

Supporting Information

Preparation of PMAA

Firstly, MAA were purified with thiophene by reduced pressure distillation. Second, potassium persulfate, purified MAA and deionized water were poured into a three-necked flask and stirred under nitrogen atmosphere at 80 °C. After polymerization reaction was kept for 8h, product was cooled at room temperature and subsequently washed by ethyl ether and DI water repeatedly. Finally, PMAA was dried in vacuum oven at 50 °C for 48 h.

Effect of environmental conditions

The adsorption capacity of Pb(II) at different pH values were evaluated in Fig. S1a. The results showed the adsorption capacity was low at pH 2 to 4. The reason was when pH value was lower than pKa of PMAA (pKa=4.2-4.9),⁵¹ high H⁺ concentration would inhibit the dissociation of -COOH and compete with Pb(II) for adsorption sites of the adsorbents. In addition, the carboxyl groups were more favored to bind with H⁺ instead of Pb(II) at acidic environment.²³ As pH value of the metal ion solution increased, carboxyl groups in PMAA tended to dissociate and molecule gradually stretched, increasing the electrostatic interaction with Pb(II).⁵² The result indicated the adsorption capacity of the CPNMs was affected by pH and PMAA content. With the PMAA content increasing, the adsorption capacity ascended from 37.6 to 53.8 mg g⁻¹ at pH=6.

Fast adsorption process was necessary for a qualified adsorbent in practical wastewater treatment.³⁴ The removal efficiencies of Pb(II) with different adsorption time was investigated and shown in Fig. S1b. At the initial stage, the adsorption rates of the CPNM to Pb(II) were relatively fast. That was because the carboxyl groups on the surface of the CPNM were abundant, which supplied many adsorption sites for Pb(II). As the contact time increased, Pb(II) gradually diffused into the PMAA shell. Besides, the electrostatic attraction and ion exchange interaction only took a very short time. As the contact time increased, Pb(II) gradually diffused into the PMAA shell. The Pb(II) concentration difference between the nanofibers and the ion solution led to slow adsorption rate after 20 min. All of the CPNMs can reached the adsorption equilibrium within 90 min, and the adsorption capacity of CPNM-1, CPNM-2 and CPNM-3 were 80.2, 55.4, and 43.6 mg g⁻¹, respectively.

It can be observed in Fig. 5c that the adsorption capacity of the CPNMs depended on the initial Pb(II) concentration. The q_e value witnessed a significant rise with the C_0 (Pb(II)) value ascending. Take CPNM-

1 as an example, the q_e value varied from 9.1 to 80.2 mg g^{-1} as the C_0 (Pb(II)) values went up from 10 to 100 mg L^{-1} , respectively. In the low C_0 (Pb(II)) solution (10 mg L^{-1}), the CPNM-1, CPNM-2 and CPNM-3 exhibited the optimum adsorption performance of Pb(II), and their rejection rates arrived at 95%, 87.2% and 84.3%, respectively.

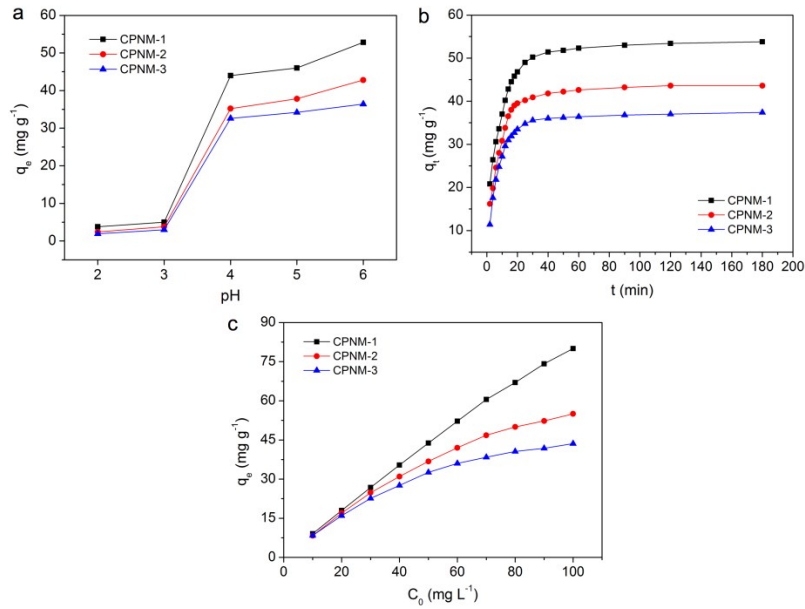


Fig. S1 Effects of (a) different pH values (25 °C, $C_0 = 60 \text{ mg L}^{-1}$, $t = 180 \text{ min}$), (b) contact time (25 °C, $C_0 = 60 \text{ mg L}^{-1}$, pH = 6), (c) initial concentration (25 °C, pH = 6, $t = 180 \text{ min}$) for Pb(II) adsorption by the CPNMs.

The metal ion adsorption capacity and removal efficiencies (% R) of the CPNM was calculated as the following equations (1) to (3):

$$q_t = \frac{(C_0 - C_t)V}{m} \quad (1)$$

$$q_e = \frac{(C_0 - C_e)V}{m} \quad (2)$$

$$\%R = \frac{(C_0 - C_e)}{C_0} \times 100 \quad (3)$$

where q_t (mg g^{-1}) is Pb(II) adsorption capacity of the CPNM at t min; q_e (mg g^{-1}) is the value at equilibrium stage.

The pseudo-first-order model, pseudo-second-order model and intra-particle-diffusion model, were established to describe the mechanism of the Pb(II) adsorption process. The linear form of the pseudo-first-

order model and pseudo-second-order model can be represented as ^{56, 57}:

$$\frac{1}{q_t} = \frac{k_1}{q_e t} + \frac{1}{q_e} \quad (4)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (5)$$

where k_1 (min^{-1}) and k_2 ($\text{g mg}^{-1} \text{min}^{-1}$) are the rate constants of the pseudo-first-order and pseudo-second-order adsorption kinetics, respectively; q_e and q_t are the adsorption capacity (mg g^{-1}) of Pb(II) at adsorption equilibrium and time t (min), respectively.

The diffusion rate of Pb(II) from the solution to the membranes was described by the intra-particle-diffusion kinetic, which was expressed as ^{58, 59}:

$$q_t = k_i t^{1/2} + C \quad (6)$$

where C is the constant parameter (mg g^{-1}) corresponding to the thickness of the boundary layer;⁵⁵ k_i is the intra-particle diffusion rate constant ($\text{mg g}^{-1} \text{min}^{-1/2}$).

The Langmuir and Freundlich models can be expressed as⁴⁴:

$$\frac{C_e}{q_e} = \frac{1}{q_{max} k_L} + \frac{C_e}{q_{max}} \quad (7)$$

$$\ln q_e = \frac{1}{n} \ln c_e + \ln k_F \quad (8)$$

where c_e (mg L^{-1}) is the residual metal concentration at equilibrium adsorption; q_e (mg g^{-1}) is the equilibrium adsorption capacity of Pb(II) on the adsorbent; and k_L (L mg^{-1}) and k_F (L mg^{-1}) are the Langmuir adsorption constant and the Freundlich constant, respectively.

The Pb(II) selectivity (α) is determined using Eq. (9) and (10). K_D is the distribution coefficient of metal ions (M^{n+}). The concentration factor (CF) of each ion is calculated using Eq. (11)^{51, 60}:

$$\alpha = \frac{K_D(\text{Pb}^{(II)})}{K_D(M^{n+})} \quad (9)$$

$$K_D = \frac{(c_0 - c_e)V}{c_e m} \quad (10)$$

$$CF = \frac{q_e(M^{n+})}{c_0(M^{n+})}$$

(11)

Table S1

Parameters of the CPNM properties.

Sample	Average D (nm)	Average d (nm)	Average t (nm)	Porosity (%)	Tensile strength (MPa)	
					Dry	Wet
CPNM-1	284.73 ± 28.85	168.07 ± 12.05	91.73 ± 1.67	77.6	1.24	1.57
CPNM-2	295.93 ± 28.16	249.55 ± 11.52	50.40 ± 1.93	76.5	1.39	1.96
CPNM-3	302.92 ± 28.09	275.01 ± 11.89	42.98 ± 1.82	75.8	1.58	2.05

Table S2

The parameters of three models for adsorption of Pb(II) by the CPNMs.

Model	Parameters	CPNM-1	CPNM-2	CPNM-3
Pseudo-first-order	R^2	0.9337	0.8494	0.7986
	$q_{e, cal}(\text{mg g}^{-1})$	16.96	14.71	10.33
	$k_1(\text{min}^{-1})$	0.0348	0.0374	0.0312
Pseudo-second-order	R^2	0.9998	0.9997	0.9999
	$q_{e, cal}(\text{mg g}^{-1})$	55.23	44.72	38.14
	$k_2(\text{g mg}^{-1} \text{min}^{-1})$	0.0046	0.0063	0.0086
Intra-particle diffusion	R_1^2	0.9989	0.9987	0.9887
	$k_1(\text{mg g}^{-1} \text{min}^{-1/2})$	9.40	8.44	8.41
	$C_1(\text{mg g}^{-1})$	7.47	4.45	0.45
	R_2^2	0.9795	0.9666	0.9758
	$k_2(\text{mg g}^{-1} \text{min}^{-1/2})$	3.86	2.68	2.50
	$C_2(\text{mg g}^{-1})$	29.33	27.18	22.09
	R_3^2	0.9218	0.8240	0.9786

k_3 (mg g ⁻¹ min ^{-1/2})	0.34	0.26	0.20
C_3 (mg g ⁻¹)	49.50	40.44	34.84

Table S3

Competitive metal ion uptake properties of CPNM-1 (pH=6; V=30 mL; m [adsorbent] = 30 mg; C₀ [Pb(II)] = 25 mg L⁻¹).

Metal ions	K _D (L g ⁻¹)	α (Pb(II)/M ⁿ⁺)	CF (L g ⁻¹)
Pb(II)	1.2656	1	0.5586
Cu(II)	0.5340	2.3697	0.5341
Cd(II)	0.1351	9.3656	0.1190
Ni(II)	0.1129	11.2098	0.1014

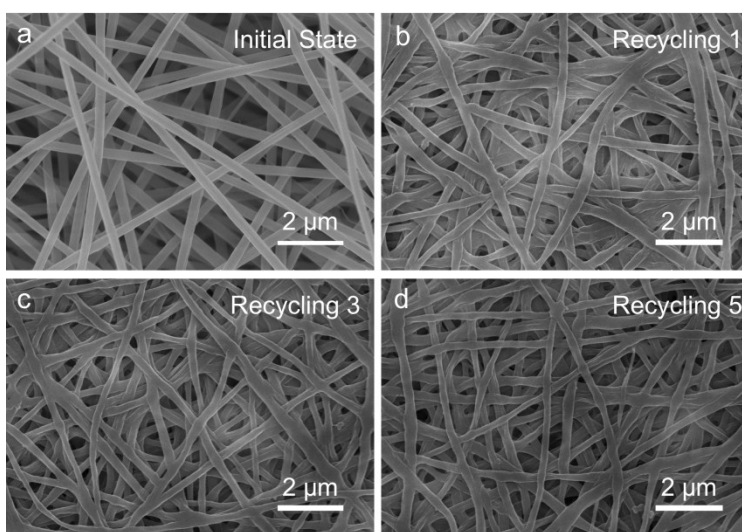


Fig. S2 SEM images of the CPNM before adsorption and after recycling 1, 3 and 5.

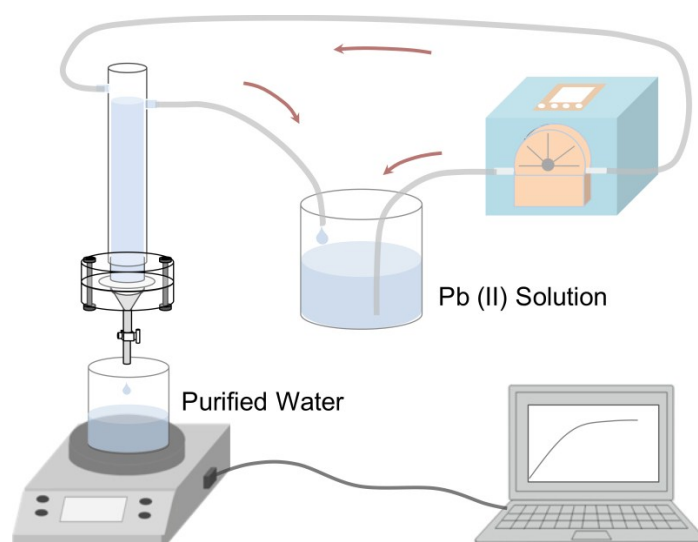


Fig. S3 Installation diagram of dynamic adsorption as a flow-through membrane.

It was known that adsorption of Pb (II) by the membrane belonged to the ion-exchange reaction in the flow-through system. In the initial stage of the adsorption process, external Pb (II) solution transported from the bulk to the surface of the membrane and then diffused through the whole membrane. The adsorption sites of the membrane were continuously occupied by Pb (II) from the solution. As a result, the ions concentration inside the membrane became gradually high, which brought about the further shrinkage of PMAA hydrogel shell and larger pore sizes of the membrane in microstructure. Therefore, it can be seen in Fig. S4 that the initial flux of the CPNM was $67.9 \text{ L m}^{-2} \text{ bar}^{-1} \text{ h}^{-1}$ and the flux gradually increased in the beginning stage of the adsorption process. In the later stage, the adsorption was mainly internal diffusion occurring inside the nanofibers, and the adsorption capacity of Pb (II) and the shrinkage of PMAA hydrogel shell tended to be stable. Therefore, after operating for 60 h, the flux tended to stabilize, and the maximum flux was $554 \text{ L m}^{-2} \text{ bar}^{-1} \text{ h}^{-1}$.

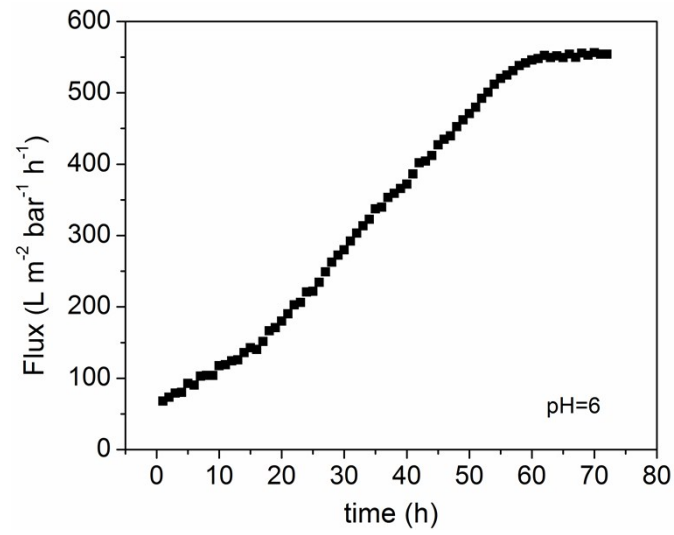


Fig. S4 Continuous filtration: flux value of the CPNM in Pb^{2+} aqueous solution (pH=6, $C_0 = 10\ mg\ L^{-1}$, $25\ ^\circ C$).