Supplementary information

Nickel hexacyanocobaltate quantum dots embedded in N-doped carbon for aqueous alkaline

battery with ultrahigh durability

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Fig. S1 Raman results of samples



Fig. S2 TEM results of CC-Ni-Co PBA. (b) and (c) HRTEM images



Fig. S3 SEM images of NC-Ni-Co PBA and CC-Ni-Co PBA after cycling tests

It could be seen that after cycling tests, NC-Ni-Co PBA and CC-Ni-Co PBA exhibited similar morphologies with sheet-like structures on the edges, which was typical morphological features of nickel (oxy)-hydroxides.



Fig. S4 TEM results of NC-Ni-Co PBA after cycling tests

TEM was conducted after cycling test to further confirm the phase transformation. We chose NC-Ni-Co PBA for TEM test. The HRTEM images in Fig. 2c showed lattice spacings of 0.207 nm and 0.208 nm, corresponding to the (422) plane of Ni-Co PBA. The existence of Ni(OH)₂ was confirmed by the enlarged HRTEM image of Fig. 2d and the SAED result. The lattice spacing of 0.234 nm in Fig. 2d could be assigned to (101) plane of Ni(OH)₂. The SAED result displayed diffraction rings with 0.156 nm and 0.218 nm, respectively, corresponding to the (110) plane of Ni(OH)₂ (PDF No. 14-0017) and (104) plane of NiOOH (PDF No. 06-0075), respectively.



Fig. S5 (a) and (b) CV curves of samples at various scan rates; (c) and (d) GCD curves of samples under various current densities



Fig. S6 CV curves of samples at different scan rates. The difference of oxidation and redox peaks potential are given.



Fig. S7 CV curves of NC-Ni-Co PBA in 1M KOH and 1M KCl.

To further confirm the energy storage mechanism of Ni-Co PBA, we conducted CV test in 1M KOH and 1M KCl and the results were shown in Fig. S7. Obviously, Ni-Co PBA showed obvious oxidation and reduction peaks in KOH, while, no current signal was observed in KCl. The results indicated that the energy storage mechanism originated from the redox reaction between Ni-Co PBA and OH⁻. Moreover, as previously discussed, corresponding nickel hydroxide and oxyhydroxides would inevitably be generated under such condition and the redox peaks were attributed to the reversible reaction between Ni²⁺/Ni³⁺.



Fig. S8 CV and GCD curves of the device

Sample	CC-Ni-Co PBA/eV	NC-Ni-Co PBA/eV
Fitting peaks		
$Ni^{2+} 2p_{3/2}$	855.8	855.95
$Ni^{3+} 2p_{3/2}$	856.6	857.32
$Ni^{2+} 2p_{1/2}$	873.1	873.67
$Ni^{3+} 2p_{1/2}$	874.6	875.21
$Co^{3+} 2p_{3/2}$	780.38	781.14
$Co^{2+} 2p_{3/2}$	781.85	781.51
${\rm Co}^{3+} 2p_{1/2}$	795.3	795.99
$Co^{2+} 2p_{1/2}$	796.9	796.78
Metal-O	529.32	531.57
O/surface hydroxyl group	531.12	532.57
O/crystal water H ₂ O	533.53	/
Absorbed oxygen	532.21	533.24
C≡N/Pyridinic N	398.45	397.98
Pyrrolic N	399.95	/
C-C	284.8	284.8
C≡N	285.5	286.15
C=0	287.7	288.2

 Table S1 XPS fitting parameters and results of CC-Ni-Co PBA and NC-Ni-Co PBA