

# Synthesis and Evaluation of Positive-Acting Photosensitive Polyimides with Phenol Moiety

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## SYNOPSIS

Novel positive-acting photosensitive polyimide resists (**1**) developable with an alkaline aqueous solution were prepared. The resists consisted of a naphthoquinone diazide (NQ) and a polyamic acid bearing hydroxyphenyl groups (PA). The hydroxyphenyl moieties are newly incorporated by adding 2-*N,N*-dimethylaminoethyl-3-hydroxybenzoate (*m*-PEA) or 2-*N,N*-dimethylaminoethyl-4-hydroxybenzoate (*p*-PEA) to the polyamic acid, through ionic interaction between the carboxylic acid and the amine. The NQ acts as a solution inhibitor for the PA by the interaction with the hydroxyphenyl moiety. This system is similar to that of phenol–novolac resin/NQ resists currently conducted in the fabrication of the Large Scale Integrated circuit (LSI). The resist (**1**) is exposed to a G-line and is developed with tetramethylammonium hydroxide aqueous solution to provide fine patterns, proving that the (**1**) is compatible with LSI-production lines currently applied. A novel polyamic acid-curing mechanism found in the course of this study is also reported. © 1995 John Wiley & Sons, Inc.

## INTRODUCTION

Of the various of high-temperature resistant polymers, the polyimides, in particular, have proved their suitability for practical use in passivation or insulation in the microelectronics industry as a result of their excellent properties including chemical and mechanical resistance, good insulation, and planarizability. Extremely, thin-film polyimides have also been used in the alignment layer of liquid crystal displays. In microelectronics applications, the polyimide layer has to be patterned. However, since polyimides have no sensitivity for pattern generation, the pattern has to be fabricated with the aid of a photoresist and multilayer etching. This is inconvenient and necessitates a multistep process. Photosensitive polyimides (including photosensitive polyimide precursors) have greatly enhanced developments in microelectronics, since they simplify polyimide layer pattern generation by eliminating the need for a photoresist. Of the various photosensitive polyimides, however, negative-acting photo-

sensitive polyimides are the most common.<sup>1-9</sup> However, these materials demand the use of much organic solvent, which causes environmental problems, in developing the relief patterns, and they also suffer from such problems as poor shelf life, shrinkage during curing, and swelling. On the other hand, development in an alkaline aqueous solution is well known to eliminate these problems.

A positive-acting photosensitive polyimide composition of a polyamic acid or a polyimide with phenol groups in the polymer chain and a naphthoquinone diazide photosensitizer has been reported.<sup>10-14</sup> A positive resist with a polyamic acid polymer was too soluble in an alkaline aqueous solution. On the other hand, the resist in which the polymer was a fully cured polyimide did not adhere strongly to the substrates. Kubota et al. dealt with positive photosensitive polyimide precursors esterified with *o*-nitrobenzyl alcohol.<sup>15</sup> Omote et al. reported a novel photoreactive polyimide based on photoinduced acidolysis<sup>16</sup> and positive polyimide resists in which naphthoquinone diazides are bonded partially to the hydroxyl groups on the polyimide skeleton.<sup>17</sup> In previous research, we reported on positive photosensitive polyimides using polyamic acid esters with

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phenol moieties.<sup>18</sup> In this article, we discuss the synthesis and characterization of positive-acting photosensitive polyimides consisting of an aromatic polyamic acid (a precursor of polyimide), a phenol derivative as a solution protector, and a naphthoquinone diazide as a photosensitizer.

## EXPERIMENT

All commercially procured compounds were used as received without further purification (unless otherwise noted). Pyromellitic dianhydride (PMDA) and 3,3',4,4'-benzophenone tetracarboxylic dianhydride (BTDA) were purchased from Daisel Chemical; 4,4'-(hexafluoroisopropylidene)bis(phthalic anhydride) (6FDA), from Hoechst; 4,4'-diaminodiphenyl ether (ODA), from Wakayama Seika; 2,2-bis(3-amino-4-hydroxyphenyl)hexafluoropropane (6FAP), from Central Glass; 2-(4-aminophenyl)-2-(4-hydroxyphenyl)propane (AHP), from Mitsui Petrochemical; 1,3-bis( $\gamma$ -aminopropyl)-1,1,3,3-tetramethyldisiloxane (TSL), from Toshiba Silicone; 3-hydroxybenzoic acid, 4-hydroxybenzoic acid, and *N*-methyl-2-pyrrolidone (NMP), from Kanto Chemical; and 2-*N,N*-dimethylaminoethanol (DAE) and borontrifluoride diethyl ether complex, from Wako Pure Chemical. Naphthoquinone diazide (4NT-300) was supplied by Toyo Gosei.

## SYNTHESIS OF PHENOL DERIVATIVE

### 2-*N,N*-Dimethylaminoethyl-4-hydroxybenzoate (*p*-PEA)

To a three-necked round-bottom flask equipped with a mechanical stirrer and a nitrogen inlet were added 4-hydroxybenzoic acid (41.4 g, 0.3 mol) and DAE (124 g, 1.4 mol). The mixture was thoroughly stirred, thereby forming a suspension. Then, a solution prepared by dissolving borontrifluoride diethyl ether complex (0.6 g) in 10 g of DAE was gradually added. The resulting mixture was slowly heated and stirred for 6 h at the DEA reflux temperature. The excess DAE was distilled off and the remaining liquid distilled under a reduced pressure, yielding 59 g of intermediate fraction (yield: 87%). The IR spectrum showed absorption peaks at 1590  $\text{cm}^{-1}$  (Ar—CO—) and 1240  $\text{cm}^{-1}$  (—CO—O—). The <sup>1</sup>H-NMR spectrum in DMSO *d*<sub>6</sub> exhibited signals at 7.13 ppm (hydroxyl proton), 6.80–7.25 ppm (aryl proton), 3.59 and 2.51 ppm (methylene proton), and 2.28 ppm (methyl proton).

### 2-*N,N*-Dimethylaminoethyl-3-hydroxybenzoate (*m*-PEA)

To a three-necked round-bottom flask equipped with a mechanical stirrer and a nitrogen inlet were added 4-hydroxybenzoic acid (41.4 g, 0.3 mol) and DAE (124 g, 1.4 mol) and the mixture was thoroughly stirred, thereby forming a suspension. To this suspension, a solution prepared by dissolving borontrifluoride diethyl ether complex (0.6 g) in 10 g of DAE was gradually added. The resulting mixture was slowly heated and stirred for 6 h at the DEA reflux temperature. The excess DAE was distilled off and the remaining liquid cooled to room temperature following precipitation. The suspension was filtered and *m*-PEA was obtained by washing with acetone (yield: 82%, mp = 82–83°C). The IR spectrum showed absorption peaks at 1560  $\text{cm}^{-1}$  (Ar—CO—) and 1370  $\text{cm}^{-1}$  (—CO—O—). The <sup>1</sup>H-NMR spectrum in DMSO-*d*<sub>6</sub> exhibited signals at 9.57 ppm (hydroxyl proton), 6.87–7.39 ppm (aryl proton), 3.67 and 2.85 ppm (methylene proton), and 2.55 ppm (methyl proton).

## SYNTHESES OF COPOLYAMIC ACIDS

### PAA-1

Nitrogen gas dried with phosphorus pentoxide was introduced into a reaction flask fitted with a stirrer and a dropping funnel. Also introduced into the flask were BTDA (21.759 g, 0.0675 mol), PMDA (4.9173 g, 0.0225 mol), and NMP (90 g). These were fully stirred, forming a mixture which was cooled to 10°C in an ice bath. Next, ODA (16.603 g, 0.0828 mol), TSL (1.3417 g, 0.0054 mol), and AHP (0.8168 g, 0.0036 mol) were dissolved in 70 g of NMP. The TSL was added to improve adhesion to the substrate. The solution was slowly dripped into the reaction flask held at 10°C. The liquid mixture was stirred for 6 h at 10–15°C, thereby synthesizing polyamic acid (PAA-1). The NMP solution (concentration 0.5 g/dL) of the polyamic acid was examined for its inherent viscosity ( $\eta_{\text{inh}}$ ) at 30°C, and this was found to be 0.53 dL/g.

### PAA-2

In the same manner, polyamic acid (PAA-2) was synthesized by using 6FDA (44.456 g, 0.10 mol), ODA (9.6145 g, 0.048 mol), 6FAP (17.571 g, 0.048 mol), TSL (0.9958 g, 0.004 mol), and NMP (220 g) as a solvent.  $\eta_{\text{inh}}$  was 0.38 dL/g.

**PAA-3**

In the same manner, polyamic acid (PAA-3) was synthesized by using 6FDA (35.557 g, 0.08 mol), ODA (11.545 g, 0.0576 mol), 6FAP (7.028 g, 0.0192 mol), TSL (0.801 g, 0.0032 mol), and NMP (160 g) as a solvent.  $\eta_{inh}$  was 0.63 dL/g.

**PAA-4**

In the same manner, polyamic acid (PAA-4) was synthesized by using 6FDA (39.983 g, 0.09 mol), ODA (16.772 g, 0.0837 mol), TSL (0.896 g, 0.0036 mol), and NMP (205 g) as a solvent.  $\eta_{inh}$  was 0.51 dL/g.

In this study, copolyimide was used for several reasons: improving adhesion to the substrate, bringing the substrate into a more desirable planarizability, and making the pattern-processing easy.

**POLYAMIC ACID RESIST**

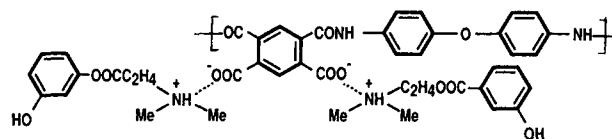
To a polyamic acid (PAA) solution (solvent: NMP), *m*- and/or *p*-PEA was added. The mixture was fully stirred, forming a homogeneous solution. To this solution, a solution prepared by dissolving 4NT-300 in NMP was added. The resulting liquid mixture was thoroughly stirred to prepare a homogeneous solution. This solution was passed through a membrane filter with a pore size of 0.5  $\mu\text{m}$ , yielding a resist with a PAA composition. The resists used in this study are summarized in Table I.

**LITHOGRAPHIC EVALUATION**

Resist was coated onto a silicon wafer and baked on a hot plate at 90–100°C for 30–90 min. The thickness of the resist film was about 5  $\mu\text{m}$ . The wafer was exposed to ultraviolet light using a PLA-501F mask aligner (Canon: superhigh-pressure mercury lamp, 250 W). The resist was developed using AD-10

**Table I** Composition of Resists

|                   | Resist-1 | Resist-2 | Resist-3 |
|-------------------|----------|----------|----------|
| PAA-2 (g)         | 2.36     |          |          |
| PAA-3 (g)         |          | 2.33     |          |
| PAA-4 (g)         |          |          | 2.49     |
| <i>m</i> -PEA (g) | 0–1.36   | 0–1.42   | 0.66     |
| <i>p</i> -PEA (g) |          |          | 0.94     |
| 4NT-300 (g)       | 0.47     | 0.46     | 0.5      |
| NMP (g)           | 8.6      | 9.6      | 8        |

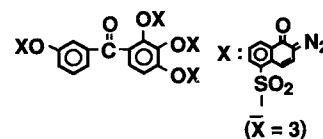
**Scheme 1** Structure of photosensitive polyamic acid derivative.

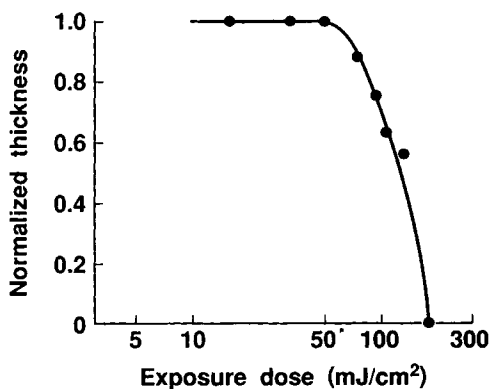
(Tama Chemical's: positive resist developer, 2.38 wt % tetramethylammonium hydroxide aqueous solution) or AD-10 diluted with water. This was followed by rinsing in water. After rinsing, the resist film was baked for full imidization at 150 and 250°C for 30 min each and 350°C for 60 min, using a Tabai Super Temp Oven STPH-100 in a nitrogen atmosphere. The solution velocity (solubilization rate) of the resist film in developing was calculated from the film thickness before and after immersion in the developer by dividing the reduction in thickness by the developing time.

**RESULTS AND DISCUSSION**

The positive-acting photosensitive polyimide consists of a polyamic acid, phenol derivatives which fill the role of solution protector, and a naphthoquinone diazide. When a phenol derivative is added to a PAA solution, the viscosity of the solution rises gradually. This means that the phenol derivative is bonded to the carboxyl group of PAA (Scheme 1). Scheme 2 shows the structure of 4NT-300 used. The sensitivity ( $D_0$ ) of the resist-3 film was evaluated from the characteristic curve, which was the plot of remaining film thickness after development against exposure dose.  $D_0$  was 176  $\text{mJ}/\text{cm}^2$  to UV light (365 nm; Fig. 1).

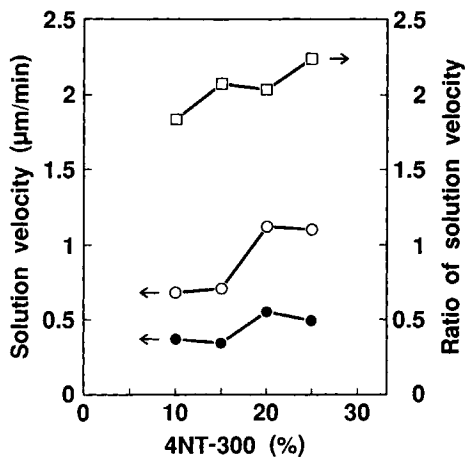
In Figure 2, the relationship between the amount of 4NT-300 and the solution velocity of exposed and unexposed parts of a resist film consisting of a polyamic acid, *m*-PEA, and 4NT-300 are shown. The polyamic acid used was PAA-4 and the *m*-PEA content was 50% equivalent of PAA-4 carboxyl groups. From this result, it was necessary for 4NT-300 to be used at 20 weight parts per 100 weight parts of PAA-4 in order to obtain good relief patterns, but,

**Scheme 2** Sensitizer (4NT-300).

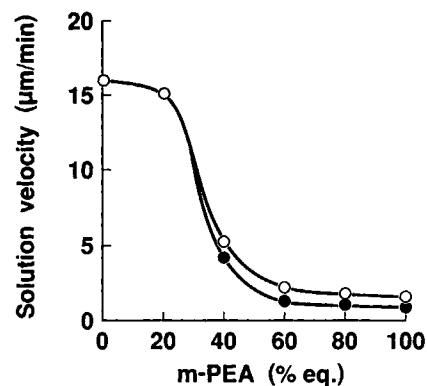


**Figure 1** UV sensitivity curve of the resist-3: prebaking temp/time, 100°C/30 min; developer, 2.38% TMAH aq (at 40°C); film thickness, 3.2  $\mu\text{m}$  (after development).

even then, the pattern contrast, i.e., the ratio of solution velocity (= 2.3), was low. Figure 3 shows the dependence of a concentration of *m*-PEA on solution velocity of exposed and unexposed parts of a resist-1 film in which the polyamic acid is PAA-2. Also, Figures 4 and 5 show the change in solution velocity of exposed and unexposed parts of a resist-1 film and a resist-2 film with an amount of *m*-PEA. The resists were baked at 100°C or 95°C for 30 min, exposed to light for 40 s, and developed with 0.595 wt % TMAH aq at 25°C. In both exposed and unexposed parts of the resists, the decrease in solution velocity is remarkable until the concentration of *m*-PEA reaches a 30% PAA carboxyl group equivalent. Also, above that level, the solution velocity does not



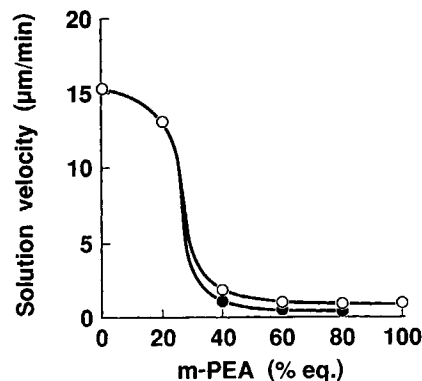
**Figure 2** Solution velocity of photosensitive polyimide: film thickness, 6.3–7.5  $\mu\text{m}$ , prebaking temp/time, 95°C/30 min; exposure dose, 500  $\text{mJ}/\text{cm}^2$ ; developer: 1.19% TMAH aq (at 25°C). (—○—) exposed; (—●—) unexposed; (—□—) ratio of solution velocity.



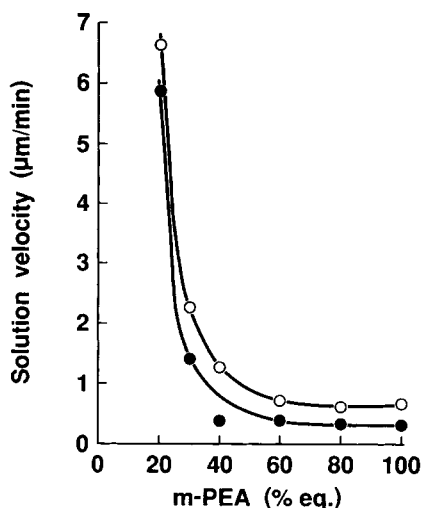
**Figure 3** Solution velocity of photosensitive polyimide (resist-1): film thickness, 3.3–4.8  $\mu\text{m}$ ; prebaking temp/time, 95°C/30 min; exposure dose, 500  $\text{mJ}/\text{cm}^2$ ; developer: 0.595% TMAH aq (at 25°C). (—○—) exposed; (—●—) unexposed.

depend on the amount of *m*-PEA and becomes very low. When a polyimide is immersed in TMAH aqueous solution, it hydrolyzes first to form a polyamic acid and dissolves soon after. A velocity of hydrolysis of a polyimide is very low. As the resist with *m*-PEA whose amount is more than a 30% PAA carboxyl group equivalent imidizes at prebaking temperature (90°C), it dissolves through hydrolysis of the polyimide in the alkaline solution. Because of this reason, the solution velocity of the resist becomes very low. On the other hand, the resist with *m*-PEA whose amount is less than 30% does not imidize at prebaking temperature, so it dissolves quickly.

Figure 6 shows the IR spectrum of PAA-1 containing *p*-PEA (100% PAA carboxyl group equiv-

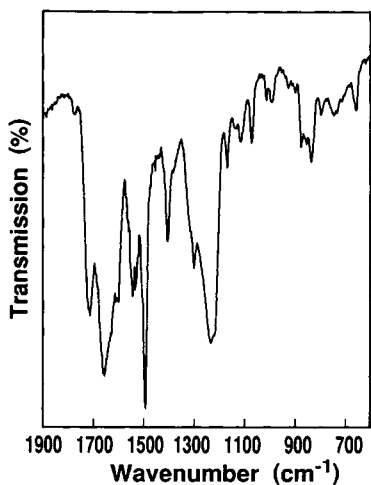


**Figure 4** Solution velocity of photosensitive polyimide (resist-1): film thickness, 4.8–7.1  $\mu\text{m}$ ; prebaking temp/time, 100°C/30 min; exposure dose, 500  $\text{mJ}/\text{cm}^2$ ; developer, 0.595% TMAH aq (at 25°C). (—○—) exposed; (—●—) unexposed.

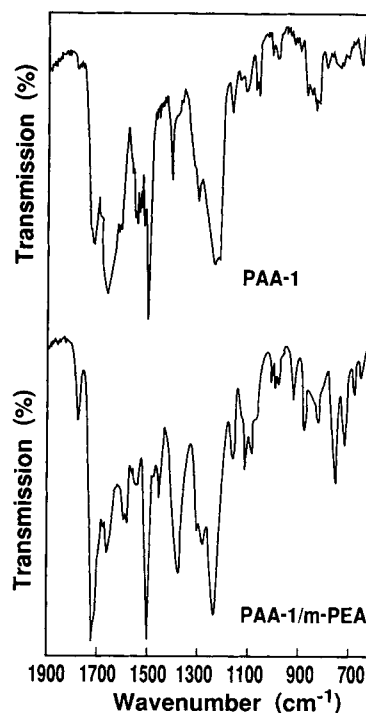


**Figure 5** Solution velocity of photosensitive polyimide (resist-2): film thickness, 6.1–7.6  $\mu\text{m}$ ; prebaking temp/time, 95°C/30 min; exposure dose, 500  $\text{mJ}/\text{cm}^2$ ; developer, 0.595% TMAH aq (at 25°C). (—○—) exposed; (—●—) unexposed.

alent) which was baked at 90°C for 1 h. This is the IR spectrum of a polyamic acid. On the other hand, in the IR spectrum of PAA-1 containing *m*-PEA (100% PAA-1 carboxyl group equivalent) baked under the same conditions, absorption peaks at 1780  $\text{cm}^{-1}$  for the two carbonyl stretching modes and at 1380 and 720  $\text{cm}^{-1}$ , respectively, for the C—N stretch and imide deformation modes are very clear (Fig. 7). This verifies that PAA-1 containing *m*-PEA imidizes at this low temperature. The combination of a solution protector and 4NT-300 does not thoroughly fulfill its function of making an unex-



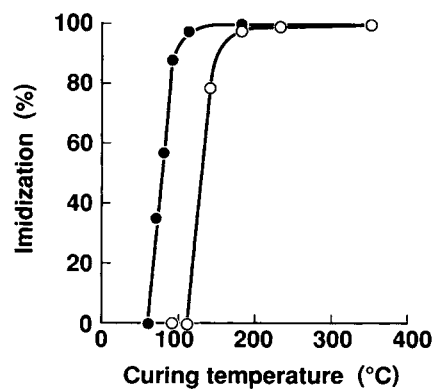
**Figure 6** IR spectrum of the polyamic acid (PAA-1) with *p*-PEA after baking at 90°C for 1 h.



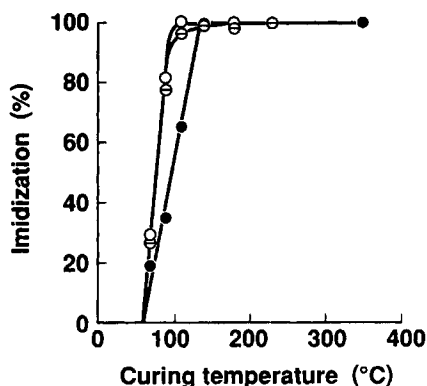
**Figure 7** IR spectra of polyamic acid (PAA-1) and PAA-1 with *m*-PEA after baking at 90°C for 1 h.

posed resist insoluble. This is also the reason for the contrast being very low.

The degree of PAA imidization was found to be dependent on both *m*-PEA concentration and temperature. Plots of the experimental results are given in Figures 8–10. The degree of imidization was measured by IR spectroscopy. Films were prepared on KBr plates and the samples were dried at 40°C for 2 h to remove the solvent. Curing then took place at the given temperature for 1 h. The percentage of imidization was calculated using the equation be-



**Figure 8** Imidization of polyamic acid vs. curing temperature: (—○—) polyamic acid (PAA-1); (—●—) PAA-1/*m*-PEA (100% equiv.).



**Figure 9** Imidization of polyamic acid (PAA-1) vs. curing temperature. *m*-PEA: (—●—) 25% equiv; (—○—) 50% equiv; (—□—) 75% equiv.

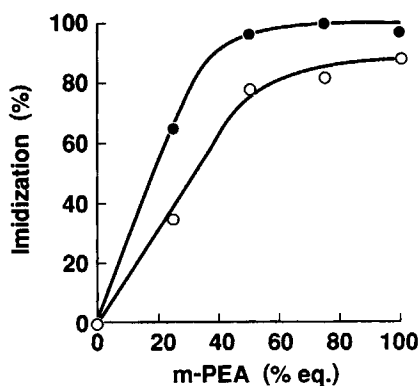
low.<sup>19</sup> Since measurements based on the 1780 and 720  $\text{cm}^{-1}$  bands suffered from dichroic effects, the 1380  $\text{cm}^{-1}$  band was used<sup>20</sup>:

Imidization (%)

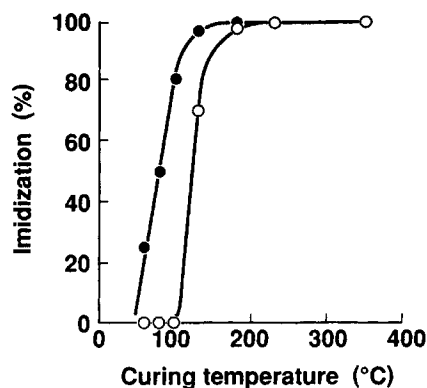
$$= \frac{Pi/Ps(t) - Pi/Ps(t=0)}{Pi/Ps(t=\infty) - Pi/Ps(t=0)} \times 100$$

$Pi$  is the absorbance of the imide peak at 1380  $\text{cm}^{-1}$ ,  $Ps$  is the absorbance of the standard reference peak at 1490  $\text{cm}^{-1}$ , and  $t = \infty$  was taken as the time beyond which no further change in the imide peak was observed at 350°C.

Figure 8 shows the relationship between imidization of polyamic acid consisting of *m*-PEA with a 100% PAA-1 carboxyl group equivalent and temperature. PAA-1 with *m*-PEA begins to imidize at 60°C, which is lower than the resist prebake temperature, and the imidization is not far from com-



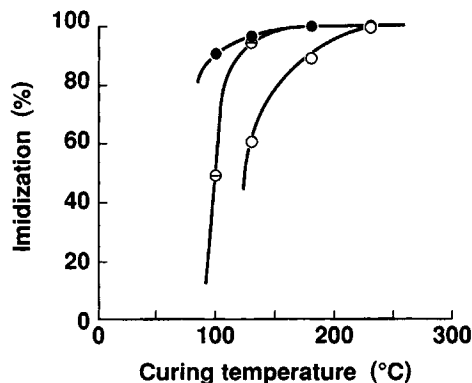
**Figure 10** Imidization of polyamic acid (PAA-1) vs. *m*-PEA concentration. Curing temperature: (—●—) 100°C; (—○—) 90°C.



**Figure 11** Imidization of polyamic acid vs. curing temperature. Polyamic acid: PMDA (1 mol)/ODA (0.94 mol)/TSL (0.06 mol);  $\eta_{inh} = 0.83$  dL/g; (—○—) polyamic acid; (—●—) polyamic acid/*m*-PEA (100% equiv).

pletion at 150°C. Meanwhile, polyamic acid by itself begins to imidize at 120°C. The percentage imidization depends on the *m*-PEA concentration, increasing linearly with *m*-PEA until a 60% PAA carboxyl group equivalent and then increasing more gradually (Figs. 9 and 10). Further, it was found that the latent curing function of *m*-PEA at low temperature did not depend on the molecular structure of the PAA (Figs. 11 and 12).

To examine the different functions of *m*-PEA and *p*-PEA, 0.01 mol aqueous solutions of these chemicals were measured for pH. The pH values of *m*-PEA and *p*-PEA were 7.78 and 9.64, respectively. The pH of *m*-PEA is not the value of tertiaryamine, so dissociation constants ( $pK_a$ ) of the two PEAs were investigated by acid-base titration. The results are shown in Table II. The dissociation of *m*-PEA took place in three steps and the  $pK_a(1)$  was 3.9,



**Figure 12** Imidization of polyamic acid vs. curing temperature. Polyamic acid: BTDA (1 mol)/ODA (0.94 mol)/TSL (0.06 mol);  $\eta_{inh} = 0.76$  dL/g. *m*-PEA: (—○—) 0% equiv; (—□—) 25% equiv; (—●—) 100% equiv.

which matches the dissociation constant of carboxylic acid compounds. This means that hydrolysis of *m*-PEA occurred, forming *m*-hydroxybenzoic acid and 2-*N,N*-dimethylaminoethanol.  $pK_a(2)$  ( $= 9.2$ ) is the dissociation constant of 2-*N,N*-dimethylaminoethanol and  $pK_a(3)$  ( $= 10.0$ ) is that of *m*-hydroxybenzoic acid. Furthermore, the water used in the hydrolysis of *m*-PEA comes from the organic solvent, the atmosphere, and the imidization by-product of PAA. These results demonstrate that *m*-hydroxybenzoic acid acts as a curing accelerant which imidizes PAA at low temperature when the resist is prebaked and it is clear that *m*-hydroxybenzoic acid converts the polyamic acid to the polyimide and does not imidize the polyamic acid ammonium salt in this resist system. On the other hand, *p*-PEA dissociates in two steps and does not hydrolyze. The reactivities of both types of PEA can be explained by Hammett's rule. Hammett's  $\sigma_p$  value of the hydroxyl group is  $-3.7$ , indicating an electron-donative group, and  $\sigma_m$  is  $0.12$ , indicating an electron-attractive group. Thus, a carbon in the *m*-PEA carbonyl group is easily attacked by hydroxyl anions being nucleophiles, following hydrolysis.

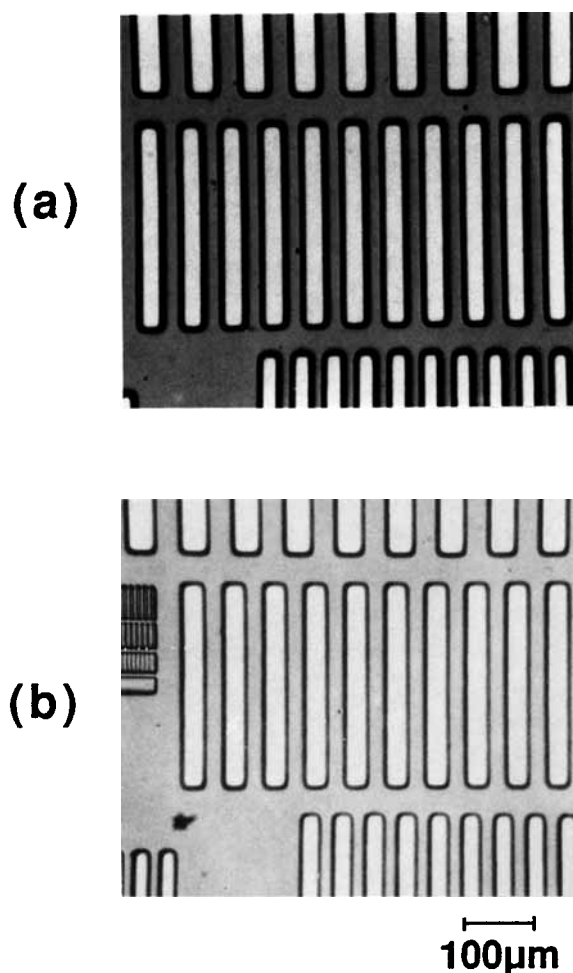
Figure 13 shows good patterns fabricated using resist-3. Lines and space patterns of less than 20 microns are resolved. To imidize the film, the wafer was heated to  $350^\circ\text{C}$ . No deformation was observed. The film thickness decreased to 70% with curing.

## CONCLUSION

Two solution protectors, 2-*N,N*-dimethylaminoethyl-3-hydroxybenzoate (*m*-PEA) and a 2-*N,N*-dimethylaminoethyl-4-hydroxybenzoate (*p*-PEA), were prepared. A novel positive-acting photosensitive polyimide consisting of a polyamic acid, *m*- and/or *p*-PEA, and a sensitizer, and developable with an alkaline aqueous solution, yielded satisfactory patterns with good profiles. The contrast, however, was low because the combination of *m*- and *p*-PEA incorporated into the polyamic acid and 4NT-300

**Table II**  $pK_a$  of PEAs (at RT)

|               |   | $pK_a$ |
|---------------|---|--------|
| <i>m</i> -PEA | 1 | 3.9    |
|               | 2 | 9.2    |
|               | 3 | 10.0   |
| <i>p</i> -PEA | 1 | 9.3    |
|               | 2 | 10.2   |



**Figure 13** Patterns fabricated by resist-3: (a) after development; (b) after curing at  $350^\circ\text{C}$ .

did not thoroughly fulfill its function of making an unexposed resist insoluble. Polyamic acid with *m*-PEA imidized partially at the prebaking temperature ( $90^\circ\text{C}$ ) and cyclized perfectly at  $150^\circ\text{C}$ , independent of the molecular structure of the polymer.

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