

Microlithographic Assessment of a Novel Family of Transparent and Etch-Resistant Chemically Amplified 193-nm Resists Based on Cyclopolymers

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The evaluation and structural optimization of a family of single layer, positive tone, chemically amplified resists for 193-nm lithography is presented. The resists are formulated from cyclopolymeric materials in which the nature, etch properties, and spatial disposition of the substituents are systematically varied. Their lithographic performance is evaluated on the basis of the interplay between chemical structure, molecular weight, and comonomer composition. These materials have good optical clarity at the desired wavelength, excellent resolution to ca. 90 nm with a phase-shifting mask, and outstanding reactive ion etch resistance.

1. Introduction

Photolithography with 248-nm KrF laser light is the current commercial standard used to form circuit features with dimensions as small as ca. 160 nm. This illumination source, however, may not be able to provide the smaller component sizes that future electronic devices will demand, necessitating a refinement of current printing technology. Alternative “unconventional” techniques¹ including some such as electron beam imaging or scanning probe lithography offering higher resolution have been proposed, but the high cost of implementing such fundamentally different processes has fueled further optimization of photolithographic methods.^{2,3} Specifically, experimental imaging with 193-nm ArF laser light appears most promising, allowing the generation of sub-100 nm dimensions with a phase-shifting mask.^{4,5} The basis for shorter wavelengths such as 193 and 157 nm providing better ultimate resolution

is fairly intuitive, and in fact, these two variables are directly proportional.

While the reduction of illumination wavelength appears to be a seemingly simple modification, in fact it presents a significant challenge because current 248-nm resists are opaque and therefore unusable at lower wavelength such as 193 nm. Clearly, resist materials with new structural features are warranted. These new materials, however, must provide not only transparency but also other important lithographic properties, including high sensitivity and reactive ion etch resistance. Sensitivity, typically provided by chemical amplification, is necessary because of the high energy of 193-nm light, which, if used at high doses, could damage the tool optics through absorption of some of the exposing radiation. Etch resistance enables the successful transfer of an imaged pattern into an underlying semiconductor substrate for circuit component formation.

Photoresists respond to incident radiation by undergoing a selective solubility change only in exposed areas. Imaging light causes either an increase in dissolution rate (positive tone resists) or a decrease (negative tone resists) relative to unexposed areas. Exploitation of this differential solubility by developer selectively removes the more soluble polymer chains and transfers a given pattern provided by previous selective illumination into

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the photoresist. Final pattern transfer into the silicon wafer is accomplished by a reactive ion etching process, with the photopolymer acting as an etch barrier for the underlying substrate.

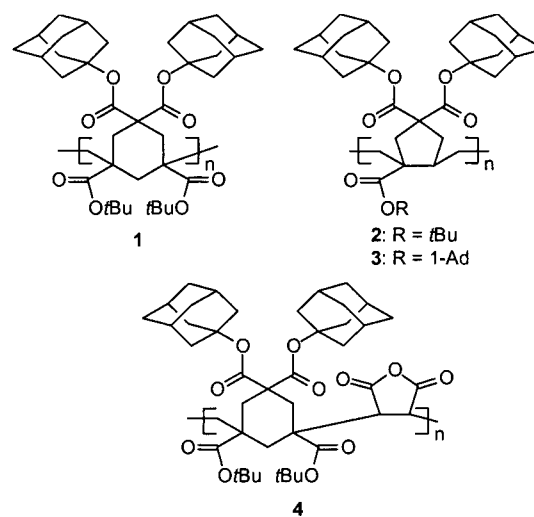
We now describe the lithographic evaluation of a family of novel 193-nm, chemically amplified, positive tone resists designed to meet the many demands of this multistep process. The key components, namely the specially designed cyclopolymers,⁶ used in formulating these resists were obtained via free-radical polymerization of suitable novel monomers, which are described in the preceding paper.⁷ Their chemical structures are summarized in Figure 1.

Multiple modular functionalities are evident in these cyclopolymers, all incorporated to provide various important lithographic properties. The various moieties and their corresponding lithographic utility include (a) tertiary esters for acid-catalyzed imaging; (b) adamantyl esters for their high carbon content, providing etch resistance; and (c) maleic anhydride and/or methacrylic acid comonomers for increased polarity, improved adhesion, and aqueous development characteristics. The first of these moieties, namely the tertiary ester, is essential for the high sensitivity provided through the chemical amplification mechanism.⁸ In the presence of a catalytic amount of photogenerated strong acid,⁹ it can be transformed from the aqueous base insoluble ester to the highly soluble carboxylic acid.⁸ In fact, because each photogenerated proton initiates the deprotection of multiple ester groups, the quantum efficiency of this overall process is greater than 1.⁸ An example of this chemically amplified resist chemistry is illustrated in Scheme 1.

2. Results and Discussion

For both the determination of material properties and the lithographic assessment of these polymers as photoresists, formulations, including various additives and a range of solvents, were spin-coated onto silicon wafers and processed as described in the Experimental Section.

Absorption of 193-nm Light. The UV absorption spectrum of a representative cyclopolymer **5**, shown in Figure 2, features an absorbance of 0.26 per micrometer of film thickness for this polymer at 193 nm. Very similar absorbance values ranging from 0.25 to 0.35 per micrometer of film thickness were found for polymers **1–6** at 193 nm. Since films with a thickness of 0.2–0.5 μm are actually used for 193-nm lithography, the films obtained from these polymers can be considered to be essentially transparent at this wavelength, enabling efficient and near-uniform distribution of the light throughout the resist film during exposure. The low absorbency of the polymers results from the absence of strongly absorbing chromophores in the polymer back-



Polymer	R	R'	R''
5	1-Ad	H	<i>t</i> Bu
6	1-Ad	Me	<i>t</i> Bu
7	<i>t</i> Bu	H	1-Ad
8	<i>t</i> Bu	Me	1-Ad
9	<i>t</i> Bu	Me	2-Me-2-Ad
10	2-Me-2-Ad	Me	2-Me-2-Ad

1-Ad =
2-Me-2-Ad =

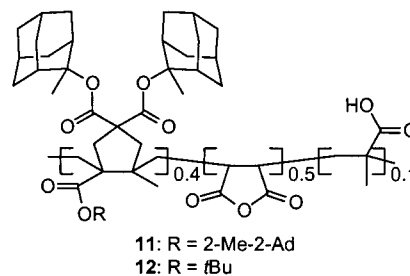


Figure 1. Cyclopolymers and cyclopolymers used as platforms for the resist formulations.

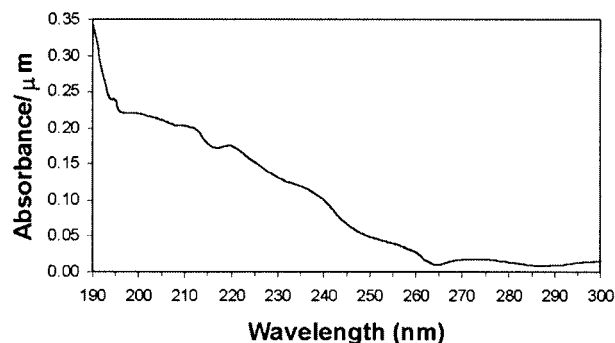


Figure 2. Absorbance of polymer **5** per micrometer of film thickness as measured by single-point ellipsometry over the range 190–300 nm. The absorbance at 193-nm is 0.26/ μm .

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bone. This further confirms the high efficiency and reliability of our synthetic cyclopolymerization protocol,⁷

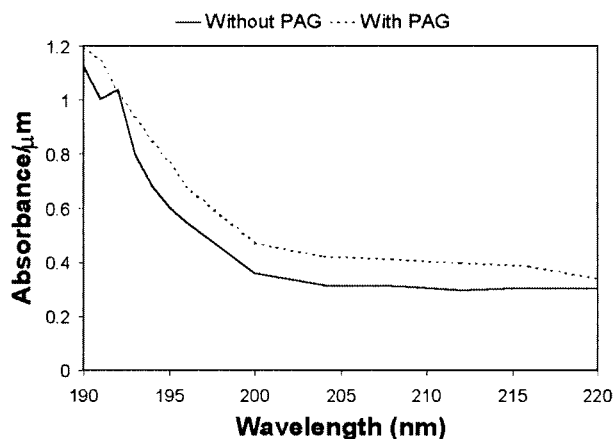
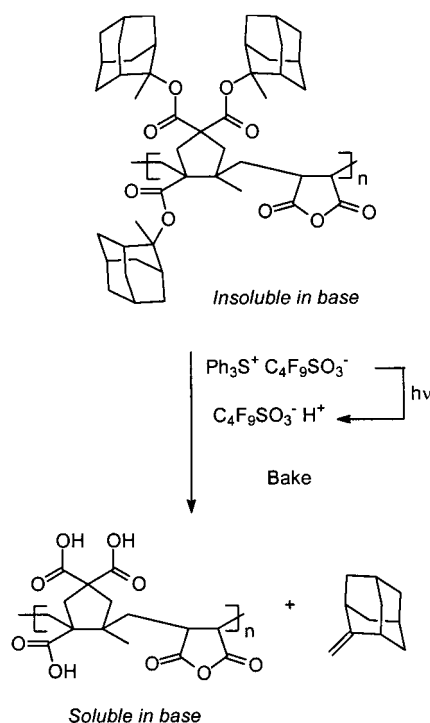


Figure 3. Absorbance of polymer **11** per μm of film thickness as measured by single-point ellipsometry over the range 190–220 nm. Full line: absorbance of the polymer matrix. Dotted line: absorbance of the resist formulation (containing 0.5 wt % PAG and 7.5 wt % *tert*-butyl cholate). The absorbances at 193 nm are $0.80/\mu\text{m}$ and $0.93/\mu\text{m}$.

Scheme 1



since unreacted double bonds would cause a large increase in absorption.

Figure 3 shows the UV spectrum of a film of cyclopolymer **11**, bearing 2-methyl-2-adamantyl esters, and of the corresponding resist formulation, containing 0.5 wt % of triphenylsulfonium nonafluorobutanesulfonate as the photoacid generator (PAG). Although the absorbance at 193 nm (0.80 per micrometer of film thickness) for this polymer is significantly higher than that for polymer **5**, it is still acceptable in terms of lithographic properties for the film thickness well below $1\ \mu\text{m}$ we use. The addition of both the highly absorbing PAG (0.5 wt % with respect to the polymer) as well as the optically transparent *tert*-butyl cholate additive (7.5 wt %) does not overly affect the final absorbance of the photoresist formulation, which reaches a value of 0.93 per micrometer of film thickness. However, these data illustrate the

effect of the PAG on total film absorbency. Each 1 wt % increment of PAG added to the photoresist formulation increases the overall absorbency by ca. 0.2 per micrometer of film thickness. Therefore, to obtain acceptable transmittance for the overall resist formulation, it is important to modify and tune the chemical properties of the polymer matrix and thus achieve highly sensitive resist systems that function efficiently with only very small amounts of PAG. An interesting modeling study outlining the effect of changes in the structure of photoacid generators on 193-nm resists has appeared recently.¹⁰

Assessment of Lithographic Imaging Performance. Carbon-rich homopolymers **1** and **2** were evaluated first. These polymers are insoluble in propylene glycol methyl ether acetate (PGMEA) but dissolve readily in cyclohexanone. Homopolymer **1** forms films that tend to crack upon development in aqueous base. Furthermore, the evaporative cooling process associated with the spin-coating procedure onto 8 in. wafers makes the polymer precipitate from the solution, yielding defects in the coating. Homopolymer **2** does not exhibit the same problems and produces good coatings on silicon wafers, probably as the result of its higher polarity when compared to homopolymer **1**. The structure of the repeat unit of homopolymer **1** is highly symmetrical; it includes a spirocarbon atom linking four structurally similar tertiary esters, two 1-adamantyl esters, and two *tert*-butyl esters. In contrast, homopolymer **2** contains repeat monomer units in which one of the tertiary ester groups has been effectively removed from the polymer backbone, increasing the polarity of the structure.

Both homopolymers are too hydrophobic to be imaged with good resolution and both required a relatively large amount of PAG (ca 10 wt %) to be imaged at relatively low doses ($20\ \text{mJ}/\text{cm}^2$). As expected, homopolymer **3**, in which the pendant groups are 1-adamantyl esters, could not be imaged, even in formulations containing as much as 20 wt % of PAG, since its 1-adamantyl ester groups cannot undergo catalyzed acidolysis, as such a process would afford 1-adamantene, an anti-Bredt, highly unstable olefin.⁷

Preliminary evaluation of several photoacid generators⁹ in related photoresists showed that best results in terms of resolution and sensitivity were obtained with triphenylsulfonium nonafluorobutanesulfonate.^{5b} The fluoroalkyl sulfonic acid that is generated photochemically has been shown to have little or no volatility and therefore provides best results in avoiding unwanted webbing and “T-top” formation in the resulting images.¹¹

Since added hydrophilicity appeared desirable, the copolymers with maleic anhydride were then evaluated. Maleic anhydride has become a popular choice as a polar comonomer in many 193-nm polymer resist systems, because of its ability to enhance the wettability and the adhesion to silicon substrates of the corresponding resist

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Table 1. Dependence of Clearing Dose on Polymer Characteristics^a

polymer	R	R'	R''	wt % PAG	mol % base	wt % cholate	M_n^b	M_w^b	PD ^b	clearing dose (mJ/cm ²)
5	1-Ad	H	<i>t</i> -Bu	4	8.5	7.5	4600	8500	1.8	18
5				4	8.5	7.5	2300	2900	1.3	7
6	1-Ad	Me	<i>t</i> -Bu	4	8.5	7.5	6900	19500	2.8	38
6				4	8.5	7.5	2900	4300	1.5	16
7	<i>t</i> -Bu	H	1-Ad	4	17	7.5	10000	16700	1.7	2
7				4	17	7.5	6900	12200	1.8	2
8	<i>t</i> -Bu	Me	1-Ad	4	17	7.5	21300	52700	2.5	4
10	2-Ad	Me	2-Ad	1	17	7.5	2700	4200	1.5	9
10	2-Ad	Me	2-Ad	1	17	7.5	7300	15200	2.1	7

^a Clearing dose defined as the minimum amount of light required to achieve full clearance of the photoresist after development. ^b As determined by GPC relative to polystyrene standards.

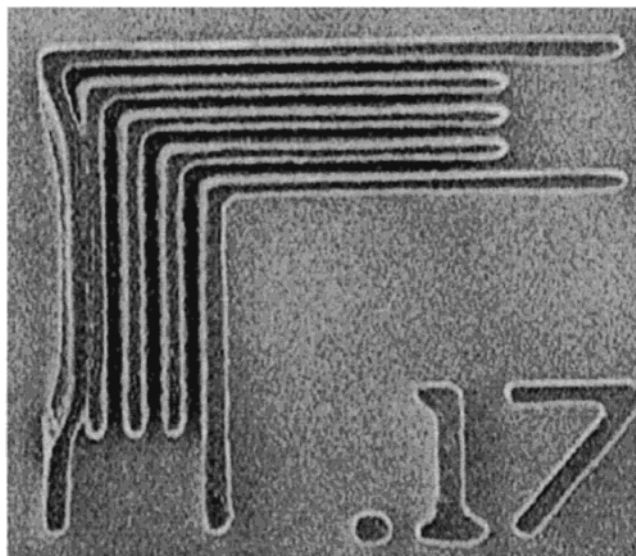


Figure 4. 170-nm 1:1 L/S features obtained by formulations containing polymer **4** after exposure using a 193-nm stepper (8 wt % PAG). Dose: 13 mJ/cm².

formulations.^{4,5,11} Resists containing copolymer **4** showed excellent resolution down to 0.18 μm 1:1 line/space (L/S) features, but they were still too hydrophobic, as they had to be developed by full immersion of the wafer in the aqueous developer. As can be seen in Figure 4, this led to pattern collapse during the develop/rinse step. In addition, the amount of PAG (8 wt %) necessary to obtain this lithographic resolution was unacceptably high for practical purposes, since the absorbance of the resulting photoresist formulation is too high.

On the other hand, resist formulations containing copolymer **5** were certainly more promising in terms of lithographic properties. They showed increased hydrophilicity when compared to equivalent formulations prepared from polymer **4**. This is demonstrated by the fact that images at the resolution shown in Figure 5 could be obtained using the industry-standard aqueous base development, with puddles of developer spread on the resist surface. The sensitivity of the resist formulations was also increased (with 5 wt % PAG the clearing dose was 2 mJ/cm²). We speculate that the increased hydrophilicity of polymer **5** when compared to polymer **4** can be attributed to the presence of a more asymmetric carbon-rich monomer with a higher dipole moment. Optimization of the lithographic conditions by addition of *tert*-butyl cholate¹² as a plasticizer⁴ and of a

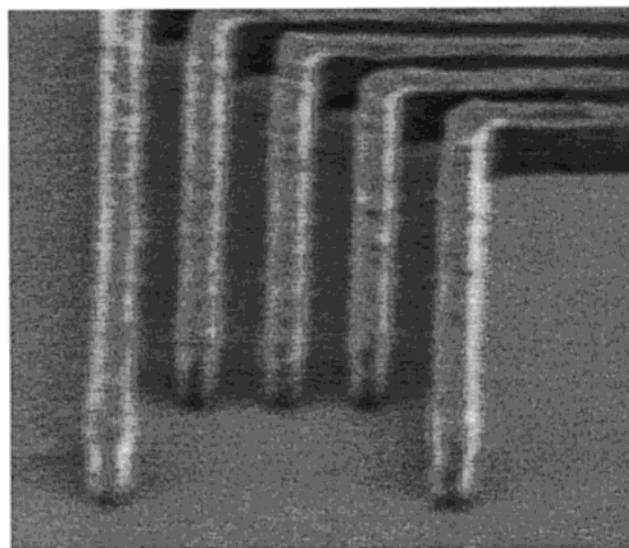


Figure 5. 170-nm 1:1 L/S features obtained by formulations containing polymer **5** after exposure using a 193-nm stepper (4 wt % PAG, 7.5 wt % *tert*-butyl cholate, 8.5 mol % 1-morpholineethanol). Dose: 17 mJ/cm².

base quencher (either 1-morpholineethanol or 1-piperidineethanol)^{11b,c} improved the ultimate resolution to 0.16–0.15 μm 1:1 L/S features, with 0.17 μm features shown in Figure 5.

An evaluation of the properties of samples of polymer **5** having different molecular weight distributions, however, revealed a steep dependence of the clearing dose (the minimum amount of light required to achieve full clearance of the photoresist after development) and of the imaging performance on the molecular weight characteristics, as shown in Table 1.

For example, two samples of polymers **5** having slightly different molecular weights (entry 1 and 2 in Table 1) showed significantly different clearing doses, when all other formulation components remained the same. For polymer **6**, the clearing dose is as high as 38 mJ/cm², with resolved features of dimensions $>0.20 \mu\text{m}$ when $M_n = 6900$, but it is less than half when $M_n = 2900$, and the resolution is improved (entry 3 and 4 in Table 1).

Lithographic evaluation of resist formulations containing polymers **7** and **8** showed once again that the materials had excellent resolution capabilities, good wettability, and high sensitivity. Furthermore, the sensitivity (i.e., the clearing dose) of these resist formulations was virtually independent of molecular weight (entries 5 and 6 and 7 in Table 1). In the case of polymer **8**, good resolution could be achieved even with very high

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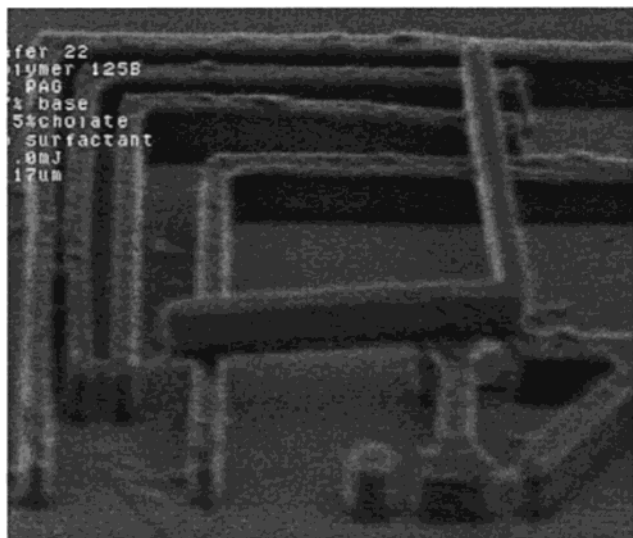


Figure 6. 170-nm 1:1 L/S features obtained by formulations containing polymer **8** after exposure using a 193-nm stepper (4 wt % PAG, 17 mol % 1-morpholineethanol, 7.5 wt % *tert*-butyl cholate). Dose: 11 mJ/cm².

molecular weight material. As can be seen in Figure 6, some lifted features could be found, indicating a loss of adhesion, perhaps the result of poor compatibility with the underlying bottom antireflective coating. Interestingly, given the small size of the lifted feature, the line shape is preserved, probably due to the rigid-rod nature of these cyclopolymeric materials. This property suggests an alternative application of these polymers in the field of nanofabrication of shape-persistent structures.

The contrast between the behavior of polymers **5** and **6** and that of polymers **7** and **8** may be related to the presence of one vs two acid-sensitive functionalities per monomer unit within the respective polymer backbones. We speculated that the more acid cleavable units that are present in a structure, the easier it is to dissolve the deprotected but still largely hydrophobic polymer chains into aqueous base developer, independently of their average chain lengths. Such speculation led us to an optimized design in which a single pendant group is selected to fulfill both the imaging function through its lability to acid and the etch resistance function through its cage structure. It is with these design criteria in mind that we selected the acid-labile 2-methyl-2-adamantyl substituent.¹³

Resist formulations containing polymers **9–12** showed excellent lithographic properties in terms of ultimate resolution, adhesion, and compatibility with aqueous base developers. Introduction of the acid-labile adamantyl substituents further enhanced the sensitivity of the resists, as formulations with as little as 0.5 wt % photoacid generator can be imaged with a radiation dose of approximately 10 mJ/cm². As expected, the lithographic performance was also virtually independent of molecular weight (see Table 1, entries 8 and 9). Formulations of polymers **11** and **12**, containing a small amount of free acid from the comonomer used, showed essentially no thickness loss in the unexposed areas after development. For example, the measured film thickness of the unexposed areas for a sample of polymer **11** was 3485 Å before and 3472 Å after development. Several different formulations with poly-

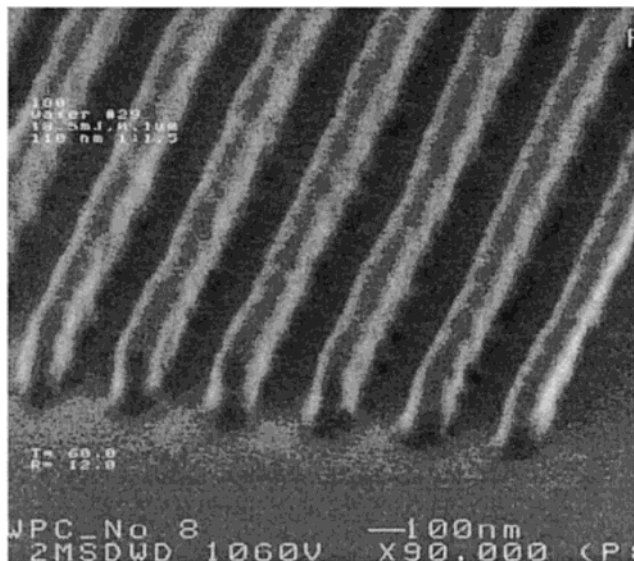


Figure 7. 110-nm 1:1.5 L/S features obtained by formulations containing polymer **11** after exposure to a 193-nm stepper (0.5 wt % PAG, 30 mol % 1-morpholineethanol). Postapplied bake 160 °C, 60 s; postexposure bake 170 °C, 60 s. Dose: 18.5 mJ/cm².

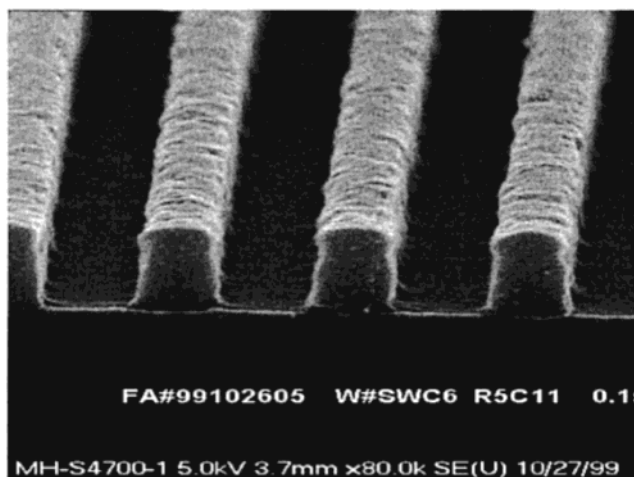


Figure 8. 150-nm 1:1 L/S features obtained by formulations containing polymer **11** after exposure using a 193-nm stepper (1 wt % PAG, 30 mol % 1-morpholineethanol). Postapplied bake 160 °C, 60 s; postexposure bake 170 °C, 60 s. Dose: 7 mJ/cm².

mer **11** were prepared for limited optimization of the lithographic properties through changes in the bake temperatures, the amount and nature of the added base quenchers, the amount of cholate plasticizer, and the amount of PAG. Micrographs obtained during these experiments are shown in Figures 7 and 8. The ultimate resolution achieved using a binary reticle and conventional illumination was 110 nm (1:1.5 L/S), and the features for nested (1:1 L/S) 150-nm lines show good, straight-wall profiles. By using phase shift mask technology, we obtained 90-nm features (1/1.5 L/S, Figure 9).

Reactive Ion Etch Resistance. The ultimate goal of photolithography is to transfer a pattern into the substrate. This is accomplished by exposing a patterned photoresist-coated wafer to a reactive ion etch process, where etching should be restricted to areas that contain no remaining photoresist. Therefore, the photoresist must etch much more slowly than the underlying wafer

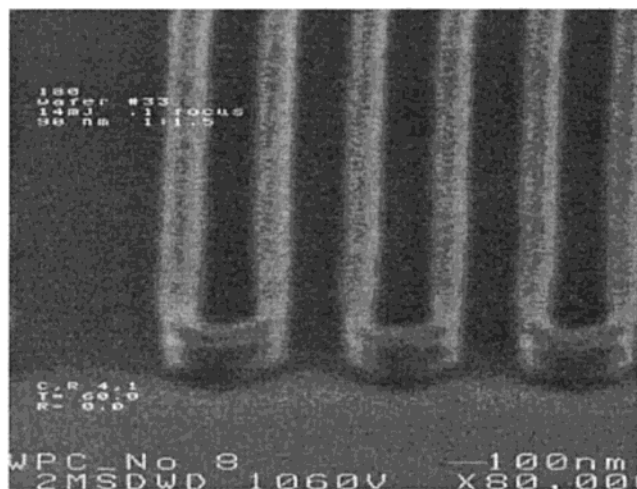


Figure 9. 90-nm 1:1.5 L/S features obtained by formulations containing polymer **11** after exposure using a 193-nm stepper using a phase-shifting mask (1 wt % PAG, 30 mol % 1-piperidineethanol). Postapplied bake 160 °C, 60 s; postexposure bake 170 °C, 60 s. Dose: 14 mJ/cm².

in order to provide good pattern transfer. With 193-nm illumination, this property, termed etch resistance, must be even better than for resists exposed with the more standard 248-nm light. The reason for this increased demand is the empirical requirement that successful imaging requires a feature height-to-width ratio of approximately 4:1. With the smaller features printed via 193-nm light, this would correspond to a film thickness of 0.52 or 0.36 μm for 0.13 and 0.09- μm features, respectively. Because this thickness is significantly below that used routinely in manufacturing today, the etch resistance of these new 193-nm materials should be superior to that of current, chemically amplified, 248-nm photoresists. The etch rate values for several resist formulations are reported in Table 2. These were all measured on the same instrument under the same harsh conditions currently used to etch polysilicon.

As shown in Table 2, the introduction of more adamantyl substituents with their inherently high C/H ratios renders the resist formulation more resistant to the etch process. Two main empirical parameters have been introduced in the specialized literature in an attempt to rationalize and predict etch values for various resist formulations. The first, named the Ohnishi parameter,¹⁴ is defined as

$$\text{ON} = N_{\text{tot}}/(N_{\text{C}} - N_{\text{O}})$$

where N_{tot} , N_{C} , and N_{O} are the total number of atoms, the number of carbon atoms, and the number of oxygen atoms, respectively, within the polymeric units. The second, called the ring parameter,¹⁵ is defined as

$$\text{RP} = M_{\text{CR}}/M_{\text{TOT}}$$

where M_{CR} and M_{TOT} are the cumulative mass of all

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Table 2. Etch Resistance of Various Resist Formulations^a

resist matrix ^a	ON ^b	RP ^b	etch rate (APEX-E = 1)
SPR510L(I-Line)			0.66
APEX-E			1
2	3.14	0.543	0.88
3	2.94	0.652	0.66
4	3.50	0.480	1.22
5	3.34	0.535	0.99
6	3.33	0.524	0.99
8	3.67	0.389	1.30
9	3.31	0.503	0.97
10	3.11	0.597	0.82
11	3.18	0.578	0.85

^a Conditions are reported in the Experimental Section. ^b ON (Ohnishi parameter) and RP (ring parameter) were calculated as described in the text considering the ring size of the corresponding polymeric structure as depicted in Figure 1.

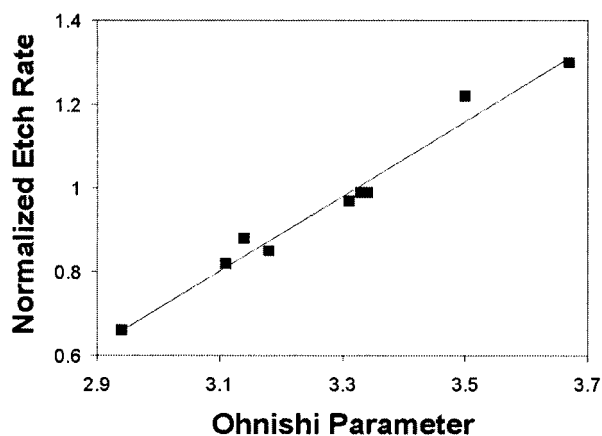


Figure 10. Plot of etch data for various resist formulations vs the Ohnishi parameter.

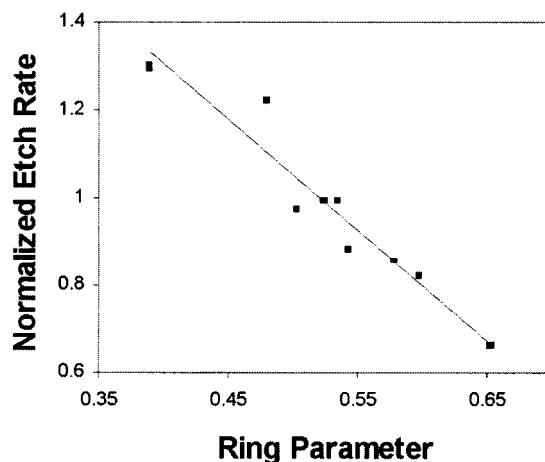


Figure 11. Plot of etch data for various resist formulations vs ring parameter.

carbon atoms contained in a ring structure in the resist and the total mass of the resist, respectively. As shown in Figures 10 and 11, within our structurally homogeneous family of cyclopolymers, a good linear correlation ($r^2 = 0.976$) was obtained between etch resistance and

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the ON parameter, whereas a slightly less satisfactory correlation was found using the RP parameter ($r^2 = 0.919$). It should therefore be possible to use these two parameters to predict with a high degree of confidence etch data for novel but structurally related cyclopolymers bearing substituents other than those we have utilized.

It should be mentioned that the data obtained for polymer **11** compare favorably with those for the best materials described to date in the literature using the same etch conditions.

3. Conclusions

We have evaluated the lithographic behavior of a series of novel cyclopolymers as resist platforms for 193-nm microlithography. Careful analysis of their lithographic performance as it relates to their chemical structure enables a correlation of lithographic properties with various polymer characteristics including structure, substituents, and molecular weight. On the basis of these data, we have designed optimized systems that feature 2-methyl-2-adamantyl substituents acting both as acid-cleavable and etch resistant groups. These systems have shown excellent promise in terms of ultimate image resolution and etch resistance. It is clear that the cyclopolymerization route we have developed for the preparation of 193-nm resists allows the rapid preparation of a variety of related structures, enabling optimization within a family of structurally related polymers.

4. Experimental Section

General. Unless stated otherwise, chemicals including solvents were purchased from Aldrich. AR-19 antireflective coating and LDD-26W developer, based on 0.26 N aqueous tetramethylammonium hydroxide (TMAH), were obtained from Shipley. The DUV-30 antireflective coating was provided by Brewer Scientific, while *tert*-butyl cholate was prepared according to a literature procedure.¹² The preparation of all of the polymers used in this study is described in the preceding report.⁷

Triphenylsulfonium Nonfluorobutanesulfonate. This compound was prepared by a double counterion exchange methodology. A solution of triphenylsulfonium hexafluoroantimonate⁹ (3 g, 6 mmol) in EtOAc (20 mL) was mixed with a solution of tetrabutylammonium chloride (Aldrich, 2.8 g, 10 mmol) in EtOAc (50 mL). A white precipitate, triphenylsulfonium chloride, immediately formed and was filtered and recrystallized (CH₂Cl₂/EtOAc). A solution of triphenylsulfonium chloride (1.6 g, 5.4 mmol) in CH₂Cl₂ (50 mL) was washed twice with a solution of potassium nonfluorobutanesulfonate (4 g, 12 mmol) in H₂O (100 mL). The organic layer was dried (MgSO₄), the solvent was removed in vacuo, and several portions of Et₂O were added to the resulting clear oil and then

removed in vacuo in order to azeotropically eliminate residual solvents. Further drying under ultrahigh vacuum for several days afforded the nonfluorobutane organic salt as a waxy, white solid (1.8 g, 54%). ¹H NMR (CDCl₃) $\delta = 7.9-7.6$ (m, 15H). Anal. Calcd for C₂₂H₁₅O₃F₉S₂: C, 47.02; H, 2.69; F, 30.43; S, 11.41. Found: C, 47.20; H, 2.65; F, 30.51; S, 11.16.

Photoresist Formulation and Wafer Coating. Photoresist formulations were prepared by dissolution of the cyclopolymer (10–20 wt % vs solvent), photoacid generator (PAG, 0–10 wt % vs polymer), base (1-morpholine ethanol or 1-piperidine ethanol, 0–30 mol % vs PAG), and *tert*-butyl cholate (0–15 wt % vs polymer) in either propylene glycol methyl acetate (PGMEA) or cyclohexanone. Solutions were passed through a 0.40 μ m Teflon filter and spin-coated at 2000–3500 rpm to afford a 3000–4500 Å thick resist film on either bare silicon wafers (for variable angle spectroscopic ellipsometer measurements only) or on wafers precoated with an approximately 800 Å thick bottom antireflective coating layer (BARC, AR-19, or DUV-30). The wafers were then baked at 140–170 °C for 60 s before use in subsequent imaging experiments.

Ultraviolet/Visible Spectroscopy. The absorbance spectra of photoresist formulations on bare silicon wafers were measured from 190 nm upward using a variable angle spectroscopic ellipsometer (VASE) from J. A. Woollam Co.

Lithographic Imaging Assessment. Photoresist-coated wafers with an underlying BARC layer were exposed to 193-nm ArF laser light on an ISI MicroStep stepper with 0.6 numerical aperture (NA) lens. Partial coherence of 0.7 was employed with binary masks and 0.3 with phase-shifting masks. After exposure, wafers were baked at 150–175 °C for 60 s before development with one 60 s puddle of LDD-26W developer, followed by a deionized water rinse. Scanning electron micrographs of whole imaged wafers were recorded on a JEOL 7550 SEM and on a Hitachi S4700 SEM for cleaved wafers.

Etch Rate Determination. The etch rate of photoresists on imaged wafers was measured on a Lam TCP9400 plasma etcher under reactive ion etch conditions with a Cl₂/HBr plasma. Flow rates of Cl₂ and HBr were 50 and 150 sccm, respectively. The film thickness both before and after etching was measured with a Prometrix SM-200 film thickness probe and directly compared to that of Shipley APEX-E, a commercial 248-nm photoresist, run at the same time under the same conditions. The data reported in Table 1 are normalized versus the thickness loss measured for APEX-E.

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