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Synthesis and characterization of new inherent photoimageable polyimides based on fluorinated tetramethyl-substituted diphenylmethanediamines

Z.G. Qian[a], Z.Y. Ge[b], Z.X. Li[b], M.H. He[b], J.G. Liu[b], Z.Z. Pang[a], L. Fan[b], S.Y. Yang[b,*]

[a]*School of Materials Science and Engineering, Beijing University of Chemical Technology, Beijing 100029, People's Republic of China*
[b]*State Key Laboratory of Engineering Plastics and Advanced Polymer Materials Laboratory, Center for Molecular Science, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100080, People's Republic of China*

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Abstract

A novel fluorinated tetramethyl-substituted diphenylmethanediamine, α,α-bis(4-amino-3,5-dimethylphenyl)-3′-trifluoromethylphenylmethane (TFPM) was synthesized via a straightforward two-step procedure with high yield, which was then employed to react with 3,3′,4,4′-benzophenonetetracarboxylic dianhydride (BTDA) to afford soluble and inherent photoimageable polyimide BTDA–TFPM [PI(BTDA–TFPM)]. Experimental results indicated that PI(BTDA–TFPM) possesses good combined chemical and physical properties, of which thermal stability with initial thermal decomposition temperature of 526 °C and glass transition temperature of 298 °C. The polyimide has excellent solubility both in strong bipolar solvents, such as NMP and DMAc, and in common organic solvents, such as THF and dioxane, etc. Strong and flexible polymer coatings or films could be prepared by just casting the polymer solution on glass substrate, followed by thermal baking at a temperature lower than 200 °C. The novel polyimide has good electric insulating properties and low dielectric constant and dissipation factor. Fine photopattern could be processed by directly exposing the polyimide coating upon UV i-line, followed by developing with an organic solution developer. © 2002 Published by Elsevier Science Ltd.

Keywords: Photoimageable polyimides; Solubility; Photosensitivity

1. Introduction

Photoimageable polyimides are currently used in advanced microelectronics packaging as passivation layers and α-particle barriers on chip, interlay dielectrics in multilayer structure and dielectric multichip module (MCM-D), stress relief buffer coatings in high-density electronic packaging, etc. [1–5]. The negative photoimaging polyimides are the commonly accepted mode, which are usually formulated from poly(amic acid)s containing photosensitive cross-linking sites in polymer backbones introduced either by chemical bonding or by salt-like linking [6]. However, the major drawbacks of negative photosensitive polyimides for advanced microelectronics applications, i.e. the heavy dimensional shrinkage and the deteriorated mechanical properties of the thermally cured polyimide photopatterns are due to significant evolution of organic volatiles derived from the photosensitive additives and solvents. There is another kind of photosensitive polyimide, the inherent photoimageable polyimide, which is a fully imidized polymer and could be directly photopatterened on exposure to 365 nm (i-line) [7–10]. In contrast to the negative photosensitive polyimide, the inherent photosensitive polyimide does not need addition of any photosensitizers and crosslinking agents, hence, less dimensional shrunken photopatterns could be obtained and the deterioration on the finally cured polyimide could be avoided. The commercially available inherent photosensitive polyimides are prepared by the polycondensation of *ortho* multialkyl-substituted aromatic diamines and 3,3′,4,4′-benzophenonetetracarboxylic dianhydride (BTDA) [11,12]. However, the polyimides usually suffer their limited solubility in organic solvent and the high moisture sensitivity, which hamper their widespread

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* Corresponding author. Tel.: +86-10-6256-4819; fax: +86-10-6255-9373.

E-mail address: public305@infoc3.icas.ac.cn (S.Y. Yang).

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applications in advanced microelectronics manufacturing process.

Recently, many reports describing fully imidized polyimides with improved solubility have appeared in the literature, where solubility enhancement is achieved either by incorporating flexible asymmetric substituents or by introducing bulky pendant groups into the rigid polymer backbone [13–22]. However, only a few papers concentrate to improve the solubility in inherent photoimageable polyimides.

A new fluorinated tetramethyl-substituted diphenylmethanediamine, α,α-bis(4-amino-3,5-dimethylphenyl)-3′-trifluoromethylphenyl methane (TFPM), was synthesized in our laboratory, which was employed in a polycondensation reaction with BTDA to afford a fully imidized polyimide. Photosensitive polyimide resin was formulated with the fully imidized polyimide, which is used to directly photoimage the structure by exposing to UV i-line, followed by developing in organic developer and thermally cured at 200 °C. High quality pattern could be obtained and the finally cured polyimide film exhibited good combined physical and chemical properties, including high thermal stability, excellent mechanical properties, high electrical insulating, low dielectric constant and dissipation factor, as well as low moisture uptake.

2. Experimental

2.1. Materials

2,6-Dimethylaniline, 3,3′,4,4′-benzophenonetetracarboxylic dianhydride and *m*-trifluoromethylbromobenzene were purchased from Acros Organics Chemicals Co. and purified by recrystallization from acetic anhydride prior to use. Benzaldehyde, hydrochloric acid and *iso*-propyl alcohol were purchased from Beijing Chemical Reagents Co. and were used without further purification. α,α-Bis(4-amino-3,5-dimethylphenyl)diphenylmethane (TMDA) was synthesized according to the literature [23]. Commercially available *N*-methyl-2-pyrrolidinone (NMP), *N*,*N*-dimethylformamide (DMF) were purified by vacuum distillation prior to use.

2.2. Monomer synthesis

2.2.1. m-Trifluoromethylbenzaldehyde

m-Trifluoromethylbromobenzene (40.0 g) dissolved in 50 ml of anhydrous ether was added dropwise to a suspension containing 10 g of magnesium and 100 ml of anhydrous ether. After the reaction mixture was processed for 4 h, *N*-methylformanilide (24.0 g) was added. The mixture was stirred for 3 h, then 20 ml of aqueous sulfuric acid (10%) was added dropwise. Finally, 100 ml of ether was added followed by the separation of aqueous layer. The organic layer was neutralized with 10% aqueous sodium carbonate, then washed twice with water and dried over anhydrous magnesium sulfate. The liquid product was obtained by distillation (36 g, 40%). Bp: 110 °C (4 mm Hg). ^{1}H NMR ($CDCl_3$, δ ppm): 10.08 (s, 1H), 8.15–8.07 (t, 1H), 7.90–7.87 (d, 1H), 7.74–7.05 (d, 1H). MS (EI intensity %) 174 (62, M +). Elemental analysis: calcd for $C_8H_5F_3O$: C, 55.18%; H, 2.89%. Found: C, 55.24%; H, 2.79%.

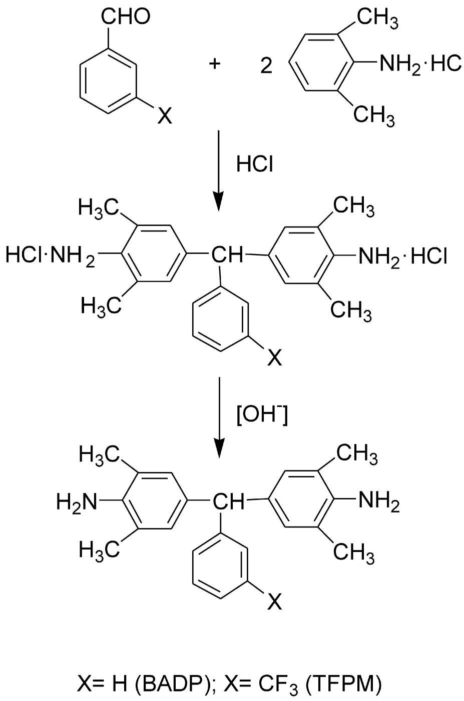
2.2.2. α,α-Bis(4-amino-3,5-dimethylphenyl)-3′-trifluoromethylphenylmethane (TFPM)

2,6-Dimethylaniline (48.47 g, 0.4 mol) was heated at 130 °C in nitrogen to which 3-trifluoromethylbenzaldehyde (21.6 g, 0.18 mol) dissolved in 33 ml (12N) of hydrochloric acid was added dropwise over a period of 1.5 h. The reaction mixture was continually refluxed at 100 °C for 18 h. After cooling to room temperature, 7.92 g of 20% aqueous sodium hydroxide was added and the resulting suspension was then poured into excess ethanol to afford pale blue precipitate. The solid was filtered, washed thoroughly with distilled water, and dried at 60 °C overnight. The product was purified by recrystallization twice in ethanol to afford pale crystal (41.7 g, 58.2%). Mp: 169 °C (DSC in air). FT-IR (KBr, cm^{-1}): 3479, 3395, 2972, 2925, 1623, 1489, 1329, 1161, 1121. ^{1}H NMR ($CDCl_3$, δ ppm): 7.53 (s, 1H), 7.50–7.45 (t, 1H), 7.36–7.33 (d, 2H), 6.50 (s, 4H), 5.25 (s, 1H), 2.00 (s, 12H). Electron-impact mass spectrometry (MS; *m*/*e*, % relative intensity): 397 (M − 1, 100). Elemental analysis: calcd for $C_{24}H_{25}F_3N_2$: C, 72.34%; H, 6.32%; N, 7.03%. Found: C, 72.58%; H, 6.13%; N, 7.11%.

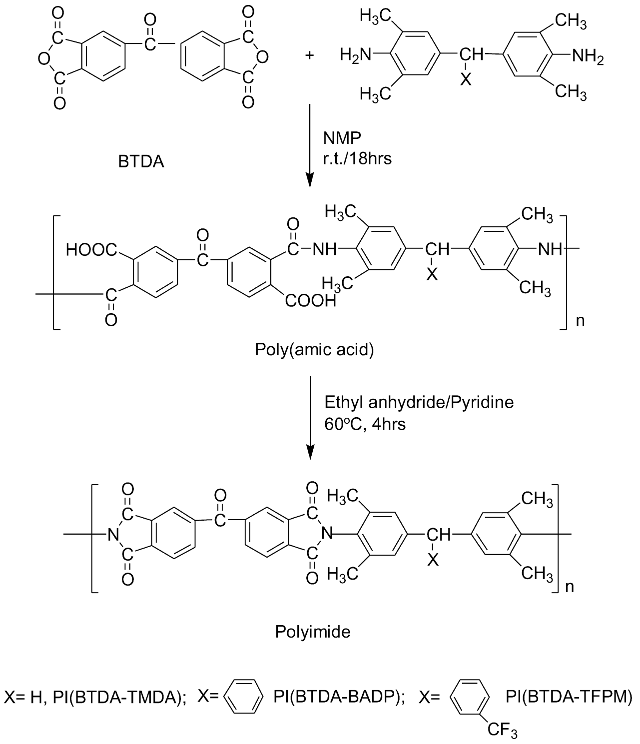
2.3. Polymer synthesis

In a typical experiment, TFPM (39.8 g, 0.1 mol) was dissolved in 280 ml of NMP to which BTDA (32.55 g, 0.1 mol) was added stepwise, followed by addition of 8 ml of NMP. The mixture was stirred in nitrogen at room temperature for 18 h to yield a viscous poly(amic acid) (PAA) solution with 20 wt% solid content. The PAA solution was chemically converted into the polyimide by the addition of 100 ml of an acetic anhydride/pyridine (5/4 v/v) mixture, followed by stirring at 60 °C for 4 h. The homogeneous polyimide solution was poured into an excess of methanol to produce a solid polymer, which was collected and washed with methanol thoroughly, and dried at 80 °C in vacuum to give polyimide (BTDA–TFPM) (67.9 g, yield 99.2%). FT-IR (KBr, cm^{-1}): 2926, 1782, 1726, 1675, 1605, 1486, 1441, 1371, 976, 863. ^{1}H NMR ($CDCl_3$, δ ppm): 8.28–8.12 (m, 6H), 7.57 (s, 1H), 7.54–7.50 (t, 1H), 7.40–7.28 (d, 2H), 6.56 (s, 4H), 5.56 (s, 1H), 2.17 (s, 12H). Elemental analysis: calcd for $(C_{41}H_{27}F_3N_2O_5)_n$: C, 71.92%; H, 3.98%; N, 4.09%. Found: C, 71.99%; H, 3.89%; N, 4.14%.

In comparison, polyimide (BTDA–TMDA) and polyimide (BTDA–BADP) were prepared in similar procedure with polyimide (BTDA–TFPM) except that TMDA or



Scheme 1. Synthesis of the tetramethyl-substituted diamine BADP and TFPM.



Scheme 2. Synthesis of inherent photoimageable polyimides.

BDAP was used instead of TFPM as the aromatic diamine monomer.

2.4. Measurements

Fourier transform infrared (FT-IR) spectra were recorded on a Perkin Elmer 782 Fourier transform spectrophotometer. Ultraviolet–visible (UV–vis) spectra were recorded on a Hitachi U-3210 spectrophotometer. Differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) were carried out with a Perkin Elmer TGA-7 thermogravimetric analyzer in nitrogen at a heating rate of 20 °C/min. Inherent viscosities were measured in 0.5% solutions in NMP at 25 °C. Moisture uptake was measured by weighing the changes in a specimen before and after immersion in boiling water for 30 min.

2.5. Photosensitivity and photopatterning

Exposure was performed on a KG-706 mask-aligner in hard contact mode and spectral output at 436, 405 and 365 nm ($g + h + I$, broadband). A combination of a high-resolution mask together with a multidensity mask was used. Spectral sensitivities were determined using Oriel Hg-line interference filters. Photosensitivity, referred to the 400 nm probe of the OAI light sensitivity meter for broadband exposure, was obtained by developing the exposed polyimide films (2.5 μm) with a mixed organic solvent of γ-butyrolactone and DMF at room temperature for 30 s, and then rinsed with de-ionized water. Photopattern was gained by exposing the polyimide film followed by development and rinse. After postbaking at 200 °C for 1 h, the micro-pattern was photographed by a scanning electron microscope. Layer thickness was controlled to ± 10% for thin layers (1.0 μm) and 5% for thicker layers (5–10 μm).

3. Results and discussion

3.1. Monomer synthesis

The synthesis of monomer *m*-trifluoromethylbenzaldehyde was prepared by reacting *m*-trifluoromethylbenzene magnesium bromide with *N*-methylformanilide according to the literature [10]. The diamine TFPM was synthesized in a one-step process (Scheme 1), in which 1 mol of *m*-trifluoromethylbenzaldehyde was coupled with 2 mol of 2,6-dimethylaniline in the presence of hydrochloride acid to afford TFPM·HCl salt. The pure TFPM compound was released by addition of aqueous sodium hydroxide solution and purified by recrystallization in anhydrous ethyl alcohol. The chemical structure of TFPM was confirmed by elemental analysis, FT-IR, ^1H NMR and mass spectrum. The elemental analysis values are in good agreement with the calculated ones and the molecular weight determined by mass spectrum is 397, being the same as the calculated one.

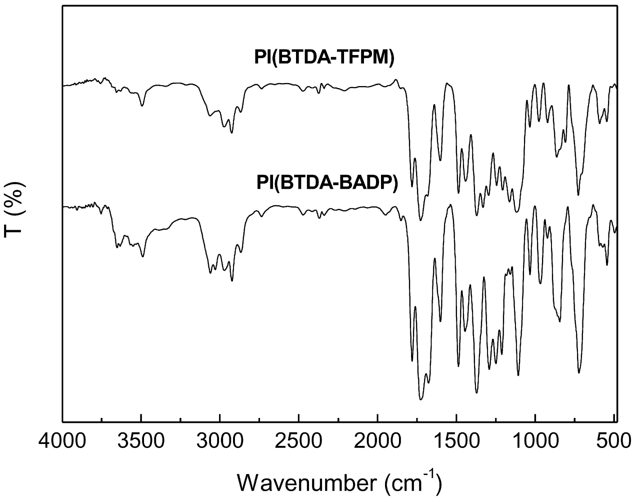


Fig. 1. FT-IR spectra of PI(BTDA–TFPM) and PI(BTDA–BADP).

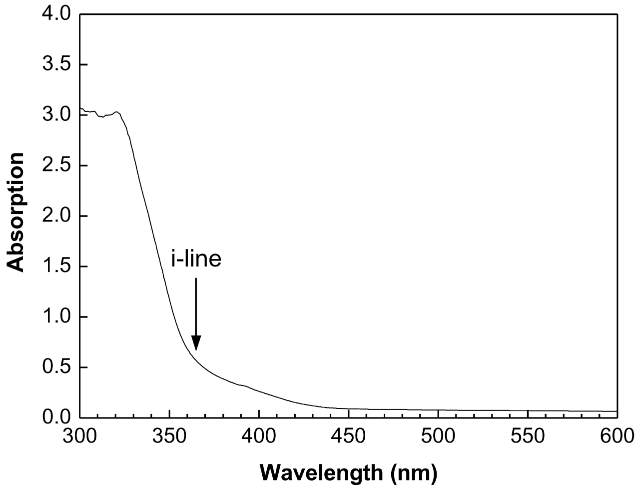


Fig. 3. UV–vis spectrum of PI(BTDA–TFPM) film.

3.2. Polymer synthesis

Polyimide was synthesized from the condensation of TFPM with BTDA by a two-step pathway (Scheme 2). Poly(amic acid) solution at 20% of solid content in NMP was first prepared at room temperature, which was then chemically imidized in the presence of acetic anhydride/pyridine mixture to produce the fully imidized polymer. The polymer was isolated and purified by precipitation of the polyimide solution in excess of ethyl alcohol, and then washed thoroughly with ethanol.

The yellowish polymers have elemental analysis values being exactly the same as expected. The inherent viscosities for the polyimides are in the range of 0.41–0.49 dl/g. Fig. 1 shows the FT-IR spectra of PI(BTDA–TFPM) compared with PI(BTDA–BADP). The characteristic absorptions for the asymmetric and symmetric C=O stretching vibrations of imide groups can be observed. The absorption at 1371 cm^{-1} is assigned to the C–N stretching vibration of imide structure. No absorption for the C=O stretching vibrations of amide groups was detected, suggesting the fully imidization of poly(amic acid).

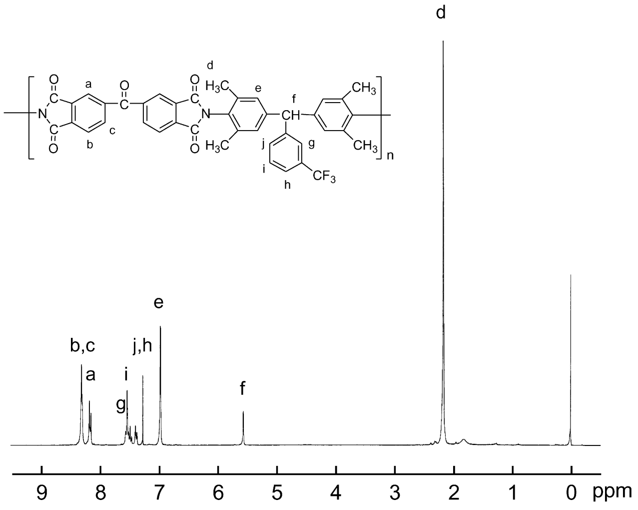


Fig. 2. ^{1}H NMR (CDCl$_3$, 300 MHz) spectrum of PI(BTDA–TFPM).

Fig. 2 shows the ^{1}H NMR spectrum of PI(BTDA–TFPM), in which the assignments of protons are in good agreement with the expected chemical structure.

The UV–vis spectrum of PI(BTDA–TFPM) film is shown in Fig. 3. The film with thickness of 2.5 μm was employed, which was prepared by spin-coating a polyimide solution on quartz surface, followed by thermally curing at 120 °C for 30 min in an oven. It can be seen that the maximum absorption appeared at 330 nm and there is a relatively strong absorption at 365 nm (i-line). The absorption cut-off wavelength is located at 425 nm, which is lower than the g-line (436 nm) absorption.

3.3. Polymer solubility

Polymer solubility was qualitatively determined by dissolving 1.5 g of solid polymers in 8.5 g of organic solvent, which was mechanically stirred under nitrogen for 24 h at room temperature to determine the solubility (Table 1). PI(BTDA–BADP) and PI (BTDA–TFPM) both could be easily dissolved in strong aprotic solvents, such as NMP, DMF, DMAc, and in common organic solvents, such as *m*-cresol, γ-butyrolactone and chloroform. On the other hand, PI(BTDA–TMDA) was partly soluble in *m*-cresol and chloroform although it could be dissolved in the strong aprotic solvents. Therefore, low boiling point organic solvents, such as cyclopentanone and chloroform, could be employed to dissolve PI(BTDA–TFPM) and PI(BTDA–BADP) to afford homogeneous polymer solutions, which could process coatings or thin films at relatively low curing temperature. For instance, PI(BTDA–BADP) and PI(BTDA–TFPM) in 1,2-dichloroethane (mp 83.5 °C) could be thermally processed to produce thin polyimide coating at lower temperature than the polymer in NMP solution (mp 202 °C). On account of the good solubility in low boiling point solvent, polyimide coatings or thin films

Table 1
The solubility of polyimides in different organic solvents

Solvent[a]	PI(BTDA–TMDA)	PI(BTDA–BADP)	PI(BTDA–TFPM)
NMP	++	++	++
DMAc	++	++	++
DMF	++	++	++
m-Cresol	+	++	++
Chloroform	+	++	++
1,2-Dichloroethane	+	++	++
γ-Butyrolactone	+	++	++
Cyclopentane	++	+	++
Cyclohexanone	+	+	++
THF	−	++	++
1,4-Dioxane	+	+	++
Acetone	−	+	++
Iso-propylalcohol	−	−	−
n-Hexane	−	+	+

Solubility was determined by mixing 1.5 g of polymer with 8.5 g of solvent, followed by stirring for 24 hrs. (++) soluble at room temperature; (+) soluble with heating at 80 °C; (−) dissoluble.

[a] NMP: 1-methyl-2-pyrrolidone; DMF: *N*,*N*-dimethylformamide; THF: tetrahydrofuran.

could be processed at relatively low temperature (180–200 °C) by completely removing the trace of solvent in the fully imidized polyimide solution. The excellent solubility of these two polyimides might be due to their non-coplanar structures and presence of bulky pendant phenyl groups besides the four methyl groups in the polymer backbone, resulting in the reducing of the inter-chain interaction.

3.4. Thermal stability

The results of the decomposition temperature for the polyimides, which determined by thermal gravimetric analysis (TGA), are listed in Table 2. The onset decomposition temperature (T_d) was measured at 526 °C for PI(BTDA–TFPM), only 6 °C lower than that of PI(BTDA–BADP) and PI(BTDA–TMDA). The decomposition temperature at 5% weight loss (T_5) for PI(BTDA–TFPM) is 532 °C, which is same as that for PI(BTDA–TMDA) and higher than that for PI(BTDA–BADP) (526 °C). In addition, the decomposition temperature at 10% weight loss (T_{10}) for PI(BTDA–TFPM) is 574 °C, much higher than that for PI(BTDA–TMDA) and PI(BTDA–BADP). It is indicated that the thermal stability of PI(BTDA–TFPM) is better than the latter. Moreover, according to the TGA curves, there is no trace of weight loss detected for PI(BTDA–TFPM) before it was heated at temperatures as high as 450 °C.

The DSC curves of the polyimides are shown in Fig. 4. The glass transition temperature (T_g) for PI(BTDA–TFPM) (ca. 298 °C) is about 13–20 °C lower than the others, which probably is due to the presence of trifluoromethyl functional groups in the polymer side chain. It is apparently indicated that the T_g value of polyimide is closely related to its polymer molecular structure as well as its morphology and aggregation: the more asymmetric the molecular structure in the polymer chain, the lower the T_g value of the polymer. On the other hand, PI(BTDA–BADP) showed T_g 7 °C higher than PI(BTDA–TMDA) (311 °C), implying that the thermal stability of polymer can be improved by introducing the pendant phenyl groups in polyimide backbone.

3.5. Electrical and dielectric properties

PI(BTDA–TFPM) exhibited dielectric constant (2.68 at 1 MHz) lower than the commercially available polyimide derived from PMDA and ODA polyimide (3.40 at 1 MHz), as well as that for PI(BTDA–TMDA) (2.98 at 1 MHz) and PI(BTDA–BADP) (2.89 at 1 MHz) (Table 3). The dissipation factors are of the order of $2–4 \times 10^{-3}$. The surface resistance and the volume resistance for PI(BTDA–TFPM)

Table 2
Decomposition temperatures of the polyimides derived from BTDA and different diamines

Decomposition temperature (°C)	PI(BTDA–TMDA)	PI(BTDA–BADP)	PI(BTDA–TFPM)
T_d[a]	532	532	526
T_5[b]	532	526	532
T_{10}[c]	548	569	574

[a] T_d: the onset decomposition temperature.

[b] T_5: the decomposition temperature at 5% weight loss.

[c] T_{10}: the decomposition temperature at 10% weight loss.

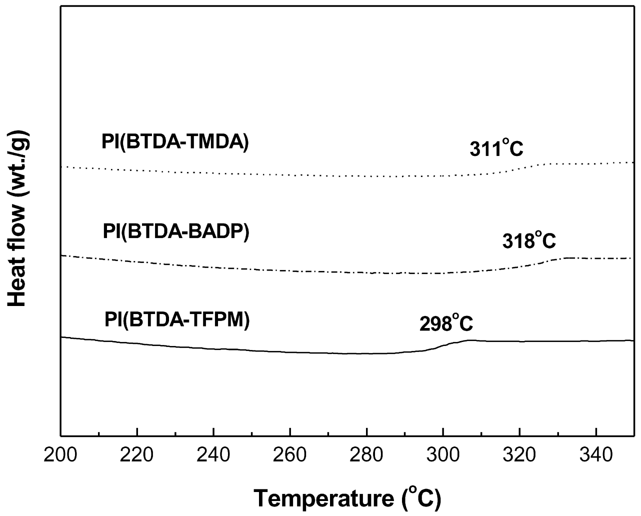


Fig. 4. DSC curves of the polyimides derived from BTDA and different diamines.

are $2.68 \times 10^{15}\ \Omega$ and $2.72 \times 10^{16}\ \Omega$ cm, respectively, higher than those for the others.

In addition, PI(BTDA–TFPM) has a much lower moisture uptake (0.21 wt%), which is half of the value for PI(BTDA–TMDA) polyimide (0.54 wt%) and much lower than the commercial Probimide 400 (2.0 wt%) [24]. The low moisture absorption could ensure the polymer possessing stable and constant electric insulating and dielectric performance. The low dielectric constant and the low moisture absorption of PI(BTDA–TFPM) are attributed to the presence of the fluorinated substitutes in the polymer chains.

3.6. Photosensitivity and photopatterning

PI(BTDA–TFPM) exhibited the UV–vis absorption peaked at 365 nm (i-line). Experimental results indicated that the material is primarily photosensitive to the i-line exposure and compatible with exposure in the contact or proximity mode. Fine patterns could be processed by UV exposure of the polymer coating on silicon wafer, followed by developing with an organic developer composed of γ-butyrolactone and DMF as main solvents (Fig. 5). The polymer coated silicon wafer was prepared by spin coating the polymer solution in NMP on silicon wafer, followed by thermal baking at 200 °C to completely remove the solvent.

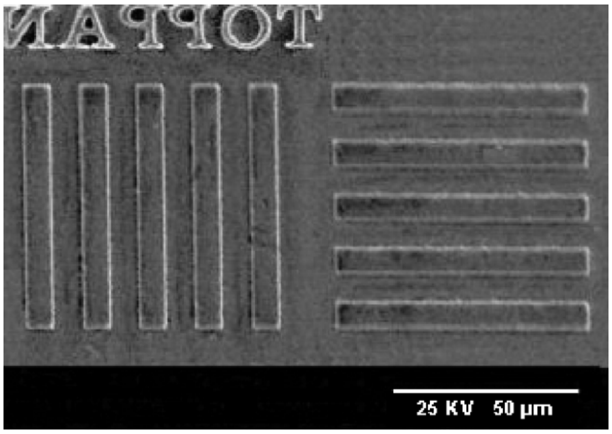


Fig. 5. SEM photograph of the pattern from the system containing PI(BTDA–TFPM).

The pattern structure has only 8–10% of dimensional shrinkage upon final thermally curing (300–350 °C), indicating that the geometrical distortion of the photopattern is minimal. Fig. 6 shows the characteristic exposure curves of the inherent photosensitive polyimides. Similar photosensitive behaviors can be observed. The photosensitivity for PI(BTDA–TFPM) was determined to be about 150 mJ/cm^2, which is lower than that for PI(BTDA–TMDA) (190 mJ/cm^2) and PI(BTDA–BADP) (170 mJ/cm^2). The aspect ratios of 1.0 and 1.5 were obtained for films over 5 and 1–5 μm thick, respectively.

4. Conclusions

A novel fluorinated diamine monomer, α,α-bis(4-amino-3,5-dimethylphenyl)-3′-trifluoromethylphenylmethane (TFPM), was synthesized via a two-step procedure, which was used to prepare inherent photoimageable polyimides with pendant phenyl groups and CF_3 groups in the polymer backbone. Experimental results indicated that the polyimides exhibited good combined chemical and physical properties. The polyimide has an excellent thermal stability with initial thermal decomposition temperature of 526 °C and glass transition temperature of 298 °C. The polyimide also showed the high electric insulating properties, low dielectric constant and dissipation factor as well as low moisture absorption. Fine pattern could be processed by exposing the polyimide coating to UV i-line, followed by

Table 3
Electrical and dielectric properties of the polyimides derived from BTDA and different diamines (25 μm films)

	PI(BTDA–TMDA)	PI(BTDA–BADP)	PI(BTDA–TFPM)
Surface resistance (Ω)	2.04×10^{15}	2.42×10^{15}	2.68×10^{15}
Volume resistance (Ω cm)	2.25×10^{16}	2.46×10^{16}	2.72×10^{16}
Dielectric constant (1 MHz)	2.98	2.89	2.68
Dissipation factor	0.0035	0.0029	0.0022
Dielectric strength (kV/mm)	100	120	128
Moisture uptake (wt%)	0.54	0.24	0.21

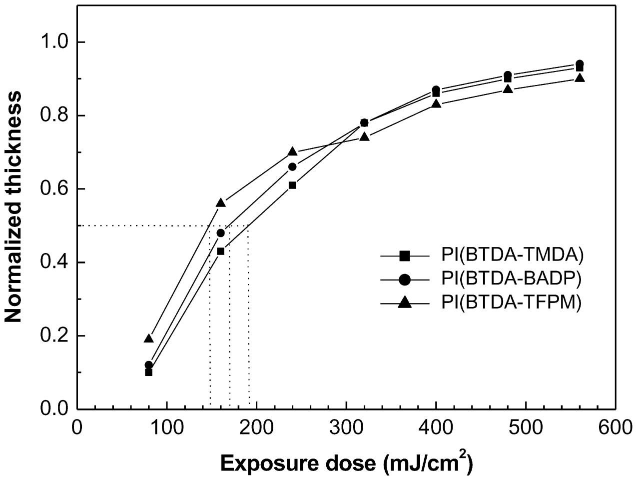


Fig. 6. Characteristic exposure curves of the polyimides derived from BTDA and different diamines.

developing with organic developer. The photosensitive polyimide exhibits minimal dimensional shrinkage of 8–12% and has a photosensitivity of about 150 mJ/cm^2.

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