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Supporting Information

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4 Removal of U (VI) from aqueous solution by effective bio-adsorbent

5 from walnut shell and cellulose composites stabilized iron sulfide

6 nanoparticle

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Preparation of biochar derived from walnut shell, FeS nanoparticles and CFeS-WS

The preparation of biochar derived from walnut shell and FeS nanoparticle were synthesized by the modified method (Lyu et al. 2018). In a word, the peanut shell was rinsed with distilled water, and dried for 24 h at 110 ℃. After cooling down, the dried walnut shell was ground into 1-2 cm pieces and carbonized for 2 h at 250 ℃ in a muffle furnace under oxygen-limited conditions. Then biochar was pulverized and sieved through a 60 meshes sieve. The biochar derived from walnut shell was gained. The iron sulfide nanoparticles (FeS) were prepared according to the reaction of $FeSO_4$ with Na_2S. That is, 50 mL $FeSO_4$ solution (0.05 mol/L) was added into 250 mL Erlenmeyer flask. Then, 50 mL Na_2S solution (0.05 mol/L) was also added and mixed for 30 min under continuous aerating N_2 and magnetic stirring condition. Then suspensions were sealed with rubber stopper and aged for 48 h. It could ensure the full growth of iron sulfide nanoparticles. Finally, the iron sulfide nanoparticles were obtained for experiments.

CFeS-WS was prepared with the composites of biochar, cellulose and FeS nanoparticles. According to the previous experimental results, it can be found that the mass ratio of ferrous sulfate, Na_2S, cellulose and biochar is 2:2:1:4, which is optimum value. That is, under the magnetic stirring condition and continuous aerating nitrogen, the 5 g $FeSO_4$ and 100 mL of anaerobic deionized water were mixed into the 250 mL Erlenmeyer flask. Then, added 10 g biochar from walnut shell to the Erlenmeyer flask and stirred them for 30 min under magnetic stirring condition. Next, added 2.5 g cellulose into the Erlenmeyer flask again, and stirred them for 30 min under magnetic stirring condition. Then, 5 g Na_2S was added into the mixture solution, which was stirred them for 30 min under magnetic stirring condition. In the process of preparation, they were conducted under magnetic stirring condition and continuous aerating nitrogen in order to avoid oxidation of Fe^{2+} ions in solution. Then, the mixture solution was sealed with rubber stopper, which was placed in a constant

62 temperature of 25℃ for 48 h. Then, they were freeze-dried for 12 h, which were

63 washed with anaerobic deionized water for three times. Subsequently, they were

64 freeze-dried for 12 h again. The efficient bio-adsorbent of CFeS-WS was obtained.

65 The obtained materials (biochar, FeS nanoparticle, FeS-WS and CFeS-WS were

66 prepared for tests.

67

68 **Calculation of the removal rate and uptake capacity**

69 The Eq.1-2 was used for the calculation of the removal rate (R (%)) and uptake

70 capacity (q (mg/g)).

71 $$R = \frac{C_0 - C_e}{C_0} \times 100\% \tag{1}$$

72 $$q = \frac{(C_0 - C_e) \times V}{m} \tag{2}$$

73 Where, V (L) and m (g) were the solution volume and the weight of adsorbent,

74 respectively. C_e (mg/L) and C_0 (mg/L) were equilibrium concentration and initial

75 concentration, respectively.

76

77 **Pseudo first-order kinetic model and Pseudo second-order kinetic model**

78 Their mathematical expressions of pseudo first-order kinetic model and pseudo

79 second-order kinetic model were shown as Eq.3-4 (Hummers & Offeman 1958,

80 Lagergren 1898).

81 $$q_t = q_e \left(1 - e^{-K_1 t}\right) \tag{3}$$

82 $$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \tag{4}$$

83 Where q_e (mg/g) and q_t (mg/g) are adsorption capacity of U(VI) ions solution by

84 CFeS-WS at adsorption time t and adsorption equilibrium respectively. K_1 (min^{-1})

85 and K_2 (min^{-1}) are the adsorption rate constant.

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Langmuir isotherm model and Freundlich isotherm model

Their mathematical expressions of Langmuir isotherm model and Freundlich isotherm model were displayed as Eq.5-6 (Langmuir 1918, Freundlich 1906).

$$q_e = \frac{q_m C_e K_L}{1 + C_e K_L} \qquad (5)$$

Where q_e (mg/g) was concentration of adsorbed U(VI) ions at equilibrium. q_m (mg/g) was the maximum uptake capacity of U(VI) ions. C_e (mg/L) was concentration of U(VI) ion at equilibrium and K_L (L/mg) was constant.

$$q_e = K_f C_e^{1/n} \qquad (6)$$

Where q_e (mg/g) was the amount of adsorbed U(VI) ions per unit mass of the adsorbent at equilibrium. C_e (mg/L) was concentration of U(VI) ion at equilibrium. K_f and $1/n$ were constant.

Calculation of thermodynamic parameters

They are Gibbs free energy (ΔG^0 (kJ/mol)), enthalpy (ΔH^0 (kJ/mol)) and entropy (ΔS^0 (J/mol/K)) respectively. They can be calculated using the following Eq.7-9:

$$\Delta G^0 = -RT \ln K_a \qquad (7)$$

$$\ln K_a = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT} \qquad (8)$$

$$K_a = \frac{q_e}{C_e} \qquad (9)$$

Where, T was the solution temperature (K), and K_a was the adsorption equilibrium constant. The value of R was the gas constant (8.314 J·mol⁻¹·K⁻¹), and q_e was the amount of adsorbate adsorbed per unit mass of adsorbate at equilibrium (mg/g). The

108 value of C_e is the equilibrium concentration of the adsorbate (mg/L).

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110 **The species distribution of U(VI) ions at different pH**

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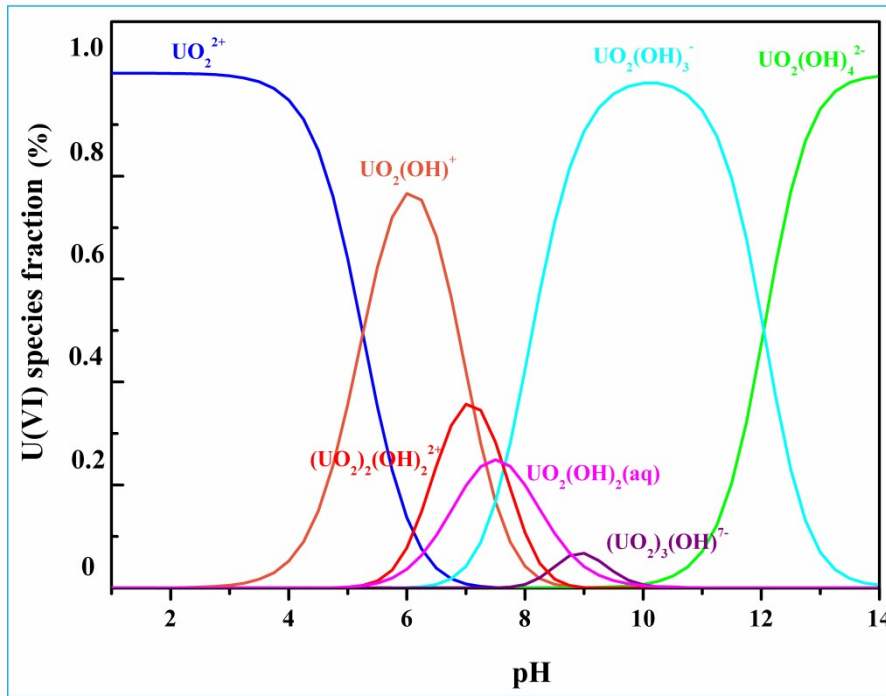
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113 Fig.9 The species distribution of U(VI) ions using Visual MINTEQ (Version 3.0).

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117 Table 1 Comparison of CFeS-WS with other adsorbents for U(VI) removal

Adsorbents	Experimental conditions	q_e (mg/g)	Refs
active carbon	pH=3.0, T=293K	28.3	(Pang et al. 2018)
magnetic biochar	pH=3.0, T=293K	54.4	(Hu et al. 2018)
nZVI/C composite	pH=3.0, T=298K	103.1	(Liu et al. 2017)
MnO$_2$@PPy	pH=5.0, T=298K	63.0	(Mellah et al. 2006)
FA@PEI	pH=5.0, T=298K	70.32	(Yao et al. 2018)
CFeS-WS	pH=6.0, T=318 K	136.24	**This work**

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