Curing Behavior and Dielectric Properties of Hyperbranched Poly(phenylene oxide)/Cyanate Ester Resins

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ABSTRACT: A novel hyperbranched poly(phenylene oxide) (HBPPO) modified 2,2′-bis(4-cyanatophenyl) isopropylidene (BCE) resin system with significantly reduced curing temperature and outstanding dielectric properties was developed, and the effect of the content of HBPPO on the curing behavior and dielectric properties as well as their origins was thoroughly investigated. Results show that BCE/HBPPO has significantly lower curing temperature than BCE owing to the different curing mechanisms between the two systems, the difference also brings different crosslinked networks and thus dielectric properties. The dielectric properties are frequency and temperature dependence, which are closely related with the content of HBPPO in the BCE/HBPPO system. BCE/2.5 HBPPO and BCE/5 HBPPO resins have lower dielectric constant than BCE resin over the whole frequency range tested, while BCE/10 HBPPO resin exhibits higher dielectric constant than BCE resin in the low frequency range ($<10^4$ Hz) at 200°C. At 150°C or higher temperature, the dielectric loss at the frequency lower than 10^2 Hz becomes sensitive to the content of HBPPO. These phenomena can be attributed to the molecular relaxation. Two relaxation processes (α- and β-relaxation processes) are observed. The β-relaxation process shifts toward higher frequency with the increase of temperature because of the polymer structure and chain flexibility; the α-relaxation process appears at high temperature resulting from the chain-mobility effects. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 121: 2113–2122, 2011

Key words: thermosets; dielectric properties; structure–property relation; hyperbranched poly(phenylene oxide)

INTRODUCTION

Cyanate ester (CE) resin, as one of the most important kinds of thermosetting resins, has been considered as the good candidate with great potentialities for developing high-performance functional/structural materials in the 21st century owing to its outstanding integrated performance.[1,2] However, no polymer is perfect to meet all requirements of actual applications, one of the biggest disadvantages of CE resin is its high curing temperature, which is generally easy to produce cured resins with big internal residual stress and thus resulting in poor stability of materials in service performance.[3] Therefore, how to overcome the curing disadvantage of CE has been a key issue since the invention of CE resins.

As we know that the biggest advantage of CE resin is its excellent dielectric properties including extremely low and stable dielectric constant and loss over wide frequency and temperature ranges, hence how to modify CE resin without scarifying this key merit is an essential aspect. However, unfortunately, this point is often neglected, that is, many literatures focused on modifying the drawback of CE resin, but seldom considered how to maintain the excellent dielectric properties.

Polyphenylene oxide (PPO) is one engineering polymer, which has been widely used in many cutting-edge industries, especially in the microelectronic industry (e.g., as the matrix for high-performance copper clad broads) owing to its excellent dielectric properties (extremely low-dielectric constant and loss), outstanding moisture and thermal resistance, and desirable toughness[4,5] However, the investigation on PPO/CE system is seldom found in literature. Wu's[6] group conducted some representative work. Specifically, they studied the curing kinetics of a blend made up of a multifunctional CE (PT-30) and PPO and found that there is an autocatalytic

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curing mechanism in the blend.[6] The similar phenomenon is also found in the CE/epoxy/PPO system.[7,8] But dielectric properties of these PPO modified systems were not discussed.

It is worth noting that PPO has high melting point and big melting viscosity, while CE will cure with the increase in temperature; hence, some solvents are generally used to prepare the PPO/CE blend to avoid the early appearance of gelation or even curing and then that must be driven off. Obviously, it is a complex and energy-consuming process; more importantly, it is generally not easy to completely remove the solvent from a viscous liquid. To overcome these drawbacks of common PPO, Edward's group synthesized unique PPO telechelic copolymers (PPOTC), which were then used to modify CE.[9] Results show that PPOTC can react with CEs to form a single-phase thermoset material with improved toughness, moisture-resistance, and decreased dielectric constant. However, the synthesis of PPOTC is so complex that is still far away from industrial production, and so the PPOTC/CE system is difficult to be applied on a large scale in the near future.

Hyperbranched polymers are considered as a kind of promising alternative modifiers for high-performance thermosets because of their unique structure and desirable properties including high solubility, low viscosity, and big reactivity compared to their linear analogues.[10,11] Hyperbranched poly(phenylene oxide) (HBPPO) is expected to inherit not only the outstanding integrated properties of linear poly(phenylene oxide) such as high thermal stability, good mechanical properties, low moisture uptake, and very good dielectric properties (low dielectric constant and loss), but also that of hyperbranched polymers.[12–14]

There are two aims of this work, one is to develop a new modified CE resin system, which has decreased curing temperature meanwhile remaining very good dielectric properties compared to original CE resin; the other is to systematically investigate the dielectric properties and their origins of the modified CE resin system and thus make new contribution to developing high performance polymers.

EXPERIMENTAL

Materials

2,2′-Bis(4-cyanatophenyl) isopropylidene (BCE) was purposed from Shangyu Chemical of Zhejiang in China. Dimethyl sulfoxide (DMSO), hexamethylene, anhydrous toluene, methanol (MeOH), and tetrahydrofuran (THF) were all commercial products (analysis grade) without further purification before use.

HBPPO was synthesized according to the procedure described in literature.[15] A typical procedure is described herein. 4-Bromo-4′,4″-dihydroxytriphenylmethane (4.0 g, 0.0113 mol), K_2CO_3 (1.5 g, 0.0107 mol) in the mixture of DMSO (160 mL), and anhydrous toluene (40.0 mL) were heated to 115–120°C and maintained in that temperature range for 3 h. After cooling to 45°C, CuCl (0.02 g, 0.0204 mmol) was added into this reaction mixture. The reaction mixture was heated under 170°C ± 2°C for 36 h and cooled to room temperature. After slow addition of cold acidic water, the reaction mixture was precipitated into the mixture of MeOH and water. The crude polymer was filtered and dried *in vacuo*, followed by dissolved in THF and filtered. After reprecipitation into hexamethylene, the resultant was filtered and dried in a vacuum oven to get the target product. The yield was 68% ± 2%.

Preparation of cured BCE resin

2,2′-Bis(4-cyanatophenyl) isopropylidene (BCE) was put into a flask equipped with a mechanical stirrer and thermometer at 120°C with stirring till a clear liquid was obtained. The resultant liquid was put into a preheated mold followed by degassing at 120°C for 30 min in a vacuum oven, and then the mold was moved into an oven for curing via the procedure of 130°C/2 h + 150°C/2 h + 180°C/2 h + 220°C/2 h. The resultant resin was demolded and then postcured at 230°C for 4 h, successively, coded as BCE.

Preparation of cured BCE/HBPPO resins

2,2′-Bis(4-cyanatophenyl) isopropylidene (BCE) was placed in a flask equipped with a mechanical stirrer and thermometer at 120°C with stirring till a clear liquid was obtained. Then preweighted hyperbranched poly(phenylene oxide) (HBPPO) was added into the flask with stirring. The mixture was maintained at 120°C for 30 min to obtain a brown–red transparent liquid. The resultant liquid was put into a preheated mold followed by degassing at 120°C for 30 min in a vacuum oven, and then the mold was moved into an oven for curing and postcuring via the procedures of 130°C/2 h + 150°C/2 h + 180°C/2 h + 220°C/2 h, and 230°C/4 h, successively. The resultant resin was coded as BCE/nHBPPO, where *n* represents the weight loading of HBPPO.

Measurements

Fourier transform infrared (FTIR) spectra were measured using a Nicolet (USA) FTIR spectrometer. The specimen was prepared by coating the sample on a potassium bromide disk and subsequently heated in the oven.

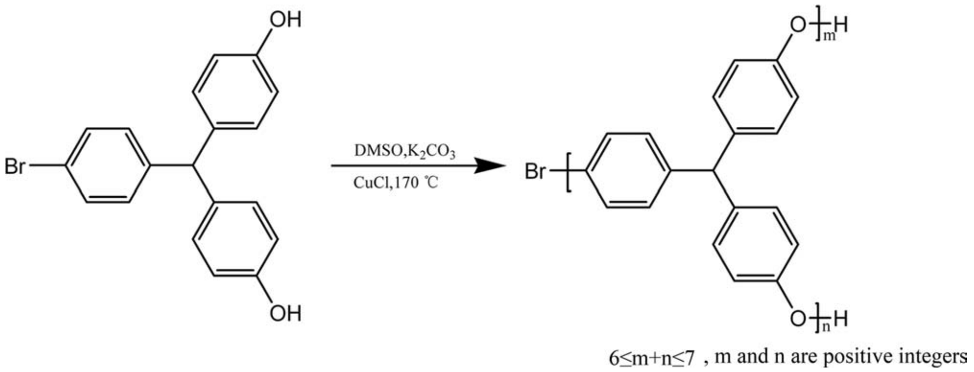

Figure 1 Synthesis of HBPPO.

Nuclear magnetic resonance (NMR) spectra were recorded on a Unity Inova 400 NMR spectrometry (USA), and DMSO-d_6 was used as the solvent.

Gel permeation chromatography (GPC) was carried out on a Waters GPC 515–2410 system (USA) equipped with a RI detector and packing column (Waters Styragel HT3-5-6E). THF was used as the elution solvent.

Differential scanning calorimeter (DSC) was recorded using a TA calorimetry (2910 MDSC, USA) from room temperature to 300°C with a heating rate of 15°C/min under nitrogen atmosphere.

The fractured surfaces were observed using a Hitachi S-4700 (Japan) scanning electron microscope (SEM). The BCE/HBPPO samples were etched in THF for 24 h and then washed by ethanol and distilled water, successively. After that, the resultant samples were air dried.

Dynamic mechanical analyses (DMA) were conducted using TA DMA Q800 apparatus from TA Instruments (USA). A single cantilever clamping geometry was used. DMA tests were carried out from room temperature to 350°C with a heating rate of 3°C/min at 1 Hz. The glass transition temperature (T_g) is defined as the peak (maximum) temperature in the tan δ-temperature plot. The dimensions of each sample were (35 ± 0.1) × (12 ± 0.1) × (2 ± 0.1) mm^3.

The dielectric properties were measured using a Broadband Dielectric Spectrometer (Novocontrol Concept 80 analyzer, Germany) at the frequency between 1 and 10^6 Hz and a temperature range from −50 to 200°C. The dimensions of each sample were (20 ± 0.1) × (20 ± 0.1) × (2 ± 0.1) mm^3.

RESULTS AND DISCUSSION

The structure of HBPPO

Figure 1 shows the molecular structure of hyperbranched poly(phenylene oxide) (HBPPO), of which the repeat unit consists of three phenyl groups which symmetrically arrange around a central methylidyne carbon atom. In addition, numerous terminal phenolic groups exist in the molecule of HBPPO. All these groups can be reflected in the spectra. Figure 2 gives the FTIR spectrum of HBPPO. The obvious absorption peak at 3420 cm^{-1} is assigned to the stretching vibration of hydroxyl group, the broad peaks at ~ 1263 and 823 cm^{-1} belong to the stretching vibration Ph—O—Ph and Ph—H, respectively. The structure of HBPPO was further analyzed by ^{1}H-NMR spectroscopy, as shown in Figure 3. It can be observed that multiplet resonances at ~ 5.20, 7.0, and 9.3ppm attribute to the proton in methine, phenyl ring, and terminal phenolic group, respectively.

The weight average molecular mass (M_w) of HBPPO determined by GPC is 2200–2300, and so the value of "$m + n$" is calculated to be 6 or 7.

Curing behavior and cured structure of BCE/HBPPO system

For a thermosetting system, its curing behavior determines the structure, and thus the performance of cured resin, and so it is necessary to first investigate the effect of hyperbranched poly(phenylene

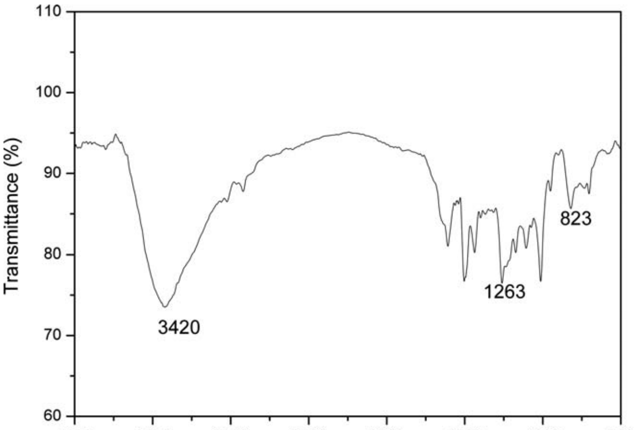

Figure 2 The FTIR spectrum of HBPPO.

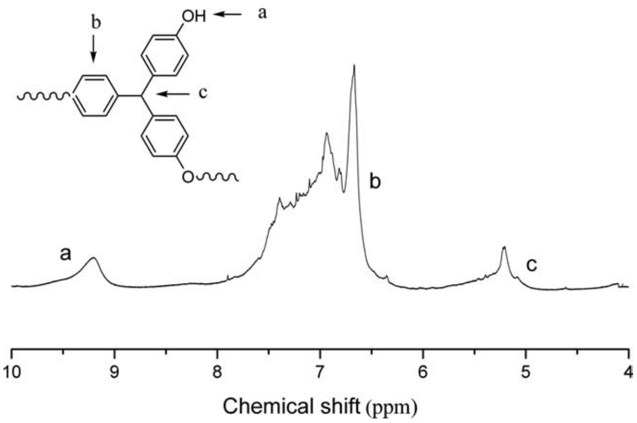

Figure 3 The ^{1}H-NMR spectrum of HBPPO.

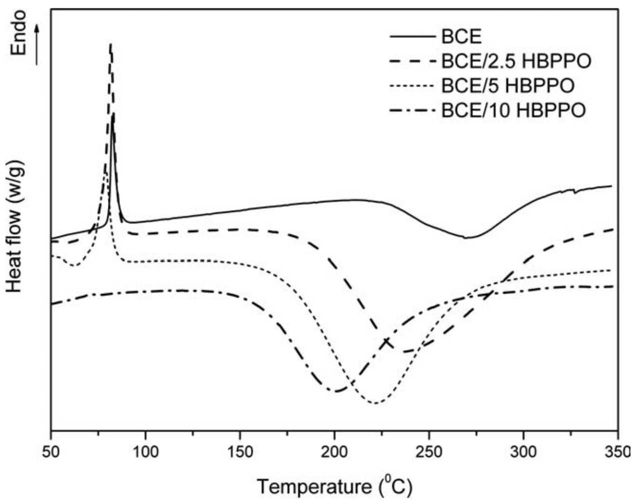

Figure 4 DSC curves of BCE and BCE/HBPPO systems.

oxide) (HBPPO) on the curing behavior and cured structure of the resultant resin.

Figure 4 shows DSC scans of BCE and BCE/HBPPO system. Each sample has one exothermic peak; however, compared to the peak of BCE, the whole peak of BCE/HBPPO system appears at significantly lower temperature. In the case of the maximum exothermic temperature, it shifts from about 272°C to 235, 221, or 200°C when 2.5, 5, or 10 wt % HBPPO is added to BCE, respectively, suggesting that the presence of HBPPO can significantly decrease the curing temperature of BCE. This is easy to be understood, because terminal-phenolic hydroxyl not only has evidently catalytic effect on the homopolymerization of BCE but also can copolymerize with —OCN group.[16–18]

The curing mechanism of BCE/HBPPO system is complicated,[18] which contains four pathways as shown in Figure 5. The critical first step is the formation of an iminocarbonate via a reaction between a CE (—OCN) group and a phenolic hydroxyl (—OH) group. It is worth noting that the chemical

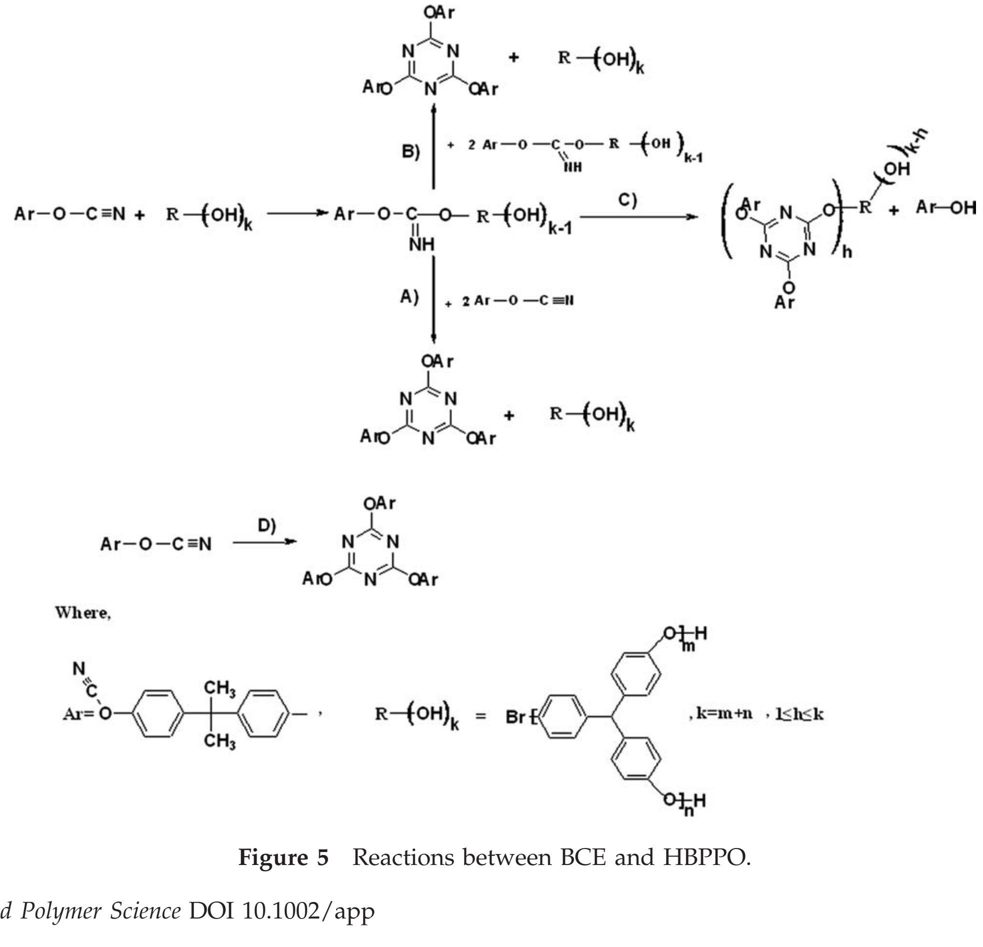

Figure 5 Reactions between BCE and HBPPO.

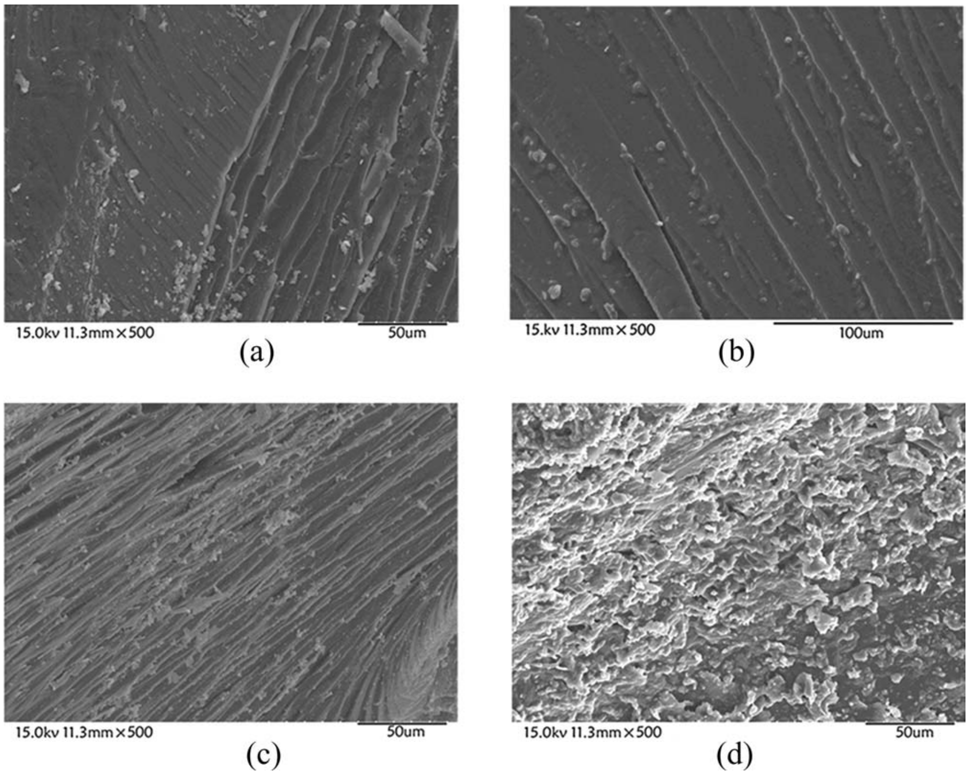

Figure 6 SEM micrographs of the fracture surfaces of cured resins: BCE (a), BCE/10 HBPPO (b), etched BCE/5 HBPPO (c), and etched BCE/10HBPPO (d).

bonding (Pathway C) between BCE and HBPPO not only brings a good miscibility between HBPPO and BCE resin but also leads to a good dispersion of HBPPO in the BCE resin, this can be further confirmed by the SEM images (Fig. 6) of the fractured surfaces for cured BCE/5 HBPPO and BCE/10 HBPPO resins after etched in THF for 24 h. It can be found that the HBPPO phase (white part) is homogenously dispersed in the BCE resin phase (black part); meanwhile, the domain of HBPPO in BCE/5 HBPPO resin is smaller than that in BCE/10 HBPPO resin.

DMA analysis measures the modulus (stiffness) and damping (energy dissipation) properties of materials as they are deformed under periodic stress. It detects essentially all changes in the state of molecular motion as temperature is scanned,[19] and so DMA is used to get more information about the crosslinked structure of BCE/HBPPO system.

From the overlay storage modulus–temperature plots as shown in Figure 7, the crosslinking density (degree) of each resin can be determined according to the rubber elasticity theory as eq. (1)[20]:

$$\rho = G'/3RT \tag{1}$$

where G' is the storage modulus of the cured resin at the temperature T ($T = T_g + 30°C$) and R is the gas constant.

The calculated values are summarized in Table I. BCE/HBPPO resins have significantly smaller crosslinking density than cured BCE resin; in addition, interestingly, BCE/2.5 HBPPO and BCE/5 HBPPO resins show similar crosslinking densities, which are much larger than the corresponding value of BCE/10 HBPPO resin, suggesting that BCE/10 HBPPO resin has special crosslinked structure compared to other two BCE/HBPPO resins. These results can be interpreted by the curing mechanism. As shown in Figure

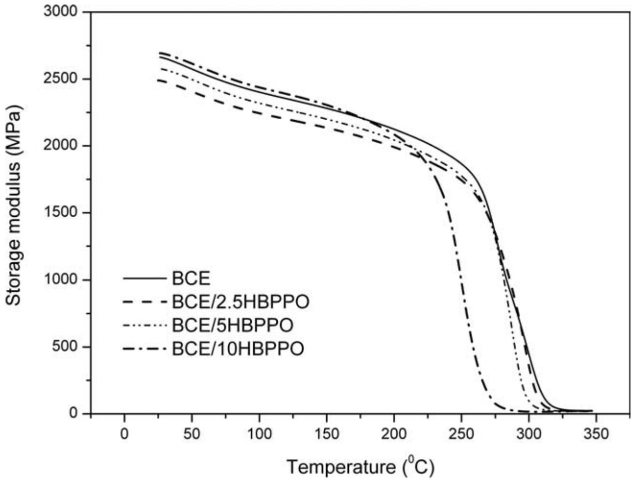

Figure 7 Overlay plots of storage modulus versus temperature for cured BCE and BCE/HBPPO resins.

TABLE I
Crosslinking Densities and T_g Values of Cured BCE and BCE/HBPPO Resins

Resin	T_g (°C)	ρ (mol/m^3)
BCE	312	4.68
BCE/2.5 HBPPO	308	4.10
BCE/5.0 HBPPO	298	4.08
BCE/10 HBPPO	273	3.23

3, Pathway C forms two products, one is a phenolic derivative of the original CE monomer, which is thought to be equally active in promoting further reactions; the other is triazine ring with trapped PPO chains, which can cause chain-termination reactions, and thus resulting in decreased crosslink density. The more is phenolic hydroxyl groups, the smaller is the crosslinking density of the modified BCE resin. Because BCE/2.5 HBPPO and BCE/5 HBPPO resins have much less content of phenolic hydroxyl groups than BCE/10 HBPPO resin, therefore, the former has larger crosslinking density than the latter.

Figure 8 shows the dependence of tan δ on temperature for cured BCE and BCE/HBPPO resins, it can be seen that each system has a single and sharp tan δ peak, which gradually shifts toward lower temperature with the addition of HBPPO to BCE resin as well as the increase of the content of HBPPO in the modified system, suggesting that compared to BCE resin, BCE/HBPPO resin tends to have decreased T_g value. This phenomenon is attributed to the introduction of flexible Ph—O—Ph chains and decreased amount of triazine network.

It is known that the shape of tan δ peak may be used as a convenient indicator of the morphological state of the crosslinked networks. Compared to cured BCE resin, although only one damping peak can be observed for all resins, the tan δ peak of BCE/10 HBPPO resin becomes broader, suggesting an increasing possibility of the existence of more than one phase in the network of BCE/10 HBPPO resin. However, the observed predominant single damping peak in DMA spectrum indicates that BCE/10 HBPPO resin is still molecularly homogeneous at least up to the scale of the dynamic mechanical test. In other words, the fact that the T_g of the investigated samples is well defined, and only slightly broadened shape indicates that the phase domains, if exist, are smaller than the size of the segments that are responsible for the primary molecular relaxation.

Dielectric properties

Frequency and temperature dependence of dielectric constant

The frequency dependence of dielectric constant for cured BCE and BCE/HBPPO resins at different temperatures is shown in Figure 9. At a given temperature lower than 200°C, each resin shows similar and good stability of dielectric constant over the whole frequency range; while with the increase of temperature, dielectric constant gradually increases.

Note that when the temperature sequentially increases to 200°C, the dielectric constant of each resin (especially BCE/10 HBPPO) becomes sensitive to frequency; that is, each resin has large dielectric constant at low frequency and then gradually decreases to a relative stable value with the increase of frequency. In the case of the magnitude of dielectric constant, significant difference mainly appears in the low frequency range ($<10^4$ Hz). Specifically, BCE/2.5 HBPPO and BCE/5 HBPPO resins show smaller dielectric constant than BCE resin over the whole frequency range, while BCE/10 HBPPO resin exhibits larger dielectric constant than BCE resin.

It is known that the dielectric constant of a polymer depends on the orientation and relaxation of dipoles of the polymer, and so the polarity and activity of dipoles of the polymer play a dominant role on determining the dielectric constant.[21] Because the movement of polymer chain segments is greatly dependent on the temperature, hence dielectric properties are significantly influenced by the temperature. When a polymer stays at a temperature much lower than T_g, the polar groups of the polymer are difficult to orient and relax in the applied electric field owing to the frozen molecular chain segments in glassy state. With the increase in temperature, the chain segment mobility increases, and polar groups begin to move in response to the applied electric field, leading to the increase of the orientation polarization, and thus the enlarged dielectric constant of the polymer. When the temperature approaches T_g, the dielectric constant increases dramatically, because the dipoles attached rigidly to

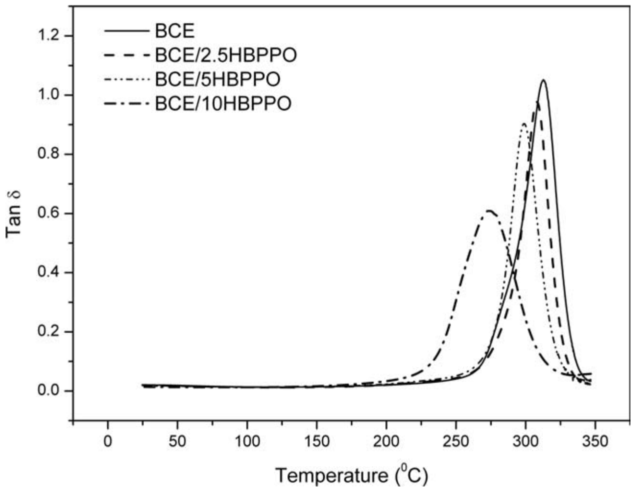

Figure 8 Overlay plots of tan δ versus temperature for cured BCE and BCE/HBPPO resins.

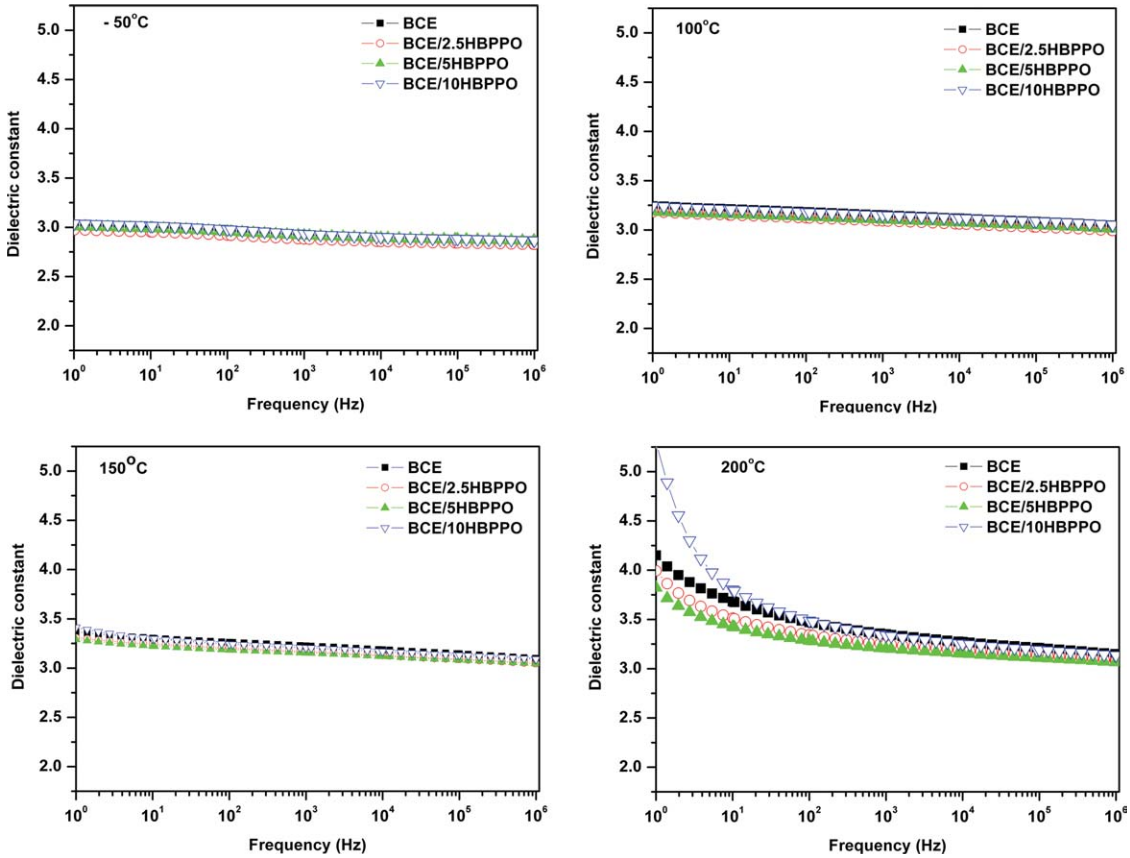

Figure 9 The dielectric constant as a function of frequency for cured BCE and BCE/HBPPO resins at different temperatures. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the polymer backbone gain sufficient mobility and become free to orient in an electric field.

Note that at a given temperature, the influence of temperature discussed above can be neglected, hence the dielectric constant is mainly dependent on the polarity of the polymer. The structure of HBPPO has low polarity and good symmetry, and so the addition of HBPPO to BCE resin is beneficial to decrease dielectric constant; however, on the other hand, the presence of HBPPO will also reduce the crosslinked degree as discussed earlier and thus tends to increase the dielectric constant. These two opposite effects play a combined role, which is related with the content of HBPPO as shown in Figure 9.

Based on the above discussion, it can be stated that the content of HBPPO almost does not affect the dielectric constant at lower temperature (e.g., ≤150°C), while which plays a significant role at higher temperature. This phenomenon is attributed to the difference of the molecular mobility at different temperatures.

Frequency and temperature dependence of dielectric loss

The overlay plots of dielectric loss versus frequency for cured BCE and BCE/HBPPO resins at different temperatures are shown in Figure 10. It can be observed that the dielectric loss of each resin is greatly temperature-dependent. When the temperature is not higher than 100°C, the dielectric loss is not dependent on the content of HBPPO, that is, all resins have similar values of dielectric loss; however, at 150°C or higher temperature, the dielectric loss at low frequency ($<10^2$ Hz) becomes sensitive to the content of HBPPO.

The dependence of dielectric loss on frequency is complicate, which is greatly related to the temperature. At −50°C, there is a broad upward peak in the range from 1 to 10^5 Hz; with the increase of the temperature, the peak turns narrow and shifts toward higher frequency. When the temperature increases to 100°C, the dielectric loss of each resin is almost independent on the frequency. Further increase the temperature to 150 or 200°C, the dielectric loss is large at low frequency, which decreases with the increase of frequency and then almost levels off. There phenomena can be explained from the molecular relaxation.

Relaxation process

From the view of molecular movement, the frequency and temperature-dependence of dielectric

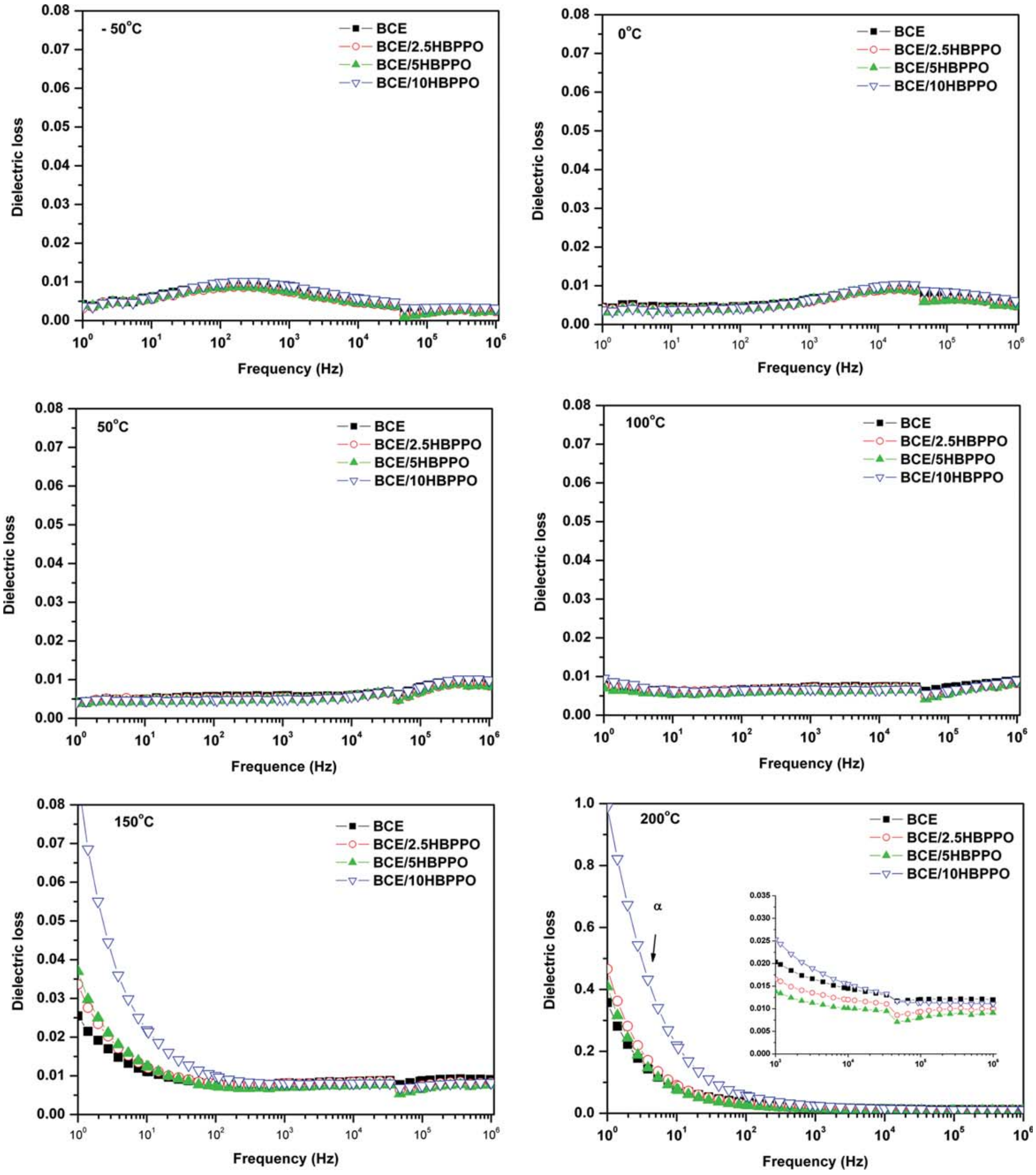

Figure 10 The dielectric loss as a function of frequency for cured BCE and BCE/HBPPO resins at different temperatures. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

loss reflect the molecular relaxation.[22,23] A polymer contains several molecular relaxation processes named as α, β, and γ. According to different relaxation mechanisms, α-relaxation process is related with the glass transition temperature, and this process invariably has a characteristic signature and is extremely broad for amorphous polymers[23]; β-relaxation process is associated to the molecular rearrangements[24] or local motions of dipolar groups.[22,25]

To confirm the temperature for each relaxation, the overlay plots of dielectric loss at 10^4 Hz versus temperature for cured BCE and BCE/HBPPO resins are depicted in Figure 11. There are two obvious relaxation processes, one appears in the temperature range from about −50 to 50°C, and the relaxation peak is about at 0°C, assigning to β-relaxation; the other occurs at about 200°C, attributing to α-relaxation process.

In the case of β-relaxation, the intensity of β-relaxation peak for each resin shows very small difference; however, with careful observation, which follows an order of BCE/10 HBPPO > BCE > BCE/5 HBPPO ≈ BCE/2.5 HBPPO. This phenomenon can be explained from the effect of HBPPO content on the structure of resultant resins. First, the curing mechanism of BCE/HBPPO resins points out that

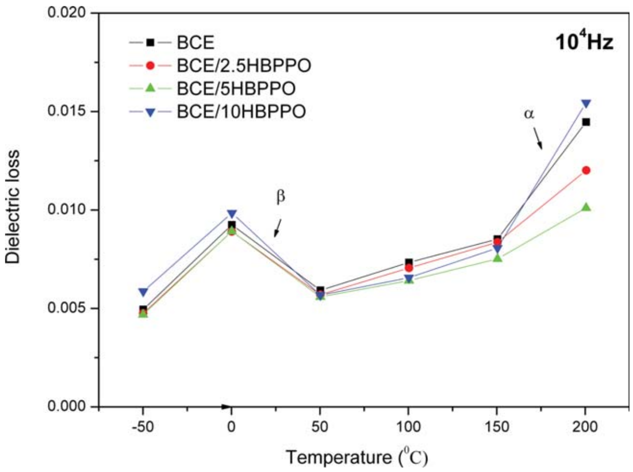

Figure 11 The dielectric loss as a function of temperature for cured BCE and BCE/HBPPO resins at 10^4 Hz. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

abundant —CH(C_6H_5) segments tend to enter into phenyl-triazine ether linkages through the coreaction between HBPPO and BCE, and these large-scale segmental chains can hinder the torsional rotation around the Ph—O bond and thus result in β-relaxation with lower intensity. Contrarily, second, the decreased crosslinked density of BCE/HBPPO resins also leads to increased intensity of β-relaxation; third, compared to BCE resin, there are a larger amount of polar groups and flexible bonds in BCE/HBPPO resins, which are beneficial to get increased β-relaxation. Obviously, these aspects counteract each other, resulting in a very small effect of HBPPO on the β-relaxation.

The α relaxation reflects the glass transition, hence when the temperature nears the T_g value (Table I), the dielectric loss significantly increases (especially at low frequency) as shown in Figure 8. Because the orientation of dipoles needs a longer time than electronic and ionic polarizations, meaning that with the increase of frequency, the orient polarization decreases, leading to decreased dielectric loss.

The magnitude of dielectric loss at high temperature follows the similar order as that of dielectric constant. Besides the factors attributing to the dielectric constant, there is additional effect resulting from the glass transition temperature. As shown in Table I that BCE/HBPPO resin has lower T_g value than BCE resin, and the larger is the content of HBPPO, the lower is the T_g value. The BCE/HBPPO resin with decreased T_g value tends to have bigger

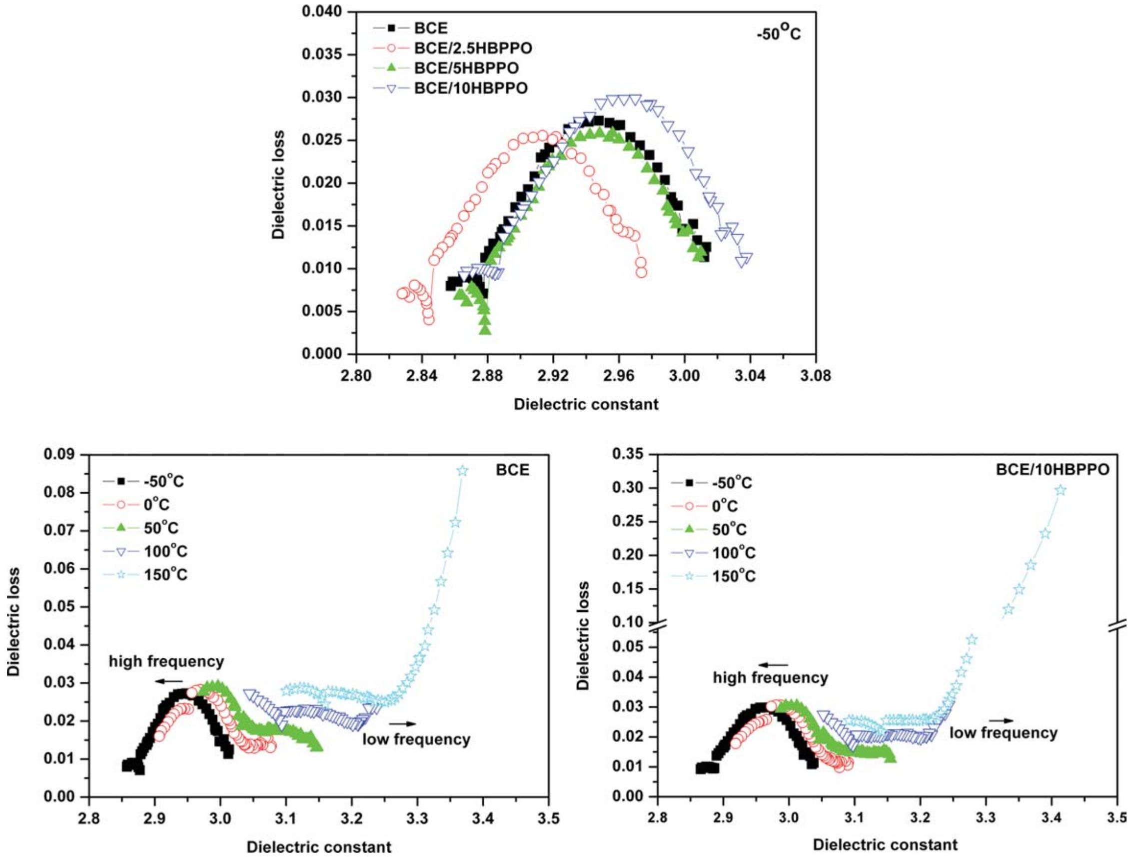

Figure 12 Cole–Cole diagrams for cured BCE and BCE/HBPPO resins at different temperatures. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

chain mobility at a given temperature near T_g value, resulting in larger dielectric loss.

The Cole–Cole equation can be used to study the relaxation processes in polymer materials having local dipolar motions at the monomer or unit cell scale. Figure 12 presents the Cole–Cole diagrams of cured BCE and BCE/HBPPO resins at different temperatures. It can be seen that all resins show semicircular curves at −50 and 0°C, suggesting that their dielectric properties obey the Debye relaxation process[26] or a single relaxation process. With the increase of temperature, these semicircles appear distortion, reflecting that the simple polarization–relaxation model is not applied. At 150°C, the curves of all resins completely change owing to the presence of α-relaxation.

CONCLUSION

A new HBPPO-modified BCE resin system with significantly decreased curing temperature and outstanding dielectric properties are successfully developed. Compared to BCE resin, BCE/HBPPO resin has different curing mechanism and thus different crosslinked networks and macroperformance. The dielectric properties are frequency and temperature dependence, while which is closely related with the content of HBPPO in the BCE/HBPPO system. Dielectric properties of any resin obey Debye behavior only at very low temperature. The molecular relaxation can be used to explain these results.

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