## **Supporting Information**

# Alginate-based supermacroporous hydrogel fabricated by cryo-polymerization for uranium extraction from seawater

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#### **Details of adsorption experiment**

To prepare the uranium-containing aqueous solution of specific concentration, first, accurately weigh 2.11 g of UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O and dissolve it in 100 mL of deionized water. Next, transfer the solution to a volumetric flask and adjust the volume to 1 L to obtain a 1000 mg L<sup>-1</sup> U(VI) stock solution. Dilute the stock solution to prepare the desired concentration of the uranium-containing aqueous solution for subsequent use. To conduct the adsorption experiment in the uranium-containing aqueous solution, add a certain mass of adsorbent into a conical flask filled with 50 mL or 100 mL of the specific concentration of uranium-containing aqueous solution. Place the flask in a constant temperature water bath shaker for a certain period of time, with the temperature set to 25°C and the rotation speed set to 160 r min<sup>-1</sup>. Measure the U(VI) concentration before and after the adsorption reaction using a UV-Vis spectrophotometer or ICP-MS. Finally, calculate the adsorption capacity of U(VI) using Eq. (S1) and (S2).

$$R = \frac{C_0 - C_e}{C_0} \times 100\%$$
 (S1)

$$q_e = \frac{\left(C_0 - C_e\right) \times V}{m} \tag{S2}$$

where R (%) is the removal rate,  $q_e (\text{mg g}^{-1})$  is the adsorption capacity,  $C_0 (\text{mg L}^{-1})$ <sup>1</sup>) and  $C_e (\text{mg L}^{-1})$  are the initial and equilibrium concentrations of U(VI), V (L) is the solution volume, and m (g) is the mass of the adsorbent.

To determine the optimal amount of adsorbent for the material, an aqueous solution containing U(VI) was used with a concentration of 50 mg g<sup>-1</sup>, a volume of 50 mL, and varying amounts of adsorbent (5, 7.5, 10, 15, 20, 30, 40, and 50 g). To identify the optimal adsorption pH for the material, the optimal adsorbent dosage was used and the pH of the aqueous solution containing U(VI) was adjusted to 2.0, 3.0, 4.0, 5.0, 6.0, 7.0, 8.0, and 9.0 using 0.5 M HNO<sub>3</sub> and 0.05 M Na<sub>2</sub>CO<sub>3</sub>. To study the adsorption kinetics during the process, the pH of the aqueous solution containing U(VI) was adjusted to the optimal adsorption pH, and small samples were taken at intervals after adsorption began (1, 2, 3, 4, 6, 8, 12, 16, 24, 36, 48, 60, and 72 h). To reduce the influence of sampling on the solution volume, the solution volume was set to 100 mL. To evaluate the adsorption kinetics of the materials, the pseudo-first-order adsorption kinetics <sup>1</sup>, pseudo-second-order adsorption kinetics <sup>2</sup>, and internal diffusion model <sup>3</sup> were used to fit the adsorption kinetics, according to Eqs. (S3), (S4), and (S5), respectively.

$$\ln\left(q_{\rm e}-q_{\rm t}\right) = \ln q_{\rm e} - k_{\rm l}t \tag{S3}$$

$$\frac{t}{q_{t}} = \frac{t}{q_{e}} + \frac{1}{k_{2}q_{e}^{2}}$$
(S4)

$$q_{\rm e} = k_{ip} t^{1/2} + C \tag{S5}$$

where  $q_t (\text{mg g}^{-1})$  is the adsorption amounts at equilibrium time and contact time,  $k_1 (h^{-1})$  and  $k_2 g (\text{mg} \cdot h)^{-1}$  are the pseudo primary adsorption kinetic constants and pseudo secondary adsorption kinetic constants, respectively.  $k_{ip} \text{ mg } (g \cdot h^{0.5})$  is the internal diffusion constant.

In order to examine the effect of temperature and initial concentration on the adsorption performance of the materials, initial concentrations of 50, 100, 150, 200, 250, 300, 350, and 400 mg L<sup>-1</sup> were tested at temperatures of 298.15 K, 308.15 K, and 318.15 K. To evaluate the adsorption isotherms of the materials, Langmuir <sup>4</sup>, Freundlich <sup>5</sup>, and Temkin <sup>6</sup> adsorption isotherm models were fitted using Eq. (S6), (S7), and (S8), respectively.

$$\frac{C_{\rm e}}{q_{\rm e}} = \frac{C_{\rm e}}{q_{\rm m}} + \frac{1}{K_{\rm L}q_{\rm m}} \tag{S6}$$

$$\ln q_{\rm e} = \frac{1}{n} \ln C_{\rm e} + \ln K_{\rm F} \tag{S7}$$

$$q_{\rm e} = \frac{\mathbf{R}T}{b_{\rm T}} \ln C_{\rm e} + \frac{\mathbf{R}T}{b_{\rm T}} \ln K_{\rm T}$$
(S8)

where  $q_{\rm m}$  (mg g<sup>-1</sup>) are the adsorption amount at equilibrium and the theoretical maximum adsorption amount,  $K_{\rm L}$  (L mg<sup>-1</sup>) is the Langmuir adsorption constant,  $K_{\rm F}$  [(mg g<sup>-1</sup>)·(L mg<sup>-1</sup>)<sup>1/n</sup>] is the Freundlich adsorption constant, *n* is the expression function of adsorption strength,  $K_{\rm T}$  (L mg<sup>-1</sup>) is for the Temkin isotherm constant, *b* (J mol<sup>-1</sup>)·(g mg<sup>-1</sup>) is for the Temkin constant related to the heat of adsorption, and *R* [J (mol·K)<sup>-1</sup>] is for the ideal gas constant 8.314.

To evaluate the thermodynamics of the adsorption process, the standard equilibrium constant  $K^{\Theta}$  <sup>7,8</sup> was calculated using the Langmuir adsorption constant based on Eqs. (S6) and (S9). The van't Hoff isotherm equation and the Gibbs free energy

defining equation were then applied according to Eq. (S10) and (S11) for further analysis.

$$K^{\Theta} = K_{\rm L} \times M_{\rm ad} \times 10^3 \times C^{\Theta} \tag{S9}$$

$$\ln K^{\Theta} = \frac{\Delta S^{\Theta}}{R} - \frac{\Delta H^{\Theta}}{RT}$$
(S10)

$$\Delta G^{\Theta} = \Delta H^{\Theta} - T \Delta S^{\Theta} \tag{S11}$$

where  $K^{\Theta}$  is the standard equilibrium constant,  $M_{ad}$  is the molecular weight of the adsorbent,  $C^{\Theta}$  (mol L<sup>-1</sup>) is the standard concentration 1,  $\Delta H^{\Theta}$  (kJ mol<sup>-1</sup>) is the standard enthalpy change,  $\Delta S^{\Theta}$  J (mol·K)<sup>-1</sup> is the standard entropy change,  $\Delta G^{\Theta}$  (kJ mol<sup>-1</sup>) is the standard Gibbs free energy change, and T (K) is the ambient temperature.

To screen for the best desorbent, adsorption experiments were conducted using a 50 mg L<sup>-1</sup> aqueous solution containing U(VI), and the adsorbed material was eluted with 50 mL of H<sub>2</sub>O, 0.1 M HNO<sub>3</sub>, 0.1 M NaOH, 0.1 M Na<sub>2</sub>EDTA, and 1 M Na<sub>2</sub>CO<sub>3</sub>+0.1 M H<sub>2</sub>O<sub>2</sub>. The eluted material was then placed in a constant temperature water bath oscillator for a specified time period. To investigate the adsorption-desorption cycle performance of the material, the adsorption experiment was first conducted using a 50 mg L<sup>-1</sup> aqueous solution containing U(VI), and then the best desorption solution was selected for elution. After the elution, the material was washed with deionized water until neutral, dried in an oven at 90°C, and subjected to another adsorption-desorption cycle experiment. The equilibrium adsorption amount  $q_e$  and desorption rate  $D_s$  were calculated using Eq. (S2) and (S12), respectively.

$$D_{\rm s} = \frac{C_{\rm d}}{C_0 - C_{\rm e}} \times 100\%$$
(S12)

where,  $D_{\rm s}$  (%) is the desorption rate and  $C_{\rm d}$  (mg g<sup>-1</sup>) is the concentration of U(VI) in the desorption solution.

To investigate the selective adsorption performance of the material towards U(VI), a 0.2 mM ion competition solution was prepared containing Na<sup>+</sup>, Mg<sup>2+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Sr<sup>2+</sup>, Ba<sup>2+</sup>, Co<sup>2+</sup>, Ni<sup>2+</sup>, Zn<sup>2+</sup> cations (all nitrates) and U(VI). The adsorption experiments were conducted and the equilibrium adsorption amount ( $q_e$ ) of each ion was calculated individually using Eq. (S2). The distribution coefficient ( $K_d$ ) of each ion was also calculated using Eq. (S13).

$$K_{\rm d} = \frac{C_0 - C_{\rm e}}{C_{\rm e}} \times \frac{V}{m}$$
(S13)

where  $K_d$  (mL g<sup>-1</sup>) is the distribution coefficient.

Furthermore, VO<sub>3</sub><sup>+</sup>, Mn<sup>2+</sup>, Fe<sup>3+</sup>, Co<sup>2+</sup>, Ni<sup>2+</sup>, Cu<sup>2+</sup> and Zn<sup>2+</sup>, which have similar concentrations to U(VI) in seawater, were chosen for adsorption experiments using a competitive solution with U(VI) formulated at 100 times the ion concentration in seawater <sup>9,10</sup>. To assess the adsorption selectivity of the materials, the equilibrium adsorption amount  $q_e$  for each ion was calculated using Eq. (S2), and the distribution coefficient  $K_d$  for each ion was calculated using Eq. (S13).

To investigate the adsorption performance of the materials on U(VI) in real seawater, 35 g of sea salt was added to 1 L of deionized water to prepare real seawater. A 1 mg L<sup>-1</sup> U(VI) stock solution was added to 50 mL of real seawater to prepare spiked 10, 20, 50, 100, and 200  $\mu$ g L<sup>-1</sup> real seawater. The pH of real seawater containing U(VI) was adjusted to 8 using 0.05 M Na<sub>2</sub>CO<sub>3</sub>. After reaching adsorption equilibrium, the remaining U(VI) concentration was detected using ICP-MS.

In addition, a dynamic seawater adsorption device was built, as shown in **Fig. S10**. The device consists of an adsorption column, a diaphragm pump, a storage tank, and a controller. The adsorption column comprises two cylindrical polyurethane sponges sandwiching the AO-PAM/Alg hydrogel in the middle. The remaining space inside the column is filled with glass beads. The pH value of the real seawater was adjusted to 8.0 using 0.05 M Na<sub>2</sub>CO<sub>3</sub>. A small amount of the solution was periodically taken, and the U(VI) concentration was detected by ICP-MS.

#### **Theoretical calculation**

Simulations were conducted using Gaussian16 software <sup>11</sup>. Initially, we optimized the geometry of all models using the density functional theory (DFT) B3LYP exchangecorrelation functional. Standard 6-311G(d) basis sets with dispersion functions were employed to describe carbon, hydrogen, oxygen, and nitrogen atoms <sup>12,13</sup>. The uranium atom was described using the Stuttgart quasi-relativistic effective core potential (RECP) along with the corresponding optimized basis set, which was combined with a small core RECP to represent the 60 core electrons in uranium for geometric optimization and single-point energy calculations<sup>14</sup>. Subsequently, to enhance the accuracy of energy calculations, we used the PBE1PBE functional for all models, and the all-electron basis set def2-TZVP was employed to describe light atoms <sup>15</sup>. To incorporate dispersion interactions, we calculated empirical corrections for all models using the DFT-D3 method. All calculations were performed using the solvation model based on density (SMD) for the water model. The binding energy  $(E_{ad})$  of the coordination structure model was calculated using Eq. (S14).

$$E_{\rm ad} = E_{\rm AO-PAM/Alg-U(VI)} - E_{\rm AO-PAM/Alg} - E_{\rm U(VI)}$$
(S14)

The surface electrostatic potential analysis of the adsorbed mass was conducted through a combination of the wave function analysis program Multiwfn 3.7 <sup>16</sup>and Gaussian 16 <sup>11</sup> software. The resulting isosurface maps were rendered using the VMD software <sup>17</sup>.



Fig. S1 The pore size distribution of PAM/Alg and AO-PAM/Alg hydrogel



Fig. S2 TGA (a) and DTG (b) curves of PAM/Alg and AO-PAM/Alg



Fig. S3 Photos of AO-PAM/Alg hydrogel after soaking in 1 M NaOH (a) and HCl (b)

for 5 days



Fig. S4 SEM images of AO-PAM/Alg hydrogel after soaking in 1 M NaOH (a and b)

and HCl (c and d) for 5 days



Fig. S5 FT-IR curves of AO-PAM/Alg hydrogels after immersion in 1 M NaOH and

HCl for 5 days



Fig. S6 Zeta potentials of PAM/Alg and AO-PAM/Alg hydrogel at different pH.



Fig. S7 Photos of AO-PAM/Alg before and after adsorption and elution



Fig. S8 FT-IR curves of AO-PAM/Alg and AO-PAM/Alg-U



Fig. S9 XRD curves of AO-PAM/Alg and AO-PAM/Alg-U



Fig. S10 Schematic diagram of continuous dynamic real seawater experiment

equipment

	Median pore	Total intrusion	Total pore		
Samples	diameter (area) volume		area	Porosity	
	μm	mL g <sup>-1</sup>	$m^2 g^{-1}$	%	
PAM/Alg	43.80	2.39	0.19	78.88	
AO-PAM/Alg	59.19	1.90	0.12	73.06	

Table S1 The mercury intrusion porosimetry (MIP) analysis results

Total		Langmuir		Freundlich			Temkin		
Isotnerm	$q_m$	$K_L$	23	$K_F$		53	$b_T$	$K_T$	D
model	mg g <sup>-1</sup>	L mg <sup>-1</sup>	$R^2$	$(mg g^{-1}) \cdot (L mg^{-1})^{1/n}$	n	K²	$(J \text{ mol}^{-1}) \cdot (g \text{ mg}^{-1})$	L mg <sup>-1</sup>	K²
298.15 K	1413.0	0.094	0.999	233.404	2.546	0.916	17.510	1.226	0.986
308.15 K	1526.6	0.161	0.999	333.003	2.791	0.906	18.120	2.798	0.979
318.15 K	1633.6	0.232	0.996	436.566	3.056	0.899	19.130	6.131	0.969

Kinetics model	Pseudo-first-order		Pseudo-second-order		Intra-particle diffusion				
	$q_e$	$k_1 \times 10^{-2}$	$R^2$	$q_e$	k <sub>2</sub> ×10 <sup>-3</sup>	$R^2$	Step	$k_{ m ip}$	$R^2$
	mg g <sup>-1</sup>	h-1	n	mg g <sup>-1</sup>	g (mg·h) -1	R	I	mg (g·h <sup>0.5</sup> ) <sup>-1</sup>	
	58.9 1.788					Step 1	9.061	0.981	
PAM/Alg		1.788	0.959	9 66.9	1.581	0.990	Step 2	5.489	0.985
							Step 3	4.971	0.994
	lg 202.4 5.419 0.958						Step 1	64.304	0.991
AO-PAM/Alg		251.3	1.163	0.999	Step 2	26.219	0.989		
					Step 3	3.422	0.802		

Table S3 Kinetics model parameters.

Т	$\Delta \ G^{\Theta}$	$\Delta H^{\Theta}$	$\Delta S^{\Theta}$
K	kJ mol <sup>-1</sup>	kJ mol <sup>-1</sup>	kJ (mol·K) <sup>-1</sup>
298.15 K	-80.496		
308.15 K	-82.532	35.831	203.619
318.15 K	-84.569		

 Table S4 Adsorption thermodynamic parameter

Coordination mode	$E_{\rm ad}$ (kcal mol <sup>-1</sup> )	no. donor <sup>a</sup>	Charge transfer (a.u.) <sup>b</sup>
mode I	-15.706	5	0.167
mode II	-20.785	5	0.251
mode III	-32.374	5	0.355
mode IV	-42.532	5	0.552
mode V	-37.051	5	0.555

Table S5 The calculated stability data of uranyl ion/complex and charge transfer

<sup>a</sup> Number of donor atoms of uranyl

<sup>b</sup> Based on Mulliken charge

	Adsorptio	$q_{ m m}$		
Adsorbent	U(VI) concentration	T	- (mg g-1)	Refs
	(mg L-1)	g L <sup>-1</sup> )		
U/Sr-IP2	40	T =298 K, pH =7.0	316.5	18
Alginate Beads	78	T =298 K, pH = 5.0	237.1	19
nZVI@Alg-Ca beads	280	T =318 K, pH = 3.0	204.1	20
Ca-alginate beads	100	T=298 K, pH = 4.0;	400.0	21
aMSP/SA	1000	T =313 K, pH = 4.0	210.0	22
SA/CMC-Ca-Al	200	T = 298  K,  pH = 4.0	101.7	23
ZIF-67/SAP <sub>0.45</sub>	200	T = 318.15  K, seawater	510.8	24
SA/PVA/PEO/ZSM-5	500	T = 318.15 K, pH = 5.0	92.7	25
MF-A aerogel	300	T = 298.15 K, pH = 4.0	211.22	26
AO-PAM/Alg hydrogel	400	<i>T</i> = 318.15 K, pH = 6.0	1633.6	This work

 Table S6 Comparison of the U(VI) adsorption capacity of AO-PAM/Alg hydrogel

with those of the previously reported adsorbents

**Table S7** Comparison of AO-PAM/Alg hydrogel and amidoxime functionalized

 adsorbents reported in the past three years in the adsorption capacity of U(VI) in real

 seawater

Adsorbents	Forms	Adsorpt	$q_{ m m}$	Refs	
		Adsorption time	Condition	- (mg g <sup>-1</sup> )	
AO-PAM/Alg	Hydrogel	25 days	V = 20  L; m = 3  mg	6.23	This work
MP-PAO	Hydrogel	28 days	V = 100  L; m = 10  mg	5.8	27
PAO-CB	Hydrogel	56 days	V = 100 L; $m = 10$ mg	8.56	28
PLMR	Resin	10 days	<i>V</i> =200 T; <i>m</i> =300 g	2.14	29
MS@PIDO/Alg	Sponge	56 days	V = 1 T; $m = 36$ mg	1.87	30
PAF-CS	Porous	60 days	$V = 100 \text{ I} \cdot m = 5 \text{ mg}$	8 07	31
TAI-C5	Powder	00 days	V = 100  L; m = 3  mg	0.72	
CI-PAO	Membrane	4 weeks	V = 100  L; m = 10  mg	6.17	32
5-AFM	Membrane	56 days	Soaked in seawater	7.46	33
PAO-BSPE	Membrane	33 days	Soaked in seawater	5.6	34
AM3	Membrane	10 weeks	Soaked in seawater	6.03	35
PAO PNM	Membrane	35 days	m = 3  mg	9.35	36
AOBS-M	Fiber	30 days	Soaked in seawater	0.97	37
PAO-co-AA	Fiber	27 days	V = 30  L; m = 10  mg	5.4	38
FF-PT	Fiber	30 days	m = 100  mg	3.22	39
CID NFs	Fiber	87 days	V = 25  L; m = 5  mg	11.39	40
PAO-PHMB-A	Fiber	30 days	Soaked in seawater	3.19	41
PAN-NH <sub>2</sub> -AO	Fiber	91 days	Soaked in seawater	0.312	42
PAO/Alg NFs	Fiber	8 weeks	V = 8 T; $m = 10$ mg	8.42	43

### Reference

- 1 H. O. Y. S. and G. Mckay, *Water Res.*, 2000, **34**, 735–742.
- 2 Y. S. Ho and G. Mckay, Process Biochem., 1999, 34, 451–465.
- 3 G. H. Graaf, H. Scholtens, E. J. Stamhuis and A. A. C. M. Beenackers, *Chem. Eng. Sci.*, 1990, **45**, 773–783.
- 4 L. I, J. Am. Chem. Soc., 1916, 38, 2221–2295.
- 5 F. H, Zeitschrift Fur Physikalische Chemie Stochiometrie Und Verwandtschaftslehre, 1906, **57**, 385–470.
- 6 J. R. D and A. F. H, Biochim. Biophys. Acta, 1995, 1247, 293–297.
- 7 H. N. Tran, S.-J. You and H.-P. Chao, J. Environ. Chem. Eng., 2016, 4, 2671–2682.
- 8 X. Zhou and X. Zhou, Chem. Eng. Commun., 2014, 201, 1459–1467.
- 9 C. W. Abney, R. T. Mayes, T. Saito and S. Dai, *Chem. Rev.*, 2017, **117**, 13935– 14013.
- 10S. Xie, X. Liu, B. Zhang, H. Ma, C. Ling, M. Yu, L. Li and J. Li, *J. Mater. Chem. A*, 2015, **3**, 2552–2558.
- 11 M. J. Frisch, G. W. Trucks, H. B. Schlegel, G. E. Scuseria, M. A. Robb, J. R. Cheeseman, G. Scalmani, V. Barone, G. A. Petersson, H. Nakatsuji, X. Li, M. Caricato, A. V. Marenich, J. Bloino, B. G. Janesko, R. Gomperts, B. Mennucci, H. P. Hratchian, J. V. Ortiz, A. F. Izmaylov, J. L. Sonnenberg, D. Williams-Young, F. Ding, F. Lipparini, F. Egidi, J. Goings, B. Peng, A. Petrone, T. Henderson, D. Ranasinghe, V. G. Zakrzewski, J. Gao, N. Rega, G. Zheng, W. Liang, M. Hada, M. Ehara, K. Toyota, R. Fukuda, J. Hasegawa, M. Ishida, T. Nakajima, Y. Honda, O. Kitao, H. Nakai, T. Vreven, K. Throssell, J. A. Montgomery, J. E. Peralta, F. Ogliaro, M. J. Bearpark, J. J. Heyd, E. N. Brothers, K. N. Kudin, V. N. Staroverov, T. A. Keith, R. Kobayashi, J. Normand, K. Raghavachari, A. P. Rendell, J. C. Burant, S. S. Iyengar, J. Tomasi, M. Cossi, J. M. Millam, M. Klene, C. Adamo, R. Cammi, J. W. Ochterski, R. L. Martin, K. Morokuma, O. Farkas, J. B. Foresman, D. J. Fox and I. Abánades Lázaro, Gaussian 16, Revision C.01 Gaussian, Inc., Wallingford, CT 2019.
- 12D. Feller, J. Comput. Chem., 1996, 17, 1571-1586.
- 13K. L. Schuchardt, B. T. Didier, T. Elsethagen, L. Sun, V. Gurumoorthi, J. Chase, J. Li and T. L. Windus, J. Chem. Inf. Model., 2007, 47, 1045–1052.
- 14M. Dolg, H. Stoll, H. Preuss and R. M. Pitzer, J Phys Chem, 1993, 97, 5852-5859.
- 15F. Weigend and R. Ahlrichs, Phys. Chem. Chem. Phys., 2005, 7, 3297-3305.
- 16T. Lu and F. Chen, J. Comput. Chem., 2012, 33, 580-592.
- 17 W. Humphrey, A. Dalke and K. Schulten, J. Mol. Graphics, 1996, 14, 33-38.
- 18J. Yin, S. Yang, W. He, T. Zhao, C. Li and D. Hua, Sep. Purif. Technol., 2021, 271, 118849.
- 19J. Yu, J. Wang and Y. Jiang, Nucl. Eng. Technol., 2017, 49, 534-540.

- 20S. Hu, X. Lin, Y. Zhang, M. Shi and X. Luo, *J. Radioanal. Nucl. Chem.*, 2017, **314**, 2405–2416.
- 21 C. Gok and S. Aytas, J. Hazard. Mater., 2009, 168, 369-375.
- 22X. Jiang, H. Wang, Q. Wang, E. Hu and Y. Duan, J. Cleaner Prod., 2020, 247, 119162.
- 23 L. Wu, X. Lin, X. Zhou and X. Luo, Appl. Surf. Sci., 2016, 384, 466-479.
- 24Z. Bai, Q. Liu, H. Zhang, J. Yu, R. Chen, J. Liu, D. Song, R. Li and J. Wang, ACS Appl. Mater. Interfaces, 2020, 12, 18012–18022.
- 25F. Zahakifar, A. R. Keshtkar and M. Talebi, *Prog. Nucl. Energy*, 2021, **134**, 103642.
- 26Y.-R. He, X.-L. Li, X.-L. Li, Z.-Y. Tan, D. Zhang and H.-B. Chen, *J. Mol. Liq.*, 2019, **289**, 111154.
- 27H. Wang, B. Zheng, T. Xu, M. Cao, F. Gao, G. Zhou, C. Ma, J. Dang, W. Yao, K. Wu, T. Liu, Y. Yuan, Q. Fu and N. Wang, *Sep. Purif. Technol.*, 2022, 289, 120823.
- 28 Y. Yuan, X. Guo, L. Feng, Q. Yu, K. Lin, T. Feng, B. Yan, K. V. Fedorovich and N. Wang, *Chem. Eng. J.*, 2021, **421**, 127878.
- 29S. Wen, Y. Sun, R. Liu, L. Chen, J. Wang, S. Peng, C. Ma, Y. Yuan, W. Gong and N. Wang, ACS Appl. Mater. Interfaces, 2021, 13, 3246–3258.
- 30D. Wang, J. Song, S. Lin, J. Wen, C. Ma, Y. Yuan, M. Lei, X. Wang, N. Wang and H. Wu, Adv. Funct. Mater., 2019, 29, 1901009.
- 31 Z. Li, Q. Meng, Y. Yang, X. Zou, Y. Yuan and G. Zhu, Chem. Sci., 2020, 11, 4747– 4752.
- 32H. Wang, T. Xu, B. Zheng, M. Cao, F. Gao, G. Zhou, C. Ma, J. Dang, W. Yao, K. Wu, T. Liu, Y. Yuan, Q. Fu and N. Wang, *J. Hazard. Mater.*, 2022, 433, 128789.
- 33 R. Yu, Y. Lu, X. Zhang, W. Chen, X. Chen and L. Li, *Desalination*, 2022, **539**, 115965.
- 34X. Xu, C. Huang, Y. Wang, X. Chen, Z. Wang, J. Han, M. Wu, G. Liu, L. Li, L. Xu and H. Ma, *Chem. Eng. J.*, 2022, 430, 133159.
- 35 R. Yu, X. Zhang, Y. Lu, W. Chen, X. Chen and L. Li, *ACS Sustainable Chem. Eng.*, 2022, **10**, 12307–12318.
- 36S. Shi, Y. Qian, P. Mei, Y. Yuan, N. Jia, M. Dong, J. Fan, Z. Guo and N. Wang, Nano Energy, 2020, 71, 104629.
- 37 Y. Wang, Z. Lin, Q. Liu, J. Zhu, J. Liu, J. Yu, R. Chen, P. Liu and J. Wang, Chem. Eng. J., 2021, 425, 131538.
- 38F. Chi, S. Zhang, J. Wen, J. Xiong and S. Hu, J. Mater. Sci., 2018, 54, 3572-3585.
- 39 Y. Pu, T. Qiang and L. Ren, Int. J. Biol. Macromol., 2022, 206, 699-707.
- 40C. Huang, L. Xu, X. Xu, L. Ma, H. Bao, J. Liao, J. Wang, J. Han, G. Xu, D. Huang,
  B. Ye, H. Zhang, M. Wu, X. Zhao and H. Ma, *Chem. Eng. J.*, 2022, 443, 136312.
- 41N. He, H. Li, L. Li, C. Cheng, X. Lu, J. Wen and X. Wang, *J. Hazard. Mater.*, 2021, **416**, 126192.
- 42 H. Gu, P. Ju, Q. Liu, G. Sun, J. Liu, R. Chen, J. Yu, J. Zhu and J. Wang, J. Colloid

Interface Sci., 2022, 610, 1015–1026.

43 X. Xu, Y. Yue, D. Cai, J. Song, C. Han, Z. Liu, D. Wang, J. Xiao and H. Wu, *Small Methods*, 2020, **4**, 2000558.