Templating Three-Dimensional Self-Assembled Structures in Bilayer Block Copolymer Films

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Previous studies have shown that using chemical or physical templating allows for the alignment and registration of different morphological features in a self-assembled monolayer film of block copolymers (BCP). Although two-dimensional (2D) patterns are useful for the fabrication of nanoscale structures, for some applications such as multi-level structures and metamaterials, three-dimensional (3D) patterns are needed. Current strategies for the fabrication of 3D structures such as stacking methods are complicated by the alignment requirements for the multiple layers. This study shows that a BCP film consisting of a bilayer of cylindrical microdomains self-assembled on a majority-block brush-coated template can form a wide range of complicated 3D structures such as mesh-shaped structures and periodic super-structures, and control of the morphology of the microdomains in both layers is possible. Moreover, we show that this method can independently control the orientation of cylindrical microdomains in each layer, forming aperiodic structures such as bends and junctions. The 3D nature of the results is supported with self-consistent field theory simulations.

The main steps of the fabrication procedure are described in Figure 1. For the experiments, a bilayer cylindrical-morphology poly(styrene-b-dimethylsiloxane) (PS-PDMS) diBCP was spin-coated on the substrates. We used arrays of PS-functionalized posts, with 20-nm diameter and 38-nm height and different periods of the posts for the experiments.

Figure 2 shows an example of a 3D structure. In the template region, ox-PDMS cylinders self-assembled into a mesh-shaped structure, which covered the whole area of the template. Outside the templated region, the top and bottom ox-PDMS cylinders share local orientation but no long-range order. For the fabrication of aperiodic structures we used two strategies: 1) altering the post periodicity in the *x*- or/and y-directions, 2) changing the post arrangement. Figure 3 shows an example of the two strategies. As can be seen in this figure, by using these two strategies the direction of cylinders in one of the layers can be changed independent of the other layer and we made bends and junctions in both the top and bottom layers.

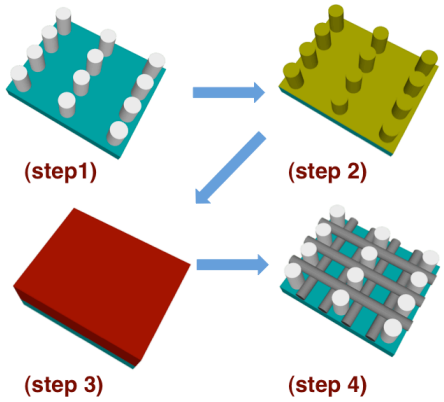

Figure 1: Diagram of the main steps of the method, (step 1) making the post array using electron beam lithography (EBL) of hydrogen silsequioxane (HSQ) resist, (step 2) functionalizing the post array using a PS brush (1 kg mol^{-1} PS brush). Next, (step 3) spin coating the PS-PDMS diBCP (45 kg mol^{-1}, 32%vol PDMS) onto the substrate and we used solvent annealing of a mixture of toluene and heptane, (step 4) using a CF_4 RIE to remove the top PDMS wetting layer and followed by oxygen RIE to remove the PS matrix leaving the ox-PDMS 3D structures remaining on the substrate.

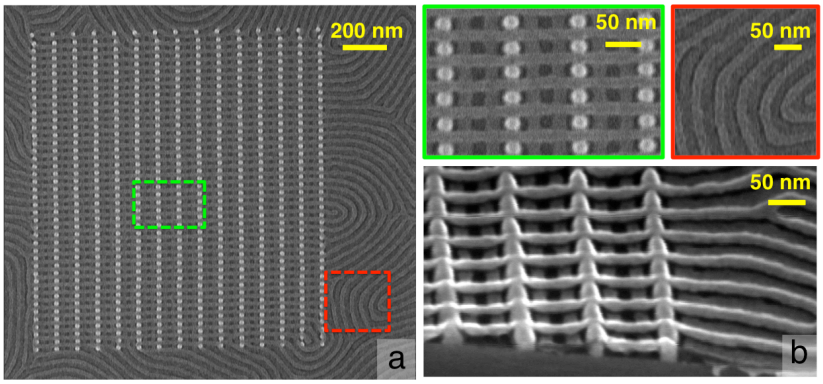

Figure 2: (a) SEM image of a templated 3D structure, (b) zoomed-in and cross-sectional images of inside and outside the template. Bright grey and white colors represent ox-PDMS and HSQ posts, respectively.

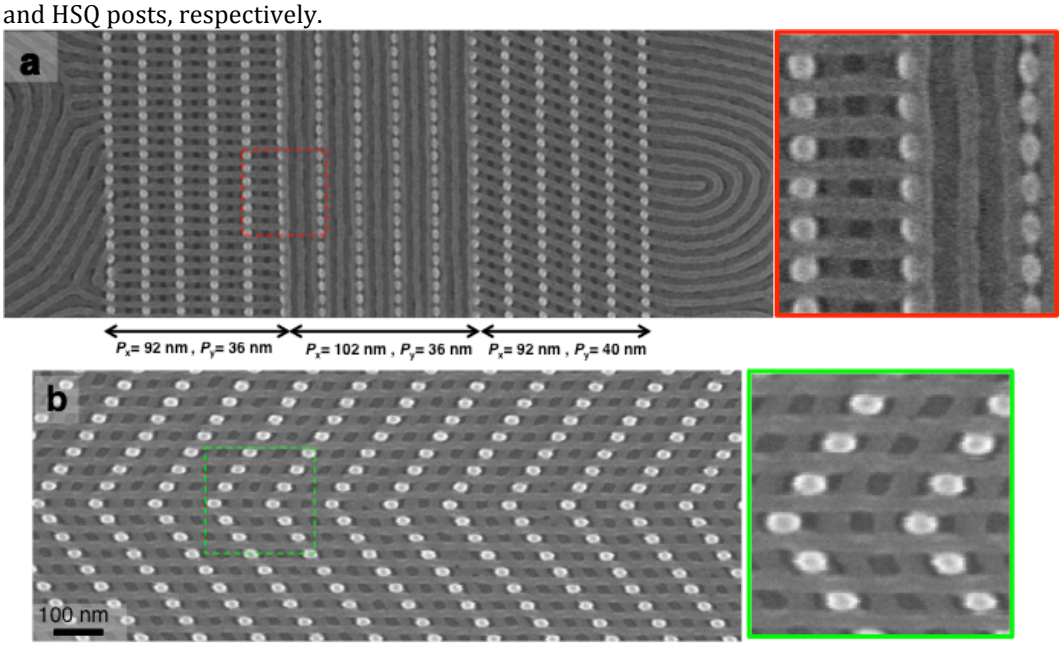

Figure 3: SEMs of locally controlled 3D structure fabricated by (a) altering the periodicity of the posts (b) changing the post arrangement. Bright grey and white colors represent ox-PDMS and HSQ posts, respectively.