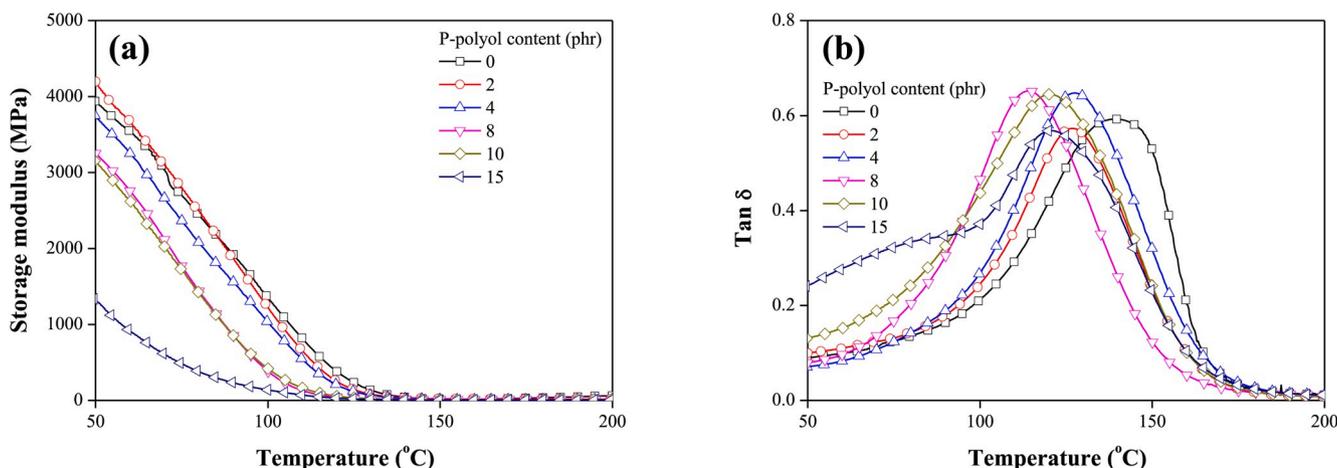


Fig. 5. DMA results of epoxy adhesives containing P-polyol: (a) storage modulus and (b)  $\tan \delta$ .

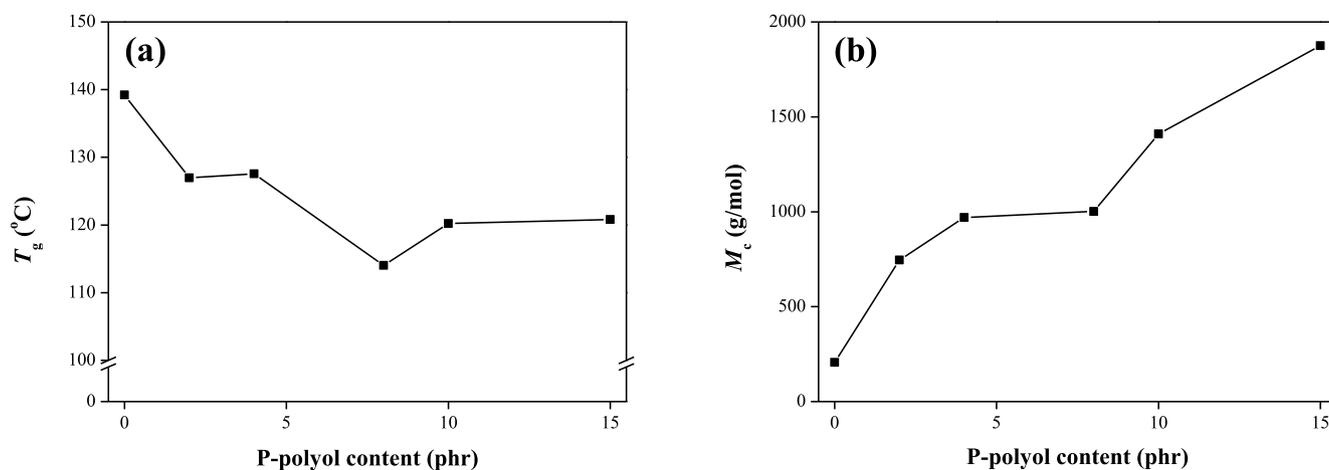


Fig. 6. Glass transition temperature ( $T_g$ ) and crosslink density ( $M_c$ ) of epoxy adhesives containing P-polyol.

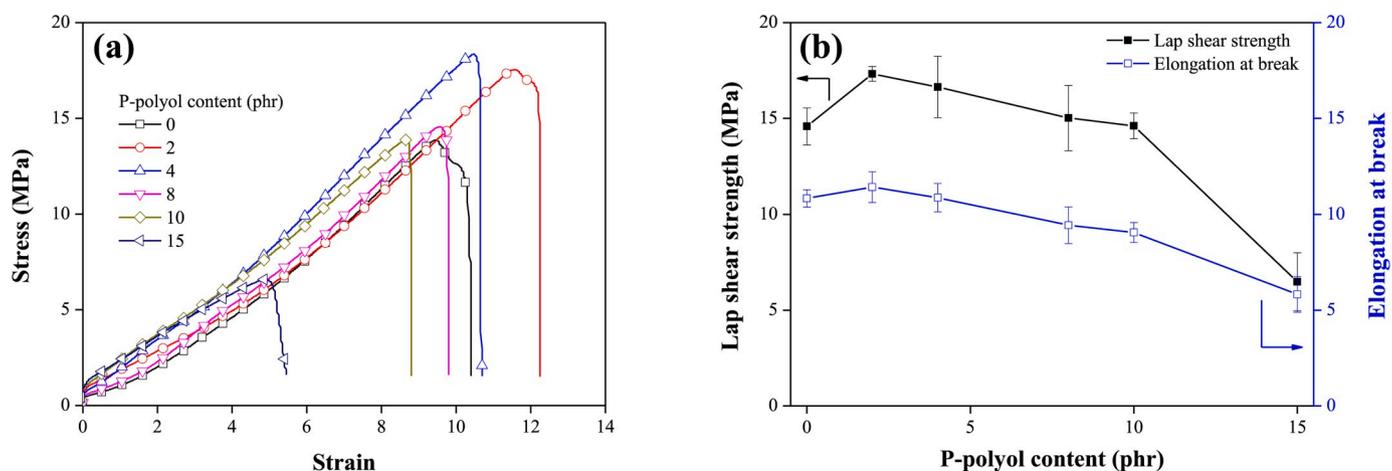


Fig. 7. Single lap shear test results: (a) stress-strain curves, (b) lap shear strength, and elongation at break.

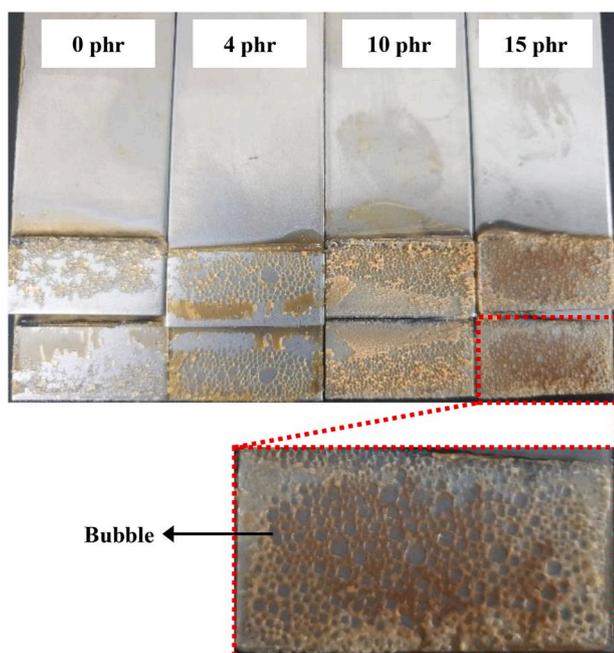


Fig. 8. Fracture surfaces from single lap shear tests.

shear strength and elongation.

### 3.4. Impact-resistant property of epoxy adhesives containing P-polyol

During impact wedge-peel testing, resistant force is measured for few milliseconds when an epoxy adhesive is cleaved by impact. As shown in Fig. 9 (a), in the case of low-impact-resistant epoxy adhesives (0, 2, 4, and 15 phr), the resistant force was maintained for less than 6 ms. On the other hand, the resistant force of high-impact-resistant epoxy adhesives was maintained for over 10 ms (8 and 10 phr). Similarly, the energy for crack growth ( $E_C$ ), which was obtained by integrating the force-time curve, increased at 8 and 10 phr but decreased at 15 phr.

As the impact resistance improved by the addition of P-polyol, the type of crack growth changed from unstable to stable (Table 2). However, excess P-polyol lowered the impact resistance of the epoxy adhesive, which caused unstable crack growth. As shown in Table 2, the dynamic resistance to cleavage (average force during cleavage) could only be obtained for the high-impact-resistant epoxy adhesives (8 and 10 phr). The average resistant force during unstable crack growth could not be compared with that during stable crack growth, and thus dynamic resistance to cleavage was not obtained for the low-impact-resistant epoxy adhesives (0, 2, 4, and 15 phr).

Incorporating an optimum amount of P-polyol improved the interfacial adhesion and lowered the brittleness of the epoxy adhesive by lowering the crosslink density, which enhanced the impact resistance. On the other hand, excess P-polyol generated many bubbles in the

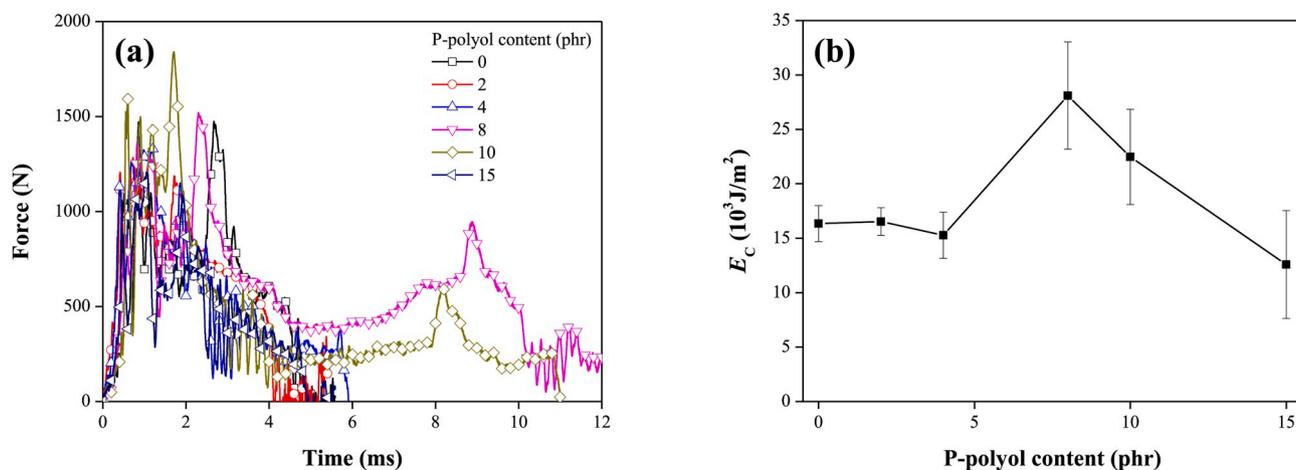


Fig. 9. Impact wedge-peel test results of epoxy adhesives containing P-polyol: (a) time-force curves and (b) energy for crack growth ( $E_C$ ).

Table 2

Type of crack growth and dynamic resistance to cleavage of epoxy adhesives containing P-polyol.

P-polyol content (phr)	Type of crack growth (Unstable/stable)	Dynamic resistance to cleavage (N/mm)
0	Unstable	–
2	Unstable	–
4	Unstable	–
8	Stable	16.9 ( $\pm 3.0$ )
10	Stable	14.2 ( $\pm 2.0$ )
15	Unstable & stable	–

adhesive layer and lowered the storage modulus, causing a deterioration in impact resistance.

### 3.5. Flame retardancy of epoxy adhesives containing P-polyol

The flame retardancy of the epoxy adhesives was characterized with TGA and PCFC. As shown in Fig. 10 (a), in the high-temperature region (600–800 °C), the decomposition curves shifted to higher temperatures with increasing P-polyol content. Similarly, char yield increased with P-polyol addition (Fig. 10 (b)), which resulted in improved flame retardancy. The decomposition temperature ( $T_{50}$ , temperature at 50% weight) decreased upon the incorporation of P-polyol, which has an aliphatic hydrocarbon chain.

According to the PCFC results, the heat release rate decreased with increasing P-polyol content (Fig. 11 (a)). Total heat release (area under the temperature-heat release rate curve) also decreased (Fig. 11 (b)), which represented an improvement in flame retardancy. Similar to the

TGA results, decomposition temperature (temperature at the maximum heat release rate) decreased by incorporating P-polyol (Fig. 11 (b)).

Based on the PCFC results, the heat release capacity, which is the best parameter for judging flame retardancy, can be calculated by dividing the maximum heat release rate by the heating rate [33–35]. As shown in Table 3, the heat release capacity steadily decreased with increasing P-polyol content, which supported that the addition of P-polyol improved the flame retardancy of the epoxy adhesives.

## 4. Conclusions

We synthesized a phosphorus-containing polyol (P-polyol) as a reactive flame retardant by step polymerization between phenylphosphonic dichloride and ethylene glycol ( $M_n \sim 900$  g/mol). With increasing addition of P-polyol, the glass transition temperature decreased and  $M_c$  (molecular weight between crosslink points) increased. As a function of P-polyol content, the lap shear strength first increased due to the high polarity of the phosphorous component, then ultimately decreased at excess P-polyol content due to the byproduct of the curing reaction ( $H_2O$ ). The flame retardancy of the epoxy adhesives improved by the incorporation of P-polyol, which was confirmed by an increase in char yield and decrease in heat release capacity. Impact wedge-peel testing was performed to characterize the impact resistance of a complete adhesive joint. Impact resistance also improved with the addition of moderate amounts of P-polyol (8, 10 phr); however, it decreased sharply at a high P-polyol content (15 phr).

Based on the results of this study, it is possible to simultaneously improve the impact resistance and flame retardancy of a structural epoxy adhesive by using P-polyol as a reactive flame retardant. Furthermore, we determined the optimum P-polyol content (8, 10 phr)

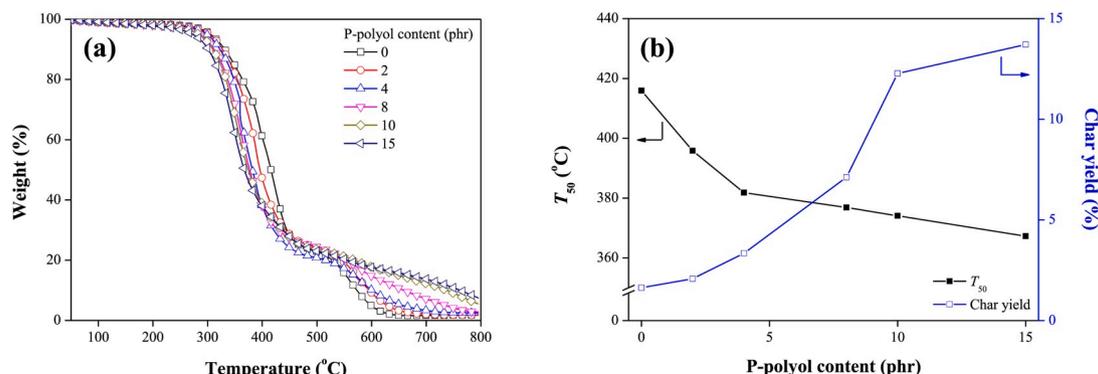


Fig. 10. TGA results of epoxy adhesives containing P-polyol in an air atmosphere: (a) temperature-weight curves, (b)  $T_{50}$ , and char yield at 700 °C.

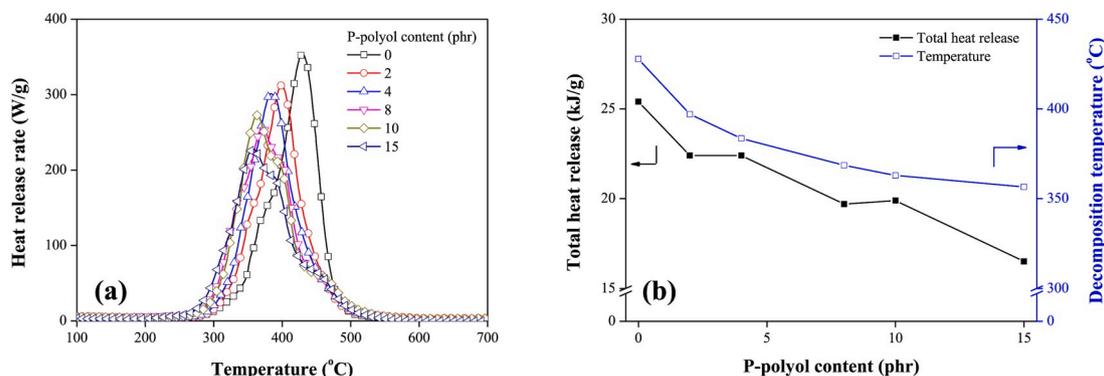


Fig. 11. PCFC results of epoxy adhesives containing P-polyol: (a) heat release rate curves, (b) total heat release, and decomposition temperature.

Table 3

Heat release capacity of epoxy adhesives containing P-polyol.

P-polyol content (phr)	Heat release capacity (J/(g·°C))
0	354.01
2	312.25
4	301.27
8	255.44
10	272.58
15	227.73

for obtaining both a high impact resistance and flame retardancy. However, we synthesized the reactive flame retardant by incorporating hydroxyl as a functional group, and thus, during the curing reaction, H<sub>2</sub>O was generated as a byproduct, which caused a decrease in mechanical strength. In future work, other functional groups such as amine and glycidyl should be incorporated to prevent this reduction in mechanical strength.

#### Declaration of competing interest

There are no conflicts of interest to declare.

#### Acknowledgements

This work was supported by the World Class 300 project (R&D) [grant number S2483588] of the SMBA (Korea) and by the Korea Research Institute of Chemical Technology [grant number SS2041-10].

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