

Supplementary Materials

Efficient Removal of Congo Red, Methylene Blue and Pb(II) by Hydrochar–MgAlLDH Nanocomposite: Synthesis, Performance and Mechanism

Yang Huang ^{1,2}, Wei Yin ¹, Tian-Lei Zhao ¹, Meng Liu ¹, Qi-Zhi Yao ³ and Gen-Tao Zhou ^{1,4,*}

¹ CAS Key Laboratory of Crust-Mantle Materials and Environments, School of Earth and Space Sciences, University of Science and Technology of China, Hefei 230026, China; esthy@mail.ustc.edu.cn (Y.H.)

² School of Environmental Engineering and Resources, University of Science and Technology of Southwest, Mianyang 621010, China

³ School of Chemistry and Materials Science, University of Science and Technology of China, Hefei 230026, China

⁴ CAS Center for Excellence in Comparative Planetology, University of Science and Technology of China, Hefei 230026, China

* Correspondence: gtzhou@ustc.edu.cn; Tel.: +86-551-63600533

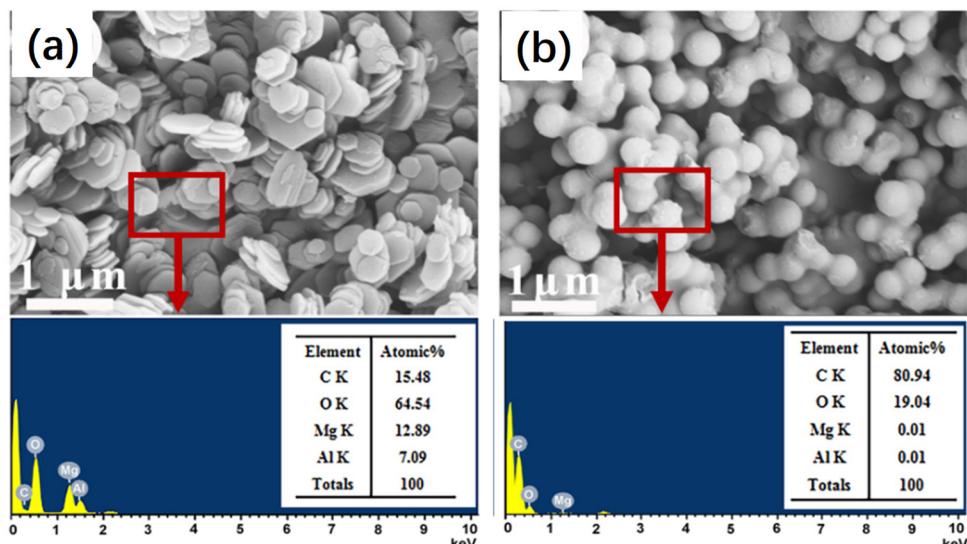


Figure S1. SEM images and EDS analyses of the synthesized MgAlLDH (a) and HC (b).

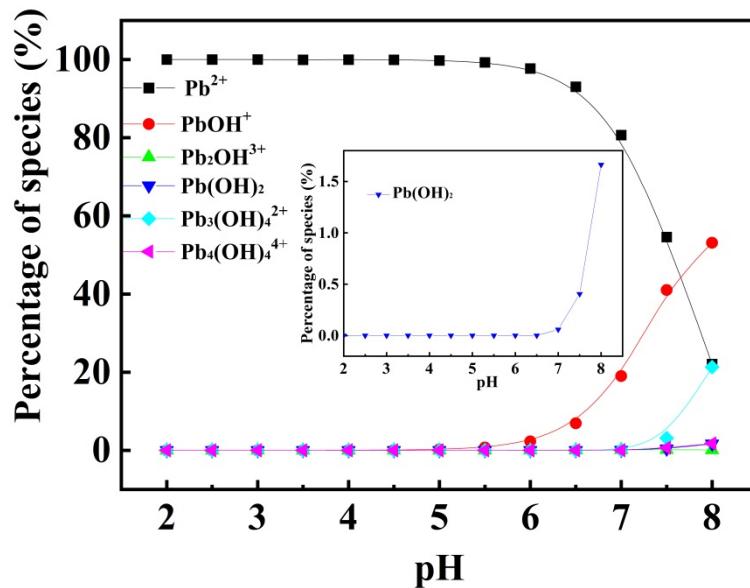


Figure S2. The species distribution of Pb(II) in aqueous solution (50 mg/L Pb(II)), calculated by a chemical equilibrium program Visual MINTEQ (version 3.1).

TEXT S1 Adsorption kinetic and isothermal models

Adsorption data at different contact times were fitted with pseudo-first-order (Equation S1) and pseudo-second-order (Equation S2) kinetics to determine the kinetic parameters [38]:

$$q_t = q_e (1 - \exp(-k_1 t)) \quad (S1)$$

$$q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e t} \quad (S2)$$

where q_e and q_t are the amounts of pollutants adsorbed (mg/g) at equilibrium and at time t (min), k_1 is the pseudo-first-order rate constant (min^{-1}), and k_2 (g/mg/min) is the rate constant of the pseudo-second-order kinetics.

The Langmuir and Freundlich isothermal models are expressed by Equations S3 and S4, respectively [38]:

$$q_e = q_m b C_e / (1 + b C_e) \quad (S3)$$

$$q_e = K_f C_e^{1/n} \quad (S4)$$

where constant b is related to the energy of adsorption (L/mg), q_m is the maximum adsorption capacity (mg/g), K_f is an indicator of the adsorption capacity, and $1/n$ is the adsorption intensity.

Table S1. Kinetic parameters of CR, MB or Pb(II) adsorption onto the HC–MgAlLDH.

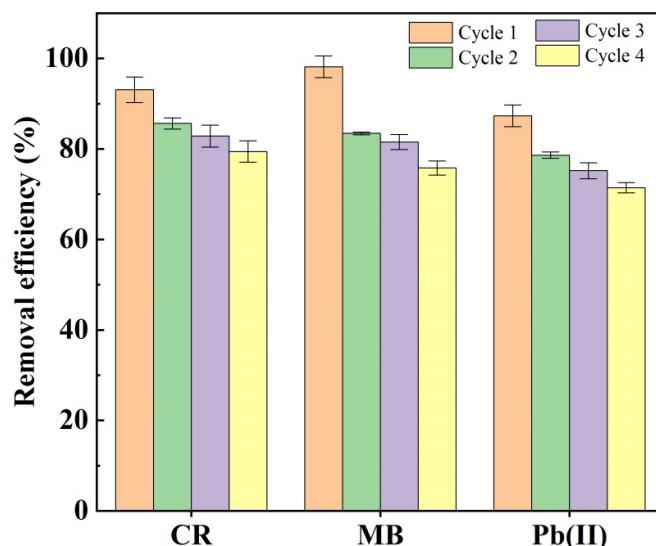
Pollutants	$q_{e,\text{exp}}$ (mg/g)	Pseudo-First-Order			Pseudo-Second-Order		
		k_1 (min^{-1})	$q_{e,\text{cal}}$ (mg/g)	R^2	k_2 (g/(mg·min))	$q_{e,\text{cal}}$ (mg/g)	R^2
CR	172.85	0.02709	156.99	0.9419	2.33×10^{-4}	167.36	0.9835
MB	161.67	0.1795	153.06	0.9670	0.0023	157.19	0.9869
Pb(II)	38.44	0.0787	35.49	0.9491	0.0034	37.05	0.9897

Table S2. Adsorption isothermal parameters of CR, MB and Pb(II) by the HC–MgAlLDH nano-composite in various systems.

Pollutant	Mono/multi-component system	Langmuir Model			Freundlich Model		
		q_m (mg/g)	b (L/mg)	R^2	K_f (mg/g)	n	R^2
CR	Only CR	348.78	0.1497	0.9857	82.9671	3.2636	0.9030
	CR+MB	332.85	0.03667	0.9793	36.0747	2.3752	0.9008
	CR+Pb(II)	337.55	0.05887	0.9938	50.0129	2.6443	0.9168
	CR+MB+Pb(II)	328.83	0.01741	0.9641	18.3591	1.9401	0.9259
MB	Only MB	256.54	0.3851	0.9799	86.7985	4.3003	0.9002
	MB+CR	242.69	0.1153	0.9706	49.2889	3.0694	0.9174
	MB+Pb(II)	236.45	0.1028	0.9673	46.7023	3.0525	0.9162
	MB+CR+Pb(II)	188.96	0.2443	0.9491	55.0183	4.0120	0.8116
Pb(II)	Only Pb(II)	33.55	0.2950	0.9376	9.0174	3.2201	0.9260
	Pb(II)+CR	56.21	0.4520	0.9561	18.5252	3.7301	0.8664
	Pb(II)+MB	47.35	0.4135	0.9754	14.9293	3.5191	0.8921
	Pb(II)+CR+MB	63.28	0.2867	0.8863	14.4048	2.5713	0.9108

Table S3. Comparison of the Langmuir removal capacities (q_m) of various LDH-based adsorbents to single CR, MB and Pb(II).

Sorbate	Adsorbent	Experimental Conditions	q_m (mg/g)	Ref.
CR	MgAlLDH	T=25°C	111.11	[33]
	Borate intercalated MgAlLDH	—	166.39	[43]
	ZnFe ₂ O ₄ /MgAlLDH	T=25°C	294.12	[44]
	MgAlLDH modified diatom	pH=7	305.80	[45]
MB	HC-MgAlLDH	pH=5, T=25°C	348.78	This work
	ZIF-67/CoAlLDH	T=30°C	57.24	[46]
	NiFeLDH decorated montmorillonite	T=25°C	99.18	[20]
	Dodecyl sulfate modified ZnAlLDH	—	113.00	[47]
	Hydrochar capped MgAlLDH	pH=7, T=30°C	226.00	[30]
Pb(II)	HC-MgAlLDH	pH=5, T=25°C	256.54	This work
	MgAlLDH	T=25°C	16.93	[48]
	MgFeLDH	T=25°C	18.45	[19]
	Tartrate intercalated MgAlLDH	—	8.40	[49]
	HC-MgAlLDH	pH=5, T=25°C	33.55	This work

**Figure S3.** Recycle test for the removal of CR, MB and Pb(II) by HC–MgAlLDH nanocomposite.

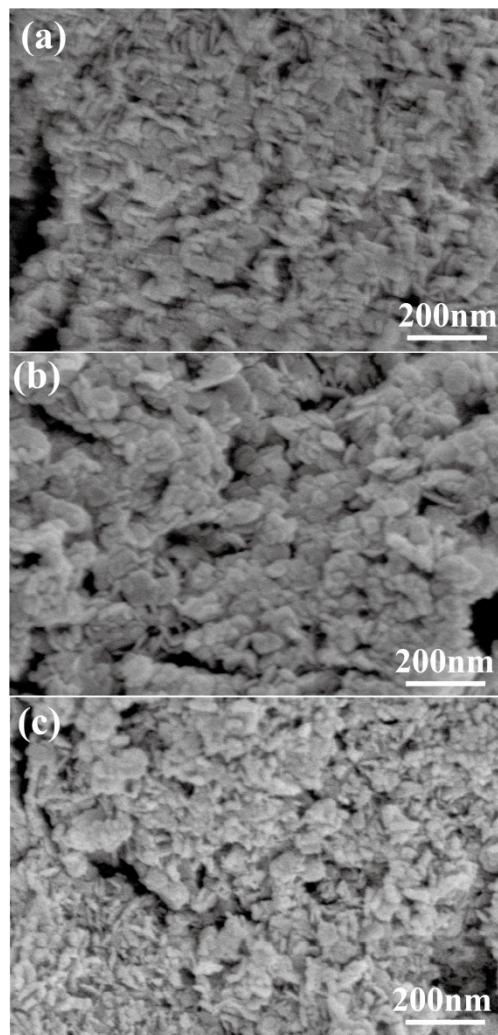


Figure S4. SEM images of regenerated HC-MgAlLDH nanocomposite after treatment of CR (a), MB (b) and Pb(II) (c).

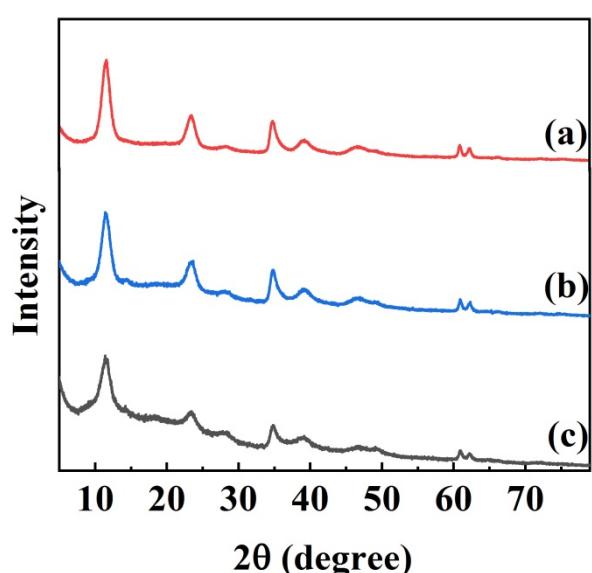


Figure S5. XRD patterns of regenerated HC-MgAlLDH nanocomposite after treatment of CR (a), MB (b) and Pb(II) (c).