Supporting Information

Urchin-like Co-doped NiS_2/C nanorod array with enriched sulfur vacancy for asymmetric supercapacitors

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First-principles Calculation: To calculate the electronic structures, first-principles calculations were carried out with the spin-polarized Generalized Gradient Approximation (GGA) by adopting the Perdew–Burke–Ernzerh (PBE) exchange-correlation parameterization to the Density Functional Theory (DFT) incorporating the LDA+U formalism using the CASTEP program. A plane-wave basis with a kinetic energy cutoff of 400.0 eV and a Monkhorst-Pack grid with a 4×4×4 k-point mesh for the Brillouin zone integration were used value of the smearing was 0.2 eV. The electronic minimization parameter of the total energy/atom convergence tolerance was 5.0×10^{-6} eV.The OH adsorption energy calculation was based on the equation：

$$E_{ads}(OH) = E_{total}(sur + OH) - E_{total}(OH) - E_{total}(sur)$$

where $E_{total}(sur + OH)$, $E_{total}(sur)$ and $E_{total}(OH)$ represent the total energy of slab after adsorption, the energy of the bare slab and the energy of the adsorbate OH^- species, respectively.

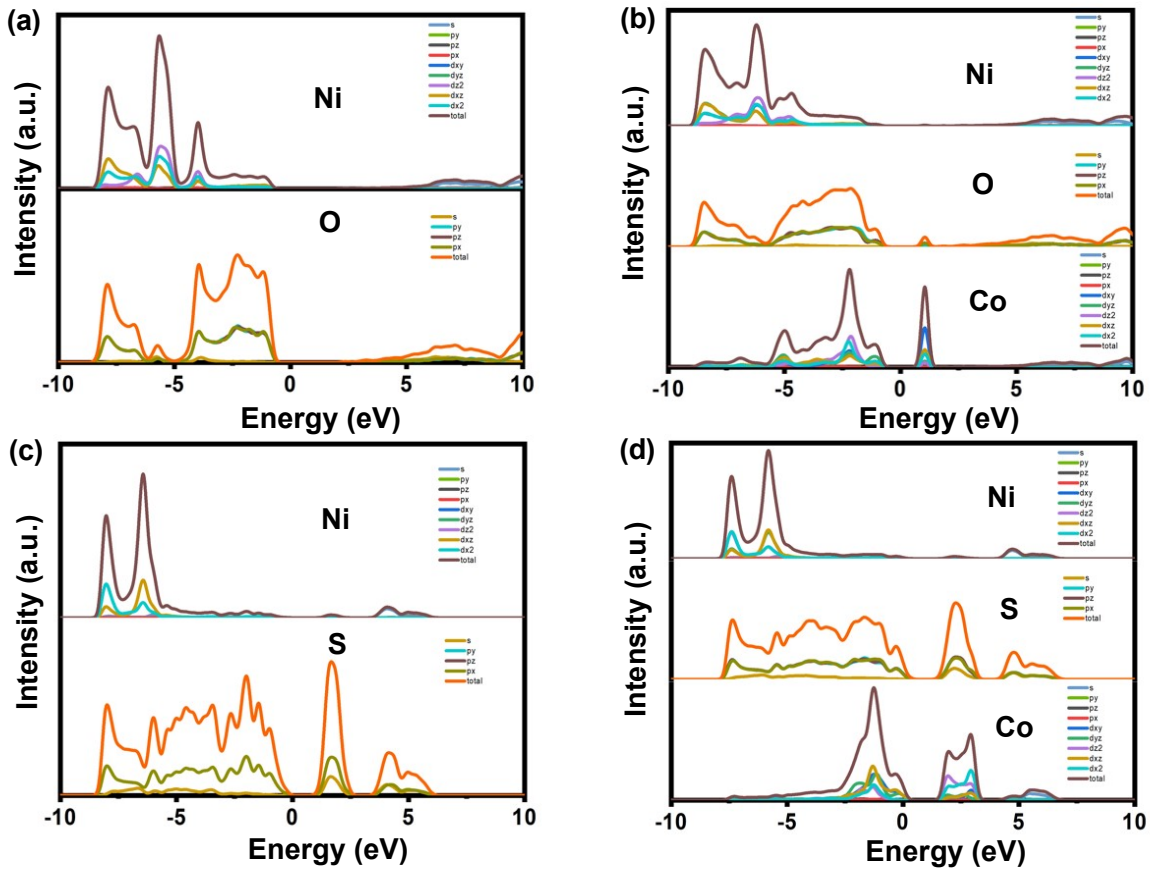

Figure S1 Total densities of states of and projected densities of states of (a) NiO, (b) Co-NiO, (c) NiS_2 and Co-NiS_2.

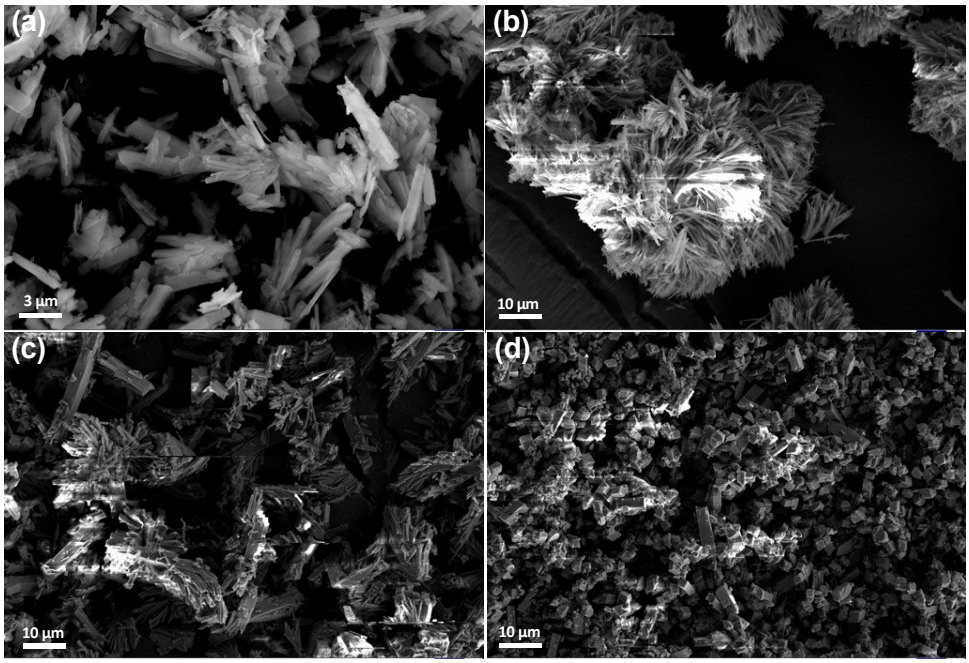

Figure S2 SEM images of Co-BTC with the Ni/Co ratio of (a) 1:0, (b) 1:0.5, (c) 1:1 and (d) 0:1.

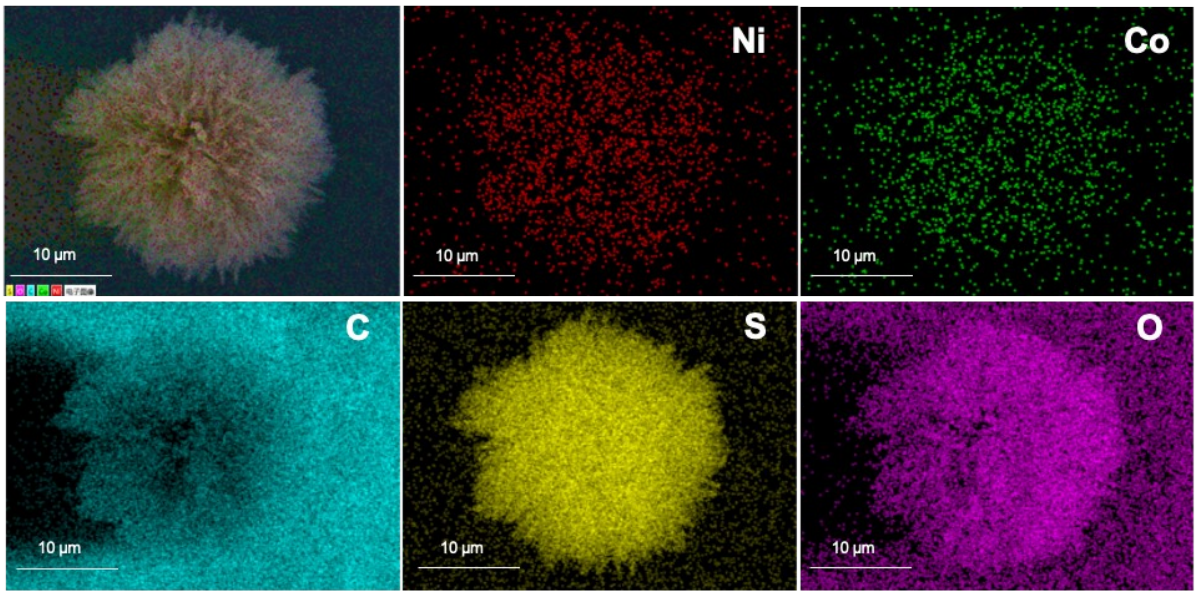

Figure S3 EDS mapping images of Co-NiS_2/C.

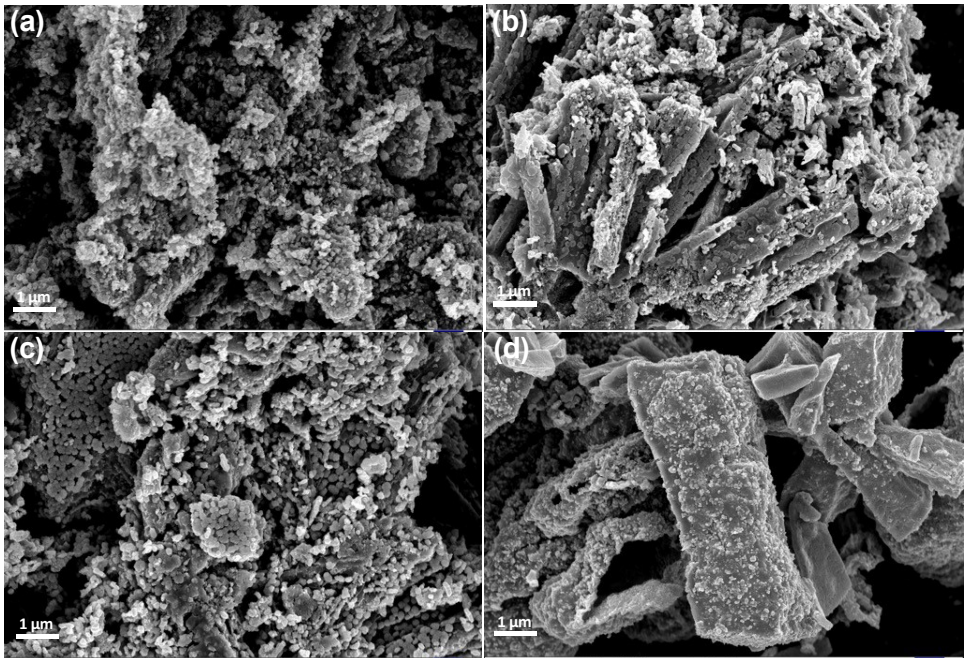

Figure S4 SEM images of Co-NiS_2/C with the Ni/Co ratio of (a)1:0, (b) 1:0.5, (c) 1:1, (d) 0:1.

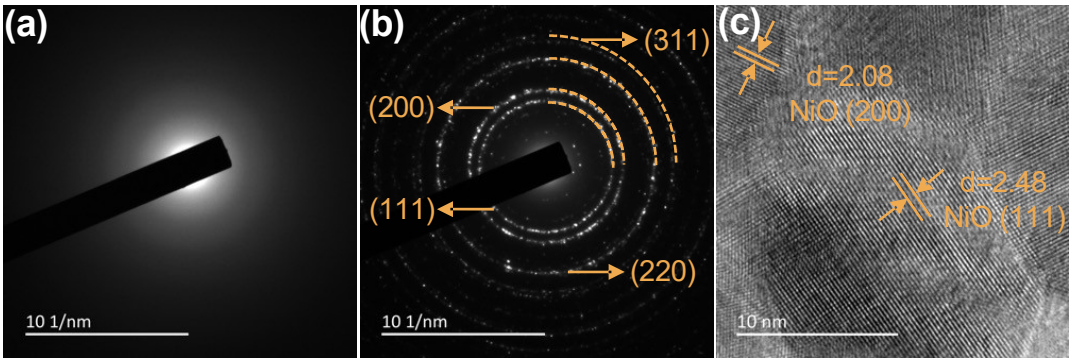

Figure S5 SAED images of (a) Co-NiBTC and (b) Co-NiO. (c) HRTEM of Co-NiO.

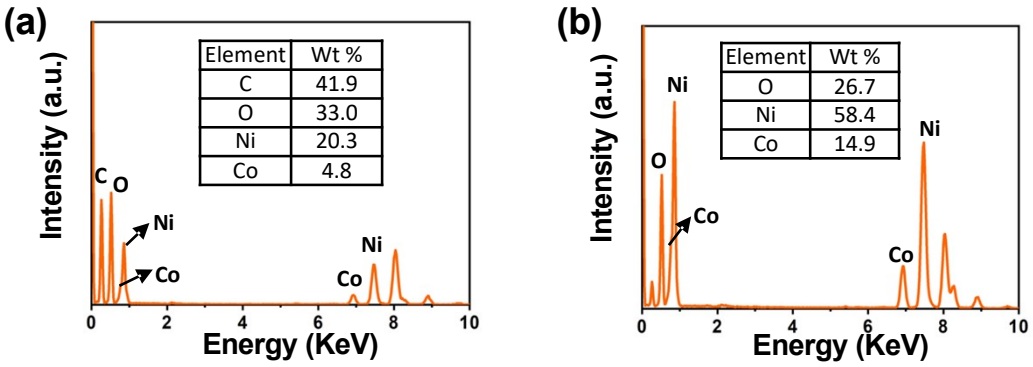

Figure S6 EDS analysis spectrum of (a) Co-NiBTC and (b) Co-NiO.

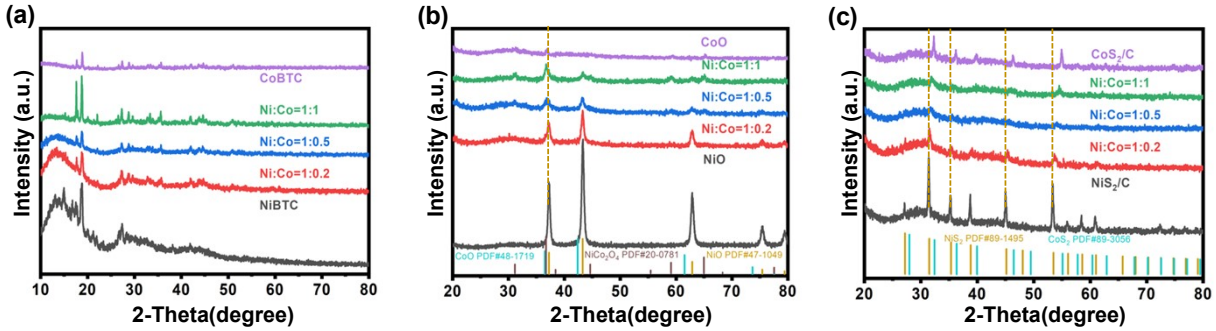

Figure S7 XRD patterns (a) Co-NiBTC, (b) Co-NiO and (c) Co-NiS_2/C with different Ni/Co ratios.

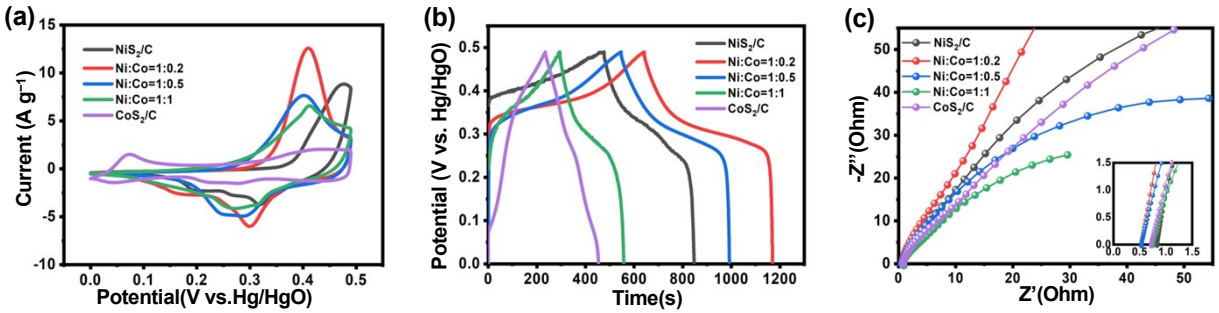

Figure S8 (a) CV curves, (b) GCD curves and (c) Nyquist plots of Co-NiS_2/C with different Ni/Co ratio of 1:0, 1:0.2, 1:0.5, 1:1 and 0:1.

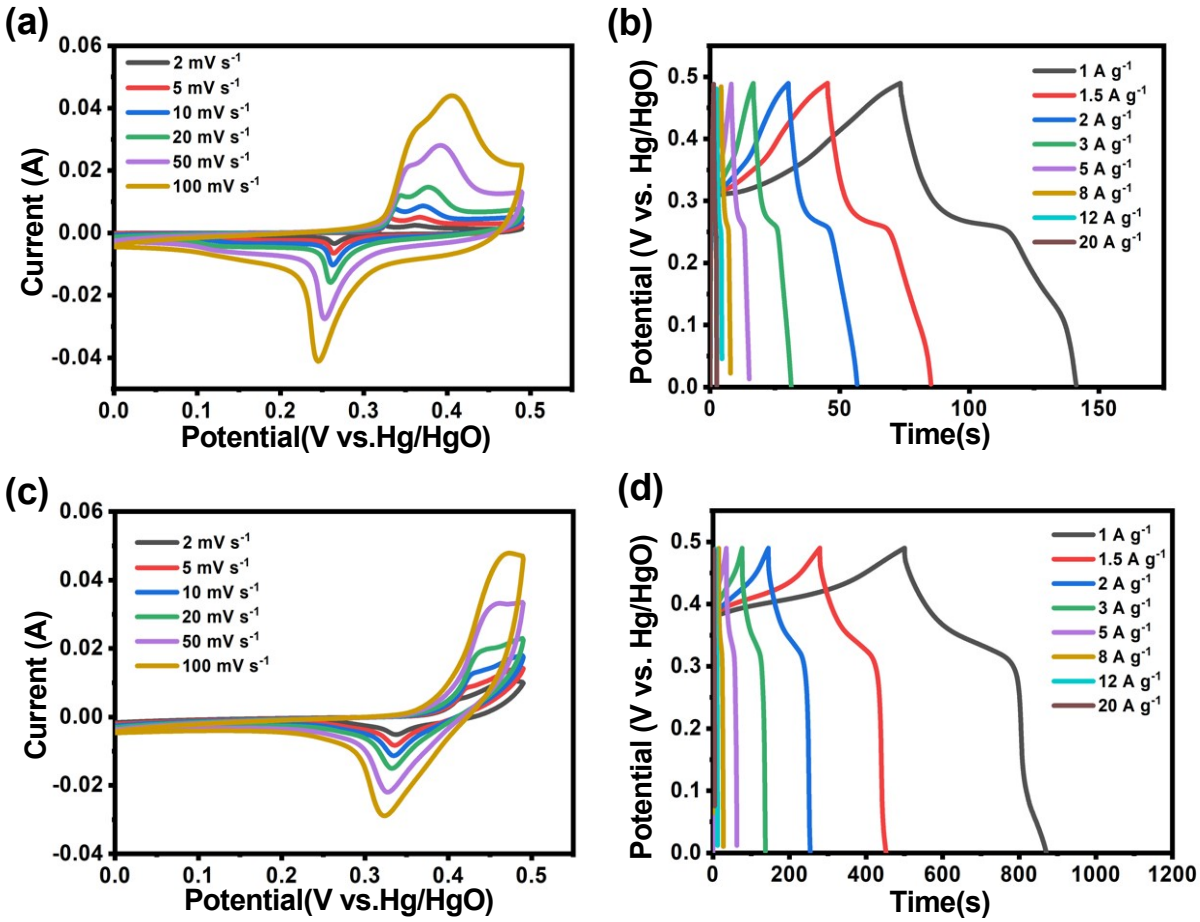

Figure S9 (a) CV curves and (b) GCD curves of Co-NiBTC. (c) CV curves and (d) GCD curves of Co-NiS_2.

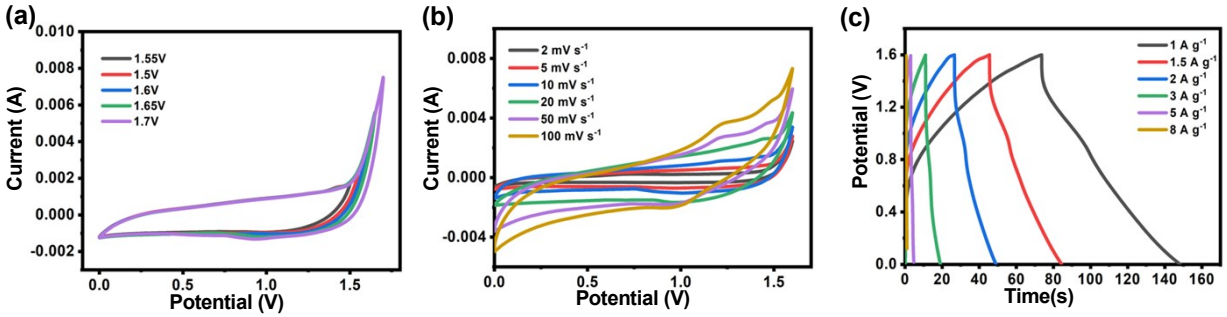

Figure S10 (a) CV curves at 10 mV s^{-1}, (b) CV curves and (c) GCD curves of Co-NiS_2//AC ASC device.

Table S1. Adsorption configurations and energies (eV) of OH^-.

Samples	NiO	Co-NiO(Ni)	Co-NiO(Co)	NiS_2	Co-NiS_2(Ni)	Co-NiS_2(Co)
Before Adsorption (eV)	-117.8322	-137.9754	-137.9754	-98.7659	-162.2247	-162.2247
After Adsorption (eV)	-125.7464	-146.1379	-145.8362	-108.6614	-173.9410	-173.5748
OH^-			-7.0816			
Adsorption energy (eV)	-0.8326	-1.0809	-0.7792	-2.8139	-4.6347	-4.2685

Table S2. The relative contents of elements in Co-NiBTC, Co-NiO, Co-NiS_2 and Co-NiS_2/C were derived from XPS data.

Samples	Co-NiBTC	Co-NiO	Co-NiS_2	Co-NiS_2/C
Co (At %)	1.36	8.52	1.16	0.79
Ni (At %)	4.78	27.71	3.93	2.17
C (At %)	58.16	10.42	38.03	39.52
O (At %)	35.7	53.35	42.8	31.8
S (At %)	/	/	14.08	25.71

Table S3. The R_S and R_{ct} of different electrodes.

Samples	Co-NiO	Co-NiS_2	Co-NiS_2/C
R_s (Ω)	0.75	0.56	0.49
R_{ct} (Ω)	0.71	0.59	0.53

Table S4 Electrochemical performance of different ASCs.

Electrode materials	P ($W \cdot kg^{-1}$)	E ($Wh \cdot kg^{-1}$)	References
Co-NiS_2/C // AC	1271.5	36.0	This work
Co-NiS_2 // AC	1283.2	26.4	This work
H-$NiMoS_4$/NiS_2 // NCO	958.6	38.6	[1]
$CuCo_2S_4$–rGO // AC	450	16.0	[2]
NiS // AC	900	31	[3]
GH@NC@Co_9S_8 // GH@NC	800	23.6	[4]
Co_9S_8-NSA // AC	828.5	20	[5]
CoNiLDH-rGO-$CoNi_2S_4$ // AC	749.9	31.6	[6]
Ni-CoP@C@CNT // GO	699.1	17.4	[7]

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