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# Synthesis and characterization of low dielectric constant polybenzoxazines with different fluorine contents

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## Abstract

The rapid development of integrated circuits puts forward higher requirements for low dielectric constant polymers. In this study, two fluorinated benzoxazine monomers, bis-(3,4-dihydro-3-(4-fluoro-3-(trifluoromethyl)phenyl)-2H-1,3-benzoxazine)isopropane (BA-ftfa) and bis-(3,4-dihydro-3-(4-fluoro-3-(trifluoromethyl)phenyl)-2H-1,3-benzoxazine)isoperfluoropropane (BAF-ftfa), were successfully synthesized. Differential scanning calorimeter (DSC) was used to monitor the thermal curing behaviors of the monomers and their mixtures. Five groups of polybenzoxazine samples with different fluorine content (25% ~ 35%) were obtained by copolymerizing the two monomers with different mass ratios. The effects of fluorine content on dielectric properties were investigated and the results show that PBAF-ftfa with the highest fluorine content has the lowest dielectric constant of 2.53 at 1 MHz. Furthermore, the results of thermogravimetric analysis (TGA) and dynamic mechanical analysis (DMA) show that all polymers possess excellent thermal stability and exhibit high glass transition temperatures over than 180°C.

**KEYWORDS**

fluorination, low dielectric constant, polybenzoxazine

## 1 | INTRODUCTION

In order to meet the development needs of high-frequency and high-speed integrated circuits, designing low-dielectric polymer materials to solve signal transmission delay and electronic cross-talk has attracted widespread attention.[1,2] The ideal low-dielectric materials for high-performance interlayer materials of large-scale integrated circuits are also expected to have good mechanical properties, thermal properties, and low water absorption.[3–5] In recent decades, numerous researches on low dielectric constant polymer materials have been developed, such as polyimides,[6,7] polytetrafluoroethylene,[8] benzoxazole,[9] benzocyclo-butene resins,[10] polybenzoxazine,[11] and epoxy resins.[12]

Polybenzoxazine, a type of thermosetting resin, has good heat resistance,[13,14] low surface free energy,[15,16] low dielectric constant, and high dimensional stability.[17] Besides, its flexibility in molecular design provides advantageous prerequisites for the manufacture of resins with low dielectric, high thermal stability, and low water absorption. Some special functional groups can be introduced by changing the phenol or amine in raw materials. Therefore, polybenzoxazine resins have received more and more attention and many studies have also shown that they have good application prospects and huge development potential in low dielectric material fields.[18–21]

From the Debye formula,[4] it can be known that the dielectric constant of the material is related to the polarization and dipole density, and their reaction can effectively reduce the dielectric constant of the material. Bringing in groups with large spatial steric resistance to increase the free volume is a method to decrease the dipole density and the dielectric constant.[22,23] Due to the low polarizability of the C—F bond, the introduction of fluorine-containing groups in polymers can reduce the dielectric constant. At the same time, bringing in fluorinated groups also enables polymers to exhibit excellent thermal stability, low water absorption, and great heat resistance.[24–27] Wu et al.[28] prepared two polybenzoxazines with octafluorocyclopentene as the center, and the results showed that the polymer with higher fluorine content performs a lower dielectric constant (2.61 at 1 MHz). Zhang et al.[29] synthesized a novel polybenzoxazine with o-trifluoroacetamide functionality which possessed great dielectric property (2.84 at 1 MHz).

In this study, two fluorinated benzoxazine monomers (BA-ftfa and BAF-ftfa) were synthesized using 4-fluoro-3-trifluoromethylaniline, paraformaldehyde and different phenolic sources (Bisphenol A and Bisphenol AF). Five groups of polybenzoxazine samples with different fluorine content were obtained by copolymerizing the two monomers with different mass ratios, and the effects of fluorine content on polymer properties were investigated. Their thermal curing behaviors were characterized by differential scanning calorimeter (DSC). The thermal stability was tested by thermogravimetric analyzer (TGA). Their surface and dielectric properties were also explored.

## 2 | EXPERIMENTAL

### 2.1 | Materials

4-Fluoro-3-trifluoromethylaniline (99%) was obtained from Shanghai Aladdin Biochemical Technology Co., Ltd. Bisphenol A (CP), dichloromethane (AR) and toluene (AR) were purchased from Sinopharm Chemical Reagent Co., Ltd. Paraformaldehyde (AR) was obtained from Shanghai Lingfeng Chemical Reagent Co., Ltd. Bisphenol AF (98%) was purchased from Shanghai Maclean Biochemical Technology Co., Ltd. All chemicals were used directly without purification.

### 2.2 | Preparation of benzoxazine monomers

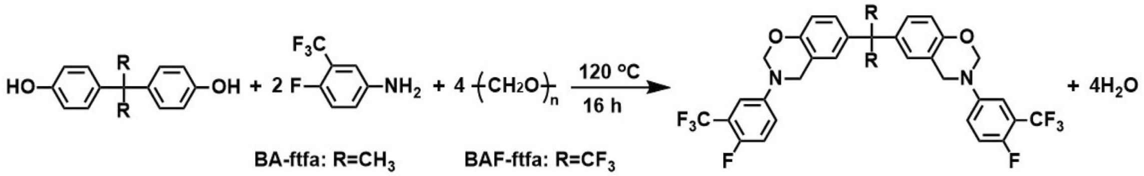
As shown in Scheme 1, two fluorinated benzoxazine monomers were synthesized using a solvent method. 2.64 g (0.088 mol) paraformaldehyde, 7.16 g (0.04 mol) 4-fluoro-3-trifluoromethylaniline and toluene were added in a four-neck flask at room temperature with nitrogen gas. The mixture was stirred at room temperature for 30 min, and then 6.72 g (0.02 mol) bisphenol AF and toluene were finally added. The system was heated to 120°C and refluxed for 16 h under nitrogen atmosphere. After the reaction solution was cooled, the toluene was evaporated with a rotary evaporator to obtain the crude product, which was then dissolved in dichloromethane and washed with NaOH aqueous solution and deionized water. Finally, the solvent was removed in a vacuum oven and a pale yellow powder named BAF-ftfa was obtained. BA-ftfa was prepared in the same way using bisphenol A. The reaction route is shown in Scheme 1.

### 2.3 | Preparation of benzoxazine copolymers

Five groups of samples with different mass ratios of BA-ftfa and BAF-ftfa were prepared and noted as BA-ftfa, AAF1, AAF2, AAF3, and BAF-ftfa. Wherein, AAF1, AAF2, and AAF3 correspond to the mass ratios of BA-ftfa and BAF-ftfa were 2:1, 1:1, and 1:2, respectively. These samples were firstly added into the molds and then put into a vacuum oven at 130°C for 1 h to remove bubbles. Afterward, they were placed in a blast drying oven to heat at 150, 180, 200, and 220°C for 1 h, 230°C for 2 h, and 240°C for 3 h. The copolymers obtained under the curing conditions were named PBA-ftfa, PAAF1, PAAF2, PAAF3, and PBAF-ftfa.

### 2.4 | Characterizations

The Fourier transform infrared (FTIR) spectra were obtained with a Thermo Fisher Scientific Nicolet IS10 FT-IR analyzer by the method of potassium bromide



SCHEME 1 Synthesis route of benzoxazine monomers.

(KBr) tablet. All spectra were scanned 32 times with a resolution of 4 $cm^{-1}$ in the range of 4000–400 $cm^{-1}$. $^1H$ NMR spectrum was recorded using a Bruker Advance III 400 (400 MHz, Germany). $^{19}F$ NMR spectrum was measured by a Bruker Ascend 600 nuclear magnetic resonance spectrometer (600 MHz, Germany). Deuterium dimethyl sulfoxide (DMSO-$d_6$) was used as the solvent. The fluorine content was obtained using a Metrohm 905 potentiometric titrator (Switzerland). DSC analyses were performed by a Q2000 differential scanning calorimeter (TA, America). All samples were tested under a nitrogen atmosphere with a heating rate of 10°C·$min^{-1}$ from 50 to 300°C. TGA was characterized using a TA SDT Q600 Thermogravimetric analyzer, the samples were heated from 40 to 800°C under a nitrogen atmosphere at a rate of 10°C·$min^{-1}$. DMA was measured using a Mettler DMA1. The surface morphology of the films was observed by a field emission scanning electron microscope (Nova NanoSEM 450, FEI). The dielectric properties of the polybenzoxazines were tested by a Concept 40 broadband dielectric impedance spectrometer (Germany), the frequency was in the range from 10 Hz to 1 MHz. All the films were dried before measurements and then tested at room temperature. The water absorption test was measured by the mass change of the polymer before and after immersion in deionized water for a day at room temperature. The water contact angle (WCA) was investigated by an OCA20 produced by the German Data physics Company, five different areas were tested on the surface and take the average as the result.

## 3 | RESULTS AND DISCUSSION

### 3.1 | Structural characterizations of benzoxazine monomers

The structures of benzoxazine monomers BA-ftfa and BAF-ftfa were characterized by FT-IR. As shown in Figure 1, the characteristic absorption peak of the oxazine ring at 953 $cm^{-1}$, the stretching vibration peak of C—O—C bond at 1091 $cm^{-1}$ and the stretching vibration peak of C—N—C bond at 1164 $cm^{-1}$ in BA-ftfa are observed clearly. The characteristic absorption peaks of the oxazine ring, the stretching vibration peak of C—O—C bond and the stretching vibration peak of C—O—C bond in BAF-ftfa are at 943, 1091, and 1170 $cm^{-1}$, respectively. Besides, the peaks at 1132 and 1319 $cm^{-1}$ and assigned to the symmetric and asymmetric of —$CF_3$ group and the absorption peak at 1238 $cm^{-1}$ assigned to Ar-F in two monomers are also present.[30] The absorption peak appeared at 1202 $cm^{-1}$ in BAF-ftfa is attributed to the vibration of the C—F bond on bisphenol AF. The information demonstrates that the benzoxazine monomers are successfully synthesized.

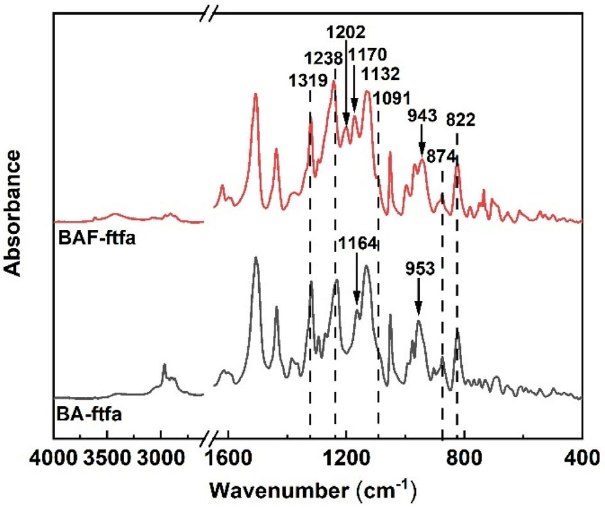


**FIGURE 1** FT-IR spectra of benzoxazine monomers BA-ftfa and BAF-ftfa. [Color figure can be viewed at wileyonlinelibrary.com]

In addition, $^1H$ NMR and $^{19}F$ NMR were also investigated to confirm the structures of benzoxazine monomers. The results of $^1H$ NMR spectra are shown in Figure 2a, the resonances at 5.4 ppm (a) and 4.6 ppm (b) in BA-ftfa are corresponded to protons of O—$CH_2$—N and Ar—$CH_2$—N in the oxazine ring, the exhibition of the peak at 1.50 ppm (c) indicates the presence of the proton in the —$CH_3$ of BA-ftfa. The resonances corresponded to protons of O—$CH_2$—N and Ar—$CH_2$—N in the oxazine ring appear at 5.6 ppm (a′) and 4.7 ppm (b′) in BAF-ftfa. The characteristic proton resonances observed between 6.6 and 7.5 ppm in two monomers are attributed to the protons of the benzene ring. $^{19}F$ NMR spectra are shown in Figure 2b, the signals at −127 ppm (a,a′) and − 60 ppm (b,b′) in both two benzoxazine monomers reveal the existence of the fluorine atom on Ar-F bond and trifluoromethyl group. Furthermore, the fluorine atoms on trifluoromethyl groups of BAF-ftfa are proved by the response peak at −63 ppm (c′). These results further prove the successful synthesis of the monomers.

### 3.2 | Curing behaviors of benzoxazines

DSC was used to study the curing behaviors of benzoxazine monomers and their mixtures. The DSC curves are shown in Figure 3 and the data are listed in Table 1. The calculated fluorine content is based on the mass ratio of the two monomers.[31] Both the calculated fluorine content and the experimental fluorine content are shown in Table 1. From Figure 3a, the benzoxazine monomers and

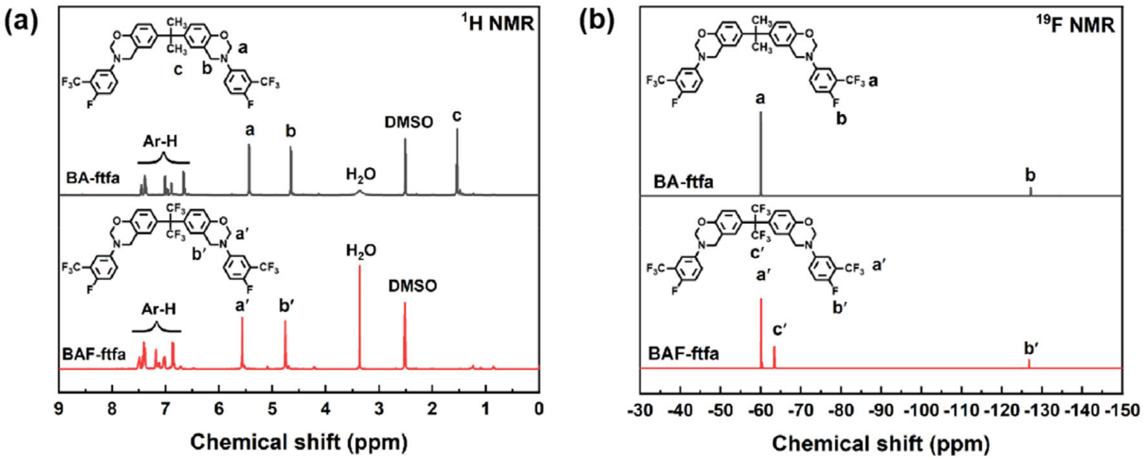


FIGURE 2 (a) $^{1}$H NMR spectra and (b) $^{19}$F NMR spectra of benzoxazine monomers. [Color figure can be viewed at wileyonlinelibrary.com]

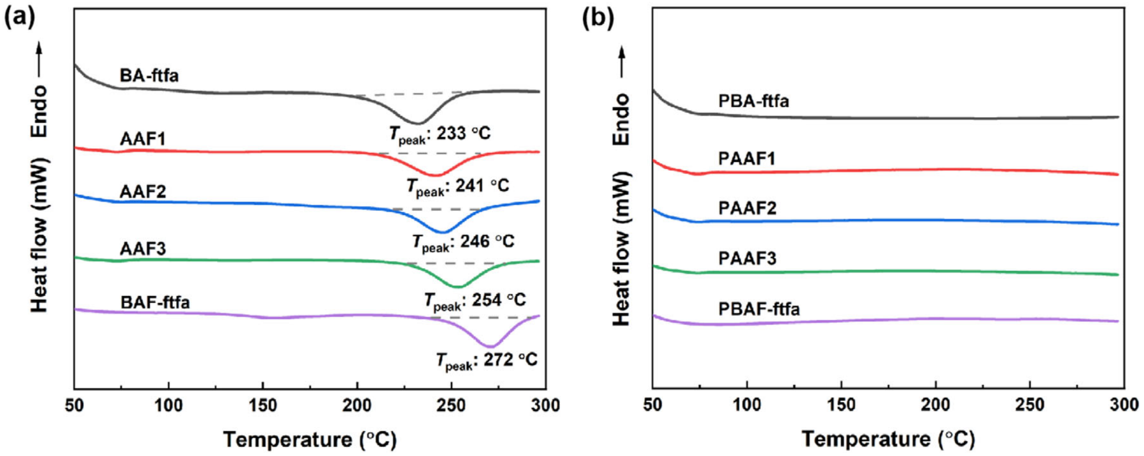


FIGURE 3 DSC curves of benzoxazine samples (a) before and (b) after curing. [Color figure can be viewed at wileyonlinelibrary.com]

TABLE 1 DSC data of benzoxazine samples.

| Samples | Mass ratio of BA-ftfa:BAF-ftfa | Fluorine content (wt.%) | | $T_{on}$ (°C) | $T_{peak}$ (°C) |
|---|---|---|---|---|---|
| | | Calculated | Found | | |
| BA-ftfa | 1:0 | 23.97 | 23.56 | 207 | 233 |
| AAF1 | 2:1 | 27.93 | 26.36 | 215 | 241 |
| AAF2 | 1:1 | 29.91 | 28.37 | 222 | 246 |
| AAF3 | 1:2 | 31.89 | 31.45 | 231 | 254 |
| BAF-ftfa | 0:1 | 35.85 | 35.09 | 246 | 272 |

their mixtures display obvious exothermic peaks in the range of 207–299°C because of the ring-opening polymerization of the oxazine ring. Among the five samples, BA-ftfa shows the lowest peak onset temperature ($T_{on}$) of 207°C and the lowest peak temperature ($T_{peak}$) of 233°C. As the fluorine content of the samples increases, the exothermic peak moves toward high temperatures gradually. The $T_{on}$ and $T_{peak}$ of BAF-ftfa are 246 and 272°C, respectively. The diphenol-based benzoxazine monomers with electron-donating bridging groups tend to polymerize at a lower temperature than the ones with electron-withdrawing bridging groups.[32] The —$C(CH_3)_2$— group in bisphenol A is the electron-donating group, while the —$C(CF_3)_2$— group in bisphenol AF is the electron-

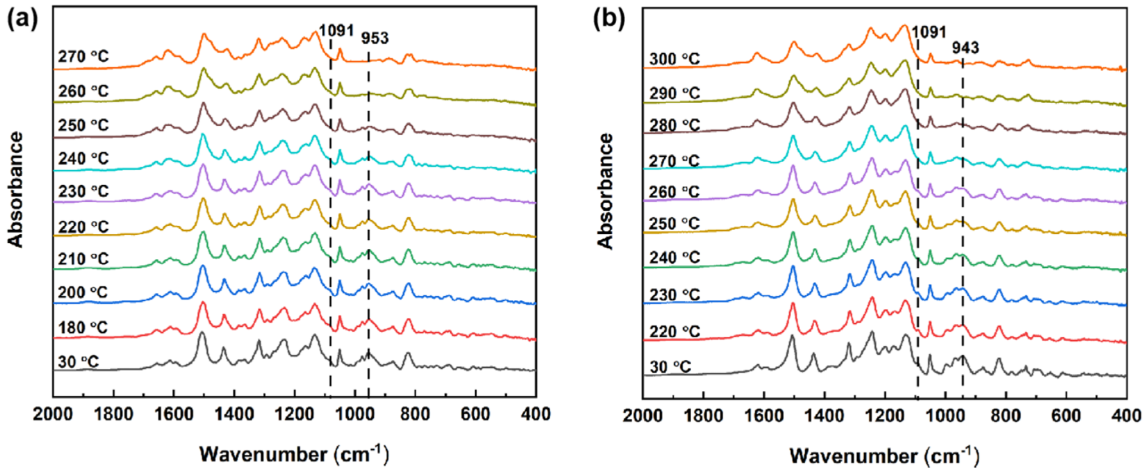


**FIGURE 4** In situ FT-IR spectra of (a) BA-ftfa and (b) BAF-ftfa at different temperatures. [Color figure can be viewed at wileyonlinelibrary.com]

withdrawing group. Therefore, the $T_{\text{peak}}$ gradually moves to higher temperatures as the fluorine content increases. As can be seen from Figure 3b, the exothermic peaks of the polybenzoxazines completely disappeared after curing, which indicates that the samples have been completely cured under the selected step temperature conditions.

In-situ FT-IR spectra analysis was further measured to confirm the changes of the structures of two benzoxazine monomers at different temperatures. From Figure 4a, it can be observed that with the temperature gradually rising, the characteristic peaks of BA-ftfa at 953 and 1091 cm$^{-1}$ associated with the oxazine ring and C—O—C stretching gradually decrease until disappear. In particular, the response peaks begin to weaken at 210°C and disappear completely at 260°C, which is consistent with the results obtained by DSC curves. As can be seen in Figure 4b, BAF-ftfa performs the same trend as BA-ftfa, except that the related bonds at 943 and 1091 cm$^{-1}$ associated with the oxazine ring and C—O—C stretching decrease starting from 250°C and completely disappear at 300°C. The changes of BAF-ftfa also verify the results of DSC.

## 3.3 | Thermal properties of polybenzoxazines

The thermomechanical properties of polybenzoxazines were investigated by the dynamic mechanical analysis (DMA). The storage modulus ($E'$) and tan $\delta$ curves of polybenzoxazines are depicted in Figure 5 and the values of the results are summarized in Table 2. From Figure 5a, the storage modulus of the five samples ranges from 3542 to 4858 MPa at 30°C, and they all present a decreasing trend with the increase of temperature. As can be seen in Figure 5b, the glass transition temperature ($T_g$, the peak temperature of tan $\delta$) of PBA-ftfa, PAAF1, PAAF2, PAAF3, and PBAF-ftfa is 211, 202, 196, 191, and 188°C. All polymers exhibit high $T_g$, which is possible owing to the presence of a large number of rigid groups.

Crosslinking density is also an important indicator for the thermosetting resins and can be calculated using Equation 1 proposed by Nielsen.[33]

$$\log_{10}\left(\frac{E'_e}{3}\right) = 7.0 + 293(\rho_x), \tag{1}$$

where $E'_e$ is the storage modulus (dyne/cm$^2$) at the temperature 40°C higher than $T_g$ and $\rho_x$ is the crosslinking density. As can be seen from Table 2, PBAF-ftfa has the lowest $\rho_x$ of $2.01 \times 10^{-3}$ mol/cm$^3$ on account of greater steric hindrance trifluoromethyl groups than methyl groups. The $\rho_x$ of PBA-ftfa cannot be obtained over the test temperature range. The crosslinking density affects the glass transition temperature of the polymer.[34,35] With the increase of fluorine content, the content of trifluoromethyl groups with greater steric hindrance in the polymer system increases, which reduces the crosslinking density of the polymer. The decrease in crosslink density lowers the glass transition temperature of the polymer.

The thermal stability of polybenzoxazines was investigated by the thermogravimetric analysis (TGA) in a nitrogen atmosphere. The curves are given in Figure 6 and the data are listed in Table 2. As can be seen in Figure 6, the temperature at 5% weight loss ($T_{5\%}$) of the five polybenzoxazines is higher than 310°C and the char yield ($CY$) of all samples is higher than 40% at 800°C. As the temperature increases, polybenzoxazines with higher fluorine content exhibit a slower weight loss process, which is

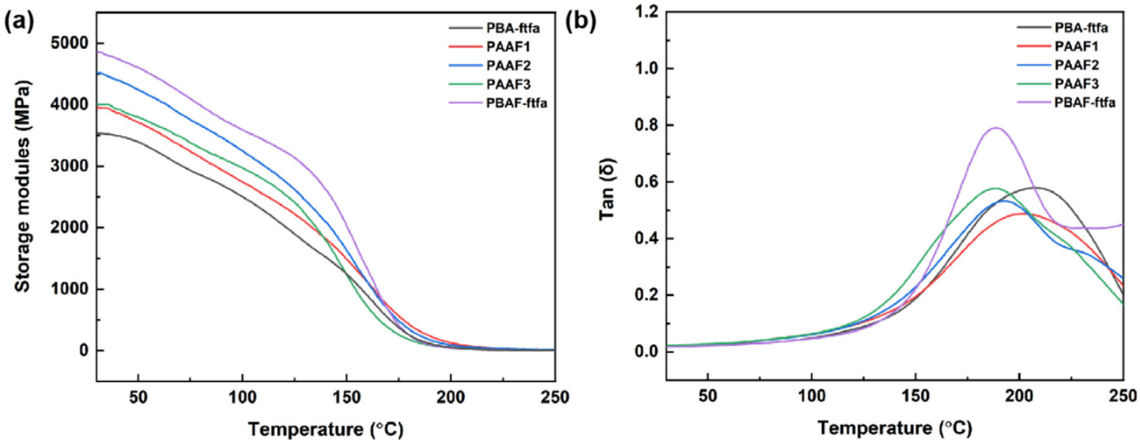


**FIGURE 5** (a) Storage modules and (b) Tan δ of polybenzoxazines. [Color figure can be viewed at wileyonlinelibrary.com]

**TABLE 2** TGA and DMA data of polybenzoxazines.

| Samples | $T_g$ (°C) | $E'$ (MPa, 30°C) | $E'_e$ (MPa) | $\rho_x$ ($\times 10^{-3}$ Mol/cm$^3$) | $T_{5\%}$ (°C) | $T_{10\%}$ (°C) | *CY* (%) |
|---|---|---|---|---|---|---|---|
| PBA-ftfa | 211 | 3542 | - | - | 314 | 341 | 46.7 |
| PAAF1 | 202 | 3952 | 18.52 | 2.70 | 318 | 353 | 46.9 |
| PAAF2 | 196 | 4519 | 19.58 | 2.78 | 326 | 360 | 46.1 |
| PAAF3 | 191 | 4007 | 12.36 | 2.10 | 328 | 367 | 46.5 |
| PBAF-ftfa | 188 | 4858 | 11.62 | 2.01 | 333 | 370 | 42.2 |

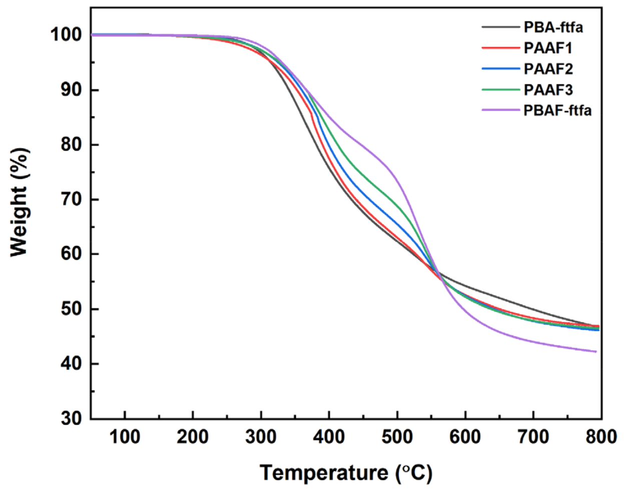


**FIGURE 6** TGA curves of polybenzoxazines. [Color figure can be viewed at wileyonlinelibrary.com]

probably due to the existence of more trifluoromethyl groups. The substitution of hydrogen atoms by fluorine atoms with larger atomic radius results in greater repulsion and spatial resistance between adjacent atoms, and C—F bonds have larger bond energy and lower polarization than C—H bonds.[36] However, due to the smallest crosslinking density of PBAF-ftfa, the *CY* of it at 800°C is the lowest. In general, the introduction of fluorine-containing groups can increase the initial weight loss temperature of the polymer.[37]

## 3.4 | Surface properties of polybenzoxazines

Due to the large dielectric constant of water, the water absorption and hydrophobicity of the polybenzoxazines have a profound influence on the dielectric properties of the material.[38] The surface properties data are listed in Table 3. Three sets of each of the five samples were soaked in deionized water for 1 week, and their mass changes were weighed every 24 h and plotted in Figure 7a. The water absorption of PBA-ftfa for 24 h and a week are 0.354% and 0.773%, which of PBAF-ftfa are 0.305% and 0.418%, and the remaining is somewhere in between. The five samples were also subjected to water contact angle (WCA) measurements to characterize their hydrophobicity, and the results are represented in Figure 7b. All polymers have contact angles higher than 90°, which means they have hydrophobic properties. In addition, it can be observed that as fluorine content increases, the water contact angle gradually increases and the water absorption tends to decrease. The results are mainly attributed to the introduction of fluorine to

TABLE 3 Surface properties and dielectric properties data of polybenzoxazines.

| Samples | Dielectric constant (1 MHz) | Tan (δ) (1 MHz) | Water absorption for 24 h (%) | Water absorption for 7 days (%) | Water contact angle (°) |
|---|---|---|---|---|---|
| PBA-ftfa | 3.28 | 0.01343 | 0.354 | 0.733 | 94.4 ± 1.1 |
| PAAF1 | 3.08 | 0.01205 | 0.353 | 0.696 | 95.1 ± 1.6 |
| PAAF2 | 2.95 | 0.01149 | 0.342 | 0.646 | 95.5 ± 1.0 |
| PAAF3 | 2.73 | 0.01143 | 0.333 | 0.574 | 96.3 ± 1.0 |
| PBAF-ftfa | 2.53 | 0.00841 | 0.305 | 0.418 | 98.2 ± 1.3 |

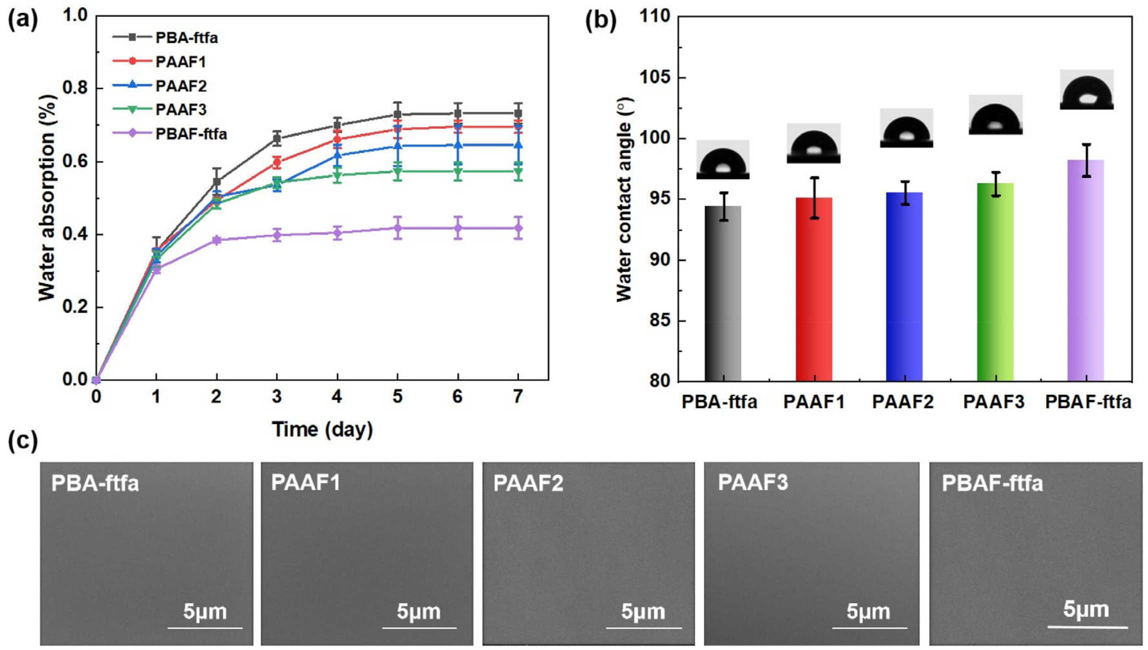


FIGURE 7 (a) Water absorption curves, (b) water contact angle, and (c) SEM images of polybenzoxazines. [Color figure can be viewed at wileyonlinelibrary.com]

reducing the surface energy of materials. The surface morphology of the prepared polybenzoxazines was observed by SEM, and it can be found that the surface was smooth without cracks. The result demonstrates that the synthesized benzoxazine monomers and their mixtures have predominant film-forming properties.

## 3.5 | Dielectric properties of polybenzoxazines

The variation of dielectric constant and dielectric loss with frequency at room temperature for polybenzoxazines is represented in Figure 8 and the data are listed in Table 3. It is well known that the dielectric constant is directly related to the polarizability of the material, which means that it is closely related to the chemical structure of the material. The dielectric constant of PBA-ftfa is 3.28 at 1 MHz, which has been significantly reduced after copolymerization with BAF-ftfa. The dielectric constant of the copolymers decreases with the increase of fluorine content. PBAF-ftfa has the lowest dielectric constant, 2.53 at 1 MHz, this is because PBAF-ftfa contains more trifluoromethyl groups, which are larger than the methyl groups to increase the free volume of the polymer.[39] Besides, C—F bonds have lower polarizability than C—H bonds, so the introduction of more fluorinated structures into polybenzoxazine resins can effectively reduce the number of polar groups per unit volume of the polymer.[6,40,41] The substitution of fluorine reduces the dielectric constant of the resin by decreasing the polarizability and hygroscopicity as well

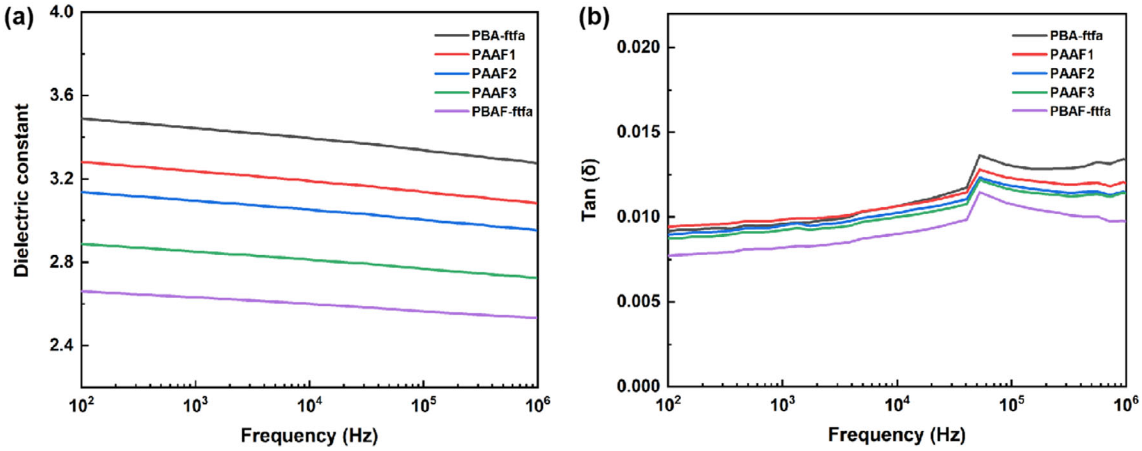


FIGURE 8 (a) Dielectric constant and (b) Tan (δ) of polybenzoxazines. [Color figure can be viewed at wileyonlinelibrary.com]

as increasing the free volume. The dielectric loss of the five samples shows the same pattern of variation as the dielectric constant, the dielectric loss of PBA-ftfa at 1 MHz is 0.01343 and that of PBAF-ftfa is 0.00841.

## 4 | CONCLUSIONS

In conclusion, polybenzoxazines with low dielectric, high heat resistance and good mechanical properties were successfully prepared and the effects of fluorine content on polymer properties were explored by copolymerizing BA-ftfa and BAF-ftfa in different mass ratios. The chemical structure of two monomers was supported by FT-IR and NMR. The results of TGA and DMA indicate that all polymers possess great thermal stability and exhibit a relatively high $T_g$ over than 180°C. Due to the low polarizability and moisture absorption of fluorine, the introduction of fluorine on polybenzoxazine can reduce its dielectric constant and enhance hydrophobic performance. As the fluorine content increases, the dielectric constant and dielectric loss of the polybenzoxazines decreases and the surface properties improve. PBAF-ftfa with the highest fluorine content gives the lowest dielectric constant of 2.53 and dielectric loss of 0.00841 at 1 MHz compared with 3.28 and 0.01343 of PBA-ftfa. Furthermore, the water contact angles of polybenzoxazines are between 94.4° and 98.2°, and the water absorption for 24 h ranges from 0.354% to 0.305%. Therefore, the fluorinated polybenzoxazines with high thermal stability and low dielectric constant were expected to be applied to the low-dielectric materials for large-scale integrated circuits.

## AUTHOR CONTRIBUTIONS

**Xiaoyun Ma:** Data curation; formal analysis; investigation; validation; visualization; writing – original draft. **Xin Lu:** Methodology; project administration); resources; writing – review and editing. **Manlin Yuan:** Validation; writing – review and editing. **Hao Lin:** Validation. **Xinyi Pan:** Validation. **Shiao-Wei Kuo:** Writing – review and editing. **Zhong Xin:** Funding acquisition; resources; supervision.

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## CONFLICT OF INTEREST STATEMENT

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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