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

Zhenxing Wang,a Shengqiang Ji,a Jin Zhang, b Qixian Liu,a Fang He,a Shaoqin Peng,a and Yuexiang Li*a

a Department of Chemistry, Nanchang University, Nanchang 330031, P. R. China,
*E-mail: liyx@ncu.edu.cn
b Institute of Carbon Materials Science, Shanxi Datong University, Datong, Shanxi Province, 037009, PR China

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Video 5: Soybean oil (dyed red) could bounce off the Fabric-OVA-TA surface (underwater).

Video 6: Soybean oil (dyed red) could bounce off the Copper mesh-OVA-TA surface (underwater).

Video 7: The excellent underwater anti-oil-adhesion performance of the as prepared PVDF-OVA-TA.

Video 8: Gravity driven oil/water separation via the Fabric-OVA-TA membrane. The water was dyed blue, while the oil was dyed red.
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 μm, 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 μm.

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⁻¹ 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. 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.

<table>
<thead>
<tr>
<th>Membranes</th>
<th>PVDF</th>
<th>PTFE</th>
<th>Fabric</th>
<th>Copper mesh</th>
</tr>
</thead>
<tbody>
<tr>
<td>Adsorption capacity</td>
<td>86.6 μg/cm²</td>
<td>97.8 μg/cm²</td>
<td>59.6 μg/cm²</td>
<td>12.6 μg/cm²</td>
</tr>
</tbody>
</table>

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**