**Supporting Information**

U(VI) adsorption behavior onto polypyrrole coated 3R-MoS2 nanosheets prepared with the molten salt electrolysis method

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**Supporting Information**

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**S1. U(VI) adsorption experiments**

The sorption behavior of 3R-MoS2 and PPy/3R-MoS2 nanosheets for U(VI) was carried out by batch technique. 10.0 mg adsorbent was added into 50 mL solution containing U(VI) ions. The solution pH was adjusted using 0.01 mol·L-1 HNO3 and NaOH. After shaken for a certain time, the solid phase was separated from solution by centrifugation at 10000 rpm for 5 min. The concentration of uranium was analyzed by Arsenazo III method with a visible spectrophotometer at 650 nm. All the batch adsorption experiments were conducted for three times, and the uncertainty of the results is obtained from the mean and standard deviation of the experimental values. The sorption capacity (*q*e, mg·g1) was calculated by Eq. (S1).

|  |  |  |
| --- | --- | --- |
|  |  | (S1) |

where *C*0 and *C*e are the initial and equilibrium concentrations of uranium ion (mg·L1), respectively. *V* is the volume of the solution (L), and *m* is the amount of the sorbent (g).

**S2. Effect of contact time**

The curve form of the pseudo first-order model and the pseudo second-order model can be expressed as Eqs. (S2) and (S3).[1]

|  |  |  |
| --- | --- | --- |
|  | $$q\_{t}=q\_{e}×\left(1e^{k\_{1}t}\right)$$ | (S2) |
|  | $$q\_{t}=\frac{q\_{e}^{2}k\_{2}t}{1+q\_{e}k\_{2}t}$$ | (S3) |

where *k*1 (min-1) and *k*2 (g·mmol-1·min-1) refer to the kinetic constants for the pseudo-first-order model and pseudo-second-order model, respectively, *qt* (mg·g1) is the amount of U(VI) adsorbed at contact time *t*, *q*e (mg·g1) is the capacity at equilibrium.

**S3. Adsorption isotherms**

Langmuir model describes the monolayer adsorption processes, which presents the adsorption at homogeneous sites, expressed as Eq. (S4).[2]

|  |  |  |
| --- | --- | --- |
|  |  | (S4) |

where *q*e is the amount of U(VI) adsorbed at equilibrium (mg∙g1) and *C*e is the equilibrium concentration (mg∙L1), *q*m is the maximum amount or the saturated adsorption amount (mg∙g1), *K*L is an equilibrium constant related to the binding strength (L∙mg1).

The essential characteristics of the Langmuir isotherm can be explained in terms of separation factor *R*L, which is defined by Eq. (S5).[3] The *R*L values under different initial U(VI) concentrations are listed in Table S5.

|  |  |  |
| --- | --- | --- |
|  |  | (S5) |

Freundlich model represents multilayer adsorption, and the adsorption performance occurs on heterogeneous surface at low concentrations. The Freundlich adsorption isotherm equation is expressed as Eq. (S6).[4].

|  |  |  |
| --- | --- | --- |
|  |  | (S6) |

where *K*F and 1/*n* stand for the Freundlich constants.

The D–R isotherm is a semi-empirical equation, which can provide more important information regarding chemical and physical sorption properties.[5] Its linear form is given in Eq. (S7).

|  |  |  |
| --- | --- | --- |
|  |  | (S7) |

where *q*e and *q*DR are the sorption amounts per mass of adsorbent (mol·g1) and the theoretical adsorption capacity (mol·g1), *β is* the constant related to the sorption energy (mol2·kJ2), *ε* is the Polanyi potential which is expressed as *ε* = R*T* ln(1 + 1/*C*e), *R* is the gas constant (kJ·mol1·K1), and *T* is the absolute temperature (K).

The mean free energy of sorption is the free energy change when one mole of ion is transferred to the surface of adsorbents from infinity in the solution, and it is calculated by Eq. (S8).

|  |  |  |
| --- | --- | --- |
|  |  | (S8) |

**S4. Effect of temperature and thermodynamic studies**

The standard free energy (Δ*G*°), standard enthalpy (Δ*H*°) and standard entropy (Δ*S*°) are calculated using Eqs. (S9)(S11).

|  |  |  |
| --- | --- | --- |
|  |  | (S9) |
|  |  | (S10) |
|  |  | (S11) |

where *K*d is the distribution coefficient (mL⋅g1), *T* and *R* are the absolute temperature (K) and the gas constant (8.314 J·mol1·K1), respectively.

**S5. Elution rate**

 $D\%=\frac{C\_{D}×V\_{D}}{q\_{e}}×100\%$ (S12)

Where: *C*D is the concentration of U(VI) after elution (mg∙L1), *V*D is the volume of eluate (L), and *q*e is the adsorption balance of PPy/3R-MoS2 nanosheets.

**UO2OH+**

**UO22+**

**(UO2)3(OH))5+**

**(UO2)2(OH)22+**

**UO2(OH)3**

**(UO2)3(OH)42+**

**(UO2)4(OH)7+**

**Fig. S1. Species distribution of uranyl ions at different pH**

**Table S1.** **Kinetic model constants for the adsorption of U(VI) onto PPy, 3R-MoS2 and PPy/3R-MoS2 nanosheets**

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Adsorbent | *q*e,exp(mg·g-1) | Pseudo-first-order model |  | Pseudo-second-order model |
| *q*1,cal / (mg·g−1) | *k*1 / min−1 | *R*2 |  | *q*2,cal / (mg·g−1) | *k*2 / (g·mg−1·min−1) | *R*2 |
| PPy | 34.12 | 34.45 | 1.571×10−2 | 0.897 |  | 34.12 | 5.819×10−4 | 0.995 |
| 3R-MoS2 | 44.51 | 42.69 | 1.235×10−2 | 0.855 |  | 46.69 | 7.373×10−4 | 0.996 |
| PPy/MoS2 | 110.43 | 100.92 | 1.641×10−2 | 0.975 |  | 115.88 | 4.033×10−4 | 0.999 |

**Table S2.** **Isothermal adsorption fitting parameters for PPy, 3R-MoS2 and PPy/3R-MoS2 nanosheets adsorption U(VI)**

|  |  |  |  |
| --- | --- | --- | --- |
| Adsorbent | Langmuir isotherm |  | Freundlich isotherm |
| *K*L | *q*m  | *R*2 |  | *K*F | *n* | *R*2 |
| PPy | 0.045 | 29.53 | 0.99 |  | 13.85 | 3.76 | 0.97 |
| 3R-MoS2 | 0.059 | 58.65 | 0.99 |  | 13.71 | 3.49 | 0.96 |
| PPy/MoS2 | 0.025 | 200.40 | 0.99 |  | 13.34 | 1.90 | 0.97 |

**Table S3.** **Comparison of adsorption capacities between PPy/3R-MoS2 nanosheets and other MoS2**

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Adsorbents | Reaction time / min | Adsorption capacity / (mg·g) | pH | Reference |
| MoS2-g-PDMA | 60 | U(VI)448.4 | Eu(III)171.2 | U(VI)4.0 | Eu(III)4.0 | [6] |
| MoS2/CeO2 | 40 | Pb(II)333 | — | Pb(II)2.0 | — | [7] |
| SDS-MoS2 | 120 | Cr(VI)63.92 | — | Cr(VI)5.0 | — | [9] |
| 2D-MoS2 | 120 | Hg(II)305 | — | Hg(II)7.0 | — | [10] |
| This workPPy/3R-MoS2  | 120 | U(VI)200.24  | — | U(VI) | — | Present study |

**Table S4.** **Thermodynamic function values of U(VI) adsorption on PPy, 3R-MoS2 and PPy/3R-MoS2 nanosheets**

|  |  |  |  |
| --- | --- | --- | --- |
| Adsorbent | Δ*H*(kJ·mol) | Δ*S*(J·mol·K) | Δ*G* (kJ·mol1) |
| 293.15(K) | 298.15(K) | 303.15(K) | 308.15(K) | 313.15(K) |
| PPy | 13.14 | 108.11 |  |  |  |  |  |
| 3R-MoS2 | 12.94 | 100.71 |  |  |  |  |  |
| PPy/3R-MoS2 | 7.67 | 95.18 |  |  |  |  |  |

**Table S5.** ***R*L with different initial concentrations of U(VI)**

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Initial concentration of U(VI) (mg∙L1) | 10 | 20 | 30 | 40 | 50 | 60 | 70 | 80 | 90 | 100 | 110 |
| PPy | 0.56 | 0.38 | 0.31 | 0.18 | 0.21 | 0.18 | 0.14 | 0.12 | 0.11 | 0.12 | 0.11 |
| 3R-MoS2 | 0.63 | 0.46 | 0.36 | 0.29 | 0.25 | 0.22 | 0.19 | 0.17 | 0.16 | 0.15 | 0.13 |
| PPy/3R-MoS2 | 0.81 | 0.66 | 0.57 | 0.57 | 0.44 | 0.41 | 0.36 | 0.33 | 0.30 | 0.28 | 0.26 |

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