Supporting Information

Delicate control of crystallographic Cu_2O derived Ni-Co amorphous double hydroxides nanocages for high-performance hybrid supercapacitors: An experimental and computational investigation

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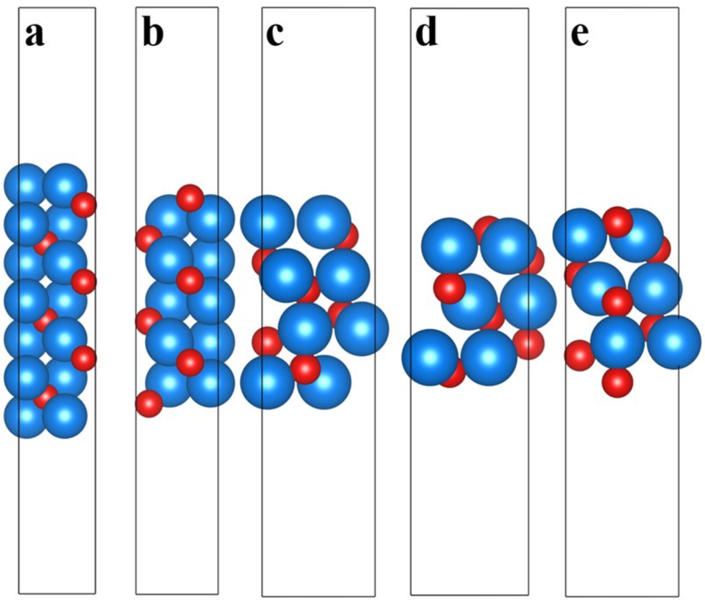


Figure S1. Atomic structures of Cu_2O surface: (100)-Cu (a), (100)-O (b), (111)-Cu (c), (111)-O (d), (111)-OO (e). Cu, blue; O, red.

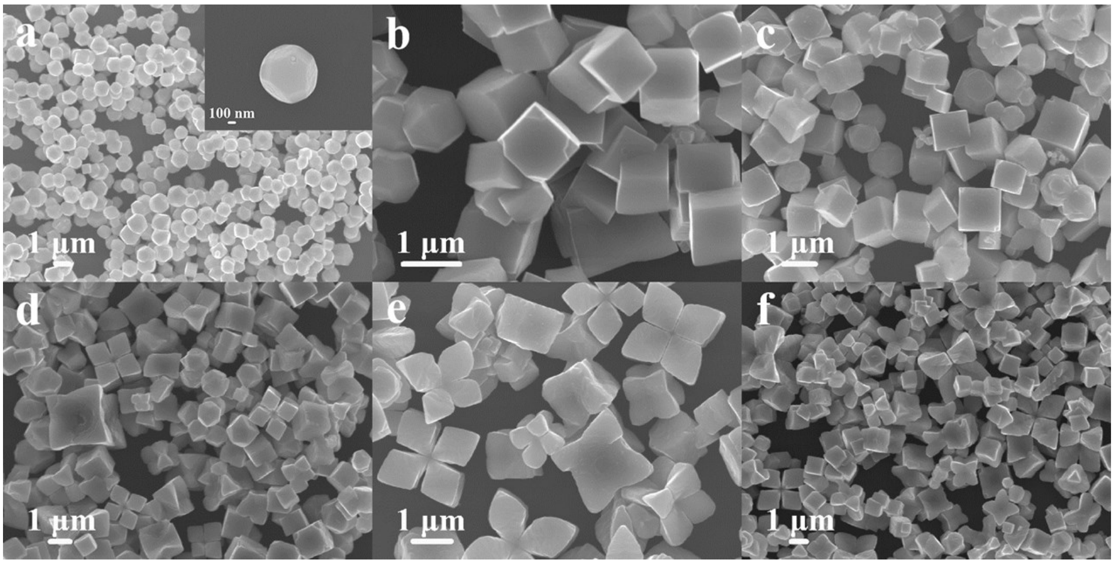


Figure S2. SEM images of the Cu_2O crystals with different aging time synthesized at 80℃: 1 h (a), 1.5 h (b), 2 h (c), 2.5 h (d), 3 h (e) and 3.5 h (f). The inset of (a) shows the quasi-spherical Cu_2O crystals at high magnification.

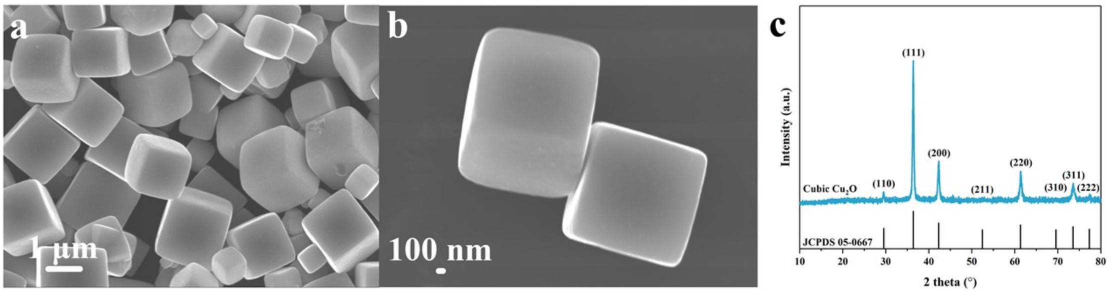


Figure S3. SEM images (a, b) and XRD patterns (c) of the Cu_2O crystals synthesized at room temperature.

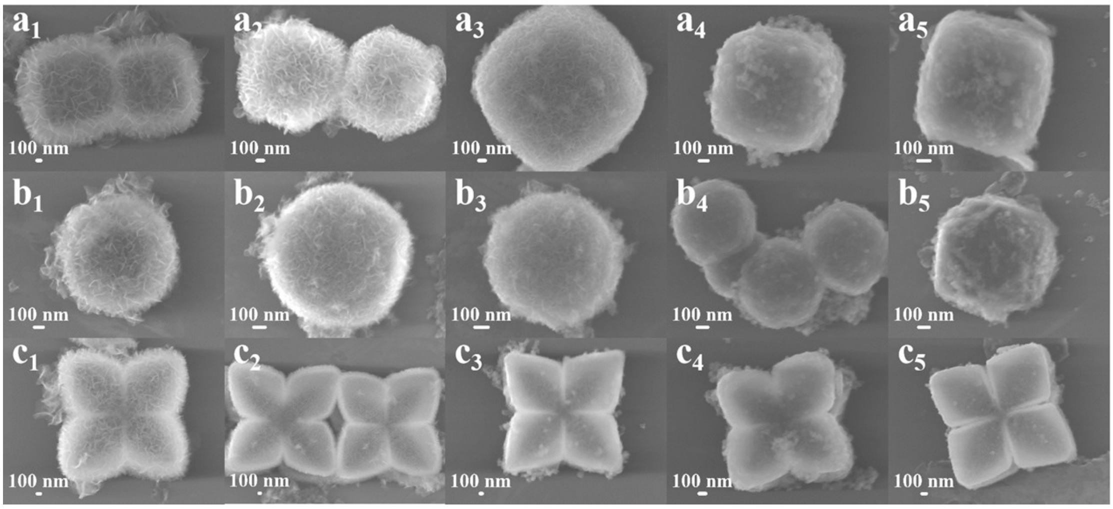


Figure S4. SEM images of $Ni(OH)_2$, Ni_7-Co_3 ADHs, Ni_1-Co_1 ADHs, Ni_3-Co_7 ADHs, $Co(OH)_2$ with different Ni/Co ratio: 1-5 represent 0:10, 3:7, 1:1, 7:3 and 10:0 (Ni:Co) respectively; a-c represent cube, sphere and flower structure respectively.

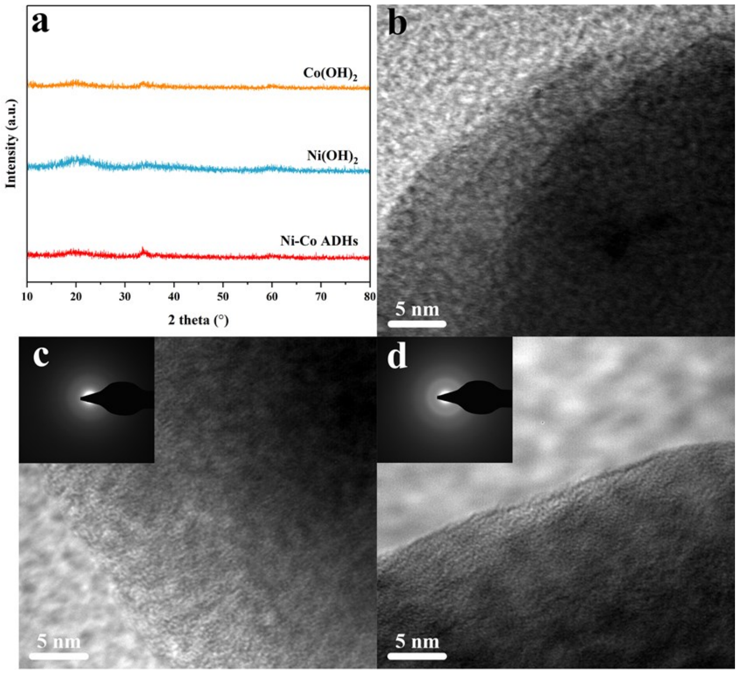


Figure S5. XRD patterns of Ni-Co ADHs, $Ni(OH)_2$ and $Co(OH)_2$ nanocages (a). HRTEM image of Ni-Co ADHs nanocages (b). HRTEM and SAED images of $Ni(OH)_2$ nanocages (c). HRTEM and SAED images of $Co(OH)_2$ nanocages (d).

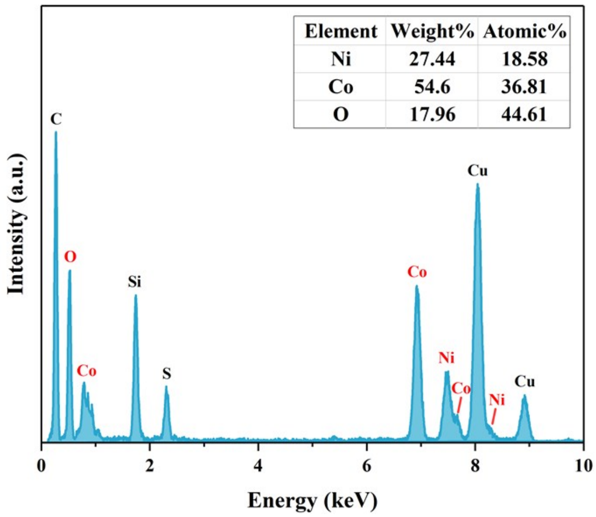


Figure S6. EDS measurements of the typical Ni–Co ADHs nanocages.

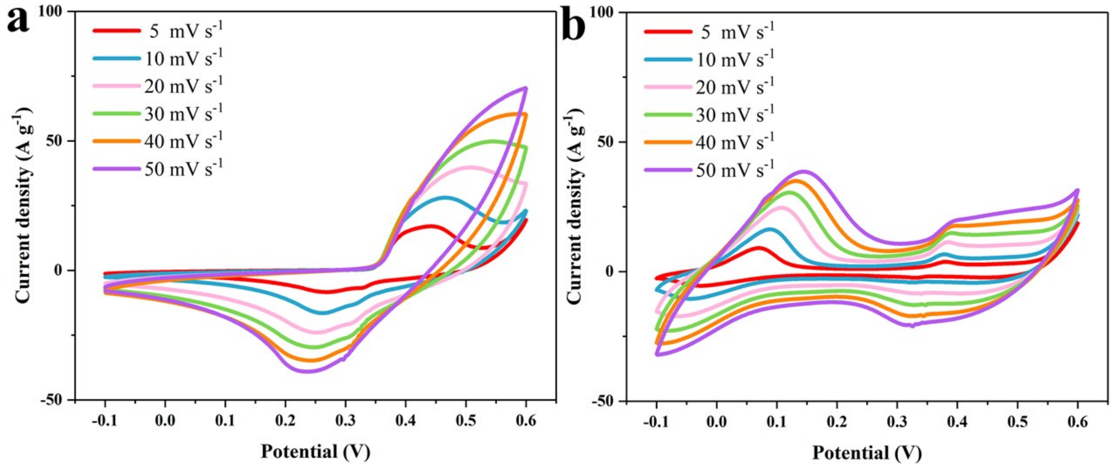


Figure S7. CV curves of $Ni(OH)_2$ (a) and $Co(OH)_2$ (b) nanocages at different scan rates.

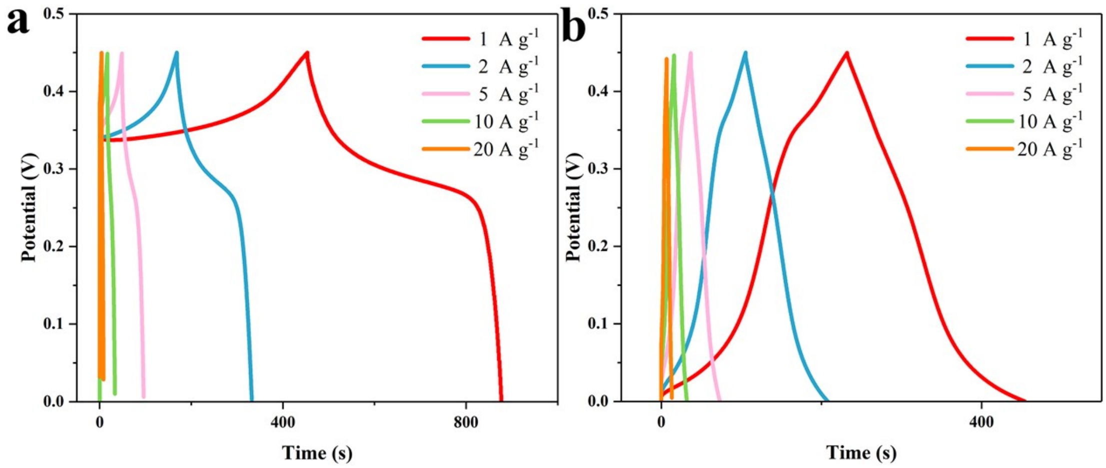


Figure S8. GCD curves of $Ni(OH)_2$ (a) and $Co(OH)_2$ (b) nanocages at different current densities.

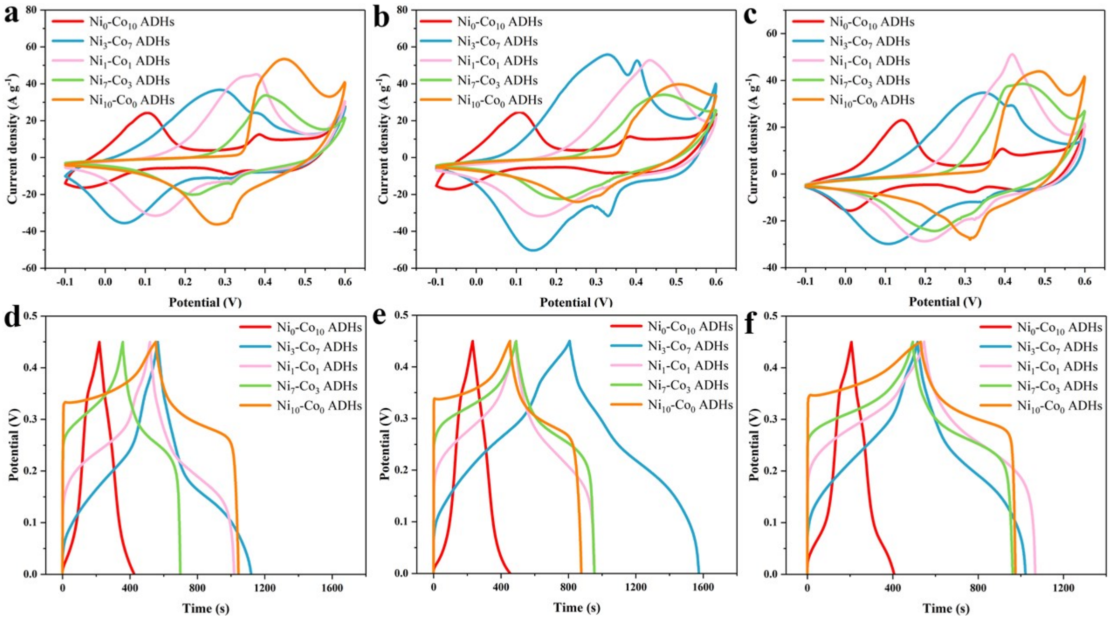


Figure S9. CV (a-c) and GCD (d-f) curves of the amorphous hydroxide electrodes with different morphologies and Ni/Co ratios. Cube: a, d; Sphere: b, e; Flower: c, f.

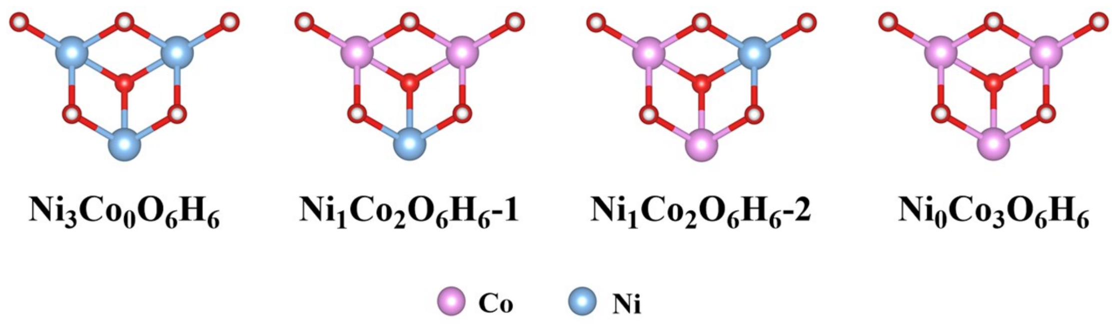


Figure S10. Basic model of hydroxides clusters. $Ni(OH)_2$, Ni-Co ADHs and $Co(OH)_2$ are represented by $Ni_3Co_0O_6H_6$, $Ni_1Co_2O_6H_6$ and $Ni_0Co_3O_6H_6$ respectively. Clusters with unsaturated Ni and Co sites are recorded as $Ni_1Co_2O_6H_6$-1 and $Ni_1Co_2O_6H_6$-2, respectively.

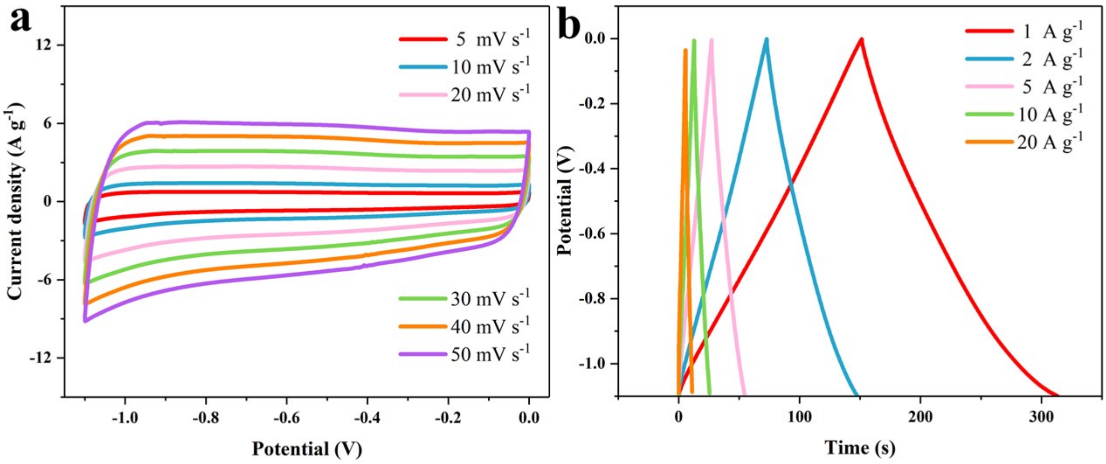


Figure S11. CV curves of AC at different scan rates (a) and GCD curves of AC at different current densities (b).

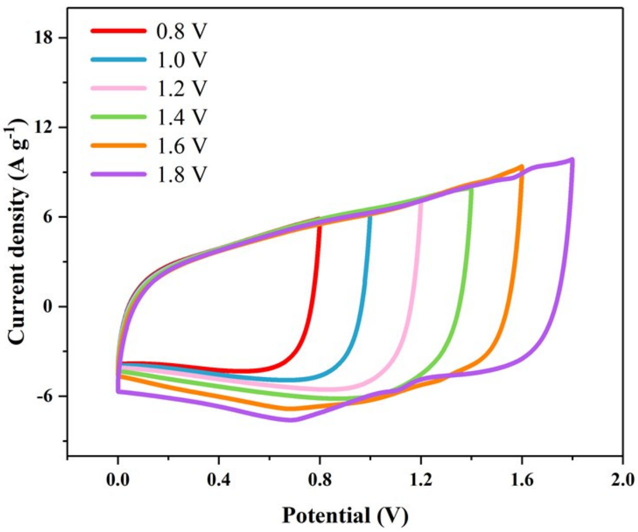


Figure S12. CV curves of Ni-Co ADHs//AC HSC with different potential windows tested at a scan rate of 80 mV s^{-1}.

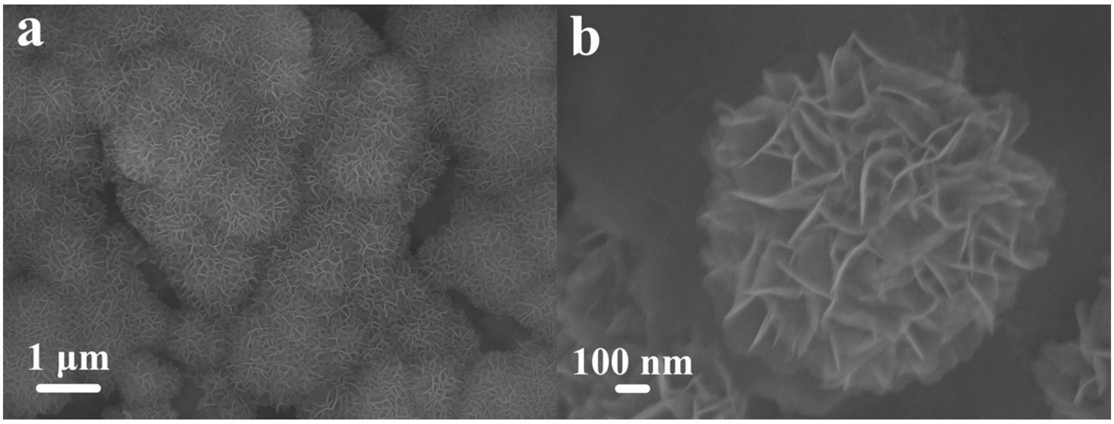
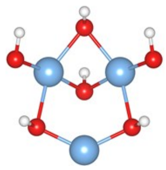
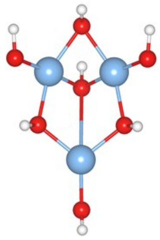
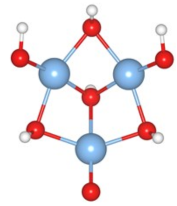
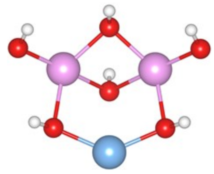
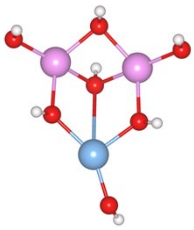
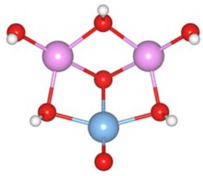
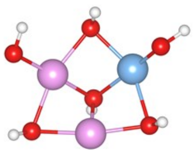
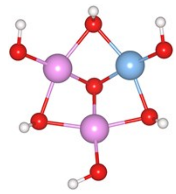
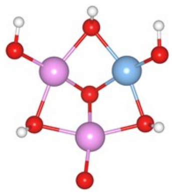
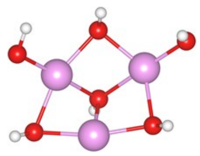
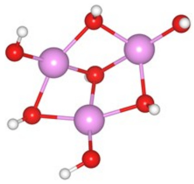
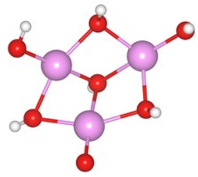


Figure S13. SEM images of Ni-Co ADHs after cycles.

Table S1. The specific capacitance (at 1 ~ 20 A g^{-1}) of the amorphous hydroxide prepared with different morphologies and Ni/Co ratio.

Morphologies	Ni/Co ratio	Specific capacitance (F g^{-1})				
		Current density (A g^{-1})				
		1	2	5	10	20
Cube	$Ni(OH)_2$	1091	907	741	578	436
	Ni_7-Co_3 ADHs	760	702	589	489	373
	Ni_1-Co_1 ADHs	1120	1080	1000	918	804
	Ni_3-Co_7 ADHs	1240	1218	1133	1058	933
	$Co(OH)_2$	462	440	378	336	280
heterogeneous cube (2 h)	Ni_3-Co_7 ADHs	1002	969	900	833	729
Sphere	$Ni(OH)_2$	944	729	522	369	187
	Ni_7-Co_3 ADHs	1040	924	767	598	436
	Ni_1-Co_1 ADHs	1067	1048	940	784	640
	Ni_3-Co_7 ADHs	1707	1636	1523	1411	1280
	$Co(OH)_2$	496	458	400	356	289
Flower	$Ni(OH)_2$	991	751	518	378	231
	Ni_7-Co_3 ADHs	1051	964	789	638	480
	Ni_1-Co_1 ADHs	1176	1138	1022	891	737
	Ni_3-Co_7 ADHs	1129	1098	1022	933	804
	$Co(OH)_2$	449	418	378	349	276

Table S2. Optimal hydroxides models (HM), adsorption models (AM) and relevant energies simulated by DFT. Ni, blue; Co, pink; O, red; H, white.

HM1	AM1-OH	AM1-O	HM2-1
			
$E_{cluster}$ = -67.51 eV	E_{Total} = -78.17 eV	E_{Total} = -72.70 eV	$E_{cluster}$ = -70.05 eV
AM2-1-OH	**AM2-1-O**	**HM2-2**	**AM2-2-OH**
			
E_{Total} = -81.17 eV	E_{Total} = -75.59 eV	$E_{cluster}$ = -71.03 eV	E_{Total} = -82.00 eV
AM2-2-O	**HM3**	**AM3-OH**	**AM3-O**
			
E_{Total} = -76.80 eV	$E_{cluster}$ = -71.98 eV	E_{Total} = -83.73 eV	E_{Total} = -78.11 eV