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Supporting Information

Unveiling the dual roles of the intercalation of $[MoS_4]^{2-}$ clusters in boosting heavy metal capture by Ca-Al layered double hydroxide

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This document contains *six* figures and *six* tables.

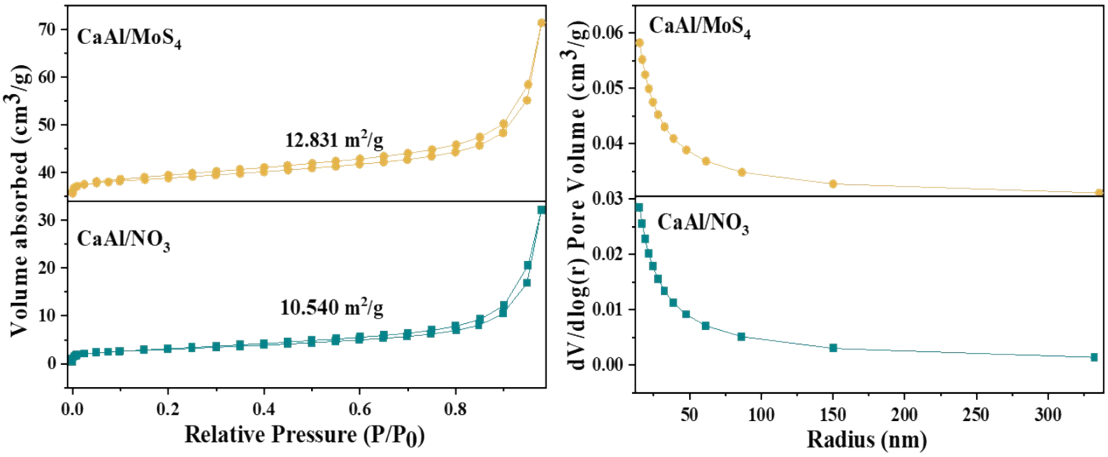

Fig. S1. N_2 adsorption-desorption isotherms (left) and pore-size distribution (right) of LDH/NO_3 and LDH/MoS_4.

Table S1 Summary of BET analysis results of LDH/NO_3 and LDH/MoS_4.

Adsorbents	BET surface (m^2/g)	Pore volume (cm^3/g)	Average pore diameter (nm)
LDH/NO_3	10.540	0.048	94.25
LDH/MoS_4	12.831	0.053	86.66

Table S2

Kinetics parameters for the adsorption of Pb(II) and Cd(II) onto LDH/NO_3 and LDH/MoS_4.

Heavy metal	Material	pseudo-first-order model			pseudo-second-order model		
		q_e (mg/g)	K_1 1/min	R^2	q_e mg/g	K_2 mg/(g·min)	R^2
Pb(II)	LDH/NO_3	322.4	0.030	0.880	368.6	9.074×10^{-5}	0.939

	LDH/MoS_4	1147.2	0.076	0.879	1202.1	9.792×10^{-5}	0.964
Cd(II)	LDH/NO_3	285.8	0.009	0.977	304	4.443×10^{-5}	0.986
	LDH/MoS_4	639.8	0.021	0.969	678.3	4.530×10^{-5}	0.980

Table S3

Intraparticle diffusion modeling for the adsorption of Pb(II) and Cd(II) onto LDH/NO_3 and LDH/MoS_4.

Heavy metal	Material	K_a	C_1	R^2	K_b	C_2	R^2	K_c	C_3	R^2
Pb(II)	LDH/NO_3	245.11	9.50	0.91	48.16	194.40	0.99	7.56	333.88	0.59
	LDH/MoS_4	1309.03	103.15	0.89	192.08	818.13	0.99	0.15	1201.02	0.49
Cd(II)	LDH/NO_3	201.17	75.38	0.99	161.03	-33.42	0.99	67.46	88.40	0.99
	LDH/MoS_4	507.53	-47.65	0.95	74.45	440.99	0.95	3.97	658.84	0.99

Table S4

Comparison of Pb(II) and Cd(II) adsorption capacities of various adsorbent with LDH/MoS_4

Pollutant	Adsorbent	q_m (mg/g)	Refs
Pb(II)	Ti/Zr-DBMD	175.0	Ref.[1]
	$MgFe_2O_4$-NH_2@sRHB	198.9	Ref.[2]
	TMMs	303.2	Ref.[3]
	Co-Al-LDH @ Fe_2O_3/3DPCNF	426.8	Ref.[4]
	BC-LDH	294.0	Ref.[5]
	GOCS	208.9	Ref.[6]
	MnFe-LDH/$MnFe_2O_3$@3DNF	591.8	Ref.[7]
	Biochar/Zn-Al LDH	226.1	Ref.[8]
	LDH/MoS_4	1202.1	This work
Cd(II)	B-LDHs-BC	119.0	Ref.[9]
	PVA/BC/$CaCO_3$	238.6	Ref.[10]
	BC-FM	120.8	Ref.[11]
	Fe_3O_4-$FeMoS_4$	140.5	Ref.[12]
	Mg-Fe LDH/GO	174.8	Ref.[13]
	SA/PEI	464.2	Ref.[14]
	CSs–LDHs	475.0	Ref.[15]
	LDH@GO-SH	102.8	Ref.[16]
	LDH/MoS_4	678.3	This work

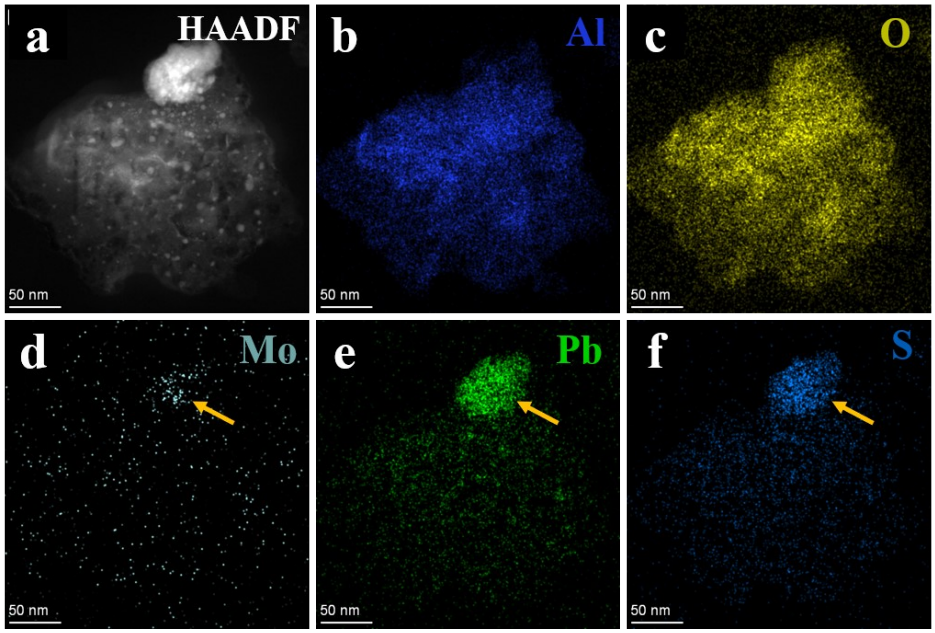

Fig. S2. TEM and element mapping of LDH/MoS_4-Pb.

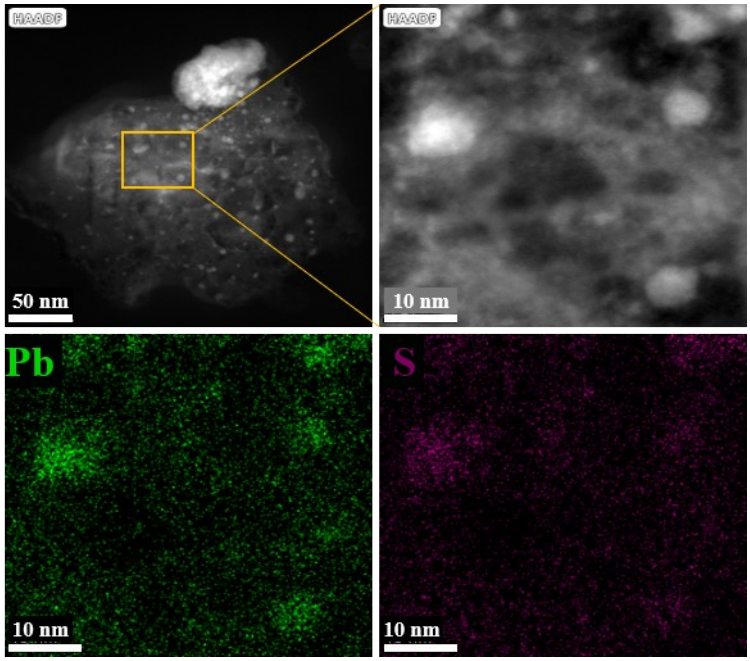

Fig. S3. TEM and element mapping of LDH/MoS_4-Pb.

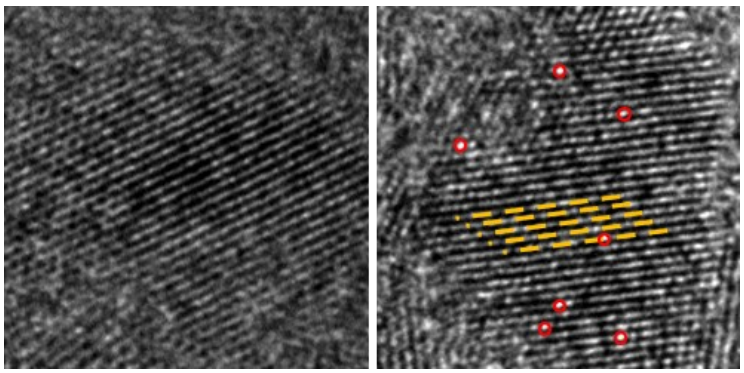

Fig. S4. HRTEM images of LDH/MoS_4 (left) and LDH/MoS_4-Cd (right).

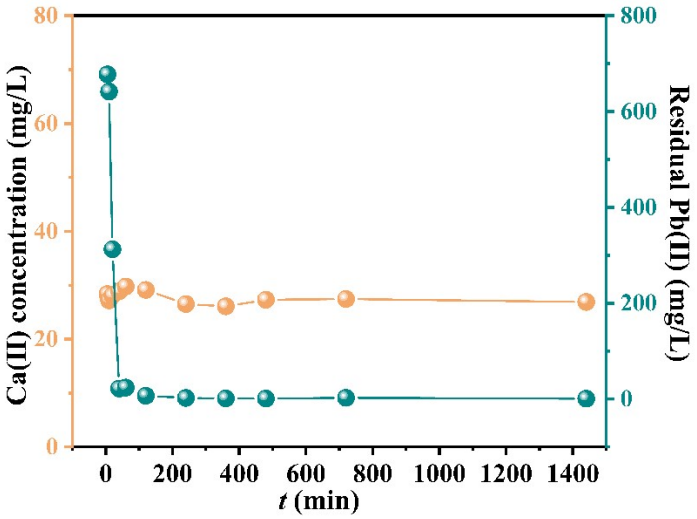

Fig. S5. Effect of time variation on the concentrations of the residual Pb(II) and the leached Ca(II) in solution (pH: 5, adsorbent dosage: 1 g/L)

Table S5

O, S, Mo, Cd and Pb content on the surface of LDH/MoS_4-Pb and LDH/MoS_4-Cd (based on XPS study).

	Content, %				
	O	S	Mo	Cd	Pb
LDH/MoS_4-Pb	32.69	16.92	1.19	-	7.31
LDH/MoS_4-Cd	36.27	2.70	0.58	14.55	-

Table S6 The calculated adsorption energy for LDH/MoS_4 toward Pb(II) and Cd(II).

Heavy metal ions	$E_{LDH/MoS4}$ (eV)	E_M (eV)	E_b (eV)
Pb(II)	-645.64	-3.71	-1.68
Cd(II)	-642.33	-0.68	-1.40

$E_{LDH/MoS4}$: the calculated corresponding energies for different systems；

E_M： Energy of a single metal ion；

E_b: the binding energy.

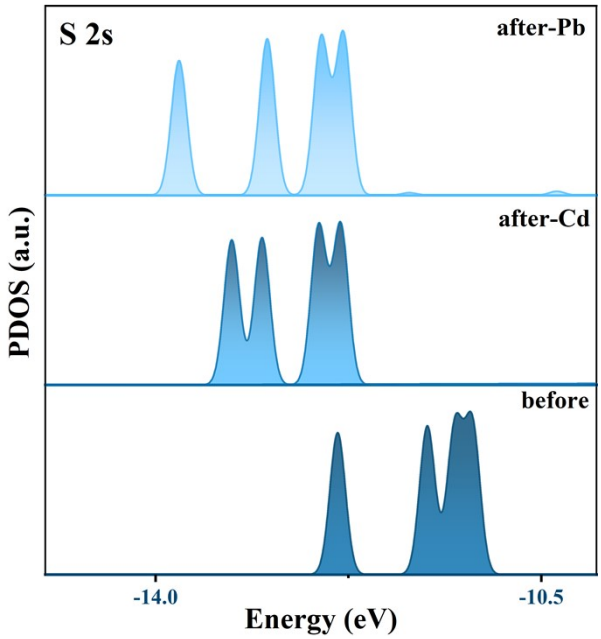

Fig. S6. PDOS for S 2s of LDH/MoS_4 before and after adsorption of Pb(II) and Cd(II).

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