

Supplementary Information for

**Smart surfaces with switchable superoleophilicity and
superoleophobicity in aqueous media: toward controllable
oil/water separation**

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Supplementary Information:

This file contains Supplementary Figures S1, S2, and S3 with legends.

S1. XPS spectrum of raw non-woven textile and the functionalized textile after deposition of silica nanoparticles and block copolymer grafting.

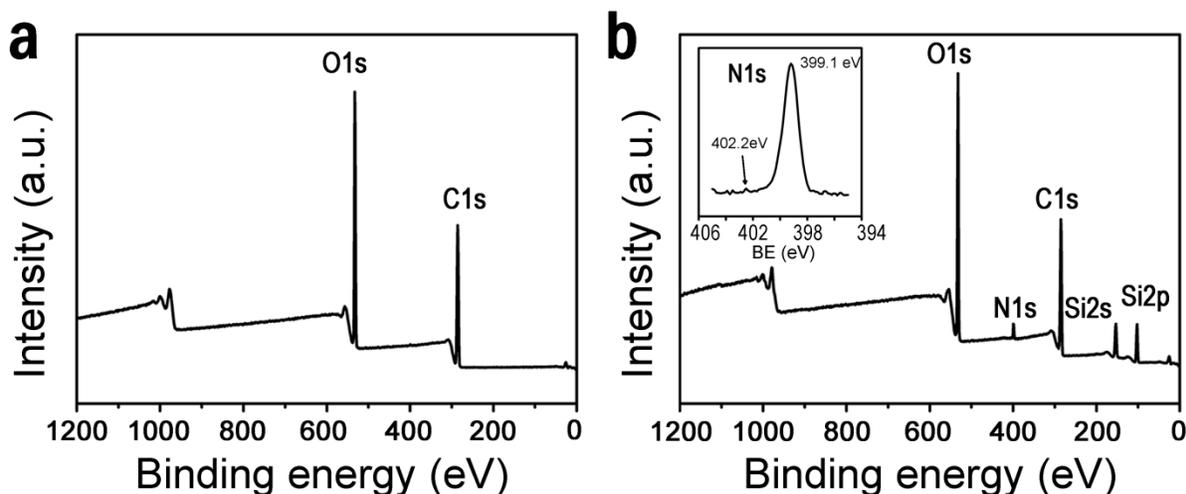


Figure S1. a. XPS spectrum of raw non-woven textile. **b.** XPS spectrum of the functionalized textile after deposition of silica nanoparticles and block copolymer grafting. The appearance of a nitrogen signal associated with P2VP confirmed the success of the grafting of the P2VP-*b*-PDMS block copolymer on the surface of the textile. Inset in **b** shows the N 1s XPS spectrum of the functionalized textile. The N 1s XPS spectrum of P2VP-*b*-PDMS grafted textile showed a strong peak located at 399.1 eV and a weak peak at 402.2 eV, which were assigned to the nitrogen of the neutral pyridine and that of the quaternized pyridine, respectively¹. The amount of quaternized pyridine was less than 1 %, as calculated from the peak area. This provided evidence for grafting the block copolymer via very small number of 2-vinylpyridine monomer units, and most of the pyridyl groups on the polymer chains remained unchanged after the grafting process, which could then move to the outmost surface upon protonation in the acidic water ($\text{pH} \leq 2.0$).

S2. Water contact angles on the functionalized textile as a function of pH of the applied water droplets.

As shown in Fig. S2, the functionalized textile can be wetted by a water droplet with $\text{pH} \leq 2.0$, and exhibits superhydrophilicity to these water droplets. For instance, when a water droplet with a pH of 1.0 was applied on the functionalized textile in air, it spread out completely within about 10 seconds and wetted the textile. After wetting with the acidic water ($\text{pH} \leq 2.0$), most of the pyridyl groups on the functionalized textile can be protonated, considering the $\text{p}K_a$ (3.6) of P2VP. While for water droplets with $\text{pH} \geq 3.0$, even after 15 min still sitting of the water droplets on the

textile surface, the functionalized textile exhibited a stable superhydrophobicity to these water droplets.

The hydrophilic transition of the P2VP blocks, which is induced by the protonation of their pyridyl groups, is controlled by the diffusion of protons from the bulk water droplets into the grafted block copolymer layer. The presence of the liquid-like hydrophobic PDMS blocks on the exterior of the grafted copolymers, together with the hierarchical structures on the textile, restricted the effective contact between the water droplets and the P2VP blocks as well as the protonation diffusion process of the P2VP. Therefore, although P2VP has a pK_a of 3.6, it is reasonable that only at a significantly low pH of the water droplets (high proton concentration), the P2VP could be protonated and changed into hydrophilic. In water with a higher pH ($pH \geq 3.0$), the non-wettable property and the presence of the liquid-like hydrophobic PDMS blocks on the exterior of the grafted copolymers prevented the contact between the water droplets and the P2VP blocks, and therefore almost no pyridyl group on the P2VP was protonated.

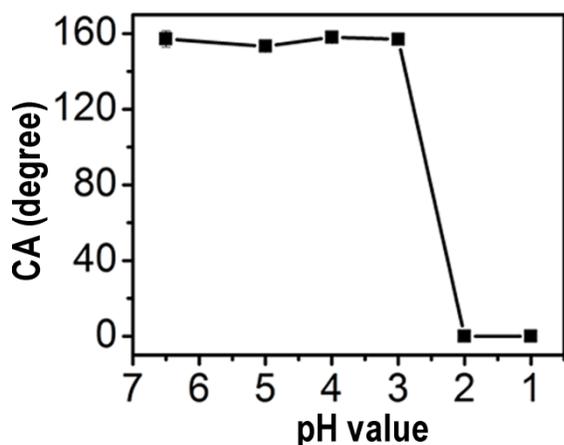


Figure S2. Water contact angles on the functionalized textile as a function of pH of the applied water droplets.

S3. Switchable superoleophilicity and superoleophobicity of the functionalized sponge in aqueous media with different pH.



Figure S3. a, Still images from video CA measurements of a DCE droplet on the functionalized sponge in water with a pH of 6.5. Upon contacting, the oil droplet was quickly sucked up by the sponge, indicating the superoleophilicity. **b**, Image of a DCE droplet applied on the surface of the functionalized sponge in water with a pH of 2.0, showing the superoleophobicity of the surface in the acidic water.

Reference.

1. Hayward, R. C., Chmelka, B. F. & Kramer, E. J. Template cross-linking effects on morphologies of swellable block copolymer and mesostructured silica thin films. *Macromolecules* **38**, 7768–7783 (2005).