Supporting Information

Carbon quantum dot regulated the electrochemical activation of Co_{0.03}Ni_{0.97}LDH for energy storage

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Text S1. Detailed deduction of b value and capacitive distribution

The charge storage mechanism of the A-CQD/Co_{0.03}Ni_{0.97}LDH electrode can be semi-quantitatively resolved according to the redox peak potential in the current response of CV curves at different scan rates. Normally, the peak current *i* and the scanning speed *v* from the CV curves of the tested electrodes can be expressed according to the following Equation (5):

$$i = av^b \tag{5}$$

wherein the value of the kinetic parameter b lies between 0.5 to 1. When b equals to 0.5, it suggests that the electrode reaction is completely diffusion-controlled process, and when b is equivalent to 1, it means that the electrode reaction is surface-reaction controlled process. The logarithm of the current values of the cathode or anode peaks is plotted against the logarithm of the scanning rate, and the b value is then acquired from the slope of the fitted line. The kinetics hysteresis of the sample electrodes can be estimated by the b values. Correspondingly, the response current can also be divided into the following Equation (6):

$$i = k_1 v + k_2 v^{1/2} \tag{6}$$

where k_1v represents for the capacitive controlled current and $k_2 v^{1/2}$ for the diffusion controlled current. The above Equation (6) could be transformed into Equation (7):

$$\frac{i}{v^{1/2}} = k_1 v^{1/2} + k_2 \tag{7}$$

The coefficients k_1 and k_2 can be obtained by linear fitting of a series of equations at various potential points. Then, the contribution proportions of above two kinds of current can be obtained on account of $k_1 v$ or $k_2 v^{1/2}$.

Text S2. Specific theoretical calculation process

DFT-based first-principles calculations were performed using the CASTEP package of Materials Studio based on the generalized gradient approximation (GGA) in the form of Perdew–Burke– Ernzerh (PBE). An energy cutoff of 400 eV was used, and a k-point sampling set of $2 \times 2 \times 1$ was tested for convergence. The convergence criterion for the maximum energy change was 1.0×10^{-5} eV/atom during the geometry optimization. After geometry optimization, the density of states (DOS) was computed with the most stable configurations.





Figure S1 displays the CV curves of 200 cycles activation at 50 mV s⁻¹ for Co/CQD and CQD/Ni(OH)₂. From Figure S1a, three pairs of redox peaks can be observed. The pair of redox peaks between 0.05 and 0.18 V are attributed to the reversible conversion of Co^{2+}/Co^{3+} derived from adsorbed Co ions, and the pair of redox peaks between 0.2 and 0.4 V are assigned to and the sulfur and oxygen functional groups on CQD[1]. The redox peak at 0.4 to 0.5 V can be attributed to Co^{3+}/Co^{4+} . However, the area of CV curve did not increase with the CV cycle activation at 50 mV s⁻¹, indicating that the electrochemical active of Co/CQD is stable. Figure S1b-c presents the CV curves of 200 cycles activation at 50 mV s⁻¹ for Ni/CQD and CQD/Ni(OH)₂. The absence of the pair of redox peaks between 0.05 and 0.18 V further suggests that they are originated from the conversion of Co^{2+}/Co^{3+} . The area of CV curve obviously increased at the 10th cycle and remained stagnant with the continual increase of the CV activation to the 200th cycle.



Figure S2. CV curves for 200 cycles of activation at 50 mV s⁻¹ for (a) $Co_{0.2}Ni_{0.8}LDH$, (b) $Co_{0.3}Ni_{0.7}LDH$, (c) $Co_{0.4}Ni_{0.6}LDH$, (d) $Co_{0.5}Ni_{0.5}LDH$, (e) $Co_{0.7}Ni_{0.3}LDH$ and (f) $Co(OH)_2$.

Figure S2 presents the CV curves for 200 cycles of activation at 50 mV s⁻¹ for Co_{0.2}Ni_{0.8}LDH, Co_{0.3}Ni_{0.7}LDH, Co_{0.4}Ni_{0.6}LDH, Co_{0.5}Ni_{0.5}LDH, Co_{0.7}Ni_{0.3}LDH and Co(OH)₂. We can observe that an irreversible oxide peak appears only in the 1st CV curve of the bimetallic CoNiLDH. In addition, the increased area of CV curve after the CV activation at 50 mV s⁻¹ increases with the increase of Co content in the bimetallic CoNiLDH until Co/Ni equal to 1. In addition, the oxide peak presented in Co(OH)₂ is reversible, but the peak current after the CV activation at 50 mV s⁻¹ is obviously lower than the bimetallic CoNiLDH, suggesting that the high energy storage density of the bimetallic CoNiLDH is mainly attributed to the Ni element in LDH[2].



Figure S3. (a) CV curves of CoNiLDH and Co(OH)₂, (b) nonlinear fitting and (c) linear fitting curves for peak current versus Co content.

Figure S3 displays the part of the 1st CV curves and the correlations between the peak current and the Ni to Co ratio. The peak current increased with the increase of Co amount until Co/Ni equal to 1 (Figure S3a), which is consistent with the previous report that the optimal electrochemical performance is achieved at the Co/Ni ratio of 1 (Figure S3b) [3]. Then, the current decreased with the continual increase of Co amount, which is attributed to the decrease from Co^{2+} to Co^{3+} caused by the reduced Ni content. The high positive correlation of R^2 =0.947 in Figure S3c indicates that the peak current at about 0.12 V would be used to estimate the conversion from Co^{2+} to Co^{3+} in Co/Ni bimetallic LDH when the Co/Ni ratio is less than 1.



Figure S4. (a, c, e, g, i) CV curves at different scan rates and (b, d, f, h, j) GCD curves at different current densities of CoNiLDH electrodes after 200 cycles of activation at 50 mV s⁻¹.

Figure S4 demonstrates the electrochemical properties of the as-prepared CoNiLDH electrodes with different Ni/Co ratios, which indicates that the $Co_{0.5}Ni_{0.5}LDH$ electrode shows the best excellent electrochemical properties in all electrodes.



Figure S5. (a) XRD pattern of Co/CQD, (b) FTIR spectra of S/CQD and Co/CQD.

XRD pattern of Co/CQD is shown in Figure S5a, in which the broad peaks of carbon in CQD can be observed. FTIR spectra (Figure S5b) show that the S-H stretching signal at 2061 cm⁻¹ disappear after the adsorption of Co ions[4]. The IR spectrum of Co/CQD is slightly red-shifted due to the introduction of Co. Furthermore, the presence of C-O-C (1097 cm⁻¹) and C-O (1023 cm⁻¹) [5] functional groups on Co/CQD. In addition, the Co-S signal at 1102 cm⁻¹ and the Co-O signal at 500 cm⁻¹ can be observed in Co/CQD [6, 7], confirming the strong interactions between Co ions and the sulfur and oxygen functional groups on CQD.



Figure S6 (a-c) N₂ adsorption/desorption isotherms and (d-f) pore size distribution curves of Ni(OH)₂, Co_{0.03}Ni_{0.97}LDH and CQD/Co_{0.03}Ni_{0.97}LDH before and after activation measured at 77K.

The specific surface area and pore volume, calculated by the Brunauer-Emmett-Teller (BET) and (Barret-Joyner-Halenda) methods (Figure S6). No significant change in the specific surface area of each material after electrochemical activation. Moreover, the calculated S_{BET} of CQD/Co_{0.03}Ni_{0.97}LDH is maximised before and after electrochemical activation. Besides, the pore size distribution curves in Figure S6d-f confirm that CQD did not change the pore size distribution significantly. The higher S_{BET} and pore volume means the higher active sites and more convenient channel of A- CQD/Co_{0.03}Ni_{0.97}LDH for energy storage.



Figure S7. (a-b) TEM and (c-e) HRTEM images of Co/CQD; (f) HAADF-STEM and EDS mapping of Co/CQD.

As shown in Figure S7, compared with the well dispersed bare CQDs reported in our previous work[8], TEM images (Figure S7a and S7b) of Co/CQD indicate that the Co ions-modification CQDs get together because of the change of surface charge of CQDs after Co ions adsorption. From HRTEM presented in Figure S7c, we can observe many clear lattice stripes. The amplified HRTEM shown in Figure S7d and S7e indicate that the interplanar spacing is 0.26 nm, matching well with the (100) plane of CQD[8]. HAADF-STEM image (Figure S7f) indicates that the texture of the Co/CQD is homogeneous and no metal Co or cobalt compounds are formed. The corresponding EDS mapping display that C, O, Co, and S elements are uniform distributed in the as-prepared Co/CQD.



Figure S8. (a) TEM and (b-d) HRTEM images of CQD/Co_{0.03}Ni_{0.97}LDH; (e) HAADF-STEM and EDS mapping corresponding to CQD/Co_{0.03}Ni_{0.97}LDH.

Figure S8 shows the TEM images of CQD/Co_{0.03}Ni_{0.97}LDH. As shown in Figure S8a, the nanoflower structure is formed by the self-assembly of thinner nanosheets. HRTEM image shown in Figure S8c-d taken from the labeled area in Figure S8b presents lattice fringes with the spacings of 0.24 nm, which can be assigned to the (111) plane of Ni(OH)₂·0.75H₂O (JCPDS#38–0715) and the (100) plane of CQD, respectively. Confirming the presence of CQD on the surface of Co_{0.03}Ni_{0.97}LDH nanosheets. In addition, the EDS mapping (Figure S8e) of the CQD/Co_{0.03}Ni_{0.97}LDH reveal that the Ni, Co, C, O and S are uniformly distributed in CQD/Co_{0.03}Ni_{0.97}LDH nanosheets, indicates that the CQD is uniformly dispersed on the surface of Co_{0.03}Ni_{0.97}LDH nanosheets.



Figure S9. (a) TEM and (b-d) HRTEM images of A-CQD/Co_{0.03}Ni_{0.97}LDH; (e) HAADF-STEM and EDS mapping corresponding to A-CQD/Co_{0.03}Ni_{0.97}LDH.

Figure S9 shows the TEM images of A-CQD/Co_{0.03}Ni_{0.97}LDH. After electrochemical activation, nanosheets become more curved and thinner (Figure S9a). Moreover, it is still observed that the (100) plane of CQD and the (111) plane of Ni (OH)₂·0.75H₂O between the tiny nanosheets (Figure S9b-d), confirmed that CQD can be stably present on the surface of Co_{0.03}Ni_{0.97}LDH nanosheets during electrochemical activation. Furthermore, the EDS mapping of A-CQD/Co_{0.03}Ni_{0.97}LDH (Figure S9e) show that Ni, Co, C, and O are uniformly distributed in the nanosheets of A-CQD/Co_{0.03}Ni_{0.97}LDH, indicating that the CQD remain uniformly dispersed on the surface of A-Co_{0.03}Ni_{0.97}LDH nanosheets, and the S signal are weakened, which confirms that S species will be transformed and detached during the electrochemical activation process.



Figure S10. XPS spectra of Co/CQD. (a) Co 2p3/2, (b) C 1s, (c) S 2p, (d) O 1s.

Figure S10a presents the Co 2p spectrum of Co/CQD, indicating that Co^{2+} and Co^{3+} co-exist in Co/CQD. The high-resolution C 1s spectrum presented in Figure S10b can be deconvoluted to three species, including C-C (284.8 eV), C-O (285.7 eV) and O-C=O (288.8 eV). XPS spectra of S 1s for Co/CQD can be fitted to three species of M=S (162.5 eV), C-S (163.3 eV) and S-O (167.8 eV). The O 1s spectra of Co/CQD shown in Figure S10d can be fitted to three peaks of O-H (530.5 eV), C-O (532.0 eV), O-C=O (534.6 eV), indicating the abundance of oxygen-containing functional groups on Co/CQD.



Figure S11. Full XPS spectra of CQD/Co_{0.03}Ni_{0.97}LDH and A-CQD/Co_{0.03}Ni_{0.97}LDH. The full XPS spectra are presented in Fig. S11, showing the coexistence of Ni, Co, O and C elements in the samples before and after electrochemical activation. Moreover, the S signal is hardly observed in both materials, which proves that the content of S in the materials is very low.



Figure S12. EXAFS k³ x(k) oscillation curves and R space fitting results of Ni K edge for (a, b) CQD/Co_{0.03}Ni_{0.97}LDH and (c, d) A-CQD/Co_{0.03}Ni_{0.97}LDH.

Figure S12 shows the EXAFS $k^3 x(k)$ oscillation curves and R space fitting results of Ni K edge for the CQD/Co_{0.03}Ni_{0.97}LDH and A-CQD/Co_{0.03}Ni_{0.97}LDH, which indicates that the geometric structure of Ni atoms in CQD/Co_{0.03}Ni_{0.97}LDH and A-CQD/Co_{0.03}Ni_{0.97}LDH is almost the same.



Figure S13. EXAFS k³ x(k) oscillation curves and R space fitting results of Co K edge for (a, b) CQD/Co_{0.03}Ni_{0.97}LDH and (c, d) A-CQD/Co_{0.03}Ni_{0.97}LDH.

Figure S13 presents the EXAFS $k^3 x(k)$ oscillation curves and R space fitting results of Co K edge for the CQD/Co_{0.03}Ni_{0.97}LDH and A-CQD/Co_{0.03}Ni_{0.97}LDH. Significant change in peaks intensity and emergence of new peaks after electrochemical activation. These evidences further indicate that Co atoms are most susceptible to be oxidized to high valence state during the electrochemical activation and the coordination of Co atoms may change significantly.



Figure S14. Three-dimensional charge density of (a) A-Ni(OH)₂, (b) A-Co_{0.03}Ni_{0.97}LDH and (c) A-CQD/Co_{0.03}Ni_{0.97}LDH.

Figure S14 displays the three-dimensional charge density of A-Ni(OH)₂, A-Co_{0.03}Ni_{0.97}LDH and A-CQD/Co_{0.03}Ni_{0.97}LDH. Among them, A-Ni(OH)₂ and A-Co_{0.03}Ni_{0.97}LDH mainly rely on M-O to realize the inter-elemental charge transfer, while the charge transfers in A-CQD/Co_{0.03}Ni_{0.97}LDH can be achieved by CQD. CQD avoids oxidation of Ni²⁺ to Ni³⁺ due to better electron transport capability thereby reducing the oxidative properties of Co^{3+} .



Figure S15. CV curves of electrodes at different scan rates. (a) A-Ni(OH)₂, (b) A-Co_{0.03}Ni_{0.97}LDH, (c) A-CQD/Co_{0.03}Ni_{0.97}LDH.

Figure S15 presents the CV curves of A-Ni(OH)₂, A-Co_{0.03}Ni_{0.97}LDH, and A-CQD/Co_{0.03}Ni_{0.97}LDH at different scan rates of $3\sim 20 \text{ mV s}^{-1}$. The current increase of A-CQD/Co_{0.03}Ni_{0.97}LDH is higher than that of A-Ni(OH)₂ and A-Co_{0.03}Ni_{0.97}LDH, indicating it better capacitive performance than A-Ni(OH)₂ and A-Co_{0.03}Ni_{0.97}LDH.



Figure S16. A-Co/CQD electrodes: (a) CV curves at different scan rates; (b) GCD curves at different current densities; (c) Nyquist plots.

Figure S16 displays the capacitive properties of A-Co/CQD. The increase current with the increase of scan rate indicates that the ideal capacitive behavior of A-Co/CQD. According to the discharge curves shown in Figure S16b, A-Co/CQD shows a low specific capacity of 75 F g⁻¹ in the potential window of 0-0.5 V under a current density of 1A g⁻¹. Furthermore, the A-Co/CQD electrode displays an excellent conductivity with a low R_s of 1 and a low R_{ct} of 0.9 Ω .



Figure S17. A-CQD/Ni(OH)₂ electrodes: (a) CV curves at different scan rates; (b) GCD curves at different current densities; (c) Nyquist plots.

Figure S17 displays the capacitive properties of A-CQD/Ni(OH)₂. With the increase of scanning rate, the CV images of A-CQD/Ni(OH)₂ remained relatively intact and did not undergo significant polarisation, indicating that it has desirable capacitive properties. According to the discharge curves shown in Figure S17b, A-CQD/Ni(OH)₂ shows specific capacity of 1376 F g⁻¹ in the potential window of 0-0.5 V under a current density of 1A g⁻¹. Furthermore, the A-CQD/Ni(OH)₂ electrode displays an excellent conductivity with a low R_s of 1.1 and a low R_{ct} of 1.5 Ω .



Figure S18. GCD curves of (a) A-Ni(OH)₂, (b) A-Co_{0.03}Ni_{0.97}LDH, (c) A-CQD/Co_{0.03}Ni_{0.97}LDH electrode at different current densities.

Figure S18 demonstrates the GCD curves of A-Ni(OH)₂, A-Co_{0.03}Ni_{0.97}LDH and A-CQD/Co_{0.03}Ni_{0.97}LDH in the voltage range of 0-0.5 V at different current densities. The clear potential platforms indicate that the battery-type characterizes of A-Ni(OH)₂, A-Co_{0.03}Ni_{0.97}LDH and A-CQD/Co_{0.03}Ni_{0.97}LDH. In addition, compared with the A-Ni(OH)₂ and A-Co_{0.03}Ni_{0.97}LDH electrodes, the longer discharge time for A-CQD/Co_{0.03}Ni_{0.97}LDH indicates that the higher specific capacity of A-CQD/Co_{0.03}Ni_{0.97}LDH than that of A-Ni(OH)₂ and A-Co_{0.03}Ni_{0.97}LDH.



Figure S19. Nyquist plot of A-CoNiLDH electrodes

Figure S19 presents the EIS plots of A-CoNiLDH electrodes with different cobalt contents. A-CoNiLDH electrodes display the lower than that of A-Ni(OH)₂, indicating the Co doping can efficiently improve the conductivity of A-CoNiLDH electrodes.



Figure S20. Equivalent circuit diagram for fitting Nyquist plot.



Figure S21. Fitting plots of log (scan rate) versus log of anodic and cathodic peaks current densities for the calculation of b-values of the A-CQD/Co_{0.03}Ni_{0.97}LDH electrode.

As shown in Figure S21, the b values were obtained by the slope of a linear fitting line of log(v) vs. log(i) from the CV curves. When the b value is 1, it represents capacitive-type electrochemical behavior, and when the b value is 0.5, it represents the diffusion-controlled charge storage behavior. From the redox peak current, the b value of A-CQD/Co_{0.03}Ni_{0.97}LDH is 0.375. It shows that the battery-type oxidation-reduction kinetics is dominant, and its charge storage capacitances is controlled by slow semi-infinite diffusion.



Figure S22. Capacitive contribution (shaded areas) in the CV curve of A-CQD/Co_{0.03}Ni_{0.97}LDH at (a) 3 mV s⁻¹, (b) 5 mV s⁻¹, (c) 8 mV s⁻¹, (d) 10 mV s⁻¹, (e) 15 mV s⁻¹, (f) 20 mV s⁻¹.

Figure S22 shows the capacitive contribution (shaded areas) in the CV curve of the A-CQD/Co_{0.03}Ni_{0.97}LDH at different scan rates. With the increase of scan rate, the capacitive contribution of the A-CQD/Co_{0.03}Ni_{0.97}LDH increases.



Figure S23. Structure of A-CQD/Co_{0.03}Ni_{0.97}LDH//ACC ASC.

As shown in Figure S23, the A-CQD/Co_{0.03}Ni_{0.97}LDH//ACC ASC devices were assembled using the A-CQD/Co_{0.03}Ni_{0.97}LDH electrodes as positive electrode, activated carbon cloth (ACC) as negative electrode, 6 M KOH as the electrolyte and glass fiber filter paper (GF) as separator.



Figure S24. (a) CV and (b) GCD curves of ACC electrode.

Figure S24 displays the CV and GCD curves of activated carbon cloth (ACC). As shown in CV curves, the quasi-rectangular indicates the typical double layer capacitance. The calculate areal capacitance of ACC can reach up to 1.8 F cm^{-2} at 3 mA cm⁻² and maintain 1.3 F cm^{-2} at 30 mA cm⁻² (Figure S24b), showing a high capacitance retention of 72.2%.



Figure S25. A-CQD/Co_{0.03}Ni_{0.97}LDH//ACC ASC (a) CV curves at different scan rates; (b) GCD curves at different current densities.

Figure S25a exhibits the CV profiles of A-CQD/Co_{0.03}Ni_{0.97}LDH//ACC ASC at the scanning rates from 3 to 20 mV s⁻¹, indicating that there are both electric double-layer capacitance and batterytype capacitance at 0–1.5 V. Figure S25b presents the GCD profiles of A-CQD/Co_{0.03}Ni_{0.97}LDH//ACC ASC, and the A-CQD/Co_{0.03}Ni_{0.97}LDH//ACC ASC is 390 F g⁻¹ (0.81 F cm⁻²) at 3 mA cm⁻² and remained 243 F g⁻¹ (0.68 F cm⁻²) at 30 mA cm⁻².



Figure S26. (a) CV curves, (b) GCD curves and (c) rate performance of A-Ni(OH)₂//Zn cell.

The CV curves (Figure S26a) exhibit the obvious redox peaks from 0.1 to 1 mV s⁻¹. Furthermore, Figure S26b represents the GCD curves from 0.1 to 1 A g⁻¹. Accordingly, the specific capacity is calculated to be 117 mAh g⁻¹ at 0.1 A g⁻¹ and 30 mAh g⁻¹ at 1 A g⁻¹, which is lower than A-CQD/Co_{0.03}Ni_{0.97}LDH//Zn. The rate capacities with the corresponding columbic efficiency are shown in Figure S26c at various current densities. At high current densities, the capacity of A-Ni(OH)₂//Zn decreases significantly and the coulombic efficiency at 0.1 A g⁻¹ is significantly lower than A-CQD/Co_{0.03}Ni_{0.97}LDH//Zn.

Samples	BET Surface Area (m ³ g ⁻¹)
Ni(OH) ₂	41.0
A-Ni(OH) ₂	48.6
Co _{0.03} Ni _{0.97} LDH	64.6
A-Co _{0.03} Ni _{0.97} LDH	91.0
CQD/Co _{0.03} Ni _{0.97} LDH	118.7
A-CQD/Co _{0.03} Ni _{0.97} LDH	122.5

 $\label{eq:stable} \textbf{Table S1.} Specific surface area calculated from N_2 adsorption-desorption isotherms.$

Sample	Path	Ν	DIÅI	σ ² [10 ⁻	$\Delta E_0 (eV)$	R
			K [A]	³ Å ²]		factor
CQD/Co _{0.03} Ni _{0.97} LDH	Ni-O	5.9±0.2	2.01±0.02	7.26±1.9	5 1 1 0 5	0.0266
	Ni-Ni/Co	5.8±0.3	3.08±0.04	14.01±1.1	-3.1±0.3	
A-	Ni-O	6.0±0.2	2.07±0.02	7.19±2.3	64:08	0.0214
CQD/Co _{0.03} Ni _{0.97} LDH	Ni-Ni/Co	6.1±0.4	3.06±0.06	11.6±3.2	-0.4±0.8	0.0314

Table S2. EXAFS fitting results for the structural parameters around Ni atoms.

Sample	Path	Ν	Β ΓÅ1	σ ² [10 ⁻	$\Delta \mathbf{F}_{\alpha}(\mathbf{a}\mathbf{V})$	R
			K [A]	Å ²]	$\Delta E_0 (ev)$	factor
	Со-О	3.1±0.4	2.16±0.02	9.53±3.1		0.042
CQD/Co _{0.03} Ni _{0.97} LDH	Co-C/S	1.2±0.6	2.27±0.11	10.33±4.3	-6.3±1.5	0.043
	Co-Co/Ni	5.4±0.3	3.14±0.04	14.01±1.1		7
A-CQD/Co _{0.03} Ni _{0.97} LDH	Со-О	3.8±0.3	2.07±0.04	9.19±5.8		0.022
	Co-C/S	2.4±0.8	2.23±0.18	14.03±4.7	-7.2±1.1	0.055
	Co-Co/Ni	5.3±0.4	3.03±0.43	12.8±4.2		4

Table S3. EXAFS fitting results for the structural parameters around Co atoms.

N is the coordination number; R is interatomic distance (the bond length between Co central atoms and surrounding coordination atoms); σ^2 is Debye-Waller factor (represents the thermal and static disorder in absorber-scatterer distances); ΔE_0 is edge energy shift (the difference between the zero kinetic energy value of the sample and that of the theoretical model); R factor is used to assess the goodness of the fitting.

Samples	Nickel (%)	Cobalt (%)
Co/CQD	0	5.16
CQD/Co _{0.03} Ni _{0.97} LDH	96.93	3.07
Co _{0.03} Ni _{0.97} LDH	96.29	3.71
Co _{0.2} Ni _{0.8} LDH	78.11	21.89
Co _{0.3} Ni _{0.7} LDH	73.10	26.90
Co _{0.4} Ni _{0.6} LDH	59.11	40.89
Co _{0.5} Ni _{0.5} LDH	49.80	50.20
Co _{0.7} Ni _{0.3} LDH	70.60	29.40

Table S4. ICP results for individual electrode materials.

Samples	$R_{ m s}\left(\Omega ight)$	$R_{ ext{ct}}\left(\Omega ight)$
A-Ni(OH) ₂	1.2	2.3
A-Co(OH) ₂	0.9	0.6
A-Co _{0.03} Ni _{0.97} LDH	1.2	2.0
A-Co _{0.2} Ni _{0.8} LDH	1.0	1.5
A-Co _{0.3} Ni _{0.7} LDH	1.0	1.3
A-Co _{0.4} Ni _{0.6} LDH	1.1	1.1
A-Co _{0.5} Ni _{0.5} LDH	1.0	0.9
A-Co _{0.7} Ni _{0.3} LDH	1.1	0.8
A-Co/CQD	1.0	0.9
A-CQD/Co _{0.03} Ni _{0.97} LDH	1.1	0.9

Table S5. *Rs* and *Rct* fitted from the EIS curve.

Table S6. Comparison of our asymmetric capacitors with recently reported high performance

Devices	Electrolyte	Voltage (V)	Power density (mW cm ⁻²)	Energy density (mWh cm ⁻²)	Cycling stability	Ref.
CuCo ₂ O ₄ @Ni (OH) ₂ //AC	2 M KOH	1.5	1.2	0.125	84.0% at 20 mA cm ⁻² for 2000 cycles	[9]
CuCo ₂ S ₄ @NiMn- LDH//AC	6 M KOH	1.5	4.66	0.183	87.6% at 10 A g ⁻¹ for 10000 cycles	[10]
Ni1Co2/NF//AC	1 M KOH	1.6	8	0.035	85.0% at 10 mA cm ⁻² for 5000 cycles	[11]
Ni(OH)2// V2O5	2 M KOH	1.3	4.66	0.025	81.4% at 100 mV s ⁻¹ for 10000 cycles	[12]
NC LDH NSs@Ag@CC//AC	1 M KOH	1.6	12.1	0.040	88.1% at 5 mA cm ⁻² for 2000 cycles	[13]
A- CQD/Co _{0.03} Ni _{0.97} LD H//ACC	6 M KOH	1.5	2.25	0.254	94.2% at 30 mA cm ⁻² for 2000 cycles	This work

ASCs.

Devices	Electrolyte	Voltage (V)	Energy density (mWh cm ⁻²)	Power density (mW cm ⁻²)	Cycling stability	Ref.
NiCo ₂ O ₄ //Zn	1 M KOH+0.2 M Zn(Ac) ₂	1.9	0.954	0.29	82.7% at 6.4 A g ⁻¹ for 3500 cycles	[14]
NiSe//Zn-U	6 M KOH +0.2 M ZnCl ₂	1.9	0.64	1.22	64.8% at 5 mA cm ⁻² for 1300 cycles	[15]
P-NiCo ₂ O ₄ - x//Zn	1 M KOH+0.5 M Zn(Ac) ₂	1.9	0.41	3.5	72.3% at 25.3 A g ⁻¹ for 5000 cycles	[16]
Co ₃ O ₄ //Zn	1 M KOH+0.1 M Zn(Ac) ₂	1.9	0.48	4	80.0% at 10 A g ⁻¹ for 2000 cycles	[17]
A-Ni/NC//Zn	6 M KOH+0.5 M Zn(Ac) ₂	1.9	0.04	28	120% at 10 mA cm ⁻² for 36000 cycles	[18]
A- CQD/Co _{0.03} Ni _{0.9} 7LDH //Zn	6 M KOH+0.5 M Zn(Ac) ₂	1.9	1.44	0.72	95.7% at 0.3 A g ⁻¹ for 500 cycles	This work

 Table S7. Comparison of our aqueous zinc-nickel cells with those recently reported.

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