## Electronic Supplementary Information

Interfacial assembly of durable superhydrophobic polyurethane sponge with "scalelike" structures for efficient oily emulsions separation

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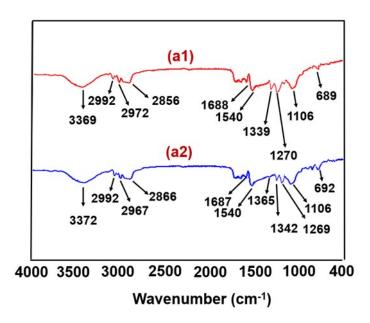


Fig. S1. FTIR spectra of (a1) PU/PDA/SiO<sub>2</sub>@H-CNTs sponge after adsorption of DCM, (a2) PU/PDA/SiO<sub>2</sub>@H-CNTs sponge after adsorption of xylene.

Two different densities of oils (DCM and xylene) were selected for oil removal studies. As shown in Fig. S1, there is no significant change in the main characteristic peaks of the separated sponges of two types oils. The FTIR results demonstrated that

the sponge after oil removal studies have no obvious impact on the composition of sponges. This can be explained by the fact that the oil/water separation process did not affect the chemical composition of the sponge surface, and the prepared sponge has prominent chemical stability.

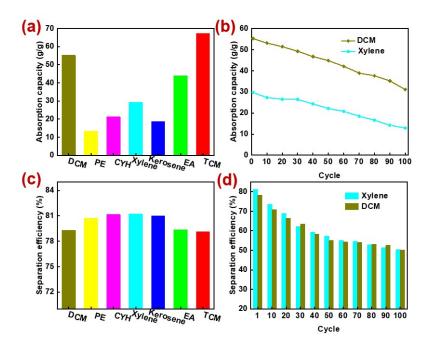


Fig. S2. (a) PU/SiO<sub>2</sub>@H-CNTs sponge's absorption capacity for various oils, (b)
PU/SiO<sub>2</sub>@H-CNTs sponge's cycle absorption capacity for xylene and DCM. (c)
PU/SiO<sub>2</sub>@H-CNTs sponge's separation efficiency for various oils, (d) PU/SiO<sub>2</sub>@H-

CNTs sponge's cycle separation efficiency for xylene and DCM.

Besides, we have conducted leaching experiments by gravimetry to conclude quantitatively. PU/SiO<sub>2</sub>@H-CNTs sponge without modification of dopamine was prepared using the same preparation process of PU/PDA/SiO<sub>2</sub>@H-CNTs sponge. As shown in Fig. S2 a and b, there is a significant decrease in adsorption capacity and separation efficiency. Meanwhile, the average adsorption capacity of the PU/SiO<sub>2</sub>@H-CNTs sponge decreased by 50.11% and the average separation efficiency can only be remained 62.97% after 100 separation cycles, which indicated that the recyclability of this sponge involved above was lower than PU/PDA/SiO<sub>2</sub>@H-CNTs sponge (Fig. S2 c and d). This was chiefly because that the SiO<sub>2</sub>@H-CNTs particles without dopamine constraints are prone to detachment on the surface of sponges after cycles, causing a decrease in absorption capacity and reusability. Besides, the H-CNTs weaken the needle demulsification process of the sponge surface, leading to a decrease in oil/water separation efficiency. Therefore, surface modification of sponge with dopamine is an important processing step.

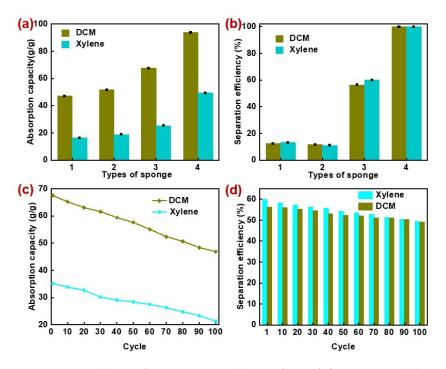


Fig. S3. PU sponge (1), PU/PDA sponge (2), PU/PDA/SiO<sub>2</sub> sponge (3) and PU/PDA/SiO<sub>2</sub>@H-CNTs sponge's (4) (a) absorption capacity, (b) separation efficiency. (c) PU/PDA/SiO<sub>2</sub> sponge's cycle absorption capacity for DCM and xylene, (d) PU/PDA/SiO<sub>2</sub> sponge's cycle separation efficiency for DCM and xylene.

To further investigate the role of H-CNTs, we prepared sponges using the same process without adding H-CNTs. Meanwhile, to simplicity the drawing, we marked PU sponge, PU/PDA sponge, PU/PDA/SiO<sub>2</sub> sponge and PU/PDA/SiO<sub>2</sub>@H-CNTs sponge as 1, 2, 3, 4, respectively. As evidenced in Fig. S3 a and b, the PU/PDA/SiO<sub>2</sub>@H-CNTs sponge's absorption capacity and separation efficiency were significantly higher than that of PU sponge, PU/PDA sponge and PU/PDA/SiO<sub>2</sub> sponge. This fully demonstrated the needle demulsification and diversion of H-CNTs, as well as the role of constructing hierarchical micro/nano structures to increase adsorption capacity. Besides, the adsorption capacity of the PU/PDA/SiO<sub>2</sub> sponge decreased by 34.86% and the separation efficiency can only be remained 84.82% after 100 separation cycles, which evidenced that the recyclability of the mentioned sponge was lower than PU/PDA/SiO<sub>2</sub>@H-CNTs sponge (Fig. S3 c and d). Therefore, the addition of H-CNTs is beneficial to improve the oily emulsion separation efficiency of the prepared sponge.