Crude-Oil-Repellent Membranes by Atomic Layer Deposition: Oxide Interface Engineering

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Oxide	Precursor A	Precursor B
ZnO	Diethyl zinc (DEZ)	H ₂ O
Al_2O_3	Trimethyl aluminum (TMA)	H_2O
TiO_2	Titanium tetrachloride (TTC)	H_2O
SnO_2	Tetrakis(dimethylamino) tin(IV) (TDMASn)	H_2O

Table S1 ALD precursors used for the different oxides.

Oxide	ALD cycles	Thickness (nm)*	Growth rate (Å/cyc)
ZnO	58	9.8 ± 0.13	1.69
Al_2O_3	77	9.5 ± 0.15	1.23
TiO ₂	100	10.6 ± 0.12	1.06
SnO ₂	78	10.2 ± 0.11	1.28

Table S2 ALD cycle numbers, growth rate, and thickness of different oxides

* Thickness of oxide layers measured on silicon wafers, which may differ from that on membranes.

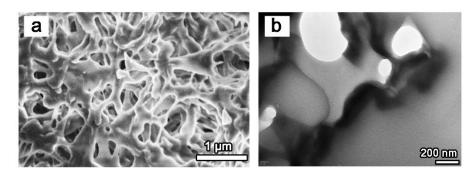


Figure S1. a) SEM and b) TEM images of the nascent PVDF membrane

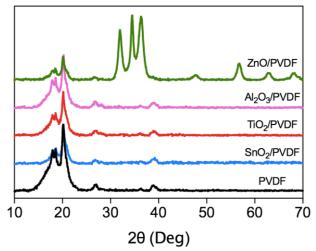


Figure S2. XRD spectra of nascent and oxide-coated membranes

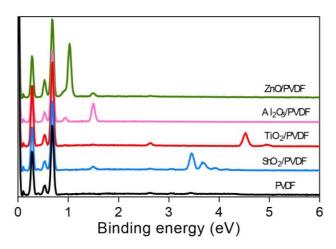


Figure S3. EDS spectra of membranes

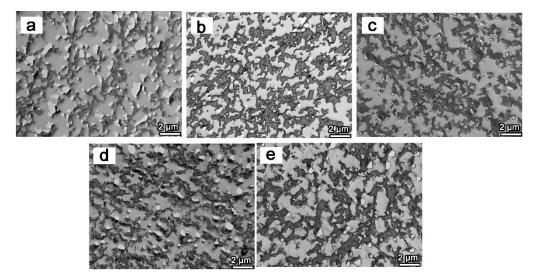


Figure S4. TEM images of the slices of a) nascent, b) ZnO-coated, c) Al_2O_3 -coated, d) TiO₂-coated, and e) SnO₂-coated membranes at low magnification.

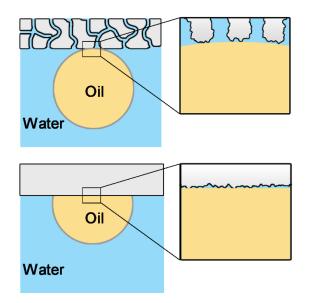


Figure S5. Scheme of the oil contact angles on membrane and silicon water surfaces. Despite minimal roughness on the silicon surface, it can be also regarded as an ideal "composite surface" for an adhesion work calculation.

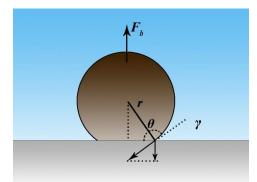


Figure S6 Force analysis of oil on a membrane surface under water. As shown in Figure SX, F_b is the buoyancy, γ is the counterforce of surface tension acting on oil by the membrane surface, θ is the oil contact angle, and *r* the radius of the oil droplet. The lifting fore is the buoyance $F_b \approx \rho g \frac{4}{3} \pi r^3$, while the adhesion force is $F_a = 2\pi r\gamma \sin^2 \theta$, which achieves a balance when $F_a = F_b$. We can conclude from these equations that the adhesion force can be reduced by increasing the oil contact angle and decreasing the contact area/triple phase line.



Figure S7. Crude oil in permeate solution in the case of nascent PVDF membrane

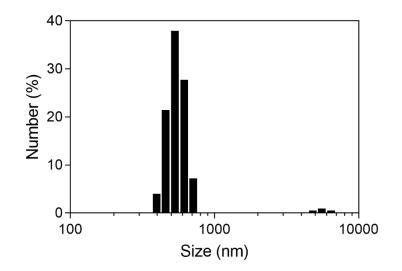


Figure S8 Size distribution of oil droplets in oil/water mixture. The permeate shows no peaks.