Field-theoretic Simulations of Directed Self-assembly for Contact Multiplication

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We use three-dimensional self-consistent field theory (SCFT) to study the self-assembly of cylinder forming diblock copolymers confined in elongated templates. This situation arises in contact holes where the goal is to produce a contact hole with reduced dimensions as well as with narrowed pitch of the center-to-center distance of cylinders. In this study, we focus on systems where two minor-block cylinders form in inside of the elongated templates. A defective bridge structure is extensively studied in this work and we evaluate the defect in various wall affinities such as “all PMMA-attractive templates”, “all neutral templates” and “PMMA-attractive sidewall with the neutral substrate”. According to our SCFT simulations, the defect formation energy of the bridge is typically above 20 kT, or fewer than 2 defects per billion in the “all neutral” template and “PMMA-attractive sidewall with the neutral substrate”, while the defect preferably forms in the “all PMMA attractive” template.

Keywords: Self-Consistent Field Theory (SCFT), graphoepitaxy, directed self-assembly, hole shrink, pitch multiplication, bridge

1. Introduction

Conventional lithography is rapidly approaching its scaling limits. Directed self-assembly (DSA) of block copolymers in particular has received significant attention as a candidate for the next generation lithography in the last decade\textsuperscript{1-4} and numerous studies to be a high-resolution patterning technique have been done not only experimentally but also numerically\textsuperscript{6-10}. The self-assembly of block copolymers have been successfully applied to line and space (L/S) and contact holes with low defect densities. With respect to vertical interconnect access lithography (VIAL), many researches have demonstrated that DSA of cylinder-forming block copolymers such as poly(styrene-b-methyl methacrylate) (PS-PMMA) can be used to produce high-resolution contact holes with reduced critical dimensions (CDs) relative to a prepattern made by the conventional top-down lithography\textsuperscript{11-13}. In addition to the hole shrink problem with PS-PMMA block copolymers, recent studies have focused on contact-hole multiplications for the purpose to decrease the lithography mask cost.

Defectivity, however, has been an issue to be solved. The experimental studies show highly desirable pairs of inner PMMA cylinders in an elongated template, while a cross-sectional image reveals an undesired morphology where two cylinders are connected at their bottom by a PMMA bridge\textsuperscript{14}. Although our previous study\textsuperscript{8} treats the bridge configuration as a perfect state since a highly anisotropic dry etching can transfer their patterns to the substrate, the bridge defect would be an issue to any pattern transfer procedure in an isotropic wet etching process which is mostly used to remove PMMA cylinders. As for the reason, the bridge morphology should be avoided, however, it has received little attention. Extensive studies of large parameter spaces are still to be carried out.

In this study, we focus in particular on the bridge morphology even though the other types of defects should be considered. Ideally, hole templates with neutral substrates would greatly favor the
formation of standing PMMA cylinders spanning from the surface to the substrate. Therefore we focus on contact multiplication with a pair of vertically-oriented PMMA cylinders in all PMMA-attractive templates (PMMA templates), all neutral templates (neutral templates) and PMMA-attractive sidewalls and neutral substrates (mix templates). We use three-dimensional self-consistent field theory (SCFT) simulations to self-assembly configurations in nano-scaled confinements and establish process windows where no defects form. Moreover, we discover the properties of DSA morphologies, such as CDs of PMMA cylinders and pitch for the center-to-center distance of the cylinders.

2. Self-consistent Field Theory

Three-dimensional self-consistent field theory (SCFT) was used for these studies as described elsewhere. The field-based SCFT is a powerful and reliable tool to discover self-assembling morphologies of block copolymers so that most of phase diagrams have been studied in the last a few decades. In addition to that, the free energy can be calculated with no further calculations. Since the total free energy of a resulting morphology is readily available from our SCFT calculations, the stability of defective and perfect structures can be established from measurements of the defect formation energy, computed as \( \Delta F = F_{\text{defect}} - F_{\text{perfect}} \).

Our molten block copolymers have \( N \) monomers per chain, \( N = N_{\text{PS}} + N_{\text{PMMA}} \), and the volume fraction of the minor PMMA block is denoted, \( f \). In addition, the interaction between monomers of different types are modeled using a Flory parameter, \( \chi \), resulting in a dimensionless segregation strength, \( \chi N \). Furthermore, masking walls provide the flexible designs for templates with various selectivity conditions using a Flory-like parameter \( \chi_w \). In this study, we are in particular interested in wall affinity effect to the defectivity so that we vary \( \chi_w N \) as described below: All PMMA–attractive and all neutral templates are modeled with \( \chi_w N = -15 \) and \( \chi_w N = 0 \), respectively. The mix templates have PMMA attractive sidewalls (\( \chi_w N = -15 \)) and neutral substrate (\( \chi_w N = 0 \)). The air is considered to be neutral to PS and PMMA in all calculation in this study.

3. Results and Discussion

The cylinder forming block copolymers in

![Fig. 1 Top view of elongated template. The Major CD and minor CD are defined as described.](image1)

![Fig. 2 PMMA density profiles of the perfect structures in (a) PMMA templates, (b) neutral templates and (c) mix templates. The bright area represents enriched PMMA density. The top image corresponds to a top view and the bottom one represents a cross-sectional view. (d) 3D-image of the perfect structures in the neutral templates.](image2)

![Fig. 3 PMMA density profiles of the bridge defect in (a) PMMA templates, (b) neutral templates and (c) mix templates. The bright area represents enriched PMMA density. The top image corresponds to a top view and the bottom one represents a cross-sectional view. (d) 3D-image of the defective bridge in the neutral templates.](image3)
between 51 nm and 66 nm are best to form a pair of perfect cylinders. While the cylinders do not reach to the substrate in PMMA templates, the cylinders span from the top to the substrate in neutral templates and mix templates as shown in Fig. 2. Our previous study in the neutral templates interestingly revealed the two cylinders with hexagonally-packed cylindrical configurations shown in Fig. 2 (b) and (d), which are similar to hexagonal cylinder morphologies in bulk.

Defective morphologies of interest in several wall affinities are summarized in Fig. 3. Similar to the perfect structure case, we also found the bridge morphologies with hexagonally-packed hemicylinders along the sidewall in the neutral templates (Fig. 3 (b) and (d)).

Using seeds corresponding to the perfect structures and the bridge defective structures, we conducted the SCFT to calculate the defect formation energy in templates varying the major and minor CDs from 88 nm to 102 nm and 51 nm to 66 nm, respectively. For the purpose of discussing a process margin, we prepare the process windows with respect to the template dimensions, minor CDs and major CDs. Since above 20 kT of the defect formation energy corresponds to less 2 defects per billion, defect-free process windows should have above 20 kT of defect energy, shown in Fig. 4 as “white area”. The process window of the PMMA templates is dominated by “red area”, or the negative defect energy, corresponding to that the bridge defects are more stable than the perfect states. As the terminal of the discontinuous cylinders has high curvature resulting of entropy loss on the polymer, it presumably destabilized the perfect structures. Note that the bridge morphology would be considered as the perfect structures with highly anisotropic dry etching procedure. While the PMMA domains in the PMMA templates form the bridge structures, the neutral templates have the wider window to form the perfect structures. It turns to be that the perfect two cylinders are stable with above 22 kT, which is beyond the ITRS threshold of ~ 20 kT, in the region we considered. More extensive computational study for the neutral case can be found elsewhere. As for the process window for the mix case, relatively wide window is found from our calculation. The optimal minor CD for the simple hole shrink problem with the block copolymer, \( f = 0.3 \) and \( \chi N = 25 \), is around 58 nm while the optimal minor CD to form the perfect structures interestingly shifts toward large minor CDs.

![Fig. 4 Process windows of various major CDs and minor CDs. These windows consider only the bridge defects. The number in each box indicates the defect formation energy in units of kT. The red, yellow and white area show that the defect formation energy is less than 15 kT, 20 kT and more than 20 kT, respectively. (a) in PMMA templates (b) in neutral templates (c) in mix templates.](image)

![Fig. 5 Defect formation energy of the bridge defects as a function of \( \chi N \) for the side and the substrate. The major and minor CD are 95 nm and 58 nm, respectively.](image)
CD. In terms of the optimal major CDs, it is independent on the wall affinity so that the defect energy varying major CDs have always the highest at ~95 nm. To conclude, we found that the perfect cylinders in the neutral template are the most stable among the three wall conditions that we studied. Precise control of the wall affinities, however, still remains as an issue.

Therefore we next turn our attention to the defect energy with respect to the wall affinities raging from strongly PMMA-attractive templates to slightly PS-attractive templates. The plot in Fig. 5 shows us that slight differences of the $\chi_w N$ result in drastic change in the defect energy around the neutral templates. As approaching towards the PMMA attractive, the defect energy of the bridge structure becomes negative indicating that the bridge is more favorable than a pair of the cylinders. Then the energy turns to be independent on the $\chi_w N$.

In this paragraph, we discuss the properties of the PMMA cylinders in the PMMA templates, the neutral templates and the mix templates. Several properties of the PMMA domains including the VIA CD and the pitch denoted as H2H distance, shown in Fig. 6. Despite of the wall affinities, we find that the VIA CD and the pitch linearly increase for all minor CDs when the major CD increases. Besides of the above findings, we find that the VIA CDs and the pitch for the PMMA templates and the mix templates are almost identical while the VIA CDs and the pitch for the neutral templates are slightly large. Note that, since we measure the VIA CDs by fitting the PMMA domains as a circle and the domains are elongated with increasing the major CDs, the values of the VIA CD are over-estimated. The variations of the pitch are independent of the minor CDs.

4. Summary and Conclusions

We used three-dimensional SCFT of cylinder forming block copolymers to investigate the process windows of the bridge defects in templates with various wall affinities, such as the PMMA
templates, the neutral templates and the mix templates. We found that the bridge structures are stable rather than the perfect states in templates varying both minor and major CDs, however, the perfect configurations are more stable in the neutral templates despite of the template CDs. Not only the self-assembly in the neutral preferably form the perfect structures, but also the defect formation energy is beyond the ITRS threshold so that it is presumably promising for the defect-free contact multiplication with a wide window. A pair of vertically-oriented PMMA cylinders in the mix templates has the promising process window, however, it is relatively narrow in comparison with the neutral one. Moreover, it is still challenging to assemble the substrate and the sidewall with different wall affinities.

In terms of the properties in the templates that we studies, over all, significant difference between different wall types is not found. The VIA CDs and pitch in the neutral templates are relatively small while they are almost identical in the PMMA and the neutral templates.

We reveal not only optimal conditions with respect to the templates CDs and the wall affinities. We hope that it will provide useful guidance in DSA lithography.

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