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# A New Three-Component Photoresist Based on Calix[4]resorcinarene Derivative, a Cross-linker, and a Photo-acid Generator

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Calix[4]resorcinarene (2,8,14,20-tetramethylcalix[4]arene-4,6,10,12,16,18,22,24-octol, abbrev. to C4-RA) derivative (**4**) having *p*-hydroxybenzyl groups on its exterior was prepared by the condensation of C4-RA and *p*-(allyloxy)benzyl bromide, followed by the cleavage of allyl groups with palladium catalyst and ammonium formate. Compound **4** having high transparency to UV-light above 300 nm was considered for a new resist matrix. A three-component photoresist consisting of **4**, 2,6-bis(hydroxymethyl)-4-methylphenol (BHMP), and diphenyliodonium 9,10-dimethoxyanthracene-2-sulfonate (DIAS) showed a sensitivity of 19 mJ cm$^{-2}$ ($D^{1/2}$) and a contrast of 3.0 ($\gamma^{1/2}$) when it was exposed to 365 nm light and post-exposure baked (PEB) at 110 °C for 5 min, followed by developing with a 0.2 wt% aqueous tetramethylammonium hydroxide (TMAH) solution. A fine negative image featuring 1 μm of minimum line and space patterns was observed on film of the photoresist exposed to 40 mJ cm$^{-2}$ of UV-light at 365 nm with a scanning electron microscope.

To increase the density of semiconductor devices, the lithography process to produce high-resolution images on substrates have been one of the most significant processes in semiconductor manufacturing.[1] Therefore, the high-performance photoresists which are essential for the lithography process have also been one of the key technologies for the next chip development. Nevertheless, a combination of novolac resins and 2-diazo-1(2*H*)-naphthalenone (DNQ) well-known as an authentic photoresist for 365 nm lithography is still the workhorse for the high-resolution imaging.

*o*-Cresol novolac resins are generally composed of various molecules having undefined molecular weights and structures connected with *o*,*o*- and *o*,*p*-methylene bridges. Their structural properties seriously affect the dissolution behaviors of the resist films in the aqueous alkaline developer.[2—5] In contrast to *o*-cresol novolac resins, macrocyclic oligomers called calixarenes, which are prepared by the base-catalyzed condensation of *p*-alkylphenol with formaldehyde, have the definite structure composed of 4—8 repeating units.[6—8] Their uniform macrocycles are easily characterized by NMR spectroscopy, and remove some structural problems occurring in cresol novolacs. E-beam resist based on *O*-acetylated *p*-methylcalix[6]arene showed high resolution images,[9] but it doesn't belong to the alkaline developable resists such as the novolac/DNQ resists. Calixarenes are not suitable for photoresists because their low solubility both in common organic solvents and aqueous alkaline solutions. We have been interested in calix[4]resorcinarenes (2,8,14,20-tetramethylcalix[4]arene-4,6,10,12,16,18,22,24-octol, abbrev. to C4-RA) which is easily prepared by the acid-catalyzed condensation of resorcinol and acetaldehyde. C4-RAs have been reported to have a good solubility for organic solvents and aqueous alkaline solutions.

In a preceding paper,[10] we reported the development of the negative working, alkaline developable photoresist based on C4-RA, a polyfunctional benzylic alcohol as a cross-linker, and a photoacid generator.

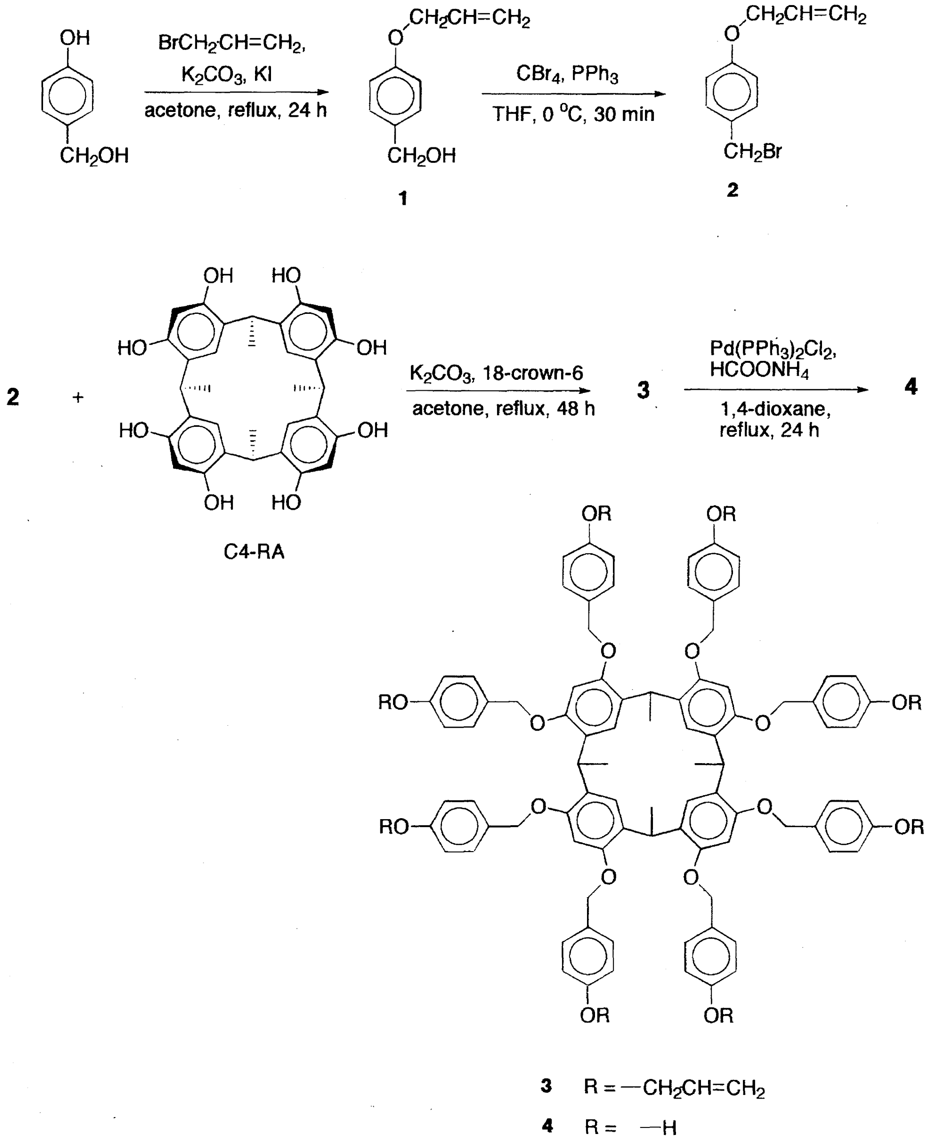
However, there are still some problems about the properties of C4-RA, especially the film-forming capability, due to its low molecular weight and relatively high crystallinity.

Recently, Shirota et al. reported a guideline for molecular design of low-molecular weight amorphous materials (called amorphous molecular materials), that is, the number of conformers can be increased by lowering the symmetry of nonplanar molecules.[11] Fortunately, C4-RA is a nonplanar molecule which provides various conformations by some modifications such as acetylation.[12] Therefore, an attachment of substituents to the hydroxy groups of C4-RA was expected to give amorphous molecular materials by increasing the number of conformers.

In this paper, we report the synthesis of *O*-(*p*-hydroxybenzyl)-C4-RA (**4**) as a new matrix and a lithographic evaluation of three-component photoresist consisting of **4**, 2,6-bis(hydroxymethyl)-4-methylphenol (BHMP) as a cross-linker, and diphenyliodonium 9,10-dimethoxyanthracene-2-sulfonate (DIAS) as a photo-acid generator.

## Results and Discussion

**Synthesis of *O*-(*p*-Hydroxybenzyl)-C4-RA 4.** *O*-[*p*-(allyloxy)benzyl]-C4-RA **3** was synthesized by the condensation of *p*-(allyloxy)benzyl bromide **2**, which was made from *p*-hydroxybenzyl alcohol **1** via the three-step reactions, and calix[4]resorcinarene (C4-RA) derived from resorcinol and aldehydes.[12,13] Then, allyl groups of **3** were cleaved with $Pd(PPh_3)_2Cl_2$ and ammonium formate to give *O*-(*p*-hydroxybenzyl)-C4-RA **4** (Scheme 1).



Scheme 1. Synthesis of *O*-(*p*-hydroxybenzyl)-C4-RA(**4**).

The structure of **4** was characterized by IR and NMR spectroscopies and elemental analysis. The IR spectrum showed characteristic absorptions at 3400 and 1225 $\text{cm}^{-1}$ due to OH and C–O–C stretching, respectively.

The condensation of resorcinol and aldehydes generally gives C4-RAs which include two stereoisomers with all-*cis* or *cis-trans-trans* configuration.[14] In case of using acetaldehyde, the stereoisomer having all-*cis* configuration was obtained as the major product.[12] Actually, the NMR spectrum of C4-RA showed four sharp peaks at 1.75, 4.50, 6.21, and 7.63 ppm, assigned to methyl, methine, aromatic, and phenolic hydroxy groups, respectively, which indicate the existence of a cone-structure formed with all-*cis* configuration only. After *p*-(allyloxy)benzyl groups were attached to phenolic hydroxy groups of C4-RA, the $^{1}\text{H}$ NMR spectrum of **3** changed dramatically. Signals at 4.74 and 1.50 ppm due to methine and methyl became broad peaks. In addition, new peaks which appeared by formation of *p*-allyloxybenzyl ethers were also broad or multiple because of slow exchange rate between conformers with bulky groups such as *p*-(allyoxy)benzyl ether. A similar phenomenon was observed in the $^{1}\text{H}$ NMR spectrum of *O*-acetylated-C4-RA.[12] Thus, compound **3** would be a mixture of several conformers. The broad signal at 5.27—5.44 and 6.11 based on allyl groups of compound **3** disappeared completely after the deallylation of **3** with a palladium catalyst. Nevertheless, the influence of substituents on the structure was observed in the $^{1}\text{H}$ NMR spectrum of **4** (Fig. 1). The $^{1}\text{H}$ NMR spectrum was measured at $-30$ °C in acetone-$d_6$. Two doublet peaks and one broad peak due to the methyl groups at 1.90, 1.85, and 1.50 ppm, and broad multiplet peaks assigned to phenolic hydroxy groups at around 8.6 ppm were observed. These peaks indicate that compound **4** has several types of conformations, as does compound **3**. The elemental analyses also supported the formation of the expected compounds.

Compound **4** showed a good solubility in organic solvents, such as dipolar aprotic solvents and acetone and especially in alcohols at room temperature. Transparent films were

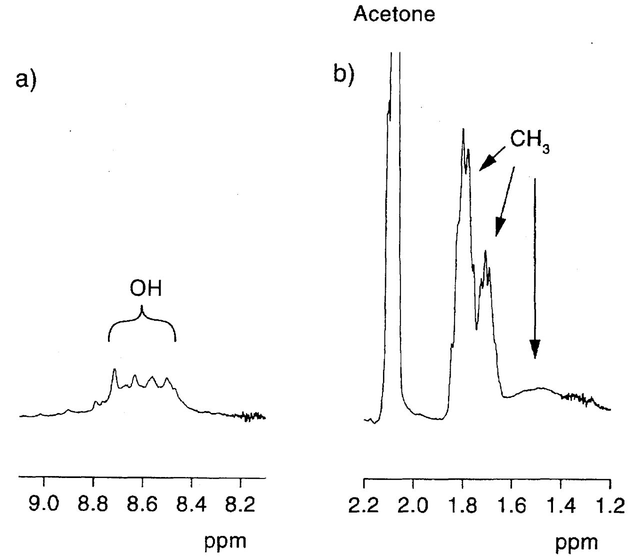


Fig. 1. $^1$H NMR spectra of **4** measured in acetone-$d_6$ at −30 °C in the region of (a) hydroxy protons, and (b) methyl protons.

cast from the solutions on non treated silicon wafers. When unsubstituted C4-RA was used as a resist matrix, the films which had stood for several hours often changed to the partially crystallized or less transparent films because of the features of the low molecular-weight macorcycle having the uniform structure. On the other hand, the films obtained from compound **4** were stable for a long time. No crystallization of **4** was observed, probably due to the structural features of the nonplanar molecule that has the bulky groups on its exterior.

**Thermal Properties.** The thermal behavior of **4** was estimated by thermogravimetry (TG) and differential scanning calorimetry (DSC). Compound **4** showed a good thermal stability: a 10% weight loss temperature ($Td_{10}$) was observed at 310 °C. DSC showed no endothermic peak due to the melting point below 200 °C and a glass transition temperature at 85 °C on the 2nd heating (Fig. 2). These results suggest that compound **4** is an amorphous material with good film properties.

**Lithographic Evaluation.** The UV spectrum of compound **4** film (1 μm thick) is shown in Fig. 3. Compound **4** has two strong absorptions at 216 and 285 nm due to π–π* transitions and is almost transparent around 250 nm and above 300 nm. This optical property is similar to that of novolac resins. Thus, this material was utilized as a matrix of phtotoresist by combining with photosensitizers absorbing at 365 nm.

DNQ is the most popular photosensitizer for the 365 nm positive-type photoresist. Therefore, the compound **4** film including DNQ was expected to act as a 365 nm photoresist by dissolution inhibition of DNQ toward an aqueous alkaline developer. Even 30 wt% loading of DNQ didn't, however, give enough contrast to produce fine images because the solubility of the matrix in the aqueous alkaline developer was too high. Thus, we decided to develop a three-component resist composed of an alkaline developable matrix, a photoacid generator such as an onium salt, and an acid-reactive cross-linker.

Onium salts are well-known as photo-acid generators for various Deep-UV resists.[15] However, there are few onium salts that have an absorption band at wavelengths longer than 300 nm. Therefore, based on the previous study,[10] diphenyliodonium 9,10-dimethoxyanthracene-2-sulfonate (DIAS) was employed as a photoacid generator which has a strong absorption in the range of 300 to 450 nm.

Many articles[16] describing various cross-linkers for negative resists have been published. Particularly the cross-linkers offering a high sensitivity and contrast by acid-catalyzed cross-linking have attracted our interest on development of high performance photoresists. Fréchet et al. reported that the polyfunctional benzylic alcohols derived from phenols functioned as good cross-linkers in the chemical amplified negative-type resists.[17] Thus, 2,6-bis(hydroxymethyl)-4-methylphenol (BHMP) was selected as the desirable cross-linker owing to its availability and sensitivity.

To optimize the conditions for generation of high-contrast images, the dissolution behaviors of exposed and unexposed

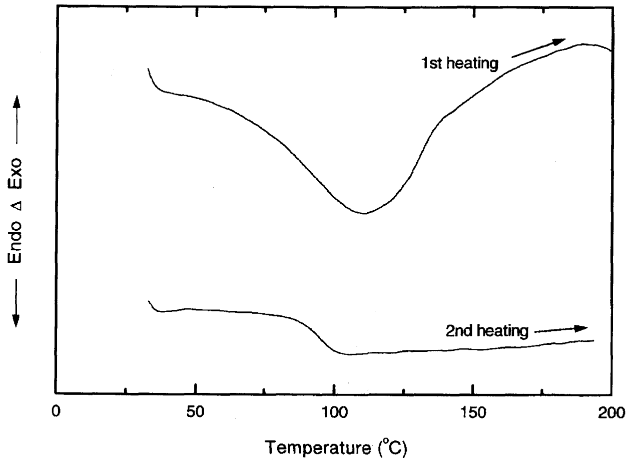


Fig. 2. DSC curve of compound **4** measured at a heating rate of 20 °C min$^{-1}$ under nitrogen.

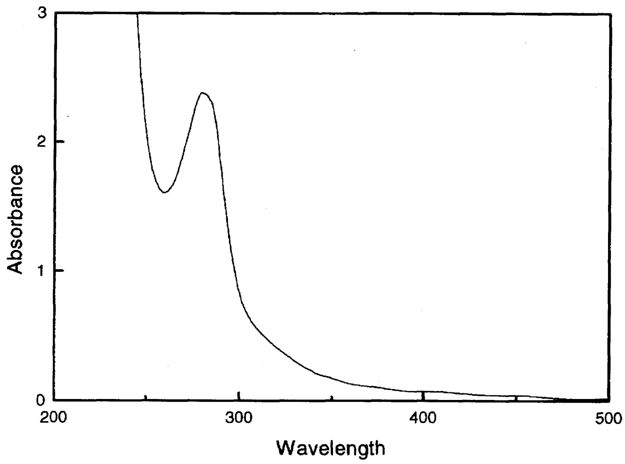


Fig. 3. UV-vis spectrum of 1 μm-thick film of compound **4** cast from cyclohexanone.

parts were studied. Since a post-exposure bake (PEB) generally influences sensitivity and contrast of the resist in a chemically amplified system, we investigated the effects of a PEB time and a PEB temperature on the dissolution rates of the resist films. The dissolution rate was estimated by measuring the film thickness after the development. The difference of the dissolution rate between exposed and unexposed parts indicates the dissolution contrast. The resist was formulated by mixing compound **4** (70 wt%), BHMP (20 wt%), and DIAS (10 wt%) in cyclohexanone. The relationship between the dissolution rate and the PEB temperature is shown in Fig. 4, where the film was exposed 40 mJ cm$^{-2}$ to 365 nm UV light, postbaked for 5 min at elevated temperatures from 60 to 130 °C, and developed with a 0.2 wt% aqueous tetramethylammonium hydroxide (TMAH) solution for 20 s. The dissolution rate of the exposed part decreased with increasing PEB temperature. On the other hand, the solubility of the unexposed area was almost unchanged until 115 °C, and then started to decrease. The difference of the dissolution rates between exposed and unexposed parts reached about 100 times at 100 °C.

The effects of PEB time, as well as the PEB temperature, were investigated by observing the changes of the dissolution rate of the resist films exposed 40 mJ cm$^{-2}$, postbaked at 110 °C, and developed with the 0.2 wt% aqueous TMAH solution (Fig. 5). The dissolution rate of the exposed area rapidly decreased with increasing the PEB time. The minimum dissolution rate was reached after the PEB time for 3 min. The dissolution rate of the unexposed parts was almost unchanged while PEB time was increased. These results indicate that increasing both PEB temperature and PEB time accelerates the probable cross-linking process via acid-catalyzed electrophilic aromatic substitution occurring on the exposed parts. However, a temperature higher than the melting point of the cross-linker caused a decrease of the dissolution rate of the unexposed area.

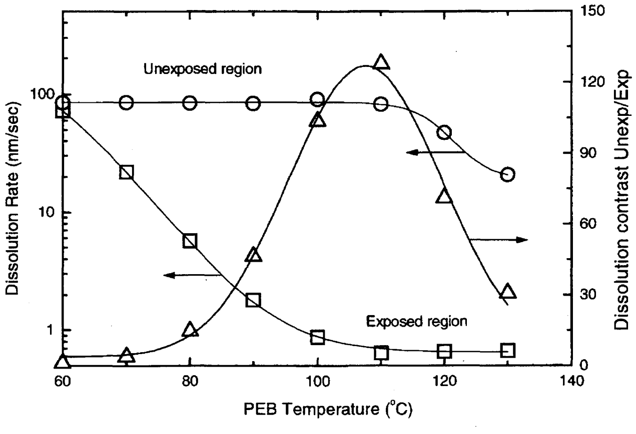


Fig. 4. The effect of post exposure bake temperature for 5 min on the dissolution rate of the resist film consisting of **4** (70 wt%), BHMP (20 wt%), and DIAS (10 wt%). Dissolution rate of (○) unexposed region and (□) exposed region to 40 mJ cm$^{-2}$. Dissolution contrast (△) between exposed region and unexposed region.

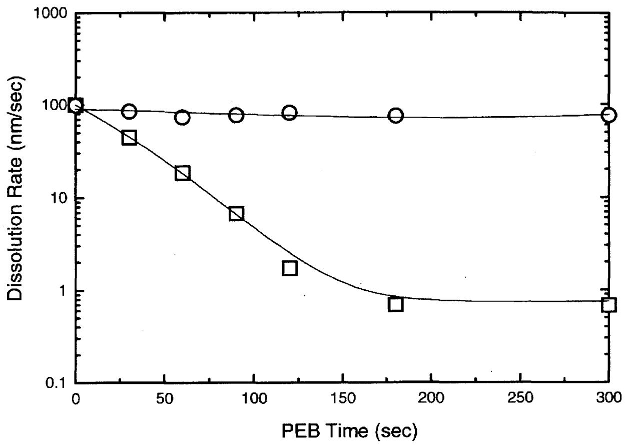


Fig. 5. The effect of post exposure bake time at 110 °C on the dissolution rate of the resist film consisting of **4** (70 wt%), BHMP (20 wt%), and DIAS (10 wt%). Dissolution rate of (○) unexposed region and (□) exposed region to 40 mJ cm$^{-2}$.

After these preliminary optimization studies involving the PEB temperature and the PEB time, we formulated a resist system consisting of calixarene (70 wt%), BHMP (20 wt%), and DIAS (10 wt%) in cyclohexanone. The film spin-cast on a silicon wafer was prebaked at 80 °C for 10 min (thickness of 1 μm), exposed to 365 nm UV radiation, postbaked at 110 °C for 3 min, and developed in a negative mode with the 0.2% aqueous TMAH solution at room temperature. The sensitivity curve was consistent with the dissolution behavior studied above, indicating that the sensitivity ($D^{1/2}$) and the contrast ($\gamma^{1/2}$) were 19 mJ cm$^{-2}$ and 3.0, respectively (Fig. 6).

Figure 7 presents a scanning electron micrograph of the contact-printed image that was observed on the resist film by exposure to 40 mJ cm$^{-2}$ and PEB at 120 °C for 3 min, followed by developing with the 0.2% aqueous TMAH solution. The high-resolution negative images featuring 1 μm line and space patterns, which is the limitation of our exposure system, were obtained on the 1 μm thick film.

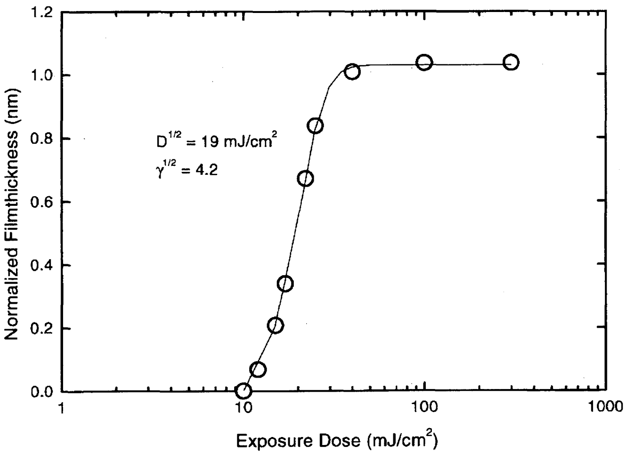


Fig. 6. Exposure characteristic curve of the system based on **4** (70 wt%), BHMP (20 wt%), and DIAS (10 wt%), using the 0.2 wt% aqueous TMAH solution as a developer.

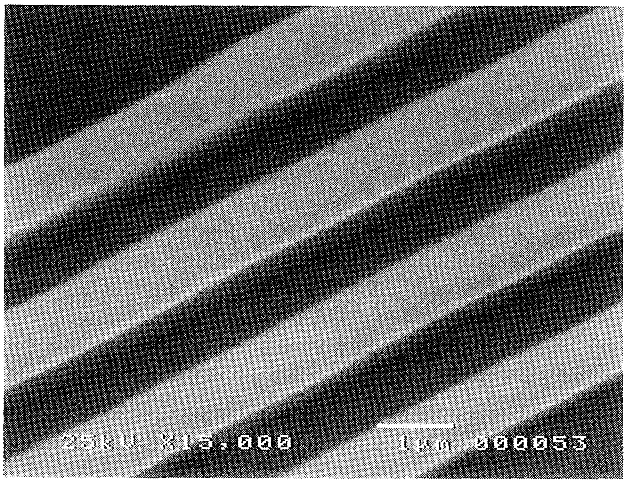


Fig. 7. A scanning electron micrograph of negative images contact-printed in the resist.

The mechanism of this negative pattern formation is same as the one previously reported based on C4-RA, a cross-linker, and a photo-acid generator.[18] A photogenerated acid catalyzes the formation of benzylic carbocation species, which undergo electrophilic aromatic substitution to produce *C*- and *O*-alkylated polymers. This reaction converts the soluble an *O*-(*p*-hydroxybenzyl)-C4-RA to an insoluble cross-linked polymer.

In conclusion, we successfully developed a new amorphous compound having the advantage of use as a resist material by the condensation of C4-RA and *p*-(allyloxy)benzyl bromide, followed by the deallylation. The new negative working alkaline developable resist consisting of compound **4** (70%), BHMP (20 wt%) as the cross-linker, and the photoacid generator DIAS (10 wt%) showed the sensitivity of 19 mJ cm$^{-2}$ ($D^{1/2}$) and the contrast of 3.0 ($\gamma^{1/2}$) to UV-light. The concept of producing amorphous molecular materials will be helpful to develop new materials based on C4-RAs for wide applications.

## Experimental

**Materials.** C4-RA was prepared by the acid-catalyzed condensation of resorcinol with acetaldehyde according to the reported procedure.[12]

2,6-Bis(hydroxymethyl)-4-methylphenol (BHMP) was synthesized by the hydroxymethylation of *p*-cresol in the presence of potassium carbonate, followed by neutralization with carbon dioxide.[19]

Diphenyliodonium 9,10-dimethoxyanthracene-2-sulfonate (DIAS) was obtained by the reaction of diphenyliodonium chloride with sodium 9,10-dimethoxyanthracene-2-sulfonate, which was produced by the reduction of anthraquinone sulfonate with zinc powder and sodium hydroxide, followed by methylation with dimethyl sulfate.[20]

**Preparation of *p*-(Allyloxy)benzyl Alcohol (1).** 4-Hydroxybenzyl alcohol (25 g, 0.201 mol), potassium carbonate (36.1 g, 0.262 mol), and potassium iodide (0.43 g, 0.0026 mol) in dry acetone (140 mL) was placed in a 500 ml three-necked flask equipped with a reflux condenser, a nitrogen inlet, and a dropping funnel. To this suspension was added dropwise a solution of 3-bromopropene (22.7 mL, 0.262 mol) in dry acetone (140 mL) under nitrogen. The reaction mixture was refluxed for 24 h, and then an insoluble part was filtered off. The filtration was dried over $MgSO_4$ and evaporated to give a yellow liquid. The yield was 27.7 g (84%). IR (NaCl) $\nu$ 1220 and 1020 cm$^{-1}$ (C–O–C), 1000 and 925 cm$^{-1}$ (C–H of allyl); $^1$H NMR ($CDCl_3$) $\delta$ = 6.85, 7.21 (d, $J$ = 8.1 Hz, ArH, 4H), 5.98—6.12 (m, –CH=, 1H), 5.26, 5.39 (dd, $J$ = 18.8, 1.6, 11.9, 1. Hz, =$CH_2$, 2H), 4.60 (s, –$CH_2$–, 2H), 4.54 (d, $J$ = 5.3 Hz, –$CH_2$–, 2H). Anal. Calcd for $C_{10}H_{12}O_2$: C, 73.15; H, 7.37%. Found: C, 72.96; H, 7.64%.

**Preparation of *p*-(Allyloxy)benzyl Bromide (2).** To a cold solution of **1** (10.7 g, 64.9 mmol) and tetrabromomethane (26.9 g, 81.1 mmol) in THF (70 mL) placed in a 300 ml three-necked flask was added slowly triphenylphosphine (21.3 g, 81.1 mmol) under nitrogen. The reaction mixture cooled with an ice bath was stirred for 30 min, and then the filtrate which had passed through a filter paper was evaporated. The residue was washed with hexane and then purified by column chromatography on silica gel using hexane/ethyl acetate (15/1) as an eluent to give **2** as a yellow oil. The yield was 11.8 g (80%). IR (NaCl) $\nu$ 1225 and 1020 cm$^{-1}$ (C–O–C); $^1$H NMR($CDCl_3$) $\delta$ = 6.85, 7.29 (d, $J$ = 8.6 Hz, ArH, 4H), 5.95—6.09 (m, –CH=, 1H), 5.27, 5.39 (dd, $J$ = 19.1, 1.6, 11.8, 1.3 Hz, =$CH_2$, 2H), 4.50 (d, $J$ = 5.3 Hz, –$CH_2$–, 2H), 4.47 (s, –$CH_2$–, 2H). Anal. Calcd for $C_{10}H_{11}OBr$: C, 52.89; H, 4.88. Found: C, 53.19; H, 5.03.

**Synthesis of *O*-[*p*-(Allyloxy)benzyl]-C4-RA (3).** A mixture of C4-RA (5.99 g, 11.0 mmol), **2** (22.0 g, 96.9 mmol), 18-crown-6 (2.56 g, 9.68 mmol), and potassium carbonate (40.2 g, 290 mmol) in freshly distilled acetone (130 mL) was refluxed for 48 h. After the precipitate was filtered off, the solvent was removed under pressure. The solution of the residue dissolved in ethyl acetate was extracted with aqueous potassium carbonate solution to remove unreacted C4-RA and washed with an saturated aqueous sodium chloride solution. Then the organic phase was dried over $MgSO_4$ and evaporated to produce a brown oil. The crude product was purified by column chromatography on silica gel using hexane/ethyl acetate (5/2) as an eluent to yield **3** as a yellow oil. The yield was 7.02 g (37%). IR (NaCl) $\nu$ 1240 and 1020 cm$^{-1}$ (C–O–C); $^1$H NMR ($CDCl_3$) $\delta$ = 6.55—7.26 (m, ArH, 40H), 6.11 (br, –CH=, 16H), 5.27—5.44 (m, –$CH_2$–, 8H), 4.74 (br, CH, 4H), 4.27—4.52 (m, $CH_2$, 32H), 1.50 (br, $CH_3$, 12H). Anal. Calcd for $C_{112}H_{112}O_{16}\cdot 1.2H_2O$: C, 77.50; H, 6.64%. Found: C, 77.48; H, 6.73%.

**Synthesis of *O*-(*p*-Hydroxybenzyl)-C4-RA (4).** Dichlorobis(triphenylphosphine)palladium(II) (0.28 g, 0.40 mmol) and ammonium formate (5.10 g, 80.9 mmol) were placed in a 300 ml three-necked flask equipped with a condenser; this was filled with argon. To the solution was added a solution of **3** (4.34 g, 2.53 mmol) in dehydrated 1,4-dioxane immediately and this mixture was refluxed for 24 h. Then the reaction solution was poured into a 10 wt% aqueous sodium hydroxide solution and unreacted compounds were extracted with diethyl ether from the aqueous solution. To the aqueous layer was added concd hydrochloric acid until the pH of the solution became 3, and then the product was extracted with ethyl acetate. The organic layer was dried over $MgSO_4$ and evaporated. The obtained brown oil was poured into deionized water to give **4** (2.52 g, 71%) as a brown solid. IR (NaCl) $\nu$ 3400 cm$^{-1}$ (–OH), 1225 and 1100 cm$^{-1}$ (C–O–C); $^1$H NMR-(DMSO-$d_6$) $\delta$ = 9.31 (br, OH, 8H), 6.23—7.14 (m, ArH, 40H), 4.42 (br, CH, 4H), 3.73 (br, $CH_2$, 16H), 1.40 (br, –$CH_3$, 12H). Anal. Calcd for $C_{88}H_{80}O_{16}\cdot 2.7H_2O$: C, 73.29; H, 5.96%. Found: C, 73.25; H, 6.06%.

**Photosensitivity.** Compound **4** was dissolved in cyclohexanone (20 wt%) at room temperature, and to this solution were added

BHMP and DIAS. Films spin-cast at 1700 rpm for 10 s on silicon wafers were prebaked at 80 °C for 10 min and exposed through a filtered super-high pressure mercury lamp (Ushio USH-200DP). Imagewise exposure through a mask was carried out in a contact-printing mode.

**Dissolution Rate.** The 1 μm films on a silicon wafer were exposed at 365 nm wavelength to the filtered super high-pressure mercury lamp, developed in a 0.2% aqueous tetramethylammonium hydroxide (TMAH) solution at room temperature, and rinsed with water. The characteristic curve of the normalized film thickness against exposure energy was obtained.

**Measurement.** Infrared spectra were recorded on a Horiba FT-210 spectrophotometer. $^1$H NMR spectra were recorded on a JEOL EX 270 spectrometer. UV-spectra were obtained on a JASCO V-560 spectrophotometer. Thermal analyses were performed on a Seiko SSS 5200-TG/DTA 220 instrument at a heating rate of 10 $^\circ\mathrm{C\,min^{-1}}$ (TG) and a Seiko SSS 5200-DSC 220 at a heating rate of 20 $^\circ\mathrm{C\,min^{-1}}$ for differential scanning calorimetry (DSC) under nitrogen. The film thicknesses were measured by a Dectak 3030 system (Vecco Instruments Inc.). Scanning electron micrographs were obtained by a JEOL-5300 microscope. Most manipulations were carried out either under dry, oxygen-free argon or nitrogen or in vacuo with Schlenk-type flasks. Solvents were dried and purified in the usual manner, and stored under an atmosphere of argon.

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