Supporting Information (SI) on

High effective enrichment of U(VI) from aqueous solution on versatile crystalline carbohydrate polymers functionalized graphene oxide

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Synthesis of graphene oxide (GO)

GO was synthesized by modified Hummers method [1]. Briefly, 3.0 g graphite and 1.5 g $NaNO_3$ were added into 300 mL concentrated H_2SO_4 under vigorous stirring and ice-water bath conditions, then 9.0 g of $KMnO_4$ was slowly added into aforementioned suspension for 2 h. The suspensions were reacted at ambient conditions for 5 days under vigorous stirring conditions. Then suspensions were heated to 98 °C and 500 mL 5 wt % H_2SO_4 was added for 2 h under vigorous stirring. After the temperature was cooled to 60 °C, 12 mL H_2O_2 (30 wt %) was added in the suspensions, and the mixtures were re-reacted for 2 h at room temperature. After centrifugation at 6000 rpm for 15 min, the solid phase was washed by 5 wt % H_2SO_4 and then DI water several times until pH 4.0. Then the solid pastes were treated under ultrasonic conditions (power of 140 W) for 30 min. The centrifugation and ultrasonication were recycled for several times, and then GO solution was rinsed with Milli-Q water until the solution was neutral. The desired products were dried in a vacuum tank at room temperature.

Calculation of specific capacitance

The specific capacitance (Cs, F/g) can be calculated from the galvanostatic charging/ discharging curves using following Eqn. (S1):

$$Cs = \frac{2I\Delta t}{m\Delta U} \qquad\qquad (S1)$$

where I and m are the discharge current (A) and the mass (g) of the active material in the one electrodes, respectively. Δ t and ΔU are the discharge time (s) and the potential window (V), respectively.

Pseudo-first-order and pseudo-second-order kinetic models

The equations of pseudo-first-order [2] and pseudo-second-order kinetic models [3]

can be described as Eqns. (S2) and (S3):

$$\ln(q_e - q_t) = \ln q_e - k_1 \times t \qquad (S2)$$

$$t/q_t = 1/(k_2 \times q_e^2) + t/q_e \qquad (S3)$$

where q_e and q_t (mg/g) are the amount of U(VI) adsorbed at equilibrium and at time t,

respectively. k_1 and k_2 are the pseudo-first-order and pseudo-second-order kinetic rate

constants, respectively.

Table S1 Optimized parameters of pseudo-first-order and pseudo-second-order

kinetic models of U(VI) on COF/GO

Pseudo-first-order kinetic model			Pseudo-second-order kinetic model		
ln q_e(mg/g)	k_1(h^{-1})	R^2	q_e(mg/g)	k_2(g/mg/h)	R^2
-0.2938	0.0812	0.3416	20	0.5556	1.0000

Distribution of U(VI) in aqueous solution

The distribution of U(VI) in aqueous solution was fitted by MINTEQ mode 2.6 [4].

The equilibrated constants and distribution of U(VI) species were summarized in

Table S2 and Figure S1A, respectively.

Determination of zeta potentials

The zeta potentials of COF/GO were determined by Nano-ZS Zetasizer (Malvern

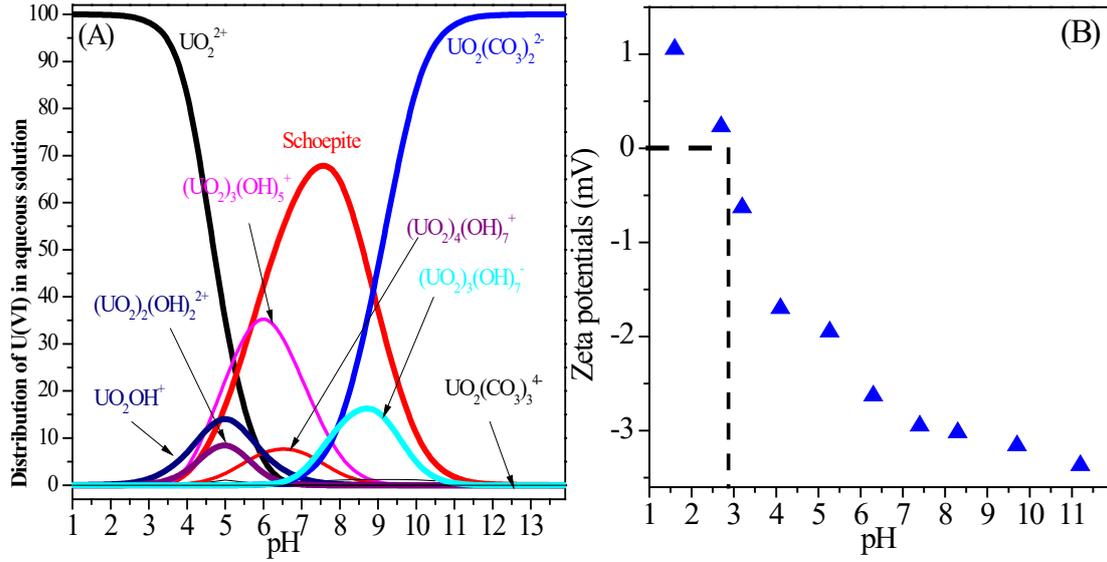Instruments Ltd., Worcestershire, UK).

Figure S1. Distribution of U(VI) species in aqueous solution (A) and zeta potentials

(B) of COF/GO under different pH.

Table S2. Equilibrate constants for U(VI) species in aqueous solutions

Reactions	Log K	Ref.
$UO_2^{2+} + H_2O = UO_2OH^+ + H^+$	-5.20	(S3) [5]
$UO_2^{2+} + 2H_2O = UO_2(OH)_2 + 2H^+$	-12.10	(S3) [5]
$UO_2^{2+} + 3H_2O = UO_2(OH)_3^- + 3H^+$	-19.20	(S3) [5]
$UO_2^{2+} + 4H_2O = UO_2(OH)_4^{2-} + 4H^+$	-33.00	(S3) [5]
$3UO_2^{2+} + 5H_2O = (UO_2)_3(OH)_5^+ + 5H^+$	-15.55	(S3) [5]
$3UO_2^{2+} + 7H_2O = (UO_2)_3(OH)_7^- + 7H^+$	-28.34	(S3) [5]
$UO_2^{2+} + 2CO_3^{2-} = UO_2(CO_3)_2^{2-}$	17.0	(S4) [6]
$UO_2^{2+} + 3CO_3^{2-} = UO_2(CO_3)_3^{4-}$	21.40	(S4) [6]

Langmuir and Freundlich equations

The Langmuir [7] and the Freundlich [8] equation can be expressed by Eqns. (S4) and (S5):

$$\frac{C_e}{Q_e} = \frac{1}{Q_m \times K_L} + \frac{C_e}{Q_m} \qquad (S4)$$

$$Q_e = K_f \times C^n \qquad (S5)$$

where Qe (mg/g) and Ce (mg/L) are the amount of adsorbed ^{238}U(VI)/^{241}Am(III) on AO/CNF and the equilibrium concentration in solution, respectively; Q_m (mg/g) is the maximum adsorption capacity; K_f ($mg^{1-n}g^{-1}L^n$) and n refer to an empirical constants related to adsorption capacity.

Table S3. Parameters of Langmuir and Freundlich models for U(VI) removal on COF/GO

	Langmuir			Freundlich		
	K_a (L/mg)	q_{max} (mg/g)	R^2	$\ln K_F$ (mg/g)/(mg/g)^n	$1/n$	R^2
293 K	0.8763	117.67	0.9993	3.8085	0.4644	0.9210
313 K	0.8182	138.89	0.9995	3.9880	0.4347	0.9117
333 K	0.8919	151.52	0.9994	4.0234	0.4809	0.9435

Calculation of thermodynamic parameters

The thermodynamic parameters (i.e., the standard Gibbs free energy change-ΔG^0, the standard enthalpy change-ΔH^0, the standard entropy change- ΔS^0) were calculated by Eqns. (S6) and (S7):

$$\Delta G^0 = -RT \ln K^0 \tag{S5}$$

$$\ln K^0 = \Delta S^0/R - \Delta H^0/(RT) \tag{S6}$$

where R and T are universal gas constant (8.314 J/(mol·K)) and Kelvin temperature (K), respectively. ln K^0 is adsorption equilibrium constants, which can be calculated by plotting ln K_d vs C_e and extrapolating C_e to zero. The thermodynamic parameters were showed in Table S4.

Table S4. Thermodynamic parameters for U(VI) removal on COF/GO

Temperature	ΔG^0(kJ/mol)	ΔH^0(kJ/mol)	ΔS^0(J/(mol·K))
293 K	28.63		
313 K	30.77	6.26	118.85
333 K	33.41		

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