

A Photosensitive Semi-Alicyclic Poly(benzoxazole) with High Transparency and Low Dielectric Constant

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ABSTRACT: A photosensitive semi-alicyclic poly(benzoxazole) based on poly(*o*-hydroxy amide) containing adamantyl units (PAHA) and 1,3,5-tris[(2-vinyloxy)ethoxy]benzene (TVEB) as an acidolytic de-cross-linker and a photoacid generator, diphenyliodonium 9,10-dimethoxyanthracene-2-sulfonate (DIAS), has been developed. PAHA with a weight-average molecular weight of 24 100 was prepared from 1,3-adamantanedicarbonyl chloride (ADC) and 4,4'-(hexafluoroisopropylidene)bis(*o*-aminophenol) (6FAP) in the presence of lithium chloride in *N*-methylpyrrolidinone (NMP) at 0 °C for 12 h. By thermal treatment, PAHA was easily converted to semi-alicyclic poly(benzoxazole) (PABO) that was insoluble in organic solvents and showed high thermal stability ($T_{d5} = 520$ °C under N_2). The UV-vis spectra of PAHA and PABO indicated excellent transparency at wavelengths above 320 and 400 nm, respectively. The average refractive index of PABO was 1.523, and the dielectric constant estimated from the refractive index was 2.55 at 1 MHz. This value is significantly lower than those of conventional wholly aromatic PBOs. The photosensitive PABO precursor, PAHA containing 15 wt % TVEB and 5 wt % DIAS, showed a sensitivity of 40 mJ/cm² and a contrast of 4.0 when it was exposed to a 365 nm light (i-line) and developed with a 2.38 wt % aqueous tetramethylammonium hydroxide solution (TMAH_{aq}) at 25 °C. A fine positive image of 10 μm line-and-space pattern was also printed in a film which was exposed to 70 mJ/cm² of i-line by contact-printing mode. The positive image in PAHA was converted to the positive image in the PABO film by the thermal treatment without pattern deformation.

Introduction

Poly(benzoxazole)s (PBOs) have been used as protection and insulation layers for very large scale integration circuits (VLSI) and multichip modules for computers owing to their high thermal stability and excellent mechanical property.^{1–4} Photosensitive PBOs (PSPBOs)^{5–7} have been developed to simplify the processing steps because phenolic hydroxyl groups in a precursor of the PBO provide adequate solubility toward an aqueous alkaline developer. Up to now PSPBO based on poly(*o*-hydroxy amide) (PHA) derived from 4,4'-(hexafluoroisopropylidene)bis(*o*-aminophenol) (6FAP) and 4,4'-oxybis(benzoic acid) derivatives^{8,9} with a photosensitive compound is widely used.

Since the lower dielectric constants are required to increase the circuit speed,⁴ many researchers have reported candidates for low dielectric materials,^{10–12} most of which have thermal stability but do not possess the function of photosensitivity. Generally, PBO is considered as relatively lower dielectric material than polyimide due to the absence of carbonyl group in the structure. Furthermore, high sensitivity of photopolymers is an important parameter for the design of photoresist materials. Highly sensitive resists require the high transparency of the matrices at certain irradiation wavelengths, which is desirable to maximize the throughput of wafers in exposure tools. Alicyclic PBOs (PABOs) are interesting materials for optoelectronics and interlayer dielectrics. Adamantyl units will be effective to decrease a conjugation in a main chain and a molecular density because of their sp³ character of carbons and bulkiness,^{13–15} respectively, giving PABOs with high transparency and low dielectric constant,

while maintaining a high thermal stability of the polymer. In the field of photolithographic techniques, chemical amplification is crucial to improve both sensitivity and contrast. Especially, chemical amplification in positive working is more requested than that in negative working due to effective elimination of dark spots on a chip after development.

In this paper, we report the synthesis of PABO with high transparency and low dielectric constant based on corresponding semi-alicyclic PHA (PAHA) which is prepared from 1,3-adamantanedicarbonyl chloride (ADC) and 6FAP. And its application to a new PSPBO consisting of PAHA, 1,3,5-tris[(2-vinyloxy)ethoxy]benzene (TVEB) as an acidolytic decross-linker, and diphenyliodonium 9,10-dimethoxyanthracene-2-sulfonate (DIAS) as a photoacid generator in positive-tone chemically amplification is described here as well.

Experimental Section

Materials. Tetrahydrofuran (THF) was distilled from sodium metal and benzophenone ketyl. *N*-Methyl-2-pyrrolidinone (NMP) was dried over calcium hydride, distilled under reduced pressure, and stored over 4 Å molecular sieves. 6FAP was purified by recrystallization from THF. ADC was prepared by reaction of 1,3-adamantanedicarboxylic acid with excess amounts of thionyl chloride, followed by recrystallization from *n*-hexane. A photoacid generator DIAS and an acidolytic decross-linker TVEB were kindly donated by Toyo Gosei Co. Ltd. and Tokyo Ohka Kogyo Co. Ltd., respectively. The other reagents and solvents were used as received.

Synthesis of 5,5'-(Hexafluoroisopropylidene)bis(2-hydroxyadamantanecarboanilide). To a solution of lithium chloride (0.653 g, 15.4 mmol) and 6FAP (2.56 g, 7.00 mmol) in 20 mL of NMP was added adamantane-1-carbonyl chloride (2.92 g, 14.7 mmol). The solution mixture was stirred at 0 °C for 3 h and then poured into water. The precipitated solid was collected and dried in vacuo at 100 °C for 8 h. The product

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was purified by recrystallization from mixture of toluene and THF. The yield was 3.1 g (64%). Sublimation point: 348–350 °C. $^1\text{H NMR}$ (DMSO- d_6 , δ , ppm): 10.32 (s, 2H, OH), 8.39 (s, 2H, NHCO), 8.11 (s, 2H, Ar), 6.87 (dd, $J = 12.9, 8.4$ Hz, 4H, Ar), 2.01 (s, 6H, CH), 1.88 (s, 12H, COC(CH $_2$) $_3$), 1.69 (s, 12H, CHCH $_2$ CH). Elemental analysis: Calcd for C $_{37}$ H $_{40}$ F $_6$ N $_2$ O $_4$: C, 64.34; H, 5.84; N, 4.06; Found: C, 64.62; H, 6.07; N, 4.07.

Synthesis of PAHA from 6FAP and ADC. To a solution of lithium chloride (0.0933 g, 2.20 mmol) and 6FAP (0.366 g, 1.00 mmol) in 1.5 mL of NMP under nitrogen was added ADC (0.261 g, 1.00 mmol) at once. And the mixture was stirred at 0 °C for 12 h under a nitrogen atmosphere. The resulting solution was poured into water. The precipitated polymer was filtered off and washed with water several times, followed by drying in vacuo. The yield was almost quantitative (>98%). $^1\text{H NMR}$ (DMSO- d_6 , δ , ppm): 10.28 (s, 2H, OH), 8.53 (s, 2H, NHCO), 8.08 (s, 2H, Ar), 6.94 (d, $J = 8.7$ Hz, 2H, Ar), 6.87 (d, $J = 8.7$ Hz, 2H, Ar), 2.19–1.67 (m, 14H, CH $_2$ or CH). IR (KBr, ν , cm $^{-1}$): 3424 (OH), 2915, 2857 (CH $_2$, CH), 1654 (C=O), 1253 (CF $_3$). Elemental analysis: Calcd for C $_{27}$ H $_{24}$ F $_6$ N $_2$ O $_4$ ·1.13H $_2$ O: C, 56.41; H, 4.61; N, 4.87; Found: C, 56.98; H, 4.50; N, 4.30.

Dissolution Rate. TVEB and DIAS were added to a PAHA solution in cyclohexanone to construct a photosensitive polymer. The polymer film spin-cast from the solution (15 wt % concentration) on a silicon wafer was prebaked at 90–140 °C for 5 min and then exposed to a filtered super-high-pressure mercury lamp at 365 nm (i-line), followed by postexposure baking at the same temperature for 5 min. The exposed film was developed with 2.38 wt % of tetramethylammonium hydroxide (TMAH $_{aq}$) solution at 25 °C to determine a dissolution rate ($\text{\AA}/\text{s}$).

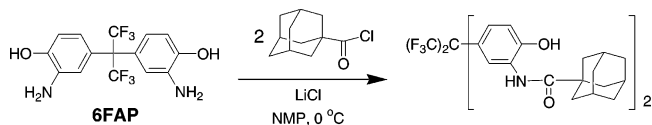
Photosensitivity. A 2.0 μm thick photosensitive polymer film on a silicon wafer was exposed to i-line and then developed with 2.38 wt % of TMAH $_{aq}$ at 25 °C for 60 s followed by rinsing with water. A characteristic curve was obtained by plotting a normalized film thickness as a function of exposure dose (mJ/cm 2).

Measurements. The infrared spectroscopy (IR) was taken with Horiba FT-210 spectrophotometer. The ^1H nuclear magnetic resonance (NMR) spectrum was recorded on a Bruker GPX300 (300 MHz) spectrometer. Thermogravimetric (TG) and differential scanning calorimetry (DSC) were performed on a Seiko TG/DTA 6300 and DSC 6200, respectively, at a heating rate 5 °C/min under a nitrogen stream. Ultraviolet–visible spectroscopy (UV–vis) was performed on a Jasco V-650 spectrophotometer. Number- and weight-average molecular weights (M_n and M_w) were determined by gel permeation chromatography (GPC) with a Tosoh HLC-8120 GPC system equipped with polystyrene gel columns (TSK GELS; GMH $_{HR}$ -M, and GMH $_{HR}$ -L) at 40 °C in DMF (containing 0.01 M LiBr) at a flow rate of 1.0 mL/min, calibrated with polystyrene standards. The film thickness on silicon wafers was measured by a Veeco Instrument Dektak 3 surface profiler. The field emission scanning electron microscope (SEM) was taken with a Hitachi S-800 scanning electron microscope with 15 kV accelerating voltage for imaging. Refractive indices of PABO films formed on quartz substrates were measured at a wavelength of 1.320 μm at room temperature with a Metricon model PC-2000 prism coupler. Using linearly polarized laser with parallel (TE: transverse electric) and perpendicular (TM: transverse magnetic) polarization to the film plane, the in-plane (n_{TE}) and out-of-plane (n_{TM}) refractive indices and the film thickness of the samples were determined. The dielectric constant (ϵ) at 1.0 MHz frequency was calculated from the equation as follows: $\epsilon = 1.10 n_{AV}^2$, where n_{AV} is average refractive index (i.e., $n_{AV} = (2n_{TE} + n_{TM})/3$).

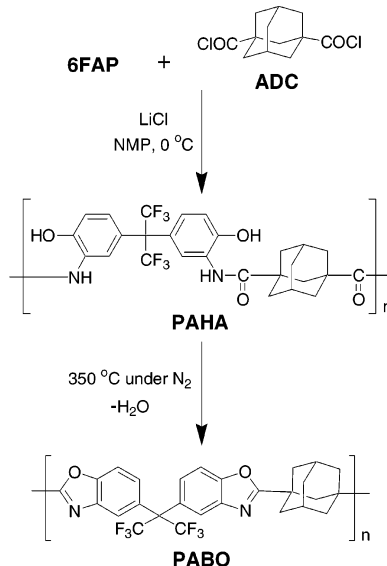
Results and Discussion

Since i-line is used as exposure source in PSPBOs, the transparency of PAHA in the region of i-line is important to improve the sensitivity. When *N*-substituent groups of *o*-hydroxybenzamide are replaced from aryl derivatives to alicyclic ones, the transparency of

Scheme 1. Synthesis of *N*-Adamantyl-2-hydroxybenzamide Derivative as a PAHA Model



Scheme 2. Synthesis of PAHA and PABO



the corresponding *o*-hydroxybenzamide increases in the area of i-line because of decreasing conjugation between 2-hydroxybenzamide and the *N*-substituent group. Adamantane is known as a robust alicyclic compound with relatively high thermal resistance. Thus, an adamantyl group was selected as an *N*-substituent (Scheme 1). The UV–vis spectra of *N*-phenyl-2-hydroxybenzamide and *N*-adamantyl-2-hydroxybenzamide derivatives shown in Figure 1 clearly indicate that the *N*-adamantyl group is very effective to improve the transparency of *N*-substituent-*o*-hydroxybenzamide.

Syntheses and Characterizations of PAHA and PABO. Polycondensation of 6FAP with ADC was carried out in the presence of lithium chloride in NMP at 0 °C for 12 h (Scheme 2). Polycondensation proceeded smoothly to give a desired PAHA with a weight-average molecular weight of 24 100 and a polydispersity of 3.2.

The structure of the polymer was identified as the corresponding PAHA by ^1H NMR and IR spectroscopy.

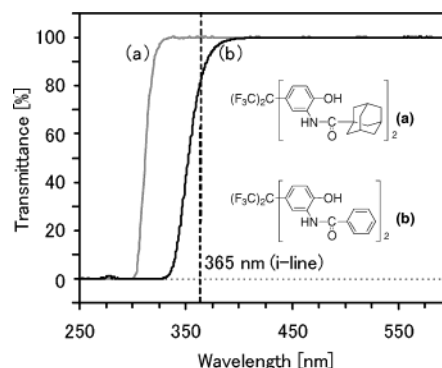


Figure 1. Transparencies of (a) alicyclic compound (adamantyl) and (b) aromatic compound (phenyl) as *N*-substituent groups of *o*-hydroxybenzamide in THF at 1.0 μM . Inset shows chemical structures of (a) and (b).

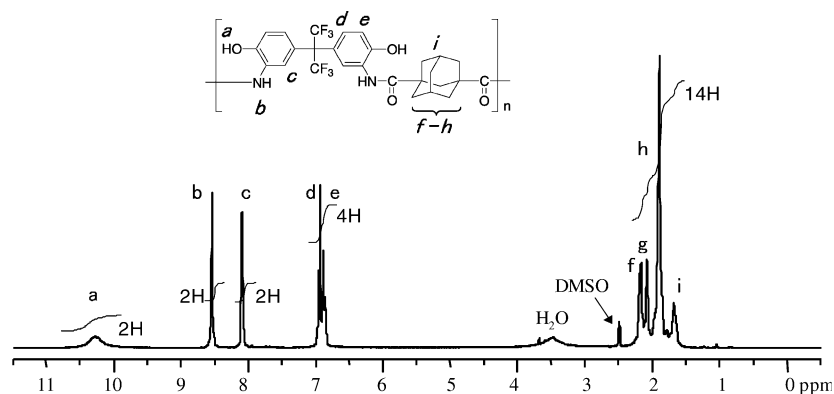


Figure 2. ^1H NMR spectrum of PAHA in $\text{DMSO-}d_6$ at $40\text{ }^\circ\text{C}$.

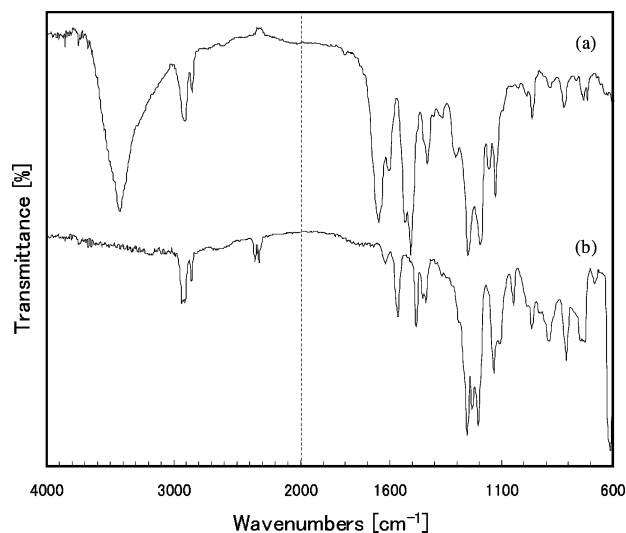


Figure 3. IR spectra of (a) PAHA and (b) PABO on a silicon wafer.

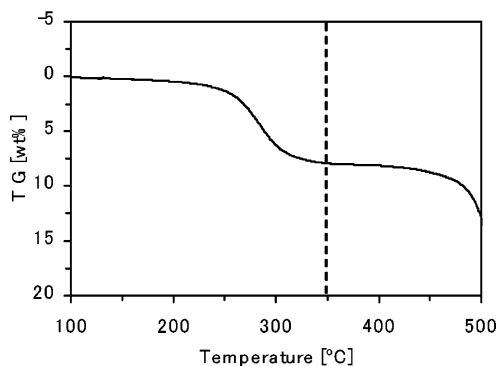


Figure 4. TG trace of PAHA. The heating rate was $5\text{ }^\circ\text{C}/\text{min}$ under nitrogen.

The ^1H NMR spectrum of PAHA is presented in Figure 2 with a good assignment of each resonance. The peaks due to the hydroxyl, amide, and aromatic protons are observed at 10.28 (broad), 8.53 (singlet), and 8.08–8.84 ppm (singlet and quartet), respectively, while adamantyl protons appeared at 2.19–1.67 ppm. The IR spectrum shows characteristic O–H and amide absorptions at 3400 and 1654 cm^{-1} , and no peak corresponding to the ester unit is observed as shown in Figure 3a. These results clearly indicate that the addition of lithium chloride is effective in promoting the chemoselective amidation in this system.¹⁶

The trace of TG for PAHA is shown in Figure 4. A rapid weight loss was observed at 250–350 $^\circ\text{C}$. In this

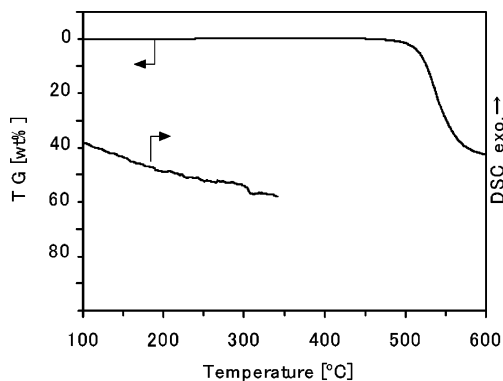


Figure 5. TG and DSC curve of PABO. The heating rate was $5\text{ }^\circ\text{C}/\text{min}$ under nitrogen.

Table 1. Solubility of Poly(*o*-hydroxyamide) (PAHA) and Polybenzoxazole (PABO)^a

solvent	solubility		solvent	solubility	
	PAHA	PABO		PAHA	PABO
acetone	±	–	HMPA ^d	+	–
chloroform	–	–	<i>n</i> -hexane	–	–
<i>m</i> -cresol	+	–	methanol	±	–
cyclohexanone	++	–	2-methoxy-ethanol	++	–
diethyl ether	–	–	2-propanol	–	–
DIGLYM ^b	++	–	PGMEA ^e	–	–
DMF ^c	++	–	sulfuric acid	++	±
1,4-dioxane	–	–	1,1,2,2-tetra-chloroethane	–	–
ethyl acetate	–	–	THF ^f	++	–
ethyl lactate	–	–	toluene	–	–

^a Symbols: ++, soluble at room temperature; +, soluble by heating; ±, partially soluble or swelling; –, insoluble. ^b Diethylene glycol dimethyl ether. ^c *N,N*-Dimethylformamide. ^d Hexamethylphosphoramide. ^e Propylene glycol–1-monomethyl ether 2-acetate. ^f Tetrahydrofuran.

range, the weight loss of 7.5% is in good agreement with the value calculated from the elimination of water due to cyclization (6.5%). The IR spectrum of thermally treated PAHA exhibits a characteristic benzoxazole ring absorption at 1620 cm^{-1} , and the peaks due to the amide (1654 cm^{-1}) and hydroxyl (around 3400 cm^{-1}) groups of PAHA completely disappear (Figure 3b). Thus, PAHA is readily converted to the corresponding PABO by thermal treatment at $350\text{ }^\circ\text{C}$ under nitrogen for 1 h. A 5% weight loss temperature (T_{d5}) of PABO is $518\text{ }^\circ\text{C}$ (Figure 5), which indicates a high thermal stability of PABO. The DSC curve of PABO in Figure 5 shows a glass transition temperature, T_g , at $302\text{ }^\circ\text{C}$ whereas it is enough for microelectronics application. Table 1 shows the results of qualitative solubility both for PAHA and

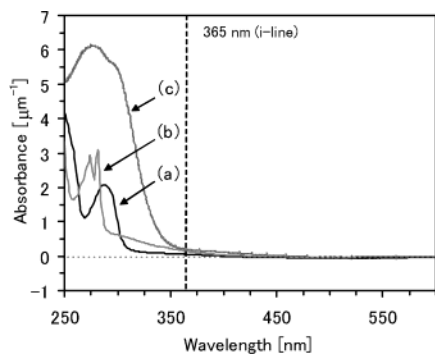


Figure 6. UV-vis spectra of (a) PAHA film, (b) PABO film, and (c) PHA on a quartz plate. The absorbance of each film is set by film thickness, μm^{-1} .

PABO. The latter was prepared by casting the solution of PAHA on a silicon wafer and then cured up to 350 °C for 1 h under nitrogen. PAHA exhibits excellent solubility in various solvents compared to that of the corresponding PABO.

The transmission UV-vis spectra (film on quartz) of PAHA and PHA which was prepared from 6FAP and 4,4'-oxybis(benzoic acid) derivatives are shown in Figure 6a,c. PAHA and PHA films show cutoff at 320 and 370 nm, respectively. Therefore, PAHA is very transparent in the UV-vis region, especially at 365 nm (i-line), compared to that of PHA. A cutoff wavelength is defined here as the point where the transmittance becomes below 1% in the spectrum. In addition, the PABO film shows relative high transparency although the small absorption is extended to 400 nm (Figure 6b).

Dielectric Constant of PABO. The average refractive indices (n_{AV}) of PABO was determined as 1.523. A dielectric constant (ϵ) of the material at optical frequencies can be estimated from the refractive index (n) according to Maxwell's equation, $\epsilon = n^2$. The ϵ around 1 MHz has been evaluated as $\epsilon = 1.10n_{\text{AV}}^2$, including an additional contribution of approximately 10% from the infrared absorption. The n_{AV} of 1.523 can be translated into the dielectric constant of 2.55. This value is considerably lower than the optically estimated ϵ of

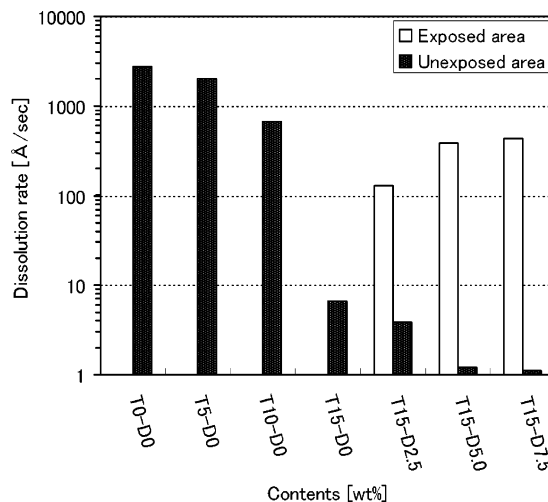


Figure 7. Effect of TVEB and DIAS loadings on the dissolution rate in 2.38 wt % TMAH_{aq} . Each film was prebaked at 120 °C for 5 min, exposed to i-line of 100 mJ/cm^2 , and postexposure baked at 120 °C for 5 min.

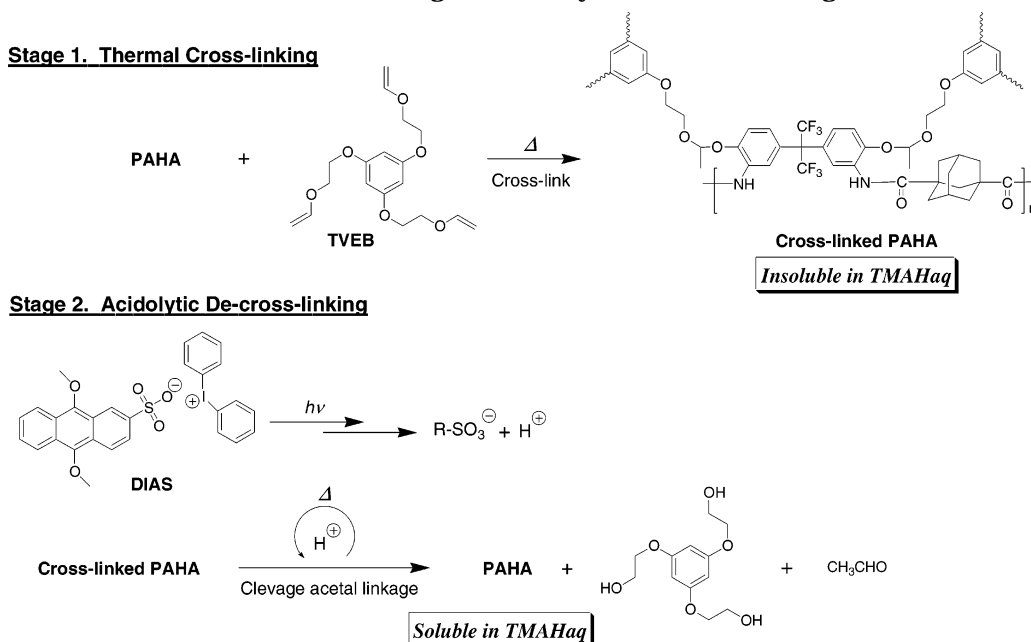
the other wholly aromatic PBOs even which contains hexafluoroisopropylidene groups ($\epsilon =$ around 2.8)^{17,18} due to the introduction of an alicyclic bulky unit, adamantyl, in the polymer structure.

Lithographic Evaluation of PAHA, TVEB, and DIAS. As no absorption in the UV-vis spectrum of PAHA was observed above 320 nm, DIAS having a strong absorption in the range 300–420 nm was selected as a photoacid generator.¹⁹ On the other hand, TVEB was selected as an acidolytic de-cross-linker for positive-tone photoresists because it is useful for a thermal cross-linking and the following acidolytic de-cross-linking²⁰ (Scheme 3).

The vinyl ether groups of TVEB react with the hydroxy group of phenol at high temperature to form cross-linked networks with acetal structures, and then, these cross-linked junctions are cleaved acid-catalytically by a photogenerated acid.

The photosensitive polymer system consisting of PAHA, TVEB, and DIAS in cyclohexanone was formu-

Scheme 3. Mechanism for Cross-Linking and Acidolytic De-Cross-Linking of PAHA with TVEB



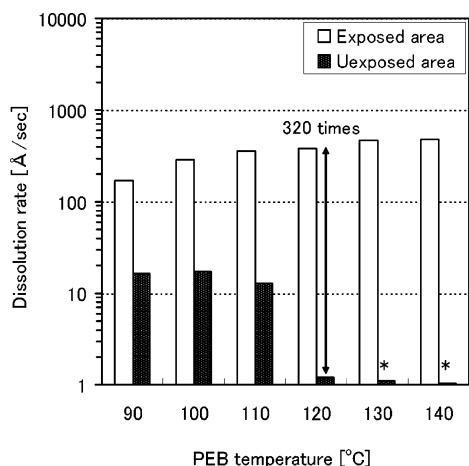


Figure 8. Effect of postexposure bake (PEB) temperature on the dissolution rate in 2.38 wt % TMAH_{aq}. PAHA:TVEB:DIAS (wt %) = 80:15:5. Both prebake and PEB time were set for 5 min at each desired temperature. When PEB was subjected at 130 or 140 °C, the remained unexposed surface showed rough significantly (marked with *).

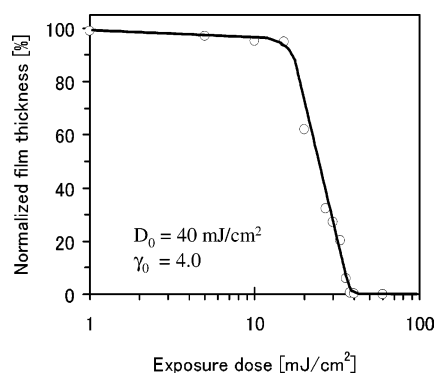


Figure 9. Photosensitive curve of the PABO resist film (thickness: 2.0 μm) consisting of PAHA (85 wt %), TVEB (15 wt %), and DIAS (5 wt %). Condition was fixed as follows: prebaked temperature, 120 °C for 5 min; PEB temperature, 120 °C for 5 min; development, 2.38 wt % TMAH_{aq} for 60 s at 20 °C.

lated. The film spin-cast on a silicon wafer was prebaked, exposed to a filtered super-high-pressure mercury lamp (365 nm), and developed with 2.38 wt % TMAH_{aq}. The dissolution rate was calculated by measuring the change of the film thickness after the development.

To clarify a difference of dissolution behaviors between the exposed and unexposed area, the effect of TVEB and DIAS loadings on the dissolution rate in 2.38 wt % TMAH_{aq} was investigated (Figure 7), where each film was prebaked at 120 °C for 5 min, exposed to 100 mJ/cm² of i-line, and then postexposure baked at 120 °C for 5 min. The difference of the dissolution rates between the exposed and unexposed area reaches 320 times when 15 wt % of TVEB and 5 wt % of DIAS were loaded.

Next, the effect of postexposure bake (PEB) temperature on the dissolution rate in 2.38 wt % TMAH_{aq} was studied. The results are shown in Figure 8. By changing the PEB temperature from 90 to 140 °C, an adequate dissolution contrast (320 times) was obtained at 120 °C. When the PEB temperature was set above 120 °C, the film surface became rough probably due to a decomposition of DIAS.

On the basis of the findings described above, a resist system was formulated as 85 wt % of PAHA, 15 wt % of TVEB, and 5 wt % of DIAS. A sensitivity curve for the 2.0 μm thick photopolymer film shown in Figure 9 indicated a sensitivity of 40 mJ/cm² and a contrast of 4.0.

Figure 10 shows scanning electron micrographs (SEMs) of the contact printed positive image after exposure to 70 mJ/cm² of i-line through a photomask and the following annealed to convert into PABO. The resist was capable of resolving 10 μm featured patterns where a 2.3 μm thickness film was used (Figure 10a). The printed film was converted to the PABO film by heating at elevated temperature up to 350 °C for 1 h under nitrogen (Figure 10b). The formation of the PABO was confirmed by the IR spectrum, where the characteristic oxazole ring absorption appeared at 1616 cm⁻¹ and the absorptions due to the hydroxyl and amide carbonyl groups disappeared at 3300 and 1650 cm⁻¹ in PAHA, respectively.

Conclusion

PAHA containing adamantyl units in the main chain was prepared from ADC and 6FAP in the presence of lithium chloride in NMP. Thermal treatment of PAHA easily produced PABO. A T_{d5} of the PABO was 518 °C under nitrogen. The optically estimated dielectric constant is 2.55, which is lower than that of conventional aromatic PBOs. Since PAHA has a high transparency in the UV–vis region, PAHA containing 15 wt % TVEB

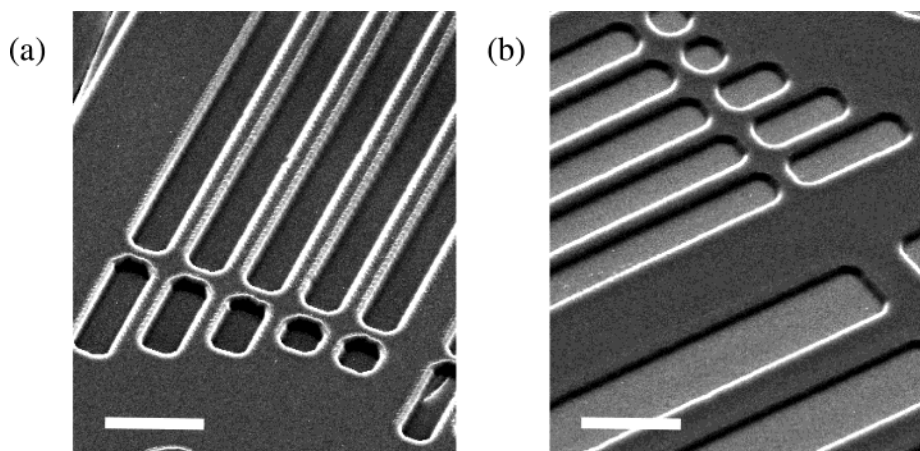


Figure 10. SEM images of (a) PAHA film (thickness: 2.3 μm) and (b) PABO film (thickness: 1.9 μm) after curing at 350 °C under nitrogen. A scale bar indicates 30 μm in each micrograph.

and 5 wt % DIAS functioned as a positive-type photosensitive polymer, showing its sensitivity and contrast 40 mJ/cm² and 4.0 with 365 nm light exposure, respectively. The positive image of PAHA film by contact-printing mode was converted to the corresponding PABO image successfully by thermal treatment at 350 °C. The incorporation of the adamantyl units is advantageous to PSPBO because it provides the high transparency and low dielectric constant as well as high thermal stability.

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