

## Supporting information

# Two-dimensional Lamellar Magnesium Silicate with large spacing as Excellent Adsorbent for Uranium Immobilization

Weixue Wang, Zhe Chen,\* Haijiang Zhou, Yifei Zhang and Xiangke Wang\*

College of Environmental Science and Engineering, North China Electric Power University, Beijing, 102206, PR China

Email: chenz@ncepu.edu.cn (Z. Chen); xkwang@ncepu.edu.cn (X.K. Wang)

### Calculation method

The adsorption percentage was described as (equation 1):

$$R = \frac{(C_0 - C_e)}{C_0} 100\% \quad (1)$$

Where  $C_0$  was the initial U(VI) concentration (mg/L) and  $C_e$  was the equilibrium U(VI) concentration (mg/L).

The distribution coefficient ( $K_d$ ), was defined as (equation 2):

$$K_d = \frac{V(C_0 - C_e)}{m C_e} \quad (2)$$

Where  $m$  (g) was the mass of adsorbent added to the glass vials,  $V$  (mL) was the volume of the suspension,  $C_0$  and  $C_e$  represent the initial and equilibrium concentrations.

The sorption capacity  $Q_e$  (mg/g) was calculated from the following equations:

$$Q_e = \frac{(C_0 - C_e)V}{m} \quad (3)$$

Where  $V$  (mL) was the total volume of the suspension in the test tubes,  $m$  (g) was the mass of the adsorbent in the test tube, and  $C_0$  (mg/L) and  $C_e$  (mg/L) were the initial and equilibrium U(VI) concentrations after reaction, respectively.

### **The pseudo-first-order model and pseudo-second-order model**

The pseudo-first-order kinetic model is defined as (equation 4):

$$\ln (Q_e - Q_t) = \ln Q_e - k_1 t \quad (4)$$

The pseudo-second-order kinetic model is defined as (equation 5):

$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{t}{Q_e} \quad (5)$$

Where  $k_1$  ( $\text{min}^{-1}$ ) and  $k_2$  ( $\text{g/mg}\cdot\text{min}$ ) were the pseudo-first-order and pseudo-second-order rate constant of U(VI) sorption, and  $t$  was adsorption time (min). The  $Q_e$  and  $Q_t$  were the amounts of U(VI) adsorbed (mg/g) at equilibrium and at contact time  $t$ , respectively.

### **The Langmuir and Freundlich models**

Langmuir model:

$$Q_e = Q_m \frac{bC_e}{1 + bC_e} \quad (4)$$

Where  $C_e$  denotes the equilibrium concentration of U(VI) (mg/L),  $b$  (L/mg) is the Langmuir constant related to the enthalpy of sorption and affinity of adsorbent,  $Q_e$  refers to the amount of U(VI) adsorbed per unit weight of the adsorbent at equilibrium (mg/g),  $Q_m$  (mg/g) is the maximum adsorption capacity.

Freundlich model:

$$Q_e = k_F C_e^n \quad (5)$$

Where  $n$  and  $k_F$  ( $\text{mg}^{1-n} \text{L}^n \text{g}^{-1}$ ) are Freundlich parameters,  $n$  is the level depending on removal equilibrium concentration;  $k_F$  is generally related to the adsorption ability of the adsorbent when the value of the adsorbate concentration is equal to 1.

## The thermodynamic studies calculation method

The Gibbs free energy change ( $\Delta G^0$ ) can be expressed by the following equation:

$$\Delta G^0 = -RT \ln k^0 \quad (6)$$

Where  $R$  is the ideal gas constant ( $8.3145 \text{ J/mol}\cdot\text{K}$ ),  $k^0$  was the sorption equilibrium constant at different temperature.

The average standard enthalpy change ( $\Delta H^0$ ) could be calculated by van't Hoff equation:

$$\ln k^0(T_2) - \ln k^0(T_1) = \frac{-\Delta H^0}{R} \left( \frac{1}{T_2} - \frac{1}{T_1} \right) \quad (7)$$

The standard entropy change ( $\Delta S^0$ ) was obtained by Eq 8:

$$\Delta S^0 = -\frac{\Delta G^0 - \Delta H^0}{T} \quad (8)$$

## Table S1

Kinetic parameters of U(VI) sorption on magnesium silicate.

| Species | Pseudo-first-order          |                                      |       | Pseudo-second-order                    |                                      |       |
|---------|-----------------------------|--------------------------------------|-------|--|--------------------------------------|-------|
|         | $k_1$ ( $\text{min}^{-1}$ ) | $Q_{e,\text{cal}}$ ( $\text{mg/g}$ ) | $R^2$ | $k_2$ ( $\text{g/mg}\cdot\text{min}$ ) | $Q_{e,\text{cal}}$ ( $\text{mg/g}$ ) | $R^2$ |
| U(VI)   | 0.0599                      | 75.02                                | 0.859 | 0.0004945                              | 168.35                               | 0.995 |

## Table S2

Comparison of U(VI) sorption capacity on magnesium silicate with other materials.

| Adsorbents                            | $T$ (K) | $Q_{\text{max}}$ ( $\text{mg/g}$ ) | $\Delta G^0$ ( $\text{kJ/mol}$ ) | $\Delta H^0$ ( $\text{kJ/mol}$ ) | $\Delta S^0$ ( $\text{J/mol}\cdot\text{K}$ ) | Reference |
|---------------------------------------|---------|------------------------------------|----------------------------------|----------------------------------|--|-----------|
| CaTiO <sub>x</sub>                    | 313.15  | 294.30                             | -3.69                            | 0.037                            | 7.85   | 1         |
| TiO <sub>2-x</sub>                    | 313     | 45.84                              | -1.51                            | 7.793                            | 0.03   | 2         |
| MgFeAl LDH                            | 298     | 167.61                             | -13.57                           | 10.23                            | 79.87  | 3         |
| SiO <sub>2</sub> @ LDH nanocomposites | 298.15  | 303.1                              | -11.56                           | 7.16                             | 62.88  | 4         |

|   |        |        |        |       |       |           |
|---|--------|--------|--------|-------|-------|-----------|
| Layered titanate nanowires                                    | 313    | 384    | -1.74  | 0.408 | 6.86  | 5         |
| Sepiolite   | 313    | 15     | -9.06  | 2.23  | 37.96 | 6         |
| <i>l</i> -C <sub>3</sub> N <sub>4</sub> /PDA/PEI <sub>3</sub> | 298    | 60.51  | -3.36  | 23.31 | 89.49 | 7         |
| nature talc   | 298    | 41.6   |        |       |       | 8         |
| magnesium silicate  | 313.15 | 626.92 | -12.76 | 11.43 | 77.25 | This work |

(LDH refers to layered double hydroxides)

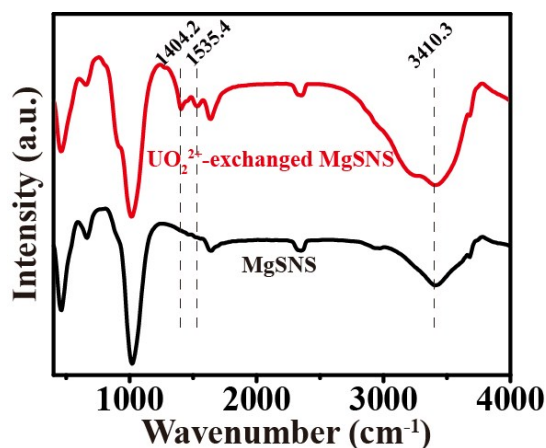


Fig. S1 FTIR spectra for magnesium silicate and U(VI)-reacted.

## Reference

1. Y. Hu, X. Wang, Y. Zou, T. Wen, X. Wang, A. Alsaedi, T. Hayat and X. Wang, *Chem. Eng. J.*, 2017, 316, 419-428.
2. S. Song, S. Huang, R. Zhang, Z. Chen, T. Wen, S. Wang, T. Hayat, A. Alsaedi and X. Wang, *Chem. Eng. J.*, 2017, 325, 576-587.
3. S. Song, L. Yin, X. Wang, L. Liu, S. Huang, R. Zhang, T. Wen, S. Yu, D. Fu, T. Hayat and X. Wang, *Chem. Eng. J.*, 2018, 338, 579-590.
4. D. Yang, S. Song, Y. Zou, X. Wang, S. Yu, T. Wen, H. Wang, T. Hayat, A. Alsaedi and X. Wang, *Chem. Eng. J.*, 2017, 323, 143-152.
5. L. Yin, P. Wang, T. Wen, S. Yu, X. Wang, T. Hayat, A. Alsaedi and X. Wang, *Environ. Pollut.*, 2017, 226, 125-134.
6. Y. Sun, J. Li and X. Wang, *Geochim. Cosmochim. Ac.*, 2014, 140, 621-643.
7. P. Wang, L. Yin, J. Wang, C. Xu, Y. Liang, W. Yao, X. Wang, S. Yu, J. Chen, Y. Sun and X. Wang, *Chem. Eng. J.*, 2017, 326, 863-874.
8. M. Sprynskyy, T. Kowalkowski, H. Tutu, E. M. Cukrowska and B. Buszewski, *Chem. Eng. J.*, 2011, 171, 1185-1193.