ADVANCED MATERIALS

**Supporting Information**

for

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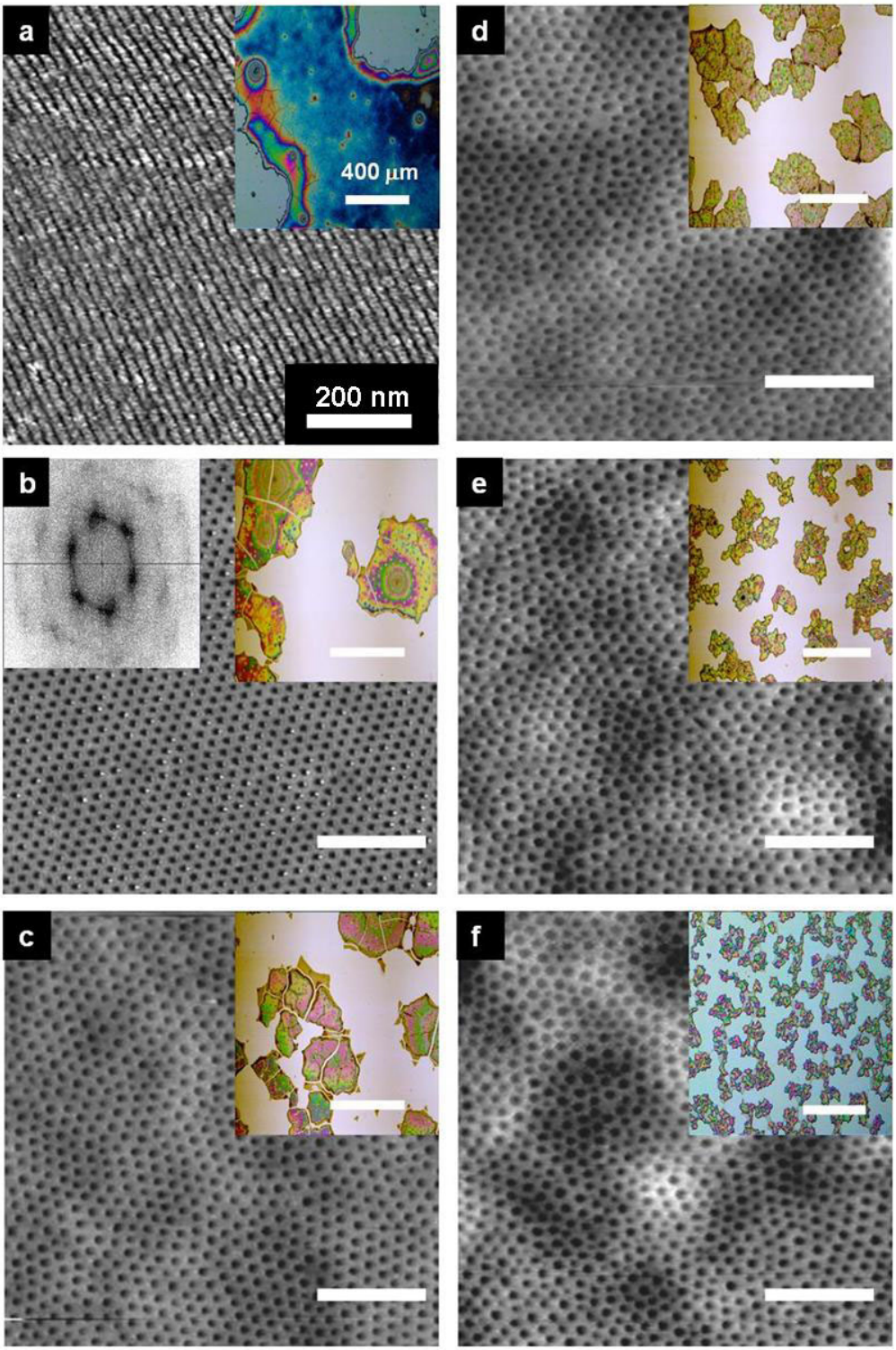
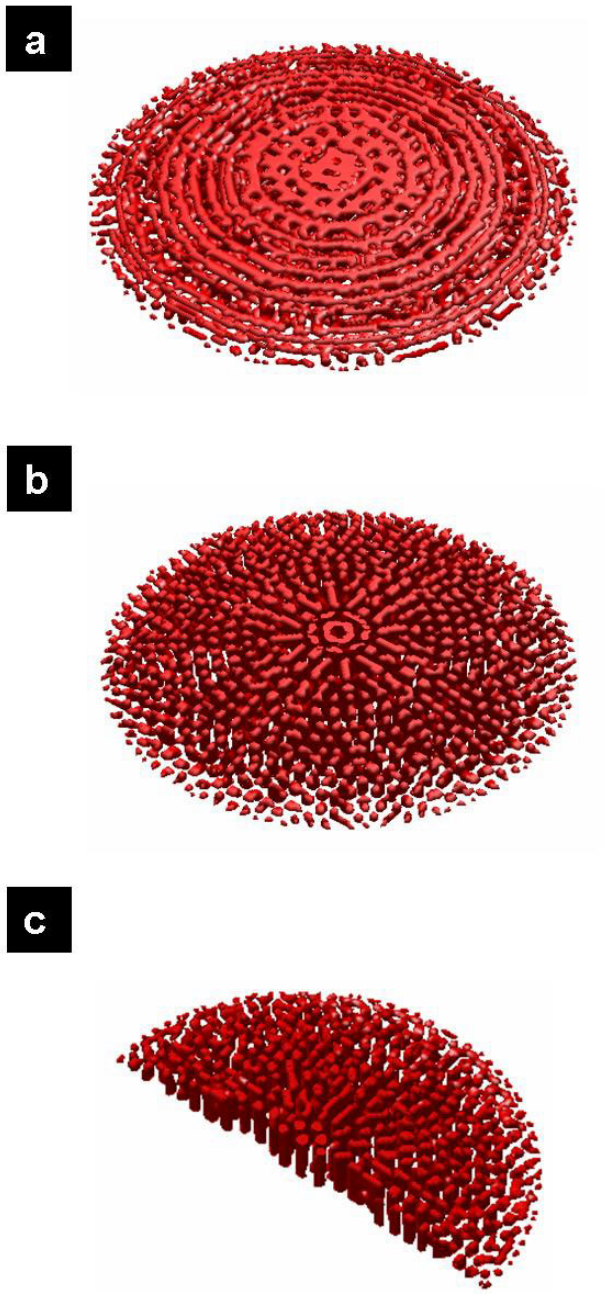
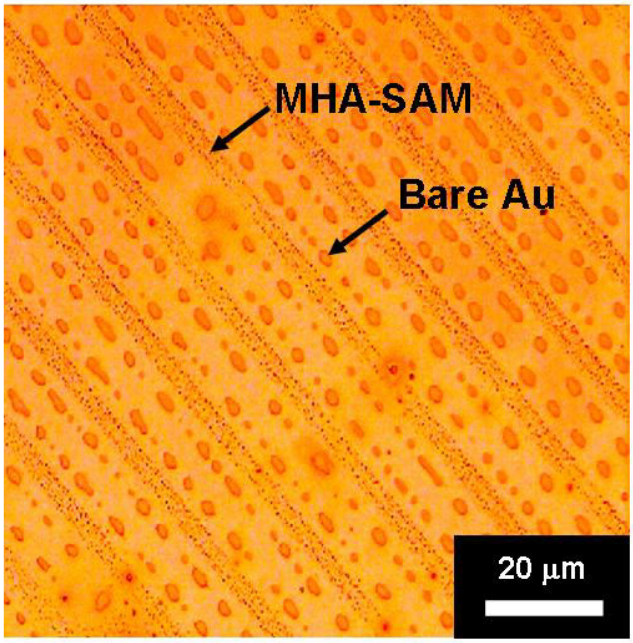



**S-1.** Tapping mode AFM images in height contrast of PS-*b*-PEO thin films prepared by benzene/water co-solvent treatment with various water fraction: (a) 0.0, (b) 0.05, (c) 0.10, (d) 0.15, (e) 0.28, and (f) 0.5. Quality of the solvent treated films is visualized in the inset of each AFM image by optical microscope. FFT image is shown in the inset of (b).




**S-2.** Simulated morphology of a diblock copolymer confined into a spherical cap equilibrated before (a) and after (b) solvent annealing with the parameters of $\varepsilon_{substrate\text{-}minor\ red\ block} = -0.002$, $\varepsilon_{air\text{-}minor\ red\ block} = -0.002$, $\varepsilon_{substrate\text{-}the\ other\ major\ block} = \varepsilon_{air\text{-}the\ other\ major\ block} = 0$. Red minor block corresponds to the simulated PEO block. The preferential interaction of PEO block with air induces the concentric rings of in-plane red cylinders equilibrated before solvent annealing which can make the cylinders ordered perpendicular to the substrate. (b) The subsequent solvent annealing transforms the in-plane cylinders into an ordered spherical structure similar to one observed in Figure 4d and 4e. In thermodynamically equilibrium, however, we have found that a structure with cylindrical domains perpendicular to the surface is more favorable as shown in the cross sectional view of the structure in (c).




**S-3.** An OM image of the micropatterned PS-*b*-PEO thin film with controlled dewetting. A 16-mercaptohexadecanoic acid (MHA) SAMs with hydrophilic carboxyl terminal group was micropatterned on Au surface with a PDMS mold having periodic lines with the width and period of 10 and 20 μm, respectively. Spin casting of the block copolymer solution results in a homogeneous thin film formation on a MHA patterned Au substrate due to both polar characteristic of bare Au and MHA surface. The following solvent annealing gave rise to a controlled dewetting structure of PS-*b*-PEO film where the larger domains were dominantly formed on bare Au regions while much smaller ones observed in MHA regions which are more hydrophilic.