Supporting Information

The Ultrathin Fluoroacrylic Coating Wrapped Microporous Non-Woven Fabrics as Efficient Air Filtration Membranes

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1. General Information

2,2,3,3-tetrafluoropropyl acrylate (TFPA), 1H,1H,5H-octafluoropentyl acrylate (OFPA), and acrylic acid (AA), analytical pure, were purchased from Bide Pharmatech Ltd. Methyl methacrylate (MMA), butyl acrylate (BA), and glycidyl methacrylate (GMA), analytical pure, were purchased from Shanghai Macklin Biochemical Co., Ltd. Dodecafluoroheptyl methacrylate (DFMA) was purchased from Shang Fluoro Technology Co., Ltd. Ammonium persulfate (APS) and sodium bicarbonate (NaHCO₃), analytical pure, were purchased from Sinopharm Chemical Reagent Co., Ltd. Ethanol (EtOH, 75%) was purchased from Shanghai Gaoyun Chemical Co., Ltd. Sodium dodecyl sulfate (SDS) and octylphenol polyoxyethylene ether (OP-10), analytical pure, were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd.

The viscosity was measured using a rotational viscometer (NDJ-5S, Shanghai Lichen, China). Functional group analysis was carried out *via* Fourier transform infrared spectrometer (Thermo Nicolet AVATAR-370, Nicolet, America). The zeta potential, particle size, and particle size distribution were determined by ZETASIZER Nano (ZETASIZER 3000 HSA, Malvern, UK). The internal morphology of the emulsion was performed with transmission electron microscopy (JEM 2100F, JEOL, Japan). The molecular weight was obtained by gel permeation chromatography (Agilent PL-GPC50, Agilent, USA). The glass transition temperature (Tg) was tested using differential scanning calorimetry (DSC 200F3, NETZSCH, Germany). The atomic compositions were performed on X-ray photoelectron spectroscopy (Thermo Scientific K-Alpha+, Thermo Fisher, USA). Scanning electron microscopy (SIGMA 300, CARL ZEISS, Germany) observed the surfaces of different fabrics. The distinct three-dimensional (3D) imaging of the coating surface morphology was observed by atomic force microscope (Dimension Icon, Bruker, Germany). The contact angle was measured with an optical goniometer (OCA 15EC, Dataphysics, Germany). The filtration efficiency was tested by an automated filter tester (LTAO-506, Shanghai Litao Automation Technology Co., Ltd, China).

2. Experimental Procedures

2.1 Preparation of the fluoroacrylic copolymer emulsion with different fluorine-containing monomers

The pre-emulsified semi-continuous seeded emulsion polymerization was used to synthesize the emulsion to obtain core-shell emulsion with uniform particle and narrow particle size distribution. Deionized water and the hybrid emulsifier (OP-10/SDS) was added to a 100ml beaker, and the hybrid emulsifier solution was prepared after stirring.

2.1.1 Preparation of core phase pre-emulsion

The core monomers and 1/3 of the hybrid emulsifier solution were added to a 250ml beaker. The core phase pre-emulsion was obtained by mechanical stirrer at 1500r/min for 1hr. It should be noted that the emulsion in the reaction flask was prone to stratification during the dropping in the pre-emulsion if the pre-emulsification time is less than 1hr.

2.1.2 Seeded emulsion polymerization

1/3 of core phase pre-emulsion, 1/3 of the hybrid emulsifier solution, 1/3 of initiator solution, and NaHCO₃ solution were added into a 250ml four-necked flask equipped with a reflux condenser, mechanical stirrer, thermometer and constant pressure drop funnel. The reaction system was heated to 80°C. When the emulsion appeared with blue fluorescence in the flask, the seeded emulsion was obtained after 30minutes of continuous reaction.

2.1.3 Core phase emulsion polymerization

The remaining core-phase pre-emulsion and 2/3 of the initiator solution were added to the reaction system simultaneously within 2hrs at 80°C. Then the system was held at 80°C for 1hr to obtain the core phase emulsion.

2.1.4 Preparation of shell phase pre-emulsion

The shell m	onomers and 1/3 of the hybrid emulsifien	solution	n were added to a 250ml beaker. The	shell phase pre-emulsion was obta	ined by
mechanical	stirrer	at	1500r/min	for	1hr.

2.1.5 Core-shell emulsion polymerization

The initiator solution and shell phase pre-emulsion were dropped into the reaction system simultaneously within 2hrs at 80°C. Then the system was heated to 85°C and held for 1hr. The emulsion was prepared after the mixture was cooled and filtered. Figure S1 displays the schematic diagram of the polymerization process for fluoroacrylic copolymer emulsions.



Figure S1 Schematic diagram of the polymerization process for fluoroacrylic copolymer emulsions.

The structure of the different fluorine-containing monomers was displayed in Figure S2.



Figure S2 The structure of the different fluorine-containing monomers.

The viscosity of emulsions was measured by a rotational viscometer (NDJ-5S, Shanghai Lichen, China) at ambient temperature with 0# spindle and 60rpm speed.

The coagulation was the ratio of condensates weight to the total weight of emulsion. The condensates were collected from the flask, magneton, and the filter screen and dried at 100°C to constant weight.

About 1g of the emulsion was dropped on the polypropylene sheet. The emulsion was cured at ambient temperature and then dried at 100°C to constant weight. The solid content and the conversion were reliable from the following equations.¹ Solid content(%) = $(M_2 - M_0)/(M_1 - M_0) \times 100\%$

Where M₀ was the weight of the polypropylene sheet, M₁ was the total weight of the polypropylene sheet and the coating before drying, and M_2 was the total weight of the polypropylene sheet and the coating after drying. $Conversion(\%) = (Solid content \times M_3)/M_4 \times 100\%$

Where M₃ was the total weight of all the reactants, and M₄ was the total weight of monomers.

The water absorption was used to characterize the water resistance of coating. The water absorption of the coating was calculated from the following formula. Water absorption(%) = $(M_5 - M_2)/(M_2 - M_0) \times 100\%$

M₅ was the total weight of the polypropylene sheet and the coating after soaking in water for 24hrs.

The	recipes for	fluoroacrylic copolymer	emulsions with	different fluorine	-containing	monomers and	characteristics	for the	emulsions,
coatings	and	wrap-coated	non-woven	fabrics	are	displayed	in	Table	S1.

Fluorine-o	containing monomers	TF	PA	OF	PA	DFN	DFMA		
		Core	Shell	Core	Shell	Core	Shell		
	MMA (g)	5.6	2.7	5.6	2.7	5.6	2.7		
	BA (g)	2.8	5.4	2.8	5.4	2.8	5.4		
	AA (g)	0.6	0	0.6	0	0.6	0		
	GMA (g)	0	0.9	0	0.9	0	0.9		
	TFPA (g)	0	12	0	0	0	0		
	OFPA (g)	0	0	0	12	0	0		
	DFMA (g)	0	0	0	0	0	12		
	OP-10 (g)	0	.5	0.	.5	0.5	0.5		
	SDS (g)	0.	25	0.3	25	0.2	5		
Eluorocerulic	APS (g)	0	.3	0.	.3	0.3			
copolymer	NaHCO₃ (g)	0	.3	0.	.3	0.3			
enuisions	H ₂ O (g)	68	.65	68.	.65	68.6	55		
	Amounts (g)	1	00	10	00	10	0		
	Appearance	Wh	ite ^[a]	Whi	te ^[a]	White ^[a]			
	Solid content (%)	29	9.5	29	.8	29.8			
	Conversion (%)	g	8	9	9	99)		
	Coagulation (%)	0.	65	0.	74	0.8	9		
	Viscosity (mPa·s)	4	.1	4.	.5	4.0)		
	Zeta potential (mV)	-4	1.6	-43	3.8	-54	.7		
	Particle size (nm)	11	3.3	14	1.2	138	.1		
	PDI ^[b]	0.	02	0.0	01	0.0	2		
Coatings ^[c]	Water absorption (%)	2	.2	2.	3	1.2	2		
E-L. (d)	Water contact angle (°)	12	5.2	12	9.3	135	.3		
Fabricsion	Oil contact angle (°)	3:	1.3	33	.8	59.	4		

Table S1 The recipes for the emulsions and characteristics for the emulsions, coatings and wrap-coated non-woven fabrics.

[a] White with blue fluorescence. [b] Particle size distribution index. [c] Wrap-coatings on polypropylene sheet. [d] The non-woven fabrics wrap-coated by fluoroacrylic copolymer emulsions with different fluorine-containing monomers.

2.2 Preparation of the fluorine-free acrylic copolymer emulsion

The preparation processes of the DFMA-free acrylic copolymer emulsion were the same as 2.1. The schematic diagram of the polymerization process for fluorine-free acrylic copolymer emulsion is shown in Figure S3.



2.3 Preparation of the DFMA-acrylic copolymer emulsions with different weight ratio of DFMA

The DFMA-acrylic copolymer emulsions with different weight ratio of DFMA were synthesized by the same method. The preparation processes were the same as 2.1. The schematic diagram of the polymerization process for the DFMA-acrylic copolymer emulsions with the different weight ratio of DFMA is shown in Figure S1.

The recipes for the DFMA-acrylic copolymer emulsions with different weight ratio of DFMA and characteristics for the emulsions, coatings, and wrap-coated non-woven fabrics are displayed in Table S2.

The wei total core-s	The weight ratio of DFMA in 0 5 otal core-shell monomers (wt%)		1	.0	2	20	3	0	4	10	60				
		Core	Shell	Core	Shell	Core	Shell	Core	Shell	Core	Shell	Core	Shell	Core	Shell
	MMA (g)	9.5	4.8	9	4.5	8.5	4.3	7.5	3.8	6.5	3.3	5.6	2.7	3.5	1.8
	BA (g)	4.7	9.5	4.5	9	4.2	8.5	3.7	7.5	3.2	6.5	2.8	5.4	1.8	3.5
	AA (g)	0.6	0	0.6	0	0.6	0	0.6	0	0.6	0	0.6	0	0.6	0
	GMA (g)	0	0.9	0	0.9	0	0.9	0	0.9	0	0.9	0	0.9	0	0.9
	DFMA (g)	0	0	0	1.5	0	3	0	6	0	9	0	12	0	18
	OP-10 (g)	0	.5	0	.5	0	.5	0	.5	0	.5	0	.5	0	.5
	SDS (g)	0.	25	0.	25	0.	25	0.	25	0.	25	0.	25	0.	25
	APS (g)	0	.3	0	.3	0	.3	0	.3	0	.3	0	.3	0	.3
	NaHCO₃ (g)	0	.3	0	.3	0	.3	0	.3	0	.3	0	.3	0	.3
	H ₂ O (g)	68	.65	68	.65	68	.65	68	.65	68	.65	68	.65	68	.65
Emulsions	Amounts (g)	10	00	1	00	1	00	10	00	1	00	1	00	1	00
	Appearance	Wł	nite	W	nite	W	nite	Wh	ite ^[a]						
	Solid content (%)	26	5.1	27	7.1	2	7.5	27	7.9	28	3.0	29	9.8	29	9.6
	Conversion (%)	8	7	9	90	ç	2	9	3	ç	3	g	99	ç	9
	Coagulation (%)	2.	31	2.	01	1.	95	1.	75	1.	38	0.	89	1.	35
	Viscosity (mPa·s)	4	.0	4	.8	5	.0	5	.5	4	.6	4	.0	4	.0
	Zeta potential (mV)		-		-		-		-		-	-5	4.7		-
	Particle size (nm)		-		-		-		-		-	13	8.1		-
	PDI ^[b]		-		-		-		-		-	0.	02		-
	M _p (g/mol)		-		-		-		-		-	19	964		-
	M _n (g/mol)		-		-		-		-		-	16	56		-
	M _w (g/mol)		-		-		-		-		-	18	339		-
	M _v (g/mol)		-		-		-		-		-	18	805		-
	Mz		-		-		-		-		-	20)76		-
	M _{z+1}		-		-		-		-		-	23	358		-
	PD (M _w /M _n)		-		-		-		-		-	1.	11		-
Coatings ^[c]	Water absorption (%)		-		-		-				-	1	.2		
Fabricald	Water contact angle (°)	11	5.9	12	1.7	12	4.9	12	7.1	13	1.6	13	5.3	13	2.0
Fadrics ^{iuj}	Oil contact angle (°)	(D	29	9.4	3	5.8	48	3.8	53	3.5	59	9.4	63	1.0

Table S2 The recipes for the DFMA-acrylic copolymer emulsions and characteristics for the emulsions, coatings and wrap-coated non-woven fabrics.

[a] White with blue fluorescence. [b] Particle size distribution index. [c] Wrap-coatings on polypropylene sheet. [d] The non-woven fabrics wrap-coated by DFMA-acrylic copolymer emulsions with different weight ratio of DFMA.

The preparation process of the DFMA-acrylic copolymer emulsion (5kg scale) was the same as 2.1. The schematic diagram of the polymerization process for DFMA-acrylic copolymer emulsion (40wt% DFMA) is shown in Figure S1.

The recipes and properties for the DFMA-acrylic copolymer emulsion (5kg scale) are shown in Table S3.

Table S3 The recipes and properties for the DFMA-acrylic copolymer emulsion (5kg scale).						
The weight ratio of DFMA in	n total core-shell monomers (wt%)	40				
		Core	Shell			
	MMA (g)	280	135			
	BA (g)	140	270			
	AA (g)	30	0			
	GMA (g)	0	45			
	DFMA (g)	0	600			
	OP-10 (g)	25				
	SDS (g)	12.5				
DFMA-acrylic copolymer	APS (g)	15				
emulsion	NaHCO ₃ (g)	15				
	H ₂ O (g)	3432.5				
	Amounts (g)	5000				
	Appearance	White with blue fluc	rescence			
	Solid content (%)	29.8				
	Conversion (%)	99				
	Coagulation (%)	0.89				
	Viscosity (mPa·s)	4.0				

2.4 Preparation of the DFMA-acrylic copolymer emulsions with different weight ratio of OP-10/SDS

The DFMA-acrylic copolymer emulsions with different weight ratio of OP-10/SDS were synthesized by the same method. The preparation processes of the DFMA-acrylic copolymer emulsions were the same as 2.1. The schematic diagram of the polymerization process for the DFMA-acrylic copolymer emulsions (the different weight ratio of OP-10/SDS) is shown in Figure S1.

The recipes for the DFMA-acrylic copolymer emulsions with different weight ratio of OP-10/SDS and properties for emulsions, coatings and wrap-coated non-woven fabrics are displayed in Table S4.

Griffin introduced the HLB value to measure the equilibrium relationship between hydrophilic and oleophilic groups in surfactant molecules.² The following formula can be used to calculate the HLB value of the hydrod emulsifier: $HLB_{AB} = (HLB_{A} \times W_{A} + HLB_{B} \times W_{B})/(W_{A} + W_{B})$

 HLB_A is the HLB value of the emulsifier A, HLB_B is the HLB value of the emulsifier B, W_A is the weight ratio of the emulsifier A, and W_B is the weight ratio of the emulsifier B. B.

The weight ratio of OP-10/SDS		1:3		1:2		1:1		2:1		3:1	
		Core	Shell	Core	Shell	Core	Shell	Core	Shell	Core	Shell
	MMA (g)	5.6	2.7	5.6	2.7	5.6	2.7	5.6	2.7	5.6	2.7
	BA (g)	2.8	5.4	2.8	5.4	2.8	5.4	2.8	5.4	2.8	5.4
	AA (g)	0.6	0	0.6	0	0.6	0	0.6	0	0.6	0
	GMA (g)	0	0.9	0	0.9	0	0.9	0	0.9	0	0.9
	TFPA (g)	0	12	0	0	0	0	0	0	0	0
	OFPA (g)	0	0	0	12	0	0	0	12	0	0
	DFMA (g)	0	0	0	0	0	12	0	0	0	12
	OP-10 (g)	0.19		0.25		0.375		0.5		0.56	
DFMA-acrylic	SDS (g)	0.56		0.5		0.375		0.25		0.19	
emulsions	APS (g)	0	.3	0	.3	0	3	0.	3	0.	3
	$NaHCO_3$ (g)	0	.3	0	.3	0	3	0.	3	0.	3
	H ₂ O (g)	68	.65	68.65		68.65		68.65		68.65	
	Amounts (g)	10	00	100		100		100		100	
	Appearance	Cle	ear	Cle	ear	Whi	te ^[a]	Whi	te ^[a]	Whi	te ^[a]
	Coagulation (%)	2.	28	1.	16	1.	08	0.8	39	1.	75
	HLB	33	.6	31	5	27	.3	23	.0	20	.8
	Particle size (nm)	10	1.3	12	0.3	13	1.3	13	3.1	15	3.5
	PDI ^[b]	0.	02	0.	02	0.	03	0.0	02	0.0	03
Coatings ^[c]	Water absorption (%)	1	2	1	.2	1	2	1.	2	1	1
Fabrics ^[d]	Water contact angle (°)	13	3.3	13	5.3	13	4.2	13	5.3	13	9.3

 Table S4 The recipes for the DFMA-acrylic copolymer emulsions with different weight ratio of OP-10/SDS and characteristics for the emulsions, coatings and wrap-coated non-woven fabrics.

[a] White with blue fluorescence. [b] Particle size distribution index. [c] Wrap-coatings on polypropylene sheet. [d] The non-woven fabrics wrap-coated by DFMA-acrylic copolymer emulsions with different weight ratio of OP-10/SDS.

2.5 Wrap-coated non-woven fabrics

Non-woven fabric was washed with 75% ethanol to dust deposits and then were dried at 60°C for 24hrs. The fluoroacrylic copolymer emulsions were coated on non-woven fabric using the ultrasonic continuous dip-coating method at 1.5m/min speed. Then the wrap-coated fabric was left at ambient temperature for 2days and were dried at 80°C for 24hrs.

2.6 Preparation of the coatings on polypropylene sheet

The polypropylene sheets were placed in the DFMA-acrylic copolymer emulsion (40wt% DFMA) and vibrated in ultrasonic apparatus for 0.5h. The coated polypropylene sheets were cured at 25°C and 65%RH (relative humidity) to form the coatings for 24h, and then dried at 100°C for 40minutes.

3. Characterization and Results

3.1 Fourier transform infrared spectrometer (FTIR)

FTIR (Thermo Nicolet AVATAR-370, Nicolet, America) was used for the determination of the spectra of DFMA-acrylic copolymer emulsion. The spectra were recorded in wave number range from 4000 to 400cm⁻¹.



Figure S4 FTIR spectra of DFMA-acrylic copolymer emulsion (40wt% DFMA).

The peak at 2958cm⁻¹ is attributed to the stretching vibration of the C-H bond. The weak absorption at 2875cm⁻¹ is associated with the C-H bond symmetric stretching of $-CH_2$. The band at 1735cm⁻¹ corresponds to the C=O bond stretching vibration. The bands at 1459 and 1389cm⁻¹ are assigned to C-H bond wagging. Broader peaks between 1165 and 1247cm⁻¹ are due to the strong absorption of the C-O-C bond overlapped by stretching vibrations of the C-F bond.⁴ The peak at 842cm⁻¹ is assigned to the C-H bond vibration of the $-OC_4H_9$ group. The peak at 689cm⁻¹ corresponds to the C-F bond stretching vibration of $-CF_3$.

3.2 Zeta potential and particle size

The zeta potential, particle size, and particle size distribution of the DFMA-acrylic copolymer emulsion (40wt% DFMA) were determined by ZETASIZER Nano (ZETASIZER 3000 HSA, Malvern, UK). Before measurement, the emulsion was diluted to clear with deionized water.



Figure S5 Particle size and particle size distribution of the DFMA-acrylic copolymer emulsion (40wt% DFMA).

3.3 Transmission electron microscopy (TEM)

3.3.1 TEM for DFMA-acrylic copolymer emulsion

The internal morphology of the DFMA-acrylic copolymer emulsion (40wt% DFMA) was performed with TEM (JEM 2100F, JEOL, Japan) at an acceleration voltage of 120kV. The emulsion was diluted with deionized water, dropped on a carbon-coated copper grid, and then negatively stained with an aqueous solution of 1.5wt% phosphotungstic acid before testing.

3.3.2 TEM for wrap-coated non-woven fibers

The cross-section of the wrap-coated fibers was analyzed with TEM (JEM 2100F, JEOL, Japan) at an acceleration voltage of 200kV to determine the thickness of fluoroacrylic wrap coating on the non-woven fiber surface.



B Figure S6 The cross-section of the wrap-coated fibers.

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3.5 Gel permeation chromatography (GPC)

The molecular weight and molecular weight distribution of the DFMA-acrylic copolymer emulsion (40wt% DFMA) were determined by GPC (Agilent PL-GPC50, Agilent, USA) using deionized water as eluent with a flow rate of 1.0mL/min, and the column system was calibrated by monodisperse linear polystyrene standards.⁵

Table S5 The GPC results of DFMA-acrylic copolymer emulsion (40wt% DFMA).

M _p (g/mol)	M _n (g/mol)	M _w (g/mol)	M _z (g/mol)	M _{z+1} (g/mol)	M _v (g/mol)	(M _w /M _n)
1964	1656	1839	2076	2358	1805	1.11



Figure S7 The GPC spectra of DFMA-acrylic copolymer emulsion (40wt% DFMA).

3.6 Differential scanning calorimetry (DSC)

The DFMA-acrylic copolymer emulsion (40wt% DFMA) was weighed and DSC (DSC 200F3, NETZSCH, Germany) was taken at heating rate of 20°C/min. Thermal behaviors of the emulsion were examined under nitrogen from -80°C to 150°C. The glass transition temperature (Tg) was taken at the onset of the corresponding heat capacity jump.



Figure S8 DSC curves of DFMA-acrylic copolymer emulsion (40wt% DFMA).

3.7 X-ray photoelectron spectroscopy (XPS)

XPS (Thermo Scientific K-Alpha+, Thermo Fisher, USA) with a monochromatic Al K α X-ray source (72W, 12kV) provided data of bond energy of 688eV corresponded to the fluorine with sp³-hybridization in the shell. The deviation of the binding energy was revised by the surface contamination C_{1s} (248.8eV).⁶ The coatings were removed from the PP sheet before testing.

Table S6 XPS analysis of atomic proportions at different coating interface. [a]

Interface	F atom/mol% (XPS/Calc.)	Catom/mol% (XPS/Calc.)	O atom/mol% (XPS/Calc.)
Coating-air	31.4/16.2 ^[b]	51.9/58.6 ^[b]	16.7/24.0 ^[b]
Coating-polypropylene film	13.9/16.2 ^[b]	61.6/58.6 ^[b]	24.5/24.0 ^[b]

[a] The total proportion of the three elements in the shell was counted as 100% (the hydrogens were not counted). [b] Calculated proportions in the shell.



Figure S9a XPS data of different interfaces of DFMA-acrylic coating (40wt% DFMA) on polypropylene film.



Figure S9b F₁₅ signals of different interfaces of DFMA-acrylic coating (40wt% DFMA) on polypropylene film.

3.8 Scanning electron microscopy (SEM)

The surfaces of the DFMA-acrylic-coated non-woven fabric (40wt% DFMA), blank non-woven fabric, and the ePTFE filtration membrane were observed through SEM (SIGMA 300, CARL ZEISS, Germany). Before SEM observations, a thin layer of conducting-coated Au layer was sputtered on each sample.⁷





Figure S10 SEM images of different fabrics. [A] DFMA-acrylic-coated non-woven fabrics (40wt% DFMA). [B] Blank non-woven fabrics. [C] ePTFE filtration membranes.

3.9 Atomic force microscopy (AFM)

The distinct three-dimensional (3D) imaging of the coating surface morphology was observed by AFM (Dimension Icon, Bruker, Germany) in the tapping mode with a scan size of $2\mu m \times 2\mu m$ at ambient temperature, and the relative humidity was 55%.⁸⁻⁹

3.10 Contact angle

The wrap-coated non-woven fabrics water/oil (n-dodecane) contact angles were measured with an optical goniometer (OCA 15EC, Dataphysics, Germany). n-Dodecane was selected as the oil droplet for measurements. The injection volume of liquid was 5µl, and the average of three readings was used as the final contact angle of each sample.



Figure S11 Water and oil contact angles of the non-woven fabrics wrap-coated by DFMA-acrylic copolymer emulsions with the different weight ratios of DFMA.

3.11 Washing-resistance of wrap-coated non-woven fabric

Ethanol was chosen as the detergent with a concentration of 75%. The non-woven fabrics wrap-coated by DFMA-acrylic emulsion (40wt% DFMA) were washed with flowing ethanol twice for 2minutes every time. After dehydration, one washing process was accomplished. After washing 1 to 20 times, the wrap-coated non-woven fabrics water/oil (*n*-dodecane) contact angles were measured.



Figure S12 The effect of washing times on the contact angle of water and oil of wrap-coated non-woven fabrics.



Figure S13 The effect of washing times on the respiratory resistance of wrap-coated non-woven fabrics.



Figure S14 The filtration efficiency for 0.3µm NaCl and DEHS aerosol particles of wrap-coated non-woven fabrics.

3.12 Assessment of antibacterial and antifungal properties

According to *the Pharmacopoeia of the People's Republic of China*, the antibacterial and antifungal properties of the fluorine-free acrylic copolymer coating and the DFMA-acrylic copolymer emulsion (40wt% DFMA) against Staphylococcus aureus (*S. aureus*), Escherichia coli (*E. coli*), and Candida albicans (*C. albicans*) were evaluated utilizing the method of plate-counting. The total number of grown colonies/cm² were assayed after 24hrs. The assay details refer to our previous reported paper.¹⁰

	Coating on glass dish	E. coli	C. albicans	S. aureus
	DFMA-acrylic copolymer coating (40wt% DFMA in shell)	_[a]	_[a]	0.009
	Fluorine-free acrylic copolymer coating	13.4	14.0	126.7
[a]	No	colony	was	observed.

3.13 Wrap-coated non-woven fabric with L-menthol ingredient

1.5g of L-menthol was dissolved in cyclohexane. The L-menthol/cyclohexane solution was pre-emulsified with OP-10/SDS hybrid emulsifier (OP-10:SDS=2:1, 0.024g of OP-10 and 0.012g of SDS). The pre-emulsified L-menthol/cyclohexane solution was obtained by mechanical stirring at 1500r/min for 1hr. Then pre-emulsified L-menthol was added to DFMA-acrylic copolymer emulsion (40wt% DFMA). The non-woven fabrics were coated the L-menthol-added DFMA-acrylic copolymer emulsion. The weight reduction averages of two samples in each testing group of L-menthol-added wrap coated filtration membranes were calculated roughly as L-menthol loss. Samples were resealed in plastic bags right after testing.

Coated non-woven fabric samples at 20 L/min air flow, 8hrs/day

Normally placed and sealed coated non-woven fabric samples
 Normally placed and sealed coated non-woven fabric samples
 NOPHUGUE
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Figure S15 The weight residual percentages of L-menthol in the fluoroacrylic copolymer emulsion coating.

3.14 The filtration efficiency

The filtration efficiency of the wrap-coated non-woven fabric by DFMA-acrylic copolymer emulsion (40wt% DFMA) was tested by an automated filter tester (LTAO-506, Shanghai Litao Automation Technology Co., Ltd, China), which could generate NaCl and DEHS aerosol particles with a controlled flow rate (95L/min) and air resistance (88.87Pa). The aerosol particles were delivered through the fabrics and clamped by a filter holder with an effective area of 100cm².

	Filtrati	on efficiency (%)
Aerosoi particles size (µm)	NaCl	DEHS
0.3	99.9	85.9 (99.9%, double-layer)
0.5	99.9	89.5 (99.9%, double-layer)
1.0	99.9	99.0
2.5	99.9	99.9
5.0	99.9	99.9
10.0	99.9	99.9

Table S8 Filtration efficiency of the fabric treated by DFMA-acrylic copolymer emulsion (40wt% DFMA).

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