Available online at www.sciencedirect.com

ScienceDirect

Polymer 47 (2006) 6652–6658

polymer

www.elsevier.com/locate/polymer

A chemically amplified positive-working photosensitive polyimide based on a blend of poly(amic acid ethoxymethyl ester) and poly(amic acid)

Myung-Sup Jung [a,b], Won-Jae Joo [a], Byoung-Ki Choi [a], Hee-Tae Jung [b,*]

[a] *Materials Center, Samsung Advanced Institute of Technology, Mt. 14-1, Nongseo-dong, Giheung-gu, Yongin-si, Gyeonggi-do 446-712, Republic of Korea*

[b] *Department of Chemical and Biomolecular Engineering, Korea Advanced Institute of Science and Technology, 373-1 Guseong-dong, Yuseong-gu, Daejon 305-701, Republic of Korea*

Received 3 February 2006; received in revised form 2 June 2006; accepted 18 June 2006

Abstract

We prepared a novel chemically amplified photosensitive polyimide based on a blend of poly(amic acid ethoxymethyl ester) (PAAE) and poly(amic acid); this blend produces polyimide (PI) films with improved mechanical properties after imidization with photoacid generator (PAG). PAAE and poly(amic acid) were end-capped with 5-norbornene-2,3-dicarboxylic dianhydride and 2,3-dimethyl maleic anhydride, respectively, to lower their molecular weights without compromising the properties of the resulting PI films. As a result of the blending of these PI precursors, the mechanical properties of the PI films were found to be less affected by the strong acid generated from the PAG than PI films fabricated by imidization of PAAE alone. The relatively high solubility of the blended PI precursor film in basic aqueous solutions was found to be effectively controlled by the use of a high-temperature post-exposure bake process to partially imidize the end-capped PAA. It was found that a 10-μm-thick film of the PSPI precursor system containing 13 wt% PAGs exhibits a sensitivity (D^0) of 700 mJ/cm^2 when developed with 2.38 wt% aqueous tetramethyl ammonium hydroxide solution at room temperature. A fine positive pattern was fabricated in a 12 μm thick film with 1000 mJ/cm^2 of *i*-line exposure. The resultant PI film was also found to exhibit excellent mechanical and thermal properties, which are critical to its practical use as a stress buffer layer in semiconductor packaging.

Keywords: Photosensitive; Polyimide; Lithography

1. Introduction

Photosensitive polyimides (PSPIs) have attracted much attention in semiconductor manufacturing because their processing does not require the use of a classical photoresist [1]. Positive-working PSPIs have recently been used as replacements for conventional polyimides in stress buffer layers in semiconductor packaging. In such applications, PSPIs have to exhibit good mechanical properties such as high elongation at break and high tensile strength to protect bare chips from the stresses induced by fillers in molding compounds, and by the thermal mismatches that occur between passivation layers and molding compounds [2,3]. Another important requirement of stress buffer layers is high resolution and sensitivity, which are critical factors for the patterning of small features such as fuse windows in high-thick films (over ∼10 μm thick before curing) [3].

There are many reports of the fabrication of positive-working PSPIs, which are mainly prepared from PI precursors and dissolution inhibitors such as diazonaphthoquinone [4] and nifedipine [5]. Intrinsically PSPI with cyclobutane structures [6], chemically amplified PSPI with PAG [7], and the direct attachment of photosensitive groups to the PI precursors [8,9] have also been reported. Among these methods for fabricating PSPI precursor systems, chemical amplification, which enables advanced photoresist design, is an effective method for obtaining high resolution and sensitivity [10]. However, very few studies have explored the use of chemical amplification, and

---

* Corresponding author. Tel.: +82 42 869 3971; fax: +82 42 869 3910.
E-mail address: heetae@kaist.ac.kr (H.-T. Jung).

doi:10.1016/j.polymer.2006.06.039

moreover, those that have been carried out were limited in aspect of lithographic performance.

In comparison with regular chemically amplified deep-UV photoresists, chemically amplified PSPI systems need to use large amounts of PAG [7]. This might be due to the fact that the photogenerated acid is captured by the residual basic solvent, such as *N*-methyl-2-pyrrolidinone (NMP), and by the amide groups of the polymer backbone through acid–base reactions, which reduces the efficiency of the PAG. The use of large amounts of PAG and the presence of acid-labile groups in the PI precursor strongly affect the morphological structure of the PI precursor in the condensed state as well as the imidization kinetics, which results in variations of the structures and properties of the resulting PI films [11]. Therefore, although high resolution and sensitivity are easily obtained by the use of chemical amplification, it is hard to ensure the good mechanical properties of the resulting PI films. Thus, if the chemical amplification is to be used as a method for obtaining high resolution, new chemically amplified PSPI precursor systems that preserve the mechanical properties of the resulting PI films must be developed for the practical applications.

In a previous study, we incorporated reactive end-cappers into the ends of low molecular weight PAAE chains in order to improve the mechanical properties of the resulting PI films [12]. However, although the PI film imidized from end-capped PAAE (EC-PAAE) alone exhibits good mechanical properties in spite of the low molecular weight of its precursor, the mechanical properties of PI film imidized with PAG are still insufficient for the practical applications. As a continuation of our work on the development of aqueous-developable positive-working PSPIs, EC-PAAE was blended with end-capped poly(amic acid) (EC-PAA) in an attempt to improve the mechanical properties of the resulting PI films imidized with PAGs. This PSPI precursor system was found to produce PI films with high resolution and excellent mechanical properties.

2. Experimental section

2.1. Materials

4,4′-Oxydiphthalic anhydride (ODPA, Shanghai Research Institute, China) was used after vacuum-drying at 130 °C for 5 h. 4,4′-Oxydianiline (ODA, Wakayama Seika Kogyo Co. Ltd, Japan) and 1,3-bis(3-aminopropyl) tetramethyl disiloxane (SDA, Shin-Etsu Chemical Co. Ltd, Japan) were used as diamines. 5-Norbornene-2,3-dicarboxylic anhydride (NDA, Tokyo Kasei Kogyo Co. Ltd, Japan) and 2,3-dimethyl maleic anhydride (DMA, Aldrich) were purified by recrystallization from xylene, and used as reactive end-cappers. Anhydrous *N*-methyl-2-pyrrolidinone (NMP, BASF) was dehydrated with 4 Å molecular sieves before use. Diphenyliodonium 5-hydroxynaphthalene-1-sulfonate (DINS) was prepared with the method described in our previous report [13]. Dimethyl(4,7-dihydroxynaphthalene) sulfonium-*p*-toluene sulfonate (DDTS) was purchased from Uray Co. (Korea) and used without further purification. A 2.38 wt% tetramethyl ammonium hydroxide (TMAH) aqueous solution was purchased from Clariant Korea Ltd and used as a developer.

2.2. Synthesis and characterization of the PI precursors

EC-PAAE based on ODPA, ODA, and NDA was synthesized via the two-step procedure described in a previous report [12]. To provide an EC-PAAE solution with 30 wt% solid content, 3 g of EC-PAAE was dissolved in 7 g of NMP. EC-PAA was synthesized in the same manner as used in the polymerization step of the EC-PAAE. ODPA (0.0425 mol), ODA (0.0475 mol), and 1,3-bis(3-aminopropyl) tetramethyl disiloxane (SDA, Shin-Etsu Chemical Co. Ltd, Japan) (2.5 mmol) were used as monomers, and DMA (0.015 mol) was used as a reactive end-capper. Polymerization was carried out in 59 g of NMP to provide an EC-PAA solution with 30 wt% solid content. The imidization ratios of the PI precursors were analyzed by Fourier transform infrared (FT-IR) spectrometer (Bio-Rad FTS 3000) with attenuated total reflectance method.

2.3. Preparation and lithographic evaluation of the PSPI precursor system

As a base polymer for PSPI precursor system, 4 g of EC-PAAE solution (30 wt% in NMP) was blended with 6 g of EC-PAA solution (30 wt% in NMP). The PAG mixture (0.45 g) consisting of DINS and DDTS, and 0.04 g of 3-aminopropyltriethoxysilane solution (10 wt% in NMP) as an adhesion promoter were added to the PI precursor mixture solution and dissolved. The resulting solution was then filtered through a 1 μm poly(tetrafluoroethylene) filter to provide the PSPI precursor system. It was then spin-coated onto an Si wafer and prebaked at 90 °C for 4 min. A photomask was vacuum-pressed against the coated silicon wafer, which was then irradiated with filtered UV light with a wavelength of 365 nm from a high-pressure mercury lamp (ORIEL Instruments). The wafer was then post-exposure-baked at 130 °C for 3 min, developed with 2.38 wt% TMAH aqueous solution for 4 min, and washed with distilled water to give a clear-patterned film. The thickness of the patterned PSPI film was 12 μm after development, as measured with a surface profiler (TENCOR® P-10). The dissolution rate was determined by dividing the change in thickness of the developed film by the developing time, and a characteristic curve was obtained by plotting the normalized film thickness against the exposure energy. The pattern profiles were obtained with scanning electron microscopy (SEM, JEOL JSM-5600).

2.4. Characterization of the PI film

The PSPI precursor solution was spin-coated onto an Si wafer, and then heat-treated on a hot plate at 80 °C for 10 min. The PSPI precursor film was then thermally imidized in an electronic furnace (KDF S-80) with a multi-step process: heating at 5 °C/min up to 200 °C, heating at 200 °C for 30 min, heating at 4 °C/min up to 310 °C, and heating at 310 °C for 30 min in nitrogen ambient. The cured film was peeled from the wafer by soaking it in an aqueous solution of hydrofluoric acid (2 wt%) for 30 min. The peeled PI films were cut into test pieces 1 cm in width and 8 cm in length, and their mechanical properties were measured using a universal testing machine (Shimadzu AGS-G) in the tensile test mode with a testing rate of 3 mm/min. The film thickness was measured with a digital micrometer (Mitutoyo). The glass transition temperature (T_g) and thermal expansion coefficient (TEC) were measured with a thermomechanical analyzer (TA instruments TMA 2940) at a heating rate of 10 °C/min with 10 mN force. The thermal degradation temperature was measured with a thermogravimetric analyzer (TA instruments TGA 2950) at a heating rate of 10 °C/min from 50 °C to 800 °C.

3. Results and discussion

3.1. Synthesis of the PI precursors

Our PSPI developable with basic aqueous solutions was obtained from EC-PAAE, EC-PAA, and the *i*-line PAGs. EC-PAAE was synthesized with a two-step procedure as reported previously [12,13]. The protection ratio of the ethoxymethyl groups was optimized at about ~60%, at which point EC-PAAE is slightly soluble in the TMAH developer, which maximizes the difference between the dissolution rates of the exposed and unexposed areas. The NDA content was chosen to provide a 0.3 molar ratio with respect to ODA on the basis of the mechanical properties of the resulting PI film and the molecular weight of EC-PAAE. Therefore, the optimized EC-PAAE has an NDA molar ratio of 0.3 and an acetal protection ratio of 60%. Its number average molecular weight, measured at a sample concentration of 0.2 g/dL in NMP with an Ubbelohde viscometer at 30 °C, is 3700. EC-PAA was also synthesized via the condensation polymerization of ODPA, ODA, and SDA with DMA as a reactive end-capper. To improve the adhesion of the PSPI films to the Si wafers, a small amount (5 mol% relative to the diamine) of SDA was copolymerized. The structures of these PI precursors are shown in Scheme 1.

3.2. The blends of EC-PAAE with EC-PAA

PI films imidized from EC-PAAE have good mechanical properties in spite of the low molecular weight of their precursor, as reported previously [12]. However, the mechanical properties of PI films were found to be seriously affected when formulated with PAG, because the imidization of EC-PAAE is inhibited by the strong acid generated by the thermal degradation of the PAG during the high-temperature curing process. Fig. 1 shows the TGA curves of DINS and DDTS, which were used as *i*-line PAGs. They begin to decompose at around 220 °C and then generate strong acid. To investigate the imidization degree of the two precursors before the decomposition of PAG, the precursor films were imidized as follows: heating at 5 °C/min up to 220 °C and heating at 220 °C for 30 min. Then, the imidization degrees of EC-PAAE and EC-PAA were compared by the FT-IR analysis (Fig. 2). In the case of EC-PAA, strong imide absorption at 1780 cm^{-1} appeared and amide carbonyl absorption at around 1660 cm^{-1} completely disappeared, indicating complete imidization of EC-PAA. However, in the case of EC-PAAE, in spite of the appearance

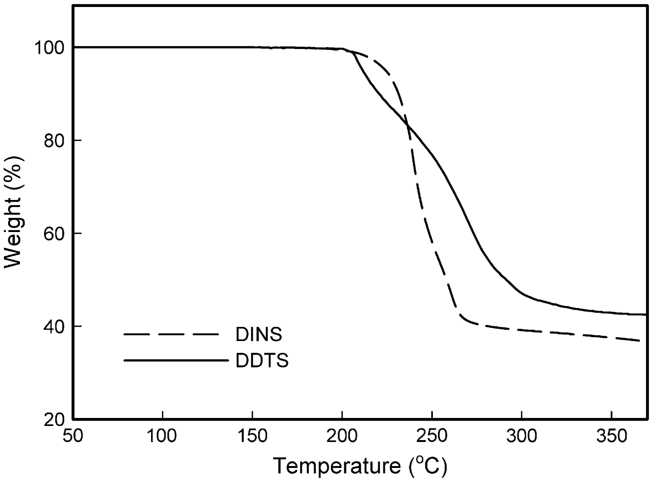
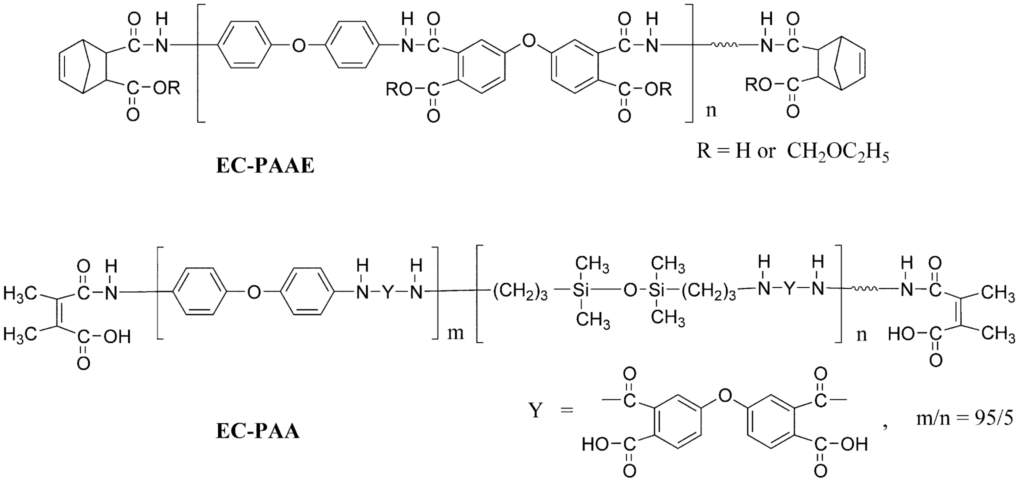


Fig. 1. TGA curves of DINS and DDTS.



Scheme 1. Structures of EC-PAAE and EC-PAA.

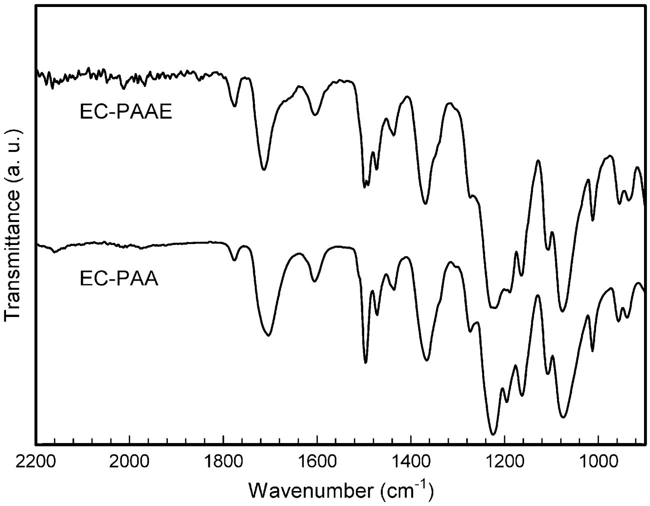


Fig. 2. FT-IR spectra of PI films imidized from EC-PAAE and EC-PAA at 220 °C.

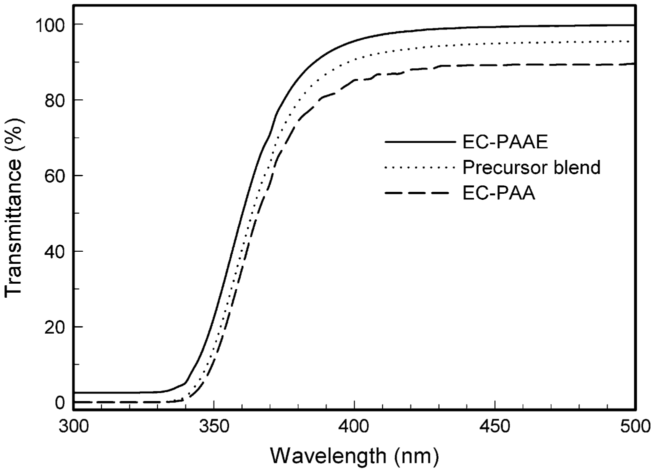


Fig. 3. UV-visible spectra of the PSPI precursors, for 10-μm-thick films. The blending ratio of the mixture was 4/6 (EC-PAAE/EC-PAA).

of imide absorption at 1776 cm^{-1}, amide carbonyl absorption at around 1660 cm^{-1} did not clearly disappear. This result clearly shows that imidization of amic ester segment in EC-PAAE is much slower than that of amic acid segment. In general, poly(amic ester) requires a higher temperature for imidization than poly(amic acid), because of the slow loss of ester groups from the precursor [14]. Therefore, the degree of imidization of EC-PAAE is very low at below 250 °C, and will increase at above 250 °C during the high-temperature (>300 °C) curing process. Thus, in PSPI precursor systems based on EC-PAAE and PAG, the imidization of EC-PAAE is strongly inhibited by the acid that is generated by the thermal degradation of the PAG at around 220 °C during the curing process. For this reason, although the elongation at break of PI films imidized from EC-PAAE alone is ~20%, it was found to be lowered to ~5% by the addition of 16 wt% PAG. Therefore, we decided to attempt to improve the mechanical properties of the resulting PI films by increasing the imidization ratio below the degradation temperature of the PAG, so we blended EC-PAAE with EC-PAA. Because EC-PAA is imidized to a significant extent below the degradation temperature of the PAG, the mechanical properties of the resulting PI films are less affected by the strong acid generated from the thermal degradation of the PAG than those of PI films obtained by imidizing PAAE alone. The blending ratio of EC-PAAE to EC-PAA was optimized to 4/6 (wt/wt) on the basis of their dissolution rates in TMAH developer and the mechanical properties of the PI films produced by imidization with PAGs.

The UV-visible spectra of 10 μm thick films of EC-PAAE, EC-PAA and their blends were obtained with a Jasco V-560 UV-visible spectrophotometer (Fig. 3). The transmittance of the blend at 365 nm is ~53%, which is not high, but is sufficient for the fabrication of high-resolution patterns in films with a thickness greater than 10 μm. The monomer composition, ODPA and ODA, was chosen by considering these factors, including the mechanical properties and transmittance.

3.3. Photolithographic evaluation

As discussed above, 60 wt% was chosen as the proportion of EC-PAA in the PI precursor mixture. At this high concentration of EC-PAA that has high solubility in basic aqueous solutions, the solubility of the blend in alkaline developer was too high for it to be used as a base polymer for a positive resist. Thus, the solubility of the blend must be lowered so that the unexposed region is not completely washed away by the alkaline developer. To control the dissolution rate of the blend, we used the high-temperature post-exposure bake (HIT-PEB) method in the photolithographic processing of the PSPI precursor system. It has been reported that the HIT-PEB process effectively lowers the solubility of poly(amic acid)s by inducing partial imidization [15–17]. Fig. 4 shows the relationship between the PEB temperature and the dissolution rate of the PI precursor mixture containing 13 wt% DINS. The dissolution rate was measured after 1000 mJ/cm^{-2} of UV exposure. In the low PEB temperature region, 110–120 °C, the dissolution rate of the unexposed area was found to be too fast. However, the dissolution

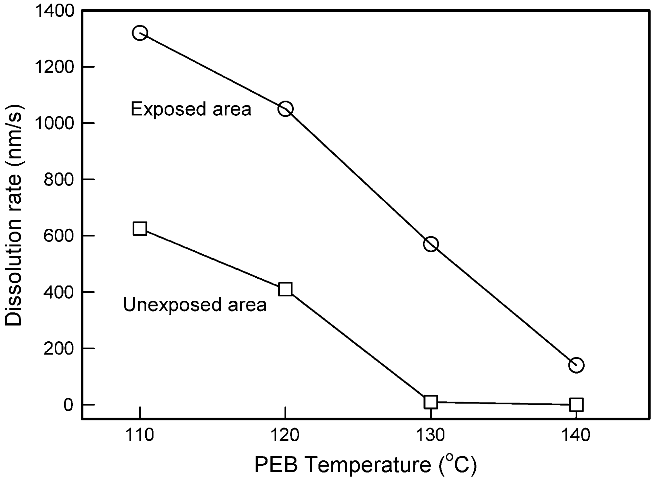


Fig. 4. Relationship between the PEB temperature and dissolution rate of a blended PSPI precursor film containing 13 wt% DINS in 2.38 wt% TMAH aqueous solution at 25 °C.

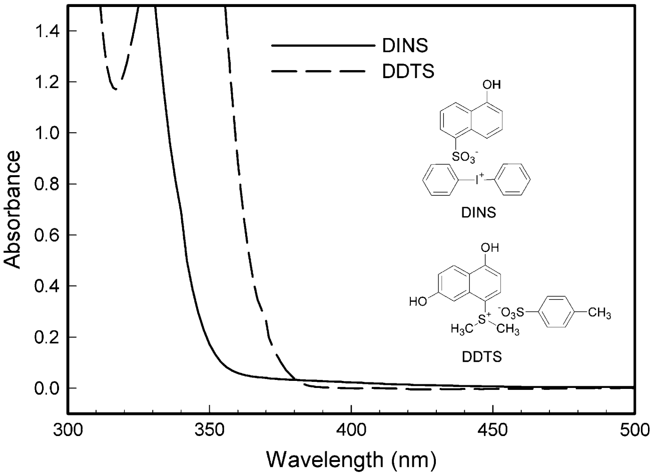


Fig. 5. UV-visible spectra of DINS and DDTS in MeOH.

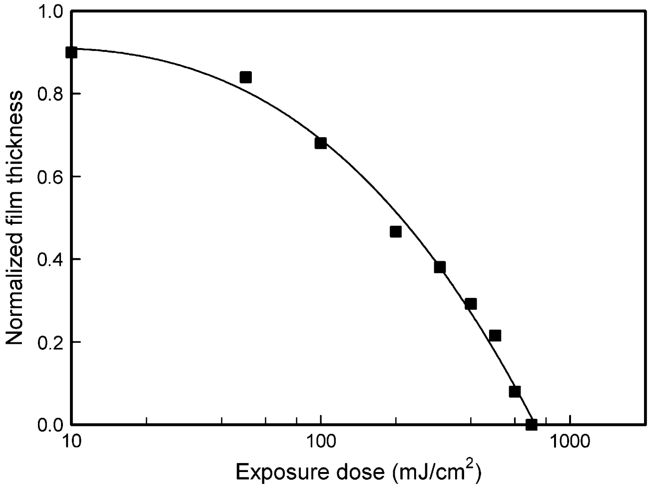


Fig. 6. Exposure characteristic curve of a blended PSPI precursor containing 13 wt% PAGs.

rate of the unexposed area was lowered at 130 °C to a satisfactory level for the fabrication of high-resolution patterns. Although the dissolution rate of the unexposed area was found to decrease to near zero after a PEB at 140 °C, the dissolution rate of the exposed area was then too slow. These results indicate that the EC-PAA in the unexposed area was slightly imidized at around 130 °C, which properly controlled the dissolution rate to fabricate high resolution patterns.

After these preliminary optimization studies, we prepared a PSPI precursor system based on the precursor mixture (EC-PAAE:EC-PAA = 4/6 (wt/wt)) and 13 wt% of the PAG mixture. The *i*-line PAG mixture consists of DINS and DDTS, which are highly soluble in NMP due to their hydroxyl groups. Fig. 5 shows the UV-visible spectra of DINS and DDTS in MeOH. These PAGs have absorption bands at 365 nm, and produce sulfonic acids on exposure to UV light. In the case of DINS, if the 5-hydroxynaphthalene moiety of DINS is excited, it donates an electron to the diphenyliodonium cation to form a neutral radical, which decomposes instantly to produce 5-hydroxynaphthalene-1-sulfonic acid. In the case of DDTS, the dimethyl(4,7-dihydroxynaphthalene) sulfonium moiety directly decomposes to produce *p*-toluenesulfonic acid. The generated acids deprotect the acid-labile acetal groups of PAAE during the HIT-PEB process.

The viscosity of the prepared PSPI precursor solution was measured with a viscometer (Brookfield LVDV II+) at 25 °C, and found to be 3210 cp with 35 wt% solid content including PI precursors and PAGs. This is an extremely low viscosity considering the high solid content, which results from the low molecular weights of the EC-PAAE and EC-PAA. This moderate viscosity with high solid content indicates that an effective single step spin-coating process will be possible for the fabrication of films with a thickness greater than 10 µm. It also means that purification processes, such as the filtration of the PSPI precursor solutions to remove dust particles or ionic impurities, are simplified.

The sensitivity curve for a 10 µm thick film is shown in Fig. 6, which indicates that its sensitivity (D^0) and contrast (γ^0) is 700 mJ/cm^2 and 0.87, respectively. Thus, relatively thick films of this PSPI precursor system exhibit higher sensitivity than those previously reported for chemically amplified PSPIs [7]. Fig. 7 shows SEM images of a positive pattern in a 12 µm

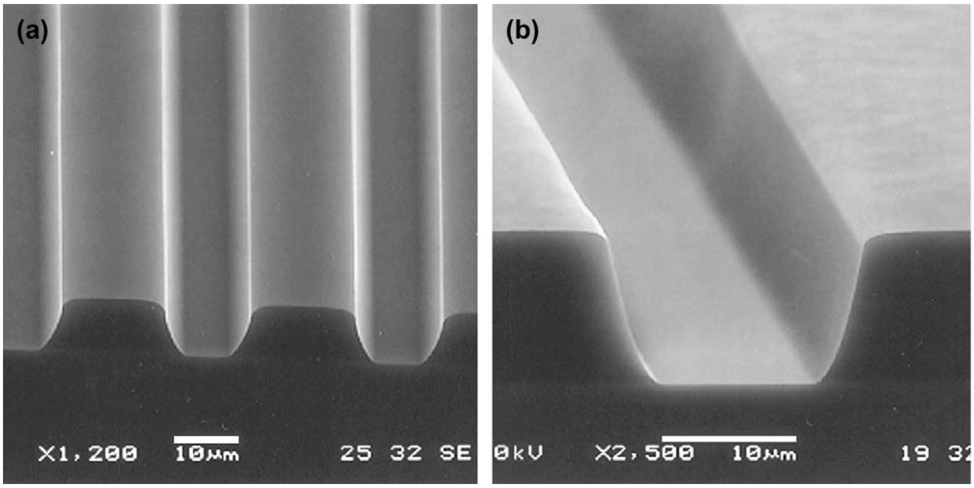


Fig. 7. SEM images of line patterns: (a) 6 µm space, (b) 10 µm space in 11.8 µm thick films.

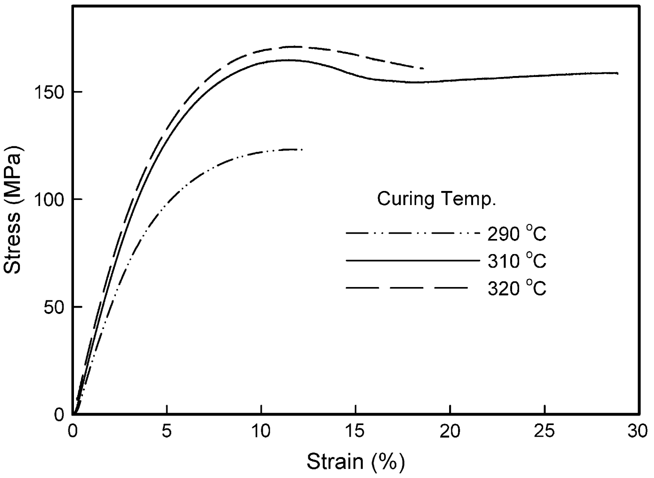


Fig. 8. Stress–strain curves of PI films obtained by curing the PSPI precursor system.

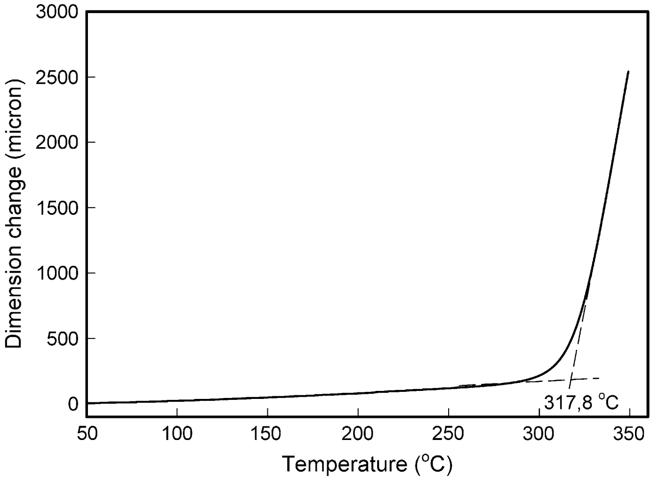


Fig. 9. TMA curve of the PSPI film imidized at 310 °C.

thick film of the PSPI precursor mixture. It was processed with PEB at 130 °C for 3 min after exposure of the film to 1000 mJ/cm^3. Fig. 6a and b shows vertical images of well-defined 6 and 10 μm positive line patterns in an 11.8 μm thick film, respectively. These results confirm that chemical amplification with the HIT-PEB process effectively controls the dissolution rates of the exposed and unexposed areas.

3.4. Physical properties of the PSPI film

For practical applications such as stress buffers and the dielectric layers of semiconductor packaging, positive-working PSPIs must exhibit excellent mechanical and thermal properties with high resolution. Fig. 8 shows the mechanical properties of PI films obtained from the precursor system containing the polymer blend and 13 wt% PAGs for various curing temperatures. The PI film imidized at 310 °C (PSPI-310) has good mechanical properties with plastic deformation. This result confirms that the imidization of EC-PAA is less affected by the acid generated from the thermal degradation of the PAGs, and that it plays an important role in improving the mechanical properties. The PI film imidized at 290 °C has poor mechanical properties due to insufficient imidization, and the PI film imidized at 320 °C exhibits rigid-brittle behavior due to further thermal reactions between the reactive end-cappers. The optimum imidization temperature is thus around 310 °C. PSPI-310 exhibits a high T_g and a low thermal expansion coefficient (Fig. 9). The T_g was found to be 317 °C, and TCE was calculated to be 36 ppm (50–100 °C) and 58 ppm (100–200 °C). These results are higher and lower, respectively than those for conventional polyimides because of the highly cross-linked structure of the PSPI films formed by thermal reactions between the reactive end-cappers. The 5% thermal degradation temperature ($Td_{5\%}$) was measured with TGA and found to be 480 °C. This value is lower than that of conventional polyimides, probably because of the aliphatic cross-linking structure and partially unimidized bonding of EC-PAAEs. Finally, PSPI-310 exhibits good thermal stability and thermomechanical properties because of its chain-extended and cross-linked structure. Fig. 10 shows the PI pattern obtained after the thermal curing of the PI film at 310 °C. Fig. 10a and b shows 10 μm L/S and 10 μm diameter hole patterns of the PI film, respectively. The patterned film thickness was reduced by ~29% compared to that of the precursor film by the thermal curing process. This film shrinkage is probably due to the imidization reaction, the loss of the ethoxymethyl

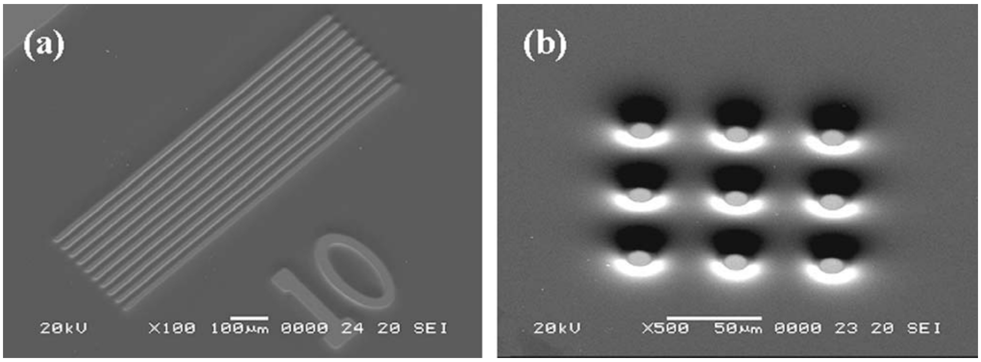


Fig. 10. SEM images of PI patterns after thermal curing at 310 °C: (a) 10 μm lines and spaces, and (b) 10 μm holes.

groups in the polymer, and the decomposition of the PAGs. However, no distortion or deformation of the pattern was observed after thermal curing. Further studies into chemically amplified PSPIs bearing reactive end-capper groups with high resolution and excellent physical properties are in progress, particularly with respect to the practical use of thick films in applications such as stress buffer layer and dielectric layer in semiconductor packaging.

4. Conclusions

To enhance the mechanical properties of PI films imidized from a chemically amplified PSPI precursor system containing large amounts of PAGs, EC-PAAE was blended with EC-PAA. However, although the thermal and mechanical properties of the resulting PI films are excellent, the dissolution rate of the blend in alkaline developer was found to be too high for it to be used as a base polymer. Therefore, the dissolution rates were controlled to the levels required for obtaining high-resolution patterns by using an HIT-PEB process (130 °C for 3 min). After the optimizations of photolithographic processes, a high-resolution pattern with 6 μm spaced lines was fabricated in an ~12 μm thick film by using the PSPI system; its resulting PI film exhibited ~30% elongation at break and a T_g of 317 °C. This is the first report of chemically amplified positive-working PSPIs satisfying two important requirements, high resolution and excellent mechanical properties, which make them useful as stress buffer layers, dielectric layers, and bond-pad-redistribution layers in semiconductor packaging.

Acknowledgments

This work was supported by the Mid-Term Technological Development Program of the Korea Ministry of Commerce, Industry and Energy. Partial support from the Brain Korea 21 program and RTI04-01-04:KMCIE are also acknowledged.

References

[1] Horie K, Yamashita T. Photosensitive polyimides, fundamentals and applications. Lancaster: Technomic; 1995.
[2] (a) Culver R. Solid State Technol November 1997:151;
(b) Prack E, Tran Z, Franklin C, Tanner S, Murray DO. Adv Elec Packaging 1997;1:1141.
[3] Flack WW, Flores GE, Christensen L, Newman G. Proc SPIE 1996; 2726:169.
[4] (a) Seino H, Haba O, Ueda M, Mochizuki A. Polymer 1999;40:551;
(b) Morita K, Ebara K, Shibasaki Y, Ueda M, Goto K, Tamai S. Polymer 2003;44:6235;
(c) Hsu SLC, Chen HT, Tsai ST. J Polym Sci Part A Polym Chem 2004;42:5990.
[5] Omote T, Yamaoka T. Polym Eng Sci 1992;32:1634.
[6] Padmanaban M, Toriumi M, Kakimoto MA, Imai Y. J Polym Sci Part A Polym Chem 1990;28:3261.
[7] (a) Omote T, Koseki K, Yamaoka T. Macromolecules 1990;23:4788;
(b) Watanabe Y, Shibasaki Y, Ando S, Ueda M. Chem Mater 2002;14: 1762;
(c) Ho BC, Chen JH, Perng WC, Lin CL, Chen LM. J Appl Polym Sci 1998;67:1313.
[8] Omote T, Mochizuki H, Koseki K, Yamaoka T. Macromolecules 1990;23:4796.
[9] Feng K, Matsmoto T, Kurosaki T. Chem Mater 1997;9:1362.
[10] (a) Reichmanis E, Houlihan FM, Nalamasu O, Neeman TX. Chem Mater 1991;3:394;
(b) Cameron JF, Aaza SL, Xu G, Yueh W. J Photopolym Sci Tech 1999;12:607.
[11] Ree MH, Nunes TL, Chen KJR. J Polym Sci Part B Polym Phys 1995;33:453.
[12] Yang SY, Park CE, Jung MS. Polymer 2003;44:3243.
[13] (a) Jung MS, Lee SK, Hyeon-Lee J, Park MK, Jung HT. J Polym Sci Part A Polym Chem 2005;43:5520;
(b) Jung MS, Seo SJ. US Patent 6541178; 2003.
[14] Coburn JC, Pottiger MT. Thermal curing in polyimide films and coatings. In: Ghosh MK, Mittal KL, editors. Polyimides, fundamentals and applications. New York: Marcel Dekker; 1996. p. 239.
[15] Hayase S, Mikogami Y, Takano K, Nakano YJ, Hayase R. Polym Adv Tech 1993;4:302.
[16] Hayase S, Takano K, Mikogami Y, Nakano Y. J Electrochem Soc 1991;138:3625.
[17] Takano K, Mikogami Y, Nakano Y, Hayase R, Hayase S. J Appl Polym Sci 1992;46:1137.