Electronic Supplementary Information (ESI)

Metallic Atomic Thickness $CuCo_2S_4$ Nanosheets as Efficient Bifunctional Electrocatalyst for Portable-Flexible Zn−Air Battery

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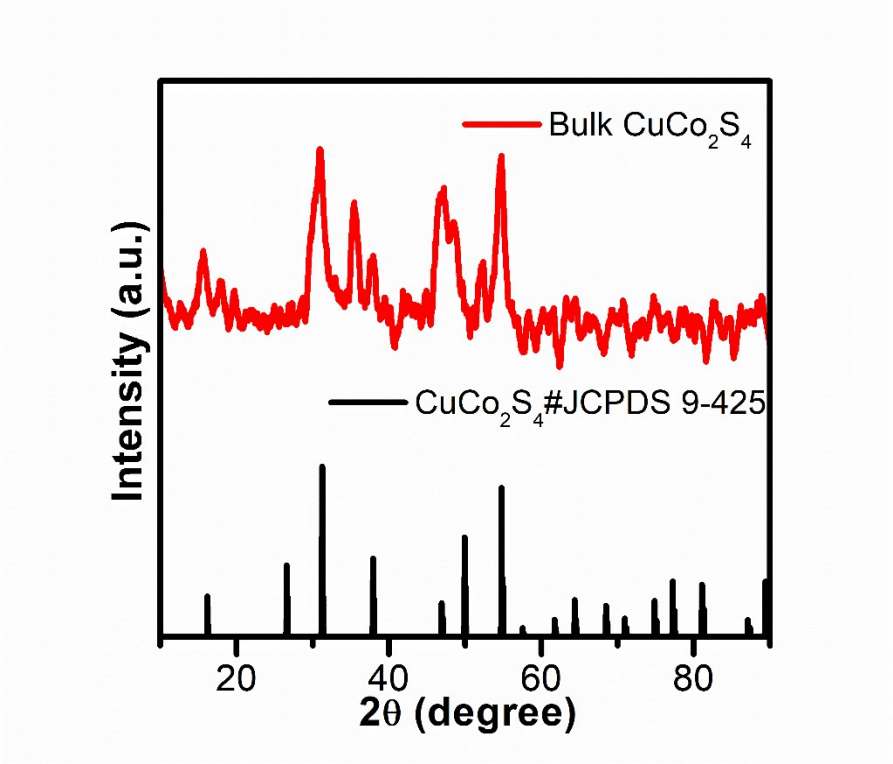

Figure S1. XRD patterns of bulk $CuCo_2S_4$.

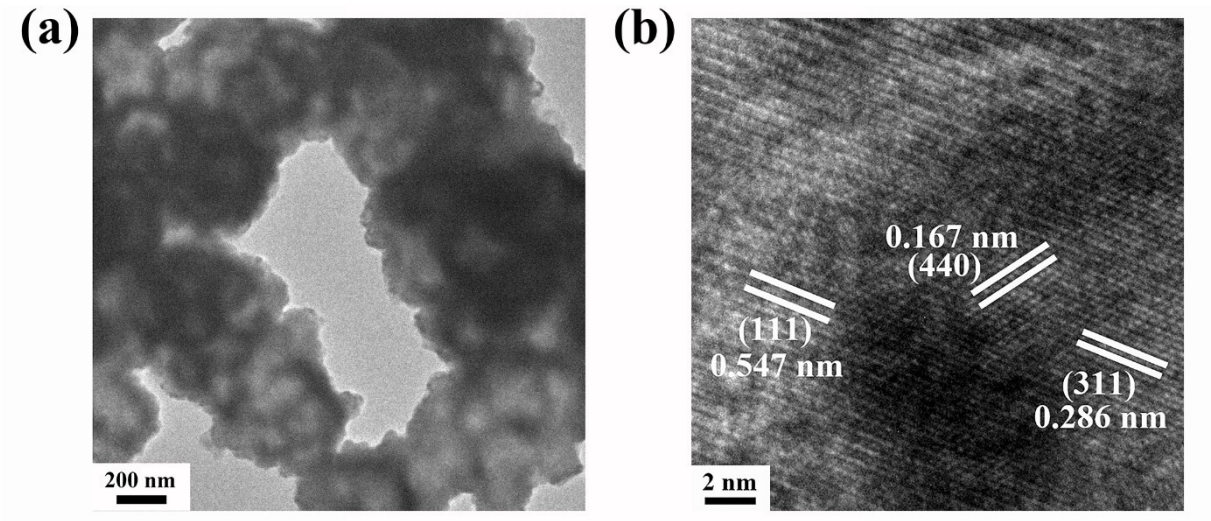

Figure S2. (a) TEM and (b) HRTEM images of bulk $CuCo_2S_4$.

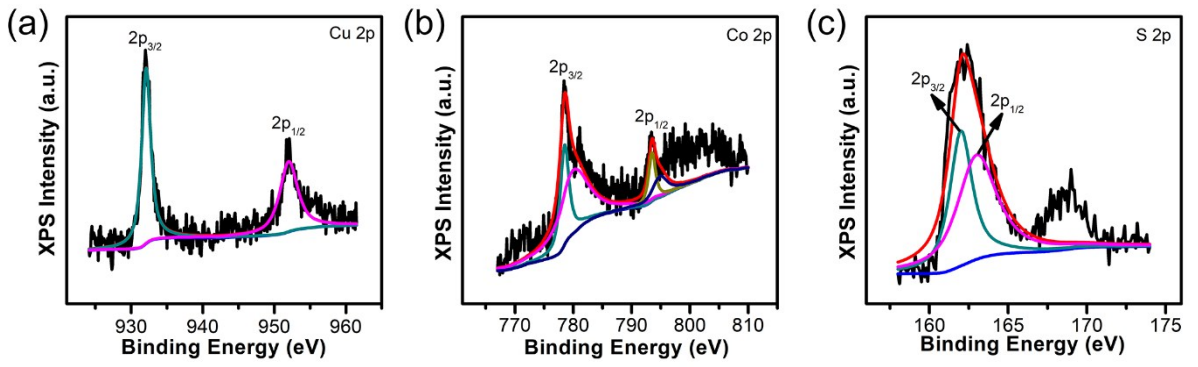

Figure S3. XPS spectra of (a) Cu 2p, (b) Co 2p and (c) S 2p of bulk $CuCo_2S_4$.

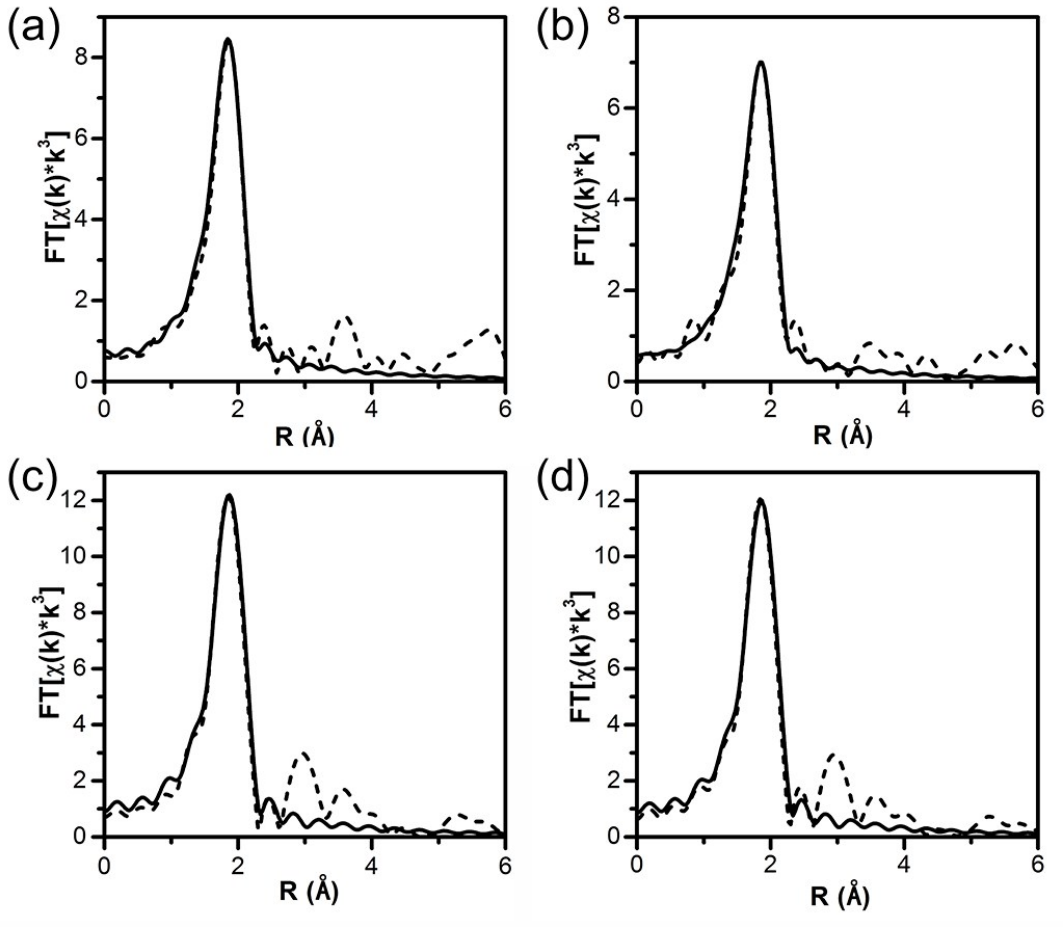

Figure S4. EXAFS data (dotted line) and fits (solid line) in R-space for (a) bulk $CuCo_2S_4$ Cu K-Edge, (b) $CuCo_2S_4$ NSs Cu K-Edge, (c) bulk $CuCo_2S_4$ Co K-Edge and (d) $CuCo_2S_4$ NSs Co K-Edge.

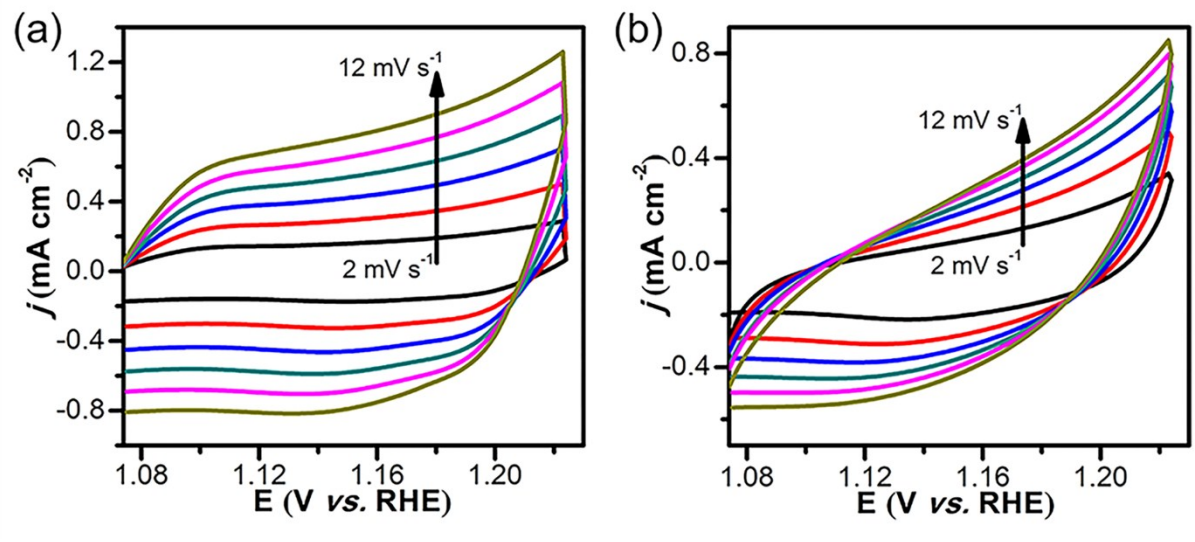

Figure S5. Cyclic voltammograms of (a) $CuCo_2S_4$ NSs and (b) bulk $CuCo_2S_4$ at different scan rate.

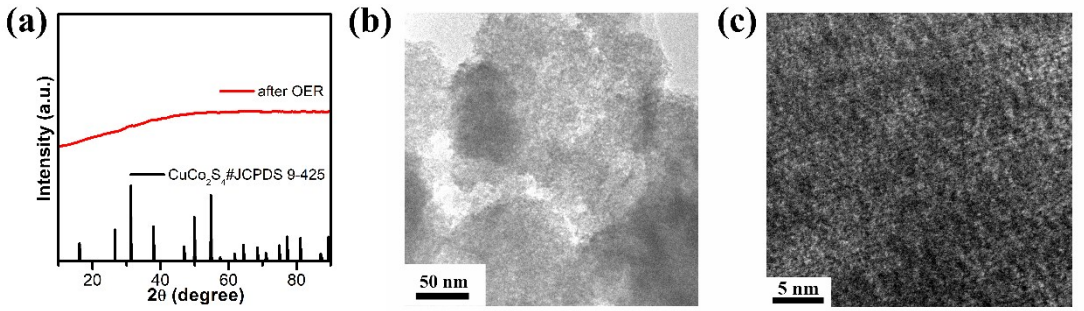

Figure S6. (a) The XRD patterns, (b) TEM image and (c) HRTEM image of the $CuCo_2S_4$ NSs after the OER test.

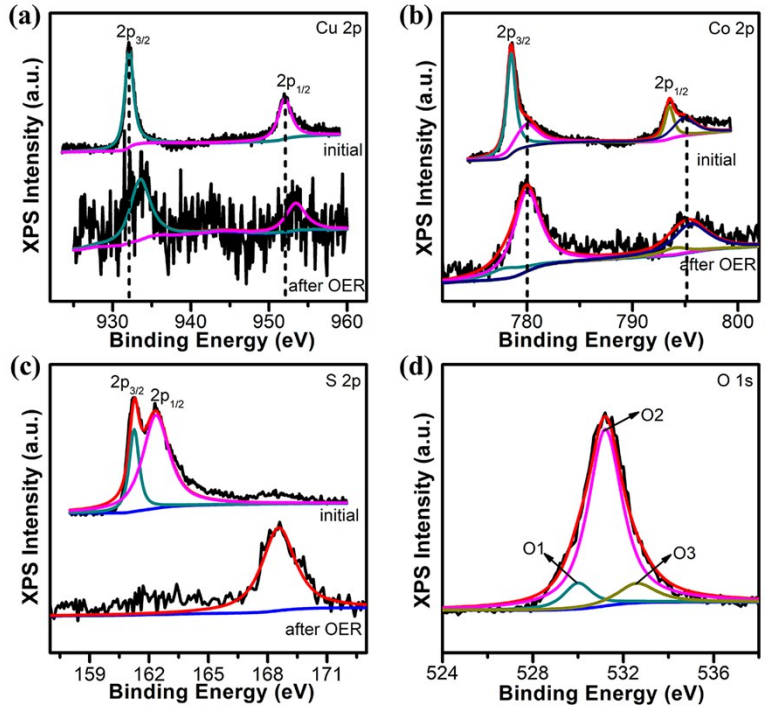

Figure S7. XPS spectra of (a) Cu 2p, (b) Co 2p, (c) S 2p and (d) O 1s of the $CuCo_2S_4$ NSs after the OER test.

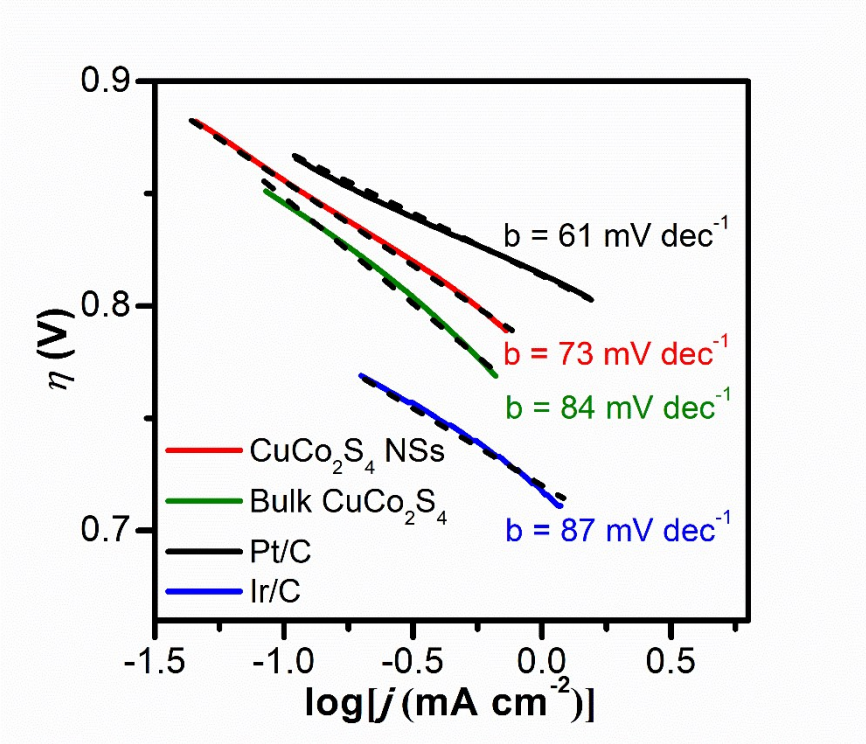

Figure S8. ORR Tafel plots of the $CuCo_2S_4$ NSs, bulk $CuCo_2S_4$, Ir/C (20 %) and Pt/C (20 %).

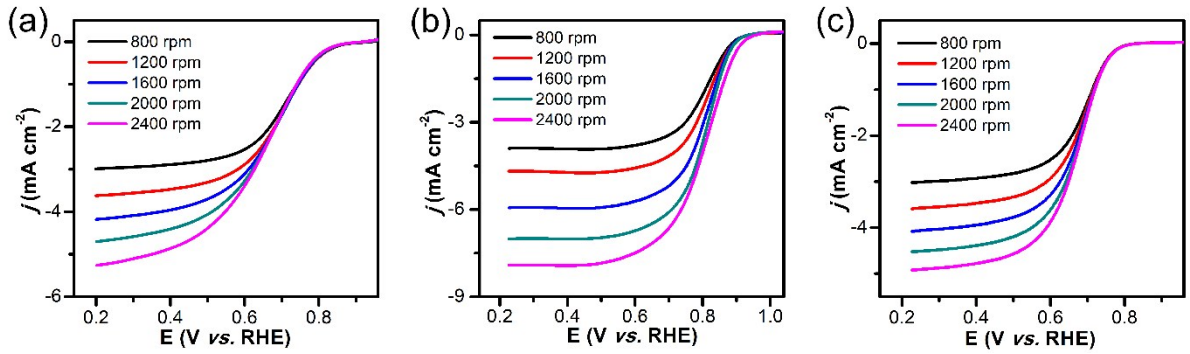

Figure S9. LSV curves of (a) bulk $CuCo_2S_4$, (b) Pt/C (20 %) and (c) Ir/C (20 %) at different rotating speeds.

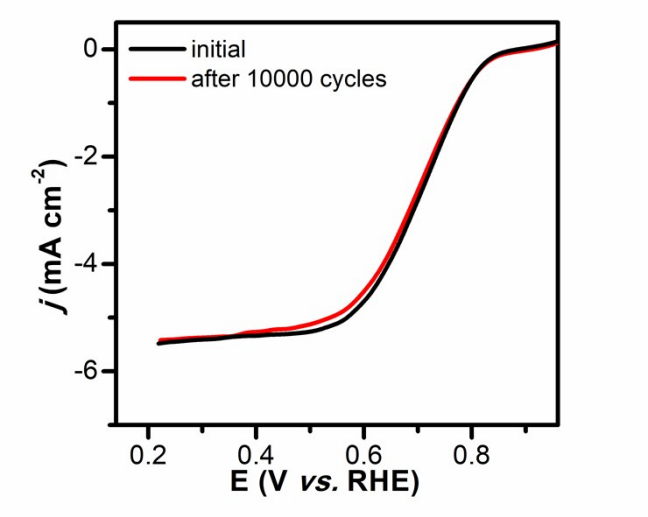

Figure S10. LSV curves of the $CuCo_2S_4$ NSs before and after 10 000 CV cycles for ORR.

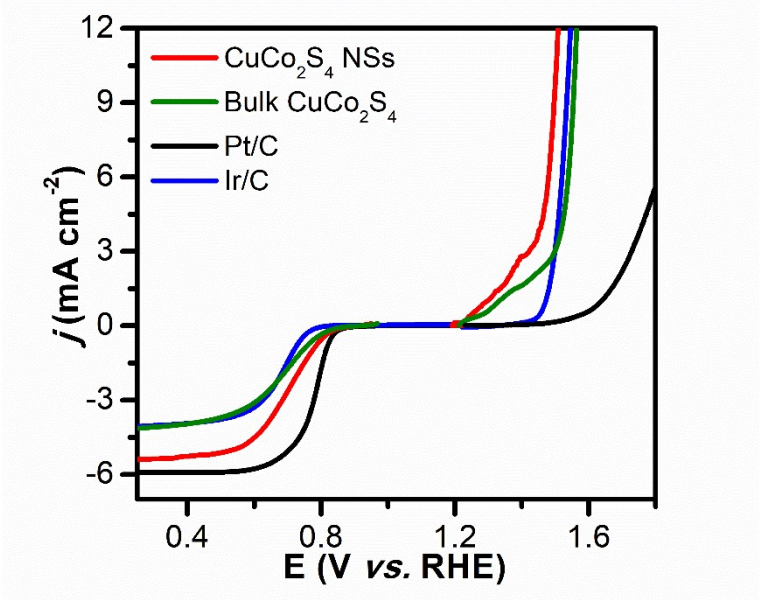

Figure S11. Polarization curves of Pt/C (20 %), Ir/C (20 %), the $CuCo_2S_4$ NSs and bulk $CuCo_2S_4$ in the whole region for OER and ORR.

Table S1. EXAFS Fitting results for Cu-center (top row) and Co center (bottom row) for bulk $CuCo_2S_4$ and the $CuCo_2S_4$ NSs. CN = Coordination Numbers, R = Vector distance (± 0.02 Å), σ^2 = Debye-Waller factor (± 0.001 Å^2).

Vector	Bulk $CuCo_2S_4$			$CuCo_2S_4$ NSs		
	CNs	**R(Å)**	**σ^2(Å^2)**	**CNs**	**R(Å)**	**σ^2(Å^2)**
Cu-S	4	2.25	0.009	3.1	2.26	0.009
Co-S	5.4	2.27	0.008	5.4	2.27	0.008

Table S2. Comparison of the OER performance for all used catalysts.

Catalysts	η_{onset} (mV *vs.* RHE)	η_{10} (mV *vs.* RHE)	Tafel slope (mV dec^{-1})	TOF at η = 400 mV (s^{-1})	Mass activity at η = 400 mV (A g^{-1})
$CuCo_2S_4$ NSs	242	287	46	0.230	489.2
Bulk $CuCo_2S_4$	281	330	79	0.134	285.1
Ir/C (20 %)	255	310	51	0.119	239.6

Table S3. Comparison of the ORR performance for all used catalysts.

Catalysts	Onset Potential (V *vs.* RHE)	$E_{1/2}$ (V *vs.* RHE)	Tafel slope (mV dec^{-1})	Limiting current density (mA cm^{-2})	Electron transfer number (*n*)
$CuCo_2S_4$ NSs	0.90	0.70	73	5.28	3.9
Bulk $CuCo_2S_4$	0.86	0.68	84	3.95	2.9
Pt/C (20 %)	0.94	0.78	61	5.91	4.1
Ir/C (20 %)	0.84	0.68	87	3.95	3.2

Table S4. The ΔE comparison of the $CuCo_2S_4$ NSs, bulk $CuCo_2S_4$, Pt/C (20 %) and Ir/C (20 %).

Catalysts	E_{OER} at *j* = 10 mA cm^{-2} (V *vs.* RHE)	$E_{1/2}$ (V *vs.* RHE)	$\Delta E = E_{OER} - E_{ORR}$ (V *vs.* RHE)
$CuCo_2S_4$ NSs	1.52	0.70	0.82
Bulk $CuCo_2S_4$	1.56	0.68	0.88
Pt/C (20 %)	1.91	0.78	1.13
Ir/C (20 %)	1.54	0.68	0.86