Supplementary Information for

Large-scale preparation of micro-gradient structured sub-micro fibrous membrane with narrow diameter distribution for high-efficiency air purification

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Table S1 The properties of PAN precursor solutions with different concentrations.

Solutions	Electrical Conductivity (μS cm^{-1})	Surface tension (mN m^{-1})	Viscosity (Pa s)
10.5%PAN	111.8	37.67	5.03
13%PAN	127.3	37.82	6.11
16%PAN	136.2	38.09	7.26

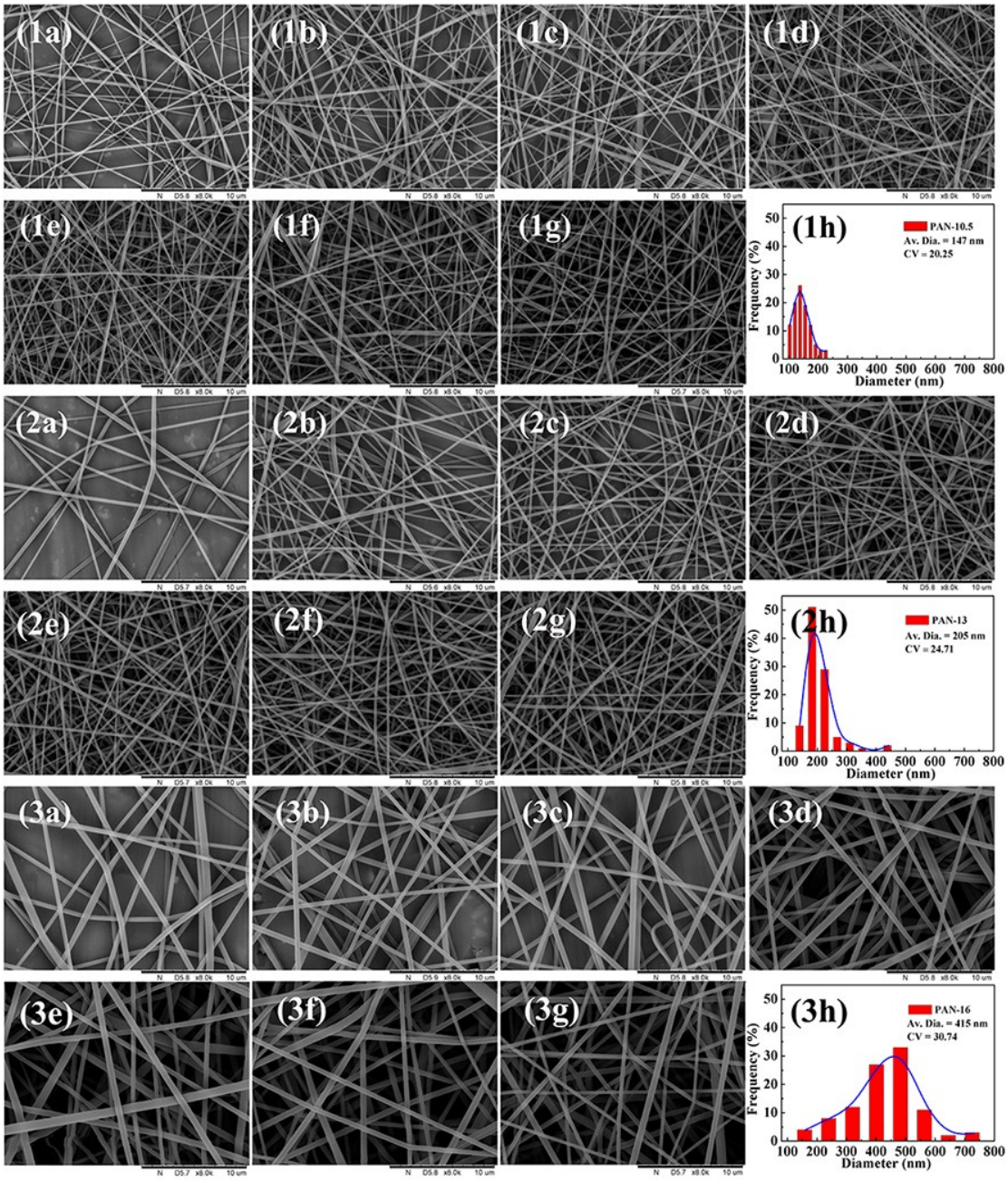

Fig. S1 SEM images of PAN-10 (1), PAN-13 (2), PAN-16 (3) sub-micro fibers with different fabric substrate feeding speeds of (a) 0.8 m min^{-1}, (b) 0.7 m min^{-1}, (c) 0.6 m min^{-1}, (d) 0.5 m min^{-1}, (e) 0.4 m min^{-1}, (f) 0.3 m min^{-1}, and (g) 0.2 m min^{-1}; (h) the diameter distributions of the sub-micro fibers.

Table S2 Structure attributes of PAN sub-micro fibrous membranes with fabric substrate feeding speeds of 0.6 m min^{-1}, 0.4 m min^{-1}, and 0.2 m min^{-1}.

Samples		Feeding speed of fabric substrate (m min^{-1})	Thickness (μm)	Basis density (g m^{-2})	Packing density
PAN-10.5	I	0.6	1.9375	0.196	0.0854
	II	0.4	3.25	0.644	0.1674
	III	0.2	5.9375	1.032	0.147
PAN-13	I	0.6	2.0625	0.224	0.0917
	II	0.4	3.75	0.596	0.1342
	III	0.2	6.5	0.956	0.1242
PAN-16	I	0.6	4.125	0.72	0.1474
	II	0.4	8.1875	1.656	0.1708
	III	0.2	10.5	2.42	0.1947

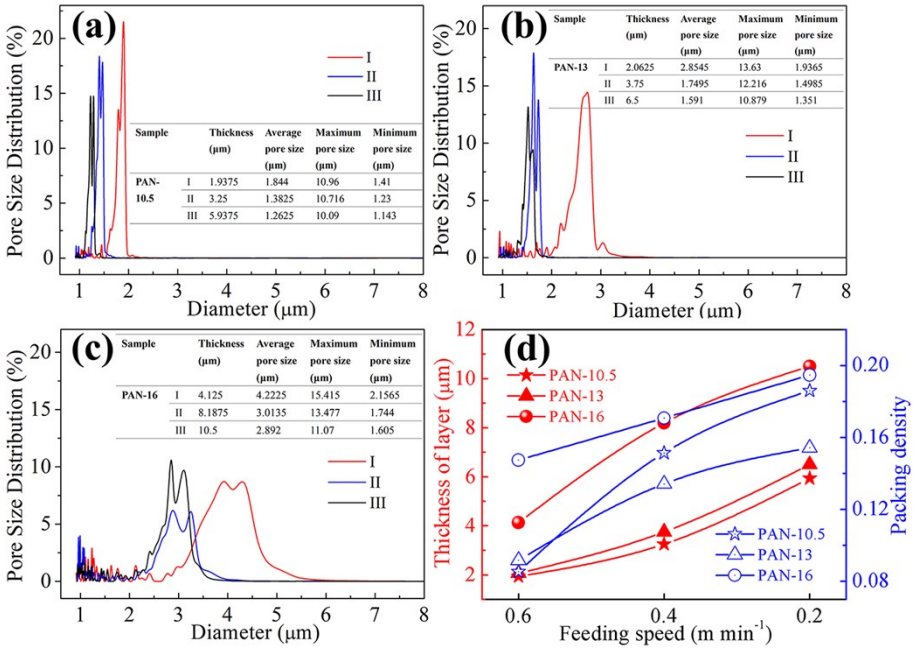

Fig. S2 Pore size distributions of PAN-10.5 (a), PAN-13 (b), and PAN-16 (c) sub-micro fibrous webs with different fabric substrate feeding speeds of 0.6 m min^{-1}, 0.4 m min^{-1}, and 0.2 m min^{-1}, thickness and packing density curves (d) of three kinds of PAN fibrous webs.

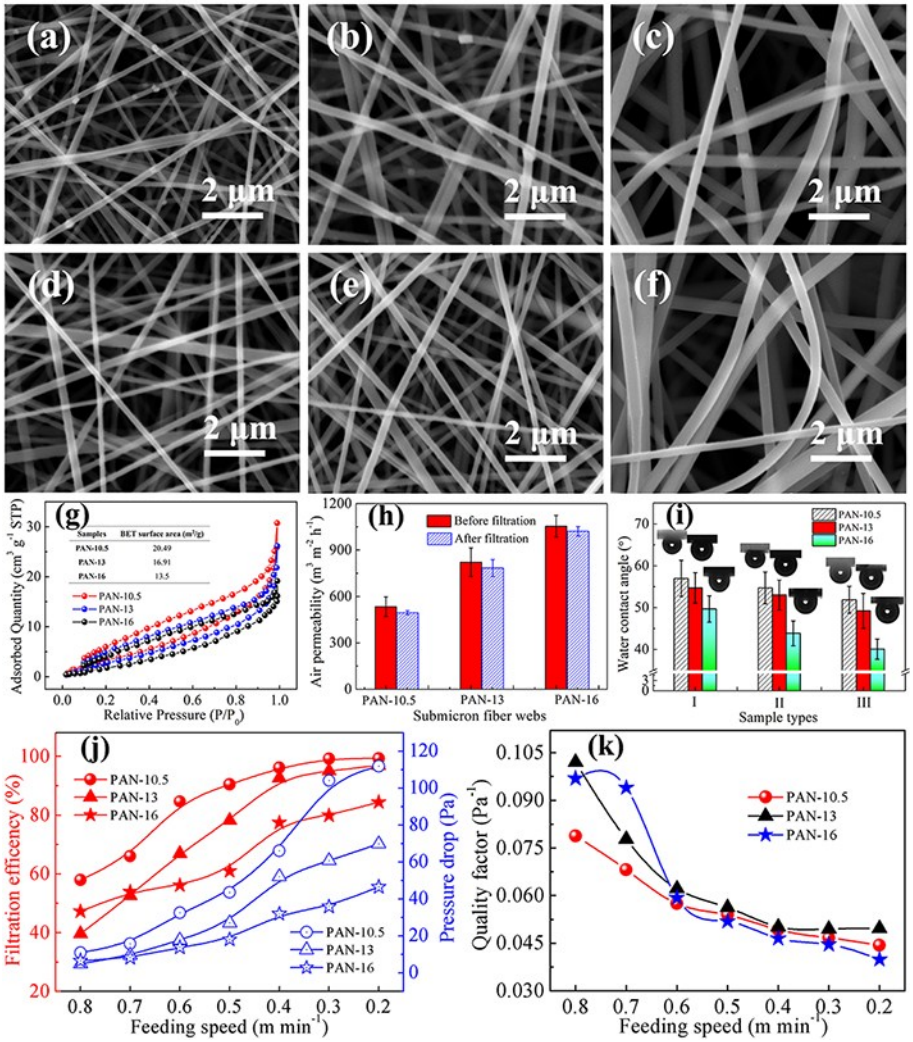

Fig. S3 SEM images of windward (a, b, c) and leeward (d, e, f) sides of PAN-10.5 (a, d), PAN-13 (b, e), and PAN-16 (c, f) fibrous webs for III type, N_2 adsorption-desorption isotherms (g), the air permeability before and after filtration (h), and the water contact angles (i) of PAN-III fibrous webs, the filtration efficiency and pressure drop (j), and Qf values (k) of PAN-10.5, PAN-13, and PAN-16 fibrous webs with different fabric substrate feeding speeds.

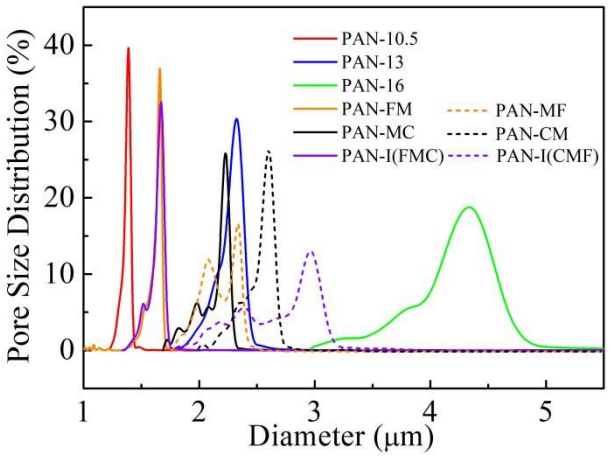

Fig. S4 Pore size distribution curves of PAN-I sample and various layers.

Table S3 Structure attributes and filtration performance of PAN-II composite membranes with different arrangements of three layers (C, M, F).

Sample	Structure	Packing density (g cm^{-2})	Thickness (μm)	MFP size (μm)	Pore size range (μm)	Filtration efficiency (%)	Pressure drop (Pa)	Qf (Pa^{-1})
PAN-II	CMF	1.792	8.25	2.29	1.70～3.717	99	32	0.0879
	FMC			1.544	1.384～2.331	99.278	55.86	0.0883
	MCF	1.76	8.31	1.583	1.293～2.63	99.85	145.0	0.0448
	FCM			1.346	1.245～2.028	99.88	146.0	0.046
	CFM	1.724	8.38	1.162	1.043～1.73	99.38	104.9	0.0485
	MFC			1.064	0.9553～1.58	99.46	113.7	0.0459

Table S4 Structure attributes and filtration performance of each layer, double layers and micro-gradient layers for PAN-II composite membrane with MCF structure.

Sample	Structure	Packing density (g cm^{-2})	Thickness (μm)	Filtration efficiency (%)	Pressure drop (Pa)	Qf (Pa^{-1})
PAN-II	C	1.32	4	54.8	16.66	0.0477
	M	0.34	3	92.09	48.02	0.0528
	F	0.304	1.69	97.85	60.76	0.0631
	FC	1.67	5.68	98.61	78.4	0.0545
	FCM	1.76	8.31	99.88	146.0	0.0461
	MCF			99.85	145.0	0.0448

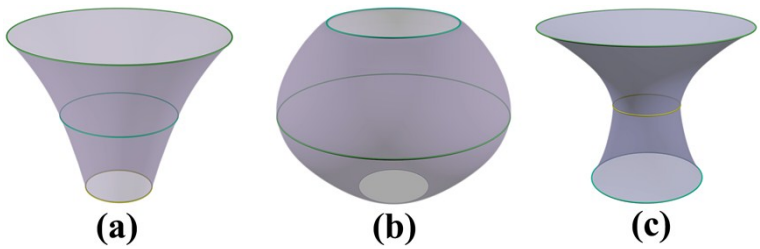

Fig. S5 Equivalent pore size diagrams of CMF (a), MCF (b), and CFM (c) structures for PAN-II hybrid membranes with different arrangements of C, M, and F fibrous layers.

Table S5 Pore size parameters of micro-gradient composite PAN/PMMA membrane for double sides.

Samples		MFP size (μm)	Pore size range (μm)
PAN/PMMA	FMC	1	0.9345～9.224
	CMF	1.4	1.27～9.662

The simple setup of PM filtration equipment was displayed in Fig. S6(a). A moxa stick burning was placed in the right bottle as the PMs generating device, which of the right side was sealed with plastic wrap with several small holes to ensure connecting with the atmosphere, and the left side was connected to the filter medium clamp and fibrous filter was inserted in it with effective area of 15.9 cm^2, the left bottle was connected with a circulating water pump providing circulating airflow passing through the test device. Various kinds of toxic gases were released by burning moxa stick, such as SO_2, CO, CO, benzene, xylenes, and so on, which of size was distributed broadly from 0.3 μm to 10 μm.[1] The white smoke with high PMs mass concerntration of >500 μg m^{-3} produced could diffuse from right bottle to left bottle without fibrous filter (Fig. S6(b)), conversely was blocked with fibrous filter, so the left bottle was transparent (Fig. S6(c)), reflecting the excellent filtration performance of prepared fibrous membrane.

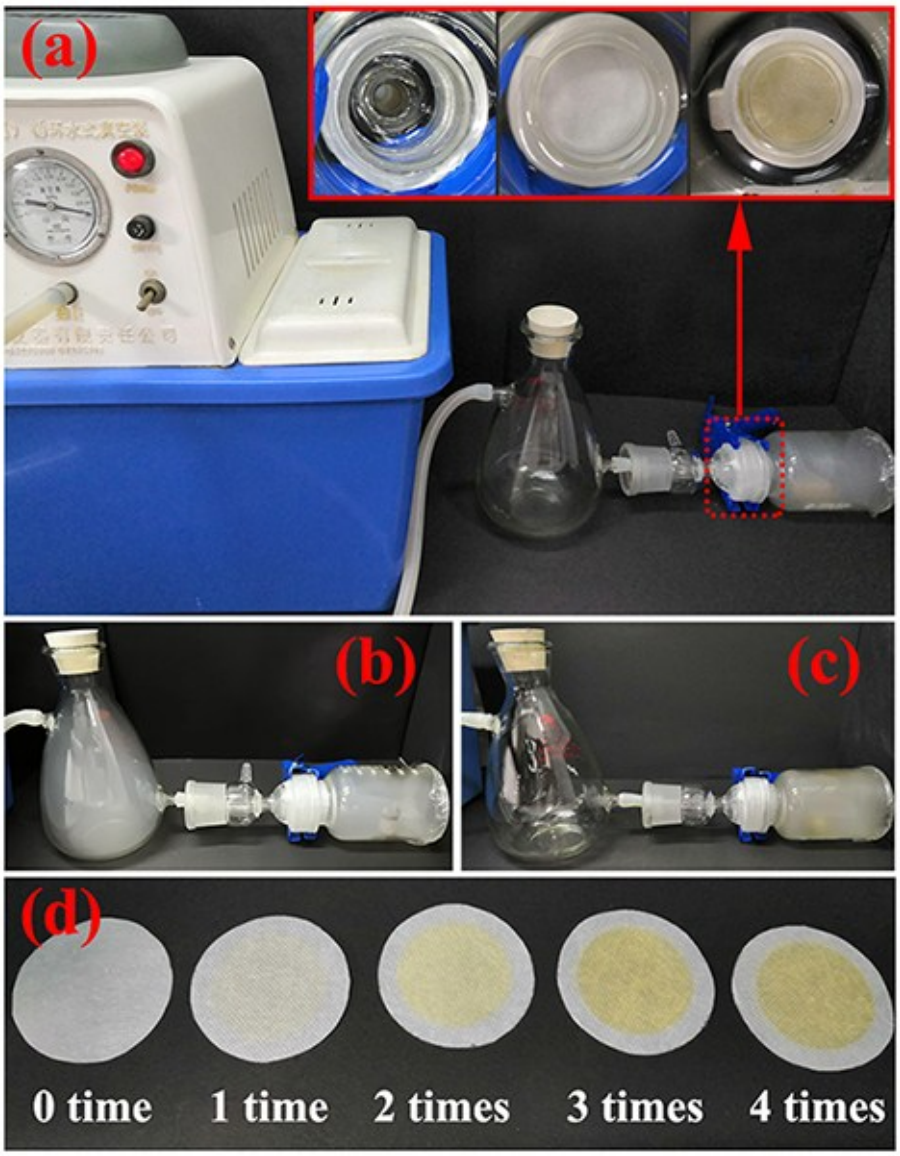

Fig. S6 PMs filtration equipment (a), the smoke diffused from right bottle to left bottle without fibrous filter (b), the smoke was blocked with prepared fibrous filter (c), and pictures of windward surface of fibrous filter after different filtration times.

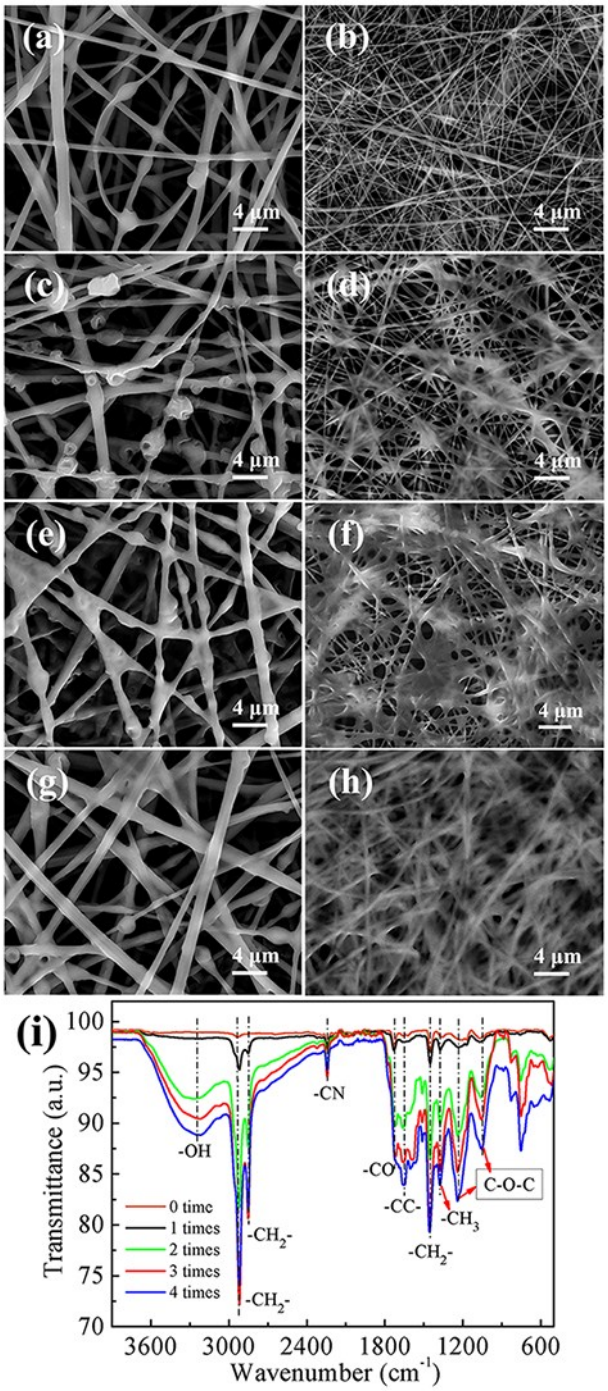

Fig. S7 SEM images of PAN-II composite membranes after $PM_{2.5}$ dynamic filtration with different filtration times of (a) and (b) 1 time, (c) and (d) 2 times, (e) and (f) 3 times, and (g) and (h) 4 times, among of (a, c, e, g) were the windward surface and (b, d, f, h) were the leeward surface; (i) FTIR spectroscopy for PAN-II composite membranes after filtration.

Table S6 Knudsen number and equilibrium factor values of fibrous layers for micro-gradient PAN/TiO_2, PAN/SWCNTs, and PAN/PMMA composite membranes.

Samples	PAN/TiO_2	PAN/SWCNTs	Micro-gradient PAN/PMMA composite		
Fibrous layer	F	F	C	M	F
d_f/nm	140	80	200	110	72
K_n	0.933	1.633	0.653	1.187	1.814

References

1. H. Gao, Y. Yang, A. Obed, J. Hou, H. Zhang, X. Qin, A low filtration resistance three-dimensional composite membrane fabricated via free surface electrospinning for effective PM 2.5 capture, *Environ. Sci. Nano*, 2017, **4**, 864-875.