Supporting Information

**Electrospun Smart Fabrics that display pH-responsive tunable wettability**

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**Experimental**

**Sample preparation:** The PS polymer used in this study was purchased from LG Chemical Inc. and was used as received. Other reagents and starting materials were purchased from Aldrich Chemical Inc. A 13 wt % solution of the PS homopolymer (Mw 43,000 g/mol, PDI = 1.7) was dissolved in a mixture of the solvents tetrahydrofuran (THF) and N,N-dimethylformamide (DMF) with a weight ratio of 1:4 under gentle stirring for at least 24 h at 60°C. The solution was subsequently cooled to room temperature prior to electrospinning. Under the electrostatic field conditions, the PS solutions were electrospun into fibrous webs using an electrospinning system equipped with a high voltage generator (HARb-1006, Matsusada), which was used to generate a potential difference. The needle tip-to-plate distance and the voltage were, respectively, 15 cm and 13 kV for the 13 wt % PS solution. These conditions yielded randomly oriented nanofibrous mats composed of highly porous fibers. The experiments were conducted at 30°C under a relative humidity of 60%. 2-(Diisopropylamino)ethyl methacrylate (DPAEMA, 97%) was purchased from Aldrich and was passed through an alumina base column for removing the inhibitor prior to use. 3-(Trimethoxysilyl)propyl methacrylate (TSPM, 98%) was used without further purification. α, α-azobisisobutyronitrile (AIBN) was recrystallized from methanol. A mixture of DPAEMA
(17.06 g, 80 mmol), TSPM (4.96 g, 20 mmol), and AIBN (0.16 g, 0.1 mmol) dissolved in anhydrous 2-methoxyethanol (150 g) was placed in a round-bottom flask. The solution was degassed by the freeze-thaw method, repeated three times. The sealed reaction flask was heated at 60°C for 24 h. The polymerized mixture was precipitated twice in a large volume of n-hexane, then vacuum-filtered. The solid powdery product was freeze-dried for 24 h under vacuum, then stored in a desiccator. The resulting polymers were dissolved in 2-methoxyethanol, and were dip-coated onto the PS nanofibrous mats. The fibrous webs were annealed at 80°C for 12 hours under humid condition (relative humidity of 60%).

**Characterization:** The fiber morphologies were imaged by field-emission scanning electron microscopy (Sigma, Carl Zeiss) and energy-dispersive X-ray spectroscopy (using a Thermo Noran System 7). X-ray photoelectron spectroscopy (XPS) was performed at the 4B1 beamline at the Pohang Accelerator Laboratory. Water contact angle (CA) measurements were conducted using a droplet shape analysis system (Krüss). The average CA value was obtained by measuring the same sample at different fiber positions. Buffered solutions with different pH values were used to investigate the pH-dependence of the wettability properties of the different substrates: pH 2, 0.2 mol L⁻¹ HCl; pH 12, 0.01 mol L⁻¹ NaOH.