Supporting Information for

Data-driven exploration of layered double hydroxide crystals exhibiting high fluoride ion adsorption properties and chemical stability

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Table S1. Explanatory variables and condition ranges for LDH candidates.

Explanatory variable	Terms
M^{2+} metal speciation	Mg^{2+}, Zn^{2+}, Ni^{2+}, Cu^{2+}, Co^{2+}, Mn^{2+}, Cr^{2+}
M^{3+} metal speciation	Al^{3+}, Fe^{3+}, Cr^{3+}, Y^{3+}, Ga^{3+}
Metal species in LDH	2 or 3 types
Heating temperature [°C]	60–120
Holding time [h]	12–22
$M_{I/II}{}^{3+}$ composition value*	0.20–0.33
F^{-} concentration [mM]	1.0
M^{2+} composition value*	0.67–0.80
Ionic radius r [Å]	0.50–0.90
Pauling electronegativity χ	1.30–1.95
Weighted r average (Å)	0.60–0.85
Weighted χ average	1.36–1.89

*LDH ($[M^{2+}{}_{1-x}M^{3+}{}_{x}(OH)_2](A^{n-})_{x/n}\cdot mH_2O$)

Table S2. Experimental conditions for the first cycle proposed using one objective variable (F^- adsorption amount). Five samples with high probability of improvement (PI) values for the acquisition function (AF) are shown.

Run	Metal speciation			Composition			Synthetic conditions				
	M^{2+}	M_I^{3+}	M_{II}^{3+}	M^{2+}	M_I^{3+}	M_{II}^{3+}	Temp. [°C]	Time [h]	F^- adsorption [$mg\cdot g^{-1}$]	Removal degree [%]	M^{2+} elution [$mg\cdot L^{-1}$]
1	Mn	Cr	Y	0.67	0.10	0.23	60	22	5.76	30.2	2.53
2	Mn	Cr	-	0.67	-	0.33	60	12	0.01	0.0	7.48
3	Mg	Y	-	0.70	-	0.30	100	12	6.40	33.9	0.07
4	Mg	Fe	Y	0.80	0.10	0.10	60	12	4.09	21.5	0.00
5	Ni	Fe	Ga	0.67	0.23	0.10	120	15	18.57	97.7	8.41

Table S3. Chemical composition of Ni-Fe-Ga LDH, as determined using ICP-OES and TG-DTA.

Run	Sample	Nominal composition			Experimental			H_2O	
		Ni	Fe	Ga	Ni	Fe	Ga	wt%	*m* in formula
5	Ni-Fe-Ga-067 LDH	0.67	0.23	0.10	0.70	0.24	0.06	4.9	0.30

Table S4. Experimental conditions for the second cycle proposed using one objective variable (F^- adsorption amount). Five samples with high PI values for the AF are shown.

Run	Metal speciation			Composition			Synthetic conditions				
	M^{2+}	M_I^{3+}	M_{II}^{3+}	M^{2+}	M_I^{3+}	M_{II}^{3+}	Temp. [°C]	Time [h]	F^- adsorption [$mg\cdot g^{-1}$]	Removal degree [%]	M^{2+} elution [$mg\cdot L^{-1}$]
6	Ni	Y	-	0.80	0.20	-	120	22		21.8	3.86
7	Ni	Fe	Cr	0.67	0.10	0.23	60	22		89.2	4.88
8	Mg	Al	Ga	0.67	0.10	0.23	120	22		91.7	7.91
9	Mn	Al	Fe	0.67	0.23	0.10	120	22		3.5	0.01
10	Ni	Al	Ga	0.67	0.10	0.23	120	22	18.42	96.9	2.53

Table S5. Experimental conditions for the first cycle proposed using two objective variables (F^- adsorption and M^{2+} leaching amounts). Five samples with high PI values for the AF are shown.

Run	Metal speciation			Composition			Synthetic conditions				
	M^{2+}	M_I^{3+}	M_{II}^{3+}	M^{2+}	M_I^{3+}	M_{II}^{3+}	Temp. [°C]	Time [h]	F^- adsorption [$mg \cdot g^{-1}$]	Removal degree [%]	M^{2+} elution [$mg \cdot L^{-1}$]
11	Zn	Al	Ga	0.80	0.10	0.10	120	22	3.58	18.8	4.51
12	Zn	Al	Y	0.80	0.10	0.10	120	22	7.01	36.8	0.28
13	Zn	Al	Fe	0.80	0.10	0.10	120	22	0.57	2.9	2.16
14	Zn	Al	Cr	0.80	0.10	0.10	120	22	11.57	60.7	7.23
15	Zn	Al	Ga	0.67	0.23	0.10	60	22	18.52	97.2	5.87

Table S6. Experimental conditions for the second cycle proposed using two objective variables (F^- adsorption and M^{2+} leaching amounts). Two samples with high PI values for the AF are shown.

Run	Metal speciation			Composition			Synthetic conditions				
	M^{2+}	M_I^{3+}	M_{II}^{3+}	M^{2+}	M_I^{3+}	M_{II}^{3+}	Temp. [°C]	Time [h]	F^- adsorption [$mg \cdot g^{-1}$]	Removal degree [%]	M^{2+} elution [$mg \cdot L^{-1}$]
16	Ni	Fe	Y	0.67	0.23	0.10	60	22	15.78	82.4	3.40
17	Ni	Cr	Y	0.67	0.23	0.10	60	21	17.09	89.2	1.48

Table S7. Competitive fluoride ion adsorption test results for Ni-Al-Ga-067, Ni-Cr-Y-067, and Mg-Al-033 LDH samples.

Sample	F^- adsorption [$mg \cdot g^{-1}$]	Removal degree [%]	K_d [ml g^{-1}]
Ni-Al-Ga-067 LDH	3.12	82.0	4564
Ni-Cr-Y-067 LDH	3.16	83.2	4951
Mg-Al-033 LDH	2.70	71.0	2448

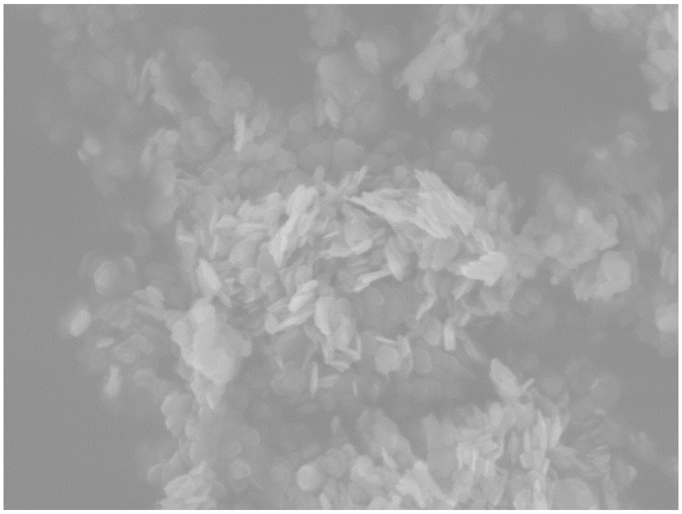

Figure S1. FE-SEM image of Ni-Fe-Ga-067 LDH.

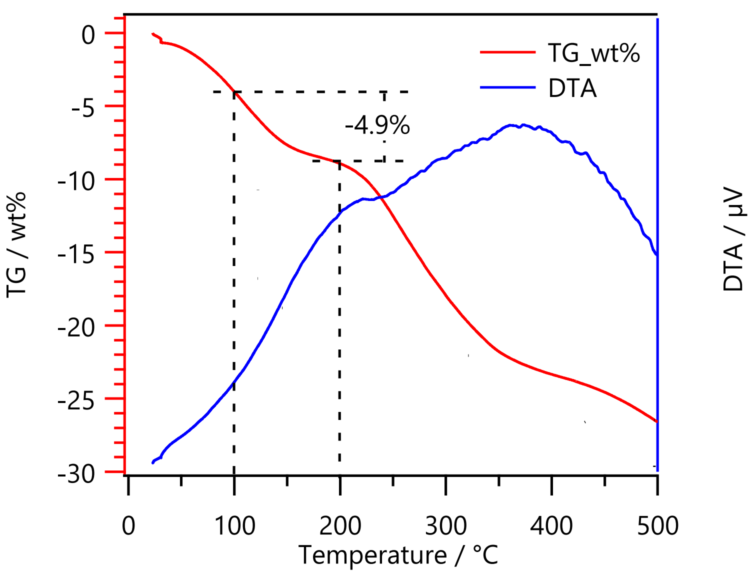

Figure S2. TG-DTA profile of Ni-Fe-Ga-067 LDH.

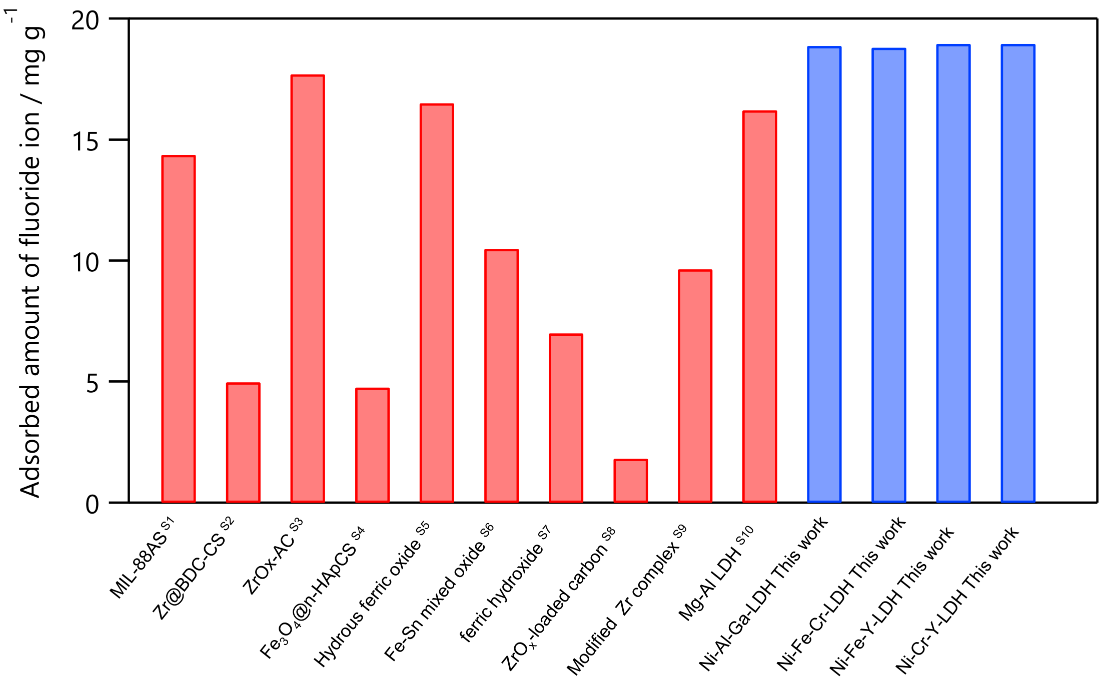

Figure S3. Comparison of F^- adsorption amounts for the LDHs prepared in this work and previously reported adsorbents.[S1–S10]

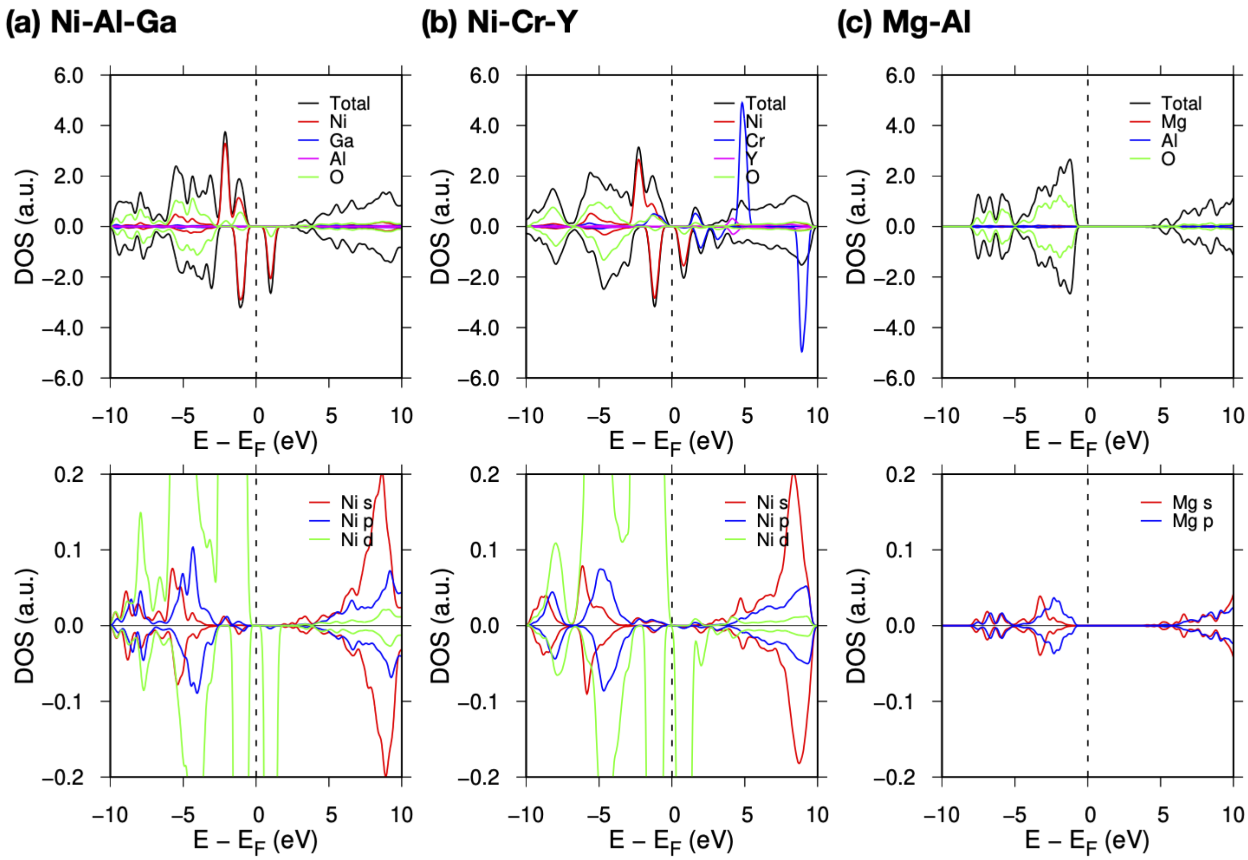

Figure S4. Projected density of states of (a) Ni-Al-Ga-, (b) Ni-Cr-Y, and (c) Mg-Al-LDHs.

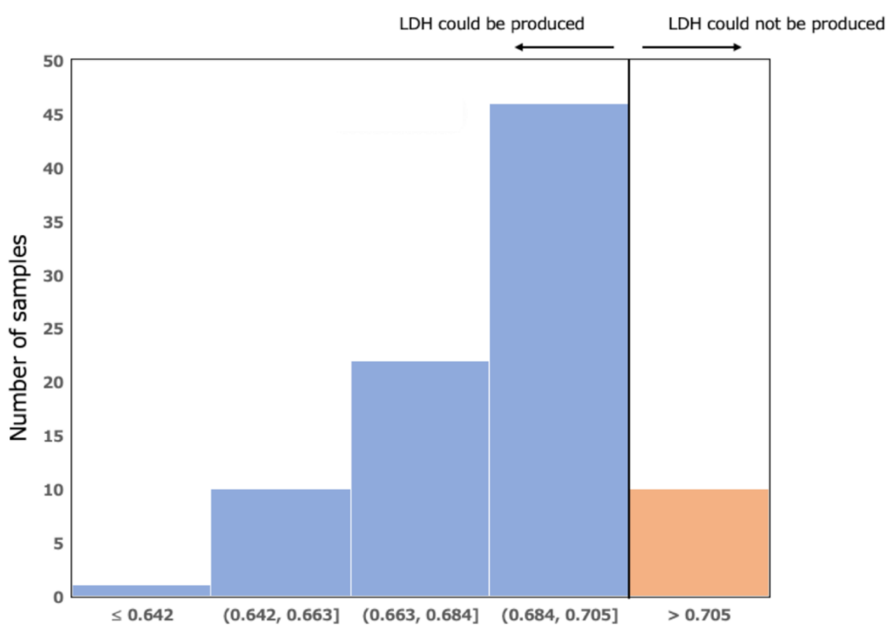

Figure S5. Histogram of weighted-average ionic radius values of metal cations for the formation of LDH structures.

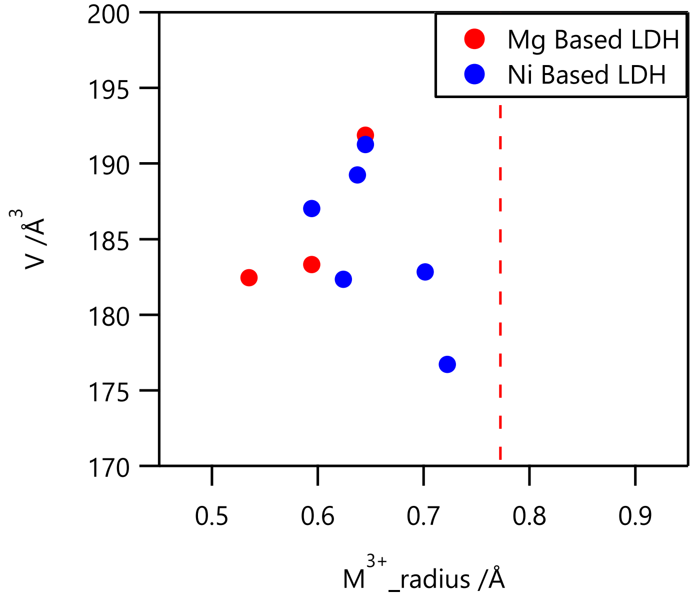

Figure S6. Effect of M^{3+} ionic radius in Mg- and Ni-based LDHs on the formation of LDHs and unit cell volume of Mg- and Ni-based LDHs.

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