Efficient Removal of Aerosol Oil-mists using Superoleophobic Filters

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Electronic Supplementary Information

Experimental Section

Materials: PFAP (perfluoroalkyl acrylic copolymer) was purchased from Chemcolor Industries Australia Pty Ltd. Glycerol propoxylate triglycidyl ether (GPTE), and 1-methylimidazole (1-MI) were purchased from Sigma-Aldrich. Distilled water, ethanol and paraffin oil were purchased from LES store. Di-ethyl-hexyl-sebacate (DEHS, surface tension 32 mN/m, density 912 kg/m^3, dynamic viscosity 23 mPa t 20 °C) was purchased from Aladdin Industrial Corporation, and olive oil was purchased from local supermarket. Glass fibre nonwoven fibrous mats (Lypore 9866, 105 g/m^2) were purchased from Lydall, Inc. The fibrous filter had a thickness of 0.56 mm under 50 kPa.

Preparation of superoleophobic coating solution: 5.0 g PFAP was dissolved in 100 ml distilled water, then stirred by a stirring bar for 30 minutes to form 5 wt. % milky emulation solution. The concentration of coating solution was chosen by best CA among 5 solution concentrations, see Table S1.

Superoleophobic coating treatment: The glass fibre mat sample was immersed in the coating solution for 10 minutes to ensure that all the fibres were completely wetted. The fibrous mat was taken out from the coating solution, and excess liquid was dripped in fume-hood. The fibrous mat was finally dried in oven at 150 °C for 1 h.

Superoleophilic coating treatment: 5.0 g glycerol propoxylate triglycidyl ether (GPTE) was dissolved in 50 ml ethanol to prepare 10 w/v % solution A. 1 g 1-methylimidazole was dissolved in 10 ml ethanol to prepare 10 w/v % solution B. The solutions A and B were mixed in a volume ratio of A: B=10:1. The as prepared mixture was diluted with ethanol to get certain 3 w/v% solution for coating treatment. The glass fibre mat sample was immersed in the coating solution for 10 minutes to ensure that all the fibres were completely wetted. The fibrous mat was taken out from the coating solution, and excess liquid was dripped in fume-hood. The fibrous mat was dried at 60 °C.

Filtration performance test: The filtration performance was evaluated on a purposed built apparatus shown in Fig. S1. Di-ethyl-hexyl-sebacate (DEHS) was used as oil model. Compressed air was pre-conditioned by passing through a HEPA (high efficiency particulate air) filter. Aerosol-mist was generated by passes the conditioned air through an aerosol generator (TSI, 9306A Six-Jet Atomizer). The total amount of air flux was stabilized at 127.2 L/min. A high inlet liquid concentration level was selected to shorten test time, and promote the inlet concentration to be the dominant factor of pressure drop and clogging to ensure the accuracy of filtration test. A vacuum pump was applied for pumping the filtered gas. The generated aerosol-mist size was controlled in the range of 0.01 - 20 μm with the most permeable particle size (MPPS) of 0.2-0.5 μm. The filtration performance (in concentration) of small particles (0.01-0.8 μm) and large particles (0.5-20 μm) was evaluated separately by SMPS (Model 3936 scanning mobility particle sizer) and APS (3221 aerodynamic particle sizer), respectively, according to the different collection mechanisms. The pressure drop was measured by a differential pressure transducer (Yokogawa, EJX-110A) and flow rate was measured by a rotameter. All filtration performance data were collected at pseudo-steady state.

Durability test: Washing durability was tested via ultrasonication the fabrics in water, ethanol, DEHS and paraffin oil for 60 minutes using an ultrasonicator (FXP10M Unisonics) at power 100 W and 40 kHz operating frequency.

Other characterisations: Contact angles were measured by a contact angle goniometer (KSV CAM101 Instruments Ltd), and 3 μL liquid droplets was applied. A purpose-made apparatus consisting of a sample holder and a digital angle meter were applied to measure the sliding angles. All contact angle (CA) and sliding angle (SA) values reported represent the mean of 5 measurements. Surface morphology was observed using a scanning electron microscope (SEM Supra 55VP) operated at an acceleration voltage of 5.0 kV. Transmission electron microscopy (TEM, JEOL-2100F, Japan) was operated at an accelerating voltage of 200 keV and the images were acquired with a Gatan image filter. Coating thickness is calculated by average 30 measurements from 2 TEM images. Fourier transform infrared spectroscopy was taken on a Burker Betex 70 FTIR spectrometer in Attenuated Total Reflection (ATR) mode ranging from 4000 cm^{-1} to 500 cm^{-1}. X-ray photoelectron spectroscopy (XPS) was applied for element and binding energy analysis, with a base pressure of 5×10^{-10} mbar. The XPS spectra were recorded using a Kratos Axis Ultra DLD spectrometer (Shimadzu, Japan) employing a monochromated Al-Ka X-ray source (1486.6eV). The C1s (284.8 eV) has been used to calibrate the binding energy (BE) scale of XPS measurements. Air permeability test was measured by FX 3300 Air Permeability tester according to the Standard (BS 5'636), with test area for 5 cm^2, test pressure of 96 Pa, and all values reported represent the mean of 5 measurements. Pore size distribution was tested by capillary flow porometer (CFP-1200 AEF, PMI Porous Materials Inc). Bursting strength was measured by Instron tester according to standard ISO3303-2, and all values reported were the average value of 5 times tests.

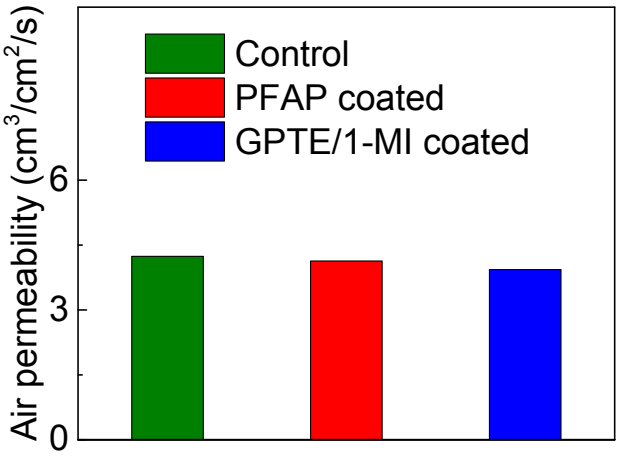

Fig. S1 Air permeability of untreated (i.e. control), PFAP coated and GPTE-1-MI coated filters.

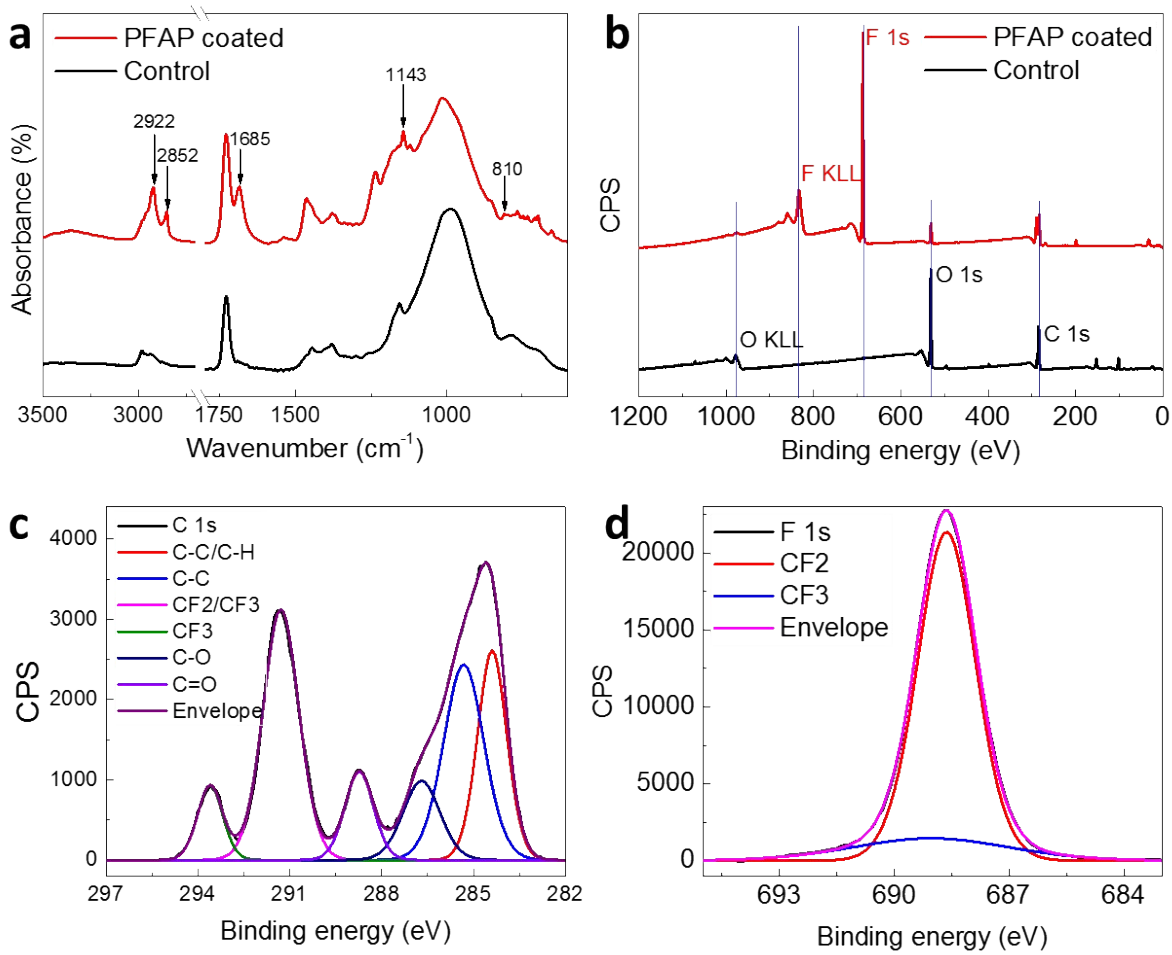

Fig. S2 Chemical characterisation of untreated and PFAP coated glass fibre nonwovens (a) FTIR spectra, (b) XPS survey spectra, (c) High-resolution C1s spectra of PFAP coated glass fibres, (d) High-resolution F1s spectra of PFAP coated (superoleophobic) glass fibres.

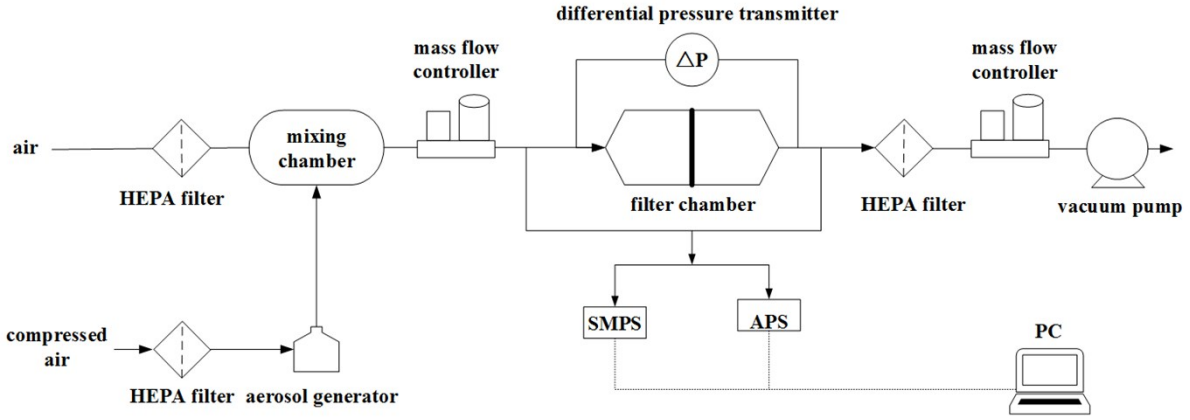

Fig. S3 Schematic diagram apparatus for filtration performance test.

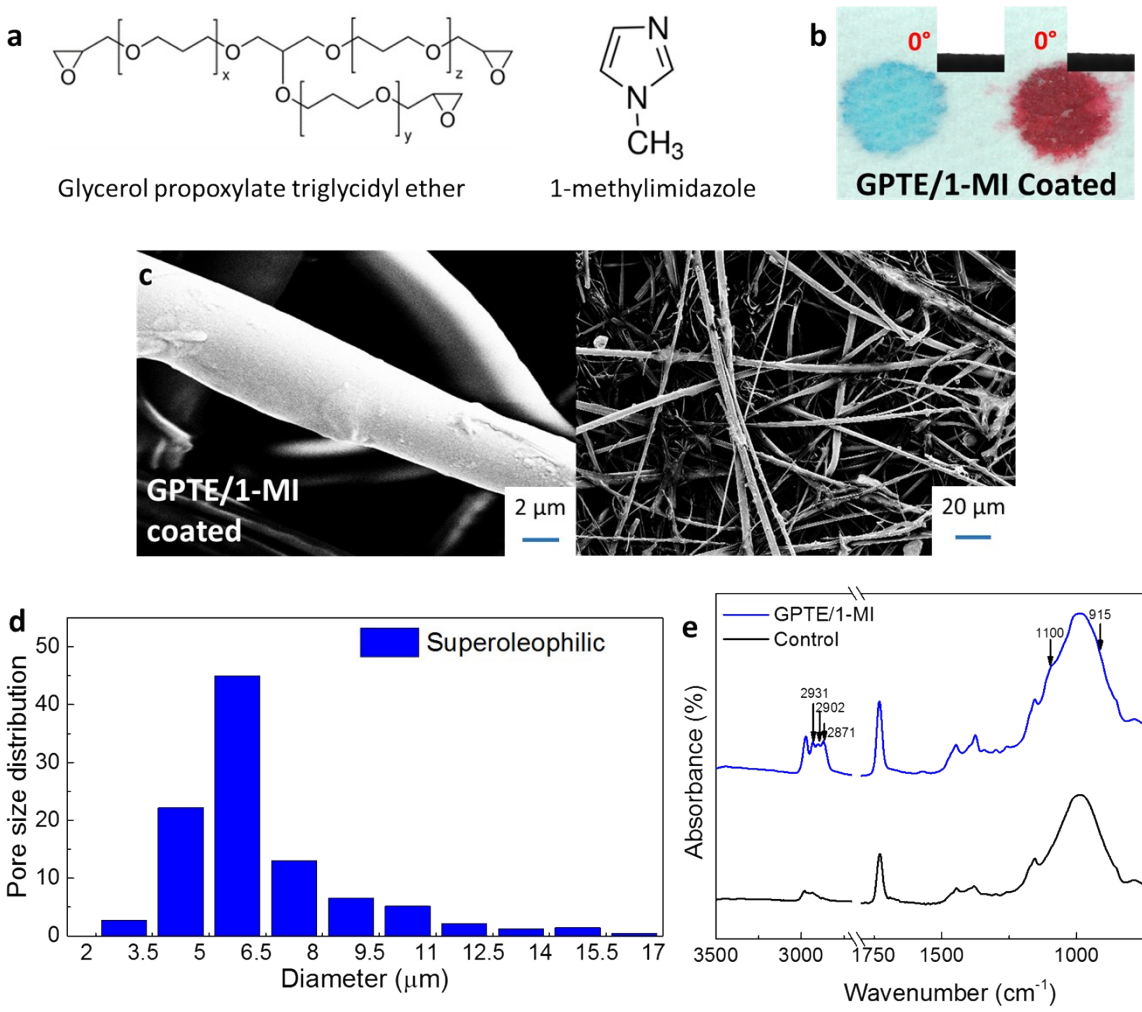

Fig. S4 (a) Structure of chemicals used to prepare superoleophilic filters. (b) Water and oil droplets on GPTE/1-MI coated glass fibre filters (blue-coloured water and red-coloured olive oil). (c) SEM image. (d) Pore size distribution (mean pore size 5.18±0.57 µm). (e) FTIR spectra.

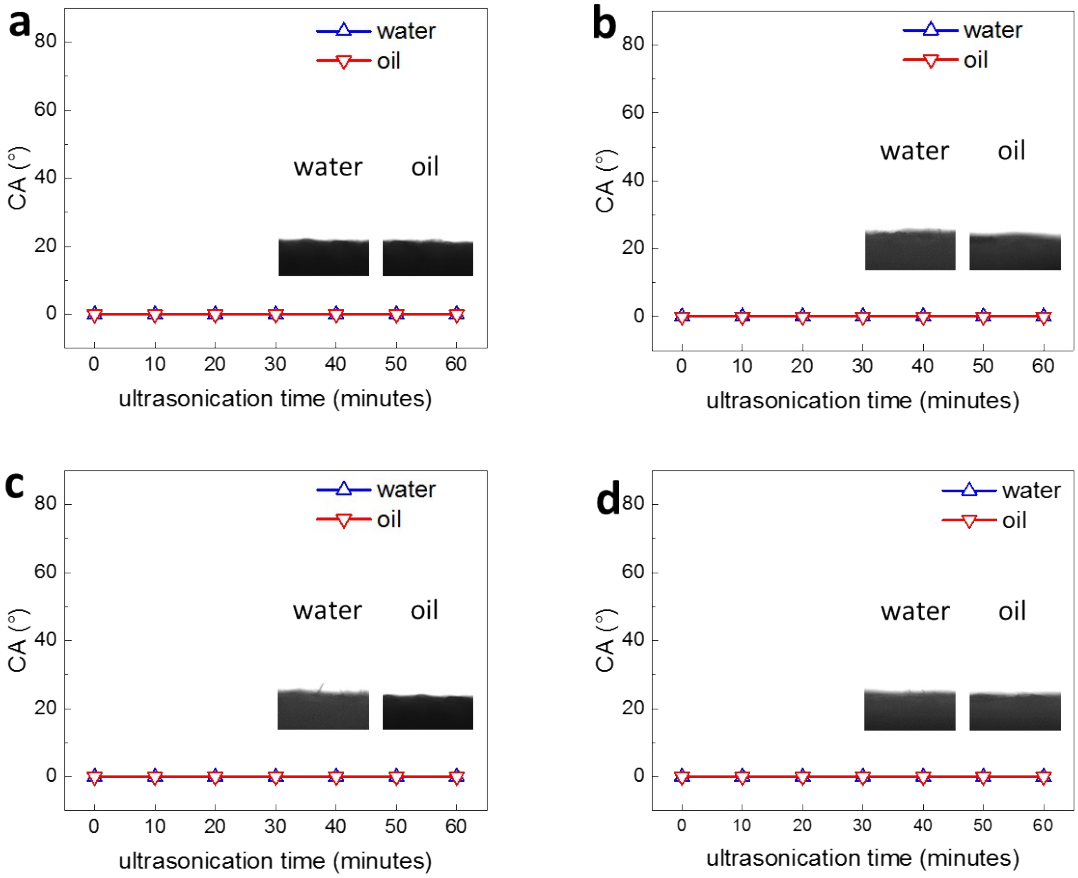

Fig. S5 Water/oil CA on superoleophilic filter after 60 minutes ultrasonication in (a) water, (b) ethanol, (c) DEHS, and (d) paraffin oil.

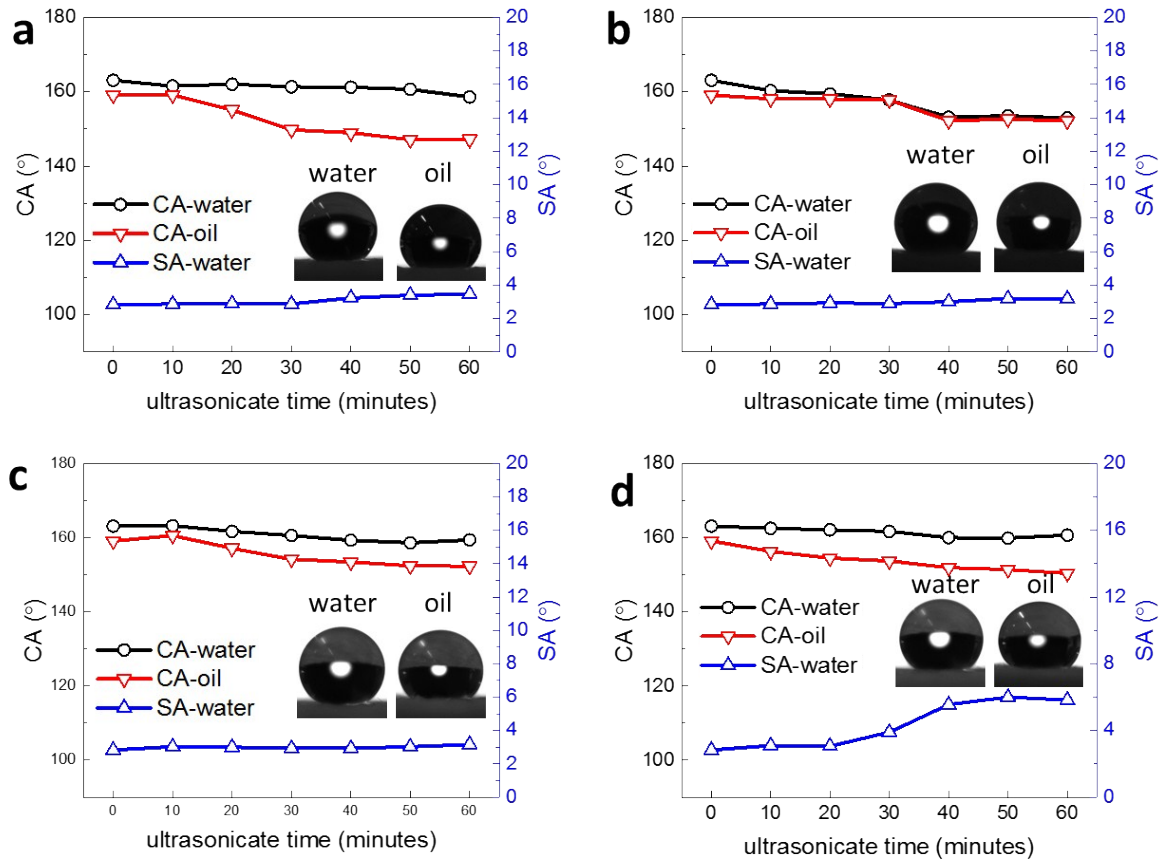

Fig. S6 Water/oil CA and water SA on superoleophobic filter after 60 minute ultrasonication in (a) water, (b) ethanol, (c) DEHS, and (d) paraffin oil.

Table S1 (a) Effect of PFAP coating treatment condition on the filter properties

Concentration (wt.%)	CA (°)		Air permeability ($cm^3/cm^2/s$)	Bursting strength (Pa)
	Water	Olive oil		
0.5%	150.6	148.1	4.48	6465.9
1%	155.8	154.8	4.39	7001.8
2.5%	160.1	157.8	4.24	7794.6
5%*	163.1	159.1	4.13	8881.8
7.5%	157.5	156.8	4.05	10571.9
10%	154.9	151.6	4.00	12811.5

* The condition used for coating treatment of fibrous filter.

(b) Oil contact angle tested on different spots of the superoleophobic filter

CA (°)	
Water	Olive oil
164.1	161.3
162.9	158.1
163.8	159.2
161.7	158.4
163.6	159.3
162.4	160.5
163.8	157.6
161.7	159.8
162.8	159.1
164.0	158.2

Based on the ten measurement results, the mean ± standard deviation is 163.1±0.87° for water contact angle, and 159.1±1.08° for oil contact angle.

Table S2 Effect of GPTE/1-MI treatment condition on filter properties

Concentration (wt.%)	Wetting time (s)		Air permeability ($cm^3/cm^2/s$)	Bursting strength (Pa)
	Water	Olive oil		
1%	3.00	4.32	3.78	1821.1
3%*	0.10	3.10	3.93	1362.5
5%	0.10	2.34	4.34	1057.3
7%	0.10	2.08	4.25	661.1
10%	0.20	2.67	4.20	545.2

* The condition used for coating treatment of fibrous filter.

Table S3 Oil-mist filtration results

Thick-ness (mm)	Pres-sure drop (kPa)	Small mists			Large mists		
		Stable efficiency (%)	Stable quality factor (kPa^{-1})	Stable downstream concentration (P/cm^3)	Stable efficiency (%)	Stable quality factor (kPa^{-1})	Stable downstream concentration (P/cm^3)
(a) Untreated filters							
0.56	6.90	87.92	0.31	253053.1	99.82	0.92	220.84
1.12	7.79	96.40	0.43	73430.9	99.92	0.92	96.81
1.68	8.69	98.84	0.51	22592.5	99.94	0.86	70.02
2.24	9.49	99.31	0.53	13483.3	99.89	0.71	149.84
2.80	10.15	99.63	0.55	7395.8	99.86	0.65	247.60
3.36	10.78	99.67	0.53	6089.4	99.88	0.62	161.73
(b) Superoleophobic filters							
0.56	7.31	97.12	0.49	61300.0	100.00	1.78	0.40
1.12	8.33	99.44	0.62	11300.0	100.00	2.12	0.00
1.68	8.87	99.84	0.72	3330.0	100.00	1.64	0.09
2.24	9.94	99.96	0.78	985.2	100.00	1.07	4.31
2.80	11.20	99.98	0.76	404.7	99.99	0.79	21.50
3.36	11.87	99.99	0.74	326.2	99.98	0.73	25.70
(c) Superoleophilic filters							
0.56	6.63	87.44	0.31	177000	99.68	0.87	268.6
1.12	7.55	97.11	0.47	57500	99.88	0.89	156.3
1.68	8.42	98.93	0.54	21200	99.92	0.84	92.8
2.24	9.08	99.56	0.60	6990	99.87	0.73	111.0
2.80	10.04	99.70	0.58	5670	99.90	0.69	121.7
3.36	10.11	99.82	0.62	2100	99.92	0.70	40.7

Table S4 Oil-mist filtration performance of double-layer filters in various layouts

Filter layout front - rear	Pressure drop (kPa)	Small mists		Large mists	
		efficiency (%)	quality factor (kPa^{-1})	efficiency (%)	quality factor (kPa^{-1})
control-control	7.792	96.40	0.42	99.92	0.92
control-superoleophobic	9.015	99.30	0.55	99.99	1.36
superoleophobic-control	11.90	98.72	0.37	99.74	0.50
superoleophobic-superoleophobic	8.329	99.44	0.62	99.99	2.12