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Supporting information

Unconventional and facile production of stimuli-responsive multifunctional system for simultaneous drug delivery and environmental remediation

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1. Batch adsorption experiments

The current study explored the adsorption performance of NCST3 fiber membrane for Pb^{2+} and Cd^{2+} ions explored under different pH, contact time, and initial concentration. All the adsorption experiments were performed at 30 °C and 120 rpm. The concentration of metal ions was measured by inductively coupled plasma atomic emission spectrometry (ICP-AES). Required concentrations of Pb^{2+} and Cd^{2+} ion standards were prepared by appropriate dilution of the above HMIs stock solution. When measuring the influence of pH, 0.1 mol/L NaOH and HCl were utilized to adjust the pH of the solution from 2 to 10. The batch adsorption studies were performed in a centrifugal tube to find the optimal pH values. The following studies, all were performed under pH 6.0. The adsorption kinetics at the initial concentrations of 25, 50, 75, and 100 mg/L were studied by setting different reaction times, and the adsorption equilibrium time was obtained. An isotherm study was conducted in the range of the initial HMIs concentration of 10-600 mg/L to explore the maximum adsorption capacity of the NCST3 membrane. All adsorption measurements were performed in triplicate, and the average of those measurements was deliberated with an error bar. The amount of HMIs adsorbed on the adsorbent was calculated by using the following equation:[1,2]

$$q_e = \frac{(C_0 - C_e)V}{M} \quad \text{(S1)}$$

$$R\% = \frac{(C_0 - C_e)}{C_0} \times 100 \quad \text{(S2)}$$

Where, q_e (mg/g) is adsorption capacity, C_0 (mg/L) is the initial concentration of HMIs solution before adsorption, C_e (mg/L) is the remaining concentration of HMIs solution after adsorption. R

(%) is the removal rate of HMIs, *M* (mg) is the mass of NCST3 and V (mL) is the volume of HMIs solution.

2. Adsorption isotherms

To explore their adsorption type, Langmuir isotherm model (S3), and Freundlich isotherm model (S4) were utilized to fit the adsorption data, the following equations:

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \quad \text{(S3)}$$

$$q_e = K_F C_e^{\frac{1}{n}} \quad \text{(S4)}$$

Among them, q_m (mg/g) is the maximum adsorption capacity. ***KL*** (L/mg) is the Langmuir equilibrium constant and is related to the affinity of the adsorbent and the adsorption site. ***KF*** ((mg/g)/(mg/L)$^{1/n}$) is the Freundlich constant related to adsorption strength and capacity, ***n*** indicates the coefficient of the Freundlich model.

The separation factor (R_L) was calculated using the following equation:

$$R_L = \frac{1}{1 + K_L C_0} \quad \text{(S5)}$$

3. Adsorption kinetics

In order to explore the adsorption kinetics process of Pb^{2+} and Cd^{2+} on NCST3 membrane, 100 mg of adsorbent and 100 mL HMIs solution with different concentration at pH 6 incubated for a certain time (10-60 min). To explore the adsorption mechanism, the adsorption kinetic data has fitted with the pseudo-first-order (S6) and the pseudo-second-order (S7) model by using the following equations,

$$\ln\left(q_e - q_t\right) = \ln q_e - k_1 t \quad \text{(S6)}$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_t} \quad \text{(S7)}$$

Among them, q_e and q_t (mg/g) are the adsorption capacities at time t and at equilibrium, respectively; k_1 and k_2 (g/mg · min) are the adsorption rate constants of the pseudo-first-order equation and the pseudo-second-order equation, respectively.

4. Selectivity tests

To a 100 mL aqueous solution of Pb^{2+}, Cd^{2+}, Li^{+}, Mg^{2+}, Cu^{2+}, $Ni^{+,}$ and Zn^{2+} ions in centrifuge tube or flask with a magnetic stir bar, NCST3 adsorbent (100 mg) was added. The resulting mixture was stirred at room temperature. At predetermined time intervals, aliquots (4 mL) were taken from the mixture, and the adsorbent was separated. The concentrations of metal ions in the resulting solutions were measured by ICP-AES.

5. Regeneration tests

For reusability tests, the NCST3 fibers were retrieved by filtration from heavy metal ions solution and washed thoroughly with DI water to eliminate any residual metal solution. Next, the metal ions adsorbed composite fibers were submerged into an HCl solution (0.1 M) and shaken at 120 rpm for 20 min. ICP-AES was used to determine the desorbed metal ions. The adsorption/desorption cycle was reiterated six times using the same NCST3 fibers to evaluate the reusability performance.

NMR spectra of f-CNT

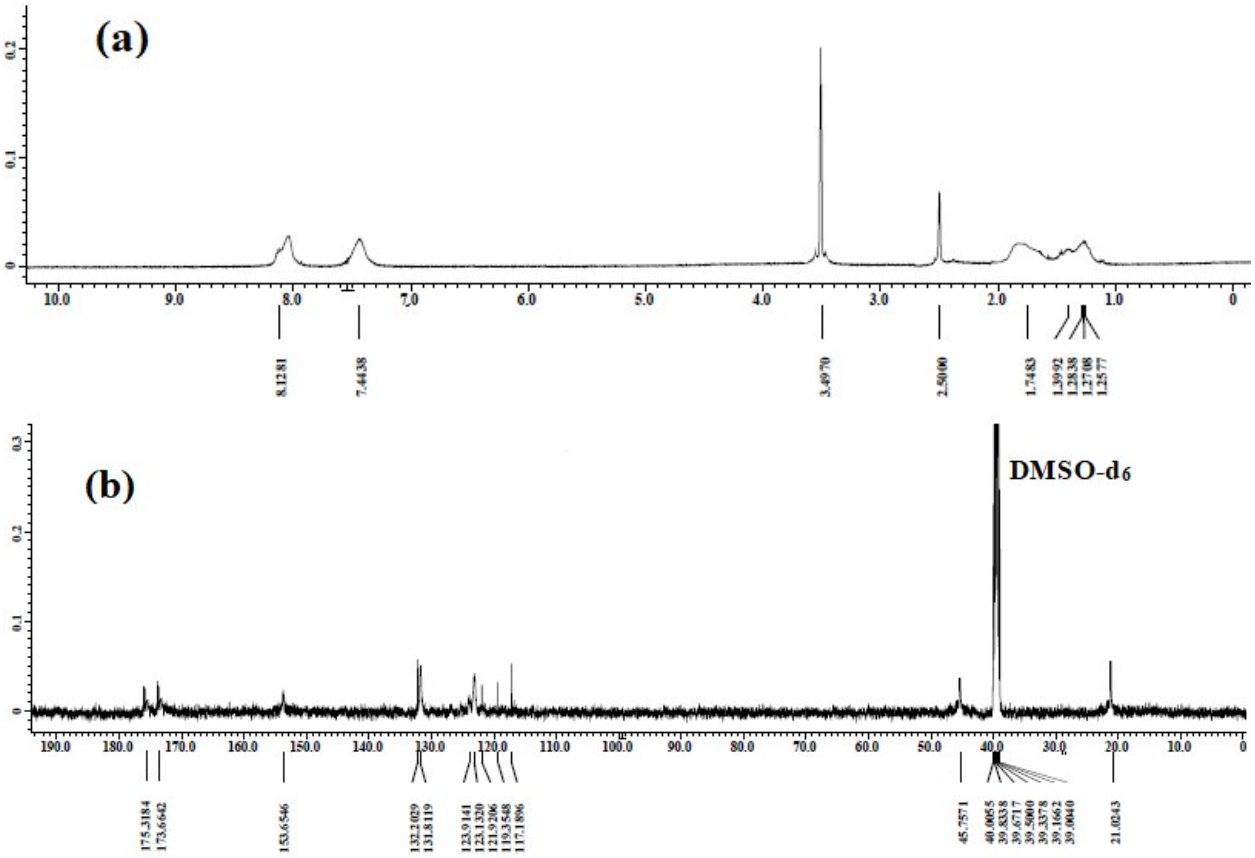

Figure S1. (a) ^{1}H-NMR and (c) ^{13}C-NMR of f-CNT.

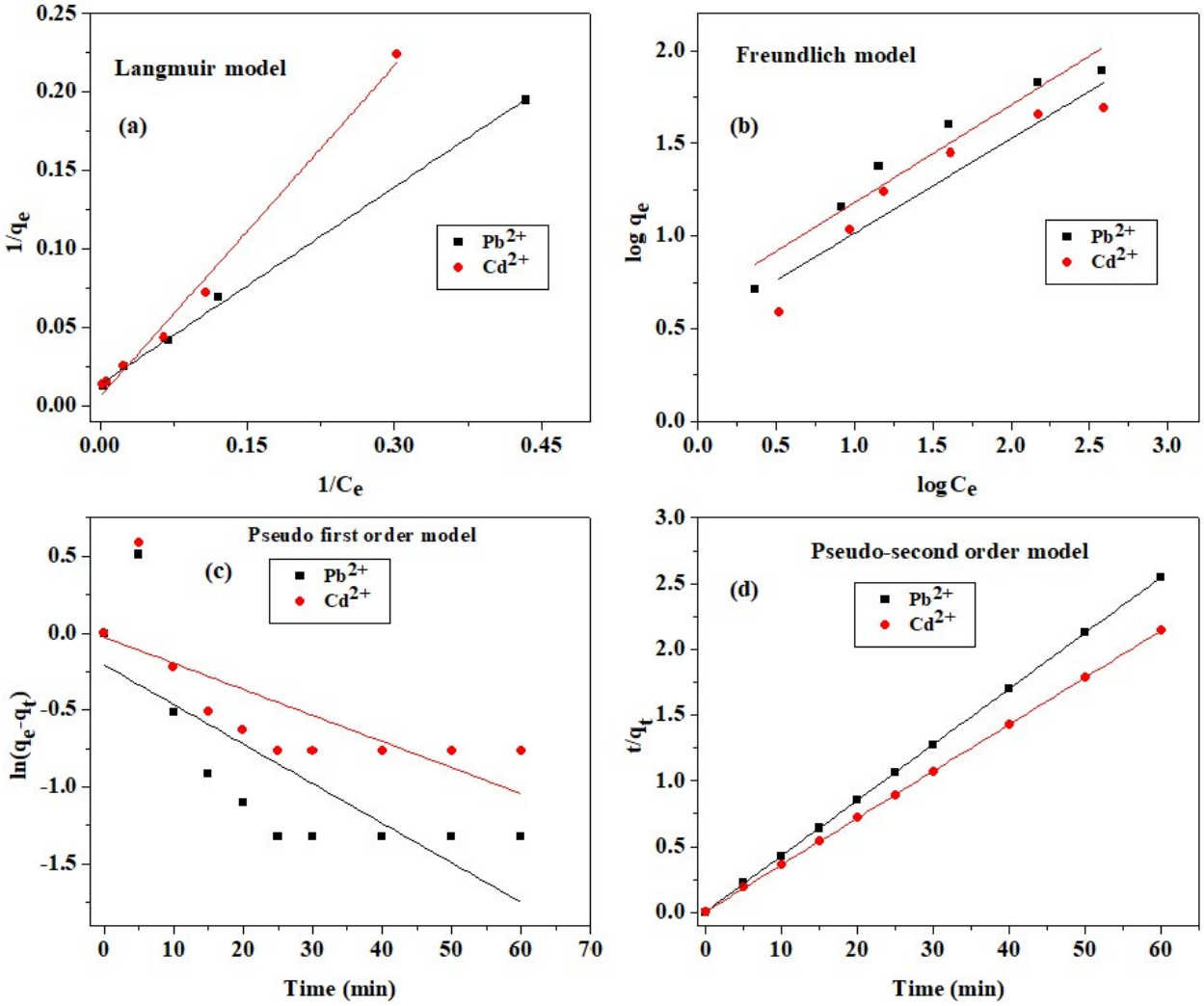

Figure S2. Langmuir (a), and Freundlich (b) isotherm models of HMIs onto NCST3 membrane. (c) Pseudo-first-order and (d) pseudo-second-order kinetic models for adsorption Pb^{2+} and Cd^{2+} ions from wastewater using NCST3 adsorbent.

Table S1. Isotherm model constants for the adsorption of Pb^{2+} and Cd^{2+} ions

HMIs		**Langmuir isotherm**			**Freundlich isotherm**		
	q_{max} (mg/g)	**K_L**	**R_L**	**R^2**	**K_f**	**$1/n$**	**R^2**
Pb^{2+}	211.9395	0.0327	0.3795	0.999	4.5236	0.5269	0.929
Cd^{2+}	167.224	0.0085	0.7017	0.991	2.4951	0.4013	0.877

Table S2. Parameters of Pseudo-first-order and Pseudo-second-order Kinetic models

HMIs			**Pseudo-first-order**			**Pseudo-second-order**		
	Conc. (ppm)	$q_{e\ Exp.}$ (mg/g)	$q_{e\ Cal.}$ (mg/g)	K_1 (min^{-1})	R^2	$q_{e\ Cal.}$ (mg/g)	K_2 (min^{-1})	R^2
Pb^{2+}	25	12.1	1.1	-1.1×10^{-4}	0.344	11.9	0.63501	0.999
	50	23.8	0.81	-4.3×10^{-4}	0.544	23.6	0.31864	0.999
	75	35.7	1.2	-3.3×10^{-4}	0.438	35.6	0.21175	0.999
	100	39.8	0.81	-1.2×10^{-4}	0.181	39.6	0.18877	0.999
Cd^{2+}	25	11.5	0.74	-1.2×10^{-4}	0.495	11.7	0.65466	0.999
	50	23.1	0.97	-2.8×10^{-4}	0.480	23.4	0.32545	0.999
	75	35.0	0.35	-1.2×10^{-4}	0.125	35.1	0.21819	0.999
	100	39.1	0.85	-1.8×10^{-4}	0.455	39.3	0.6500	1.000

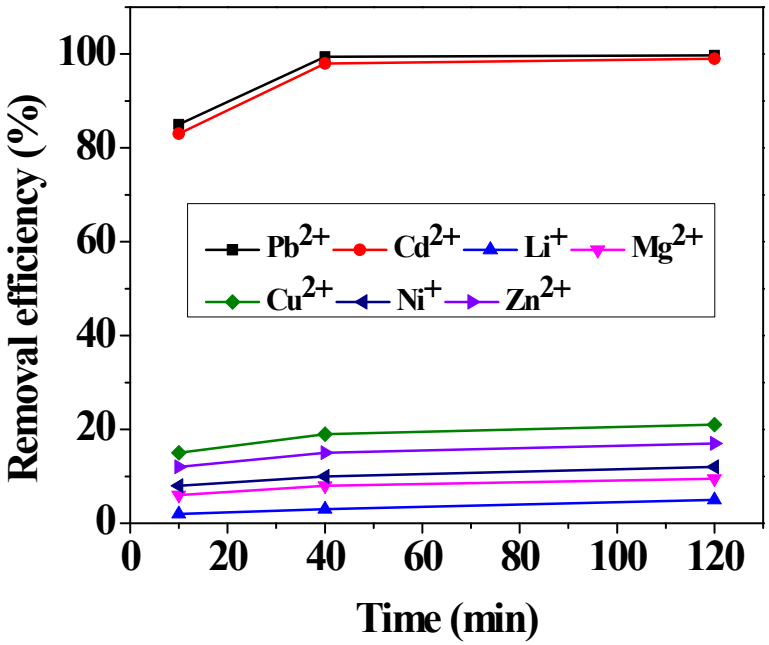

Figure S3. Time-dependent selective removal efficiency of heavy metal ions in the presence of NCST3 adsorbent.

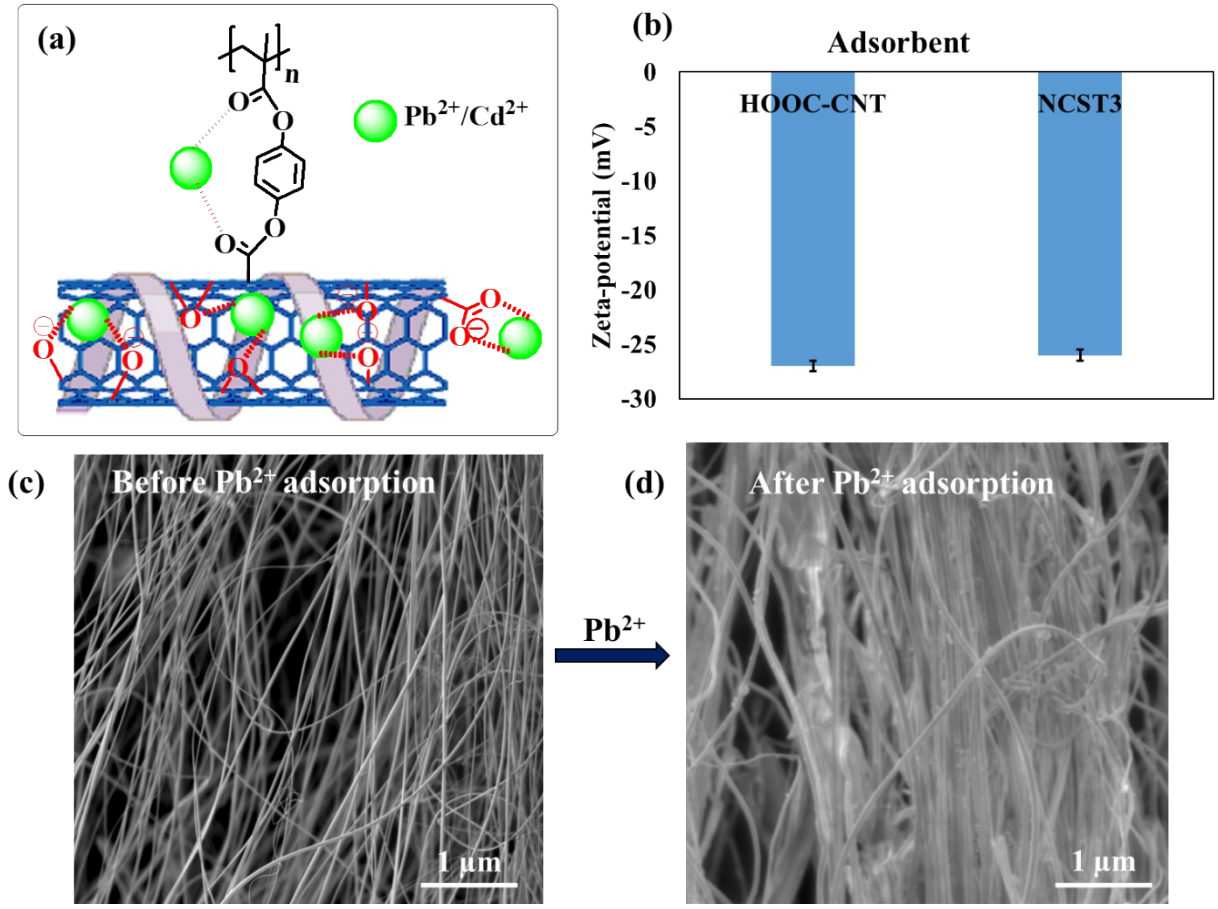

Figure S4. (a) Schematic illustration of proposed interactions between NCST3 adsorbent and HMIs, (b) Zeta potential of HOOC-CNT and NCST3 adsorbent, and (c) SEM image of NCST3 adsorbent before Pb^{2+} adsorption, and (d) SEM image of NCST3 adsorbent after Pb^{2+} adsorption.

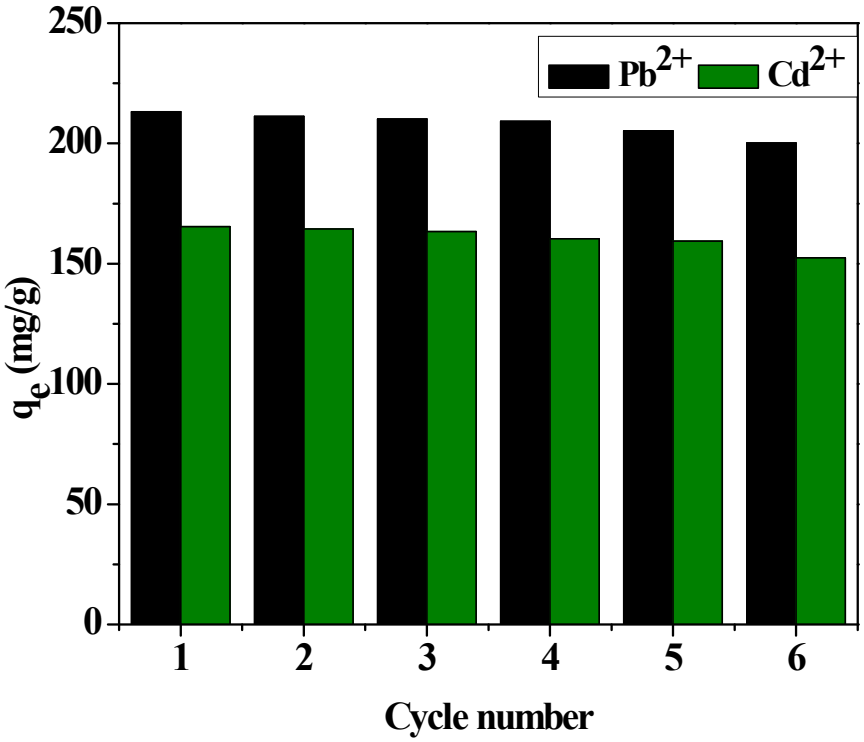

Figure S5. Adsorption capacities of Pb^{2+} and Cd^{2+} on NCST3 membrane after six times regeneration.

References

1. J. Tang, Y. Chen, M. Zhao, S. Wang and L. Zhang, Phenylthiosemicarbazide-functionalized UiO-66-NH_2 as highly efficient adsorbent for the selective removal of lead from aqueous solutions, *J. Hazard. Mater.*, 2021, 125278. https://doi.org/10.1016/j.jhazmat.2021.125278.
2. A. Ayub, Z. A. Raza, M. I. Majeed, M. R. Tariq, A. Irfan, Development of sustainable magnetic chitosan biosorbent beads for kinetic remediation of arsenic contaminated water, *International Journal of Biological Macromolecules*, 2020, **163**, 603-617.