

## Supporting Information

### In Situ Transformed CoOOH@Co<sub>3</sub>S<sub>4</sub> Heterostructured Catalyst for Highly Efficient Catalytic OER Application

Abu Talha Aqueel Ahmed <sup>1</sup>, Vijaya Gopalan Sree <sup>2</sup>, Abhishek Meena <sup>1</sup>, Akbar I. Inamdar <sup>1</sup>, Hyunsik Im <sup>1</sup>  
and Sangeun Cho <sup>1\*</sup>

<sup>1</sup> Division of System Semiconductor, Dongguk University, Seoul 04620, Republic of Korea

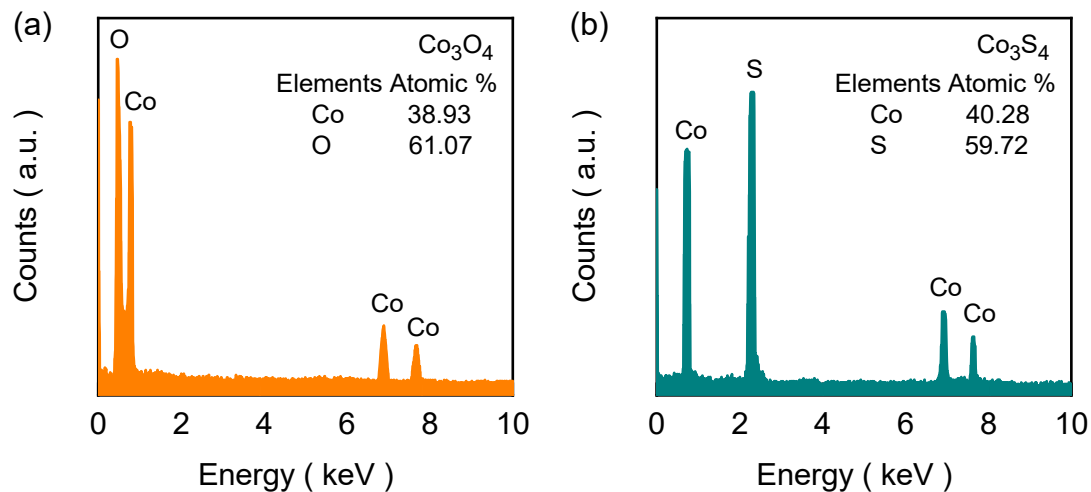
<sup>2</sup> Department of Physics, Dongguk University, Seoul 04620, Republic of Korea

**Corresponding Author:** sangeun.c@dongguk.edu

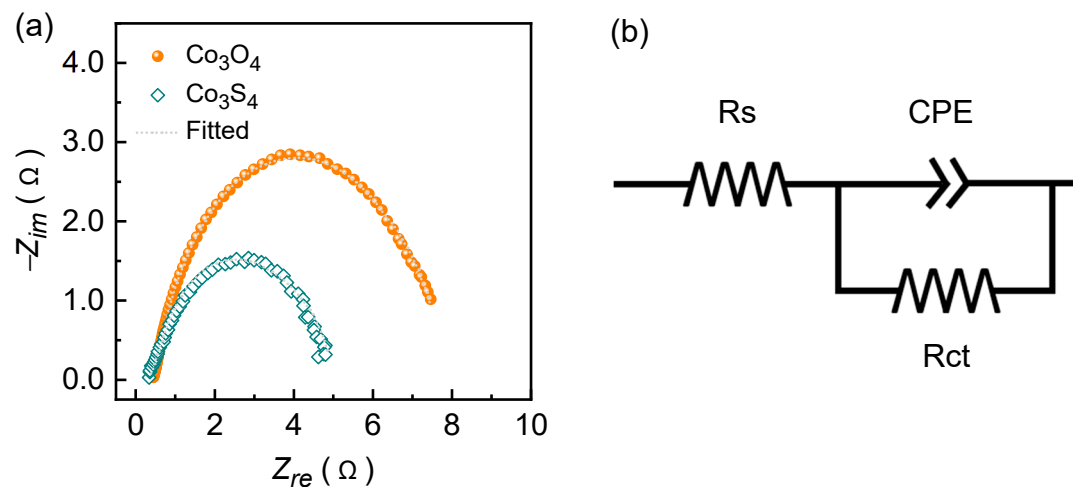
Table S1. The electrocatalytic OER performance of our optimized Co<sub>3</sub>S<sub>4</sub> catalyst films and other metal sulfide-based catalyst in alkaline electrolyte medium at 100 mA cm<sup>-2</sup>.

No.	Catalyst film	Overpotential @100 (mA cm <sup>-2</sup> )	Tafel slope (mV dec <sup>-1</sup> )	Stability at <i>J</i> ( <i>J</i> in mA cm <sup>-2</sup> )	Supporting Reference
1	Ni-Fe-OH@Ni <sub>3</sub> S <sub>2</sub> /NF	165 mV	93	50 h@100	[55]
2	Ni <sub>50</sub> Fe <sub>50</sub> -DAT	300 mV	–	72 h@100	[56]
3	NiFeSedO-NiNF	247 mV	54	~8 h@100	[57]
4	Fe <sup>0</sup> -Ni <sub>x</sub> S <sub>y</sub> /NF	186 mV	92	10 h@1.47 V	[58]
5	CFP NPs	294 mV	50	70 h@100	[59]
6	Ni <sub>3</sub> Fe(OH) <sub>9</sub> /NF	370 mV	28	10 h@100	[60]
7	CoS/MoS <sub>2</sub> heterostructure	294 mV	31	100 h@100	[61]
8	NiFe-NF	290 mV	50.1	30 h@100	[62]
9	Polyhedron-like Co <sub>3</sub> S <sub>4</sub>	292 mV	103	101	Present Work

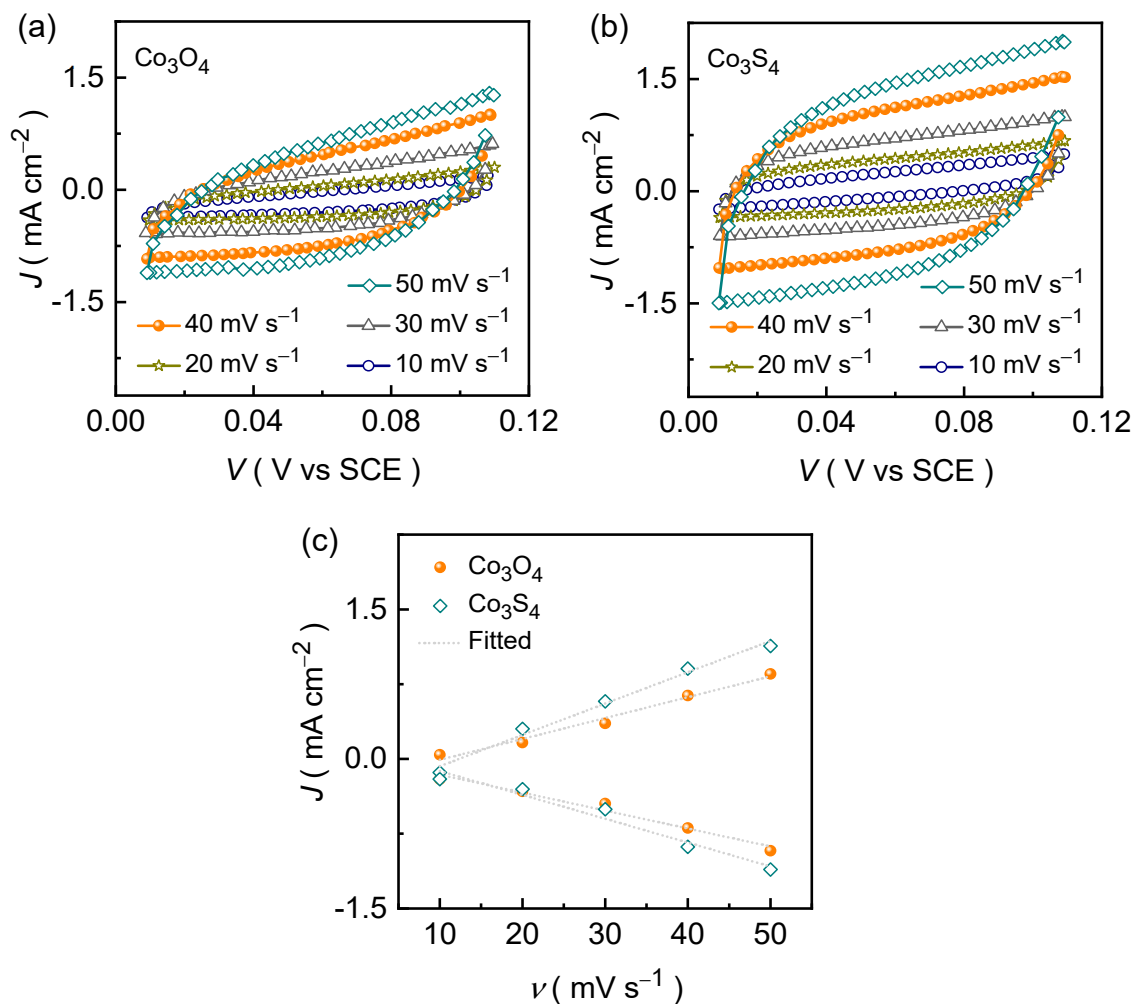
## Supporting Figures



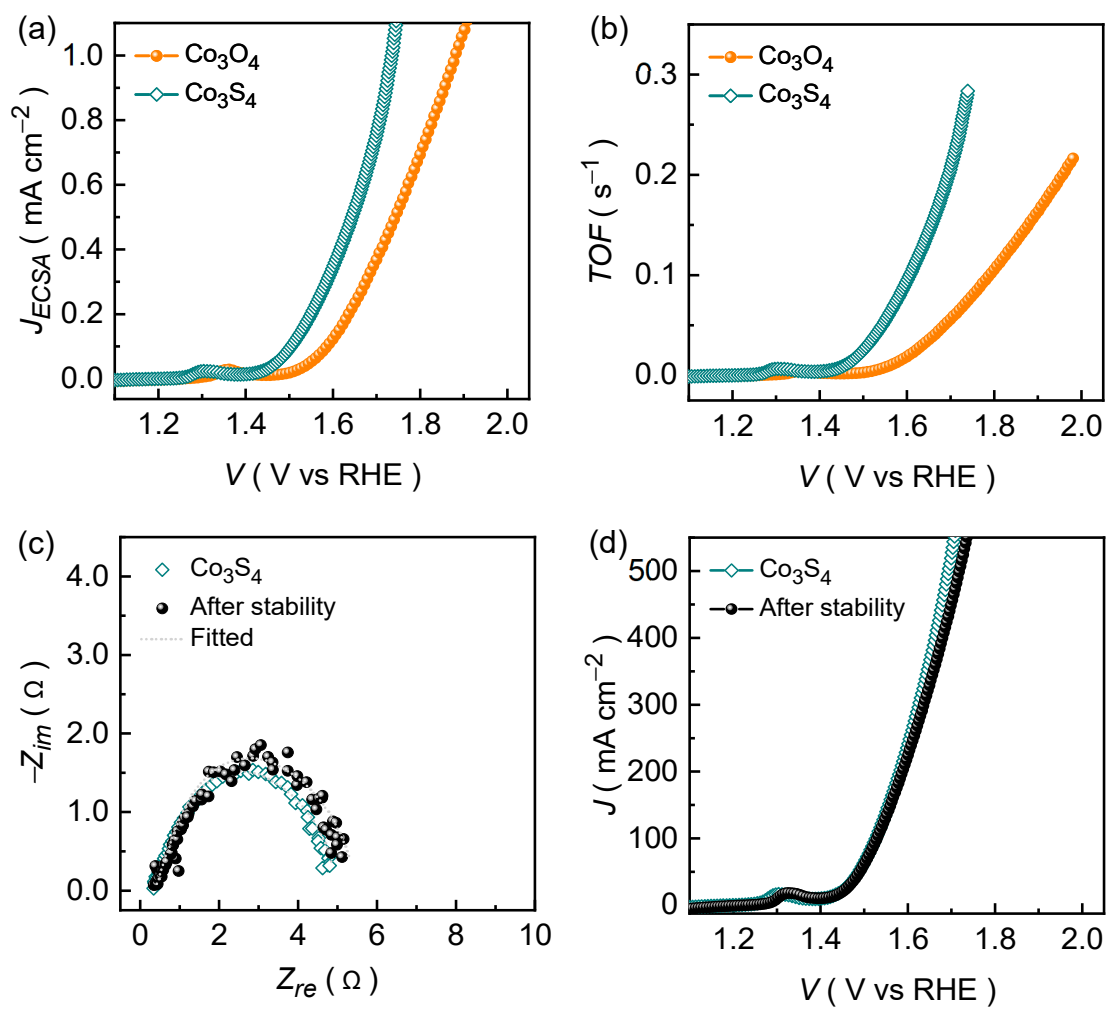
**Figure S1.** (a) EDX spectra of (a)  $\text{Co}_3\text{O}_4$  and (b)  $\text{Co}_3\text{S}_4$  electrode films. Notably, the inset table shows the obtained elemental compositions in the atomic percentage ratios.



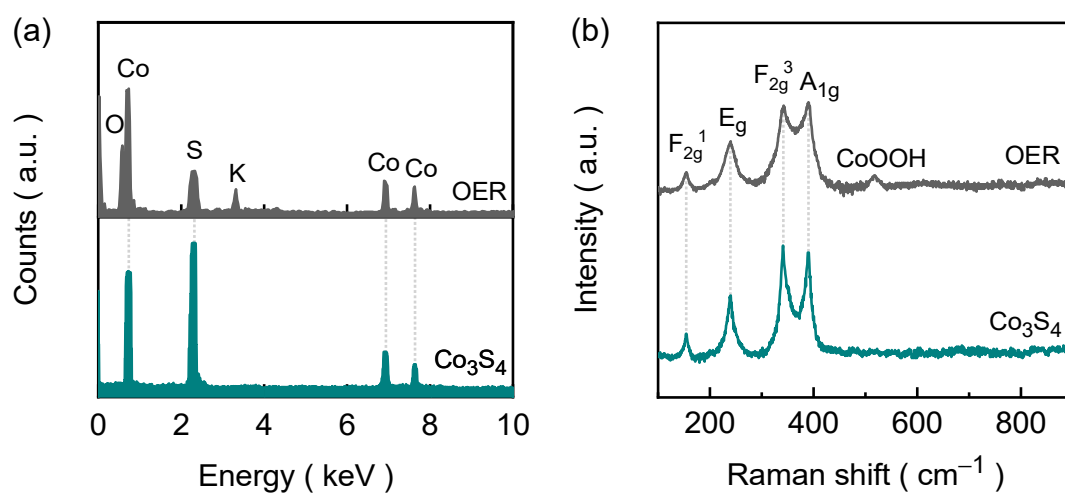
**Figure S2.** (a) EIS curves recorded for the  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{S}_4$  electrode films along with the fitting curves date marked with the grey dotted lines. (b) The formed tank-circuit used to fit the EIS curves in Z-view software.



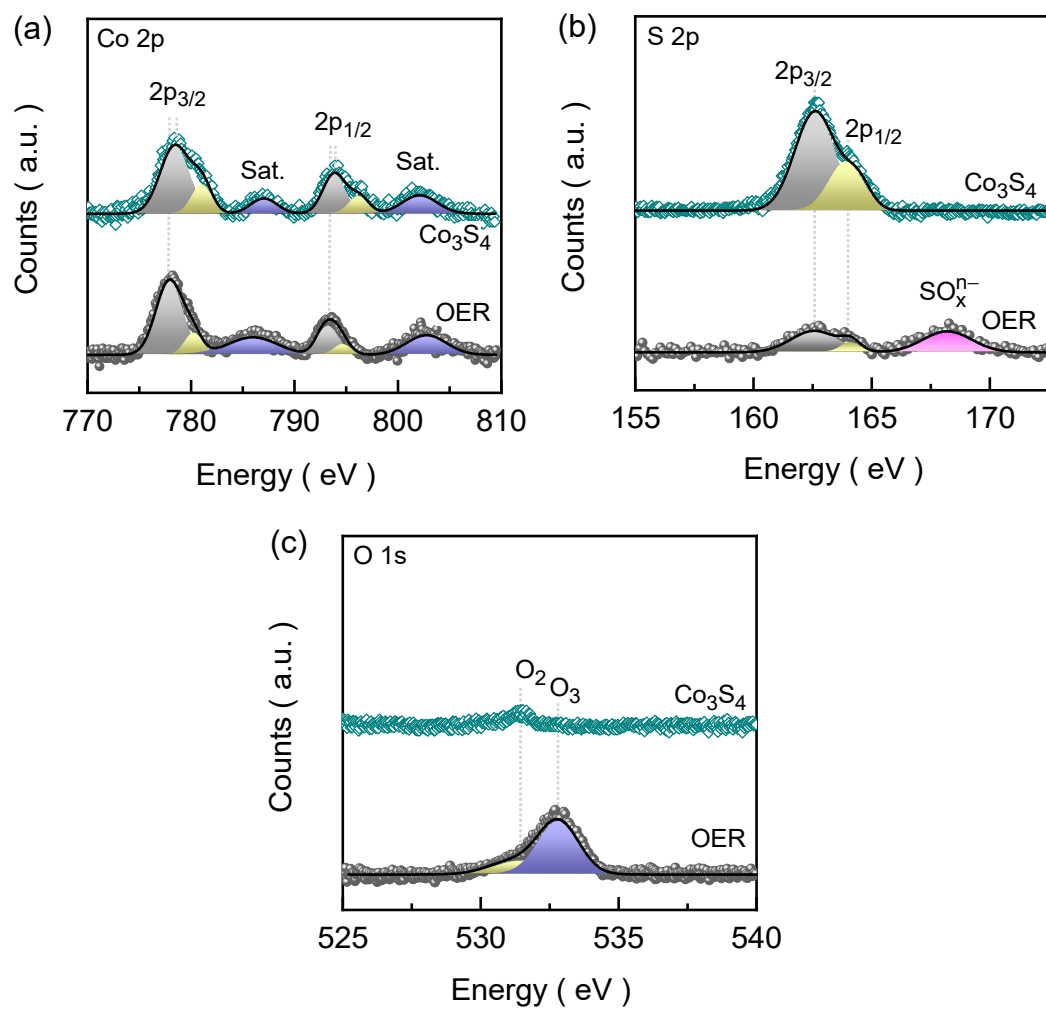
**Figure S3.** Scan rate dependent CV curves of the (a)  $\text{Co}_3\text{O}_4$  and (b)  $\text{Co}_3\text{S}_4$  catalyst films measured in non-Faradaic potential region at different scan rates. (c) “ $J$  versus  $\nu$ ” plots obtained at 0.06 V (*vs.* SCE) from non-Faradaic CV curves to calculate the double-layer capacitance and ECSA.



**Figure S4.** (a) ECSA-compensated LSV curves and (b) TOF curves of  $\text{Co}_3\text{O}_4$  and  $\text{Co}_3\text{S}_4$  catalyst films. Post-stability measured (c) EIS and (d) LSV curves of the proposed  $\text{Co}_3\text{S}_4$  catalyst film.



**Figure S5.** (a) EDX and (b) Raman spectra of Co<sub>3</sub>S<sub>4</sub> catalyst film measured after the prolonged chronopotentiometric stability test.



**Figure S6.** High-resolution (a) Co 2p, (b) S 2p, and (c) O 1s XPS emission spectra of  $\text{Co}_3\text{S}_4$  catalyst film measured after the prolonged chronopotentiometric stability test.

## Supporting References

55. Zou, X.; Liu, Y.; Li, G.-D.; Wu, Y.; Liu, D.-P.; Li, W.; Li, H.-W.; Wang, D.; Zhang, Y.; Zou, X. Ultrafast Formation of Amorphous Bimetallic Hydroxide Films on 3D Conductive Sulfide Nanoarrays for Large-Current-Density Oxygen Evolution Electrocatalysis. *Adv. Mater.* **2017**, *29*, 1700404.
56. Hoang, T.T.H.; Gewirth, A.A. High Activity Oxygen Evolution Reaction Catalysts from Additive-Controlled Electrodeposited Ni and NiFe Films. *ACS Catal.* **2016**, *6*, 1159–1164.
57. Peugeot, A.; Creissen, C.E.; Karapinar, D.; Tran, H.N.; Schreiber, M.; Fontecave, M. Benchmarking of oxygen evolution catalysts on porous nickel supports. *Joule* **2021**, *5*, 1281–1300.
58. Cheng, X.; Lei, C.; Yang, J.; Yang, B.; Li, Z.; Lu, J.; Zhang, X.; Lei, L.; Hou, Y.; Ostrikov, K. Efficient Electrocatalytic Oxygen Evolution at Extremely High Current Density over 3D Ultrasmall Zero-Valent Iron-Coupled Nickel Sulfide Nanosheets. *ChemElectroChem* **2018**, *5*, 3866–3872.
59. Meena, A.; Aqueel, A.A.T.; Narayan, S.A.; Gopalan, S.V.; Im, H.; Cho, S. Highly Efficient CoFeP Nanoparticle Catalysts for Superior Oxygen Evolution Reaction Performance. *Nanomaterials* **2024**, *14*, 1384.
60. Lu, X.; Zhao, C. Electrodeposition of hierarchically structured three-dimensional nickel–iron electrodes for efficient oxygen evolution at high current densities. *Nat. Commun.* **2015**, *6*, 6616.
61. Ahmed, A.T.A.; Lee, C.H.; Ansari, A.S.; Pawar, S.M.; Han, J.; Park, S.; Shin, G.; Yeon, S.; Cho, S.; Seol, J.; et al. Hybridized heterostructure of CoS and MoS<sub>2</sub> nanoparticles for highly-efficient and robust bifunctional water electrolysis. *Appl. Surf. Sci.* **2022**, *592*, 153196.
62. Guo, D.; Qi, J.; Zhang, W.; Cao, R. Surface Electrochemical Modification of a Nickel Substrate to Prepare a NiFe-based Electrode for Water Oxidation. *ChemSusChem* **2017**, *10*, 394–400.