

Supporting information

Robust fluorine-free superhydrophobic PDMS-Ormosil@fabrics for highly effective self-cleaning and efficient oil-water separation

*Chunyan Cao,^a Mingzheng Ge,^a Jianying Huang,^a Shuhui Li,^a Shu Deng,^a Songnan Zhang,^a Zhong Chen,^b Keqin Zhang,^{a,c} Salem S. Al-Deyab,^d and Yuekun Lai^{*a,c}*

^aNational Engineering Laboratory for Modern Silk, College of Textile and Clothing Engineering, Soochow University, Suzhou 215123, China.

^bSchool of Materials Science and Engineering, Nanyang Technological University, 50 Nanyang Avenue, 639798, Singapore.

^cResearch Center of Cooperative Innovation for Functional Organic / Polymer Material Micro/Nanofabrication, Soochow University, Suzhou, Jiangsu 215123, China.

^dDepartment of Chemistry, College of Science, King Saud University, Riyadh 11451, Saudi Arabia.

Corresponding author email: yklai@suda.edu.cn

Movie S1. Video of the selective absorption process of oil-water mixture (toluene dyed with oil red O) from water by the combination of porous sponge and superhydrophobic fabric (Me 35, 11.2 M, 1.2 wt%) with repulsive state.

Movie S2. Video of rapidly continuous absorption (d-f) process of oil-water mixture (toluene dyed with oil red O) from water by the combination of porous sponge and superhydrophobic fabric (Me 35, 11.2 M, 1.2 wt%) under the assistant of vacuum pump.

Movie S3. Video of the selective collection of oil from surfactant-free oil-in-water emulsions by the combination of porous sponge and superhydrophobic fabric (Me 35, 11.2 M, 2.8 wt%) with a dynamic stirring process.

Figure S1. SEM images showing the as-prepared Ormosil@fabric at Me 15 (a), 25 (b), 45 (c) and 65 (d) with the constant ammonium concentration of 11.2 M. Inset images are the corresponding image at low magnification.

Figure S2. SEM images of as-prepared Ormosil@fabric with different ammonium concentration at 5.6 M (a), 8.4 M (b), 11.2 M (c) with Me 35. (d), (e) and (f) are the corresponding high magnification images of (a), (b) and (c), respectively.

Figure S3. Optical images of unstable water droplet (dyed with methyl blue) on the highly hydrophobic Ormosil@fabric (a) and stable droplet on superhydrophobic PDMS-Ormosil@fabric.

Figure S4. The influence of PDMS concentration on contact angle (a), (insets are the corresponding CA images) and SEM images of modified with different concentration of PDMS (b-e).

Figure S5. Element mapping (a) and EDS spectrum (b) of Ormosil@fabric (Me35, 11.2M). Inset (b) is corresponding EDS scanning area.

Figure S6. Optical images of Ormosil@fabric (a) and PDMS-Ormosil@fabric (b) upon immersing in water solution.

Figure S7. SEM images and corresponding droplet images of cotton modified with different concentration (0.4, 1.2, 2.0 and 2.8 wt%) after immersing in pH=2 of HCl solution for 24 h.

Figure S8. SEM images and corresponding droplet images of cotton modified with different PDMS concentration (0.4, 1.2, 2.0 and 2.8 wt%) after immersing in pH=12 NaOH solution for 24 h.

Figure S9. Optical images of droplet on PDMS-Ormosil@cotton fabric and pristine cotton fabric.

To investigate the influence of methanol fraction (mol ratio of Methanol and MTMS) and ammonium concentration on the surface morphology, a series of experiments were systematically performed. It was found that methanol fraction and ammonium concentration had a great influence on the structure of Ormosil coating film. Figure S1 shows the SEM images of the as-prepared Ormosil structure obtained at different methanol fraction 15, 25, 45, 65 (denoted as Me15, Me25, Me45 and Me65) with the ammonium concentration of 11.2 M, respectively. Dense film were uniformly formed on the cotton fibre, while no obvious difference among Me 15-65 samples was observed in the low magnification (the inset images in Figure S1a-d). With the lower proportion of methanol (Me15, Me25), Ormosil clusters provide surfaces roughness with smaller pores (Figure S1a,b). As increasing mol fraction of methanol to 45 (Figure S1c), we could instinctively observe all continuous dense films had highly porous networks, and the film tended to have a structure with larger pores. However, with the methanol fraction further increased to 65 (Figure S1d), the increasing larger pores would gradually weaken the gel network and result in pore collapse, which are extremely unfavourable for constructing stable superhydrophobic surfaces.

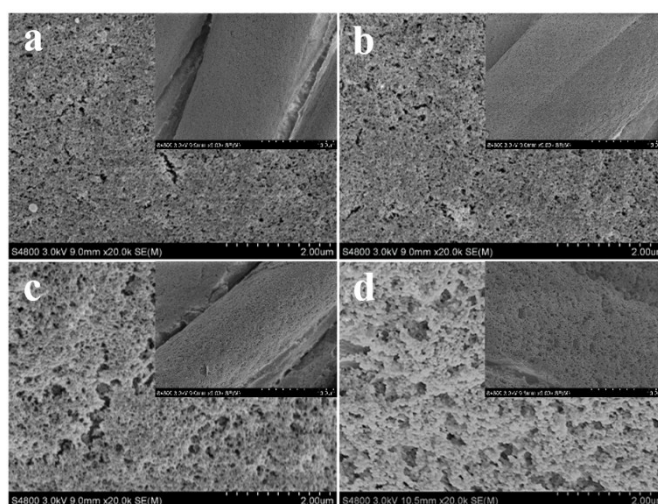


Figure S1. SEM images showing the as-prepared Ormosil@fabric at Me 15 (a), 25 (b), 45 (c) and 65 (d) with the constant ammonium concentration of 11.2 M. Inset images are the corresponding image at low magnification.

Figure S2a-c shows SEM images of the as-prepared Ormosil structure on cotton fabric obtained in the condition of absolute methanol fraction with different ammonium concentration. At a low ammonium concentration of 5.6 M (Figure S2a,d), only few micropores structures were sparsely coated on the fiber surfaces. As the concentration increased to 8.4 M (Figure S2b,e), higher density of 3D fractal-like structure with numerous loose micro-scale pores formed and almost entire cotton fibre surfaces are covered with highly porous network. When the ammonium concentration was further increased to 11.2 M (Figure S2c,f), remarkable density increment of Ormosil coating and well-defined microstructure pores have formed on the cotton yarn based on the increase of ammonium concentration. As a result, 3D-fractal-like structure with numerous micro-scale pores structure could be obtained under optimal parameters (methanol ratio: 35-45, ammonium concentration: 8.4 -11.2 M).

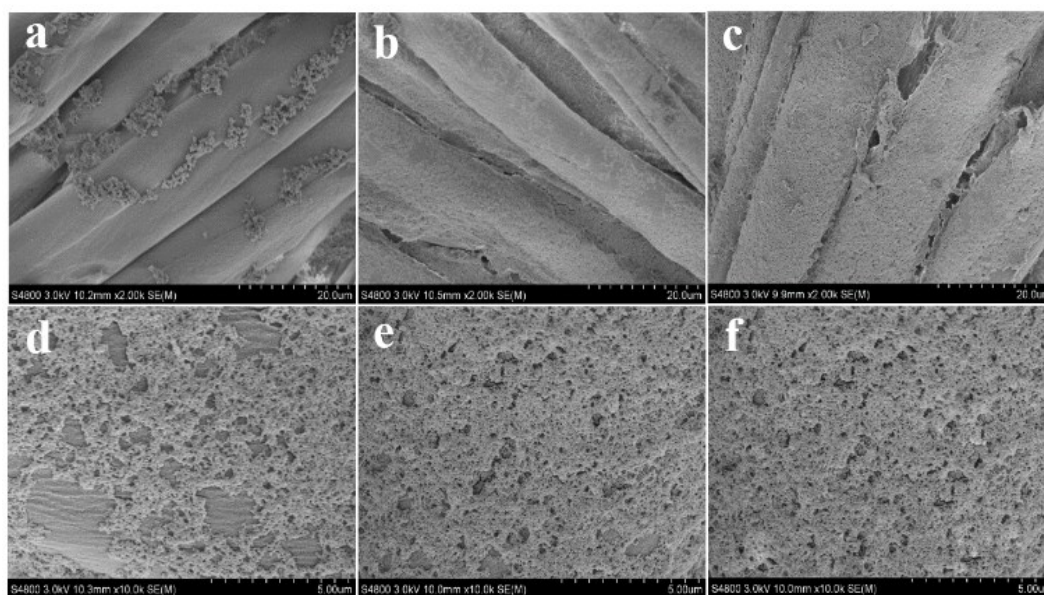


Figure S2. SEM images of as-prepared Ormosil@fabric with different ammonium concentration at 5.6 M (a), 8.4 M (b), 11.2 M (c) with Me 35. (d), (e) and (f) are the corresponding high magnification images of (a), (b) and (c), respectively.

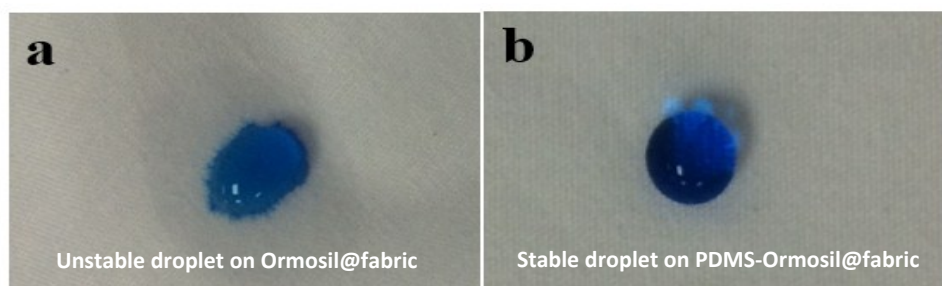


Figure S3. Optical images of unstable water droplet (dyed with methyl blue) on the highly hydrophobic Ormosil@fabric (a) and stable droplet on superhydrophobic PDMS-Ormosil@fabric.

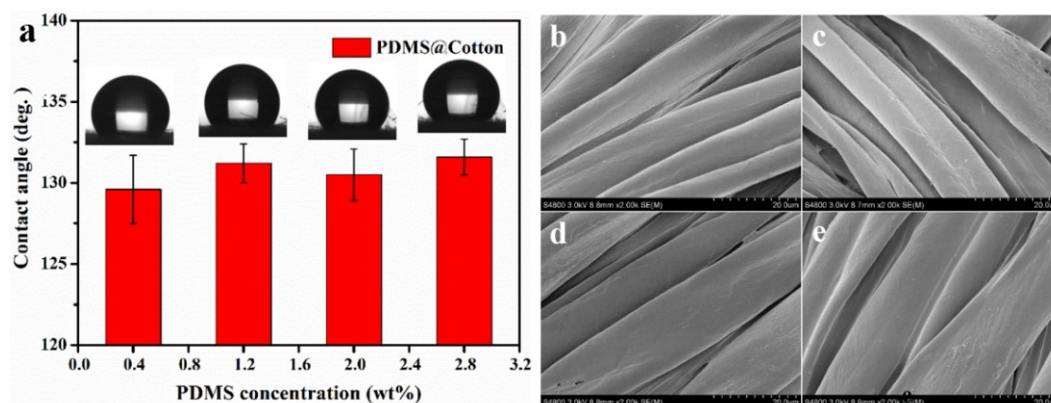


Figure S4. The influence of PDMS concentration on contact angle (a), (insets are the corresponding CA images) and SEM images of modified with different concentration of PDMS (b-e).

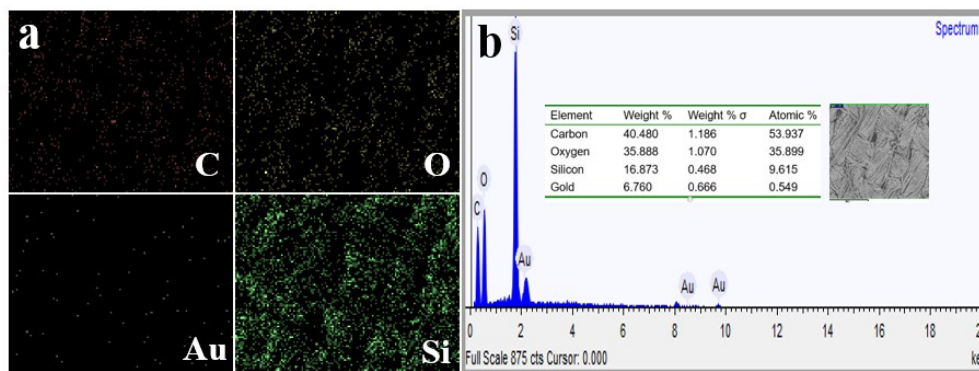


Figure S5. Element mapping (a) and EDS spectrum (b) of Ormosil@fabric (Me35, 11.2M). Inset (b) is corresponding EDS scanning area.

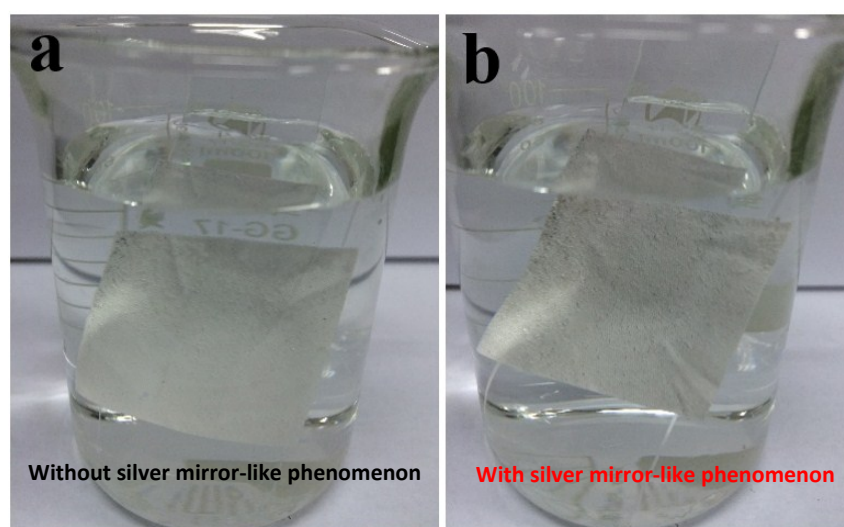


Figure S6. Optical images of Ormosil@fabric (a) and PDMS-Ormosil@fabric (b) upon immersing in water solution.

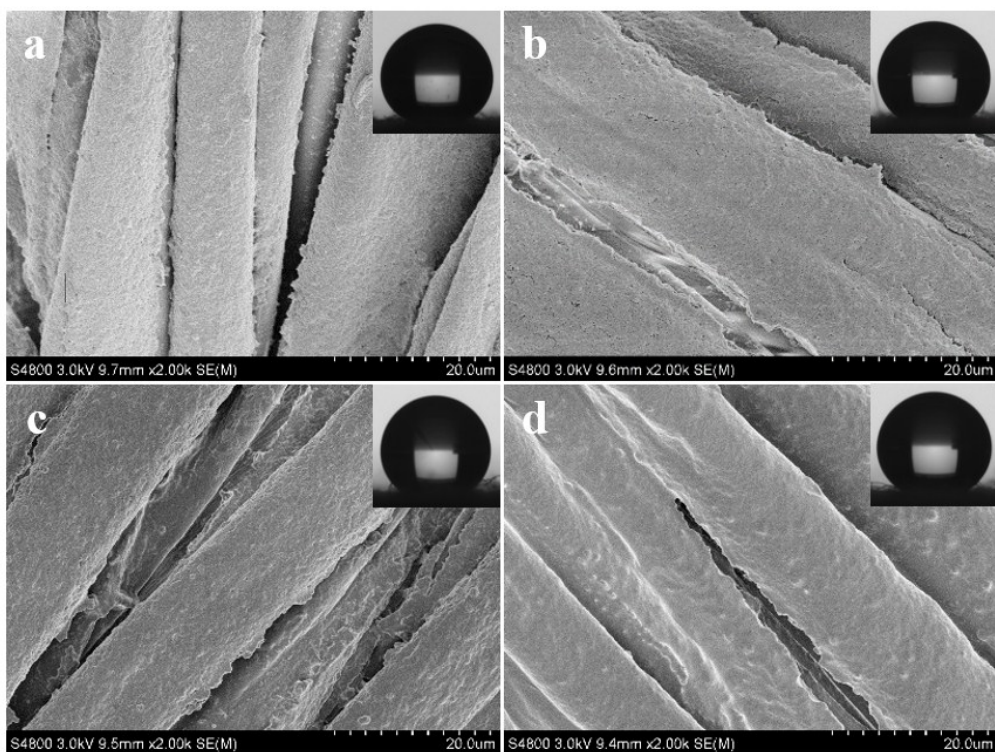


Figure S7. SEM images and corresponding droplet images of cotton modified with different concentration (0.4, 1.2, 2.0 and 2.8 wt%) after immersing in pH=2 of HCl solution for 24 h.

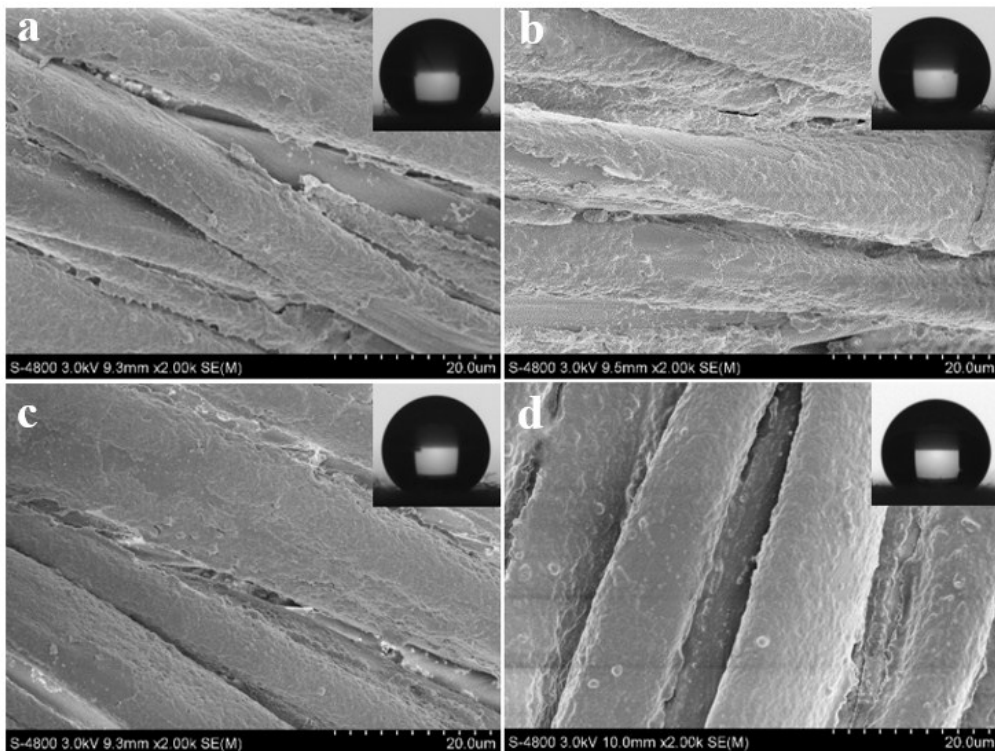


Figure S8. SEM images and corresponding droplet images of cotton modified with different PDMS concentration (0.4, 1.2, 2.0 and 2.8 wt%) after immersing in pH=12 NaOH solution for 24 h.

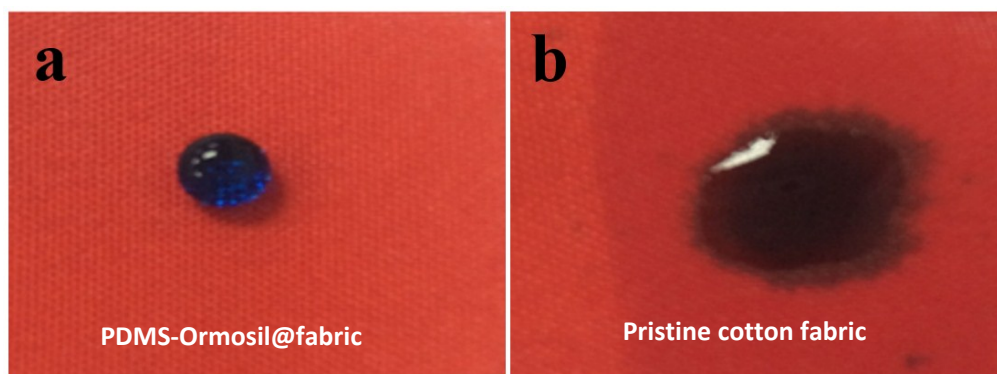


Figure S9. Optical images of droplet on PDMS-Ormosil@ cotton fabric and pristine cotton fabric.

As depicted in Figure S9, PDMS-Ormosil composite layer coated cotton fabric (Fig. S9a) shows no difference in color compared with pristine cotton fabric (Fig. S9b), convincingly confirming the high transparency of the PDMS-Ormosil coating layer. This is very beneficial for the retention of the original color expression on source colorful fabric.