Supplementary Information for

Microdomain Control in Block Copolymer-Based Supramolecular Thin Films through Varying Grafting Density of Additives

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1. In-situ Measurements of Film Thicknesses during Solvent Annealing



Figure S1. Film thicknesses as a function of time during solvent annealing: (a) A PS(20000)-*b*-P4VP(17000)(PDP)_{1.0} film originally with thickness of 117 nm increasing to 180 nm under low vapor pressure of chloroform. The swelling ration is about 1.5. (b) A PS(20000)-*b*-P4VP(17000)(DBSA)_{1.0} originally with thickness of 100 nm increasing to 240 nm under high vapor pressure of chloroform. The swelling ration is about 2.4. Note that the film was placed faceup on a stage in a sealed jar upon measurements.



2. AFM Image of PS-*b*-P4VP(PDP)_{0.8} supramolecular Thin Films

Figure S2. AFM images of a PS(20000)-*b*-P4VP(17000)(PDP)_{0.8} thin film, ~ 132 nm in thickness, showing a mixture of perpendicular and parallel PS cylinders. The *z* scale is 40 nm for height and 40° for phase.

3. TEM Images of PS-b-P4VP(DBSA) supramolecules



Figure S3. TEM images of PS-*b*-P4VP(DBSA)_{*x*} bulk samples: (a) x = 0.5 and (b) x = 1.0, both showing PS cylindrical microdomains in P4VP(DBSA) matrices.

4. AFM Images of PS-b-P4VP(PDP) Thin Film



Figure S4. AFM images of a PS(40000)-*b*-P4VP(5600)(PDP)_{2.5} thin film ~ 71 nm in thickness, showing perpendicular lamellae. The *z* scale is 10 nm for height and 60° for phase