Synthesis and Properties of Novel 4,4′-Biphenylene-Bridged Flame-Retardant Cyanate Ester Resin

Zhongyun Zhang, Ruobing Yu, Fan Wang, Jialiang Wang, Yangsheng Jiao

Key Laboratory for Specially Functional Polymeric Materials and Related Technology of the Ministry of Education, School of Materials Science and Engineering, East China University of Science and Technology, Shanghai 200237, Peolple's Republic of China

ABSTRACT: The bisphenol-containing 4,4′-biphenylene moiety was prepared by the reaction of 4,4′-bis(methoxymethyl) biphenyl with phenol in the presence of *p*-toluenesulfonic acid. The bisphenol was end-capped with the cyanate moiety by reacting with cyanogen chloride and triethylamine in dichloromethane. Their structures were confirmed by Fourier transform infrared spectroscopy, ^{1}H-NMR, and elemental analysis. Thermal behaviors of cured resin were studied by differential scanning calorimetry, dynamic mechanical analysis, and TGA. The flame retardancy of cured resin was evaluated by limiting oxygen index (LOI) and vertical burning test (UL-94 test). Because of the incorporation of rigid 4,4′-biphenylene moiety, the cyanate ester (CE) resin shows good thermal stability (T_g is 256°C, the 5% degradation temperature is 442°C, and char yield at 800°C is 64.4%). The LOI value of the CE resin is 42.5, and the UL-94 rating reaches V-0. Moreover, the CE resin shows excellent dielectric property (dielectric constant, 2.94 at 1 GHz and loss dissipation factor, 0.0037 at 1 GHz) and water resistance (1.08% immersed at boiling water for 100 h). © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 122: 2609–2615, 2011

Key words: cyanate ester resins; thermal stability; LOI; flame retardant

INTRODUCTION

Cyanate ester (CE) resins are a family of high-temperature thermosetting resins—more accurately named polycyanurates—that bridge the gap in thermal performance between engineering epoxy and polyimides.[1] CE resins have many excellent properties, such as good thermal stability, high-glass transition temperatures, low dielectric constants over a wide range of frequencies and temperatures, and outstanding adhesion to conducting metals.[2,3] These properties make them direct competitors for epoxy resins (epoxies) and bismaleimides in high-performance applications such as structure aerospace application and electronic application.[4] Presently, CE resins are consumed by the high-speed electronic industry, which was once dominated by epoxy resins.[5] However, the major drawback of most CE resins for the electronic applications is lack of flame retardancy, for which flame retardancy (UL-94 V-0) is required.[6–8]

CE resins are often modified by flame-retardant brominated epoxy resins to meet the requirement of flame retardancy while highly toxic hydrogen bromide and potentially carcinogenic brominated furans and dioxins may form during combustion.[9,10] Because of the existence of brominated epoxy resins, it is apt to corrode metal components (via the generation of halide ion in warm, humid environment), depress T_g, break down dielectric loss properties, and increase density.[11] It is urgent to investigate inherently self-extinguishing CE resins. The most effective way to flame retardancy in CE resins is the incorporation of flame-retardant group into the CE backbone. In the literature, phosphorus-containing CE resins,[12–14] commercial CE resins such as sulfur-linked AroCy T,[15] and hexafluoroisopropylidene-linked AroCy F[16] were envisioned. The phosphorus-containing CE resins show excellent flame retardacy(UL-94 V-0), while T_g and thermal stability decrease markedly with the increase of phosphorus content. Sulfur-linked CE resin can achieve self-extinguishing flammability rating (UL-94 V-0), but only cyclopentanone is stable solvent for AroCy T,[17] which limits the processability of AroCy T. AroCy F resin shows self-extinguishing behavior (UL-94 V-0), high-thermal stability, and low-dielectric properties, but the generation of toxic, halogenated degradation by-products during exposure to the fire is harmful to enviroment.[18] The best solution to flame retardancy is to develop nonhalogen, nonphosphorus flame-retardant CE resins. Anuradha and Sarojadevi[19] synthesized a series of new CEs by Betti-type reaction, with excellent flame retardancy (LOI value: 43–45), which bear quinoline

and —NH— group in the main chain and various pendant groups. In this work, we will develop a novel CE to enrich the nonhalogen flame-retardant CE resins.

To prepare flame-retardant CEs, flame-retardant mechanism is understood. All flame retardants act either in the vapor phase or the condensed phase to interfere with the combustion process during heating, pyrolysis, ignition, or flame spread.[20] For example, the flame retardants in the condensed phase promote the formation of carbonaceous char of the polymer, which acts a barrier to inhibit gaseous products from diffusing into the flame and to shield the polymer surface from heat and air.[21] Therefore, the char residue may be used as a measurement of the nonflammability, and a correlation between the char residue (CR) and the limiting oxygen index (LOI) of polymers is represented by eq. (1)[22,23]:

$$LOI \times 100 = 17.5 + 0.4CR \quad (1)$$

where CR is the char residue in wt % at 800°C.

Based on the condensed-phase mechanism of flame retardancy, it is convenient for the introduction of chemical group resulting in high char residue to achieve UL-94 V-0 rating. In this article, we envision a novel CE by the introduction of aromatic 4,4′-biphenylene moiety in the backbone, which is helpful to promote char residue of CE resin, increase the LOI value of CE resin, and improve flame retardant performance. Furthermore, hydrophobicity of 4,4′-biphenylene moiety would decrease water absorption of the CE resin. This study will focus on the synthesis of CE-containing 4,4′-biphenylene moiety in the backbone and the relationship between structure and properties.

EXPERIMENTAL

Materials

4,4′-Bis(methoxymethyl)biphenyl (BMBP) was obtained from Quzhou Rainful Chemical Co., and Cyanogen chloride, triethylamine, dichloromethane, and bishphenol A dicyanate ester (BADCy; Fig. 1) were supplied by Shanghai Huifeng Technical and Business Co. Phenol, toluene, and p-toluenesulfonic acid (PTS) were purchased from Shanghai Lingfeng Chemical Reagent Co. The other reagents were used as received without further purification.
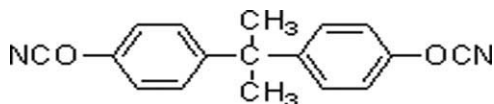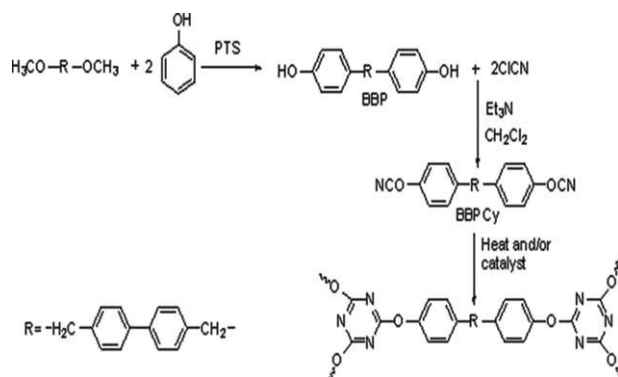

Figure 1 Chemical structure of BADCy.

Scheme 1 Synthesis and cyclotrimerization of BBPCy.

Synthesis of 4,4′-bis(4-hydroxybenzyl) biphenyl (BBP)

About 188 g (2 mol) of melted phenol, 48.4 g (0.2 mol) of BMBP, and 2.4 g of PTS were added into the 500-mL four-necked round-bottomed flask. The mixture was gradually heated to 130°C with stirring until the methanol was formed and generated out. Then, the reaction was continued at 130°C for 4 h. After the completion of the reaction, the reaction mixture was neutralized with sodium hydroxide, and excess phenol was distilled under reduced pressure to obtain crude product. Finally, the crude product was dissolved in toluene and washed with hot water several times, the precipitation was obtained when the organic phase cooled below 0°C, then filtrated the precipitation and recrystallized from toluene, and dried under vacuum to give BBP as yellowish crystals (43.9 g and 60%). m.p. = 220°C.

Fourier transform infrared spectroscopy (FTIR; KBr, cm^{-1}): 3386 (v_{OH}), 3024, 2908, 2840, 1612, 1597, 1512, 1247, 1101, 808, and 765. ^{1}H-NMR (acetone-d_6; δ, ppm): 3.90 (s, 4H), 6.75 (d, J = 8.5, 4H), 7.07 (d, J = 8.5, 4H), 7.26 (d, J = 8, 4H), 7.52 (d, J = 8.5, 4H), and 8.13 (s, 2H). ELEM. ANAL. Calcd for $C_{26}H_{22}O_2$: C, 85.22%; H, 6.05%; O, 8.73%; Found: C, 84.94%; H, 6.04%; O, 9.02%.

Synthesis of 4,4′-bis(4-cyanatobenzyl) biphenyl (BBPCy)

To a 1000-mL four-necked round-bottomed flask fitted with a stirring bar, thermometer, nitrogen inlet,
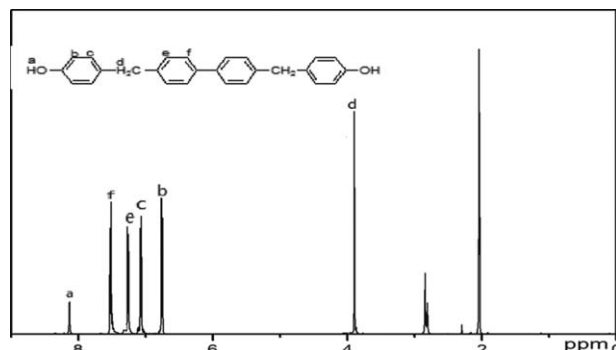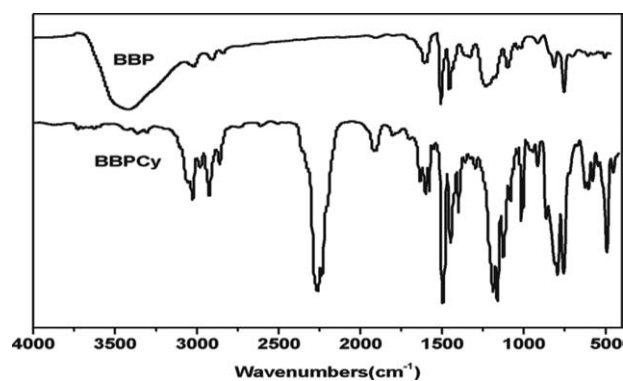

Figure 2 ^{1}H-NMR spectrum of BBP.

Figure 3 IR spectra of BBP and BBPCy.

Figure 4 ^{1}H-NMR spectrum of BBPCy.

and dropping funnel were added 11.9 g (0.194 mol) of ClCN and 300 mL of dichloromethane. The mixture was cooled at −5°C. A solution of 63 g (0.172 mol) of BBP, 19 g (0.189 mol) of triethylamine, and 200 mL of dichloromethane was transferred to the dropping funnel and added dropwise to the ClCN solution under nitrogen over 30 min and then maintained at −5°C for 2 h. After the reaction was completed, the resulting solution was filtered, and 10% sulfuric acid was added to the filtrate, following by washing with water several times. The organic phase was dried with anhydrous Na_2SO_4 and distilled under vacuum to remove the solvent to obtain the corresponding dicyanate (57.3 g ,80%) as a yellowish solid; m.p. = 163°C.

FTIR (KBr, cm^{-1}): 3026, 2924, 2860, 2263 (v_{OCN}), 1496, 1163, 1126, 794, and 756.

^{1}H-NMR (acetone-d_6, δ, ppm): 4.08 (s, 4H), 7.33 (d, $J = 8$, 4H), 7.35 (d, $J = 8.5$, 4H), 7.47 (d, $J = 6$, 4H), and 7.58 (d, $J = 8.5$, 4H). ELEM.ANAL. Calcd for $C_{28}H_{20}O_2N_2$: C, 80.75%; H, 4.84%; N, 6.73%; O, 7.68%; Found: C, 81.05%; H, 4.58%; N, 6.52%; O, 7.85%.

Sample preparation

BBPCy and BADCy resins were melted, then poured directly into the preheated molds (100°C) respectively, degassed the entrapped air or any volatiles, and subsequently cured in an air convection according to the procedure: 180°C for 4 h, 200°C for 1 h, and then followed by 3 h postcure at 230°C. The cured resins were ejected from the molds, cut, and polished according to the dimension requirements for the property measurements.
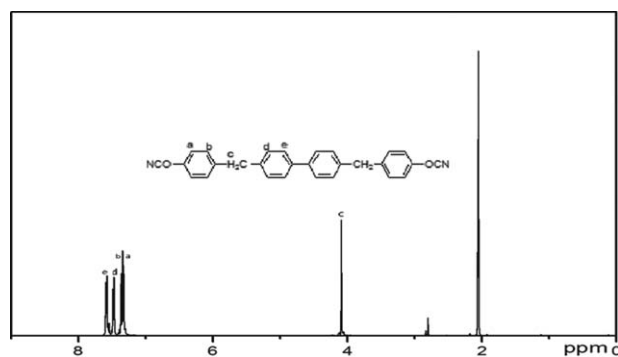
Measurements

FTIR spectra were recorded on Nicolet 5700 in the form of pellet with KBr. ^{1}H-NMR spectra were carried out on the DRX500 spectrometer (Bruker). The spectra were determined in acetone-d_6 with TMS as internal standard. Elemental analysis (EA) was determined using an Elementar Vario EL III elemental analyzer. Differential scanning calorimetric (DSC) analysis was performed on a TA Instruments modulated DSC 2910 equipment at a heating rate of 10°C min^{-1} under flowing nitrogen (50 cm^3 min^{-1}). Glass transition temperature (T_g) was measured by a TA DMA 2980 Dynamic Mechanical Analyzer at a frequency of 1 Hz and a heating rate of 3°C min^{-1} in the air. TGA was carried out on a NETZSCH STA 409 thermal analyzer at a heating rate of 10°C min^{-1} under N_2 from 25 to 800°C. LOI is defined as the minimum fraction of O_2 in a mixture of O_2 and N_2 that will just support flaming combustion. The LOI test was conducted according to the ASTM D 2836 oxygen index method using a test specimen bar 7–15 cm in length, 6.5 ± 0.5 mm in width and 3.0 ± 0.5 mm in thickness. The UL-94V flame test was performed according to the ASTM D 3801 testing procedure with the testing specimen bar of 130 mm in length, 10 mm in width, and 2 mm in thickness. The UL-94V determines the upward-burning characteristics of a solid. Five sample bars, which were suspended vertically over surgical cotton, were ignited by a Bunsen burner. Two ignitions with 10-s burning time were applied to each sample bar. Dielectric measurements were performed with an Agilent 4991B measurement at room temperature by the two-parallel-plate mode, whose applied voltage was 200 mV. The instrument was calibrated with polytertrafluoroethylene sample provided by the

TABLE I
Elemental Analysis Results for BBP and BBPCy

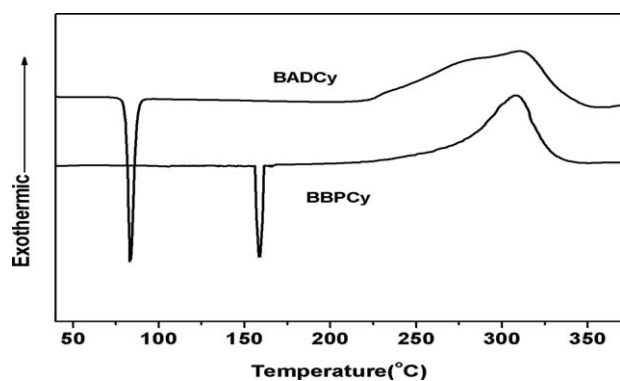| Monomers	Molecular	Calculated values	Found values
BBP	$C_{26}H_{22}O_2$	C: 85.22%, H: 6.05%, O: 8.73%	C: 84.94%, H: 6.04%, O: 9.02%
BBPCy	$C_{28}H_{20}N_2O_2$	C: 80.75%, H: 4.84%, N: 6.73%, O: 7.68%	C: 81.05%, H: 4.58%, N: 6.52%, O: 7.85%

Figure 5 DSC thermograms of BBPCy and BADCy monomers.

Figure 6 DMA spectra of cured BBPCy and cured BADCy resins.

manufacturer. Before being tested, samples (2 × 2 cm^2 and 0.2 cm thickness) were dried under vacuum at 100°C for 8 h. Long-term water absorption was performed as follows: the samples were dried *in vacuo* at 120°C until constant weight. Then, the dried samples were immersed in the boiling distilled water for the duration of the study. Samples were taken out of the bath, and surface moisture was removed with a dry cloth before weighed. The water absorption was calculated as follows: weight gain = $(W/W_0 - 1) \times 100\%$, where W is the weight of a sample after it was placed in the boiling water for some time and W_0 was the weight of a sample before it was placed in the boiling water.

RESULTS AND DISCUSSION

Characterization of BBPCy

The CE resin (BBPCy) was prepared according to the following two steps: synthesis of bisphenol precursor (BBP) and cyanation of bisphenol precursor (Scheme 1).

BBP and BBPCy were characterized by ^{1}H-NMR, FTIR, and EA. In the ^{1}H-NMR spectrum of BBP in acetone-d_6 (Fig. 2), two singlet signals of the methylene and hydroxyl proton are observed at 3.90 and 8.13 ppm. Four kinds of doublet peaks corresponding to the aromatic protons are detected at 6.75, 7.05, 7.25, and 7.52 ppm, respectively. Moreover, the chemical shift of acetone-d_6 can be observed at 2.05 ppm, and the chemical shift for water in the acetone-d_6 occurs at 2.8 ppm. FTIR spectrum of BBP is shown in Figure 3, with strong absorption peak for the vibration of hydroxyl group at 3386 cm^{-1}.

TABLE II
Thermal Cure Characteristics of BBPCy and BADCy

Dicyanates	M_p (°C)	T_i (°C)	T_p (°C)	T_f (°C)	ΔH (J/g)
BBPCy	163	230	309	339	446
BADCy	83	230	310	351	430

The EA result of BBP in Table I is in good agreement with the calculated values. These results confirm the proposed structure of BBP. For BBPCy in Figure 3, the disappearance of hydroxyl stretching at 3386 cm^{-1} and the appearance of the characteristic peak of the —OCN functionality locating at 2263 cm^{-1} confirm the formation of the corresponding dicyanate. Figure 4 shows the ^{1}H-NMR spectrum of BBPCy. Compared to Figure 2, BBPCy shows pronounced downfield shifts associated with the aromatic protons, particularly for those ortho to the cyanate group. The assignments of chemical shifts are as follow: 2.05 ppm (acetone-d_6), 2.8 ppm (water in the acetone-d_6), 4.08 ppm (methylene protons), 7.33 and 7.35 ppm (phenyl protons on the phenol ring), and 7.47 and 7.58 ppm (phenyl protons on the biphenyl ring). The EA of BBPCy is in good agreement with the calculated values (Table I).

Thermal behaviors of CE resins

The curing behaviors of CE resins are examined by DSC. The thermograms of the dicyanates are shown in Figure 5. The endothermic transition temperature (M_p), onset temperature for curing reaction (T_i), exothermic peak temperature (T_p), and end temperature
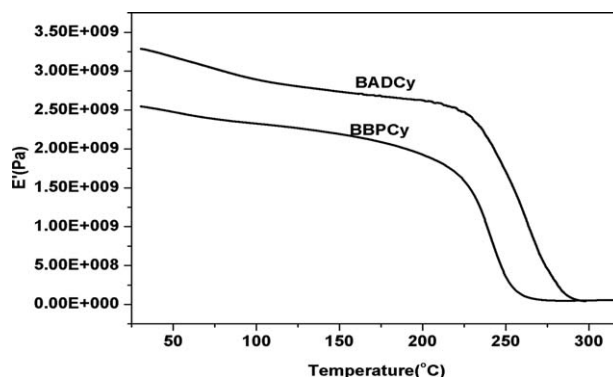

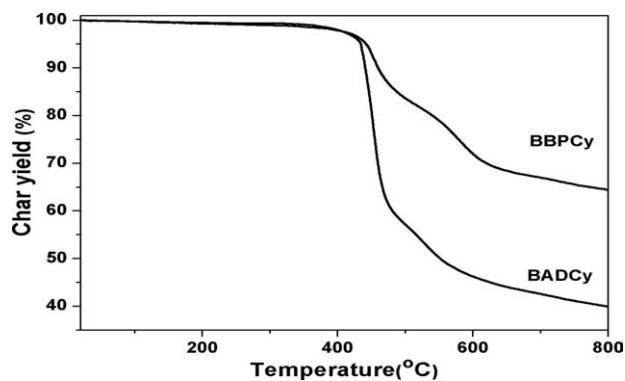**Figure 7** Storage modulus for cured BBPCy and cured BADCy resins.

Figure 8 TGA thermograms of cured BBPCy and cured BADCy resins in N_2.

for curing reaction (T_f) are reported in Table II. BADCy and BBPCy show an endothermic transition associated with melting point at 83°C and 163°C, respectively. The initial curing temperature for BADCy and BBPCy are very close. Due to the introduction of aromatic 4, 4′-biphenylene moiety (more rigid backbone than isopropylidene backbone in BADCy), BBPCy has a higher melting point than BADCy.

Dynamic mechanical properties of cured CE resins

Dynamic mechanical analysis (DMA) is a powerful technique measuring the glass transition temperature (T_g), particularly for polymers with rigid backbones. The peak temperature of tanδ is assumed to be T_g in this article. Figure 6 shows the dynamic analysis of cured BBPCy and cured BADCy resins. The introduction of stiff 4,4′-biphenylene moiety in the backbone of BBPCy resin enhances the stiffness of the chain but decreases the crosslink density of the resin simultaneously, resulting in a lower T_g than that of BADCy resin. Figure 7 shows the storage modulus of cured BBPCy and BADCy resins. The storage modulus at room temperature for cured BBPCy and BADCy resins is 2547 and 3288 MPa, respectively. All resins show a sharp reduction in
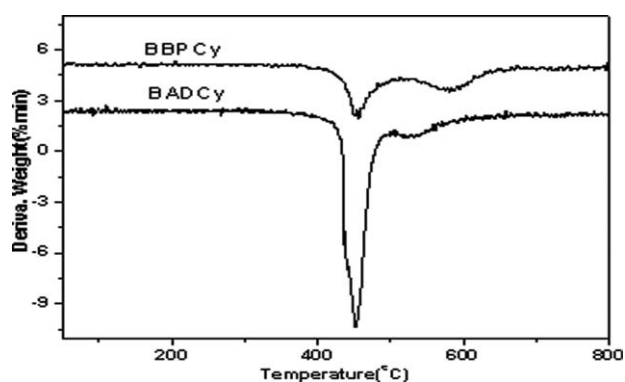

Figure 9 DTG thermograms of cured BBPCy and cured BADCy resins in N_2.

TABLE III
TGA Data of Cured BBPCy and Cured BADCy Resins

Resins	$T_{5\%}$ (°C)	$T_{10\%}$ (°C)	T_{m1}[a]	T_{m2}[a]	CY (%) at 800°C
Cured BBPCy	442	460	455	580	64.4
Cured BADCy	434	440	451	532	39.8

[a] The maximum-rate degradation temperature.

the storage modulus as the temperature increased signifying a transition between glassy state and rubbery state.[24] The slightly lower storage modulus for BBPCy resin relative to BADCy resin is probably attributable to the lower crosslink density for BBPCy resin than that for BADCy resin.

Thermal degradation behavior of cured CE resins

The thermal stability of a polymeric material is very important, which may concern the release of decomposition products and the formation of a char. The thermal stability of the cured CE resin is evaluated by TGA analysis under N_2. Figures 8 and 9 show the TGA and the corresponding DTG curves of cured BBPCy and cured BADCy resins, respectively. The onset decomposition (5% weight loss), 10% weight loss, the rapid weight loss before 800°C, and char yield (CY) at 800°C in N_2 are summarized in Table III. The 5% degradation temperature and char yield of BBPCy resin is 442°C and 64.4%, respectively, which is higher than those of BADCy resin (the 5% degradation temperature is 434°C and char yield is 39.8%).

In Figure 9, cured BBPCy resin shows two stages of weight loss in N_2, which are reflected by the two peaks in the first derivative weight loss (DTG). It is generally accepted that the degradation mechanism of CE resins proceeds in several stages including the random scission and crosslinking between 400 and 450°C, decyclization of the triazine ring between 450 and 500°C, and decomposition of the primary residue between 500 and 750°C.[25,26]

Flame retardancy of cured CE resins

To investigate the flame retardancy of the cured CE resins, the LOI values and vertical burning rating (UL-94) of CE resins were tested. These data are summarized in Table IV. Self-extinguishing is

TABLE IV
Results of Limited Oxygen Index and Vertical Burning Test (UL-94) for CE Resins

Samples	Flaming drops	Cotton ignited	UL-94 standard	LOI (%)
BBPCy	N/A[a]	N/A	V-0	42.5
BADCy	N/A	N/A	Burning	28

[a] Not available.

characteristic of LOI value above 26. The LOI value of cured BBPCy resin is 42.5, indicative of good flame retardancy, which is much higher than that of BADCy resin. The UL-94 rating of cured BBPCy reaches V-0, and no flaming drips were observed in the first and second ignition. Because of the introduction of 4,4′-biphenylene moiety in the backbone, it is apt to increase the carbonaceous char, which acts as a barrier to prevent the heat transfer between the flame zone and the burning zone, to protect the underlying material from further burning and retard the pyrolysis of polymer.

Water absorption

Water absorption will have an effect on thermal, dielectric, and mechanical properties of the resins. Because of water's contribution to polarization and energy dissipation, the dielectric constant of laminate board materials will be increased. Furthermore, moisture absorption will also reduce T_g of laminate materials. Therefore, low water absorption will be necessary to laminate materials. Figure 10 shows the water absorption of the cured CE resins in boiling water as a function of immersion time. It shows that the water absorption of cured BBPCy resin in boiling water is 1.08%. CE resins, with weak dipoles and the absence of hydrogen bonding, are characteristic of low-moisture absorption properties. In addition, the incorporation of hydrophobic 4,4′-biphenylene moiety in the backbone will further depress the water absorption of BBPCy resin, resulting in lower water absorption than that of BADCy resin.

Dielectric properties

The dielectric constant and dissipation factor of BBPCy and BADCy resins are shown in Table V. The dielectric constant and dielectric dissipation factor of BBPCy resin are 2.94 and 0.0037, respectively. The low dielectric character of CE resins is attributed
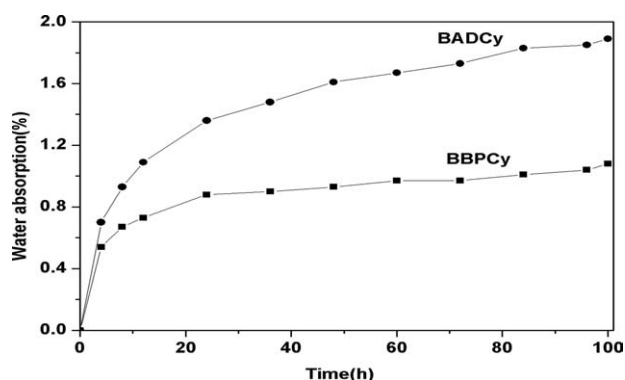
TABLE V
Dielectric Properties and Water Absorption of Cured BBPCy and Cured BADCy Resins

Resins	D_k[a]	D_f[b]	Water absorption[c] (%)
BBPCy	2.94	0.0037	1.08
BADCy	2.98	0.0083	1.89

[a] Dielectric constant at 1 GHz.
[b] Dissipation factor at 1 GHz.
[c] Water absorption at boiling water for 100 h.

to the high degree of symmetry in this linkage structure where dipoles associated with the carbon–nitrogen and carbon–oxygen bonds are counterbalanced, resulting in a low dipole moment and low energy storage in an electric field.[27] The lower dissipation factor of BBPCy resin may be due to the incorporation of lower polar 4,4′-biphenylene moiety in the backbone, which shields the interchain electronic reaction.[28]

CONCLUSIONS

Novel flame-retardant CE-containing 4,4′-biphenylene moiety was successfully synthesized through the reaction of cyanogen chloride with the corresponding bisphenol precursor. Because of the introduction of aromatic 4,4′-biphenylene moiety in the backbone, the LOI value of cured BBPCy resin is 42.5 and UL-94 rating reaches V-0. DMA and TGA results show that BBPCy resin has slightly lower T_g (258°C) and higher thermal stability ($T_{d5\%}$ is 442°C and char yield at 800°C is 64.4%) than BADCy resin. BBPCy resin exhibits low water absorption (1.08%) and excellent dielectric properties (D_k is 2.94 and D_f is 0.0037 at 1 GHz). The excellent flame retardancy and many other excellent properties make BBPCy a promising candidate for printed circuit board and electronic encapsulation.

Figure 10 Plots of water absorption versus time for cured BBPCy and cured BADCy resins.

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