Supplementary Information

Layered double hydroxide intercalated with dimethylglyoxime for highly selective and ultrafast uptake of uranium from seawater

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Supplementary Note 1. Chemicals and materials

Magnesium nitrate ($Mg(NO_3)_2 \cdot 6H_2O$, Tianjin Damao Chemical Reagent Co., Ltd, 99.0%), aluminium nitrate ($Al(NO_3)_3 \cdot 9H_2O$, Sinopharm Chemical Reagent Co., Ltd, 99.0%), dimethylglyoxime (*abbr.* DMG, with a chemical formula of $C_4H_8N_2O_2$, innochem Co., Ltd, 99.0%), uranyl nitrate ($UO_2(NO_3)_2 \cdot 6H_2O$, Shanghai Acmec Biochemical Co., Ltd, 99%), formamide (abbr. FM, with a chemical formula of CH_3NO, Xilong Scientific Reagent Co., Ltd, 99.0%). All solvents and chemical regents in the present work were of analytical reagents and were used without further purification. And deionized water was used in all experiments.

Supplementary Note 2. Characterization Techniques

Fourier-transform infrared (FT-IR) spectra were recorded with a Nicolet-380 FT-IR spectrometer using the KBr pellet method. X-ray diffraction (XRD) was performed on a PANalytical X'pert Pro MPD diffractometer with Cu-Kα radiation at room temperature, with step size of 0.0334°, scan time 20 s per step, and 2θ ranging from 5 to 80°. The morphology of the samples were characterized with scanning electron microscope (SEM) on a Hitachi S-4800 microscope. The solid samples after the adsorption experiments were performed by X-ray photoelectron spectroscopy (XPS) using an ESCALAB 250Xi spectrometer (Thermofisher). The peaks were fitted using the software Avantage.

The metal ion concentrations in supernatant solutions before and after adsorptions were analyzed using the inductively coupled plasma atomic emission spectrometer (ICP-AES, Jarrel-ASH, ICAP-9000) and inductively coupled plasma mass spectrometry (ICP-MS, PE, NEXION300X), respectively. The metal ion contents in solid samples were determined by ICP-AES, and a 0.1 M HNO_3 solution was used to dissolve them. The contents of C, H and N in solid samples were determined by

Elementar Vario EL elemental analyzer. The chemical formulas of the samples were determined from the results of ICP and CHN elemental analyses. A Sartorius universal type pH meter (PB-10) was used to monitor the pH value of the solutions before and after adsorption.

Supplementary Note 3. Chemical stability of DMG-LDH at the solution of pH=5.

We dissolved 0.02 g of DMG-LDH, containing 3.4 mg of Mg and 2.1 mg of Al based on the composition of $Mg_{0.65}Al_{0.35}(OH)_2(C_4H_6N_2O_2)_{0.075}(CO_3)_{0.1}\cdot H_2O$, in an aqueous solution at pH of 5, and kept standing for 24 h at room temperature. We found that the average concentration of Mg was 10 $mg\cdot L^{-1}$ and that of Al was 0.03 $mg\cdot L^{-1}$ in 20 mL solution, corresponding to 0.20 mg Mg and 0.0006 mg Al. Therefore, the dissolved mass fraction of Mg was 5.88% and that of Al was 0.03%. We see surely the adsorbent slightly dissolved in the weak acid environment.

Supplementary Note 4. Langmuir model.

The two equations of Langmuir model are as follows:

$$q_e = q_m \frac{bC_e}{1 + bC_e} \quad (1)$$

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{q_m K_L} \quad (2)$$

Where q_e ($mg\cdot g^{-1}$), q_m ($mg\cdot g^{-1}$), C_e ($mg\cdot L^{-1}$) and K_L ($L\cdot mg^{-1}$) are the equilibrium adsorption capacity, theoretical maximum adsorption capacity, equilibrium concentration and Langmuir constant, respectively.

Supplementary Note 5. Sorption kinetics models.

The pseudo-first-order (Eq. 3) and pseudo-second-order (Eq. 4) models, are normally used to fit the dynamic data to understand the adsorption rate and mechanism:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (3)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (4)$$

Where q_e (mg·g^{-1}): equilibrium adsorption capacity of unit mass, q_t (mg·g^{-1}): adsorption capacity at time of t, k_1 (min^{-1}) and k_2 (g·mg^{-1}·min^{-1}): equilibrium rate constants, and k_1 and k_2 can be got by $ln(q_e\text{-}q_t)$ and t/q_t, respectively.

Table S1. Composition of the DMG-LDH composite.

Chemical formula	wt%, found (calcd)				
	Mg	Al	C	H	N
$Mg_{0.65}Al_{0.35}(OH)_2(C_4H_6N_2O_2)_{0.075}(CO_3)_{0.1}\cdot H_2O$	18.20 (17.21)	10.70 (10.30)	5.31 (5.23)	4.26 (4.85)	2.00 (2.29)

The *found* data (experimental data) were obtained by CHN and ICP, and the *calcd* data (theoretical data) were determined based on the chemical formula.

Table S2. Parameters for pseudo-second-order dynamic model of U(VI) sorption on DMG-LDH.

$q_{e,\,exp}$ (mg·g^{-1})	k_2	$q_{e,\,cal}$ (mg·g^{-1})	R^2
13.90	2.97	13.90	1

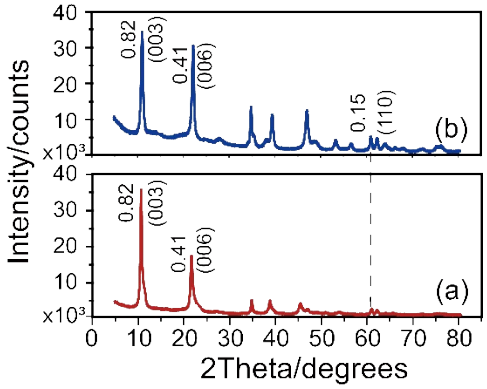

Fig. S1 (a) XRD pattern of before-immersion sample of DMG-LDH composite; (b) XRD pattern of post-immersion sample of DMG-LDH composite in deionized water of pH=5.