Electronic Supplementary Information

Water Solvent-assisted Condensation Polymerization Strategy of the Superhydrophobic Lignocellulosic Fiber for Efficient Oil/Water Separation

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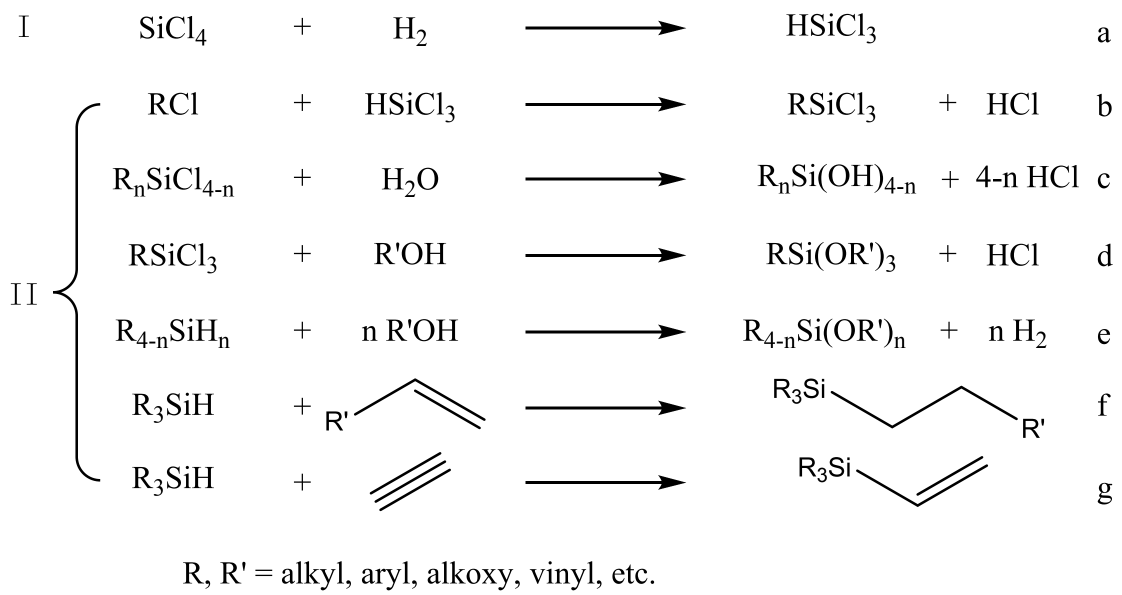
Table S1 A summary of various preparation methods of various (super)hydrophobic oil/water separation materials

Main materials	Method	Solvent used	Byproduct	Substrate	Ref.
TEOS, OTMS	Ultrasound, irradiation	Ethanol	Ethanol, methanol	Fabric Bag	1
MTES	Freeze-drying, immerse	Ethanol	Ethanol	Cellulose aerogels	2
MTMS, HDTMS	Freeze-drying	Ethanol	Methanol	Cellulose aerogels	3
MBA, urea, MTCS	Freeze-drying, vapor deposition	Water	HCl	Cellulose aerogels	4
OTCS, $ClCH_2COOH$, CH_3COOH	Freeze-drying, vapor deposition	Ethanol, isopropanol, methanol	HCl	Cellulose aerogels	5
ESO, BDE	Freeze-drying, immerse	n-hexane	-	Cellulose aerogels	6
MDI	Freeze-drying, immerse	Acetone, tert-butanol	-	Cellulose aerogels	7
DA·HCl, ODA	Freeze-drying	Water, tert-butanol	-	Cellulose aerogels	8
DADE, POM	Freeze-drying	NMP, water	-	BC aerogels	9
SiO_2, DVB, AIBN	Polymerization	Ethyl acetate	-	-	10
NaOH, $NaClO_2$, HCl, CH_3COOH, titanium isopropoxide	Freeze-drying, carbonization	Ethanol	Inorganic matter	Carbon aerogels	11
NaOH, $NaClO_2$, HCl, CH_3COOH	Freeze-drying, carbonization	Water	Inorganic matter	Carbon aerogels	12
PP, PTFE	Supercritical CO_2 foaming	-	-	Foam	13
TPU, EP, CNTs, hardener	Freeze-drying	1, 4-dioxane	-	Foam	14
OTCS, APT, HCl	Immerse	Toluene, ethanol, acetone	HCl	PU sponge	15
TEOS, MTMS, TMCS	Irradiation	Ethanol, methanol	Ethanol, methanol, HCl	Polyurethane sponge	16
SA, HCl, thiourea	Immerse	Ethanol	-	Copper mesh	17
CA, NaOH	3D-printed	Ethyl acetate, methanol	-	Cellulose mesh	18
ZnO, DFTMS, PS	Hydrothermal reaction, dip-coating	Ethanol, THF	Methanol	Cotton	19
PEI, 5Acl, ODA	Dip-coating/immerse	Ethanol, THF	-	Cotton	20

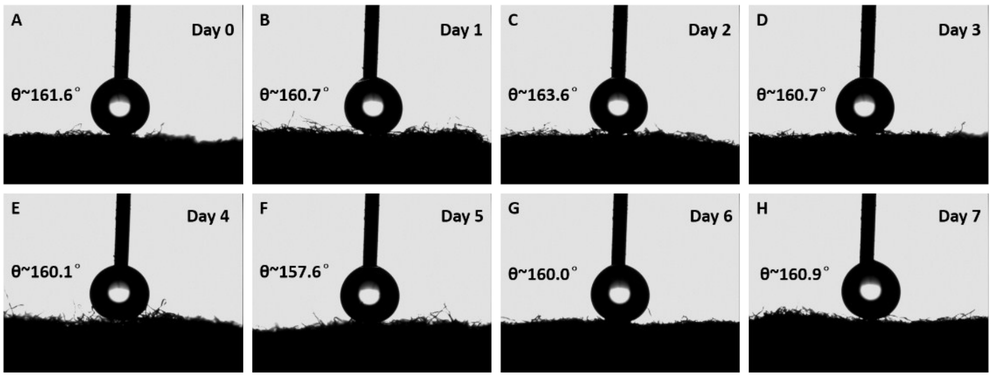


Scheme S1 Method for preparing silane coupling agent

As shown in Table S1, the low surface energy materials mainly were used in the superhydrophobic treatment in the form of foam, sponge, aerogels, and mesh materials for oil/water separation. The above materials are treated with various silane coupling agents which are assisted by organic solvents or inorganic nanoparticles as well. These processes are accompanied by the production of all kinds of by-products (hydrochloric acid, alcohols, and inorganics). Meanwhile, the oil/water separation materials of originated from biomass substrates are normally made into various cellulose aerogels or carbon aerogels. The preparation of micro-nanocellulose is energy-intensive, and the further freeze-drying process is also high energy-consuming. Thus it is difficult to expand on a large scale. In addition, the raw materials used for foam and sponge are difficult to degrade. The chemical reaction formula for the preparation of various silane coupling agents commonly used for (super) hydrophobic surfaces are given in Scheme S1.[21-28] The preparation of silane coupling agents is generally high energy consumption and heavy pollution. Based on the above reasons, it is necessary to develop more effective, environmentally friendly, bulky and cheap chemicals to (super) hydrophobicize the surface of materials.

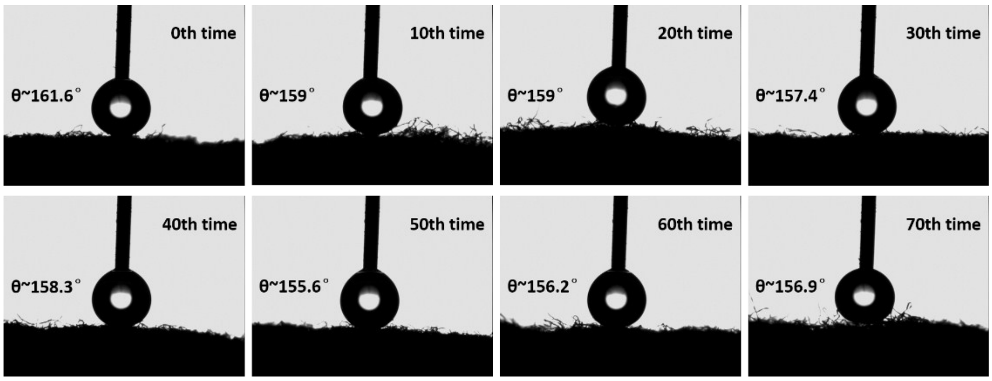
Abbreviation	
TEOS	Tetraethylorthosilicate
OTMS	Octadecyltrimethoxysilane
SA	Stearic acid
TPU	Thermoplastic polyurethane
EP	Epoxy resin
ESO	epoxidized soybean oil
MTCS	Methyltrichlorosilane
CNTs	Carbon nanotubes
PDMS	Poly(dimethylsiloxane) trimethylsiloxyterminated
DA·HCl	Dopamine hydrochloride
TMCS	Trimethylchlorosilane
BDE	1,4-butanediol diglycidyl ether
ODA	Octadecylamine
HDTMS	Hexadecyltrimethoxysilane
MTMS	Methyltrimethoxysilane
OTCS	Octyltrichlorosilane
PP	Polypropylene
PTFE	Polytetrafluoroethylene
DADE	4,4'-Diaminodiphenyl ether
DFTMS	Dodecafluoroheptyl-propyl-trimethoxysilane
PS	Polystyrene
PVA	Poly(vinyl alcohol)
POM	Paraformaldehyde
CA	Cellulose acetate
PU	Polyurethane
MDI	methylene diphenyl diisocyanate
MBA	N,N'-methylenebisacrylamide
OTCS	Octadecyltrichlorosilane
APT	Attapulgite
5Acl	Dipentaerythritol penta-/hexa-acrylate
BC	Cacterial cellulose
PEI	branched poly(ethyleneimine)
DVB	Divinylbenzene
AIBN	Azodiisobutyronitrile

UV Irradiation tests



Supplementary Figure S6. UV Irradiation tests. (A-H) The OMF lignocellulosic fiber was exposed to ultra-violet (UV) radiations of both shorter (254 nm) as well as longer (365 nm) wavelengths for 168 hours.

Filtration recycle tests



Supplementary Figure S6. Filtration recycle tests. The contact angle images illustrating the separation of a complex oil/water mixture that consists of heavy oil (25 mL of DCM) and water (25 mL) for repetitive use in gravity-driven filtration of an oil/water mixture at least 70 times, through gravity-driven filtration using OMF lignocellulosic fiber.

Movie S1

Illustrating the separation of floating droplets of motor oil by using OMF lignocellulosic fiber. 0.5 mL of oil was placed on the air/water interface in a Petri dish, then the treated pulp was placed on the water surface and the OMF lignocellulosic fiber selectively absorbed the oil from the oil/water interface within 30 seconds.

Movie S2

Illustrating the separation of floating droplets of motor oil by using original lignocellulosic fiber. 0.5 mL of oil was placed on the air/water interface in a Petri dish. The original lignocellulosic pulp could not absorb any trace of the floating oil droplets, but the original lignocellulosic pulp absorbed water quickly and became wet. The wet lignocellulosic pulp could not absorb any trace of the moving oil droplets on the air/water interface because of repellence effect.

Movie S3

Illustrating the successful collection/separation of heavy model-oil (DCM) under water (red colour aids visual inspection) using the OMF lignocellulosic fiber.

Movie S4

Illustrating the original lignocellulosic fiber was unsuitable for collecting oil under water but was for collecting the aqueous phase (red colour) selectively.Movie S5

Oil/water separation tests. Illustrating the separation of a complex oil/water mixture that consists of heavy oil (25 mL of DCM, colourless) and water (25 mL, red colour) through gravity-driven filtration using OMF lignocellulosic fibers.

Movie S6

Antifouling tests. 1. Illustrating the OMF lignocellulosic fibers were inserted into MnO_2 dyed water, the OMF lignocellulosic fibers did not get dyed due to the antifouling properties. 2. Showing that fast flowing K_2MnO_4 dyed water did not stain the OMF lignocellulosic fibers which were pasted on the "double-sided tape" substrates.

Movie S7

Illustrating the successful collection/separation of heavy model-oil (20 mL of DCM, colorless) under water (20 mL red colour aids visual inspection) using the OMF cotton fabric.

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