Electron Beam Lithographic Evaluation and Chain Scissioning Yields of Itaconate Resists*

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Synopsis

Copolymers of itaconic acid with methyl methacrylate, P(ItA–MMA), have been synthesized as promising positive working electron beam resists.[1,2] However, attempts to obtain greater electron beam sensitivity by increasing the itaconic acid content and initial molecular weight of these copolymers have heen hindered by difficulties in synthesizing itaconic acid copolymers with an ItA content greater than 50 mol % or with a molecular weight above 250,000. The usefulness of the MMA–ItA copolymers is also limited by their susceptibility to anhydride formation which makes this resist very sensitive to prebake conditions and aging times. To overcome these limitations and to develop resist materials with improved sensitivity, alkyl ester derivatives of itaconic acid have been synthesized, both as homopolymers and as copolymers with methyl methacrylate. The electron-beam chain scissioning yields, $G(s)$, of these derivatives have been determined, and the most promising of these copolymers and homopolymers have been evaluated for lithographic performance. The $G(s)$ values of the alkyl itaconate copolymers depend greatly on the structure of the alkyl group. The mono-alkyl itaconate copolymers exhibit $G(s)$ values 2–3 times greater than the corresponding dialkyl itaconate copolymers. In particular, copolymers of monomethyl itaconate (MeI) with methyl methacrylate are found to be promising resist materials with high sensitivities and compatability with processing conditions. A trend in sensitivity is observed for a series of MMA–MeI copolymers ranging from 20 to 85 mol % MeI, with a maximum sensitivity observed for the 57 and 73% MeI compositions. These copolymers exhibit improved sensitivity over that of the itaconic acid copolymers. Anhydride is formed less readily from the MeI copolymers than from the ItA copolymers, improving the stability of the resist for process conditions. Areas exposed in P(MMA–73 mol % MeI) at 4 $\mu C/cm^2$ (20 kV) were developed with less than 10% thinning of unexposed resist and with a contrast (γ) of 2. Vertical walls were observed for 1 μm wide lines using P(MMA–73% MeI) at a dose of about 6 $\mu C/cm^2$.

INTRODUCTION

Progress in the application of electron beam lithography to the manufacture of very large scale integrated circuits (VLSI) is currently limited by a lack of adequate resist materials. Specifically, resists with high sensitivity to electron beam degradation are needed in order to increase the writing speed of the electron beam and achieve an acceptable rate of throughput. Research in this area has been extensive, and investigation continues on a wide variety of materials, almost exclusively polymeric, as potential resists. Poly(methyl methacrylate) (PMMA) has received the most

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study and remains the most widely used e-beam resist because of its many favorable properties, but its sensitivity is poor (approximately 50 $\mu C/cm^2$ required for good pattern definition). The lithographic performance of PMMA has been considerably improved by copolymerization, but such copolymers have generally shown a decrease in thermal stability.[3] Two noted exceptions are the MMA–methacrylic acid copolymers[3] and the MMA–itaconic acid copolymers,[2] both of which exhibit an improvement in sensitivity over that of PMMA without a loss of thermal stability. Bargon and Hiroaka[4] reported that copolymers of poly(itaconic anhydride) resulted in better adhesion and faster development of images.

Our initial work with methyl methacrylate–itaconic acid copolymers (MMA–ItA) showed that incorporation of 35 mol % ItA raised the chain scission efficiency, $G(s)$, to nearly 5 times that of the PMMA homopolymer.[1] The itaconic acid copolymers were found to form anhydride upon prebaking, and in some cases, this anhydride formation led to improvements in sensitivity[2] similar to that reported for methacrylic acid copolymers.[3,5,6] P(MMA–35% ItA) exhibits 10% thinning of unexposed resist when developing a dose of 8 $\mu C/cm^2$, and 1 μm wide lines with nearly vertical walls can be produced with a dose of about 10 $\mu C/cm^2$ (20 kV accelerating voltage).[2] These data indicate about a fivefold improvement in sensitivity over that of PMMA and a slight improvement over the MMA–methacrylic acid copolymers.[2,6] However, further attempts to improve the sensitivity of the itaconic acid copolymers have been unsuccessful. The MMA–ItA copolymers could not be synthesized with ItA contents greater than 50 mol %, and copolymers synthesized in the 40–50% ItA range did not exhibit an appreciable improvement in sensitivity over that of P(MMA–35% ItA). It was also difficult to synthesize the itaconic acid copolymers with molecular weights greater than about 250,000, thus reducing the prospect of improving their sensitivity by increasing initial molecular weight. Another limitation of the itaconic acid copolymers is that they are very sensitive to prebake temperature, with changes in temperature of less than 5°C considerably altering the lithographic performance of the resist.

In an effort to produce a resist material with greater sensitivity and without the limitations of the itaconic acid copolymers, the methyl-,dimethyl, and ethyl esters of itaconic acid have been synthesized. The ester monomers are more reactive to propagation in polymerization than itaconic acid, allowing synthesis of itaconate–MMA copolymers with any desired ester content and with molecular weights above 10^6.

Chain scission efficiencies $G(s)$ were used for estimating the e-beam sensitivities of the itaconate copolymers, thus determining which materials merited more thorough lithographic evaluation. Electron beam $G(s)$ data for itaconic acid and dimethyl itaconate were reported previously.[1] $G(s)$ data for dimethyl itaconate, based on gamma irradiation, have also been reported.[7-9] The present work adds to this study the e-beam chain scission efficiences of the monomethyl and ethyl itaconate copolymers of methyl methacrylate.

Of particular interest are the monomethyl itaconate copolymers, P(MMA–MeI), that, based on the new $G(s)$ data, are expected to exhibit the highest sensitivity of these esters. The structure of methyl itaconate is

compared to those of methyl methacrylate, methacrylic acid, and itaconic acid in Figure 1. The methyl itaconate repeat unit retains the pendant carboxylic acid group of itaconic acid, the presence of which is believed to be largely responsible for the enhanced sensitivity of both methacrylic acid and itaconic acid copolymers. The major focus of this paper is the characterization and evaluation of a series of methyl itaconate–methyl methacrylate copolymers, as promising, highly sensitive e-beam resists.

EXPERIMENTAL

Synthesis. Monomers of the monoesters were prepared by refluxing itaconic anydride with a 3:1 excess of the appropriate alcohol in chloroform. The unconjugated carbonyl of itaconic anhydride preferentially reacts with alcohols to form 4-alkyl-2-methylene succinates.[10,11] This indicates that the carboxylic acid group directly adjacent to the polymer backbone remains unreacted, thus forming the monoester shown in Figure 1. The identity of this product was confirmed by melting point determination.

Monomers of the diesters were prepared by refluxing itaconic acid with an excess of alcohol in benzene, using a small amount of sulfuric acid as a catalyst.

The polymers were synthesized by free radical initiation using benzoyl peroxide with both solution and bulk polymerizations. The solution polymerizations were conducted in acetone or methanol at 50°C with 20–30% conversions. The bulk polymerizations at 70°C gave conversions up to 50%.

The polymers used for lithographic evaluations were fractionally precipitated from solutions in acetone or methanol by addition of water or benzene. These high molecular weight fractions were about 10–20% of the original weight of the unfractionated polymers.

Polymer Characterization. The molecular weights of those polymers that are soluble in tetrahydrofuran were determined by gel permeation chromatography (GPC). Otherwise, intrinsic viscosities were determined in methanol at 30°C as an indication of molecular size. Compositions of the monoalkyl itaconate–MMA copolymers were determined by titration of the free acid of the itaconate half esters with sodium hydroxide in methanol using a thymolphthalein indicator. Compositions of the itaconic acid and diester copolymers were determined by proton NMR.[1] The molecular

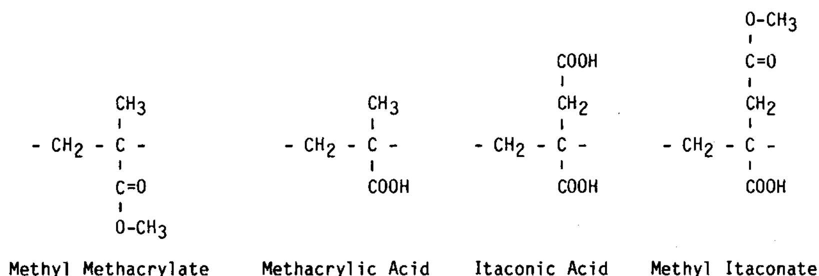

Fig. 1. Comparison of the repeat structure of methyl itaconate with those of methyl methacrylate, methacrylic acid, and itaconic acid.

TABLE I
Characterization of Methyl Itaconate–Methyl Methacrylate Copolymers

Polymer	Composition mol % MeI	Molecular weight M_n (GPC)	Molecular weight Intrinsic Viscosity (dL/g)	Casting solvent	Developing solvent
PMMA[a]	0	320,000	1.09	Chlorobenzene	Methyl isobutyl ketone
P(MMA–MeI)	38	99,000	—	2-Ethoxyethanol	2-ethoxyethyl acetate
P(MMA– MeI)	57	—	3.7	2-Ethoxyethanol[b]	2-Methoxyethyl acetate
P(MMA–MeI)	73	—	6.3	2-Methoxyethanol[b]	2-Methoxyethyl acetate
P(MMA–MeI)	85	—	2.1	2-Methoxyethanol	2-Methoxyethyl acetate
PMeI	100	—	0.9	2-Methoxyethanol	10:1 2-Propanol: water

[a] Obtained from KTI.
[b] Cast with double layers.

weights and compositions of the polymers used for lithographic evaluatons are shown in Table I.

Infrared spectra (IR) of the copolymers were recorded by direct measurement through coated silicon wafers using a Perkin-Elmer 683 Infrared Spectrophotometer. IR analysis of the monomethyl itaconate copolymers, prebaked *in vacuo* at 120°C, showed some formation of a five-membered anhydride ring similar to that formed from itaconic acid, but much less anhydride was formed in the case of the MeI copolymers than with the itaconic acid copolymers (Fig. 2). While methyl itaconate is thought to form six- and possibly seven-membered anhydride rings at higher temeratures,[12] this was not observed by IR analysis for these copolymers after the 120°C prebake. Intermolecular crosslinking did not occur to a large enough extent to effect the solubility characteristics of the films.

Thermal Analysis. Glass transition temperatures (T_g) of the MMA–MeI copolymers were determined in nitrogen by differential scanning calorimetry (DSC), using a Perkin-Elmer DSC-4, with a heating rate of 40°C/min. The T_g varied from 106°C for the PMMA homopolymer to 112°C for the PMeI homopolymer.

Thermal stability was determined by baking developed patterns at 150°C for 10 min and observing the effect of baking on line profiles.

Determination of *G*(*s*) Values. Chain scission efficiences were determined using flood electron beam exposures of coated wafers, followed by molecular weight determination by gel permeation chromatography (GPC), as described previously.[1] The molecular weight determinations were based on polystyrene equivalents. The absorbed dose was derived from the incident dose and the appropriate depth-dose function. Using this technique, the electron beam $G(s)$ for PMMA was about 0.8 scissions/100 eV, which agrees well with values reported by other researchers for electron beam exposures

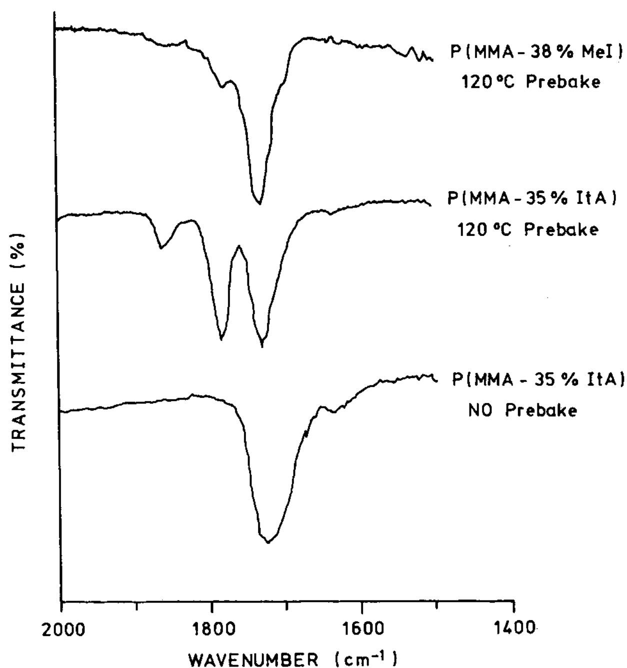

Fig. 2. Infrared spectra of methyl itaconate and itaconic acid copolymers with and without prebake at 120°C.

of PMMA.[13] $G(s)$ values for copolymers with itaconic acid or monoester contents above 50 mol % were not determined due to their insolubility in tetrahydrofuran, the solvent used for GPC analysis.

Lithographic Evaluation. Conventional spinning was used for all polymers except the very high molecular weight MeI copolymers (57 and 73% MeI) for which it was necessary to coat double layers, using low spin speeds in order to obtain adequate film thicknesses (1 μm or greater). The casting solvents used for spinning each polymer are listed in Table I. Water was an acceptable casting solvent for the PMeI homopolymer, but somewhat more uniform films were obtained using 2-methoxyethanol. Resist films were prebaked *in vacuo* at 120°C for 1 h, except for PMMA which was baked at 160°C for 1 h at atmospheric pressure. The vacuum oven baking was utilized for the copolymer films to help insure removal of the casting solvent, since the boiling points of the casting solvents were above 120°C. Film thicknesses were measured with a Tencor alpha-step.

Exposures were made with a Cambridge EBMF-II-150 at an accelerating voltage of 20 kV, without proximity correction. Test patterns and line profiles (scribed and broken under liquid nitrogen) were observed using a Cambridge SEM. Contrast and thinning data were obtained by measuring film thicknesses after development of a series of 20 μm wide lines exposed at a series of 16 incident doses.

Developing solvents, as listed in Table I, were chosen using a three-dimensional solubility mapping technique, described in a previous paper.[2]

Developing was conducted in unstirred solution and was terminated using an appropriate nonsolvent, followed by blowdrying with dry nitrogen. A 15-min postbake at 100°C was utilized after developing.

RESULTS

The monesters of itaconic acid yield much higher electron beam chain scission efficiencies $G(s)$ than the diesters (Fig. 3), presumably because of the pendant caboxylic acid group still present in the monoesters (Fig. 1). Other resist materials with carboxylic acid groups have been shown to exhibit high electron beam sensitivities, such as methacrylic acid [$G(s)$ = 3 times that of PMMA],[6] and itaconic acid, with two acid groups, which exhibits a still higher sensitivity [$G(s)$ = 4–5 times that of PMMA].[2] These high e-beam sensitivities have been attributed to the unstable characteristics of the carboxylic acid group, the removal of which induces the formation of propagating radicals.[14]

The monomethyl ester of itaconic acid exhibited a higher $G(s)$ than the monoethyl ester (Fig. 3). Harada[7] and Helbert et al.[8] also have reported slightly larger $G(s)$ values for acrylate polymers with smaller ester side groups.

For a given copolymer composition, the $G(s)$ values of the monomethyl itaconate (MeI) copolymers are lower than those of the itaconic acid copolymers (Fig. 3). However, the MeI copolymers can be synthesized with

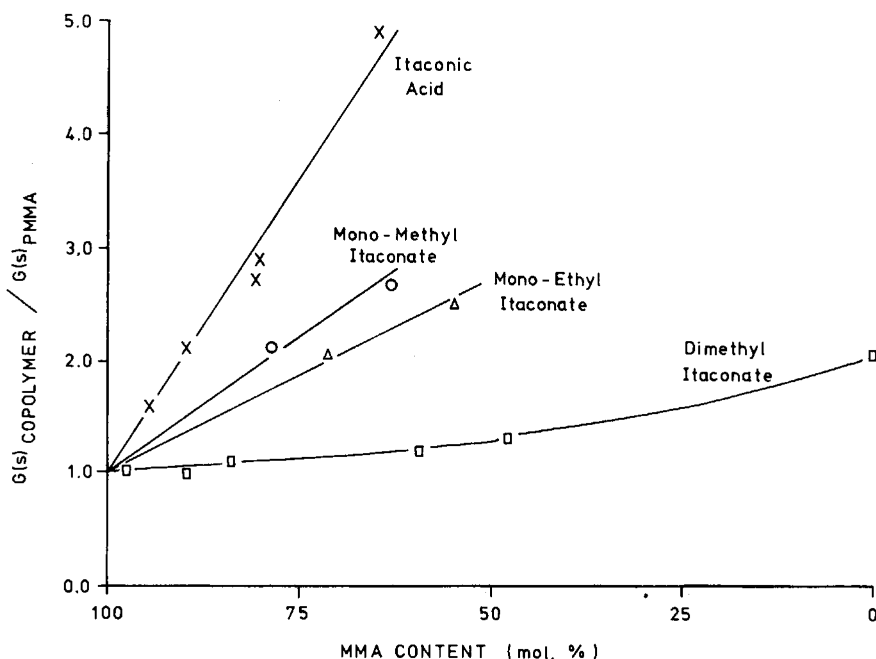

Fig. 3. Effect of copolymer composition on the electron beam $G(s)$ of itaconic acid and the methyl, ethyl, and dimethyl esters of itaconic acid. $G(s)$ data are reported relative to that of PMMA, which exhibited a $G(s)$ of 0.8 scissions/100 eV, which is consistent with reported values.[13]

MeI contents ranging up to 100% MeI, whereas the itaconic acid (ItA) copolymers could not be synthesized with ItA contents greater than 40–50 mol % ItA due to the low reactivity of the ItA monomer. The MeI copolymers with higher MeI content may be as susceptible to chain scissioning as the ItA copolymers containing 35% ItA (Fig. 3).

The MMA - MeI copolymers were synthesized with very high molecular weights, estimated at well over one million by intrinsic viscosity measurements (Table 1). High molecular weight resists have been found to exhibit certain practical limitations, including difficulty in synthesis, filtration, and spin-coating. It has also been reported that high molecular weight polymers swell during developing, thus resulting in distorted images.[15] The MMA–MeI copolymers, however, did not present these problems, with the exception of the requirement to spin-coat double layers to achieve film thicknesses of 1 μm (see Table I). Problems with swelling were not observed during development of any of the high molecular weight MeI copolymers.

In the evaluation of lithographic performance, a nonlinear relationship between copolymer composition and sensitivity was observed in which the MeI–MMA copolymers exhibited greater sensitivities than either of the homopolymers. (Fig. 4). The copolymers with the greatest sensitivities contained 57 and 73 mol % MeI. This nonlinear trend has been reported for many other copolymer resist systems,[16] such as the copolymers of methyl-α-chloroacrylate and methacrylate, which exhibit maximum sensitivity for a copolymer containing 38 mol % methyl-α-chloroacrylate.[17] The cause of these observations is uncertain because many factors are simultaneously influencing the sensitivity of the copolymers, e.g., interaction with developing system, molecular weight, and $G(s)$. For the MeI–MMA copolymers, the trend in sensitivity is most likely due to the increased susceptibility to chain scissioning of the methyl itaconate copolymers, but, unfortunately, the $G(s)$ values could not be determined for the copolymers with greater than 38 mol % MeI with the present technique. The observed trend in sensitivity (Fig. 4) also corresponds with the trend in intrinsic viscosity

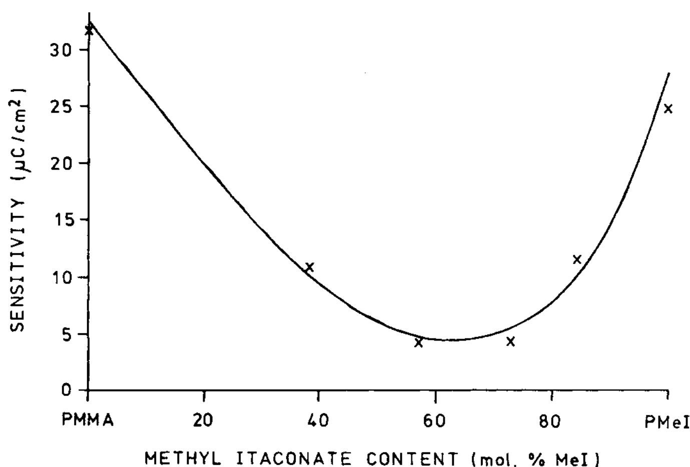

Fig. 4. Sensitivity of the methyl itaconate–methyl methacrylate copolymers as a function of copolymer composition. The sensitivity is based on 10% thinning of the unexposed resist.

measured for the copolymers (Table I), indicating the possibility of a molecular weight effect on sensitivity. However, little difference in sensitivity was observed for fractions of a P(MMA–50 mol % MeI) copolymer with intrinsic viscosities ranging from 0.8 to 2.8 dL/g. Initial molecular weight generally increases the solubility rate ratio of exposed to unexposed resist (S/S_0), particularly for low exposure doses and relatively low initial molecular weights.[15] On the basis of resist modeling techniques, it has been reported that the effect of initial molecular weight on sensitivity begins to level off for very high molecular weights.[18] The MMA–MeI copolymers evaluated all had very high molecular eights, with intrinsic viscosities of 2–6 dL/g (Table I).

The PMeI homopolymer was difficult to synthesize with a high molecular weight (Table I) and also exhibited several problems in lithography, including stress-cracking, poor adhesion, and scumming. The 85% MeI copolymer also exhibited some scumming, but none of these problems were encountered with the other MeI–MMA copolymers. Apparently, the methyl itaconate increases the sensitivity of the copolymers by contributing an e-beam sensitive link, while the methyl methacrylate helps in maintaining the good developing and film-forming characteristics of PMMA.

The MeI–MMA copolymers are less susceptible to anhydride formaton than the ItA–MMA copolymers and are thus less sensitive to prebake conditions and aging times than the ItA–MMA copolymers. Infrared analysis shows only slight anhydride formation at 120°C with MeI copolymers, whereas anhydride formation is extensive at this temperature with ItA copolymers (Fig. 2). When using high prebake temperatures, the itaconic acid copolymers exhibited solubility problems (most likely due to intermolecular anhydride formation) such as insolubility of unexposed films, difficulty developing patterns exposed at very low doses, and poor contrast. These solubility problems were not encountered with the MeI copolymers prebaked *in vacuo* at 120°C. Anhydride formation was shown to increase the thermal stability of other copolymers, including those of methacrylic acid[19] and itaconic acid.[2] In some cases, this anhydride formation also led to increased sensitivity by limiting the thinning of the unexposed resist.[3,5,6] With isothermal heating of the MeI–MMA copolymers at 140°C for 1 h, considerable anhydride formation is observed by IR analysis. Use of a prebake temperature of 140°C with the P(MMA–73% MeI) copolymer resulted in a slight improvement in thinning behavior, but this was accompanied by a loss of contrast and resolution. The remainder of the lithographic evaluations were therefore limited to the 120°C prebake for which the best pattern definitions were realized and for which fluctuations in prebake temperature were not expected to greatly alter lithographic performance.

The 57 and 73 mol % MeI copolymers exhibited only 10% thining of unexposed resist after developing 20 μm wide lines exposed at a dose of 4 $\mu C/cm^2$. A thinning curve is shown for P(MMA–73% MeI) in Figure 5. The thinning behavior of these MeI copolymers represents a considerable improvement over that previously reported for the ItA copolymers,[2] particularly with forced developing. The copolymers containing both 57 and 73 mol % MeI exhibited excellent resolution (<0.5 μm) at an exposure dose of 5 $\mu C/cm^2$ [P(MMA–73% MeI), Fig. 6]. The contrast (γ) obtained when

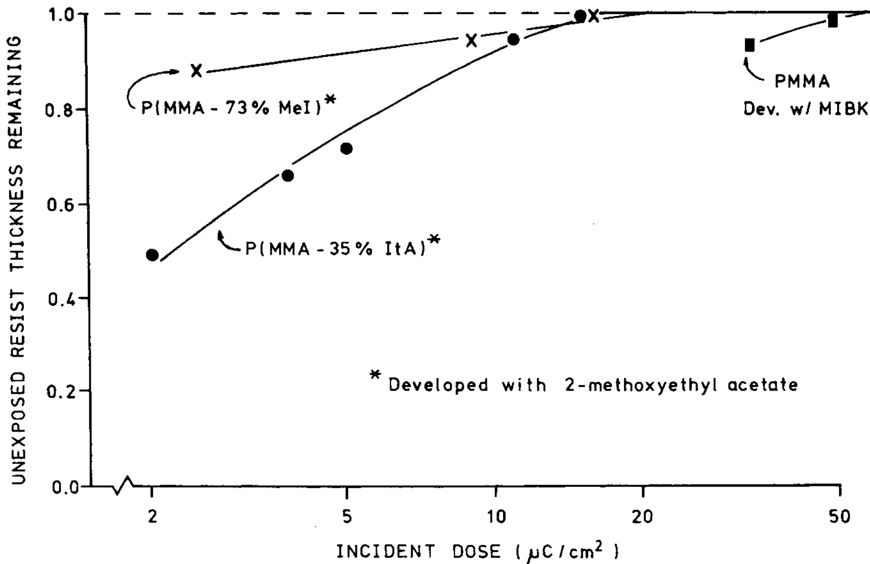

Fig. 5. Thinning characteristics of P(MMA–73% MeI) compared to that of an itaconic acid copolymer and PMMA. Shown on the ordinate is the amount of thinning of unexposed resist occurring for complete development of an area exposed at the dose given on the abscissa.

developing P(MMA–73% MeI) at 5μC/cm^2 was about 2. Profile control was better with the 73% MeI copolymer than with the 57% MeI copolymer. Nearly vertical-walled, 1μm wide line profiles were obtained with P(MMA–73% MeI) after exposure at 6μC/cm^2 and development with 2-methoxyethylacetate [Fig. 7(a)]. Undercutting, which is necessary for certain subsequent patterning steps, occurred at exposures above 10 μC/cm^2 [Fig. 7(b)]. These results represent a large improvement over those of the itaconic acid copolymers, which exhibited vertical walls with an exposure of 10 μC/cm^2 and development with 2-methoxyethyl acetate,[2] and those of methacrylic acid–methacrylic anhydride copolymers for which vertical-walled line profiles were reported at exposure doses of 14 μC/cm^2 (developed with 2-ethoxyethanol, accelerating voltage 25 kV).[6] It is important when comparing these doses to realize that the dose required to produce vertical wall profiles depends also on the developing conditions used.

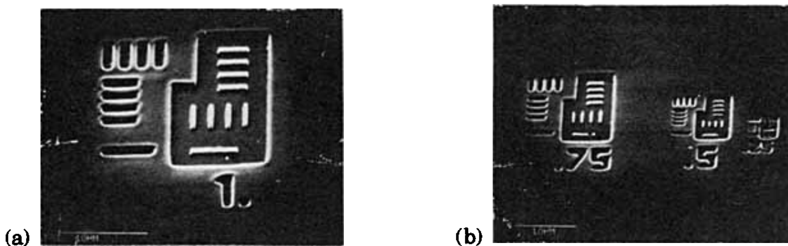

Fig. 6. Resolution of P(MMA–73% MeI) exposed with a dose of 5 μC/cm^2 at 20 KV and developed with 2-methoxyethyl acetate: (a) 1 μm pattern; (b) 0.75, 0.50, and 0.25 μm patterns.

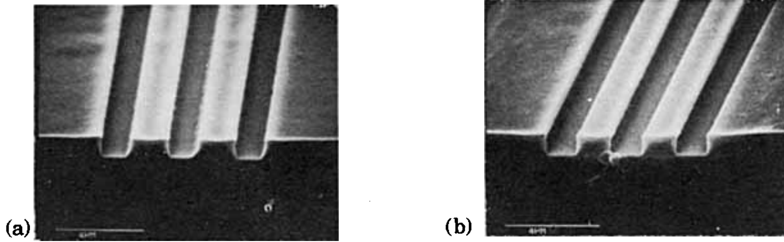

Fig. 7. Profile control with P(MMA–73% MeI): (a) nearly vertical-walled line profiles of 1 m wide lines exposed with a dose of 6 $\mu C/cm^2$; (b) undercut line profiles obtained at a dose of about 10 $\mu C/cm^2$.

The MeI–MMA copolymers exhibit good thermal stability. Patterns with nearly vertical-walled line profiles, developed in P(MMA–73% MeI), retained good pattern definition after baking for 10 min at 150°C (Fig. 8).

CONCLUSIONS

The methyl itaconate–methyl methacrylate (MeI–MMA) copolymers have demonstrated excellent lithographic performance. Development of P(MMA–73% MeI) at an exposure dose of 4 $\mu C/cm^2$ resulted in less than 10% thinning of the unexposed resist, and nearly vertical walls were obtained on 1 μm wide lines at a dose of 6 $\mu C/cm^2$. These results represent a considerable improvement in sensitivity over that of the itaconic acid copolymers. Methyl itaconate apparently contributes increased radiation sensitivity to the copolymer, while methyl methacrylate maintains good developing and film-forming properties for the copolymers.

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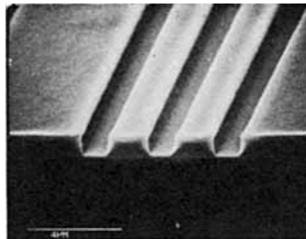

Fig. 8. Thermal stability of P(MMA–73% MeI). Original pattern with vertical-walled line profiles baked for 10 min at 150°C.

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