Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Electronic Supplementary Information

Theoretical and experimental investigation of electronic/ionic conductivity and deprotonation of $Ni_{3-x}Co_xAl$-LDHs in an electrochemical energy storage system

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Table S1 Fractional coordinates of atoms in Ni_3Al-LDHs

Element	Atom number	Fractional coordinates of atoms		
		x	y	z
H	1	0.675206	0.845668	0.295619
H	2	0.192077	0.877429	0.287087
H	3	0.279456	0.445391	0.286755
H	4	0.721652	0.590583	-0.2902
H	5	0.778083	0.05949	-0.28224
H	6	0.293591	0.09603	-0.28706
H	7	0.669446	0.443784	0.2864
H	8	0.36637	0.590109	-0.29071
O	1	0.671829	0.841651	0.155761
O	2	0.184224	0.866264	0.146838
O	3	0.217561	0.387975	0.144883
O	4	0.778834	0.611799	-0.14974
O	5	0.808999	0.120254	-0.14095
O	6	0.320124	0.142701	-0.14521
O	7	0.672049	0.387991	0.144583
O	8	0.331678	0.611029	-0.15039
Al	1	0.498737	0.497178	-0.00326
Ni	1	-0.00342	-0.00315	0.007925
Ni	2	0.499913	-0.00379	0.006978
Ni	3	-0.00169	0.496463	-0.00971

Table S2 Fractional coordinates of atoms in Ni_2CoAl-LDHs

Element	Atom number	Fractional coordinates of atoms		
		x	y	z
H	1	0.670641	0.846666	0.29708
H	2	0.189308	0.867284	0.288244
H	3	0.275129	0.446859	0.287935
H	4	0.722627	0.585445	-0.29122
H	5	0.776745	0.062083	-0.28483
H	6	0.307606	0.103316	-0.28774
H	7	0.665929	0.446691	0.28752
H	8	0.369928	0.588609	-0.28956
O	1	0.66862	0.841465	0.156996
O	2	0.185071	0.865631	0.149364
O	3	0.21688	0.389216	0.146036
O	4	0.778532	0.609373	-0.15053
O	5	0.807955	0.121334	-0.14321
O	6	0.324232	0.144911	-0.14557
O	7	0.670909	0.389769	0.145236
O	8	0.333756	0.610863	-0.14888
Al	1	0.499087	0.4975	-0.00248
Co	1	-0.0041	-0.00361	0.007594
Ni	1	0.500062	-0.00287	0.007707
Ni	2	-0.00174	0.496631	-0.01025

Table S3 Fractional coordinates of atoms in $NiCo_2Al$-LDHs

Element	Atom number	Fractional coordinates of atoms		
		x	y	z
H	1	0.698585	0.870315	0.297987
H	2	0.201782	0.885503	0.290882
H	3	0.2948	0.451519	0.289094
H	4	0.708639	0.580978	-0.28888
H	5	0.766189	0.044603	-0.28714
H	6	0.27463	0.091895	-0.28896
H	7	0.669962	0.447669	0.290321
H	8	0.358586	0.577895	-0.29248
O	1	0.684572	0.852872	0.157014
O	2	0.193131	0.878617	0.151735
O	3	0.227923	0.398667	0.145592
O	4	0.771993	0.606964	-0.14748
O	5	0.804704	0.114443	-0.14553
O	6	0.311468	0.138932	-0.1466
O	7	0.675059	0.396354	0.148657
O	8	0.330322	0.605306	-0.15249
Al	1	0.499632	0.49916	-0.00174
Co	1	0.501525	-0.00147	0.006353
Co	2	-0.00054	0.498707	-0.0102
Ni	1	-0.00069	0.000871	0.008114

Table S4 Fractional coordinates of atoms in Co_3Al-LDHs

Element	Atom number	Fractional coordinates of atoms		
		x	y	z
H	1	0.692502	0.863414	0.30054
H	2	0.202461	0.879497	0.292253
H	3	0.288812	0.450292	0.291506
H	4	0.710799	0.580537	-0.29115
H	5	0.766196	0.052331	-0.29041
H	6	0.289409	0.099615	-0.29088
H	7	0.669047	0.449513	0.289583
H	8	0.362256	0.578284	-0.291
H	9	0.415089	0.33588	0.474964
O	1	0.679063	0.848014	0.159316
O	2	0.192588	0.874061	0.152861
O	3	0.224368	0.39525	0.148418
O	4	0.772977	0.607186	-0.15041
O	5	0.803652	0.116088	-0.14794
O	6	0.317129	0.143282	-0.14803
O	7	0.673407	0.395708	0.146573
O	8	0.332157	0.605841	-0.14939
O	9	0.50666	0.521568	0.506084
Al	1	0.499677	0.499255	-0.0009
Co	1	-0.00191	-0.00089	0.007376
Co	2	0.501034	-0.00109	0.006787
Co	3	-0.00051	0.497362	-0.01063

Table S5 The crystal lattice parameters of $Ni_{3-x}Co_xAl$-LDHs using DFT and DFT-D

Sample	DFT			DFT-D		
	a (Å)	b (Å)	c (Å)	a (Å)	b (Å)	c (Å)
Ni_3Al	6.280781	6.280172	7.715130	6.226201	6.22475	7.550005
Ni_2CoAl	6.296824	6.288829	7.725056	6.237358	6.229169	7.563131
$NiCo_2Al$	6.341947	6.341126	7.727814	6.281399	6.279915	7.567417
Co_3Al	6.372119	6.321036	7.723622	6.261195	6.306874	7.562502

Table S6 Calculated effective mass of $Ni_{3-x}Co_xAl$-LDHs for three times

sample	Effective mass (m_0)			
	1	2	3	average
Ni_3Al	0.637	0.651	0.654	0.641
Ni_2CoAl	0.620	0.628	0.623	0.624
$NiCo_2Al$	0.620	0.621	0.623	0.621
Co_3Al	0.614	0.618	0.618	0.617

Table S7 The $d_{O\cdots H}$ values of $Ni_{3-x}Co_xAl$-LDHs after geometry optimization for three times

Sample	Times			
	1	2	3	average
Ni_3Al	1.780 Å	1.777 Å	1.775 Å	1.777 Å
Ni_2CoAl	1.771 Å	1.774 Å	1.771 Å	1.772 Å
$NiCo_2Al$	1.741 Å	1.736 Å	1.735 Å	1.737 Å
Co_3Al	1.739 Å	1.737 Å	1.732 Å	1.736 Å

Table S8 Mulliken charge population analysis of H atoms of LDHs layer and O atoms of interlayer OH^- ions

Sample	Mulliken charge population analysis (e/atom)	
	H	O
Ni_3Al	0.36	-0.88
Ni_2CoAl	0.39	-0.92
$NiCo_2Al$	0.44	-1.01
Co_3Al	0.38	-0.90

Table S9 The values of R_s in $Ni_{3-x}Co_xAl$-LDHs (x=0, 1, 2, 3)

Sample	R_s (Ω)
Ni_3Al	1.445
Ni_2CoAl	1.496
$NiCo_2Al$	1.355
Co_3Al	1.341

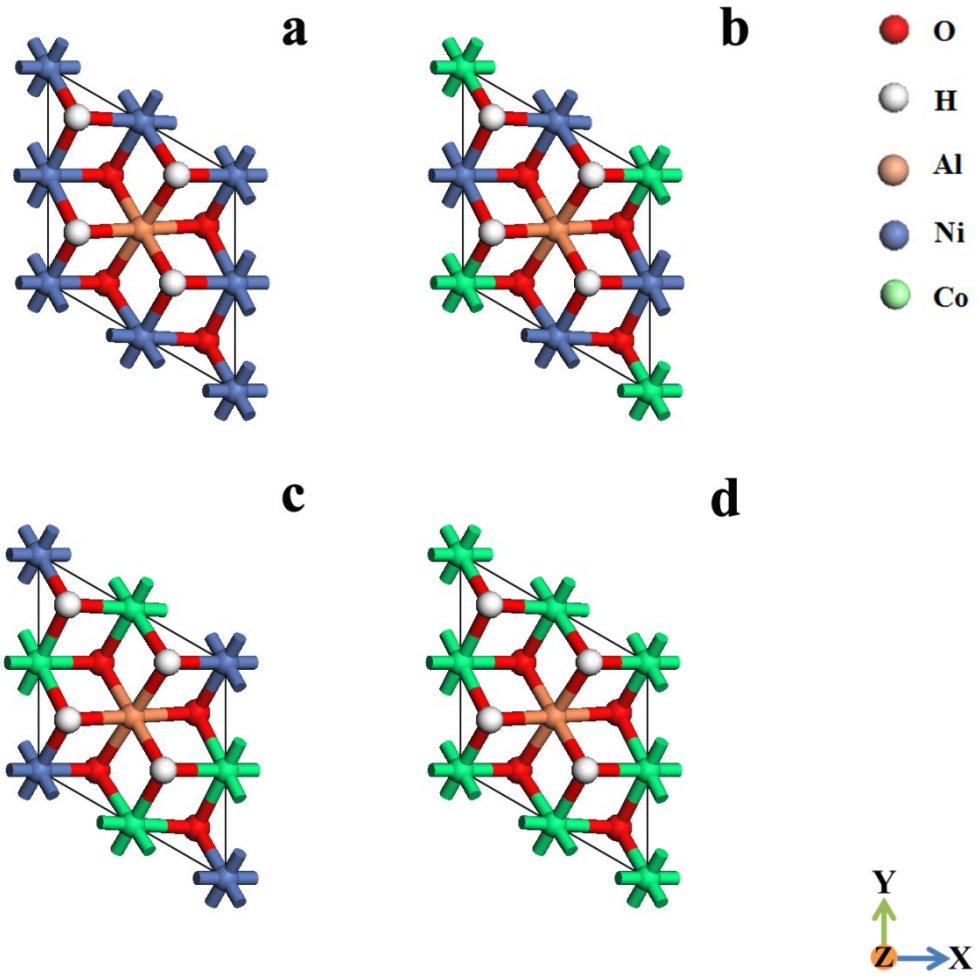

Fig. S1 Top-view schematics of $Ni_{3-x}Co_xAl$-LDHs models. (a) x=0; (b) x=1; (c) x=2; (d) x=3, respectively.

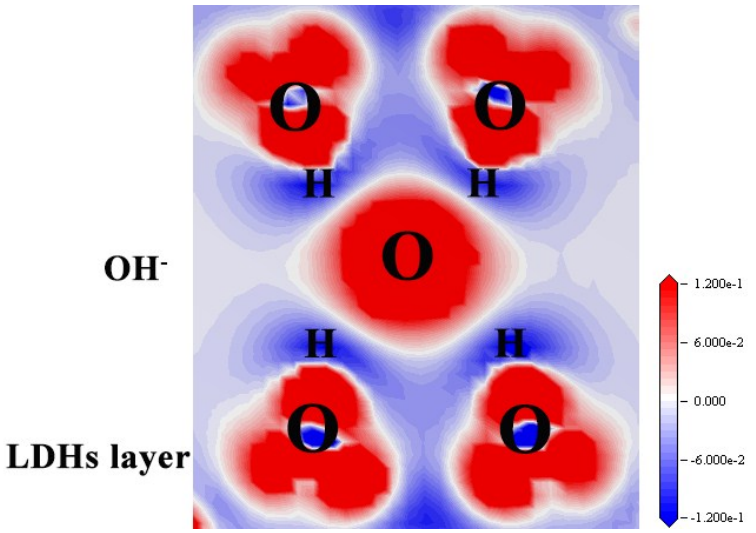

Fig. S2 Electron density difference of interlayer OH^- ions and -OH in LDHs layer