

Supporting Information

**Ni-Mn LDH decorated 3D Fe-inserted and N-doped carbon frameworks composites for efficient uranium (VI) removal**

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## **Characterization**

### **Materials Characterization**

The morphology of the samples was characterized with scanning electron microscope (SEM, JEOLJSM-6480A microscope) and transmission electron microscopy (TEM) on a FEI Tecnai G2 S-Twin with an accelerating voltage of 200 kV. The texture and surface properties of the samples were measured by nitrogen adsorption isotherms using an AUTOSORB-IQ2-MP analyzer and ESCALAB 250Xi X-ray photoelectron spectrometer. The samples were analyzed by X-ray diffraction (XRD) which was performed on a Rigaku D/max-III B diffractometer with Cu K $\alpha$  irradiation ( $\lambda = 1.54178 \text{ \AA}$ ). The X-ray source was operated at 40 kV and the current used in XRD measurements was 150 mA. Fourier-transform infrared (FT-IR) spectra were recorded with an AVATAR 360 FT-IR spectrophotometer (resolution 4 cm $\text{\AA}$ 1) using standard KBr pellets. The magnetic measurement was carried out with a vibrating sample magnetometer (VSM, Lanzhou University LakeShore 7304).

### **Adsorption Measurement**

The uranium (VI) concentration was analyzed using the inductively coupled plasma atomic emission spectrometer (ICP-AES) and inductively coupled plasma mass spectrometry (ICP-MS), which were analyzed by IRIS Intrepid II XSP and X Series instrument, respectively.

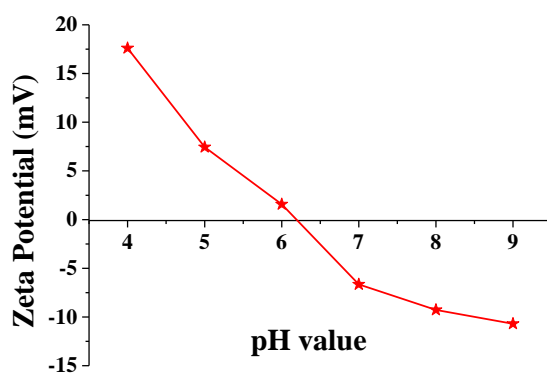


Fig. S1.  $\zeta$ -Potential of Fe-NCNF-LDH sample at different pH values.

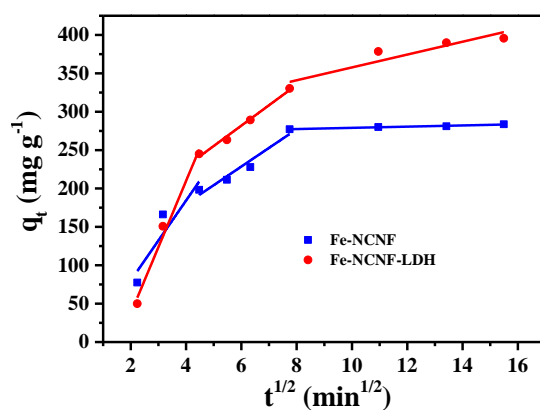


Fig. S2 Intra-particle diffusion kinetics for the adsorption of uranyl ions on Fe-NCNF and Fe-NCNF-LDH.

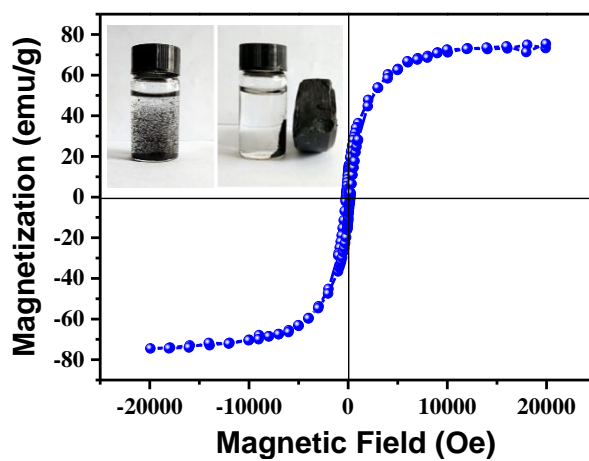
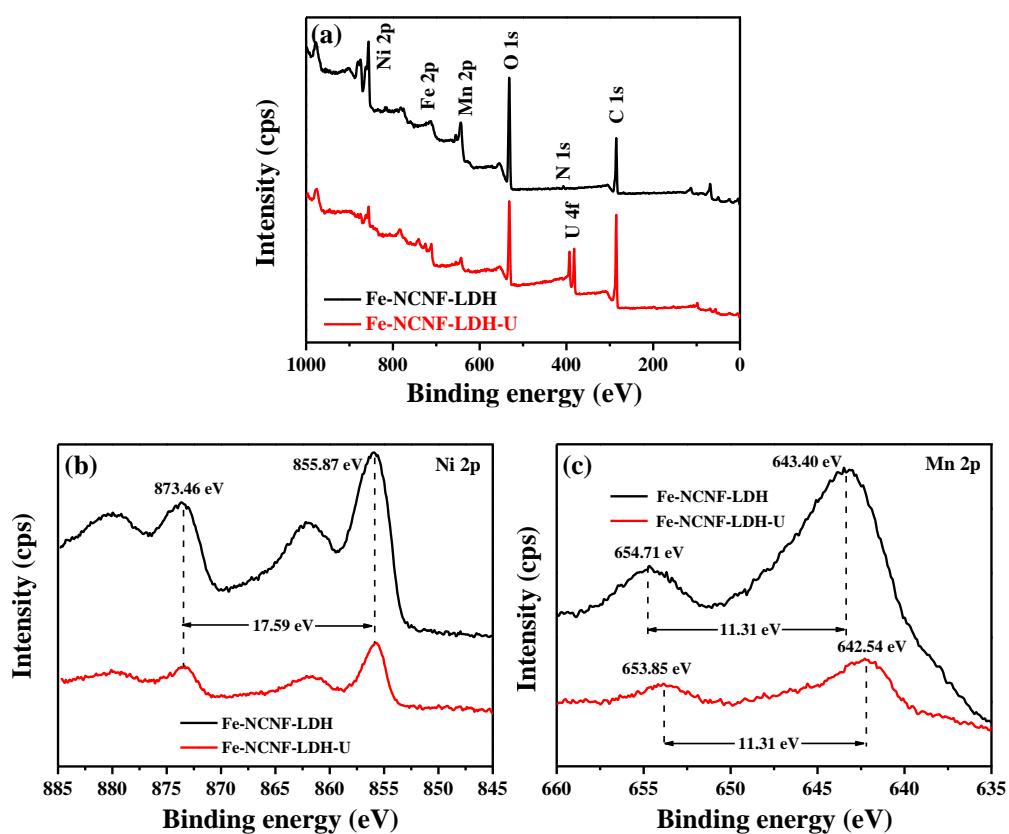
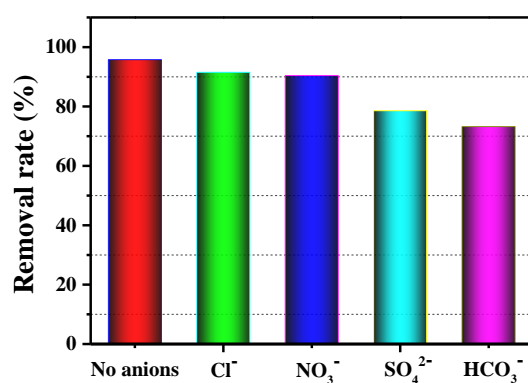


Fig. S3 Magnetic hysteresis curves and magnetic separation picture (inset) of Fe-NCNF-LDH.



**Fig. S4** XPS spectra (a), Ni 2p spectra (b), Mn 2p spectra (c) of Fe-NCNF-LDH and Fe-NCNF-LDH-U.



**Fig. S5.** Effect of coexisting anions on the adsorption of uranium (VI) by the Fe-NCNF-LDH.

**Table S1.** Pseudo-first-order and pseudo-second-order constants and values of  $R^2$  for Fe-NCNF-LDH.

T(K)	Pseudo-first-order			Pseudo-second-order		
	$k_1(\text{min}^{-1})$	$q_e (\text{mg g}^{-1})$	$R^2$	$k_2 (\text{mg min})$	$q_e (\text{mg g}^{-1})$	$R^2$
Fe-NCNF	0.066	274.1824	0.919	0.0003	301.9573	0.956
Fe-NCNF-LDH	0.0412	382.5334	0.969	0.0001	434.9932	0.977

**Table S2** Parameters of the intra-particle diffusion model of uranium (VI) adsorption on Fe-NCNF and Fe-NCNF-LDH.

	$k_{p1}$	$R_1^2$	$k_{p2}$	$R_2^2$	$k_{p3}$	$R_3^2$
<b>Fe-NCNF</b>	52.07	0.93	24.28	0.97	0.78	0.98
<b>Fe-NCNF-LDH</b>	86.21	0.99	26.54	0.99	8.36	0.94

**Table S3.** Isotherm constants and values of  $R^2$  for Fe-NCNF-LDH.

T(K)	Langmuir isotherm			Freundlich isotherm		
	$q_m (\text{mg}\cdot\text{g}^{-1})$	$b (\text{L}\cdot\text{mg}^{-1})$	$R^2$	$K (\text{L}\cdot\text{g}^{-1})$	n	$R^2$
298	598.497	0.002	0.977	77.601	0.013	0.915
308	769.532	0.001	0.971	139.402	0.007	0.837
318	851.607	0.001	0.931	229.083	0.004	0.652

**Table. S4** The maximum adsorption capacity of different adsorbents for uranium (VI).

Adsorbents	Adsorption Capacity mg-U/g-adsorbent	Conditions	Ref.
$\text{Fe}_3\text{O}_4@\text{SiO}_2$ magnetic composites	52.36	$T = 298 \text{ K}, \text{pH}=6.0$	[1]
Graphene oxide-manganese dioxide	185.2	$T = 298 \text{ K}, \text{pH} =$	[2]
Fe(II)-Al(III) layered double hydroxides	99.01	3.8	[3]
PAF magnetic sorbent	115.3	$T = 298 \text{ K}, \text{pH}=6.0$	[4]
LDH@MMT	33	$T = 298 \text{ K}, \text{pH} = 5$	[5]
AO-g-MWCNTs	145	$= 298.15\text{K}, \text{pH}=9$	[6]
GO	299	$= 298 \text{ K}, \text{pH} = 4.5$	[7]
Layered double hydroxide/graphene	277.80	$\text{RT}, \text{pH} = 4$	[8]
Fe-NCNF-LDH	598.50	$T = 298 \text{ K}, \text{pH} = 4$	this work
		$T = 298 \text{ K}, \text{pH} = 7$	

**Table S5.** Thermodynamics parameters for uranium (VI) adsorption on Fe-NCNF-LDH.

T (K)	$\Delta G$ (kJ/mol)	$\Delta H$ (kJ/mol)	$\Delta S$ (J/(mol·K))
298	-79.66		
308	-82.34	77.76	267.57
318	-85.01		

**Table S6** Selected results for the extraction of uranyl ions from simulated seawater

Elements	Ion concentration (ppb)		Removal (%)
	initial	final	
U	3.227	0.102	96.84
Fe	118.3	63.9	45.98
Al	14.42	8.333	42.21
Na	1.667*10 <sup>4</sup>	1.202*10 <sup>4</sup>	38.68
V	0.537	0.389	27.56
Ca	9.903*10 <sup>3</sup>	7.072*10 <sup>3</sup>	28.59
Mg	6.521*10 <sup>3</sup>	5.504*10 <sup>3</sup>	15.6
Cu	52.33	50.18	4.11
Ba	90.46	88.3	2.39
Zn	38.95	38.88	0.18

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