Electronic Supplementary Material

Magnetic KIT-6 nano-composite and its amino derivatives as convenient adsorbent for U(VI) sequestration

Jiafeng Ouyang[1,2], Wenlu Guo[2,3], Lin Wang[2], Changming Nie[3], Dadong Shao **(✉)**[1], Weiqun Shi[2], Liyong Yuan **(✉)**[2]

1 School of Environmental and Biological Engineering, Nanjing University of Science and Technology, Nanjing 210094, China

2 Institute of High Energy Physics, Chinese Academy of Sciences, Beijing 100049, China

3 School of Chemistry and Chemical Engineering, University of South China, Hengyang, 421001, China

E-mails: shaodadong@126.com (Shao D); yuanly@ihep.ac.cn (Yuan L)

Table S1 Compositions of the coexistent ions solution

Coexistent ion	Reagent	Reagent purity
U	$UO_2(NO_3)_2·6H_2O$	Standard reagent
Co	$Co(NO_3)_2•6H_2O$	AR
Ni	$Ni(NO_3)_2•6H_2O$	AR
Zn	$Zn(NO_3)_2•6H_2O$	AR
La	$La(NO_3)_3•6H_2O$	AR
Sm	$Sm(NO_3)_3•6H_2O$	99.9% metal basis
Sr	$Sr(NO_3)_2$	AR
Yb	$Yb(NO_3)_3•5H_2O$	99.9% metal basis
Nd	$Nd(NO_3)_3•6H_2O$	AR
Gd	$Gd(NO_3)_3•6H_2O$	AR

SI-1 Analytical techniques

The morphologies and sizes were examined with a field emission scanning electron microscopy (SEM, Hitachi S-4800). High resolution transmission electron microscopy (HRTEM) was performed with a Philips Tecnai F20 microscope operating at 200 kV (Cs = 1.2 mm, Point resolution 0.24 nm). Image was recorded using a CCD camera (GATAN 894, 2048×2048 pixels, pixel size 14×14 μm). Power X-ray diffraction (PXRD) patterns were obtained by a Bruker D8-Advance X-ray Diffractometer with a Cu Kα radiation (λ=1.5406 Å). Thermogravimetric data were recorded by a thermal gravimetric analyzer (TGA, TA Instruments, Q500) from 323-1073K by a heating rate of 5 K min^{-1} under air flow. Data of Fourier transform infrared (FT-IR) spectra of samples were record on a Bruker Tensor 27 spectrometer with a potassium bromide pellet method. The N_2 adsorption/desorption experiments were measured at a liquid nitrogen temperature (77K) using a micromeritics ASAP 2020 HD88 instrument. The specific surface areas were calculated by the Brunauer–Emmett–Teller (BET) method with prior degassing under vacuum at 423K. Zeta Potentials were obtained by Malvern

Zetasizer Naw ZS90, with the concentration of 50 mg L^{-1}. The UV–Visible spectrometry with arsenazo-III as the chromogenic agent was used at an absorption wavelength at 656 nm. Inductively coupled plasma optical emission spectrometer (ICP-OES, HORIBA, JY 2000-2) was used to analyze the initial and equilibrium concentration of the multi-ion solution.

SI-2 Sorption experiments

The sorption experiments were carried out using the batch method. The concentrations of U(VI) varied from 5–200 mg L^{-1}. The pH of solution was adjusted by adding negligible volumes of 0.1M/1M NaOH solution or 0.1M/1M HNO_3 solution. In a typical process, 4 mg adsorbent was added into 10 ml test solution in polytetrafluoroethylene-lined screw cap glass tubes (20 ml). The glass tubes were shock in Shaking Water Bath for specified time (t, min) at 288K, and then the solid phase was centrifugal separation or magnetically separated from the solution after 360 min. The control experiment was performed at the same time using the identical U(VI) solution without any adsorbent. Before the determination, test solutions filtered by needle type filter, and diluted 10-50 times for the concentration analysis. The concentrations of U(VI) in the solution were determined by UV–Visible spectrometry and the multi-ion were determined by inductively coupled plasma optical emission spectrometer (ICP-OES). All values were measured in duplicate with the uncertainty within 5%.

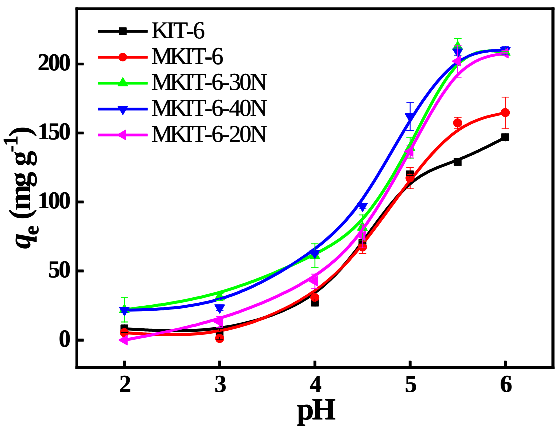

Fig. S1. Effect of pH, $m_{sorbent}/V_{solution}$=0.4 mg mL^{-1}, $[U]_{initial}$ =100 mg L^{-1}, T=288K.

SI-3 Sorption kinetic study

In order to study the sorption kinetic, the pseudo-first-order model and pseudo-second-order model were used to fit U(VI) sorption kinetics:

The pseudo-first-order equation:

$$ln(q_e - q_t) = ln\, q_e - k_1 t \tag{S1}$$

The pseudo-second-order equation:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{S2}$$

where k_1 and k_2 are the sorption rate constant (min^{-1} for first-order sorption and g mg^{-1} min^{-1} for second-order sorption); t is the contacting time (min); q_e is the sorption amount at equilibrium time; q_t is the sorption capacity at time t. Constants and correlation coefficients were shown in Table 2.

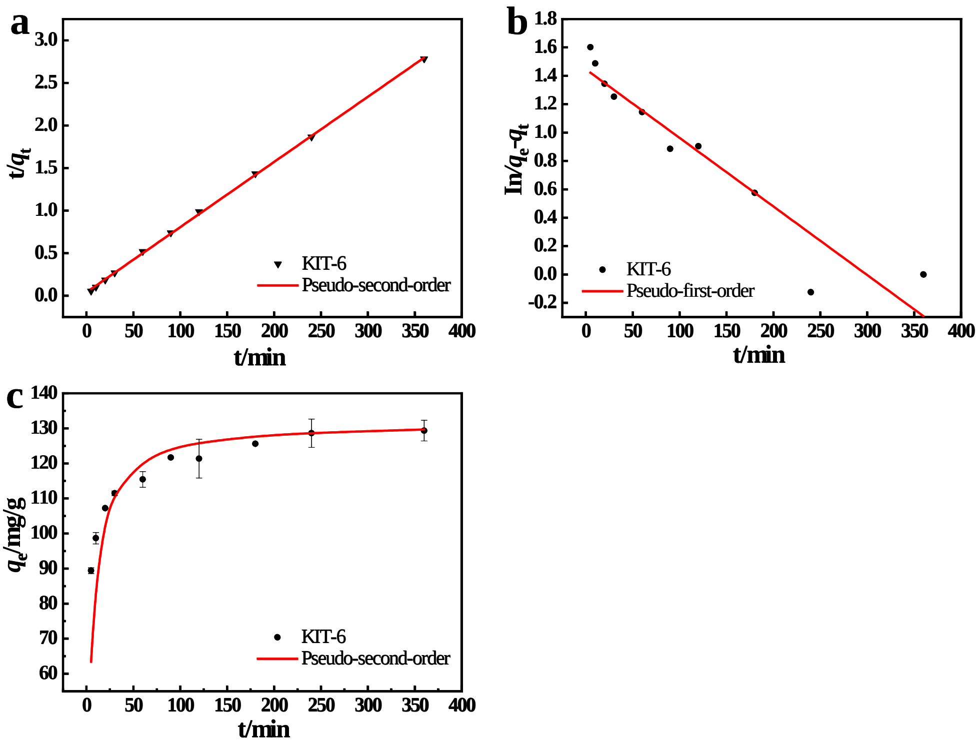

Fig. S2. (a) Shows linearity between t/q_t versus t, an indication of good matching of the experimental kinetics data with pseudo-second-order model; (b) shows linearity between ln(q_e-q_t) versus t, an indication of not good enough matching of the experimental kinetics data with pseudo-first-order model; (c) shows fitting result for KIT-6.

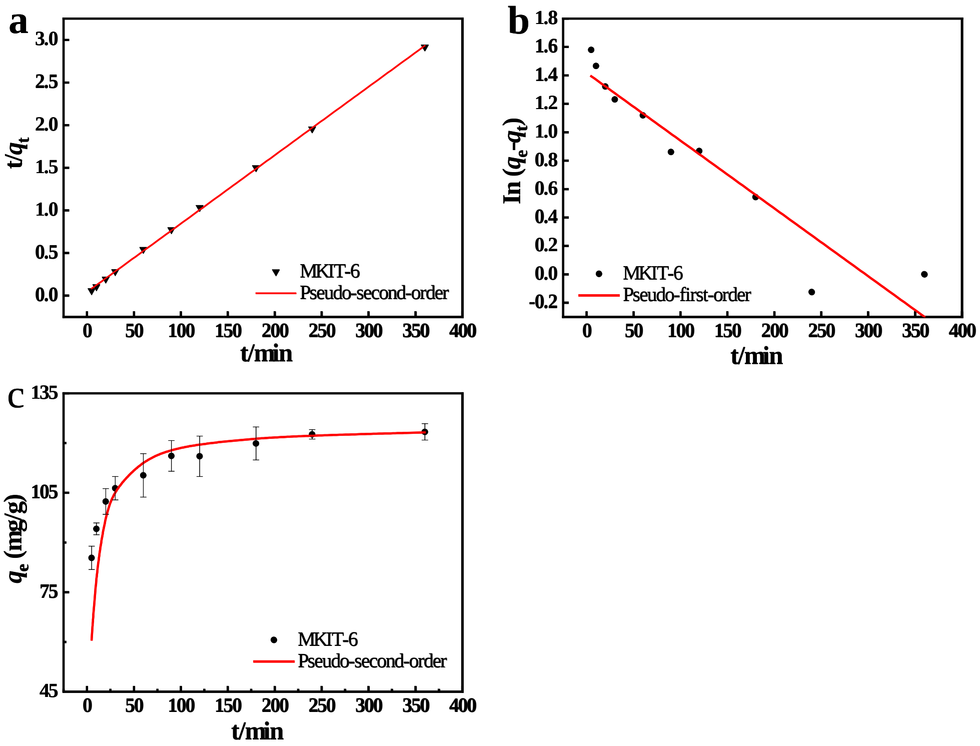

Fig. S3. (a) Shows linearity between t/q_t versus t, an indication of good matching of the experimental kinetics data with pseudo-second-order model; (b) shows linearity between ln(q_e-q_t) versus t, an indication of not good enough matching of the experimental kinetics data with pseudo-first-order model; (c) shows fitting result for MKIT-6.

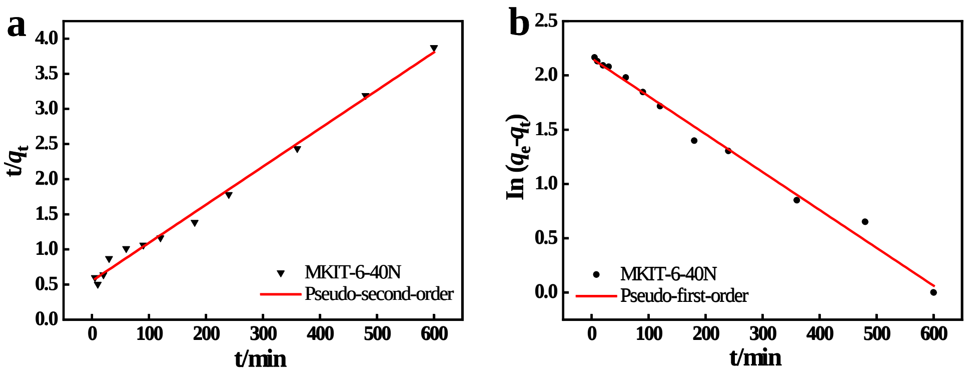

Fig. S4. (a) Shows linearity between t/q_t versus t, an indication of good matching of the experimental kinetics data with pseudo-second-order model; (b) shows linearity between ln(q_e-q_t) versus t, an indication of not good enough matching of the experimental kinetics data with pseudo-first-order model.

SI-4 Sorption isotherms study

To verify the sorption type, the batch sorption experiment data were fitted by Langmuir and Freundlich[6] models respectively. From the linear form of these isotherms model, equations can be written as follows:

$$\frac{C_e}{q_e} = \frac{1}{bq_0} + \frac{C_e}{q_0} \tag{S4}$$

$$ln\, q_e = ln\, k_F + \frac{ln\, C_e}{n} \tag{S5}$$

where q_e is the sorption capacity (mg g^{-1}) at equilibrium time, C_e is the equilibrium concentration of U(VI) ions in solution (mg L^{-1}), q_0 is the saturated sorption capacity (mg g^{-1}), b is an empirical parameter, k_F and n are the Freundlich constants related to the sorption capacity and the sorption intensity, respectively. Correlation coefficients for the sorption are listed in Table 3.

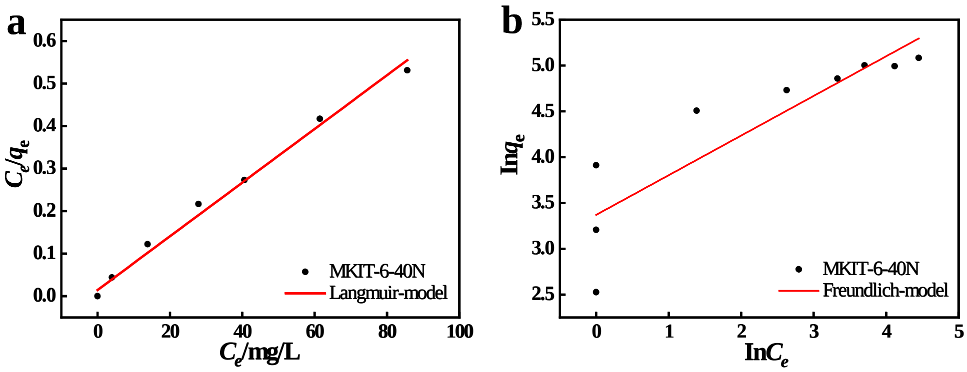

Fig. S5. (a) Shows linearity between C_e/q_e versus C_e, an indication of good matching of the experimental kinetics data with Langmuir model; (b) shows linearity between lnq_e versus lnC_e, an indication of not good enough matching of the experimental kinetics data with Freundlich model.