

Supporting information for:

Facile fabrication of modified polyamide acid porous membrane for uranium enrichment in wastewater

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1. Adsorption procedure

Uranium adsorption test of CPAA membrane

Regardless of the kinetics or the isothermal uranium adsorption of CPAA membrane, 10 mg CPAA membrane was placed into a certain concentration of uranium solution, and 0.5 mL sample solution was taken out at regular intervals until the adsorption was completed. Take the adsorption kinetic adsorption test as a representative. At 25 °C, CPAA membrane (10 mg) was transferred into a conical flask containing 250 mL, 20 ppm uranium solution, and the mixture was stirred at 300 rpm. The sample solutions (0.5 mL) were collected at 0, 0.5, 1, 2, 3, 4, 6, 8, 10, 12, 24, 36, and 48 hours, respectively.

Determination of uranium concentration

In order to obtain the concentration of uranium solution at different adsorption times, arsenazo III was used as a chromogenic agent, and the absorbance of the mixed solution at 652 nm was measured using UV-Vis. The specific steps were as follows:

Drawing of standard curve: $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (0.21097 g) was carefully transferred into a 100 mL volumetric flask, and the ultrapure water was added to obtain the uranium standard solution (1000 ppm). Afterwards, measure 50, 100, 150, 200, 250, 300 μL of the standard solution into the 10 mL volumetric flasks, and standard solutions with uranium concentrations at 5, 10, 15, 20, 25, 30 ppm can be achieved, respectively. Subsequently, the obtained uranium solution (0.5 mL), hydrochloric acid (0.1 mol/L, 0.5 mL), arsenazo III (1g/L, 1 mL) and ultrapure water (2 mL) were sufficiently mixed and poured into a quartz cuvette for UV-Vis tests. The absorbances (A) at 652 nm were recorded, and the absorbances *versus* uranium concentration linear regression equation can be thus obtained.

Similarly, for sample solution with unknown uranium concentration, sample solution (0.5 mL), hydrochloric acid (0.1 mol/L, 0.5 mL), arsenazo III (1g/L, 1 mL) and ultrapure water (2 mL) were fully mixed and transferred into a quartz cuvette to measure the absorbance at 652 nm. According to the above linear regression equation,

the uranium concentration of each sample can be achieved, and the uranium adsorption amount can be further calculated by the following equation:

$$q_t = (C_0 - C_t)V/m$$

2. Equations

Equation S1

$$q_t = k_{ip}t^{0.5} + C \quad (1)$$

Where q_t (mg/g) and t (h) are the uranium-adsorbed amount at the contact time and the contact time, respectively. k_{ip} is the internal diffusion constant. C is the intercept of the equation.

Equation S2:

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

Where q_t (mg/g) and q_e (mg/g) are the uranium-adsorbed amounts of CPAA membrane at the contact time and adsorption equilibrium time, respectively. t (min) is the contact time and k_2 (g/(mg min)) is the rate constant.

Equation S3:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (3)$$

Where q_t (mg/g) and q_e (mg/g) are the uranium-adsorbed amounts at the contact time and adsorption equilibrium time, respectively. t (min) is the contact time and k_1 (min^{-1}) is the rate constant.

Equation S4:

$$K_d = \frac{(C_0 - C_e)}{C_e} \times \frac{V}{m} \quad (4)$$

Where C_0 (mg L^{-1}) is the initial concentration of uranium, C_e (mg L^{-1}) is the concentration at equilibrium, V (mL) is the volume of simulated seawater, and m (g) is the mass of the GPAA membrane.

Equation S5 and Equation S6

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{k_L q_m} \quad (5)$$

$$q_e = k_F C_e^{\frac{1}{n}} \quad (6)$$

Where C_e (mg/L) and q_e (mg/g) are separately the uranium concentrations in the simulated seawater and the uranium adsorption amounts of the adsorbent at equilibrium; q_m (mg/g) is the adsorption capacity; k_L (L/mg) is the Langmuir adsorption constants; k_F and n are the Freundlich adsorption constants.

3. Figures

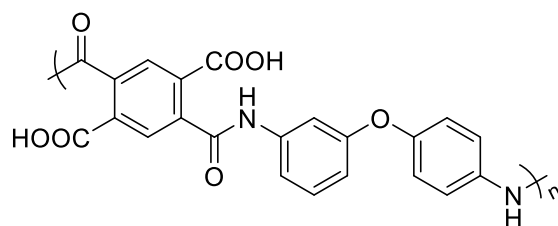


Figure S1 Molecular structure of PAA in this work.

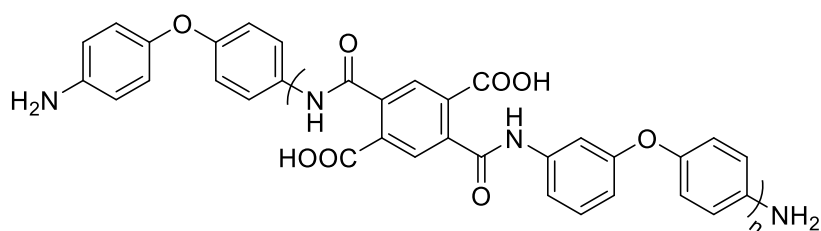


Figure S2 Molecular structure of NPAA in this work.

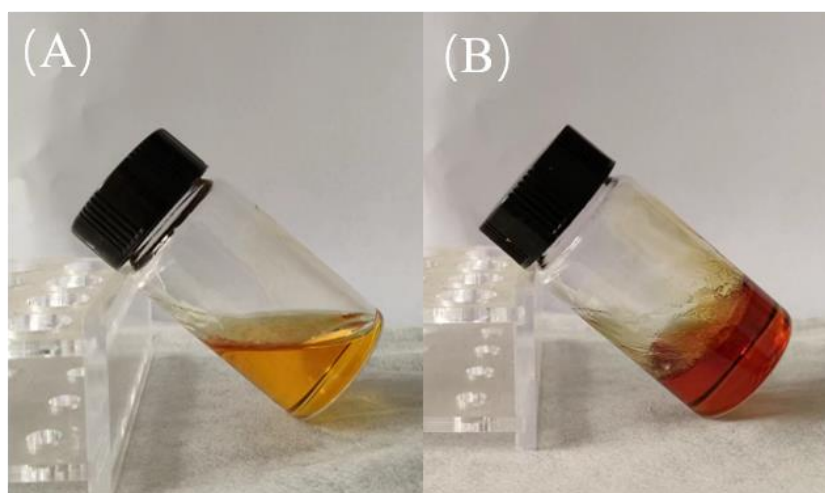


Figure S3 Digital photographs of PAA+glutaraldehyde (A) and NPAA+glutaraldehyde (B). (Note: the molar ratio in digital photograph A, PMDA:ODA:glutaraldehyde=1:1:0.035; the molar ratio in digital photograph B: PMDA:ODA:glutaraldehyde=1:1.01:0.035)

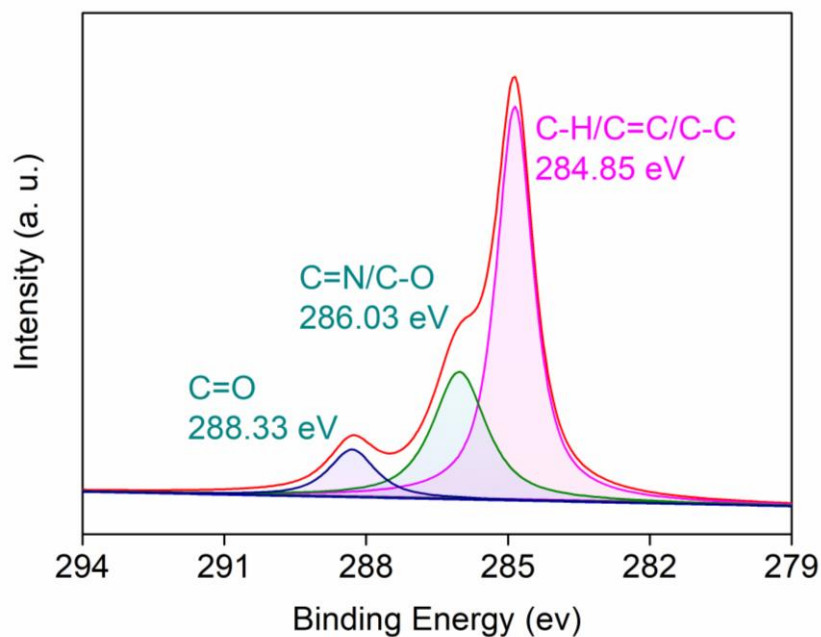


Figure S4 High resolution C1s XPS profile of GPAA membrane.

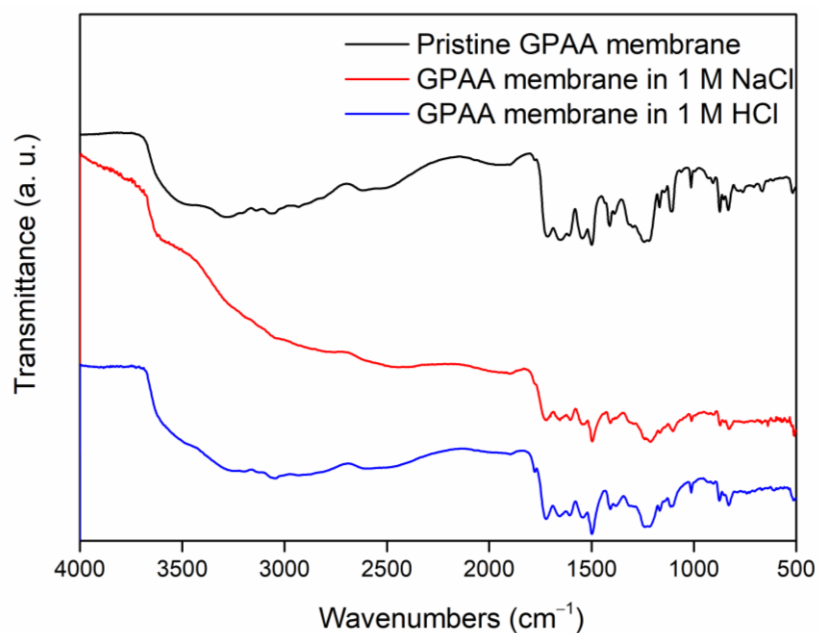


Figure S5 FTIR spectra of GPAA membranes in different treatment conditions. Pristine GPAA membrane, GPAA membranes soaked in 1 M NaCl and 1 M HCl for four weeks, respectively.

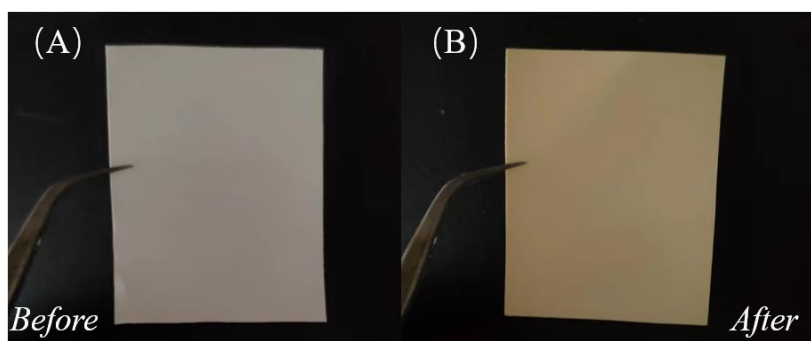


Figure S6 Digital photographs of GPAA membranes before and after uranium adsorption.

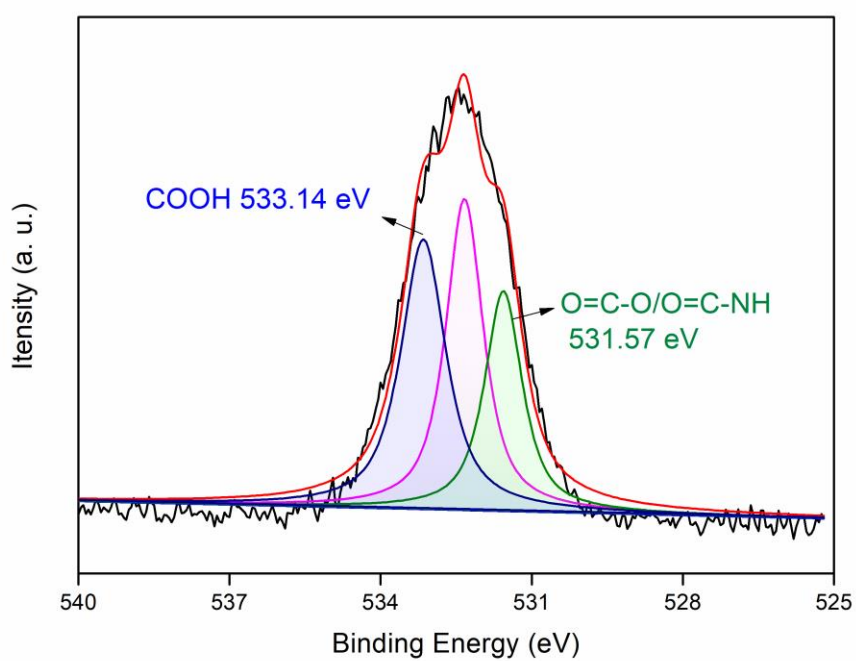


Figure S7 High resolution XPS spectra of O1s of GPAA membrane before uranium adsorption.

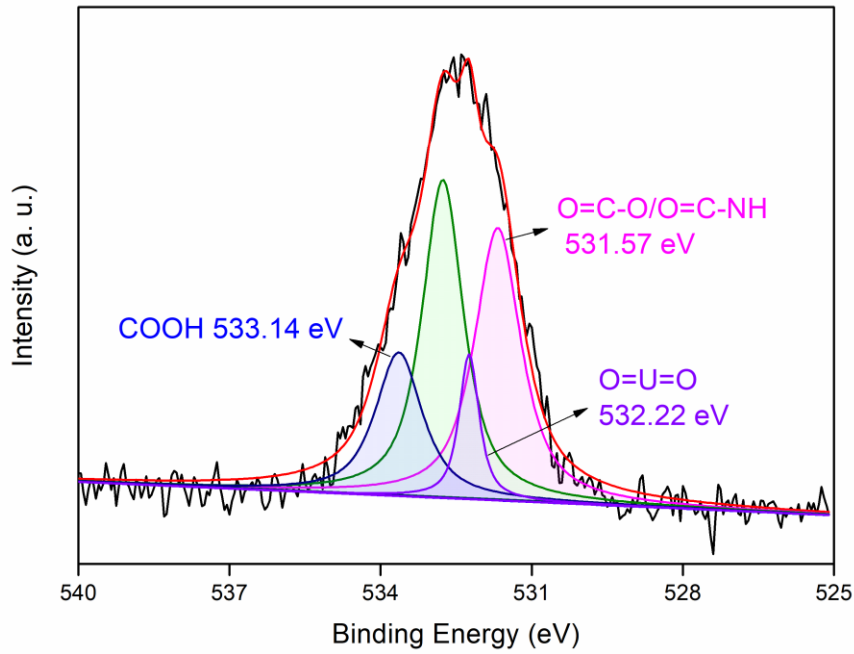


Figure S8 High resolution XPS spectra of O1s of GPAA membrane after uranium adsorption.

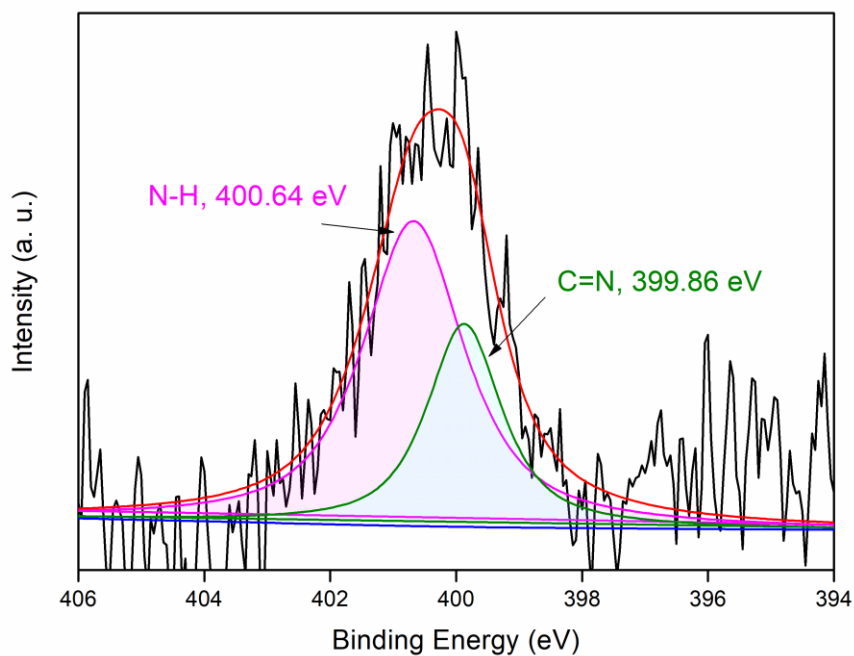


Figure S9 High resolution XPS spectra of N1s of GPAA membrane after uranium adsorption.

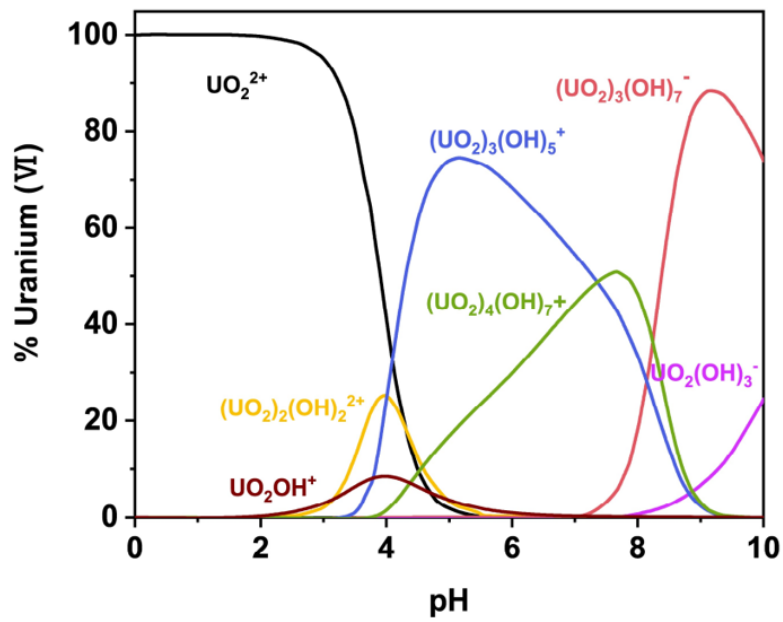


Figure S10 Effect of different pH values on the distribution of uranium species (Nat. Sustain., 2022, 5, 71-80).

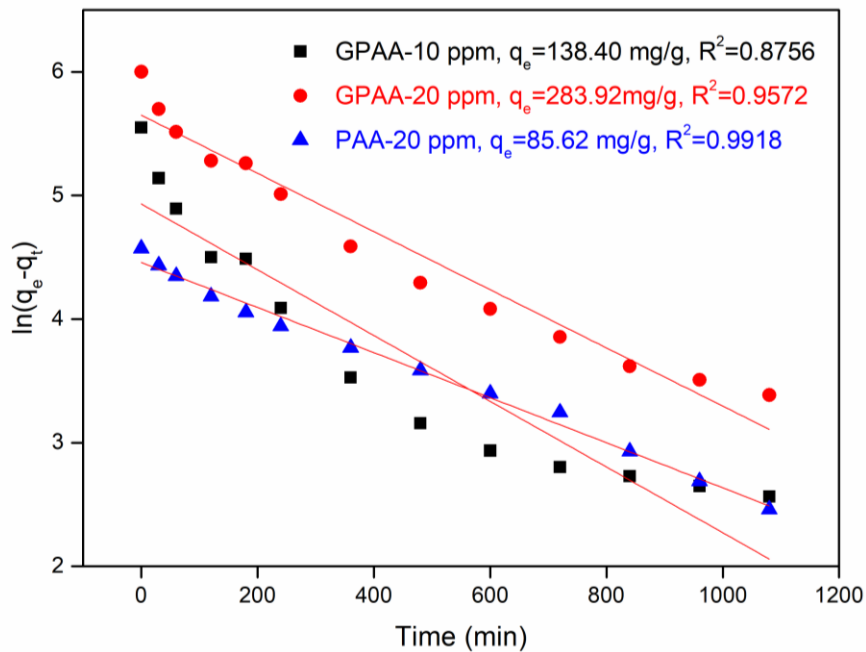


Figure S11 Uranium adsorption properties of GPAA membrane. The pseudo first order linear regression fitting curves of the adsorption kinetics with uranium initial concentrations at 10 ppm, 20 ppm for GPAA membrane, and 20 ppm for PAA membrane, respectively.

4. Tables

Table S1 Distribution coefficient and adsorption kinetic parameters.

Item	C_0 (ppm)	K_d (mL/g)	Pseudo second order			Pseudo first order		
			R^2	q_e (mg/g)	k_2 (g/(mg min))	R^2	q_e (mg/g)	k_1 (/min)
GPAA	10	1.27×10^5	0.9966	257.06	7.00×10^{-5}	0.8756	138.40	2.66×10^{-3}
GPAA	20	7.63×10^4	0.9887	403.22	2.50×10^{-5}	0.9572	238.92	1.35×10^{-3}
PAA	20	5.12×10^3	0.9523	96.72	4.70×10^{-5}	0.9918	85.62	1.82×10^{-3}

Table S2 Langmuir and Freundlich models parameters.

Langmuir model			Freundlich model		
R^2	q_m (mg/g)	k_L (L/mg)	R^2	k_F	n
0.9867	413.2	3.7337	0.8679	263.5163	6.4424