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Supporting Information

Evidence of multiple sorption modes in layered double hydroxides using Mo as structural probe

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The Supporting Information contains 21 pages, 2 texts, 7 tables and 13 figures.

20 **Text S1.** Characterization of Synthesized CaAl LDHs.

21 Freshly synthesized Cl^- and SO_4^{2-} CaAl LDHs have well-crystallized hexagonal-shaped plate
22 morphology (Figure S2) but different average sizes: around $\text{Ø}0.5 \mu\text{m}$ for AFm- Cl_2 and $\text{Ø}5 \mu\text{m}$
23 AFm- SO_4 , respectively. The specific surface areas and the average edge lengths thus differ
24 (Table S6), with lateral surface (S_l) area concentrations equal for AFm- Cl_2 and AFm- SO_4 to
25 2.19×10^{18} and $2.29 \times 10^{17} \text{ nm}^2 \text{ g}^{-1}$, respectively, as obtained from¹:

$$26 \quad S_l = \frac{P}{S \times \rho} \quad (1)$$

27 where P , S , and ρ represent CaAl LDHs particle's perimeter (nm), basal plane surface area
28 (nm^2), and density (g nm^{-3}), respectively.

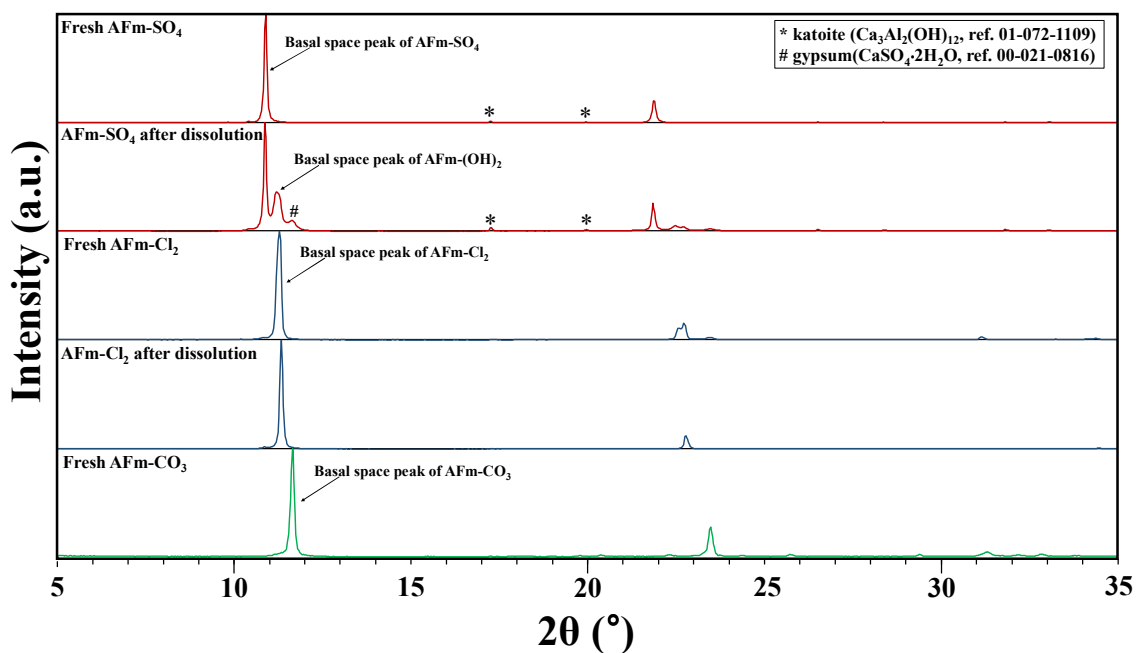
29 X-ray diffraction (XRD) was used to determine the nature and amount of eventual impurities
30 and the inter-layer distances of the synthesized CaAl LDHs (Figure S1), including AFm- SO_4 ,
31 AFm- Cl_2 , and AFm- CO_3 . Due to the sulfate-deficiency during the synthesis, no ettringite was
32 formed in the fresh AFm- SO_4 , but katoite [$\text{Ca}_3\text{Al}_2(\text{OH})_{12}$] was observed as an impurity. AFm-
33 SO_4 and AFm- Cl_2 powders were also characterized after 7 days in equilibrium with N_2 -
34 saturated water at pH 12.4. The (003) inter-layer distance of AFm- SO_4 , was initially
35 $8.128 \pm 0.006 \text{ \AA}$ at 5% relative humidity (RH) corresponding to 10.5 H_2O hydration state², and
36 it changed to a shorter distance of $7.884 \pm 0.006 \text{ \AA}$ with time. The lack of SO_4^{2-} is compensated
37 with OH^- in the inter-layer space, resulting in the formation of AFm-OH with 13 H_2O
38 molecules². Besides, a tiny diffraction peak occurred at 11.65° after equilibration of AFm- SO_4
39 in water, which is probably derived from gypsum formed during sample drying. A strict N_2
40 protection gives us confidence to exclude formation of AFm- CO_3 (i.e., CO_2 contamination) in
41 both reacted AFm- SO_4 and AFm- Cl_2 . No detectable impurity is observed with the AFm- Cl_2
42 phase, characterized by an inter-layer distance of $7.838 \pm 0.006 \text{ \AA}$.

43 Atomic ratios between each element in CaAl LDHs were estimated by digesting 100 mg CaAl
44 LDHs solid phase in a certain amount of 1% HNO₃, and by measuring total aqueous Ca, Al,
45 and S concentrations using inductively coupled plasma optical emission spectrometry (ICP-
46 OES). This nitrolysis method was applied for each CaAl LDH in duplicate. Katoite impurity
47 in AFm-SO₄ was inferred from the Ca/Al stoichiometry. Structural water content was
48 determined by thermal gravimetric analysis (TGA) and derivative thermogravimetric analysis
49 (DTG) curves (Figure S9) and dehydration and dehydroxylation were included in the total
50 water loss till 700 °C³, at which temperature the water loss is assumed to be completed. Based
51 on the calculation, 78 at.% AFm-SO₄ and 22 at.% katoite composed the product for AFm-SO₄
52 synthesis. The experimental chemical formula of AFm-SO₄ and AFm-Cl₂ were
53 Ca₄Al₂O₆(SO₄)_{0.92}(OH)_{0.08}·6.4H₂O and Ca₄Al₂O₆Cl_{1.64}(OH)_{0.36}·7.4H₂O, respectively.

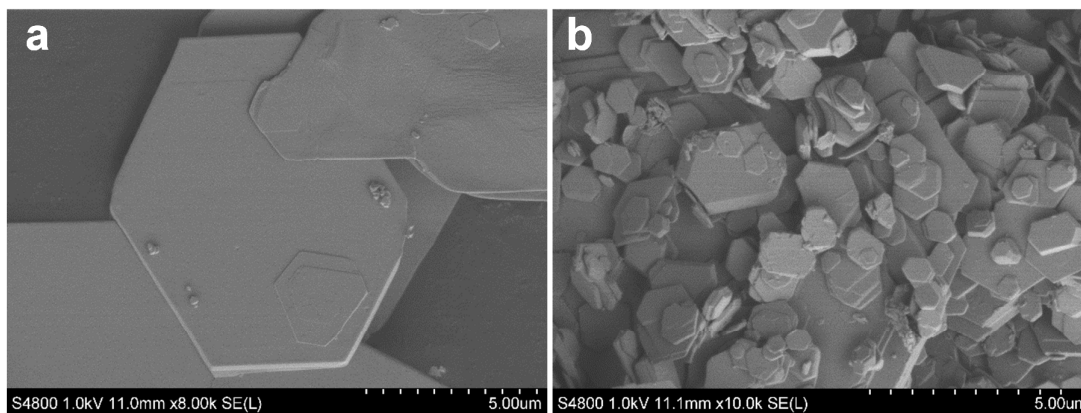
54 **Text S2.** Instability of CaAl LDHs in presence of Mo.

55 The released Cl⁻ is more than the stoichiometric amount that may be derived from the
56 corrosion of double-layer structure. This is confirmed by the newly appearance of a katoite
57 diffraction peak for Mo-reacted samples in the synchrotron based XRD patterns since sample
58 CIA (Figure S8), in agreement with saturation index (SI) computed from the aqueous phase
59 composition after reaction of both CaAl LDHs with molybdate for 48 h (Figure S10).
60 Molybdate could also affect heavily the metastability of AFm-SO₄ and result in ettringite
61 formation (Figure S8). Compared to the dissolution products for 7 days (Figure S1), phases
62 transformation seems to be promoted under the effect of molybdate. Indeed, the pair
63 distribution function (PDF) analysis of high energy XRD data conducted on six selected
64 sorption products, and compared to the calculated PDF patterns of katoite, ettringite, and
65 powellite, shows almost no apparent phase transformation, except the formation of katoite and
66 ettringite in Mo-reacted AFm-Cl₂ and AFm-SO₄, respectively (Figure S11 and Figure S12).

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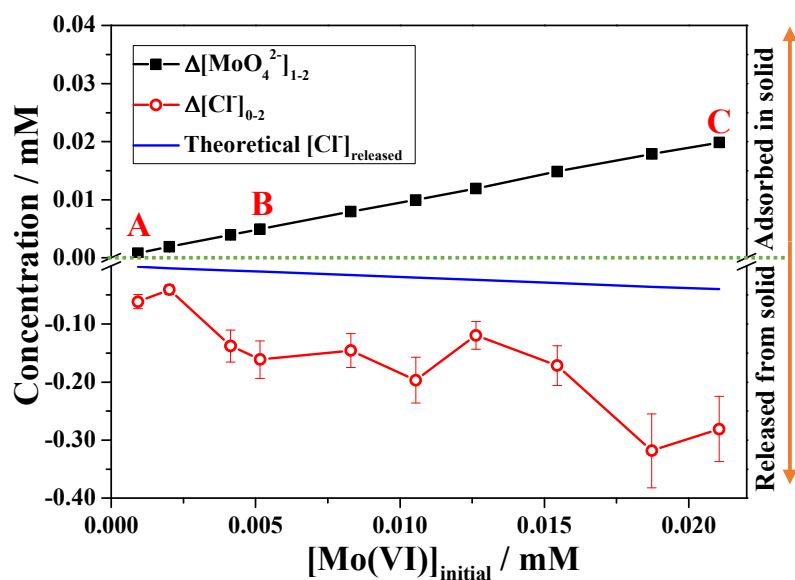


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 69 **Figure S1.** X-ray diffraction patterns of fresh CaAl LDHs and the phases after dissolution.
 70 The dissolution equilibrium was set at pH ~12.4 for 7 days and samples were mounted
 71 oriented at relative humidity of 5%. Cu K α radiation at 1.5406 Å is used.



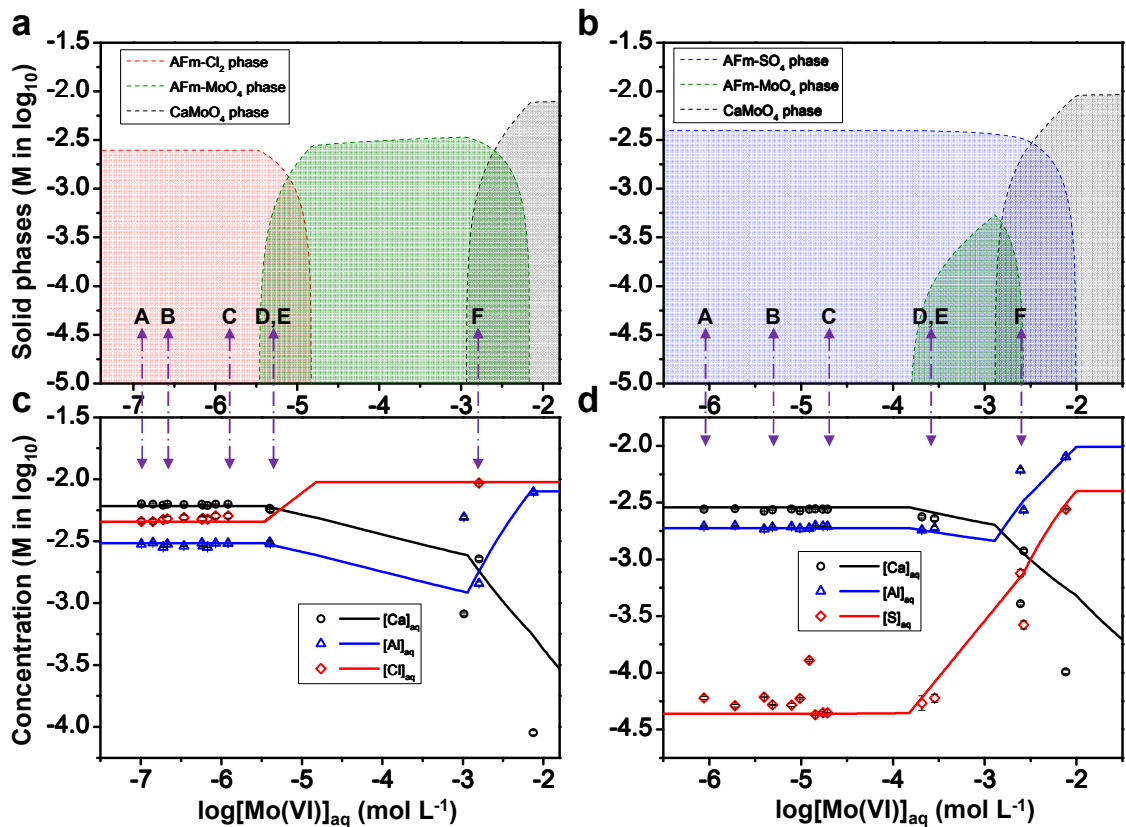
72
 73 **Figure S2.** FE-SEM micrographs of the synthesized CaAl LDHs. Scale bar, 5 µm. (a) Pristine
 74 AFm-SO₄ particles. (b) Pristine AFm-Cl₂ particles.

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77 **Figure S3.** Concentration variation profile of aqueous MoO_4^{2-} and Cl^- as a function of
 78 molybdate loading on AFm- Cl_2 . The change in concentration of adsorbed MoO_4^{2-} is plotted
 79 by the black solid dots and the released Cl^- is represented by the red hollow dots. The
 80 theoretical $[\text{Cl}^-]_{\text{released}}$ is estimated by a stoichiometric anion exchange, shown as the blue
 81 curve. The selected three adsorption products are marked by A, B, and C.



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83 **Figure S4.** PHREEQC modelling results as a function of $[\text{Mo(VI)}]_{\text{aq}}$ in equilibrium. (a)

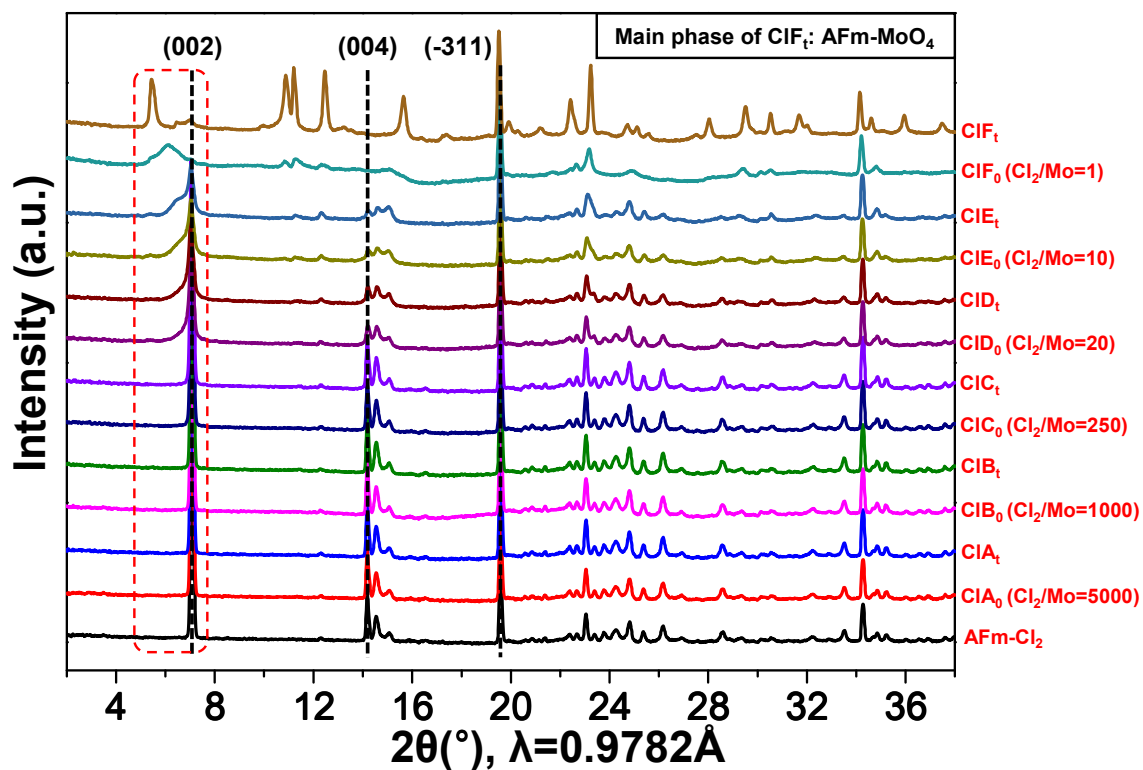
84 Dominated solid speciation in AFm-Cl₂ reactors. (b) Dominated solid speciation in AFm-SO₄

85 reactors. (c) Modelling results of aqueous Ca, Al, and Cl concentrations in AFm-Cl₂ reactors.

86 (b) Modelling results of aqueous Ca, Al, and S concentrations in AFm-SO₄ reactors. Reactors,

87 marked by A to F, correspond to the same ones in Figure 1.

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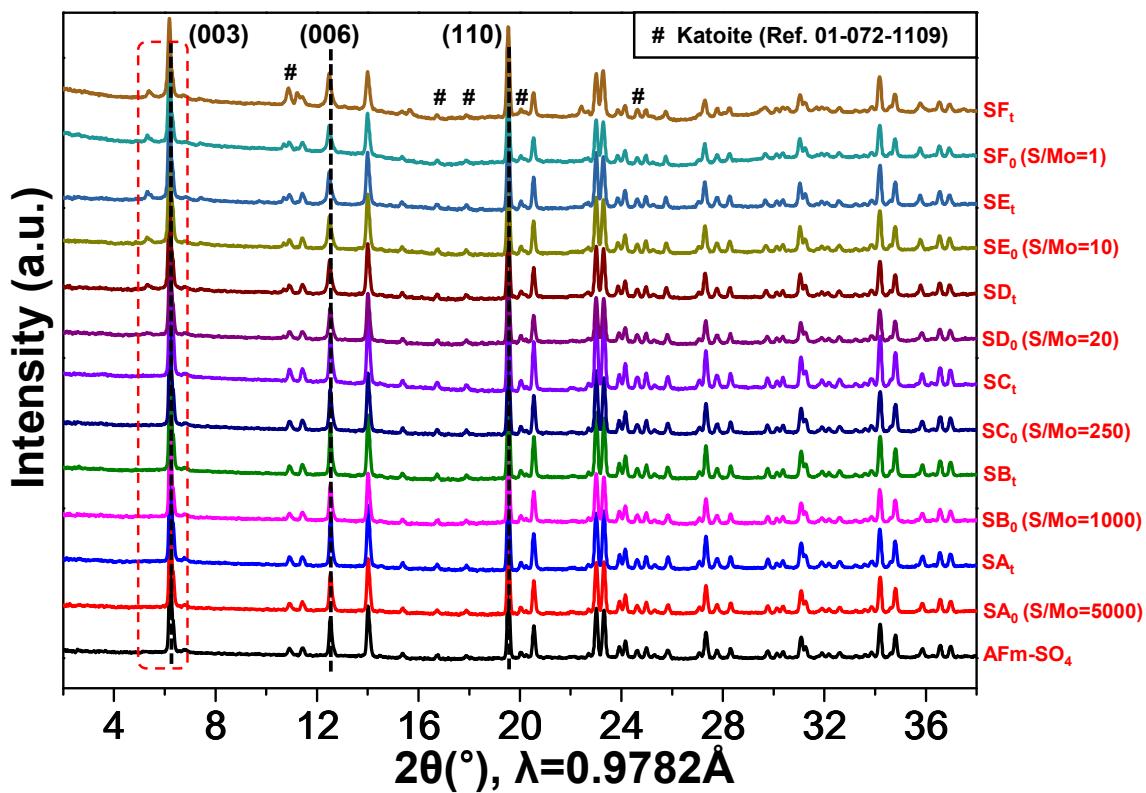
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90 **Figure S5.** In-situ time resolved XRD patterns of reacted AFm-Cl₂ at each loading step. The
 91 diffraction peak range of layer-to-layer distance (d_{002}), circled by red dotted lines, is enlarged
 92 in the main text to provide more details. The Cl₂/MoO₄ ratio at each step is shown following
 93 the sample name. The main phase of CIF_t is AFm-MoO₄, with no obvious signal of katoite
 94 and powellite observed.

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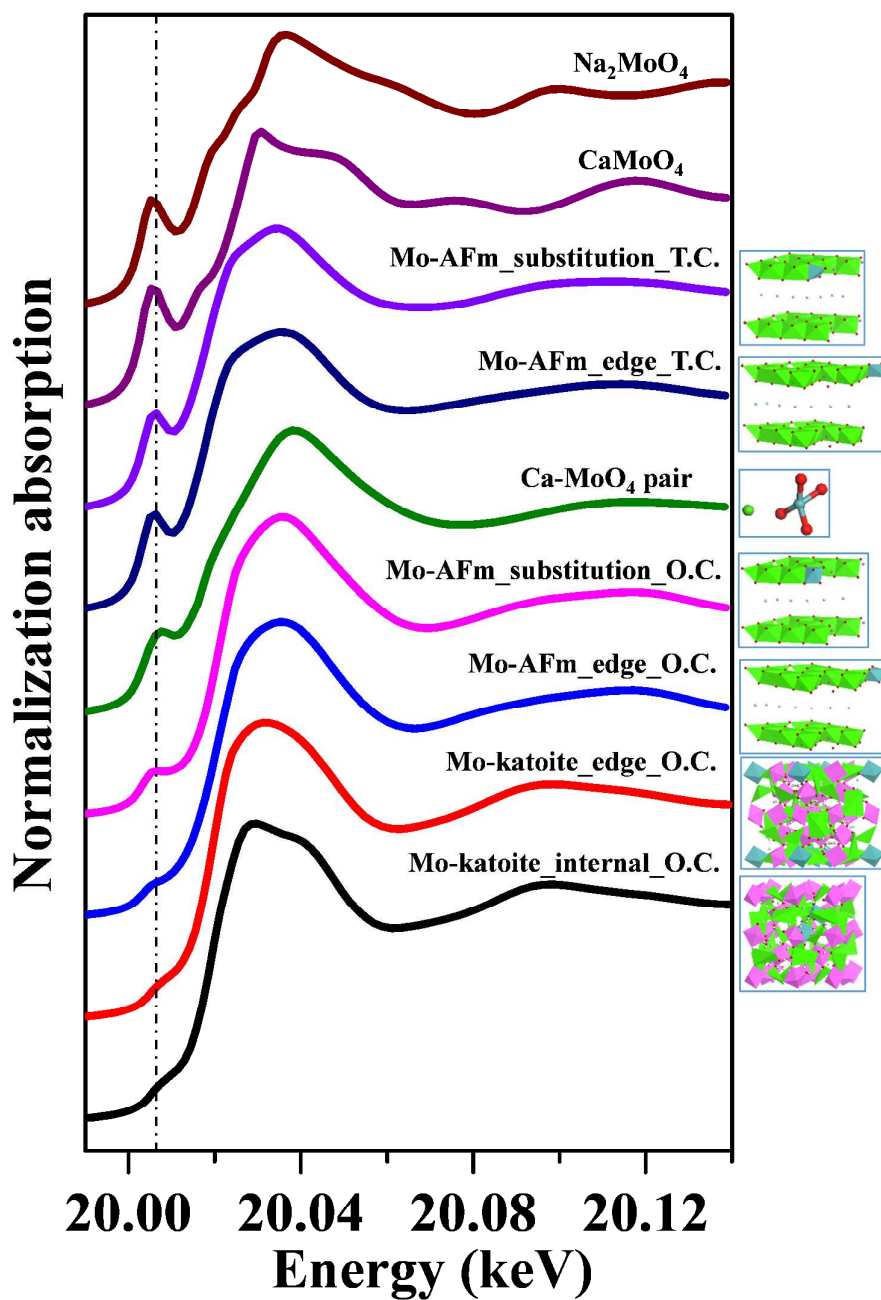
99 **Figure S6.** In-situ time resolved XRD patterns of reacted AFm-SO₄ at each loading step. The

100 diffraction peak range of layer-to-layer distance (d_{003}), circled by red dotted lines, is enlarged

101 in the main text to provide more details. The SO₄/MoO₄ ratio at each step is shown following

102 the sample name.

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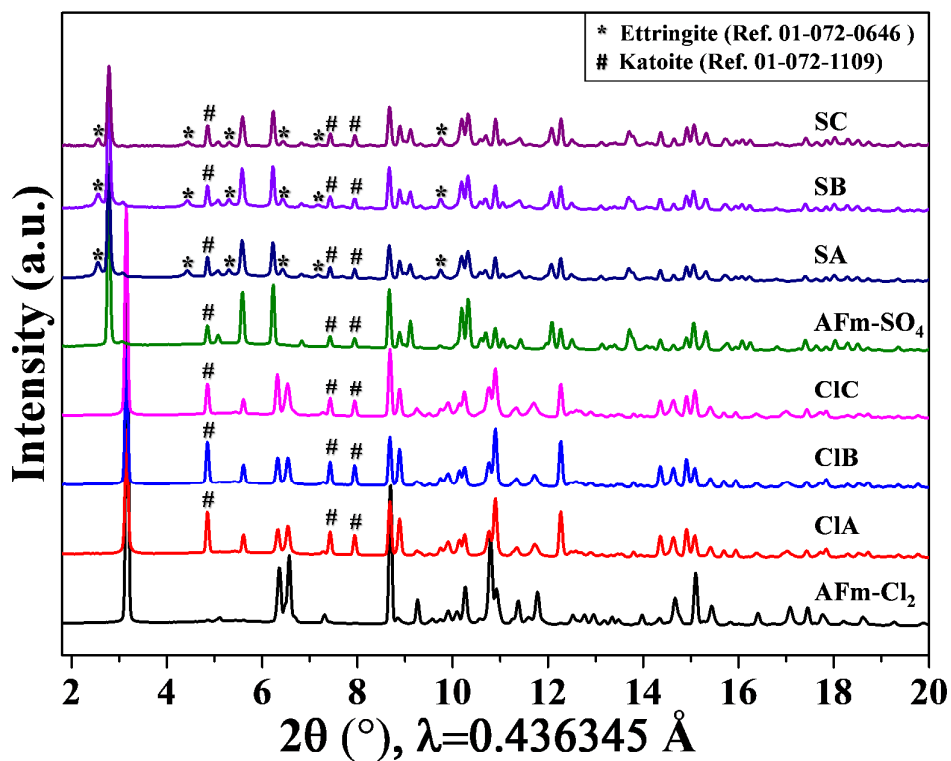


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105 **Figure S7.** Calculated Mo K-edge XANES spectra of different Mo coordination. From the
 106 bottom up, the pre-edge intensity decreases steadily and the molybdate coordination
 107 environments are pure O.C. (octahedral Mo(VI)-katoite, both inner-incorporated and edge-
 108 incorporated), distorted O.C. (octahedral Mo(VI)-AFm phases, both the substitution and
 109 epitaxial growth), distorted T.C. (Ca-MoO₄ ion pair, both the substitution and epitaxial

110 growth for tetrahedral Mo(VI)-AFm phases), and pure T.C. (CaMoO_4 and Na_2MoO_4). O.C
111 and T.C. represent octahedral coordination and tetrahedral coordination, respectively.

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114 **Figure S8.** Synchrotron based X-ray diffraction patterns of the selected adsorption products
115 for CaAl LDHs. Inter-layer peaks are located in the range of 2 - 4°.

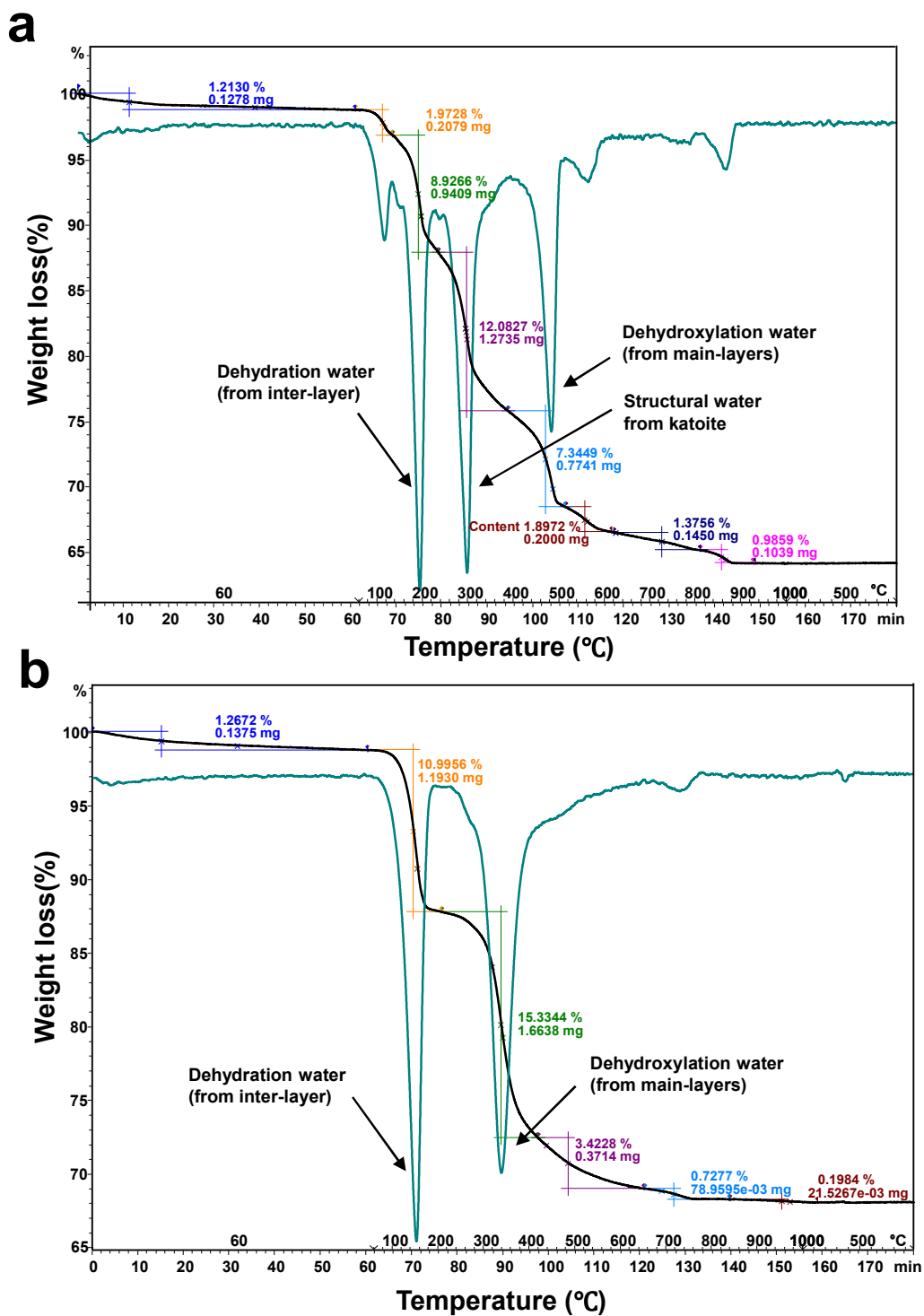
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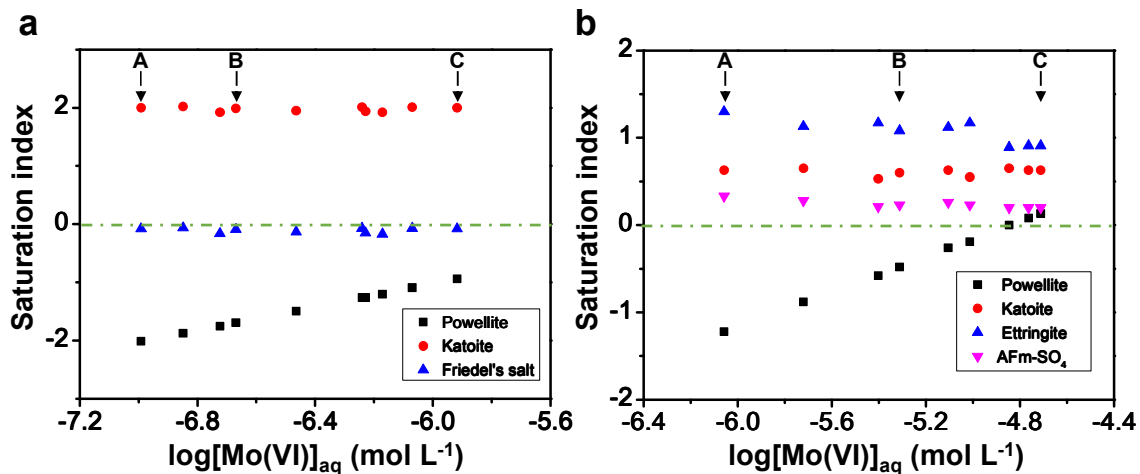
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122 **Figure S9.** Thermogravimetric analyses of the synthesized CaAl LDHs. (a) AFm-SO₄ phase.

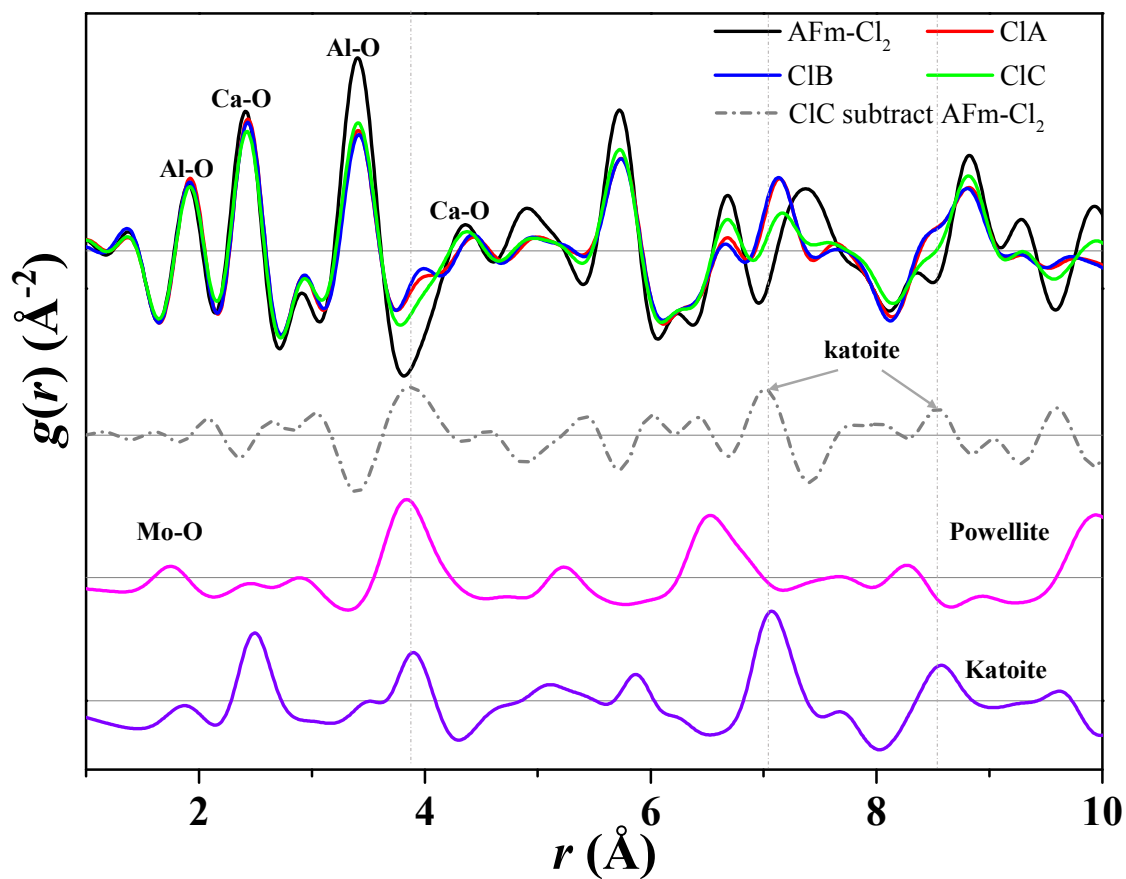
123 (b) AFm-Cl₂ phase. TGA is represented as the black curves and DTG in olive-green curves.



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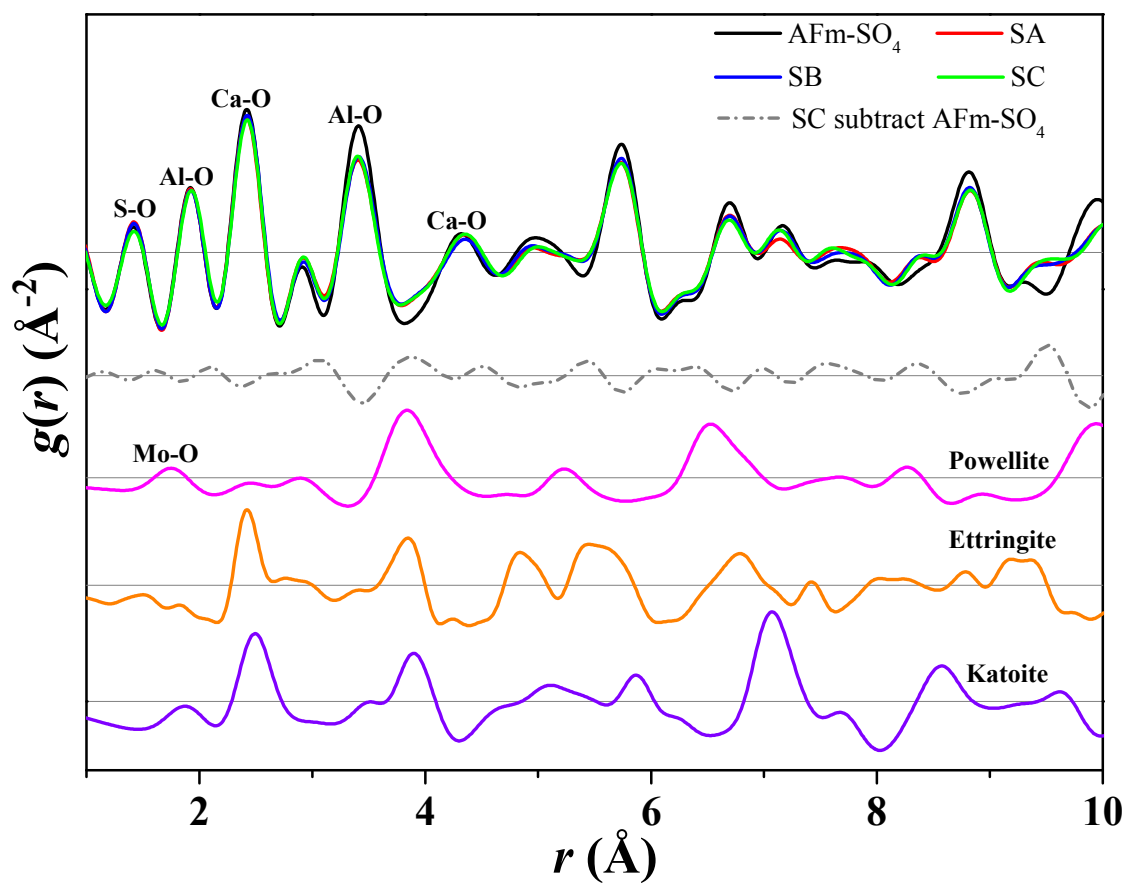
125 **Figure S10.** Saturation index of several possibly formed mineral phases as a function of
 126 $[\text{Mo(VI)}]_{\text{initial}}$. (a) For AFm-Cl₂ system. (b) For AFm-SO₄ system. Calculation is based on the
 127 aqueous data in batch sorption experiment on CaAl LDHs.

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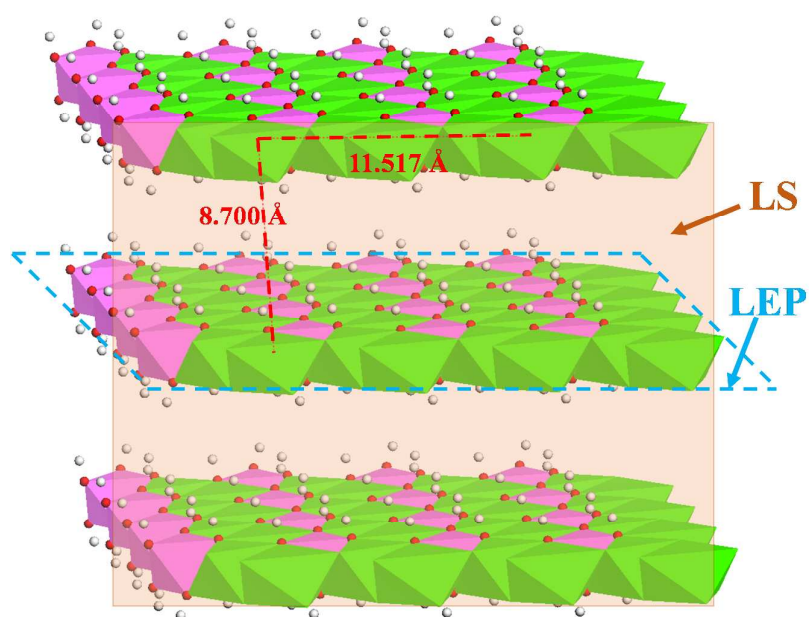
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130 **Figure S11.** Comparison of calculated and experimental PDF of katoite, powellite, and AFm-
 131 Cl₂ samples in batch sorption experiment. The grey dotted line is the subtracted signal of ClC
 132 on fresh AFm-Cl₂.



133
 134 **Figure S12.** Comparison of calculated and experimental PDF of katoite, powellite, ettringite
 135 and AFm-SO₄ samples in batch sorption experiment. The grey dotted line is the subtracted
 136 signal of SC on fresh AFm-SO₄.

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139 **Figure S13.** Schematic representation of the meaning of LS and LEP in LDHs. LS and LEP
140 represent lateral surface and lateral edge perimeter, respectively.

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157 **Table S1.** Solution content of selected elements for AFm-Cl₂ experiments before introducing
 158 MoO₄²⁻. Concentrations were determined after reaching dissolution equilibrium, which was
 159 confirmed by dissolution kinetics^a.

ID	pH	[Ca] _{aq} /mol L ⁻¹	[Al] _{aq} /mol L ⁻¹	[Cl] _{aq} /mol L ⁻¹
CIA	12.37	6.23(6)×10 ⁻³	3.25(1)×10 ⁻³	4.49(21)×10 ⁻³
	nd	6.31(7)×10 ⁻³	3.25(2)×10 ⁻³	4.54(21)×10 ⁻³
	nd	6.43(7)×10 ⁻³	3.25(2)×10 ⁻³	4.59(22)×10 ⁻³
CIB	nd	6.24(7)×10 ⁻³	3.26(2)×10 ⁻³	4.64(21)×10 ⁻³
	nd	6.36(8)×10 ⁻³	3.22(2)×10 ⁻³	4.76(23)×10 ⁻³
	nd	6.31(7)×10 ⁻³	3.29(3)×10 ⁻³	4.63(21)×10 ⁻³
	nd	6.35(6)×10 ⁻³	3.29(3)×10 ⁻³	4.64(23)×10 ⁻³
	nd	6.26(7)×10 ⁻³	3.24(1)×10 ⁻³	4.58(22)×10 ⁻³
	nd	6.28(6)×10 ⁻³	3.27(2)×10 ⁻³	4.74(21)×10 ⁻³
	nd	6.28(6)×10 ⁻³	3.27(1)×10 ⁻³	4.79(23)×10 ⁻³
CIC	nd	6.28(6)×10 ⁻³	3.27(1)×10 ⁻³	4.79(23)×10 ⁻³

160 ^a nd: not determined; Uncertainties from three times repeated analyses of identical samples
 161 are given by the number in brackets on the last digit(s).

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175 **Table S2.** Solution content of selected elements for AFm-Cl₂ experiments after equilibrium
 176 with MoO₄²⁻ for 48 h^a.

ID	pH	[Mo] _{aq} /mol L ⁻¹	<i>q</i> /mol g ⁻¹	[Ca] _{aq} /mol L ⁻¹	[Al] _{aq} /mol L ⁻¹	[Cl] _{aq} /mol L ⁻¹
CIA	12.37	1.42(17)×10 ⁻⁷	3.93(42)×10 ⁻⁷	6.30(10)×10 ⁻³	3.07(4)×10 ⁻³	4.55(21)×10 ⁻³
	nd	1.02(12)×10 ⁻⁷	9.57(51)×10 ⁻⁷	6.32(8)×10 ⁻³	3.00(2)×10 ⁻³	4.58(21)×10 ⁻³
	nd	1.89(18)×10 ⁻⁷	1.97(13)×10 ⁻⁶	6.19(7)×10 ⁻³	2.82(2)×10 ⁻³	4.73(22)×10 ⁻³
CIB	12.37	2.14(8)×10 ⁻⁷	2.47(16)×10 ⁻⁶	6.29(8)×10 ⁻³	2.99(3)×10 ⁻³	4.80(22)×10 ⁻³
	nd	3.43(3)×10 ⁻⁷	3.98(21)×10 ⁻⁶	6.25(6)×10 ⁻³	2.88(1)×10 ⁻³	4.90(22)×10 ⁻³
	nd	5.91(7)×10 ⁻⁷	4.98(32)×10 ⁻⁶	6.18(6)×10 ⁻³	2.88(1)×10 ⁻³	4.83(22)×10 ⁻³
	nd	6.74(13)×10 ⁻⁷	5.98(30)×10 ⁻⁶	6.15(8)×10 ⁻³	2.82(2)×10 ⁻³	4.76(22)×10 ⁻³
	nd	5.75(16)×10 ⁻⁷	7.43(38)×10 ⁻⁶	6.28(6)×10 ⁻³	3.06(1)×10 ⁻³	4.76(22)×10 ⁻³
	nd	8.51(16)×10 ⁻⁷	8.93(40)×10 ⁻⁶	6.28(6)×10 ⁻³	3.04(1)×10 ⁻³	5.06(23)×10 ⁻³
CIC	12.37	1.21(1)×10 ⁻⁶	9.92(44)×10 ⁻⁶	6.28(6)×10 ⁻³	3.03(2)×10 ⁻³	5.07(23)×10 ⁻³
CID	12.38	3.98(32)×10 ⁻⁶	1.35(4)×10 ⁻⁴	5.83(15)×10 ⁻³	3.10(7)×10 ⁻³	nd
CIE	12.38	4.05(28)×10 ⁻⁶	2.60(7)×10 ⁻⁴	5.68(19)×10 ⁻³	3.02(5)×10 ⁻³	nd
CIF	12.37	1.59(13)×10 ⁻³	1.52(7)×10 ⁻³	2.27(7)×10 ⁻³	1.44(4)×10 ⁻³	9.35(16)×10 ⁻³
CIG	12.41	1.03(5)×10 ⁻³	3.90(9)×10 ⁻³	8.15(16)×10 ⁻⁴	4.93(12)×10 ⁻³	nd
CIH	12.42	7.50(35)×10 ⁻³	6.14(21)×10 ⁻³	8.97(36)×10 ⁻⁵	7.86(23)×10 ⁻³	nd

177 ^a nd: not determined; Uncertainties from three times repeated analyses of identical samples
 178 are given by the number in brackets on the last digit(s).

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187 **Table S3.** Solution content of selected elements for AFm-SO₄ experiments before introducing
 188 MoO₄²⁻. Concentrations were determined after reaching dissolution equilibrium, which was
 189 confirmed by dissolution kinetics^a.

ID	pH	[Ca] _{aq} /mol L ⁻¹	[Al] _{aq} /mol L ⁻¹	[S] _{aq} /mol L ⁻¹
SA	12.35	2.69(3)×10 ⁻³	1.97(2)×10 ⁻³	5.08(22)×10 ⁻⁵
	nd	2.74(4)×10 ⁻³	1.97(2)×10 ⁻³	5.32(17)×10 ⁻⁵
	nd	2.77(3)×10 ⁻³	1.96(1)×10 ⁻³	5.08(26)×10 ⁻⁵
SB	nd	2.70(3)×10 ⁻³	1.98(1)×10 ⁻³	5.13(14)×10 ⁻⁵
	nd	2.73(3)×10 ⁻³	1.97(2)×10 ⁻³	4.88(29)×10 ⁻⁵
	nd	2.74(3)×10 ⁻³	1.97(1)×10 ⁻³	4.98(24)×10 ⁻⁵
	nd	2.68(2)×10 ⁻³	1.89(1)×10 ⁻³	6.04(16)×10 ⁻⁵
	nd	2.71(3)×10 ⁻³	1.96(1)×10 ⁻³	5.00(5)×10 ⁻⁵
	nd	2.72(3)×10 ⁻³	1.95(2)×10 ⁻³	4.93(24)×10 ⁻⁵
	nd	2.74(3)×10 ⁻³	1.97(2)×10 ⁻³	5.02(44)×10 ⁻⁵

190 ^a nd: not determined; Uncertainties from three times repeated analyses of identical samples
 191 are given by the number in brackets on the last digit(s).

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204 **Table S4.** Solution content of selected elements for AFm-SO₄ experiments after equilibrium
 205 with MoO₄²⁻ for 48 h^a.

ID	pH	[Mo] _{aq} /mol L ⁻¹	<i>q</i> /mol g ⁻¹	[Ca] _{aq} /mol L ⁻¹	[Al] _{aq} /mol L ⁻¹	[S] _{aq} /mol L ⁻¹
SA	12.34	8.75(90)×10 ⁻⁷	2.62(3.04)×10 ⁻⁸	2.77(3)×10 ⁻³	1.95(2)×10 ⁻³	6.00(17)×10 ⁻⁵
	nd	1.90(11)×10 ⁻⁶	5.61(3.67)×10 ⁻⁸	2.80(3)×10 ⁻³	1.97(2)×10 ⁻³	5.11(13)×10 ⁻⁵
	nd	3.96(26)×10 ⁻⁶	8.76(9.20)×10 ⁻⁸	2.65(3)×10 ⁻³	1.85(1)×10 ⁻³	6.09(4)×10 ⁻⁵
SB	12.35	4.88(35)×10 ⁻⁶	1.39(1.18)×10 ⁻⁷	2.74(3)×10 ⁻³	1.92(1)×10 ⁻³	5.21(4)×10 ⁻⁵
	nd	7.86(49)×10 ⁻⁶	2.21(1.59)×10 ⁻⁷	2.78(4)×10 ⁻³	1.94(3)×10 ⁻³	5.19(17)×10 ⁻⁵
	nd	9.70(6)×10 ⁻⁶	4.22(2.25)×10 ⁻⁷	2.68(3)×10 ⁻³	1.86(1)×10 ⁻³	5.95(9)×10 ⁻⁵
	nd	1.23(8)×10 ⁻⁵	1.85(2.20)×10 ⁻⁷	2.77(4)×10 ⁻³	1.88(3)×10 ⁻³	1.29(2)×10 ⁻⁴
	nd	1.43(8)×10 ⁻⁵	5.89(2.84)×10 ⁻⁷	2.79(4)×10 ⁻³	1.98(2)×10 ⁻³	4.27(9)×10 ⁻⁵
	nd	1.73(6)×10 ⁻⁵	7.29(2.58)×10 ⁻⁷	2.77(4)×10 ⁻³	1.96(2)×10 ⁻³	4.44(9)×10 ⁻⁵
SC	12.34	1.94(5)×10 ⁻⁵	8.17(2.51)×10 ⁻⁷	2.77(3)×10 ⁻³	1.95(1)×10 ⁻³	4.45(3)×10 ⁻⁵
SD	12.35	2.07(12)×10 ⁻⁴	3.30(49)×10 ⁻⁵	2.37(6)×10 ⁻³	1.81(4)×10 ⁻³	5.41(86)×10 ⁻⁵
SE	12.36	2.84(17)×10 ⁻⁴	1.20(8)×10 ⁻⁴	2.28(8)×10 ⁻³	1.88(7)×10 ⁻³	5.98(50)×10 ⁻⁵
SF	12.41	2.66(13)×10 ⁻³	9.81(72)×10 ⁻⁴	1.18(3)×10 ⁻³	2.71(8)×10 ⁻³	2.65(22)×10 ⁻⁴
SG	12.40	2.45(15)×10 ⁻³	3.19(10)×10 ⁻³	4.07(9)×10 ⁻⁴	6.10(21)×10 ⁻³	7.56(60)×10 ⁻⁴
SH	12.42	7.63(42)×10 ⁻³	6.07(22)×10 ⁻³	1.02(4)×10 ⁻⁴	8.04(26)×10 ⁻³	2.77(4)×10 ⁻³

206 ^a nd: not determined; Uncertainties from three times repeated analyses of identical samples
 207 are given by the number in brackets on the last digit(s).

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213 **Table S5.** Equilibrium constants of selected phases^a and reactions occurred on CaAl LDHs at
 214 each sorption stage. *logK* values of AFm edge sites sorption and AFm-MoO₄ formation were
 215 adapted.

	Equation	<i>logK</i>
<i>Solubility equilibrium</i>		
AFm-Cl ₂	$\text{Ca}_4\text{Al}_2\text{Cl}_2(\text{OH})_{12} \cdot 4\text{H}_2\text{O} + 12\text{H}^+ \Leftrightarrow 4\text{Ca}^{2+} + 2\text{Al}^{3+} + 2\text{Cl}^- + 16\text{H}_2\text{O}$	74.93
AFm-SO ₄	$\text{Ca}_4\text{Al}_2(\text{SO}_4)(\text{OH})_{12} \cdot 6\text{H}_2\text{O} + 12\text{H}^+ \Leftrightarrow 4\text{Ca}^{2+} + 2\text{Al}^{3+} + \text{SO}_4^{2-} + 18\text{H}_2\text{O}$	73.07
AFm-MoO ₄	$\text{Ca}_4\text{Al}_2(\text{MoO}_4)(\text{OH})_{12} \cdot 4\text{H}_2\text{O} + 12\text{H}^+ \Leftrightarrow 4\text{Ca}^{2+} + 2\text{Al}^{3+} + \text{MoO}_4^{2-} + 16\text{H}_2\text{O}$	74.00
CaMoO ₄	$\text{CaMoO}_{4(s)} \Leftrightarrow \text{Ca}^{2+} + \text{MoO}_4^{2-}$	-7.90
Katoite(C ₃ AH ₆)	$\text{Ca}_3\text{Al}_2(\text{OH})_{12} + 12\text{H}^+ \Leftrightarrow 3\text{Ca}^{2+} + 2\text{Al}^{3+} + 12\text{H}_2\text{O}$	80.32
Ettringite	$\text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12} \cdot 26\text{H}_2\text{O} + 12\text{H}^+ \Leftrightarrow 6\text{Ca}^{2+} + 2\text{Al}^{3+} + 3\text{SO}_4^{2-} + 38\text{H}_2\text{O}$	56.97
<i>Sorption stage</i>		
Edge sorption	$2\text{AFm_edge-OH} + \text{MoO}_4^{2-} \Leftrightarrow (\text{AFm_edge})_2\text{-MoO}_4 + 2\text{OH}^-$	2.80 ^b , 1.05 ^c
AFm-MoO ₄ formation	$4\text{Ca}^{2+} + 2\text{Al}^{3+} + \text{MoO}_4^{2-} + 16\text{H}_2\text{O} \Leftrightarrow \text{Ca}_4\text{Al}_2(\text{MoO}_4)(\text{OH})_{12} \cdot 4\text{H}_2\text{O} + 12\text{H}^+$	-74.00
CaMoO ₄ precipitation	$\text{Ca}^{2+} + \text{MoO}_4^{2-} \Leftrightarrow \text{CaMoO}_{4(s)}$	7.90

216 ^a *logK* values of other phases refer to THERMOCHEMIE database⁴.

217 ^b For edge sorption on AFm-Cl₂.

218 ^c For edge sorption on AFm-SO₄.

219

220 **Table S6.** Physical parameters of AFm particles.

	SA ¹ (m ² g ⁻¹)	AEL ² (μm)	Density (g cm ⁻³)	Basal area (m ²)	LEP ³ (nm g ⁻¹)	LS ⁴ (nm ² g ⁻¹)
AFm-Cl ₂	3.5	0.5	2.11	6.50×10 ⁻¹³	2.23×10 ¹⁷	2.19×10 ¹⁸
AFm-SO ₄	1.9	5.0	2.02	6.50×10 ⁻¹¹	2.26×10 ¹⁶	2.29×10 ¹⁷

221 ¹ SA represents specific surface area measured by N₂-sorption BET.

222 ² AEL represents average edge length of AFm particles measured by SEM.

223 ³ LEP represents lateral edge perimeter.

224 ⁴ LS represents lateral surface.

225 **Table S7.** Edge sites concentration and adsorbed anion amount.

	LEP length conc. (nm L ⁻¹)	LS area conc. (nm ² L ⁻¹)	LEP site conc. (mol L ⁻¹)	LS site conc. (mol L ⁻¹)	Sample	Adsorbed Mo amount (mol L ⁻¹)
					CIA	7.86×10 ⁻⁷
AFm-Cl ₂	4.45×10 ¹⁷	4.38×10 ¹⁸	7.39×10 ⁻⁷	1.94×10 ⁻⁵	CIB	4.94×10 ⁻⁶
					CIC	1.98×10 ⁻⁵
					SA	5.25×10 ⁻⁸
AFm-SO ₄	3.52×10 ¹⁶	3.57×10 ¹⁷	5.85×10 ⁻⁸	1.58×10 ⁻⁶	SB	2.78×10 ⁻⁷
					SC	1.63×10 ⁻⁶

226 * The L/S ratio of 500 kg/kg was used. Layers' edge and lateral adsorption site densities are
 227 estimated as 1.00 nm⁻¹ and 2.67 nm⁻² (2.67 Ca atoms are included per area of 8.700×11.517 Å², seeing
 228 Figure S13), respectively. LS and LEP represent lateral surface and lateral edge perimeter,
 229 respectively.
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