Multilayer Resist Systems for Lithography

M. Hatzakis

IBM Thomas J. Watson Research Center
Yorktown Heights, New York

Multilayer resist systems offer the possibility of increasing the resolution of lithography tools by the fact that only a thin (2000–3000 Å) layer is used as the pattern defining resist. Step coverage and topography variations in the workpiece are planarized by use of a thick bottom layer that can be a resist or any polymer selected for processing qualities. A review of multilayer lithographic systems is presented with emphasis on performance and advantages over single layer resists. Some examples of such systems published in the period from 1976 to the present are discussed.

Introduction

The advantages of reducing the device dimension in logic and memory VLSI circuits have been shown in numerous papers since the late 60s. These include improvements in performance as well as cost reduction, provided that the wafer throughput of new, high resolution lithography tools can be kept high. Also, in order to realize the speed improvements promised by a decrease in the device active area, conductor interconnection resistance, among other things, must be kept low. This implies that polysilicon or metal conductor thickness does not scale with decreasing lateral dimensions and therefore the aspect ratio (height-to-width) of patterns exposed and developed in the resist increases as lateral dimensions decrease. For example, in logic circuits, as the width of the first metal interconnection pattern approaches one micrometer, the metal thickness must also remain close to one micrometer in order to keep the current density within the conductor at a reasonable level, and avoid electromigration effects.

Lithographic definition of such structures is accomplished either by subtractive plasma etching techniques or by lift-off through a suitable resist. In the case of lift-off, the resist thickness must be higher than the metal thickness to ensure that in the first case, linewidth control is maintained and in the second case, successful lift-off can be accomplished. Furthermore, topography generated on the workpiece in many processes, such as double polysilicon in MOS technology, can be as high as 0.8 μm. This means that if the resist thickness required to define the metal pattern is say 1.2 μm over the 0.8 μm high topography then the total resist thickness in the low areas (valleys) can be as high as 2 μm. In order to develop a resist pattern with such a high aspect ratio it is essential that the volume of resist in the exposed area receives a uniform dose and that all areas away from the exposed pattern receive doses that are much smaller than any resist element within the pattern, regardless of the form of radiation used in the exposure tool. The preceding statement can also be expressed in terms of the exposing tool *contrast* which can be defined as the ratio of the exposure dose within the pattern to the exposure dose outside of the pattern. If the tool contrast is low and the resist thickness high, then excessive widening of the exposed pattern occurs at the top of a positive resist due to the long development time required to completely clear the exposed resist areas.

In optical (UV) tools, loss of contrast has been the direct result of efforts to reduce the minimum linewidth that can be printed to dimensions approaching one micrometer. In electron beam tools, similar contrast loss is caused by forward electron scattering in thick resist and also electron backscattering from the substrate. In x-ray tools, contrast is limited by the thickness of the opaque material (usually gold) that can be achieved with current x-ray mask technology.

If the resist thickness is kept low then development times can be short and line widening due to low tool contrast can be controlled easily. In addition, radiation scattering in the resist is reduced, especially forward electron scatterning for e-beam tools.

This realization, that improved resolution can be obtained with thin resist layers and that thick resists are essential to the device fabrication process, led to the development of multiple resist layer systems that provide some additional advantages not covered previously, as will be seen in later sections.

In general, multiple layer schemes can be divided into two main categories; those in which at least two layers are used as resists and both are exposed and developed, and those in which only one layer, usually the topmost, is used as the imaging resist and all other layers are selectively removed using the top layer as a protective mask. The two systems will be covered separately because in the former, the radiation sensitivity of at least two layers is important while in the latter system the etching medium properties are more important.

Multiple, Radiation Sensitive, Resist Systems

Early attempts to form double resist layers were based on the observation that the solubility of poly(methylmethacrylate), PMMA electron resist, after exposure depends on, among other parameters, the initial molecular weight of the polymer, especially at low exposure charge densities [1]. Utilizing this fact, W. Moreau and C. H. Ting [2] proposed that a double layer can be formed by spinning low molecular weight PMMA first to the desired thickness, baking at 160 °C and subsequently applying a second layer of high molecular weight PMMA. After electron beam exposure of the double layer at a dose of approximately 2×10^{-5} C/cm², which is one fifth of the dose normally required to produce vertical or undercut profiles in a single PMMA layer, the resist is developed in a reasonably strong developer like methylisobutylketone (MIBK). Due to the low solubility of the top layer, development proceeds very slowly in the exposed regions, until the interface between the two layers is reached. At this point the development rate increases rapidly due to the high solubility of the bottom layer, with the result that vertical or even undercut resist profiles can be obtained at this low electron dose. The technique was also investigated by Horwitz [3]. The solubility difference obtained by spinning two resist layers of different molecular weight can also be simulated in a single resist layer by surface treatment in a way that results in a drastic reduction in solubility down to a predetermined depth [4]. In this case AZ1350-J photoresist is treated after prebaking by immersion in an aromatic solvent such as chlorobenzene for a fixed time before or after pattern exposure and before development. The depth of the treated layer depends largely on the soak time and the prebake temperature. Since the treated layer develops more slowly than the unreated layer, the developed resist profiles have the appearance of two distinct layers, as shown in Fig. 1. The technique can be used with other diazo type resist systems and with UV, electron beam or x-ray exposure. So far it has been applied to the lift-off metallization technique.

Although undercut resist profiles can be obtained with the double PMMA layer system at exposure charge densities four to five times lower than with a single layer, the top resist opening continues to develop during development of the bottom layer with the result that unacceptable pattern bias occurs in many cases, especially if the bottom layer is considerably thicker than the top layer. The same is true for single layers modified by soaking and developed in the same developer. This problem can be avoided if two dissimilar resists are used which are developed with mutually exclusive solvents [5, 6]. In this case one of the PMMA layers is substituted by a copolymer of methyl methacrylate and methacrylic acid [7, 8] which even after exposure is insoluble in non-polar solvents such as toluene and chlorobenzene while PMMA developes quite rapidly in these solvents. Alternatively the copolymer can be developed in polar solvents such as methanol or ethoxyethanol in which PMMA is completely insoluble. When PMMA is used as the thick bottom layer [5], the top copolymer layer is developed in ethoxyethanol to the correct pattern size after exposure. The developer is then changed to toluene or chlorobenzene and the thick PMMA layer is developed without any further change in the pattern dimensions on the top layer. Furthermore since the thick PMMA layer planarizes the substrate, topographic variations do not affect the top thin copolymer layer, as indicated in Fig. 2. Actual scanning electron micrographs of the resist system used for metal lift-off are shown in Fig. 3.

In the previous two systems the primary lithographic tool was used to expose both resist layers simultaneously. If the sentivity of the bottom layer is much lower than that of the

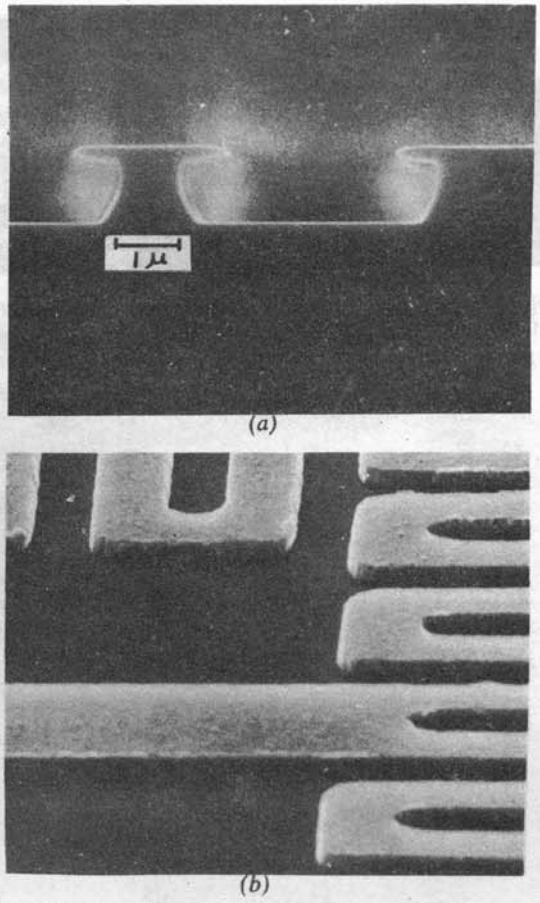


Fig. 1—(a) SEM profiles of AZ1350-J photoresist UV exposed, modified to a depth of 2000 Å by soaking in chlorobenzene and developed in AZ developer; (b) SEM photo of metal pattern obtained by use of the lift-off technique with chlorobenzene soaked AZ1350-J.

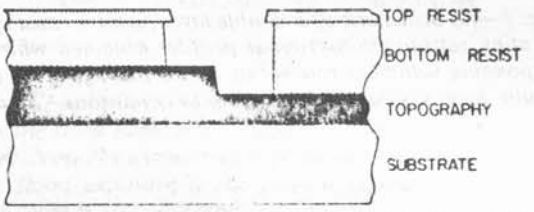
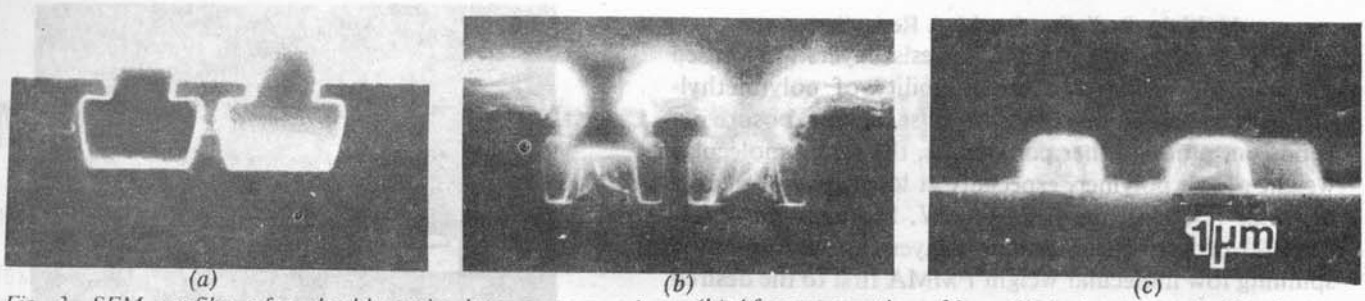


Fig. 2—Schematic of a double layer resist system indicating planarization of the bottom layer over surface topography.



(a) (b) (c)

Fig. 3—SEM profiles of a double resist layer system using PMMA, 1.4 μm thick as the bottom layer and P(MMA-co-MAA) copolymer as the top layer. (a) Resist profiles after development; (b) After evaporation of 1 μm thick aluminum; (c) Metal pattern after lift-off.

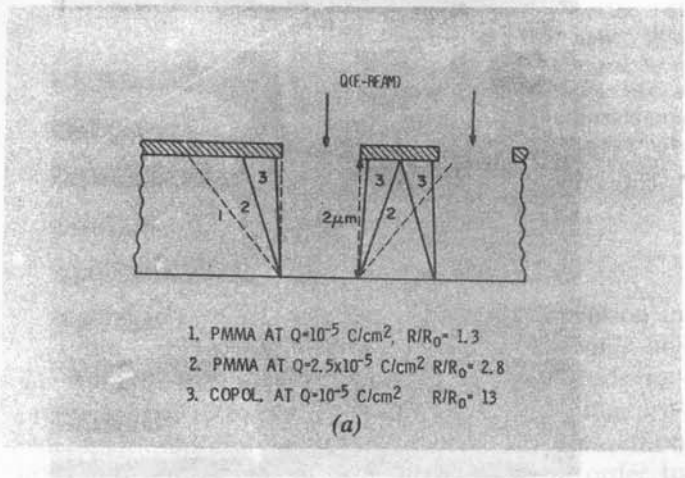


Fig. 4—(a) Schematic of a double layer positive resist system indicating various bottom resist profiles obtained with the corresponding solubility rate ratios R; (b) SEM profiles of actual double layer system corresponding to conditions "2" and "3".

top layer, then development of the bottom layer can be nearly isotropic, that is, vertical and horizontal development proceed at nearly the same rate. This means that narrow resist spaces between two exposed regions get severely undercut with the result that if the bottom resist layer is very thick such narrow spaces can be lost through excessive undercutting.

The limitations of a double layer positive resist system are shown in Fig. 4. It is assumed in the top sketch that the resist outside the main exposure is completely unexposed and that the solubility rate ratio of PMMA in toluene ($R = S_1/S_0$, where S_1 is the solubility of the exposed resist and S_0 is the solubility of the unexposed resist) equals 1.3 for a 10^{-5} C/cm² exposure and 2.8 for 2.5×10^{-5} C/cm² exposure. For copolymer as the thick bottom layer exposed at 10^{-5} C/cm² and developed in ethoxyethanol, the solubility rate ratio is 13. The three lines labeled 1, 2 and 3 represent the bottom resist profiles after development where 1 and 2 represent PMMA as the bottom layer exposed at 10^{-5} and 2.5×10^{-5} C/cm², respectively, and 3 is for copolymer as the bottom layer, exposed at 10^{-5} C/cm². In this simple approach it is assumed that sideways (horizontal) development, under the top layer, proceeds at a rate slower than vertical development by a factor equal to the solubility rate ratio. The conclusion is that in order to get vertical profiles in the bottom resist layer a value of R greater than ten must be obtained, especially if this layer is relatively thick (~2 μm). The problem can be avoided completely if a secondary exposure source is used to reexpose the bottom layer using the top layer as an in-situ mask. This approach has been developed by Lin [9] who used a thick PMMA layer as the bottom resist and a thin (~2000 Å) AZ1350-J layer as the top imaging resist. This layer can be exposed to UV or electron beam radiation and developed to the desired lithographic dimensions after which the thick PMMA layer is reexposed to deep UV light (200–220 nm) where AZ1350-J is almost completely opaque and PMMA is moderately sensitive. However, since this exposure can be done in a flood system, many wafers can be exposed simultaneously; therefore throughput is not seriously impacted. At this UV wavelength, scattering and interference effects are reduced and vertical profiles can be easily obtained. Figure 5 shows profiles of 1.9 μm thick PMMA obtained with the Portable Conformable Mask (PCM) system.

One of the main disadvantages of this system is that during the application of the AZ1350-J layer a thin layer of PMMA resist is re-dissolved and mixed with the AZ so that a thin interface layer is formed which is insoluble in both the AZ and PMMA developers. This layer has to be removed by dry plasma etching after the AZ development. Furthermore, the UV light used in the flood exposure has to be carefully filtered to eliminate wavelengths between 240–260 nm where the AZ resist has a transmission window. The filtering causes considerable loss of intensity in the desired shorter wavelengths. These concerns can be eliminated if a thin third layer is introduced between the two main resist layers. Aluminum, 1000 Å thick, has been used by Lin and Chang [9], although any other material completely opaque to UV can be used.

A similar scheme using X-rays to expose the bottom PMMA layer has been reported recently [10]. In this case a heavy metal such as gold must be used at the interface between the two resists and patterned by etching through

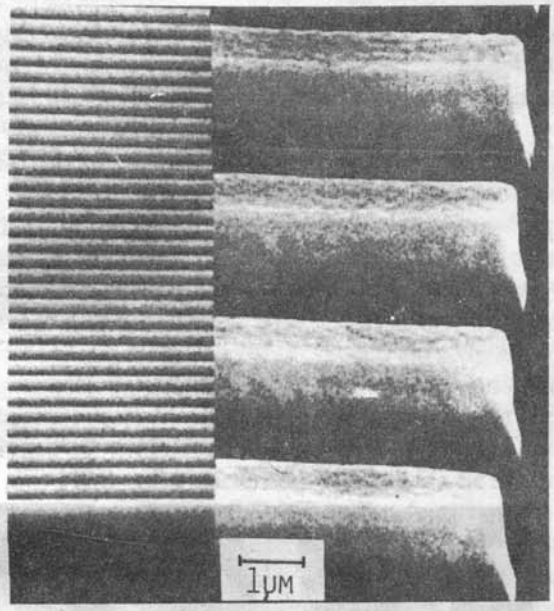


Fig. 5—SEM photos of 1.9 μm thick PMMA UV exposed through a 2000 Å thick AZ1350-J resist using the PCM (portable conformal mask) concept.

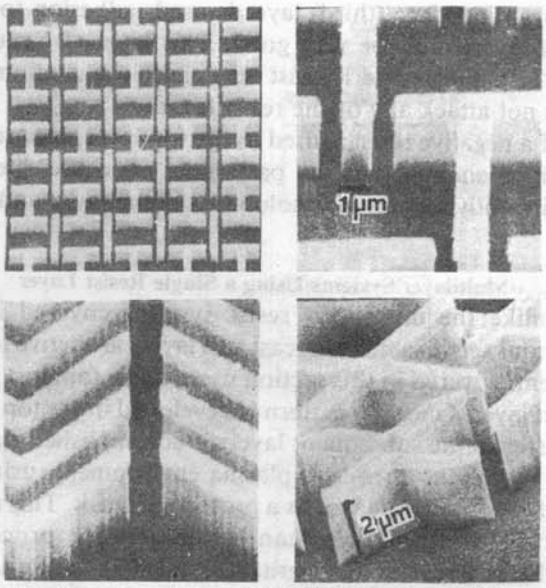


Fig. 6—SEM photos of 2 μm thick PMMA exposed through an in-situ gold x-ray mask fabricated over PMMA by e-beam exposure of a layer of AZ2400, followed by lift-off. Gold mask still in place.

the top electron-beam-exposed imaging layer or deposited by lift-off or electroplating again through the top imaging layer. Figure 6 shows an example of the resist profiles obtained with the x-ray exposure technique. Gold, 2500 Å thick, patterned with the lift-off technique was used as an in-situ x-ray mask for these experiments. As in the case of UV flood exposure, x-ray exposure dose must be sufficient to produce a solubility rate ratio greater than ten in order to obtain nearly vertical wall profiles in the PMMA.

Negative resists can also be used as imaging layers in multiple resist layer schemes. However if the bottom resist layer is also negative, the exposure dose during the first patterning step must be high enough to completely crosslink the total volume of resist under the top resist layer. With too low an exposure dose, the top layer will be removed during development because, at least with electron beam exposure, crosslinking is heavier at the resist-substrate interface and some soluble portion would exist at the resist-resist interface at lower doses.

If a positive resist is used as the bottom layer, then a second exposure is necessary to expose the bottom layer after development of the top layer. Since exposure of the top layer at least partially exposes the bottom layer also, especially with electron beam or x-ray exposure where penetration exceeds the thickness of both layers, then the flood exposure dose of the bottom layer must be sufficiently high to ensure a solubility rate ratio greater than ten. This is illustrated in Fig. 7 where the bottom layer is either a copolymer, P(PMMA-co-MAA), or PMMA. As can be seen, to avoid the excessively high solubility rates required for the bottom layer a much more sensitive negative resist must be used.

Some of the general conclusions regarding multilayer resist systems in which at least two layers are radiation sensitive, can be summarized as follows:

1. The top imaging layer should be both thick and sensitive enough so that sufficient thickness remains after development to protect the underlayer and minimize pinhole formation. (2000–4000 Å)
2. The bottom layer should be sensitive enough so that at the exposure dose used, a minimum solubility rate ratio of ten is obtained for maximum resolution and near vertical resist profiles.
3. Baking temperatures for the top resist layer must be equal to or lower than the maximum allowable temperature for the bottom layer.
4. It is preferable that the developer for the top layer also develops the bottom layer at some finite but very small rate. This will insure that no insoluble interface layer between the two resists can be formed.

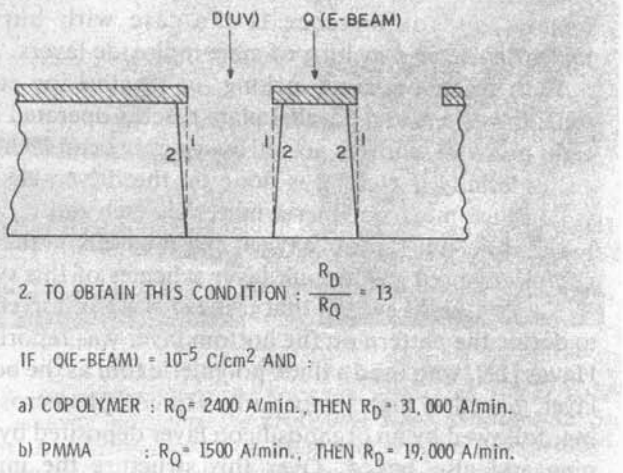


Fig. 7—Schematic of a double resist layer system using a positive resist as the thick bottom layer and a thin negative resist as the top layer. Note the excessively high solubility rates required for the UV flood exposure if the e-beam exposure dose for the negative resist is 10^{-5} coul/cm^2.

5. If an interface (third) layer is used, adhesion to both resist layers must be very good. This layer also must be free of pinholes and it must be etched in a medium that does not attack any of the resist layers.
6. If a negative resist is used as the top layer, it should be sensitive enough that the patterning exposure does not significantly increase the solubility of the layer under it.

Multilayer Systems Using a Single Resist Layer

Unlike the multilayer resist systems covered in the previous section where at least two layers are active resists, systems covered in this section use a single topmost active resist layer. Once the pattern is developed in the top layer, transfer to the subsequent layers is accomplished by etching, usually in a reactive plasma environment, using the top first or second layer as a protecting mask. This means that the bottom layer can be chosen for processing qualities such as temperature stability, adhesion to substrate, low plasma etch rate, etc., since it does not have to be an active resist. Wet chemical etching of the bottom layer is of course unacceptable because such etching is almost always isotropic and will therefore result in resist profiles that form an angle of 45 degrees with the surface normal, as for instance is the case with buffered hydrofluoric acid etching of silicon dioxide layers.

With reactive plasma etching, or reactive ion etching (RIE) however, in a parallel plate reactor operated at the right pressure, entirely anisotropic etching can be achieved, especially if etching is done on the driven electrode [11]. Since most polymeric materials etch quite rapidly (~1000 Å/min) in an oxygen plasma, this is the most commonly used gas in multilayer schemes of this type.

The first resist system that utilized reactive ion etching to define the pattern on the bottom layer was reported by Havas [12], who used a thick polymeric film as the bottom layer, baked at high temperatures to complete crosslinking, followed by an organosilicon layer deposited by spinning and also baked. Over this structure the imaging photoresist layer was applied, exposed and developed. At this point the sample was introduced into a reactive ion etching chamber and the image transferred to the organosilicon layer by a CF_4 plasma using the imaging layer as the protective mask. Subsequently the gas was changed to O_2 and the bottom layer etched using the organosilicon film as the mask. At the same time the top resist layer was also removed. Since the etch rate of organosilicon films is very nearly zero, a very thin layer (1000–2000 Å) of this film is sufficient protection when a two micrometer thick bottom layer is etched. In this application the bottom layer was overetched so that undercut profiles suitable for lift-off were obtained [13].

A similar process has been also reported by Moran and Maydan [14] who used plasma deposited silicon dioxide as the interface layer and a thick (2–3 μm) Hunt HPR-204 photoresist layer baked to 200 °C to thermally stabilize it. The top imaging layer, in this case, was a negative x-ray resist which after exposure and development was used as a mask for etching the SiO_2 layer in CHF_3 plasma. Again the pattern transfer to the thick bottom layer was accomplished via reactive ion etching in oxygen. Figure 8a shows the processing sequence and 8b the resulting profiles in the bottom thick layer.

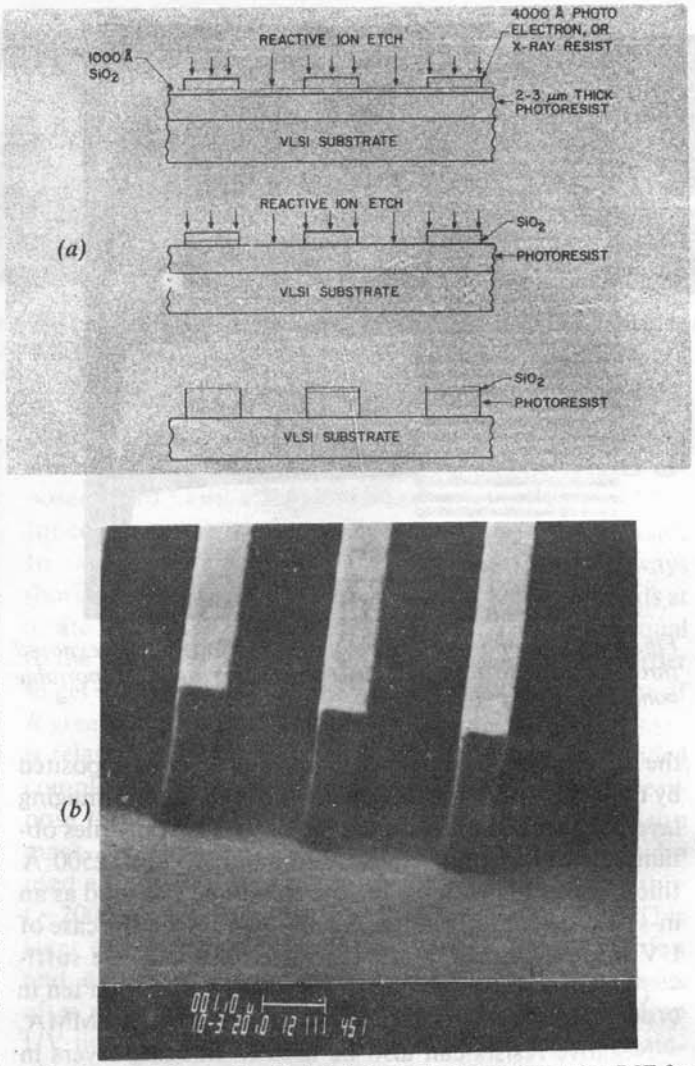


Fig. 8—(a) Schematic of a triple layer resist system using RIE for pattern transfer to the bottom layer and SiO_2 as the thin interface layer; (b) SEM profiles of high aspect ratio lines after plasma etching of the bottom layer (courtesy J. Moran, Bell Laboratories).

Many variations of the two examples of multilayer systems involving organometallic or inorganic layers and oxygen plasma have been reported but will not be covered in detail here due to lack of space. Most of these involve the use of a single top layer for pattern definition which is developed with the conventional wet process while all subsequent layers are plasma etched. Recent work on plasma developed resists shows that it is possible to develop some negative resists with a heat treatment followed by plasma etching in oxygen gas [15, 16]. If such resists are used as top imaging layers, then a completely dry-developed multilayer system becomes a reality. Such a system will avoid handling and disposal of solvents except for resist application on the workpiece, which is done by spinning from a resist solution.

Another important development that can further simplify multiple resist systems of the type discussed in

this section, concerns recent work with certain inorganic resists. These resists are, in general, vacuum deposited by evaporation and consist of thin layers of compounds such as As_2S_3 or multiple layers of $AgCl/As_2S_3$ [17] or $Ag_2Se/GeSe$ [18] that are radiation sensitive to UV, electron beams or X-rays and can be developed in solutions or by dry plasma etching. Since these compounds are inorganic and radiation sensitive, they can be patterned directly on a thick polymeric film without the need for an intermediate protecting layer. After development, the thick polymer film can be patterned directly by oxygen plasma etching without any loss of the inorganic patterning layer. Figure 9a illustrates the patterning process for reference 17 while Fig. 9b shows SEM profiles of thick (2.5 μm) HPR resist patterned with the Ge-Se system and oxygen plasma etching according to reference 18.

While most of the concerns listed at the end of the previous section also apply in this case, the main difference here is in tranferring the pattern to the bottom thick layer by oxygen plasma etching. Care therefore should be taken to ensure that:

1. The plasma etching conditions such as pressure, power, gas flow rate and substrate bias, should be chosen properly so that etching takes place only in a direction perpendicular to the substrate surface (anisotropic).

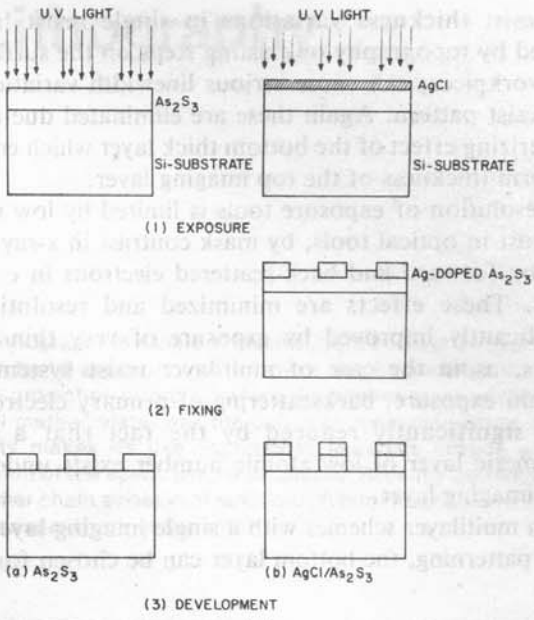
2. Since the film used as the mask for plasma etching has to be very thin to obtain high pattern resolution, it should not etch in oxygen plasma and should be coated by a process that results in a pinhole free film.

Some of the resist systems covered here are listed in Table I, for easy reference. Use of AZ1350-J or Hunt HPR 204 or any quinone-diazide type photoresist as the bottom thick layer is dictated by two main factors. First, these resists can be stabilized against flow during high temperature processing of the finished resist pattern by prebaking at temperatures between 160–220 °C. Second, these resists exhibit low RIE rates in CF_4 or other gases used for silicon or SiO_2 etching, as compared to PMMA or other vinyl copolymers. It should be remembered however, that after baking at over 160 °C, quinone diazide type resists crosslink and become insoluble in organic solvents and cannot be removed by conventional organic-based strippers.

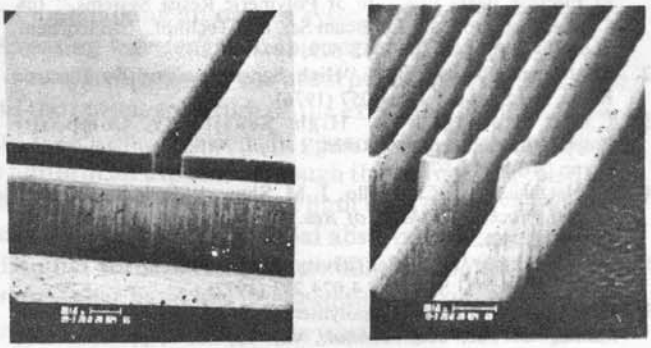
Conclusions

Multilayer resist systems appear to be gaining popularity as the need for smaller lithographic dimensions and higher aspect ratios of resist patterns is constantly increasing. The advantages of such systems over single layer resists can be summarized as follows:

1. In photolithography with step-and-repeat or one-to-one imaging systems operating at a single UV wavelength, standing waves in the resist pattern created by reflection from the substrate are increasingly causing linewidth control problems as pattern dimensions approach one micrometer. These problems are eliminated with multilayer systems where only the top thin layer is exposed while the bottom thick layer can serve as an antireflection coating.



(a)



(b)

Fig. 9—(a) Schematic of the inorganic As_2S_3 and $AgCl/As_2S_3$ resist systems indicating the processing steps (courtesy M. S. Chang, Hewlett-Packard); (b) SEM photos of the inorganic Se-Ge resist system UV exposed and developed and used as an in-situ RIE mask for plasma etching of a thick Hunt 204 resist layer. The inorganic layer is still visible over the thick bottom layers (courtesy K. L. Tai, Bell Laboratories).

Table I—Resist System Characteristics

Resist System	Exposure Tool	Application	Reference #
PMMA HIGH Mw PMMA LOW Mw	E-Beam	Lift-off Process	2, 3
P[MMA-co-MAA] PMMA	E-Beam	Lift-off Process	5, 6
Chlorobenzene soaked AZ1350-J	UV	Lift-off Process	4
AZ1350-J PMMA	UV/E-Beam	Lift-off or RIE	9
AZ1350-J or PMMA siloxane AZ1350-J	UV/E-Beam	High Temperature Lift-off	12
Hunt 204 SiO_2 Hunt 204	UV	RIE	14
$Ag_2Se/GeSe$ Hunt 204	UV	RIE	18

2. Resist thickness variations in single resist layers, caused by topography or existing steps on the surface of the workpiece, can cause serious linewidth variations in the resist pattern. Again these are eliminated due to the planarizing effect of the bottom thick layer which ensures uniform thickness of the top imaging layer.
3. Resolution of exposure tools is limited by low image contrast in optical tools, by mask contrast in x-ray tools and by forward and back-scattered electrons in e-beam tools. These effects are minimized and resolution is significantly improved by exposure of very thin resist layers, as in the case of multilayer resist systems. In e-beam exposure, backscattering of primary electrons is also significantly reduced by the fact that a thick polymeric layer of low atomic number exists under the thin imaging layer.
4. In multilayer schemes with a single imaging layer and RIE patterning, the bottom layer can be chosen for processing qualities alone since it does not have to perform as a resist, and it can be thick enough to accommodate the next processing step, without loss of pattern resolution.

Some of the disadvantages include:

1. The extra processing steps required to coat, bake and develop more than one resist layer.
2. The need for vacuum equipment on the line for evaporation of some of the imaging or interface materials.
3. The need for reactive ion etching equipment which can be very costly, especially if a large number of wafers have to be loaded in one etching cycle.

Even with these disadvantages, however, multilayer resist systems offer perhaps the only solution to micrometer or submicrometer lithography that will meet the device designer's requirements for topography coverage, pattern resolution and linewidth control.

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Mike Hatzakis received his B.S. in Electrical Engineering in 1964 and M.S. in Electrical Engineering in 1967 from New York University. He is the Manager of the Lithography Processes Group at IBM Thomas J. Watson Research Center in Yorktown Heights, New York. His responsibilities include the development of circuit and device fabrication. He has been very active since the early 60s in the field of electron beam microfabrication, including the development of new electron resists and submicrometer size devices. Mr. Hatzakis is a member of Tau Beta Pi and Eta Kappa Nu.