

Supporting Information

Controlled Phase Evolution from $\text{Cu}_{0.33}\text{Co}_{0.67}\text{S}_2$ to $\text{Cu}_3\text{Co}_6\text{S}_8$ Hexagonal Nanosheet as Oxygen Evolution Reaction Catalysts

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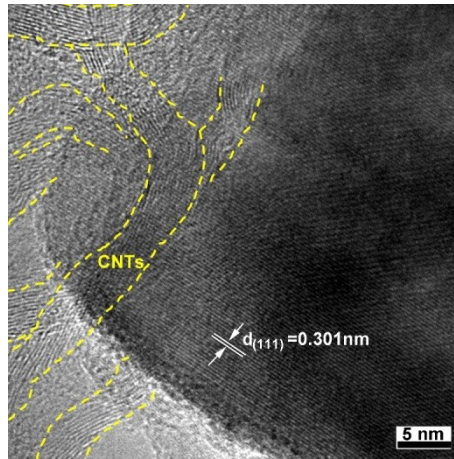


Fig. S1. high-resolution TEM image of $\text{Cu}_{0.33}\text{Co}_{0.67}\text{S}_2/\text{CNT}$.

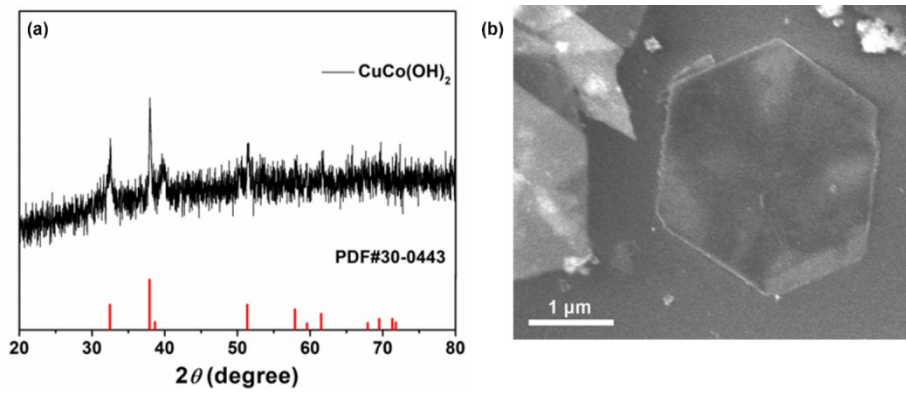


Fig. S2. (a) XRD pattern and SEM image of $\text{CuCo}(\text{OH})_2$.

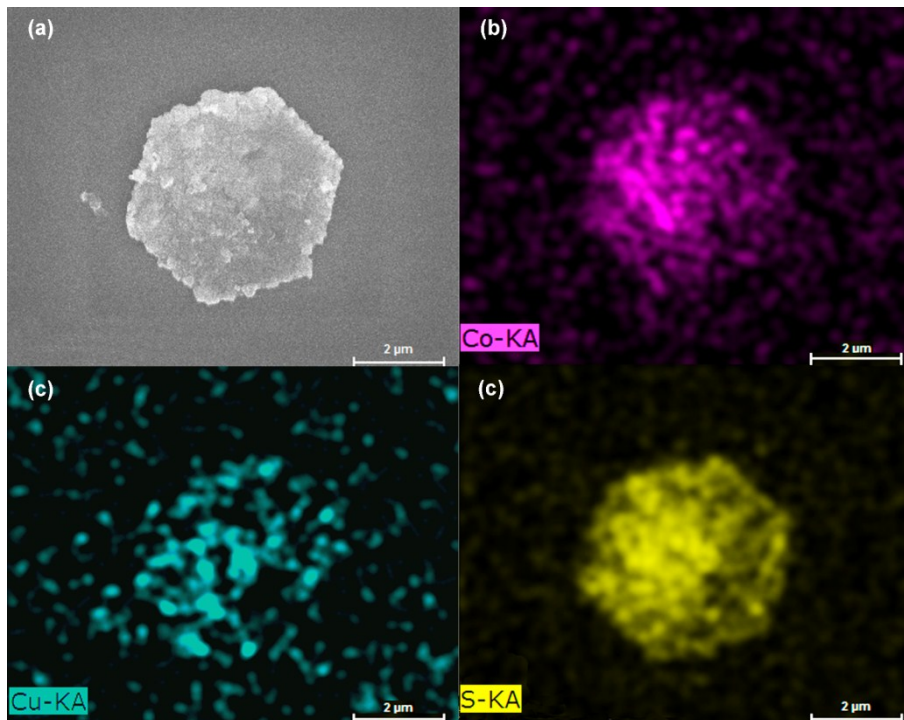


Fig.S3. (a)HAADF-SEM image and (b–d) elemental mapping images of $\text{Cu}_{0.33}\text{Co}_{0.67}\text{S}_2$.

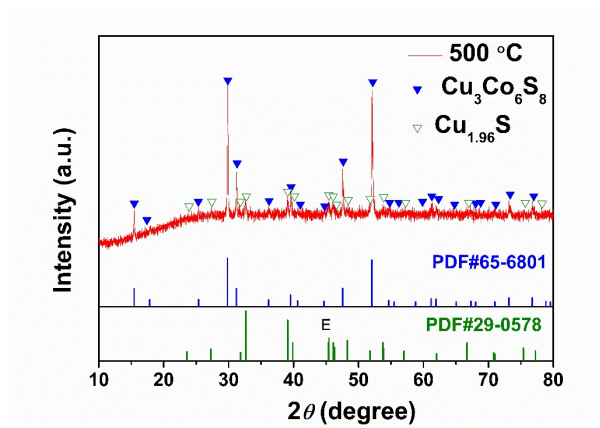


Fig. S4. XRD pattern of the Cu-Co-S product at 500 °C.

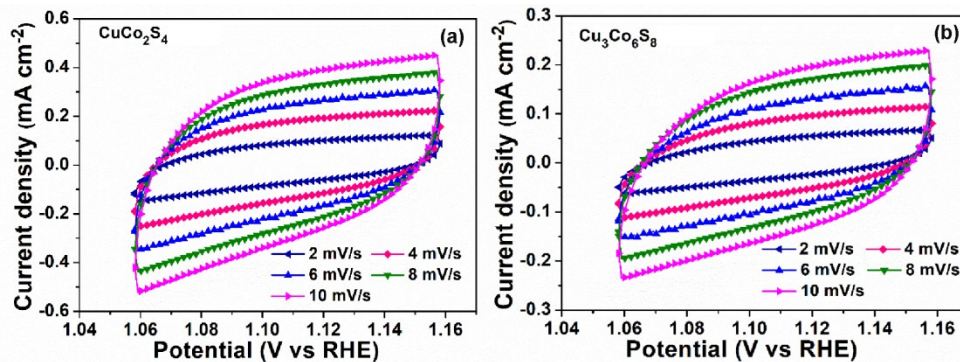
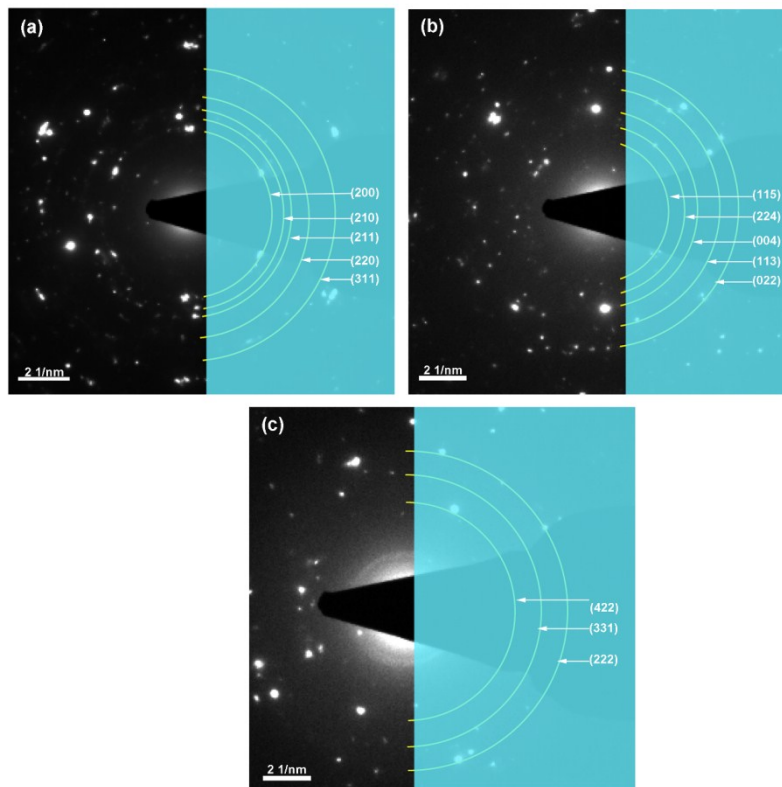


Fig. S5. the corresponding SAED pattern of (a) $\text{Cu}_{0.33}\text{Co}_{0.67}\text{S}_2$, (b) CuCo_2S_4 and (c) $\text{Cu}_3\text{Co}_6\text{S}_8$.

Fig. S6. CVs of (a) CuCo_2S_4 and (b) $\text{Cu}_3\text{Co}_6\text{S}_8$ at various scanning rates.

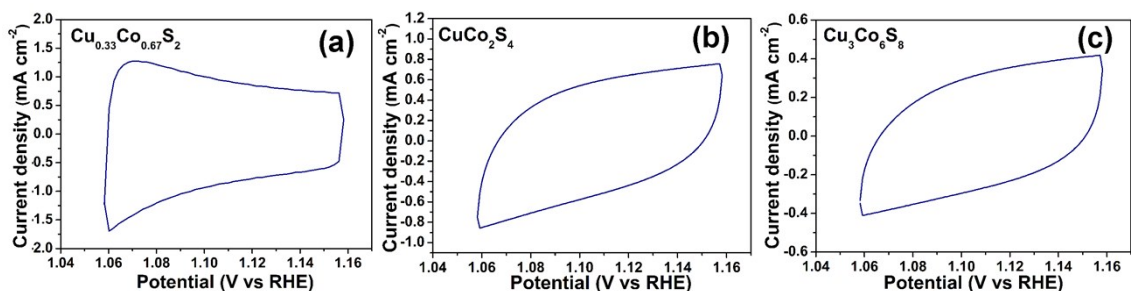


Fig.S7. Cyclic voltammograms of (a) $\text{Cu}_{0.33}\text{Co}_{0.67}\text{S}_2$, (b) CuCo_2S_4 and (c) $\text{Cu}_3\text{Co}_6\text{S}_8$ at 20 mV/s in 1 M KOH.

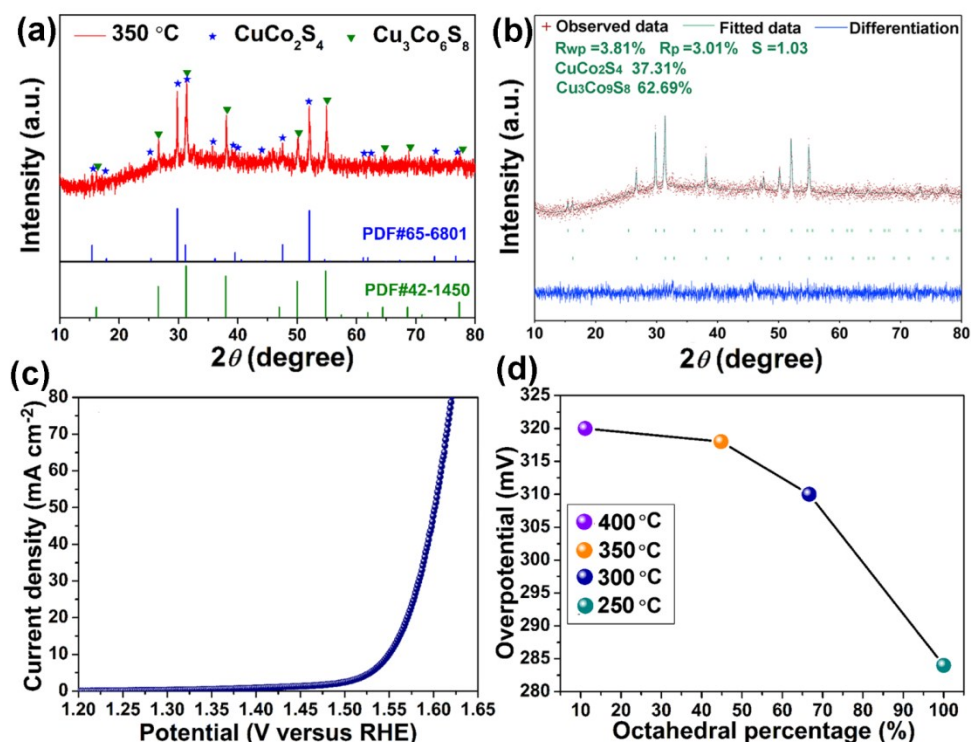


Fig.S8. (a) XRD pattern of the Cu-Co-S product at 350 °C. (b) Rietveld refinement of the XRD pattern of the Cu-Co-S product at 350°C. (c) LSV curve of the product at 350 °C. (d) The Relationship of the content of octahedron in products at different temperatures with their overpotential performance.

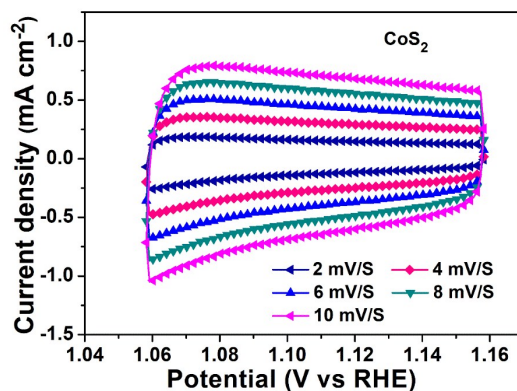


Fig. S9. CVs of CoS_2 at different scan rates.

Table. S1. Summary of the electrochemical activities of $\text{Cu}_{0.33}\text{Co}_{0.67}\text{S}_2$, CuCo_2S_4 , $\text{Cu}_3\text{Co}_6\text{S}_8$ and CoS_2 for OER.

| Catalyst | Overpotential@1 0 mA cm ⁻² (mV) | Tafel slope (mV dec ⁻¹) | C _{dl} (mF cm ⁻²) | TOF (s ⁻¹) | R _{ct} |
|--|--|--|---|--|-----------------|
| $\text{Cu}_{0.33}\text{Co}_{0.67}\text{S}_2$ | 284 | 86 | 76.32 | 13.512 s ⁻¹ @280 mV 19.814 s ⁻¹ @300 mV 32.045 s ⁻¹ @320 mV 49.940 s ⁻¹ @340 mV | 47 |
| CuCo_2S_4 | 310 | 90 | 31.26 | 6.701 s ⁻¹ @280 mV 9.799s ⁻¹ @300 mV 16.156s ⁻¹ @320 mV 27.518 s ⁻¹ @340 mV | 54 |
| $\text{Cu}_3\text{Co}_6\text{S}_8$ | 320 | 91 | 15.56 | 4.244 s ⁻¹ @280 mV 6.548 s ⁻¹ @300 mV 11.099 s ⁻¹ @320 mV 19.570 s ⁻¹ @340 mV | 72 |
| CoS_2 | 343 | 98 | 67.03 | 2.081 s ⁻¹ @280 mV 2.8112 s ⁻¹ @300 mV 4.905 s ⁻¹ @320 mV 9.638 s ⁻¹ @340 mV | 48 |

Table. S2. Rietveld refinement results for the XRD patterns of the $\text{Cu}_{0.33}\text{Co}_{0.67}\text{S}_2$ and CoS_2 .

| Sample | Phase | Space group | Lattice parameters | | | Amount (wt%) | S |
|--|----------------|-------------|--------------------|-------|-------|--------------|------|
| | | | a(Å) | b(Å) | c(Å) | | |
| CoS_2 | CoS_2 | <i>Pa-3</i> | 5.524 | 5.524 | 5.524 | 100 | 1.10 |
| $\text{Cu}_{0.33}\text{Co}_{0.67}\text{S}_2$ | CoS_2 | <i>Pa-3</i> | 5.638 | 5.638 | 5.638 | 100 | 1.05 |

Table. S3. Comparison of OER performances of $\text{Cu}_{0.33}\text{Co}_{0.67}\text{S}_2$ with other reported similar non-noble metal OER electrocatalysts.

| Material | Electrolyte (KOH) | Scan rate (mV s^{-1}) | η_{10} (mv) | Ref. |
|--|-------------------|----------------------------------|------------------|-----------|
| $\text{Co}_2\text{P@Co}_3\text{O}_4$ | 1.0 M | 5 | 335 | 1 |
| $\text{Co}_3\text{S}_4\text{@NCNTs}$ | 0.1 M | 5 | 430 | 2 |
| CoS_2 NTA/CC | 1.0 M | 10 | 276 | 3 |
| Co-doped Ni–Mn LDH | 1.0M | 10 | 310 | 4 |
| CoS_2 HNSs | 1.0 M | 10 | 290 | 5 |
| $\text{Co}_9\text{S}_8/\text{Zn}_{0.8}\text{Co}_{0.2}\text{S@C}$ | 0.1 M | 5 | 292 | 6 |
| $\text{Co}_9\text{S}_8\text{@NS-3DrGO}$ | 1.0 M | 5 | 317 | 7 |
| oxygenated- CoS_2 – MoS_2 | 1.0 M | 2 | 272 | 8 |
| $\text{Co}_x\text{S}_y\text{@C}$ | 0.1 M | 5 | 470 | 9 |
| HPMS $\text{Co}_3\text{O}_4/\text{CoS}_2$ | 1.0 M | 2 | 280 | 10 |
| $\text{Zn}_x\text{Co}_{3-x}\text{O}_4$ | 1.0 M | 10 | 435 | 11 |
| Co_3O_4 Nanoflakes | 1.0M | 5 | 451 | 12 |
| $\text{CoSe}_2\text{@C}$ | 1.0 M | 10 | 330 | 13 |
| $\text{CoSe}_2\text{@C-CNT}$ | 1.0 M | 5 | 306 | 14 |
| Zn-Doped CoSe_2 | 1.0 M | 2 | 356 | 15 |
| CoFeP NSs | 1.0 M | 5 | 305 | 16 |
| octahedral Co_3O_4 particles | 1.0 M | 2 | 301 | 17 |
| $\text{Co}_3\text{O}_4\text{@rGO}$ | 1.0 M | 2 | 313 | 18 |
| $\text{CoS}_2\text{@N,S-GO}$ | 0.1 M | 10 | 390 | 19 |
| $\text{Cu}_{0.33}\text{Co}_{0.67}\text{S}_2$ | 1 M | 10 | 284 | This work |

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