

Supporting Information

Removal of U(VI) from aqueous solution by amino

functionalized flake graphite prepared by plasma treatment

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Supporting Information

Total number of pages: 7

Total number of Figures: 6

Total number of Tables: 3

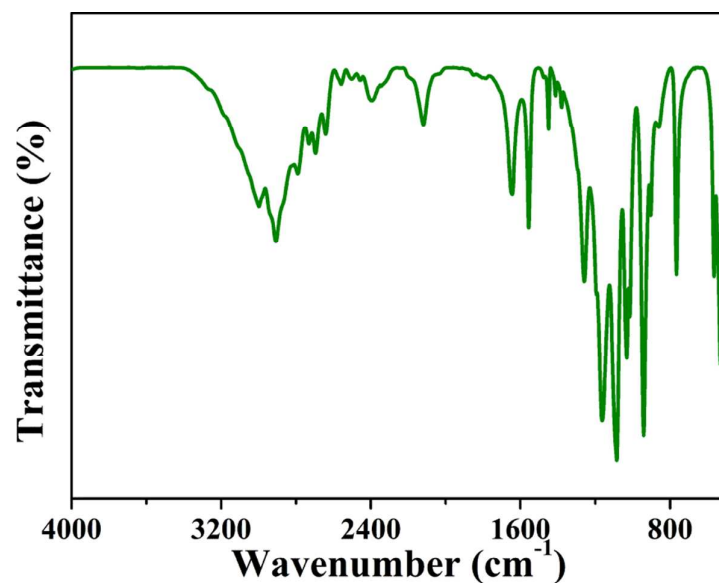


Figure S1 FT-IR spectrum of O-PEA.

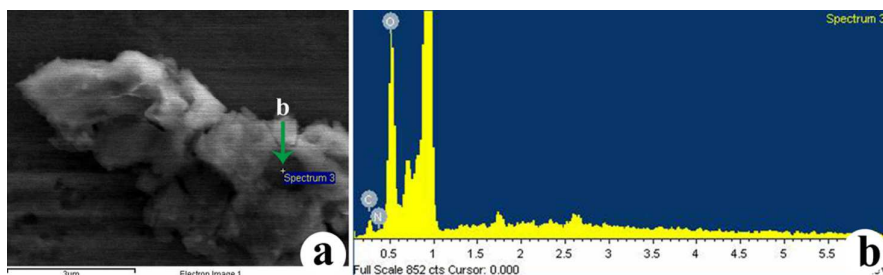


Figure S2 (a) Electron image of PTFG-4, (b) Corresponding EDS examination of PTFG-4.

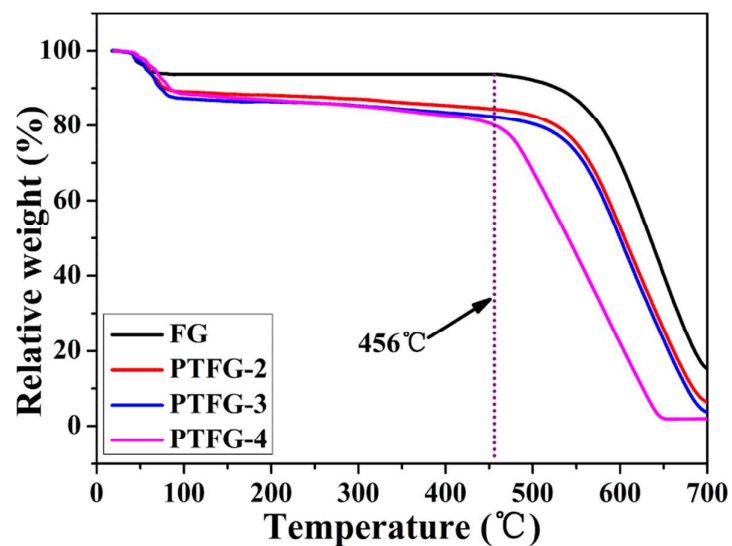


Figure S3 TGA curves of the prepared samples.

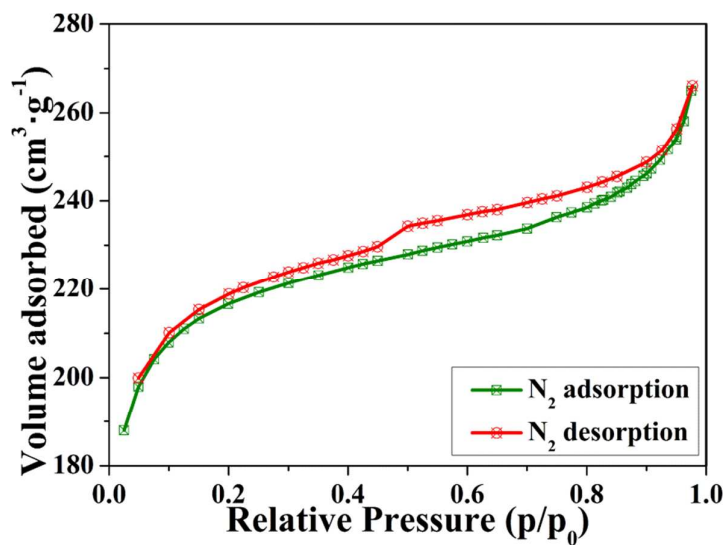


Figure S4 Nitrogen adsorption/desorption isotherms of FG.

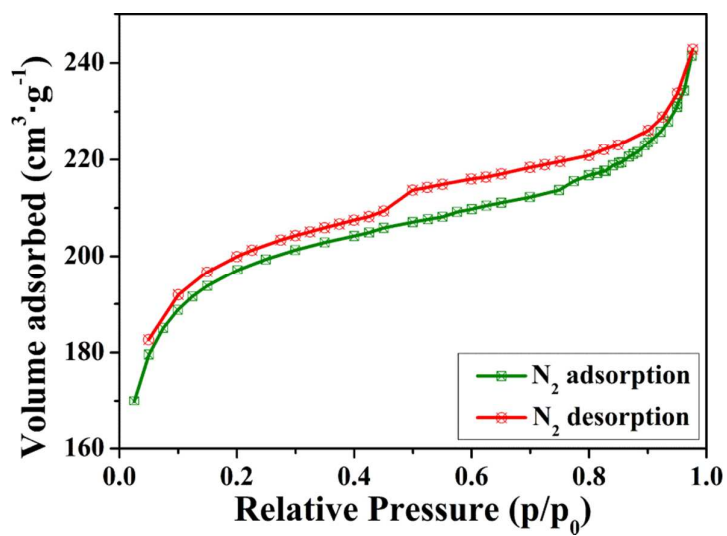


Figure S5 Nitrogen adsorption/desorption isotherms of PTFG-1.

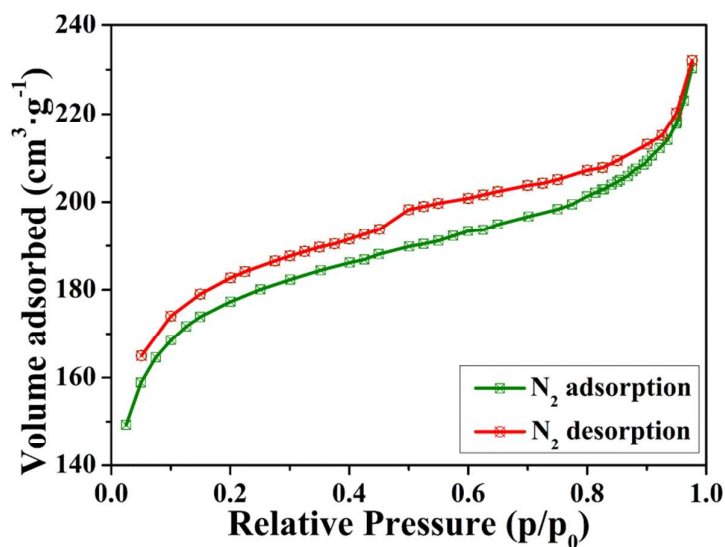


Figure S6 Nitrogen adsorption/desorption isotherms of PTFG-4.

Table S1 Equations and nomenclatures of three kinetic models (q_e and q_t refer to the amounts of U(VI) adsorbed at equilibrium and designed time t , respectively).

| Equations | nomenclatures | | |
|--|--|---|--|
| $q_t = q_e (1 - e^{-k_1 t})$ | k_1 (min^{-1}) | pseudo-first order rate constant | |
| $\frac{t}{q_t} = \frac{1}{2k_2 q_e^2} + \frac{1}{q_e} t$ | k_2 ($\text{g} \cdot \text{mg}^{-1} \cdot \text{min}^{-1}$) | pseudo-second order rate constant | |
| | k_i ($\text{mg} \cdot \text{g}^{-1} \cdot \text{min}^{1/2}$) | intra-particle diffusion constant | |
| $q_t = k_i t^{1/2} + C$ | C | intercept in the intra-particle diffusion model | |

Table S2 Aqueous complexation reactions of U(VI).

| Reactions | $\log K$ ($I = 0.01$ M, $T = 298$ K) |
|---|---------------------------------------|
| $\text{UO}_2^{2+} + \text{H}_2\text{O} = \text{UO}_2(\text{OH})^+ + \text{H}^+$ | -5.25 |
| $\text{UO}_2^{2+} + 2\text{H}_2\text{O} = \text{UO}_2(\text{OH})_2 + 2\text{H}^+$ | -12.15 |
| $\text{UO}_2^{2+} + 3\text{H}_2\text{O} = \text{UO}_2(\text{OH})_3^- + 3\text{H}^+$ | -20.25 |

| | |
|--|--------|
| $\text{UO}_2^{2+} + 4\text{H}_2\text{O} = \text{UO}_2(\text{OH})_4^{2-} + 4\text{H}^+$ | -32.4 |
| $3\text{UO}_2^{2+} + 5\text{H}_2\text{O} = (\text{UO}_2)_3(\text{OH})_5^+ + 5\text{H}^+$ | -15.55 |
| $3\text{UO}_2^{2+} + 7\text{H}_2\text{O} = (\text{UO}_2)_3(\text{OH})_7^- + 7\text{H}^+$ | -32.20 |
| $4\text{UO}_2^{2+} + 7\text{H}_2\text{O} = (\text{UO}_2)_4(\text{OH})_7^+ + 7\text{H}^+$ | -21.90 |

Table S3 Equations and nomenclatures of the four isotherm models (C_e ($\text{mg}\cdot\text{L}^{-1}$) and q_e ($\text{mg}\cdot\text{g}^{-1}$) refer to the U(VI) concentration and the adsorption capacity at equilibrium, respectively).

| Equations | Nomenclatures | |
|--|---|---|
| $q_e = \frac{Q_{\max} b C_e}{1 + b C_e}$ | b ($\text{L}\cdot\text{g}^{-1}$) | Langmuir constant related to the free energy of adsorption |
| | Q_{\max} ($\text{mg}\cdot\text{g}^{-1}$) | Langmuir monolayer adsorption capacity |
| $q_e = K_f C_e^{1/n}$ | n | Freundlich constant indicative of the heterogeneity factor |
| | K_f ($\text{mol}^{1-n}\cdot\text{L}^n\cdot\text{g}^{-1}$) | Freundlich constant indicative of the relative sorption capacity of the EIR |
| $q_e = Q_0 \exp(-\beta \varepsilon^2)$ | Q_0 ($\text{mg}\cdot\text{g}^{-1}$) | Maximum adsorption capacity based on D–R model |
| | β ($\text{mol}^2\cdot\text{kJ}^{-2}$) | D–R constant related to the adsorption energy |
| | ε | Polanyi potential |
| $q_e = B \ln K_t + B \ln C_e$ (8) | K_t ($\text{L}\cdot\text{g}^{-1}$) | Tempkin equilibrium binding constant |
| | $B = RT/b$ | Tempkin constant related to the heat of adsorption |

Description of the four isotherm models

Monolayer adsorption process and no interaction between the adsorbates on adjacent sites are two basic assumptions of Langmuir model. Based on this model, the

adsorption is restricted to monolayer coverage and beyond that adsorption cannot proceed.

$$q_e = \frac{Q_{\max} b C_e}{1 + b C_e} \quad (\text{Langmuir})$$

where C_e ($\text{mg} \cdot \text{L}^{-1}$) and q_e ($\text{mg} \cdot \text{g}^{-1}$) refer to the U(VI) concentration and the adsorption capacity at equilibrium, respectively. b ($\text{L} \cdot \text{g}^{-1}$) and Q_{\max} ($\text{mg} \cdot \text{g}^{-1}$) donate the Langmuir constant concerning the adsorption energy and the Langmuir monolayer adsorption capacity, respectively. While the Freundlich isotherm is an empirical equation and it is applied to adsorption process with non-ideal conditions, including reversible and multilayer adsorption process, under the supposition that surface sites are interdependent and the molecules binding on the surface site will affect the adjacent sites.

$$q_e = K_f C_e^{1/n} \quad (\text{Freundlich})$$

where K_f ($\text{mol}^{1-n} \cdot \text{L}^n \cdot \text{g}^{-1}$) represents the adsorption capacity when metal ion equilibrium concentration equals to 1, and n donate the degree of dependence of sorption at equilibrium concentration.

Furthermore, no limits on surface properties or constant adsorption potential of adsorbent make D-R a much more widely used isotherm model compared to Langmuir isotherm. Meanwhile, the characteristic of adsorption curve is ascribed to the porous structure of the adsorbent in this model. D-R isotherm model is presented as follows:

$$q_e = Q_0 \exp(-\beta \varepsilon^2) \quad (\text{D-R})$$

the linear form of D-R can be expressed as:

$$\ln q_e = \ln Q_0 - \beta \varepsilon^2 \quad (\text{D-R})$$

where β ($\text{mol}^2 \cdot \text{kJ}^{-2}$) represents the activity coefficient with respect to mean adsorption energy, and ε is the Polanyi potential, which is equal to:

$$\varepsilon = RT \ln\left(1 + \frac{1}{C_e}\right)$$

where R is ideal gas constant with the value of $8.314 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$, and T (K) is the

absolute temperature in Kelvin (K). E ($\text{kJ}\cdot\text{mol}^{-1}$) is regarded as mean adsorption energy, required in transmission of 1 mol of ions from solution to solid surfaces. It can be obtained according to the following relation:

$$E = \frac{1}{\sqrt{2\beta}}$$

Whether the adsorption process belongs to physical or chemical can be determined by the magnitude of E . The E value can be used to clarify whether the adsorption process is dominated by chemical ion exchange or physical forces.

Moreover, the Tempkin isotherm was adopted to analyze the equilibrium data as well with another supposition that the decrease of layer adsorption heat of adsorbates is in proportion to surface coverage because of the interactions between adsorbent and adsorbate. Meanwhile, binding energies, up to some maximum ones, with uniform distribution is the conspicuous characteristic of the adsorption process. The linear equation of Tempkin isotherm model is presented as follows:

$$q_e = B \ln K_t + B \ln C_e \quad (\text{Tempkin})$$

where K_t ($\text{L}\cdot\text{g}^{-1}$) is the equilibrium binding constant in accordance with the maximum binding energy and constant B is relevant to the adsorption heat ($B = RT/b$), of which b is the Tempkin constants calculated from a plot of q_e vs $\ln C_e$.