Multi-Layer Block Copolymer Self-Assembled Structures Using Tilted Pillar Templates

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In this paper, we used an array of spaced and tilted pillars, prepared by electron-beam lithography of an inorganic resist to control orientation and alignment of a self-assembled cylindrical morphology of PS-b-PDMS in a multi-layer.

Figure 1 explains alignment control using tilted pillars. Tilted pillars are defined by polar and azimuthal angles. As already reported by our group, an array of carefully spaced and shaped pillars can control orientation and alignment in a monolayer of cylindrical microdomains. Use of tilted pillars can provide a 3D template, and thus be expected to control the orientation in a multi-layer of cylinders. In the case of $\alpha = 0°$ and $\beta = 60°$, the self-assembled cylinders should form a close packed arrangement with a triangular pattern in a side view.

In experiments, pillars of 70 nm length and a 60° tilt angle were fabricated by direct electron-beam lithography on a pre-tilted silicon wafer using hydrogen silsesquioxane (HSQ) as shown in Fig.2. The substrate with pillars was functionalized with a PDMS brush. PS-PDMS diblock copolymer with an overall molecular weight of 45.5 kg/mol, a volume fraction of PDMS f_{pdms} = 35.5 % was spin-coated onto the substrate to a film thickness of 60 nm and solvent-annealed. The natural period L_0 of the block copolymer was ~35 nm, and thus the film contains 2 layers of cylinders.

These experiments revealed that 60° tilted HSQ posts can control alignment of two layers of PS-b-PDMS cylinders. Figure 3 (a) shows that pillars tilted in the x-z plane with lattice parameters of L_x 105 nm and L_y 35 nm (expressed as (105, 35)), were able to align cylinders in the y axis, also observed in the case of (70, 35). Figure 3 (b) shows the case for (105, 70), in which the cylinders were mostly ordered in the tilting direction, also observed in the cases of (70, 70) and (35, 35), although this case resulted in a different alignment from our expectation as shown in Fig. 2. In both cases, we can observe that adjacent cylinders in both figures seem aligned in alternating layers as expected for a bilayer close packed cylinder array.

We also conducted experiments with vertical pillars with the same lattice parameters and film thickness, but most of these experiments were not effective in templating the cylinders, presumably because the vertical pillars frustrate the closepacking geometry. We conclude that appropriately tilted pillars can control multi-layers of self-assembled block copolymer microdomains.

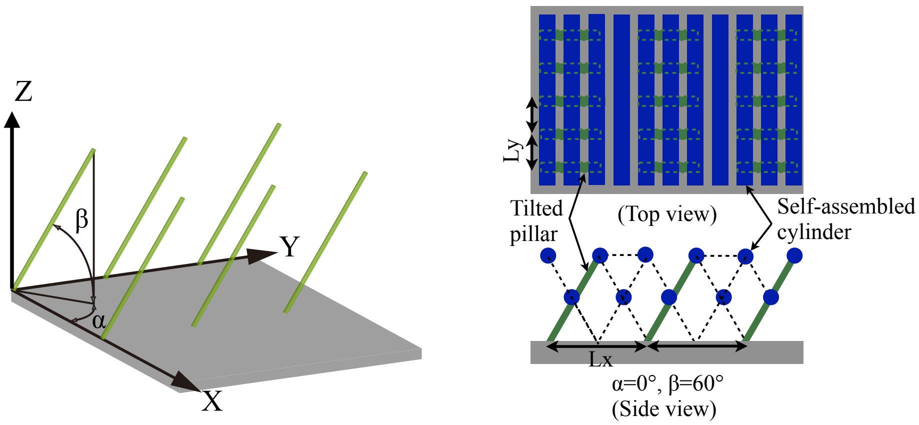

Figure 1: Alignment control of a cylindrical morphology of block copolymer using tilted pillars: Tilted pillars are defined by polar and azimuthal angles. When α=0 and β=60°, self-assembled cylinders are expected to be directed in a lattice as shown in the top-down view, and to be aligned in a multi-layer as shown in the side view.

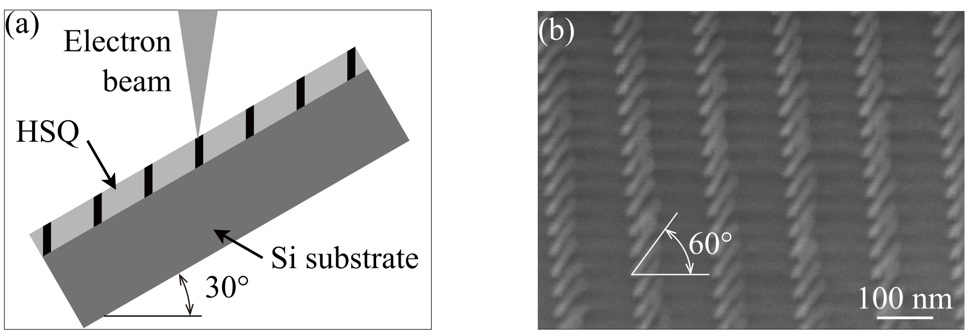

Figure 2: 60° tilting HSQ pillars: (a) The pillars were fabricated on a silicon wafer tilted at 30° using electron-beam lithography. (b) A 75° diagonal SEM micrograph of tilted HSQ pillars.

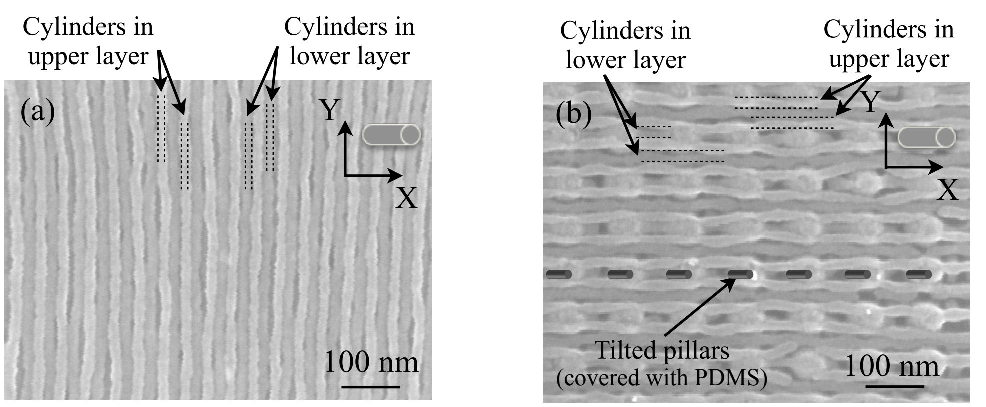

Figure 3: Demonstration of alignment control in a multi-layer of BCP cylinders using HSQ pillars tilted in the x-z plane at 60°: (a) HSQ pillars with L_x 105 nm and L_y 35 nm were able to align cylinders in the y axis. (b) HSQ pillars with L_x 105 nm and L_y 70 nm were able to align cylinders in the x axis. Adjacent cylinders in both figures were aligned in alternating layers.