## Robust Superhydrophilicity and Under-water Superoleophobicity Cellulose Sponge for Highly Effective Oil/water Separation

Gang Wang <sup>a, b, †</sup>, Yi He <sup>a, †</sup>, He Wang, <sup>a</sup> Lin Zhang <sup>a</sup>, Quanyao Yu <sup>a</sup>, Shusen Peng <sup>a</sup>, Xuedong Wu <sup>a</sup>, Tianhui Ren <sup>b,\*</sup>, Zhixiang Zeng <sup>a,\*</sup> and Qunji Xue <sup>a</sup>

Dedication († These authors contributed equally to this work.)

<sup>a</sup> Key Laboratory of Marine Materials and Related Technologies, Zhejiang Key Laboratory of Marine Materials and Protective Technologies, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo, 315201, P. R. China.

E-mail: zzx1572000@aliyun.com

<sup>b</sup> Laboratory of Green Organic and Oil Chemistry, School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, Shanghai, 200240, P. R. China.

E-mail: thren@sjtu.edu.cn

Keywords: (superoleophobicity, superhydrophilicity, cellulose sponge, antifouling, oil/water separation)



**Figure S1.** The infrared spectra of cellulose powder and cellulose sponge. The infrared spectrum of each peak is similar before and after the regeneration which indicated that the cellulose didn't have derivative reaction and kept good hydrophilic nature.



**Figure S2.** SEM images of the cellulose sponge. (a) The cellulose sponge surface with nanopore. (b) The cellulose sponge surface with macropore. (c) The cross-sectional SEM image of cellulose sponge.



**Figure S3.** (a)The separation efficiency of cellulose sponge surface with nanopore and macropore. (b) The flux of cellulose sponge surface with nanopore and macropore. (c) Droplet size distribution of filtrate by DLS after separation by nanopore surface. (d) Droplet size distribution of filtrate by DLS after separation by macropore surface.

In order to validate the top-layer with nanopore of the cellose sponge on the oil/water separation, we cut out the top-layer and test its oil/water separation efficiency, flux and the droplet size distribution of filtrate. Compared with Figure S2a, after cutting out the top-layer, there are discrete macropores of  $5~20\mu m$  in diameter disorderly distributed on the surface. The microemulsion with the sizes lower than  $20\mu m$  can easily penetrate the macropores (Figure S3b). Compared to the sponge with nanopore (Figure S3a), the cellulose sponge with macropore has relatively lower oil/water separation efficiency. Nevertheless, the separation efficiency of 96.42% is higher than that of the same pore size membrane. The honeycombed body (Figure S2c) with the pore size of  $0~10\mu m$  of the sponge will block a part of oil droplets penetrate the sponge and Figure S3d also illustrates this point.



Figure S4. The bending performance of the cellulose sponge.



Figure S5. Optical image of the crooked cellulose sponge.



**Figure S6.** (a) Droplet size distribution of oil/water emulsion by DLS. (b) Droplet size distribution of filtrate by DLS.



**Figure S7.** Digital photos of the cellulose sponge. a) Digital photos of the dry cellulose sponge. b) Digital photos of the cellulose sponge wetted by water.

Movie S1 and S2. The effect of the water layer on the  $P_{breakthrough}$ .

In Movie S1 and Movie S2, two pieces of cellulose membrane was exactly the same with thickness of 0.14mm and pore size of 300nm. To imitate the Scheme 2, the

two membranes were dipped in the water. One of the membrane was waken out from water and fixed on the separation experiment setup with water immediately to imitate the oil supported by water layer (Movie S1). The other one was taken out from water and dried the water of the surface with filter paper to imitate the oil supported by air (Movie S2).