

Multilayer resist methods for nanoimprint lithography on nonflat surfaces

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Five multilayer resist methods (three positive tones and two negative tones) have been devised for nanoimprint lithography on nonflat surfaces. Three of the methods have been demonstrated experimentally on a SiO₂ surface with 100 nm deep sharp steps. The advantages and disadvantages of each method are discussed. Our results should be applicable to nanoimprint lithography with 10 nm feature size on nonflat surfaces. © 1998 American Vacuum Society. [S0734-211X(98)10106-3]

I. INTRODUCTION

Nanoimprint lithography (NIL), a new approach to nanolithography, patterns a resist by physically deforming the resist's shape with a mold, instead of modifying the resist's chemical properties with radiation as in conventional lithography.¹ NIL has demonstrated 10 nm feature size on a flat surface and the potential for low-cost and high throughput.² One key issue for NIL to become a major lithography tool is to imprint on nonflat surfaces. This article presents five multilayer resist methods for solving this problem.

II. FIVE METHODS FOR NIL ON NONFLAT SURFACES

We have devised five multilayer resist methods for NIL on nonflat surfaces (Fig. 1). The five methods start with the same first step: surface planarization, where a resist thicker than the height of the steps on a wafer surface is cast on the wafer. This resist, called the planarization layer, can be planarized by (a) a free viscous flow of the resist at an elevated baking temperature or (b) a forced resist flow through pressing a flat mold on the resist surface while heating. The samples for our experiments have 100 nm deep sharp steps of SiO₂ on Si. Poly(methylmethacrylate) (PMMA) and modified novolak resin 250 or 300 nm thick were used as the planarization layer. The free viscous flow approach works well for PMMA, when heated up to 175 °C for 24 h, but not for the novolak resin because it is a thermal-set plastic and has a high viscosity. The forced flow approach works well for both PMMA and novolak resin, when heated at 175 °C for 20 min, with a pressure on the flat mold of 600 psi. After removing the mold, the resist over the sharp steps was planarized.

A. Trilayer resist scheme

The first method is a trilayer resist scheme that has a positive tone [Fig. 1(A)]. The bottom layer is a 250 nm thick 15 K molecular weight PMMA as the planarization layer. The middle layer is a 30 nm thick SiO₂, for the purpose of transferring patterns. The top layer is a 250 nm thick 2 k molecular weight polystyrene, which is the patterning layer

for NIL. One key point is that the glass transition temperature (T_g) of 2 k polystyrene is at about 0 °C,³ which is much lower than the T_g of the PMMA (93 °C). Therefore the top layer polystyrene can be imprinted at 90 °C without deforming the bottom layer PMMA. The process began with an imprint to pattern the top layer at a temperature of 90 °C and a pressure of 600 psi. Then the pattern in the top layer was transferred to the middle layer SiO₂ by reactive ion etching (RIE), using CHF₃ with a gas flow rate of 15 sccm, a pressure of 5 mTorr, a power of 150 W, and an etching rate of 6 nm/min. A second RIE transferred the SiO₂ pattern into PMMA using O₂ with a gas flow rate of 6 sccm, a pressure of 3 mTorr, a power of 200 W, and an etching rate of 60 nm/min. SiO₂ has an extremely low etching rate in oxygen RIE, serving as a good etching mask for PMMA.

Figure 2 shows rectangular holes in the middle layer after processing with the trilayer resist scheme. The bottom layer is invisible in the scanning electron microscope (SEM) image. Figure 3 shows a resist pattern on a 100 nm SiO₂ sharp step after processing. The resist pattern consists of a 250 nm thick PMMA layer (bottom layer), and a 30 nm thick SiO₂ layer (middle layer). As shown, the top surface of the resist at the 100 nm step is flat, the side wall is straight, and the lateral dimension of the resist pattern is unaffected by the step.

B. Imprint and lift-off scheme

The second method is an imprint and lift-off scheme that has a negative tone [Fig. 1(B)] that uses two resist layers. The bottom layer is a modified novolak resin (Brewer Science XHRi 16)⁴ previously used as antireflection coating (ARC). It is a thermal-set polymer so that it hardens after a bake. It will not dissolve in acetone, therefore it can survive a lift-off process. The top layer is 15 k molecular weight PMMA. Since the T_g of the modified novolak resin is much higher than that of the PMMA, the imprint on the top layer will not deform the bottom layer. The process started with imprinting a pattern into the top layer with a temperature of 175 °C and a pressure of 600 psi, then using lift-off to transfer it to a metal, which served as a mask for the subsequent oxygen RIE pattern transfer to the planarization layer. The metal used in this scheme is Cr, which is a good etching mask. The etching rate for the novolak resin is 30 nm/min.

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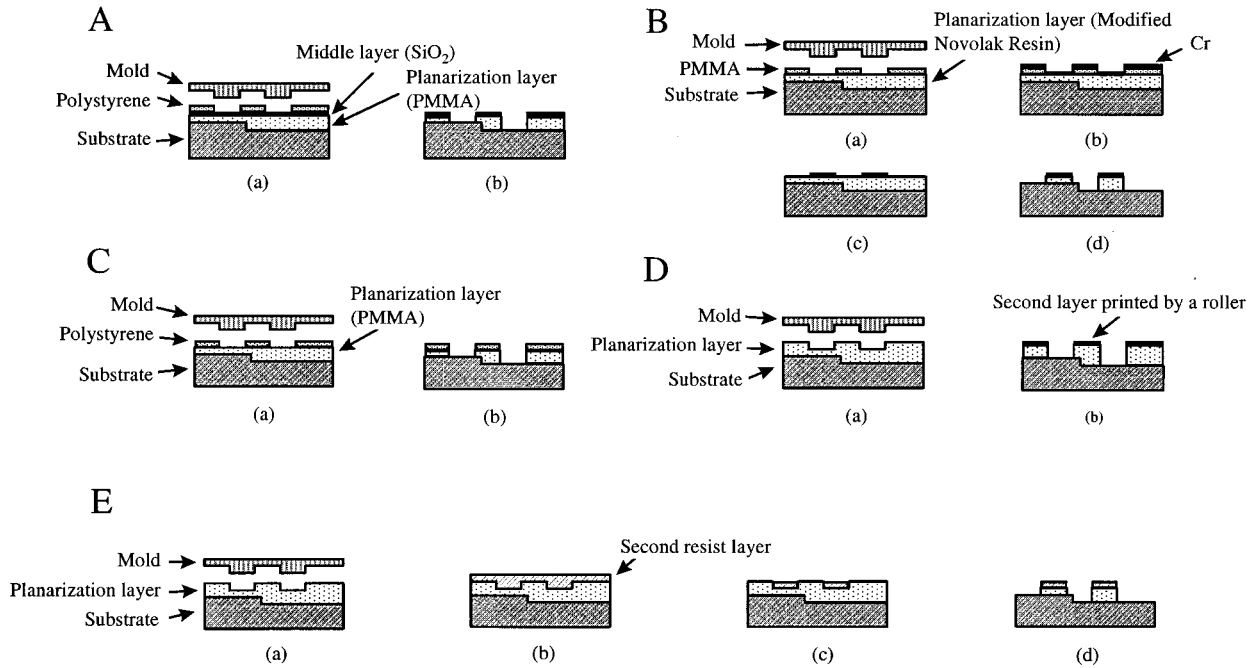


FIG. 1. (A) Trilayer resist scheme (positive tone); (B) Imprint and lift-off scheme (negative tone); (C) Imprint and RIE scheme (positive tone); (D) Imprint and print scheme (positive tone); (E) Imprint and etch back scheme (negative tone).

Figure 4 shows a resist pattern on a 100 nm SiO₂ sharp step after processing with the imprint and lift-off scheme. The resist pattern is a layer of 300 nm modified novolak resin. Again the surface of the resist at the 100 nm step is smooth, the side wall is vertical, and the lateral dimension of the resist profile is not affected by the step.

Figures 5(a) and 5(b) show a 190 nm period resist grating on a 190 nm period SiO₂ grating after processing with the imprint and lift-off scheme. Again, the steps on the substrate have no effects on the resist linewidth and the vertical sidewall.

C. Imprint and RIE scheme

The third method is an imprint and RIE scheme using double layer resists and having a positive tone [Fig. 1(C)].

The process started with an imprint to pattern the top layer. Then the pattern was transferred to the bottom using RIE with the top layer as the mask. The bottom layer is 250 nm thick PMMA with a molecular weight of 15 k. The top layer is 250 nm thick polystyrene with a molecular weight of 2 k. Since polystyrene has a lower *T_g* than PMMA, imprinting on the top layer will not deform the bottom layer. After the imprint, oxygen RIE was used to etch PMMA, with a gas flow of 6 sccm, a pressure of 3 mTorr, and a power of 150 W. In this recipe, 2 k polystyrene has an etching rate of 25 nm/min, which is lower than that of 15 k PMMA (60 nm/min), therefore it can serve as the etching mask for PMMA.

Figure 6 shows a resist pattern on a 100 nm SiO₂ step after processing with the imprint and RIE scheme. Again, the surface, the side wall, and the lateral dimension of the resist

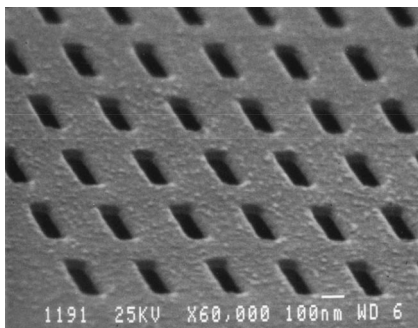


FIG. 2. Rectangular holes in the middle layer in the trilayer scheme [Fig. 1(A)]. The underneath bottom layer is invisible in the SEM photo.

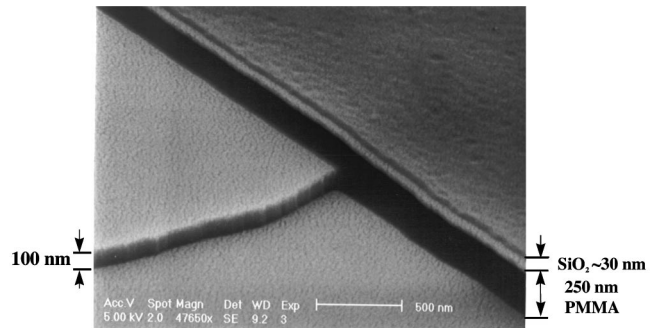


FIG. 3. Resist pattern on a 100 nm SiO₂ sharp step in the trilayer scheme [Fig. 1(A)].

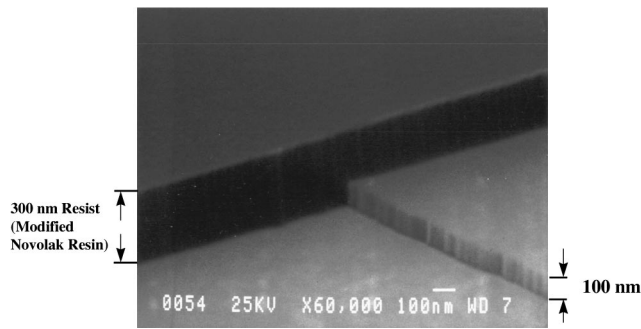
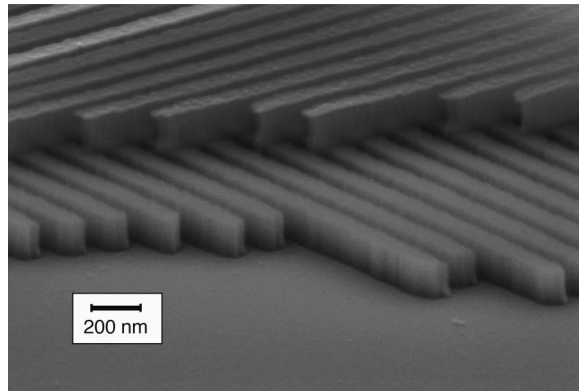


FIG. 4. Resist pattern on a 100 nm SiO₂ sharp step in the imprint and lift-off scheme [Fig. 1(B)].

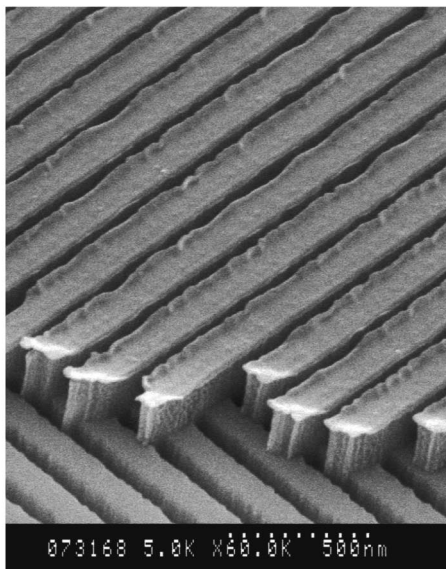
are not affected by the SiO₂ step. However, due to the erosion of oxygen RIE, the polystyrene surface is rough.

D. Imprint and print scheme

The fourth method is an imprint and print scheme that has a positive tone [Fig. 1(D)]. In this method, the planarization



(a)



(b)

FIG. 5. 190 nm period resist grating on a 190 nm period SiO₂ grating in the imprint and lift-off scheme [Fig. 1(B)], viewed from (a) 70° and (b) 40°.

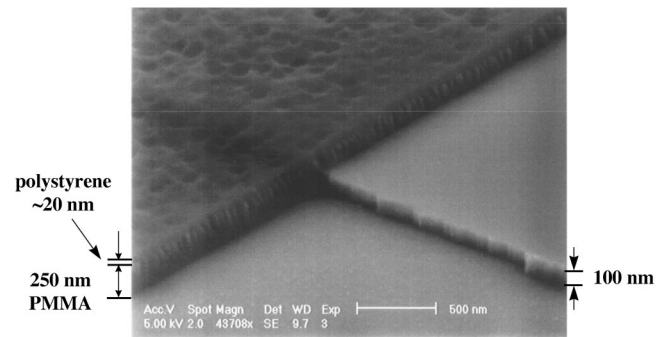


FIG. 6. Resist pattern on a 100 nm SiO₂ sharp step in the imprint and RIE scheme [Fig. 1(C)].

layer is first patterned by NIL. Then a second layer is printed only on the top surface of the imprinted pattern by a roller. The second layer will serve as the etching mask for the following RIE pattern transfer into the whole planarization layer. A careful selection of the material for the second layer and the RIE recipe can give a high RIE selectivity between the roller printed layer and the planarization layer. Thus, the roller printed layer can be used as a RIE etching mask to transfer the NIL pattern.

E. Imprint and etch back scheme

The fifth method is an imprint and etch back scheme that has a negative tone [Fig. 1(E)]. In the first step, the same as that in the fourth method, a pattern was imprinted into the thick planarization layer. Instead of applying another layer of resist by rolling, a second layer of resist was cast by spin coating and was planarized. A uniform etch back of the second layer resist transferred the pattern into the second layer, which in turn, was used as a mask in another RIE etching of the bottom layer.

III. DISCUSSIONS

The advantage of the first method is that it is very flexible and many materials can be used to achieve optimum results. The disadvantage is that the process is complex, requiring more processing steps and hence higher cost. For the second and third methods, the processes are less complicated, but they have more stringent requirements on materials. The second method requires the bottom layer to be insoluble in a chemical solvent such as acetone for the lift-off process, and requires that the T_g of the top layer be lower than that of the bottom layer for the NIL process. In the third method, the top layer should have both a lower T_g and a higher etching resistance than the bottom layer. The two requirements may be difficult to satisfy at the same time. For the fourth and fifth methods, the processes are also simple. The disadvantage for the fourth method is that it might not be easy to achieve good coverage of the top surface when the second layer is printed by a roller. The fifth method requires the surface of the second resist layer to be very flat, otherwise it is impossible to achieve good results for etch back.

For the nonflat surfaces with gradual slopes, patterns can be imprinted directly without a multilayer resist scheme. This is because the mold made of the Si wafer can be deformed. Finally, although most features shown in the article are micron in size, the methods should be applicable to 10 nm features.

IV. SUMMARY

We have devised five methods for NIL on nonflat surfaces. Three of the methods are demonstrated experimentally on a surface with 100 nm deep sharp steps. The choice of each method will be determined by applications. Our results indicate that 10 nm NIL should be achievable on nonflat surfaces.

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³L. H. Sperling, *Introduction to Physical Polymer Science*, 2nd ed. (Wiley, New York, 1992), pp. 352–353.

⁴Purchased from Brewer Science Inc., 2401 Brewer Drive, Rolla, MO 65401.