

## A Robust Bilayer Nanofilm Fabricated on Copper Foam for Oil- Water Separation with Improved Performances

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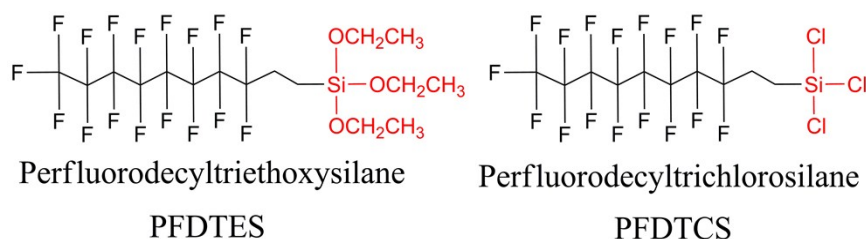


Fig. S1. The chemical structures of perfluorodecyltriethoxysilane (PFDTES) and perfluorodecyltrichlorosilane (PFDTCS).

**Video S3** Oil/ water separation experiments for light oils.

For light oils, the equipment used to separate oil/water mixture was gradient, and the mixture was poured **slowly** because the oil (such as toluene) is on the top of water.

The slope is helpful for oil-water separation [1-2].

**Video S4** Oil/ water separation experiments for heavy oils.

For heavy oils, the water is on the top of oil (such as tetrachloromethane), the mixture

was poured directly and the equipment is vertical.

**Table S1** The present chemical elements and their corresponding atomic concentrations (AC, calculated by formula 1) of TESPA powder, as well as the surfaces of the bare copper (Cu-Bare), the  $K_2S_2O_8$ -KOH oxidized copper (Cu-KK), the oxidized copper treated with TESPA (Cu-KK-TESPA), and the oxidized copper treated with TESPA and PFDTCS (Cu-KK-TESPA-PFDTCS), respectively.

Sample	Na1s (%)	Cu2p (%)	C1s (%)	O1s (%)	N1s (%)	S2p (%)	Si2p (%)	Cl2p (%)	F1s (%)
TESPA Powder	6.23	—	52.82	15.03	15.72	6.11	4.09	—	—
Cu- Bare	—	51.89	25.39	22.72	—	—	—	—	—
Cu- KK	—	42.73	30.52	26.75	—	—	—	—	—
Cu- KK-TESPA	—	37.71	26.53	25.24	7.43	1.94	1.15	—	—
Cu- KK-TESPA- PFDTCS	—	6.25	12.03	9.66	0.82	0.93	2.37	5.39	62.46

Although the numbers of N, S, and Si atoms in TESPA molecule are 4, 2, and 1 (see TESPA molecular structure in the first part or Scheme 1 in this paper), the obtained specific values in Table S1 for TESPA powder, Cu-KK-TESPA, and Cu-KK-TESPA-PFDTCS surfaces are not exact 4:2:1. Two reasons account for this phenomenon. First, XPS is just a semi-quantitative analytical technique. Second, S2s and Si2s are detectable in the survey scan, but their peak areas are not incorporated into the total areas of S and Si, because their high-resolution spectra cannot be given by individual elemental scans. Therefore, take sulphur (S) for instance, the actual content merely comprises S2p excluding S2s. However, significant tendency can be

seen. The atomic concentrations of N1s, S2p, and Si2p gradually decrease; the peak shapes of S2s and Si2s are smaller than that of S2p and Si2p, respectively. Compared with the XPS scan curves of the other four surfaces, the line of the Cu-TESPA-Heat is rough, and the C1s content is remarkably high. This is caused by heating TESPA SAM under atmospheric environment at an elevated temperature.

**Table S2** The present chemical states of Cu<sub>2p<sub>3/2</sub></sub> in the surfaces of the bare copper (Cu-Bare), the K<sub>2</sub>S<sub>2</sub>O<sub>8</sub>-KOH oxidized copper (Cu-KK), the oxidized copper treated with TESPA (Cu-KK-TESPA), as well as the oxidized copper treated with TESPA and PFDTCS (Cu-KK-TESPA-PFDTCS), respectively. The corresponding binding energy, full width at half maximum (FWHM) and atomic concentration (AC) of each chemical state are also given.

Sample	Chemical State	Binding Energy (eV)	FWHM	AC (%)
Cu-Bare	Cu(II)O	934.17	1.10	5.47
	Cu(0)/ Cu(I)O	932.58	1.10	94.53
Cu-KK	Cu(II)O	934.82	2.15	41.65
	Cu(0)/ Cu(I)O	933.30	2.10	58.35
Cu-KK-TESPA	Cu(II)O	934.78	1.70	23.86
	Cu(0)/Cu(I)	932.87	1.70	76.14
Cu-KK-TESPA-PFDTCS	Cu(II)O	934.95	1.90	34.66
	Cu(0)/Cu(I)	932.89	1.90	65.34

**Table S3** The present chemical states of S2p in TESPA powder, the K<sub>2</sub>S<sub>2</sub>O<sub>8</sub>-KOH

oxidized copper treated with TESPA (Cu-KK-TESPA), as well as the oxidized copper treated with TESPA and PFDTCS (Cu-KK-TESPA-PFDTCS), respectively. The corresponding binding energy, full width at half maximum (FWHM) and atomic concentration (AC) of each chemical state are also given.

Sample	Chemical State	Binding Energy (eV)	FWHM	AC (%)
TESPA Powder	SO <sub>3</sub> <sup>2-</sup> 2p <sub>1/2</sub>	168.32	1.00	1.53
	SO <sub>3</sub> <sup>2-</sup> 2p <sub>3/2</sub>	167.12	1.00	3.07
	SH2p <sub>1/2</sub>	162.93	1.00	9.83
	SH2p <sub>3/2</sub>	161.73	1.00	19.67
	S <sup>-</sup> 2p <sub>1/2</sub>	162.36	1.00	21.97
	S <sup>-</sup> 2p <sub>3/2</sub>	161.16	1.00	43.93
Cu-KK-TESPA	SO <sub>4</sub> <sup>2-</sup> 2p <sub>1/2</sub>	169.44	1.40	4.73
	SO <sub>4</sub> <sup>2-</sup> 2p <sub>3/2</sub>	168.24	1.40	9.45
	SS2p <sub>1/2</sub>	165.21	1.40	4.62
	SS2p <sub>3/2</sub>	164.01	1.40	9.23
	SCu2p <sub>1/2</sub>	163.60	1.40	23.99
	SCu2p <sub>3/2</sub>	162.40	1.40	47.98
Cu-KK-TESPA-PFDTCS	SO <sub>4</sub> <sup>2-</sup> 2p <sub>1/2</sub>	169.06	1.40	9.59
	SO <sub>4</sub> <sup>2-</sup> 2p <sub>3/2</sub>	167.86	1.40	19.17
	SS2p <sub>1/2</sub>	165.80	1.40	23.75
	SS2p <sub>3/2</sub>	164.60	1.40	47.49

**Table S4** The present chemical states of Si2p in TESPA powder, the K<sub>2</sub>S<sub>2</sub>O<sub>8</sub>-KOH oxidized copper treated with TESPA (Cu-KK-TESPA), as well as the oxidized copper

treated with TESPA and PFDTCS (Cu-KK-TESPA-PFDTCS), respectively. The corresponding binding energy, full width at half maximum (FWHM) and atomic concentration (AC) of each chemical state are also given.

Sample	Chemical State (Structures in Fig. 13d)	Binding Energy (eV)	FWHM	AC (%)
TESPA Powder	Structure 1	101.62	1.43	100
Cu- KK-TESPA	Structure 2	103.08	1.30	14.58
	Structure 3-6	102.23	1.30	85.42
Cu- KK-TESPA- PFDTCS	Si-Cl	104.79	1.10	6.55
	Structure 2	103.35	1.10	40.12
	Structure 3-6	102.44	1.10	53.33

## References

- [1] Weifeng Zhang, Na Liu, Yingze Cao, Yuning Chen, Liangxin Xu, Xin Lin, and Lin Feng. A Solvothermal Route Decorated on Different Substrates: Controllable Separation of an Oil/Water Mixture to a Stabilized Nanoscale Emulsion, *Advanced Materials*, 27 (2015) 7349-7355.
- [2] Chunai Dai, Na Liu, Yingze Cao, Yuning Chen, Fei Lua and Lin Feng. Fast formation of superhydrophobic octadecylphosphonic acid (ODPA) coating for selfcleaning and oil/water separation, 10 (2014) 8116-8121.