# **Supporting Information**

### Tannic acid encountering ovalbumin: a green and mild strategy for superhydrophilic

### and underwater superoleophobic modification of various hydrophobic membranes for

#### oil/water separation

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**Figure S1**. Size distribution of the oil droplets in the (a) soybean oil-in-water, (b) toluene-in-water, (c) hexane-in-water, and (d) gasoline-in-water emulsions.

Note that, for soybean oil-in-water emulsion, the size of some soybean oil droplets is larger than 10  $\mu$ m, which is out of the measuring range. Combining the data from the optical microscopy observation (BM-60XCC), the droplet sizes of the soybean oil droplets are in the range of 1-30  $\mu$ m.



Figure S2. SEM images (low resolution) of the (a) PVDF, (b) PVDF-OVA, and (c) PVDF-OVA-TA membranes.



Figure S3. SEM images of the (a) PTFE, (b) PTFE-OVA, and (c) PTFE-OVA-TA membranes.



Figure S4. SEM images of the (a) fabric, (b) fabric-OVA, and (c) fabric-OVA-TA membranes.



Figure S5. SEM images of the (a) copper mesh, (b) copper mesh-OVA, and (c) copper mesh-OVA-TA.



**Figure S6** The pore size distribution of the (a) pristine PVDF, (b) PVDF-OVA-TA, (c) PTFE, and (d) PTFE-OVA-TA membranes.



Figure S7 EDX mapping of the top surface of PVDF, PVDF-OVA, and PVDF-OVA-TA membranes, respectively.

For the PVDF-OVA, new elements such as O and N are detected, which can confirm the formation of OVA layer on the PVDF-OVA membrane. For the PVDF-OVA-TA, uniformly distributed N element is also observed, indicating the OVA still exists on membrane surface after treated by TA. These results indicate OVA and TA coatings are distributed uniformly on membrane surface.



**Figure S8**. TGA of (a) PVDF, PVDF-OVA, PVDF-OVA-TA, and (b) PTFE, PTFE-OVA, and PTFE-OVA-TA membranes (from room temperature to 800 °C at a heating rate of 10 °C min<sup>-1</sup> under nitrogen atmosphere).

As shown in **Figure S8**, for the pristine PVDF membrane, almost no weight loss was observed before 350°C, while sharply decrease of weight is observed when the temperature is higher than 350°C, which should be due to the decomposition of the PVDF at higher temperature.<sup>1</sup> At 350 °C, the weight loss of PVDF, PVDF-OVA, and PVDF-OVA-TA membranes is about 0.7 %, 2.4 %, and 3.4 % of the initial membranes, respectively. The increased weight loss (from 0.7 % to 2.4 %; from 0.7 % to 3.4 %) should be due to the decomposition of OVA and OVA-TA coatings on membrane surface. According to the results, it seems that

the weight ratio of OVA and OVA-TA on the membrane surface should be about 1.7% (2.4%-0.7%) and 2.7% (3.4%-0.7%), respectively. However, it should be noted that the OVA and OVA-TA may not be completely decomposed, and some of them may be carbonized when the temperature is above 250 °C. So it is hard to obtain the accurate weight ratio of OVA and OVA-TA on membrane, but we can speculate the real weight ratio of OVA and OVA-TA on PVDF membrane should be higher than 1.7% and 2.7%.

Similarly, for PTFE membranes, according to the TGA results, we can also estimate the weight ratio of OVA and OVA-TA on the PTFE membranes is at least 0.7 % and 1.5 %.

Table S1 The adsorption of OVA on different hydrophobic membranes.

Membranes	PVDF	PTFE	Fabric	Copper mesh
Adsorption capacity	86.6 μg/cm <sup>2</sup>	97.8 μg/cm <sup>2</sup>	59.6 μg/cm <sup>2</sup>	12.6 μg/cm <sup>2</sup>

Note: The pH value of the used ultrapure water is about 6.6-6.8, which should be due to the dissolved carbon dioxide.

#### Reference

1 F. Liu, N. A. Hashim, Y. Liu, M. R. Moghareh Abed, K. Li, J. Membr. Sci., 2011, 375, 1-27.