Rule-based patterning of a multi-state system by block copolymer self-assembly

Hyung Wan Do[1], Hong Kyoon Choi[2], Jae-Byum Chang[2], Caroline A. Ross[2], and Karl K. Berggren[1]
[1]*Department of Electrical Engineering and Computer Science,*
[2]*Department of Materials Science and Engineering,*
Massachusetts Institute of Technology, Cambridge, MA, 02139
hwdo@mit.edu

Lithographic confinement can direct the self-assembly of block copolymers to achieve nanoscale patterns with high order.[1,2] Previously, we have demonstrated that block copolymer cylinders can form a multi-state system with ladder-shaped structures rather than a one-state system with concentric rings inside a topographic polygonal confinement when bending angle of the confinement is 90° or above. In this work, we describe a design rule to determine the individual state by controlling alignment direction of the ladder-shaped structures. This work could lead to a new pattern generation technique that enables higher-throughput pattern generation.

We fabricated topographic templates consisting of square cells with one to four openings using electron-beam lithography of HSQ resist. Figures 1a-1e show five different types of the square confinements. The templates were chemically functionalized with a hydroxyl-terminated PS brush. Next, 45.5 kg/mol PS-*b*-PDMS block copolymer was spin coated on to the templated substrate and solvent annealed using a 5:1 mixture of toluene and heptane at room temperature for 5 h. The top PDMS surface and PS matrix were removed using CF_4 and O_2 plasma treatment, respectively. Without any openings, ladder-shaped block copolymer patterns were formed inside the square cell with equal probability of horizontal and vertical alignment. When one or more openings were introduced, the alignment directions were controlled by a majority rule determined by the number of horizontal and vertical openings. Figures 1f-1j show the ladder-shaped structures with measured probability of horizontal alignment denoted as 0 state.

Using the five types of design rules, we were able to fabricate an array of connected binary states where the two states are defined as horizontally (0 state) and vertically (1 state) aligned ladder-shaped structures. Two examples of complex 4 by 4 binary patterns each composed of 16 independently controlled binary states are shown in Figure 2. Figures 2a and 2d show a diagram of desired binary states. Templates were fabricated by rule-based design (Figures 2b and 2e). After block copolymer self-assembly, binary states formed inside the template matched the desired binary patterns (Figures 2c and 2f).

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[1] J. Y. Cheng et al., *Nat. Materials.* **3**, 823-828 (2004).
[2] Y. S. Jung et al., *Nano Lett.* **8**, 2975-2981 (2008).

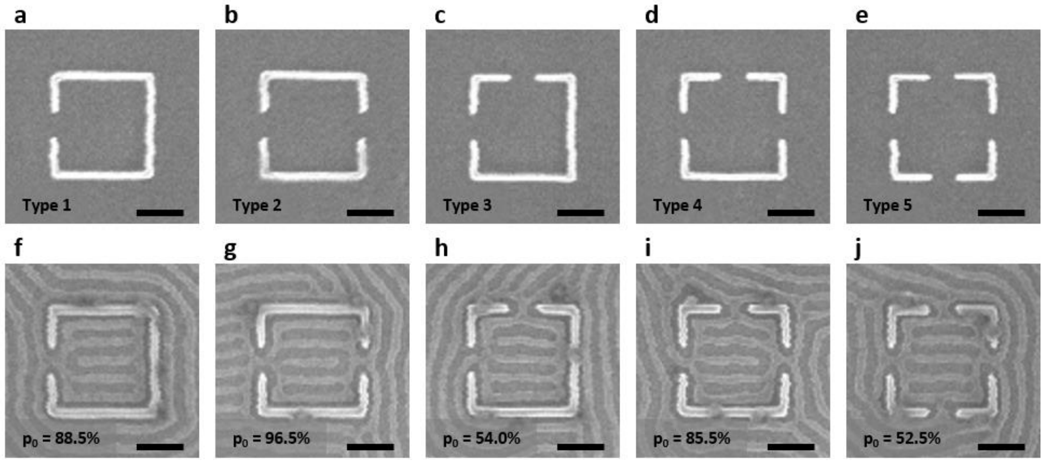

Figure 1: SEM images of square confinement with openings and resulting block copolymer patterns. Probability of 0 state for each type was measured. Binary states were determined by a design rule based on majority rule. (a,f) Square confinement with one opening. Preferential horizontal alignment (0 state) was observed. (b,g) Square confinement with two openings on non-adjacent sides. Stronger preferential horizontal alignment was observed. (c,h) Square confinement with two openings on adjacent sides. Alignment in both directions was observed with equal probability. (d,i) Square confinement with three openings. Preferential horizontal alignment was observed. (e,j) Square confinement with four openings. Alignment in both directions was observed with equal probability. Scale bars, 200 nm.

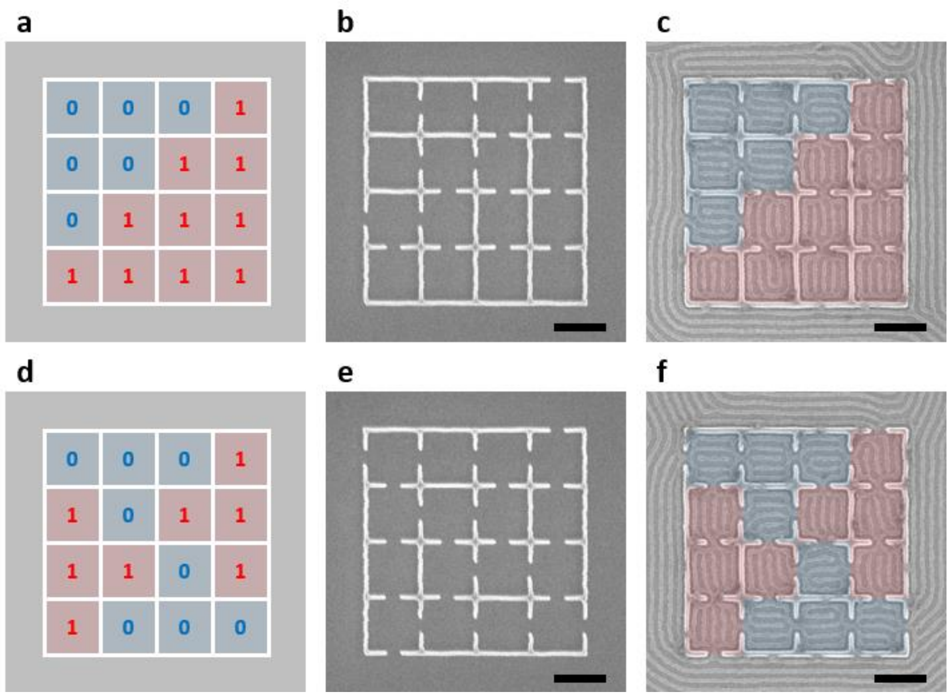

Figure 2: Fabrication of binary state arrays. (a,d) Diagram of desired 4 x 4 binary state arrays. (b,e) Templates fabricated by rule-based design. (c,f) Resulting block copolymer patterns matching the desired binary states. Red indicates 0 state and blue indicates 1 state. Scale bars, 200 nm.