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# Advanced RELACS<sup>TM</sup> (Resolution Enhancement of Lithography by Assist of Chemical Shrink) Material for 193nm Lithography

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## ABSTRACT

The controllability of iso-dense bias generated by 193nm lithography was intensively studied with novel RELACS<sup>TM</sup> material. The shrinkage, shrinkage linearity, and shrinkage bias were considerably relied on MB temperature. It is the most powerful technology that changing of mixing bake (MB) temperature can control iso-dense bias. Furthermore, AZ<sup>®</sup> Exp.R600 has several attractive advantages, which are able to improve LWS, LER, sidewall roughness of contact holes, surface roughness, and side lobe. Moreover, we have successfully developed a novel RELACS<sup>TM</sup> material to be applied for the patterning of sub-70nm contact hole.

Keywords: RELACS<sup>TM</sup>, 193nm resists, shrinkage, RET, LER, LWS, iso-dense bias

## 1. INTRODUCTION

Since 193nm resists have been developed several years ago, most of commercially available 193nm resists still have not shown enough lithographic margin of contact hole pattern to meet below 0.10 $\mu$ m technology node. Furthermore, 193nm resist suffers from line width slimming (LWS) caused by its chain scission of volatile side chain induced by e-beam irradiation<sup>1</sup> as well as line edge roughness (LER), which is correlated with the quality of the aerial image<sup>2,3</sup>. Though the correlation between LER and the lithography system are fairly clear, the relationship with resist chemistry is less understood. According to recent report, LER in chemically amplified (CA) resists stems in a large part from the phase separation of protected and deprotected polymer in the line edge regions<sup>4</sup>. This disproportional dissolution of partially deprotected polymers plays a major role in the LER of CA resists.

Several years ago, Clariant and Mitsubishi Electronics have developed AZ<sup>®</sup> R200 and AZ<sup>®</sup> R500, which was able to be applied to 0.10 $\mu$ m technology node with 248nm lithography<sup>5,6</sup>. However conventional RELACS<sup>TM</sup> cannot shrink holes on 193nm resists except pure COMA(cyclo-olefin/maleicanhydride) type<sup>7</sup> because all of commercially available 193nm resist have very low acid diffusion from the inside of resist during mixing bake (MB) processing. Actually, acid diffusion phenomena are very important in conventional RELACS<sup>TM</sup> process<sup>6</sup>. The acid diffusion is significantly influenced by the molecular structures of the acid molecule and polymer matrix. Furthermore, the acid diffusion is mostly affected by not only free-volume in the resist but the molecular interaction between the polymers and the photo-induced acid<sup>8</sup>. Especially the diffusion is retarded in the polymer matrix with bulky acrylic groups such as the adamantyl functional group in 193nm resist<sup>9</sup>. Normally, in 248nm resist, photo-induced acid at partially

exposed area as well as exposed area can be diffused to unexposed area during its post exposure bake (PEB) processing. And then this diffused acid is able to diffuse again toward the surface of resist pattern during MB process, therefore, conventional RELACS™ works so well to propagate its shrinkage. In the case of 193nm RELACS™ process with conventional one, however, the algorithm is stopped by very low acid diffusion of 193nm resist. In the long run, there is no shrinkage of the contact hole pattern with 193nm resist.

To ensure sufficient process margin for low- $k_1$  ( $<0.5$ ) imaging, resolution enhancement technology (RET) will be implemented in volume manufacturing. Furthermore, shorter wavelength, higher numerical aperture (NA) projection lenses and improvements in aberrations are necessary more than ever. To compete with the extension of 248nm lithography with proven RET, 193nm technology has to prove its production worthy and be successfully integrated into critical layers with effective cost. Most promising 193nm technology on the cutting edge can print 70nm isolated lines with RET and 140nm contact holes can be patterned without RET, as far as we have known. For sub-100nm contact hole patterning, until now, the unique solution is thermal flow of 248nm resist and RELACS™. Especially 193nm resist for RELACS™ and new RELACS™ material to shrink the contact holes patterned by 193nm resist must be developed<sup>9</sup>. Moreover, in order to optical lithography should be viable through the 70nm node, 193nm with extreme NA must be needed. As NA getting higher lens aberration is also getting worse as well as iso-dense bias. It is so critical at patterning of contact holes from the device side to overcome this huge iso-dense bias despite off-axis illumination (OAI) and aggressive mask bias can help improving depth of focus (DOF) margin as well as resolution of contact holes. The print bias between isolated and dense pattern is strongly influenced by selection of mask bias, partial coherence, and illumination condition<sup>10</sup>. Moreover, thermal flow of resist depends on polymer chains design and baking temperature. In addition, the key issues are dense-iso thermal bias, bulk effect and precision temperature controlling of hot plate<sup>11</sup>. For device manufacturing bulk effect should be compensated by precise mask correction as well as the resist shrinkage amount should match with mask size<sup>12</sup>.

In order to resolve these unwanted shortcomings of 193nm resist process and RELACS™ process, we have developed novel RELACS™ material, AZ® Exp.R600, which consists of aqueous vinyl copolymer and other additives, to shrink the contact hole without any deterioration of lithographic margin. In this paper we report the shrinkage properties of novel RELACS™ material with acrylate type 193nm resist and will discuss new concept of RELACS™ for 193nm resist. Furthermore, this paper focuses on CD bias between dense and isolated contact hole pattern and its extended process ability of 193nm resist with conventional illumination in stead of off-axis illumination, which has extremely big CD bias. Finally, we believe that novel RELACS™ material will help lithographers with its several attractive properties.

## 2. EXPERIMENTAL

AZ® AX1050p-HS was chosen for 193nm resists, which had been already studied in the previous work<sup>10</sup>. The resist solutions were spin coated on organic BARC (AZ® ArF-1C5D) coated silicon wafers, and pre-baked (115°C for 60sec) to obtain a typical film thickness of 0.40  $\mu\text{m}$  using a Tokyo Electron Clean Track ACT8. The coated wafers were exposed using a PAS5500/950 (ASML) ArF scanner with 0.63NA and 0.4  $\sigma$  of conventional illumination at IMEC. Attenuate phase shift mask (6% transparency) was used with 55nm positive bias. On the other hand, in order to pattern 100nm contact holes, we used an ASML PAS5500/1100 with 0.75 of NA and QUAZAR™ (0.85/0.55) illumination. In this case we used an other attenuate phase shift mask (6% transparency) was used with 50nm positive bias. Post exposure bakes at 110°C for 60sec and developed with OPD262 developer for 60sec. The CD measurements were performed on a Hitachi S9200 CD-SEM as follows: acceleration voltage is 600eV with 8.0pA of beam current and threshold is 50% with 4 of view flip number and 64 of integration flip number. In order to prevent LWS, freeze-and-move method was carried on the CD measurements. For the LWS measurements we made out an automatically measurable recipe with repeating cycle of focusing-measuring. The recipe worked under the same condition of previous one. In this case automatic focusing mode was I-diff at 0.14 $\mu\text{m}$  of a contact hole with 0.336 $\mu\text{m}$  of pitch. Magnification was set at 100 k.

RELACS processing, which has already studied previous work<sup>6</sup>, were carried out as follows.

- 1) AZ® Exp. R600, AZ® Exp. R602, and AZ® Exp. R604 were spin-coated on the patterned wafer formed by above conventional patterning process,
- 2) The mixing bake were varied in temperature from 100°C to 180°C for 90sec to satisfy each specific purpose,
- 3) Wafers developed and rinsed by de-ionized water (DIW) for 60sec.

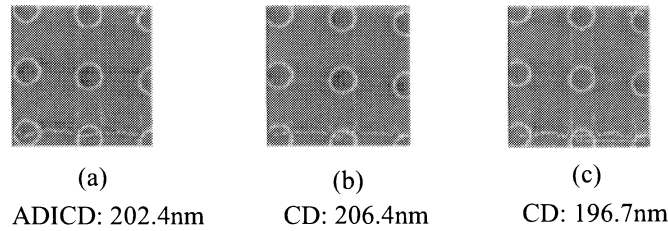


Figure 1. Shrinkage performance of 0.20 $\mu$ m contact holes with 193nm resist by the conventional RELACS™  
 (a) AZ® AX1020p alone, (b) 1<sup>st</sup> AZ® R500 @ MB 150°C/90sec, (c) 2<sup>nd</sup> AZ® R500 @ MB 120°C/90sec

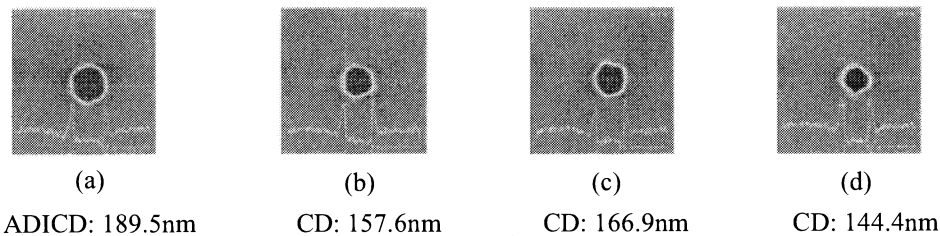


Figure 2. Shrinkage performance of 0.20 $\mu$ m contact holes with 193nm resist by the conventional RELACS™  
 (a) AZ® T926 193nm resist alone  
 (b) AZ® R500 @ MB 100°C/70sec  
 (c) AZ® R500 @ MB 110°C/70sec  
 (d) AZ® R500 @ MB 120°C/70sec

### 3. RESULTS AND DISCUSSION

#### 3.1 New concept of RELACS™ for 193nm resists

As mentioned above the RELACS™, i.e. AZ® R200 and AZ® R500, was not able to shrink the contact holes of AZ® AX1020p (acrylate type)<sup>7</sup>. Although we have done successive RELACS™ process, the shrinkage was no better than the initial CD of resist (Figure 1).

On the other hand we tried to verify whether the lack of acid diffusion in resist film might cause to cut the algorithm of conventional RELACS™ process. The crosslinker used in the conventional RELACS™ is only able to react in the presence of acid catalyst. Therefore we not only designed novel 193nm resist for the purpose of easier acid diffusion after patterning but also it has more hydrophilic side chain to enlarge the mixing boundary with the conventional RELACS™ material. Figure 2 shows the conventional AZ® R500 works so well with specially designed 193nm resist. It is an obvious smoking gun that the conventional RELACS™ cannot shrink the pattern of 193nm resists due to very low acid diffusion of 193nm resist.

By the previous evidence we have conceived new concept of RELACS™ for 193nm resist. Several criteria were established as follows.

- 1) Do not use the diffused acid from resist,
- 2) Select material to form the potential mixing boundary first,
- 3) Inquire to maximize the mixing boundary between RELACS™ and resist,
- 4) Adopt self-crosslinkable moiety in the polymer by thermal activation.

If the RELACS™ process should be carried out regardless of the acid diffusion from resist, the most important factor is the depth of mixing boundary between two polymers of RELACS™ and resist. The depth of mixing boundary means that the intensity of interaction between different field of two substance. When a substance faces to different substance, we assume, the fields of two substances are able to be mingles and makes mixing boundary each other at their interface if they have affinity. Unfortunately we cannot prove or verify this assumption to experiment and mathematical equation here, however we have had a lot of experiences through the numerous tests. Specific

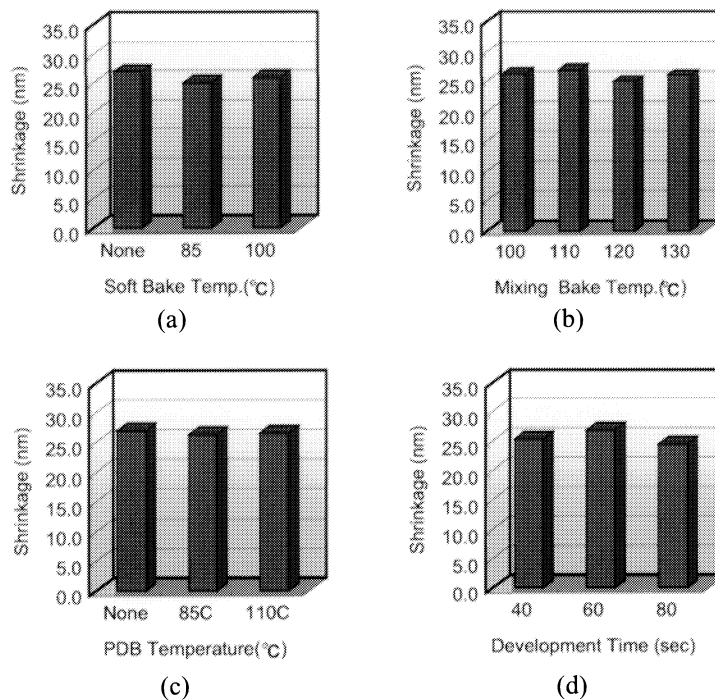


Figure 3. Shrinkage performances of contact holes with 193nm resist depend on various process parameter (Resist: AZ® AX1050p-HS, RELACS™: AZ® Exp.R600)

- (a) MB: 110 °C /90sec, PDB: none, Dev. Time:60sec
- (b) SB: none, PDB: none, Dev. Time:60sec
- (c) SB: none, MB: 110°C/90sec, Dev. Time:60sec
- (d) SB: none, MB: 110 °C /90sec, PDB: none

polymers showed deeper mixing boundary with 193nm resist, but other polymers did not. In the long run we selected several kinds of polymer which showed deepest mixing boundary. In this study we introduce new RELACS™ material based on one of them.

### 3.2 Shrinkage characteristics depend on process parameters

In previous work<sup>7</sup>, RELACS™ should be process via 3 times of bake step in its process flow. New RELACS™, AZ® Exp.R600, should be verified whether each bake step is needed or not and how much sensitive to each process parameter. Figure 3 shows the dependency of AZ® Exp.R600 on each process parameter in the view of shrinkage change. Regarding bake steps AZ® Exp.R600 is insensitive to soft bake (SB), mixing bake (MB), post development bake (PDB), and development time. In other word the steps of SB and PDB had better to skip from process flow in order to increase throughput. Needless to say the film thickness increases by skip of SB step. Although the shrinkage is invariable to change MB temperature from 100°C to 130°C for 90sec, we need to investigate at much higher MB temperature. It will be discussed later.

### 3.3 Depth of focus post RELACS™ process

As shown in Figure 4, 0.14μm contact holes with 0.336μm of pitch has 0.40μm of DOF margin using attenuated PSM (6% transparency) with 18.4%.of exposure latitude. After RELACS™ processing with AZ® Exp.R600, we have successfully achieved 0.115μm of contact holes without any change of initial DOF.

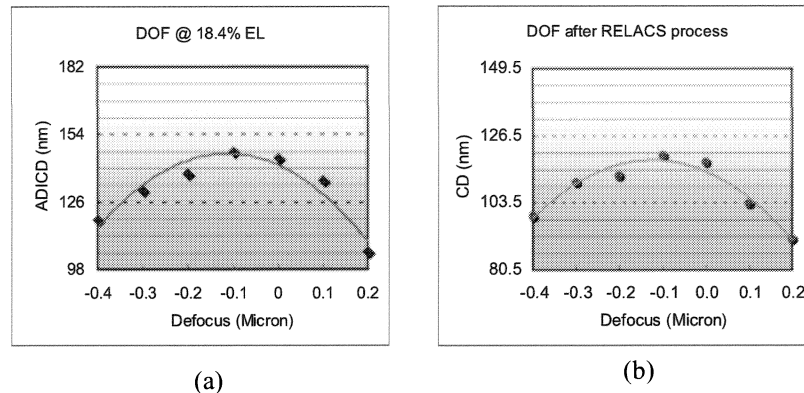


Figure 4. Change of DOF margin post RELACS™. (Resist: AZ® AX1050p-HS, RELACS™: AZ® Exp.R600)  
(a) Ante RELACS™, (b) Post RELACS™ @MB: 110°C/90sec / 0.14μm with 0.336μm of pitch

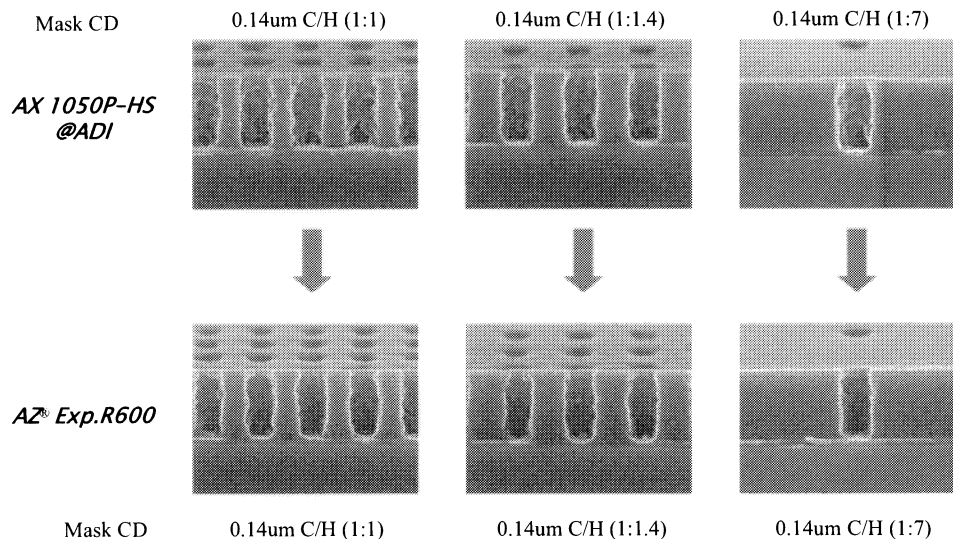


Figure 5. Cross-sectional Images ante/post RELACS™. (Resist: AZ® AX1050p-HS, RELACS™: AZ® Exp.R600)

### 3.4 Cross-sectional SEM images post RELACS™ process

Figure 5 shows cross-sectional SEM images of 0.14μm of contact holes post AZ® Exp.R600 at different pitch size. Significantly the sidewall roughness of contact holes was considerably improved. However lipping of initial profile was not disappeared clearly.

### 3.5 Improvement of Line Edge Roughness (LER)

As shown in Figure 6, the LER of initial resist pattern was 11.31nm ( $3\sigma$ ), but AZ® Exp.R600 was able to improve LER to 8.8nm by the average of 9 points. For the LER measurements the 0.12μm line/space (1:1) of AZ® AX1020p 193nm resist were patterned by an ASML PAS5500/950 with 0.63 of NA and annular illumination (0.85/0.55). At 200 K magnification LER analysis is performed by right edge mode with  $3\sigma$  on an image obtained by 32 scans using 32 measurement points and 11 sum points/line with and inspect area of 389.

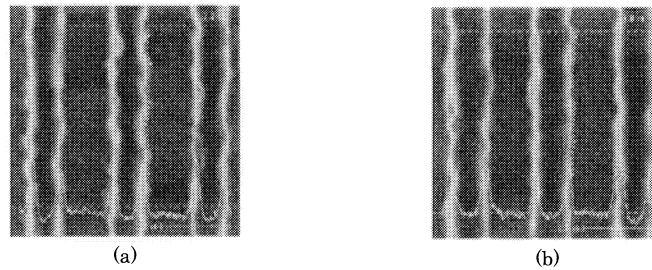


Figure 6. Improvement of LER post RELACS™.  
 (a) AZ® AX1020p alone:  $3\sigma = 11.31\text{nm}$  (Average of 9points) \*ADICD: 109.25nm  
 (b) AZ® Exp.R600:  $3\sigma = 8.8\text{nm}$  (Average of 9points) \*CD: 124.38nm

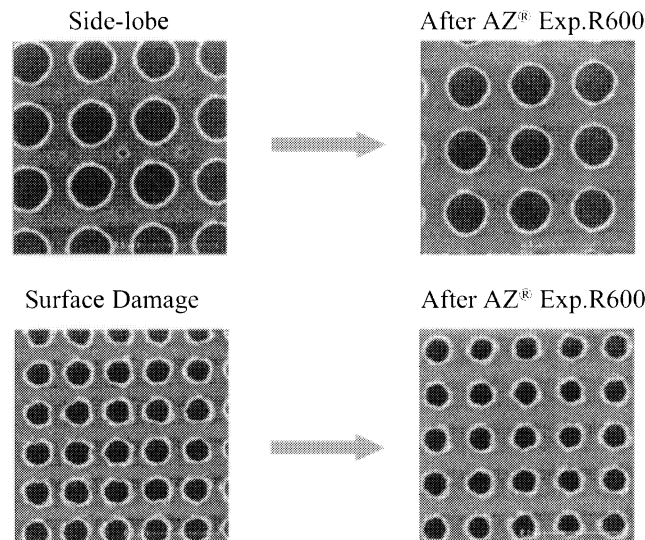


Figure 7. Improvements of side-lobe and the surface roughness post AZ® Exp.R600

### 3.6 Improvements of side lobe and surface roughness

Nevertheless attenuated PSM has been the most powerful method of RET to resolve the contact holes below sub-wavelength, the side lobe is critical issue. Furthermore the surface roughness of the resist is also becoming important. At 70nm technology node the surface roughness is going to be a bigger issue. Figure 7 shows AZ® Exp.R600 can mend side lobe as well as the surface roughness.

### 3.7 Relieving line width slimming (LWS)

Current commercially available 193nm resists contain carbonyl groups, which are decomposed by highly accelerated electrons during CD-SEM measurement<sup>13,14</sup>. It causes CD slimming and 15-30% film thickness shrinkage owing to the mass loss due to a thermal decomposition reaction. In the previous related study, although the amount of CD slimming of 193nm resist (VEMA type) was up to 20nm after 50 measurements, it could be relieved to 8nm by use of anti-shrinkage coating (ASC)<sup>15</sup>. Actually ASC was a prototype of AZ® Exp.R600. Several studies have reported that acrylate-type resists showed much slimmer than COMA or VEMA resists<sup>14,16,17,18</sup>. Previous studies have demonstrated considerable factors towards understanding what influences the LWS. Higher accelerating voltages

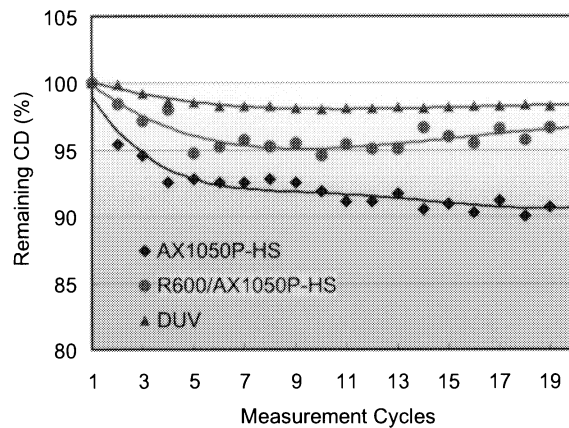


Figure 8. Relieving effect of LWS post AZ<sup>®</sup> Exp.R600 (MB : 110°C/90sec /Measured at 0.14 $\mu$ m of a contact hole of AZ<sup>®</sup> AX1050p-HS with 0.336 $\mu$ m of pitch under 600eV, 8.0pA, and 100K)

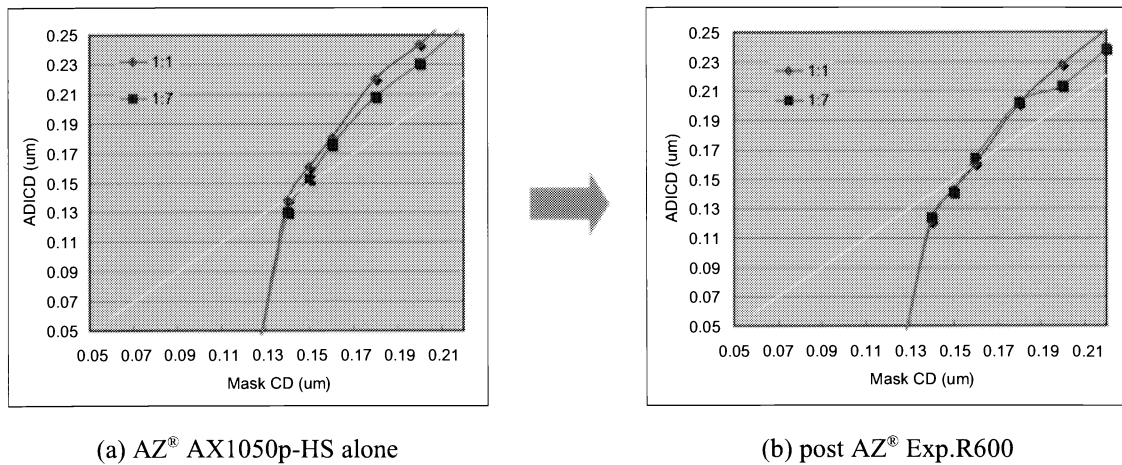


Figure 9. Improvement of iso-dense bias by conventional illumination ante/post AZ<sup>®</sup> Exp.R600  
(Eo is set at 0.14 $\mu$ m C/H (1:1) on ASML PAS/950 with 0.63NA and conventional ill. (0.4  $\sigma$ ) with att-PSM (6%)  
Mask bias: 55nm)

cause greater amounts of LWS during CD-SEM measurement and beam current appears to have insignificant effect on LWS. The magnitude of LWS appears absolute, independent of feature size, implying a surface phenomenon. Several previous studies<sup>1,19</sup> noted that LWS can be controlled by the CD-SEM measurement condition, however, the lithographers still suffer from LWS. Because, especially, the irradiated area during measurement cannot help avoiding CD slimming, it must cause the CD variation during dry etching process. Figure 8 notes how AZ<sup>®</sup> Exp.R600 relieves the CD slimming of contact hole with worst acrylate-type193nm resist. It is speculated that AZ<sup>®</sup> Exp.R600 is able to relieve the CD slimming up to 70% of 248nm resist.

### 3.8 Iso-dense bias post RELACS<sup>™</sup> process

In order to investigate the influences of optical settings on iso-dense bias ante/post RELACS<sup>™</sup>, two set of experimental substrates were prepared as follows. One is set conventional illumination (0.4  $\sigma$ ) with 0.63 of NA using attenuated PSM (6% transparency) with 55nm bias. The other is prepared QUAZAR<sup>™</sup> illumination (0.85/0.55)



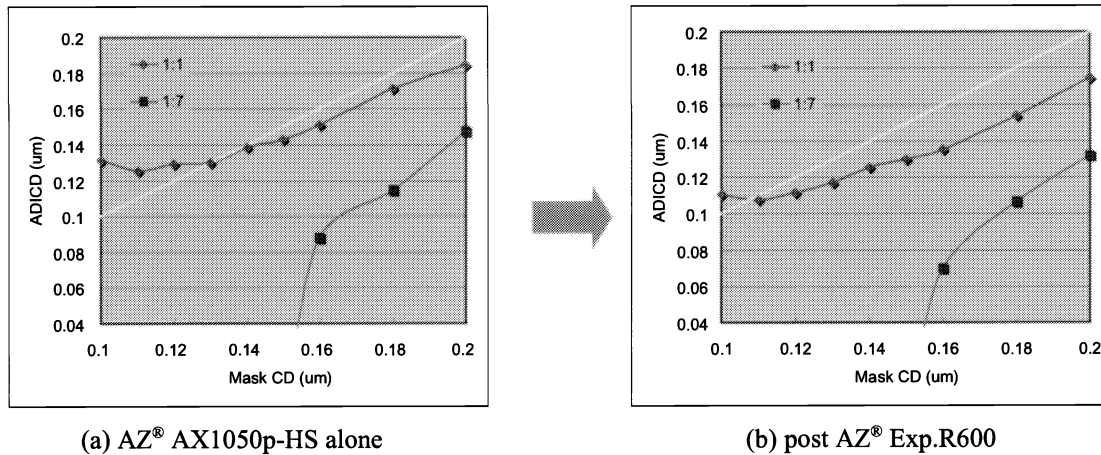


Figure 10. Improvement of iso-dense bias by conventional illumination ante/post AZ® Exp.R600 (Eo is set at 0.14μm C/H (1:1) ASML PAS/1100 (0.75NA, QUAZAR™(0.85/0.55) with HT-PSM (6%) Mask bias: 50nm)

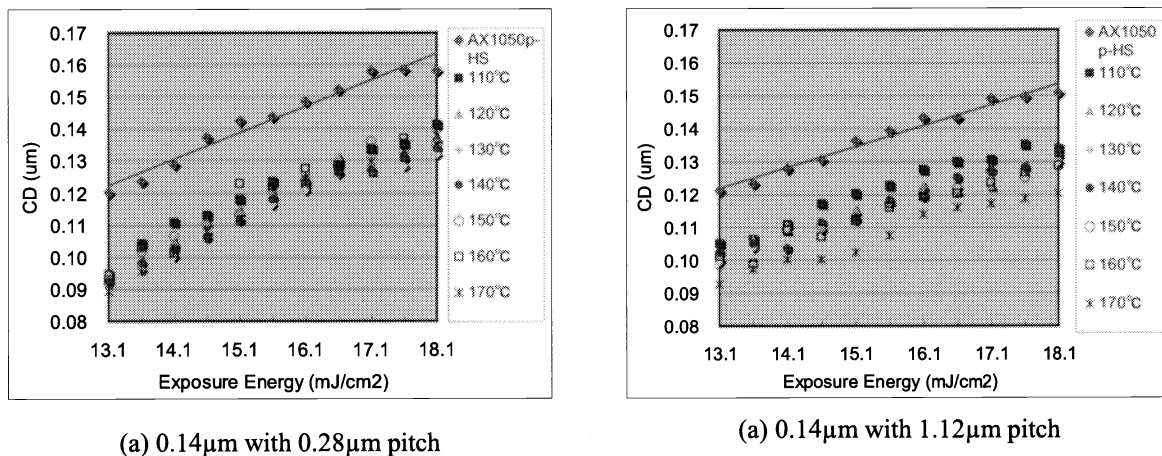


Figure 11. CD variations depend on MB temperature ante/post AZ® Exp.R600 (ASML PAS/950 (0.63NA, Conventional ill. (0.4 σ) with HT-PSM (6%) Mask bias: 55nm)

with 0.75 of NA using attenuated PSM (6% transparency) with 50nm bias. Comparing iso-dense bias different hole pitches, which are 0.14μm contact holes with 0.28μm (1:1) and 1.12μm (1:7) of pitches, are evaluated under each optical setting. Dense and isolated patterns had 0.30μm and 0.5μm of DOF, respectively. Normally iso-dense bias by QUAZAR™ illumination was much bigger than by the conventional illumination. Figure 9 shows how the iso-dense bias by conventional illumination change ante/post RELACS™ process. While iso-dense bias is insignificantly small, AZ® Exp.R600 renders the bias much smaller, especially around the optimum exposure dose, without deterioration of linearity. In Figure 10, iso-dense bias generated by the QUAZAR™ illumination is too bigger than by the conventional illumination, however, the iso-dense bias can be improved by new RELACS™

### 3.9 Shrinkage linearity and shrinkage bias

In the previous section, AZ® Exp.R600 is able to reduce iso-dense bias that is already generated by optical setting. Actually the iso-dense bias of initial resist pattern was +6.21nm at 0.14μm (Eo) and then decreased to -2.32nm post AZ® Exp.R600. What is a factor to control the reducing of iso-dense bias post RELACS™? If you find it out you can

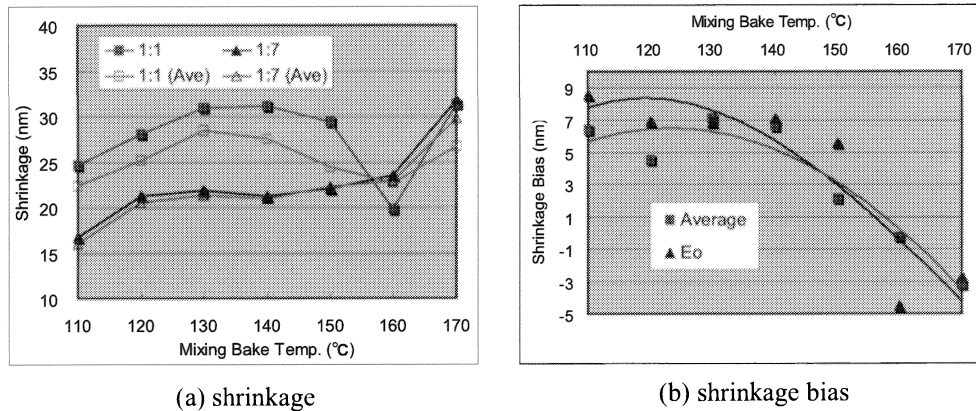


Figure 12. Dependency of shrinkage and shrinkage bias on MB temperature at 0.14iso/dense contact holes (ASML PAS/950 (0.63NA, Conventional ill. ( $0.4\sigma$ )) with HT-PSM (6%) Mask bias: 55nm)  
 \*Average (Ave.) is mean value corresponding to exposure dose

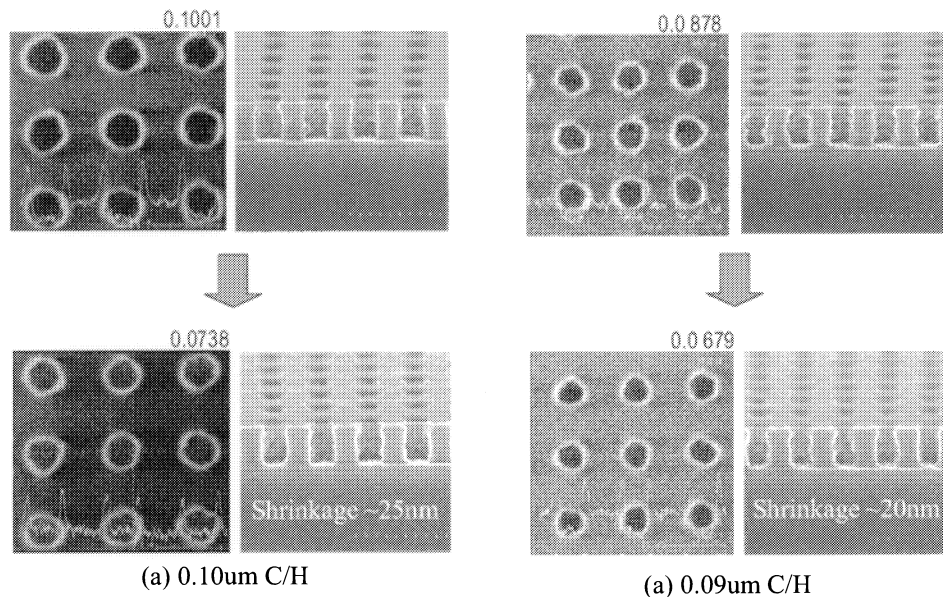
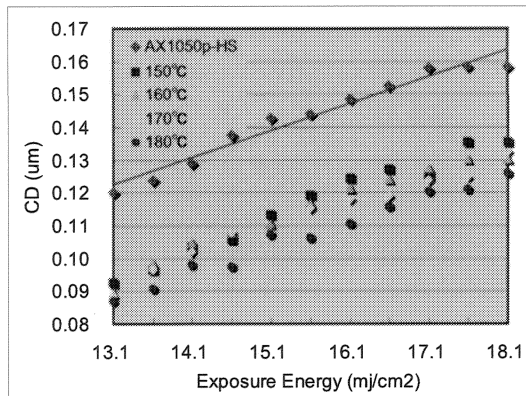


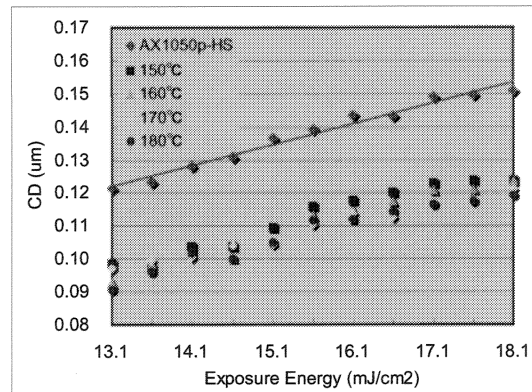
Figure 13. Shrinkage performances of AZ® Exp.R600 on 0.010um and 0.090um of contact holes (1:1.4) (ASML PAS/1100 (0.73NA, QUAZAR™ (0.85/0.55) with HT-PSM (6%) Mask bias: 50nm)

control the iso-dense bias using AZ® Exp.R600. Fortunately, we have found that shrinkage linearity and shrinkage bias between isolated and dense patterns were considerably relied on the MB temperature. It means you are able to control iso-dense bias by changing of MB temperature, in the long run. Figure 11 shows the dependency of CD at 0.14 $\mu$ m with 0.28 $\mu$ m and 1.12 $\mu$ m of pitches on MB temperature ante/post RELACS™ process, respectively. Isolated contact holes are more variable with MB temperature than dense contact holes. Consequently, we need to check more precise behavior about shrinkage and its bias depending on MB temperature.

In Figure 12, the shrinkage has a little strong dependency on MB temperature, while the shrinkage bias is remarkably rely on MB temperature. Shrinkage rate is 0.217nm/°C for 0.28 $\mu$ m pitch (1:1) and 0.251nm/°C for 1.12 $\mu$ m (1:7). Furthermore, shrinkage bias decreasing rate is 0.159nm/°C by average and 0.188nm/°C for Eo.

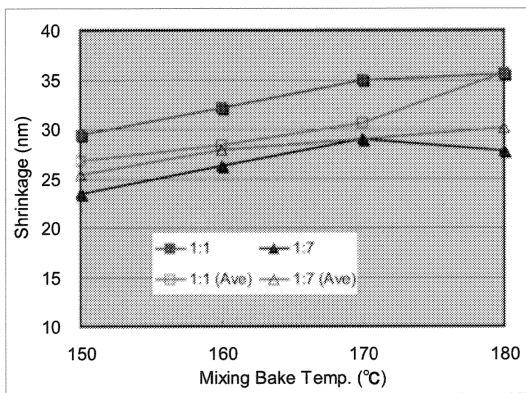


(a) 0.14 $\mu$ m with 0.28 $\mu$ m pitch

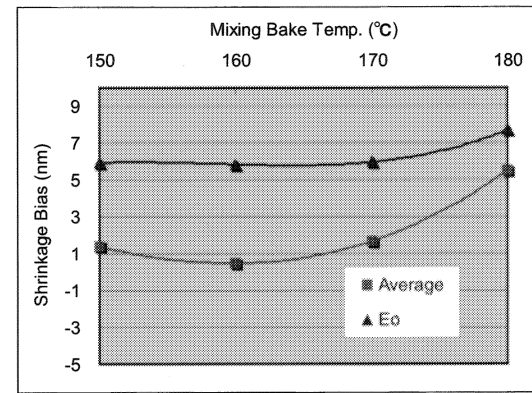


(b) 0.14 $\mu$ m with 1.12 $\mu$ m

Figure 14. CD variations depend on MB temperature ante/post AZ<sup>®</sup> LExp.R602 (ASML PAS/950 (0.63NA, Conventional ill. (0.4  $\sigma$ ) with HT-PSM (6%) Mask bias: 55nm)



(a) shrinkage



(b) shrinkage bias

Figure 15. Dependency of shrinkage and shrinkage bias on MB temperature of AZ<sup>®</sup> LExp.R602 at 0.14iso/dense contact holes (ASML PAS/950 (0.63NA, Conventional ill. (0.4  $\sigma$ ) with HT-PSM (6%) Mask bias: 55nm)

\*Average (Ave.) is mean value corresponding to exposure dose

### 3.10 Challenging to sub-70nm node contact hole

As shown in Figure 13, we have successfully shrunk 0.100 $\mu$ m and 0.088 $\mu$ m of contact holes to 0.074 and 0.068 $\mu$ m with AZ<sup>®</sup> Exp.R600, respectively. Currently, the finest contact holes to resolve by 193nm single layer resist

(SLR) with optical RET on the cutting edge will be 0.09 $\mu$ m (1:1). However we should not forget whenever the semiconductor manufacturing is not only technology driven but also the most cost-effective technology is the one used for. Novel RELACS<sup>™</sup> is the most attractive material that has good reliability to shrink the contact holes of 193nm resist as well as cost-effective.

### 3.11 Performances of modified AZ<sup>®</sup> Exp.R600

In order to increase the shrinkage of AZ<sup>®</sup> Exp.R600 we have developed AZ<sup>®</sup> LExp.R602, which is modified crosslinker with the same polymer matrix. MB temperature is relatively higher and shrinkage is bigger than AZ<sup>®</sup>

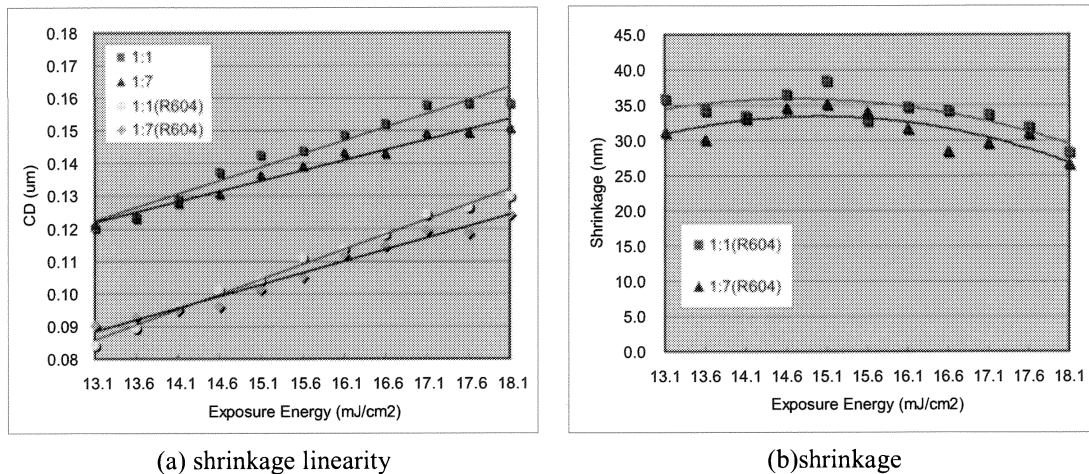


Figure 16. Shrinkage performances ante/post AZ® LExp.R604 at 0.14iso/dense contact holes (ASML PAS/950 (0.63NA, Conventional ill. ( $0.4\sigma$ )) with HT-PSM (6%) Mask bias: 55nm)

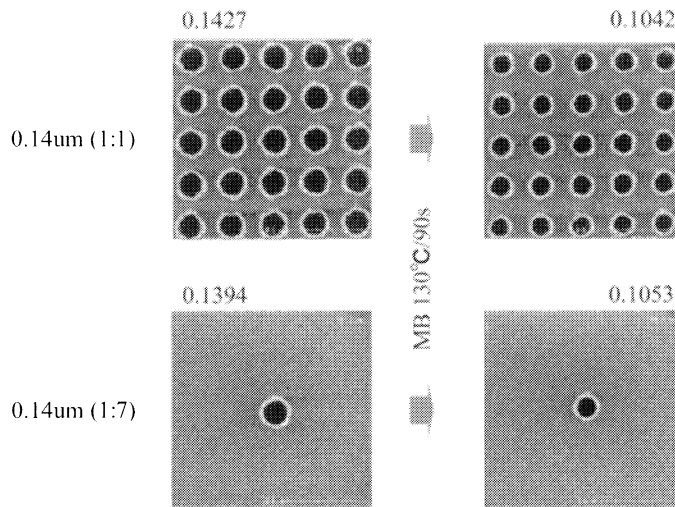


Figure 17. Top-view images ante/post AZ® LExp.R604 at 0.14iso/dense contact holes (ASML PAS/950 (0.63NA, Conventional ill. ( $0.4\sigma$ )) with HT-PSM (6%) Mask bias: 55nm)

Exp.R600. Figure 14 shows the dependency of CD variations at 0.14μm with 0.28μm and 1.12μm of pitches on MB temperature ante/post RELACS™ process, respectively. Isolated contact holes are less variable with MB temperature than dense contact holes. It is very interesting result to compare with AZ® Exp.R600. In Figure 15, while shrinkage rate is little smaller than AZ® Exp.R600, it has more linear relationship on MB temperature. The shrinkage rate is 0.203nm/°C for 0.28μm pitch (1:1) and 0.1851nm/°C for 1.12μm (1:7). Furthermore, the shrinkage bias is very small as MB temperature increased. Shrinkage bias increasing rate is 0.023nm/°C by average and 0.099nm/°C for Eo.

Recently, we have successfully developed another modified one, AZ® LExp.R604, it shows 40nm of shrinkage in spite of lower MB temperature (Figure 16, Figure 17). Unfortunately, we have no data the dependency on MB temperature, yet. In this case the iso-dense bias of initial resist pattern was +6.21nm at 0.14μm (Eo) and then decreased to +2.81nm post AZ® LExp.R604. Furthermore, its shrinkage bias was 2.59nm in average corresponding exposure dose, at the most.

#### 4. CONCLUSIONS

Novel advanced RELACS™ material, AZ® Exp.R600, has been developed for 193nm lithography. It showed 30nm of shrinkage with acrylate type 193nm resist above MB 130°C/90s. AZ® Exp.R600 has not only robust process window for shrinkage but it could improve LER, side lobe, surface roughness, and sidewall roughness of contact hole. Furthermore, it could relieve LWS of 193nm resist up to 70% of 248nm resist. We have also successfully achieved 40nm of shrinkage by modifying AZ® Exp.R600. Therefore AZ® Exp.R600 was able to print 75nm of contact hole from 100nm of initial contact hole patterned by 193nm lithography. Finally, the controllability of iso-dense bias generated by 193nm lithography by using AZ® Exp.R600 was also intensively studied. Significantly, it could control iso-dense bias by the dependency of shrinkage bias on MB temperature. The shrinkage, shrinkage linearity, and shrinkage bias were considerably relied on MB temperature. It is most powerful technology that iso-dense bias can be control by changing of MB temperature.

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