

## Supporting Information

### **Self-assembled CoSe<sub>2</sub>-FeSe<sub>2</sub> Heteronanoparticles along the Carbon Nanotubes Network for Boosted Oxygen Evolution Reaction**

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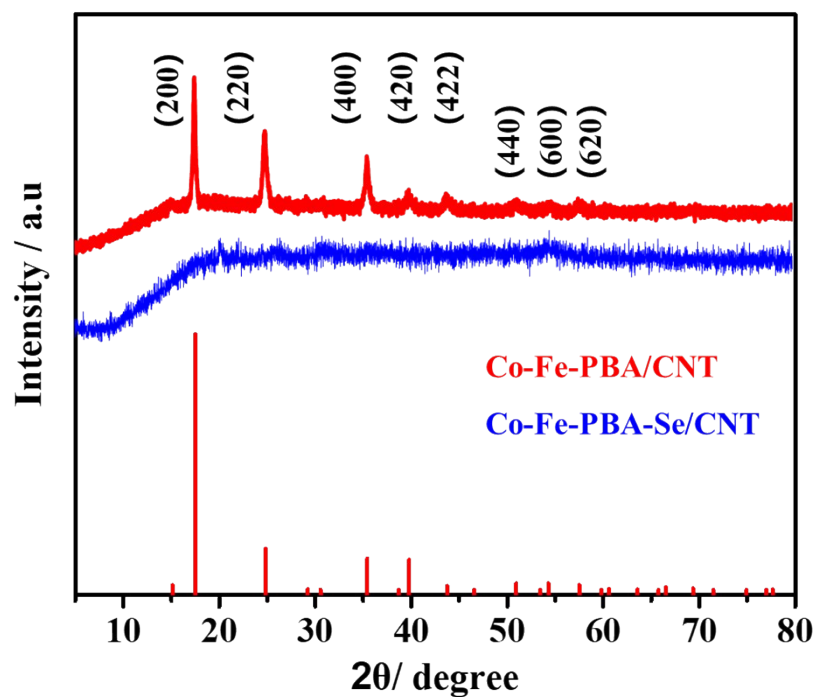
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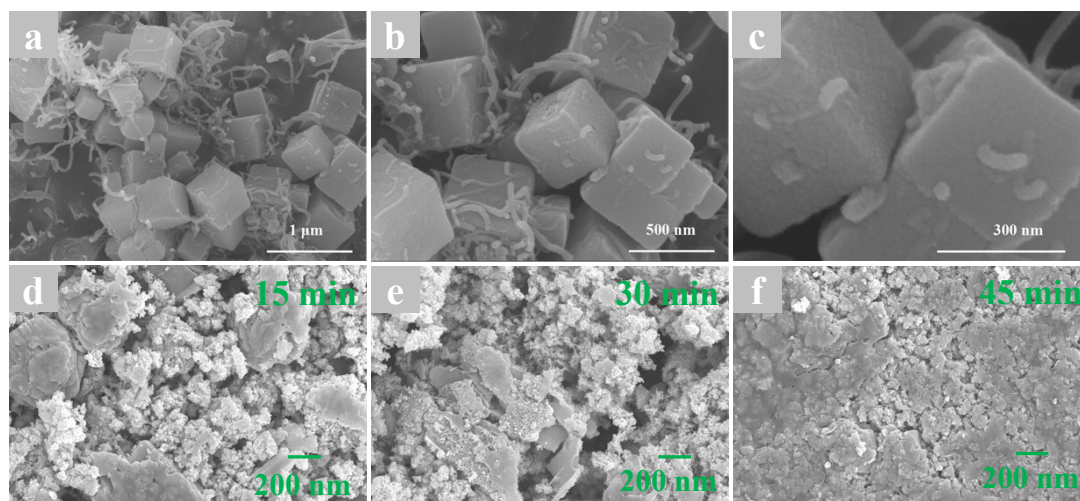
## Synthesis of precursors

**Preparation of Co-Fe-PBA/CNT:** 1 mg/mL CNT pulp suspension was first prepared by ultrasonication (1 h) of 6 g multi-walled CNT paste in 300 mL of deionized water. Subsequently, 9 mmol cobalt(II) nitrate hexahydrate (2.62 g) and 13.5 mmol trisodium citrate dihydrate (3.97 g) were dissolved in a sequence to form solution **A**. Similarly, solution **B** was prepared by dissolving 6 mmol potassium hexacyanoferrate(II) (1.975 g) in 300 ml of deionized water. The as-prepared solution **B** was slowly added to solution **A** under continuous stirring at ambient temperature and the resulting mixture was allowed stir for additional 15 minutes. Thereafter, the attained mixed suspension was aged for 24 h at room temperature and then collected by centrifugation by washing with DI water and ethanol several times. The resulting sample was dried at 65 °C overnight and denoted as Co-Fe-PBA/CNT. Similar, methodology was used in fabricating Co-Co-PBA/CNT only by replacing potassium hexacyanoferrate(II) with Potassium hexacyanocobaltate(II). Similarly, Fe-Fe-PBA/CNT was prepared by replacing cobalt(II) nitrate hexahydrate with ferric nitrate(III) nonahydrate.

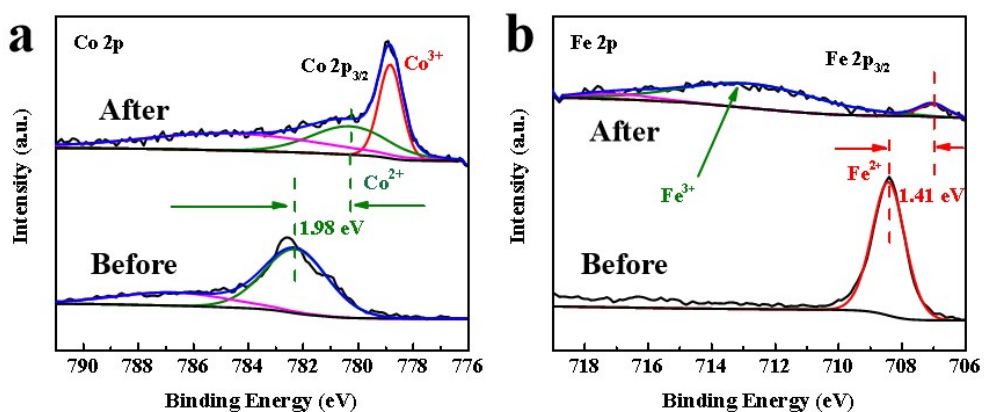
**Preparation of Co-Fe-PBA-Se/CNT:** 0.2 g of Co-Fe-PBA/CNT was dispersed in 60 ml ethylene glycol and allowed to stir at room temperature (30 min) in order to obtain a homogenous suspension. Then, controlled amounts of sodium selenite (1.13 g) was added into the homogenous suspension and stirred for additional 30 min. The resulting suspension was transferred into a three-neck flask and subjected to microwave assisted heating (120 °C) for 45 minutes. After cooling down naturally, the as-obtained precipitate was collected by centrifugation by washing with methanol and ethanol alternatively for several times. The resulting product was dried at 65 °C overnight and denoted as Co-Fe-PBA-Se/CNT.



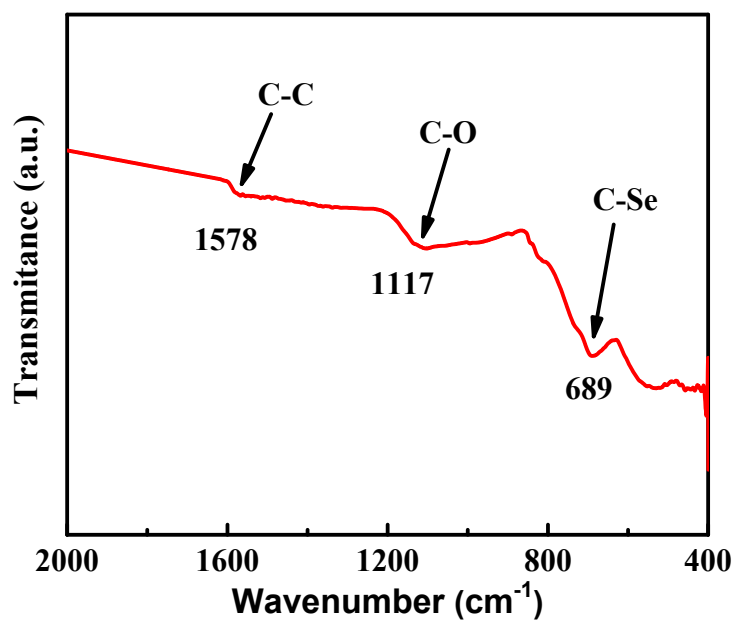
**Fig. S1** XRD pattern of Co-Fe-PBA/CNT and Co-Fe-PBA-Se/CNT.



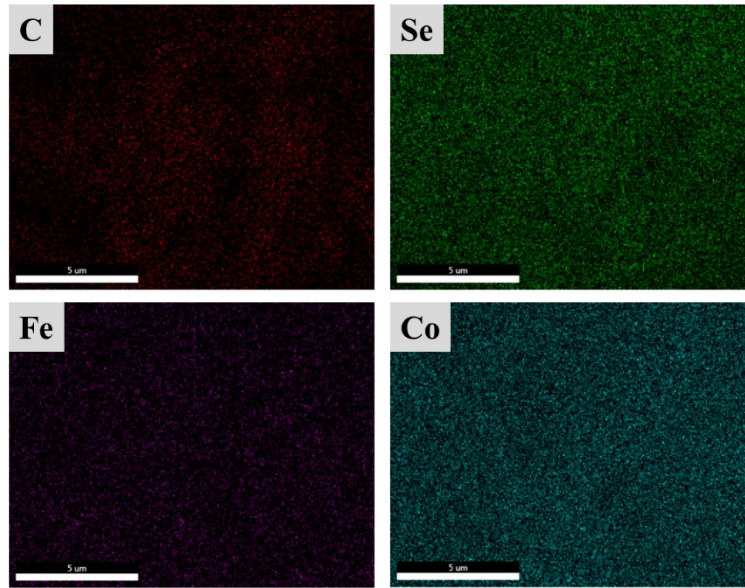
**Fig. S2** SEM image of Co-Fe-PBA/CNT (a-c), Co-Fe-PBA-Se/CNT (d-f) with different reaction time (15, 30 and 45 min, respectively).



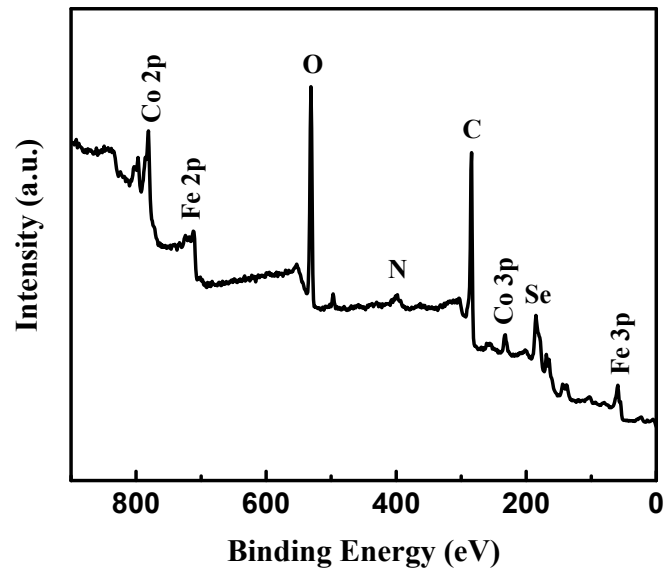
**Fig. S3** The high-resolution Co 2p (a), and Fe 2p (b) core-level XPS spectra of Co-Fe-PBA/CNT (**Before**) and Co-Fe-PBA-Se/CNT (**After**).



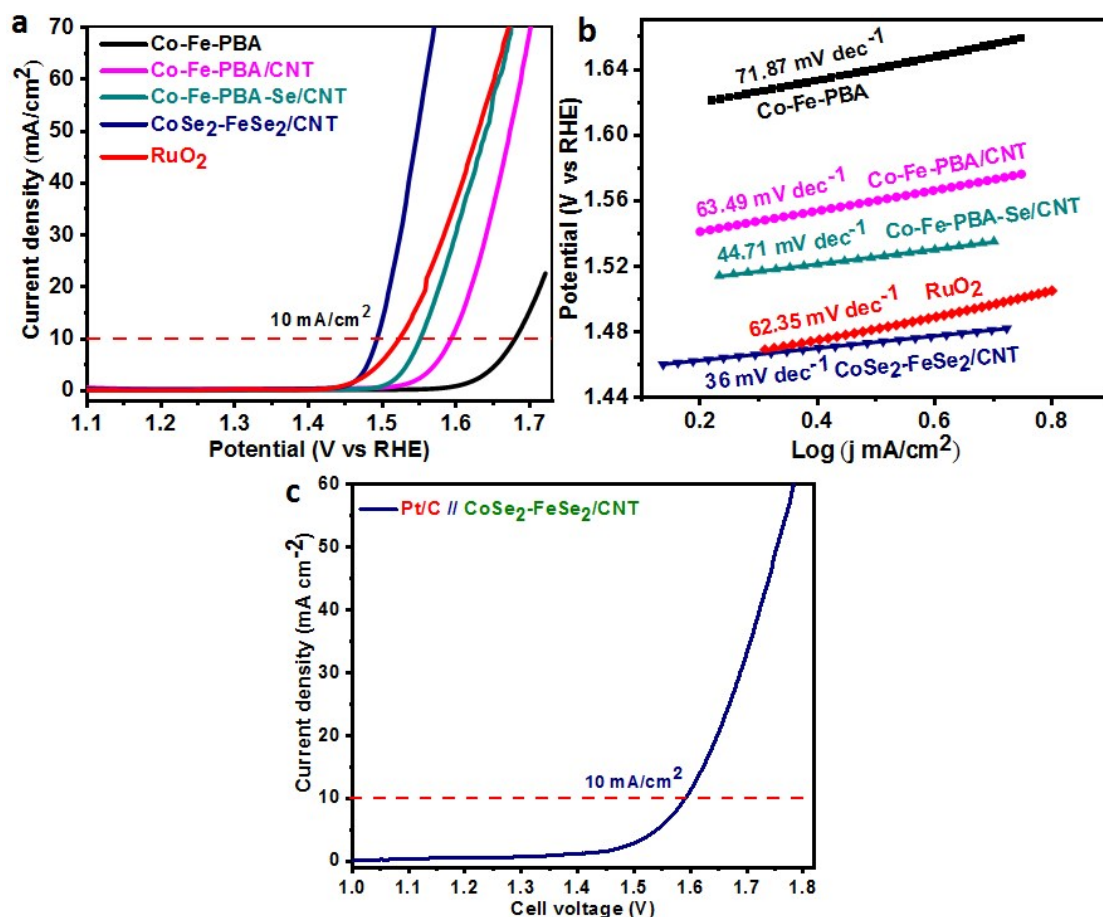
**Fig. S4** FTIR spectrum of CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT.



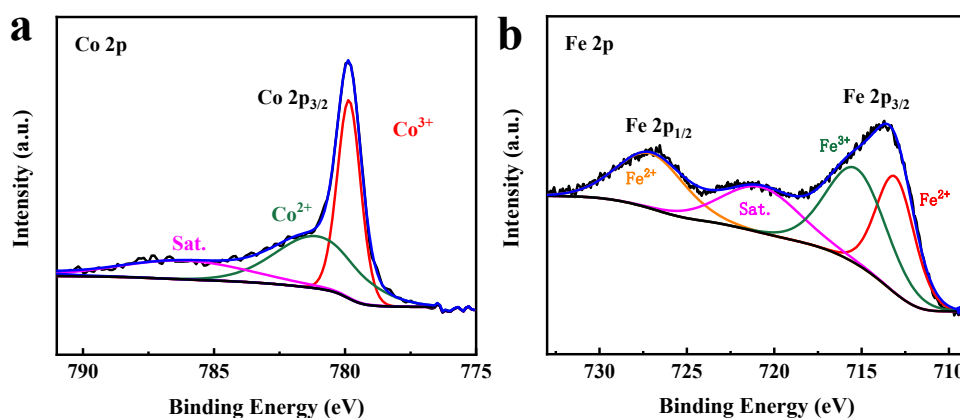
**Fig. S5** SEM mapping of element C, Se, Fe and Co in CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT.



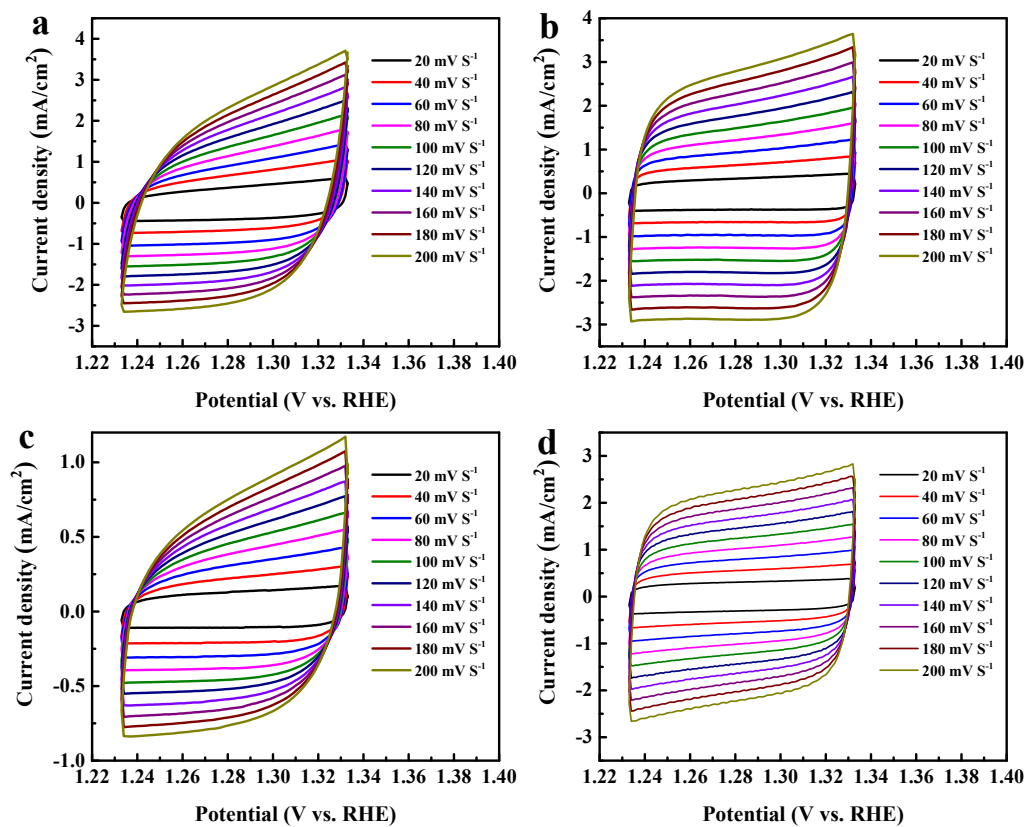
**Fig. S6** XPS survey scan of CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT.



**Fig. S7** LSV curves and corresponding Tafel plots of Co-Fe-PBA, Co-Fe-PBA/CNT, Co-Fe-PBA-Se/CNT, CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT and commercial RuO<sub>2</sub> electrocatalysts in 1.0 M KOH electrolyte (a,b), and overall water-splitting ability of CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT (anode) with state-of-the-art Pt/C (cathode) (c).



**Fig. S8** Core-level XPS spectra of Co 2p (a), and Fe 2p (b) for the CoSe<sub>2</sub>/CNT and FeSe<sub>2</sub>/CNT samples, respectively.



**Fig. S9** Voltammograms of CoSe<sub>2</sub>/CNT (a), FeSe<sub>2</sub>/CNT (b), CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT (c) and CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT (d) for OER in alkaline electrolyte.



**Calculation of Faradaic Efficiency:** In general, the amount of oxygen (O<sub>2</sub>) evolved can be estimated from the obtained gas volume in a water-drainage set-up by applying the ideal gas law. While, the theoretical quantity of O<sub>2</sub> can be calculated through Faraday law based on the fixed current. Therefore, by using this data one can evaluate the Faradic efficiency by the following equation;

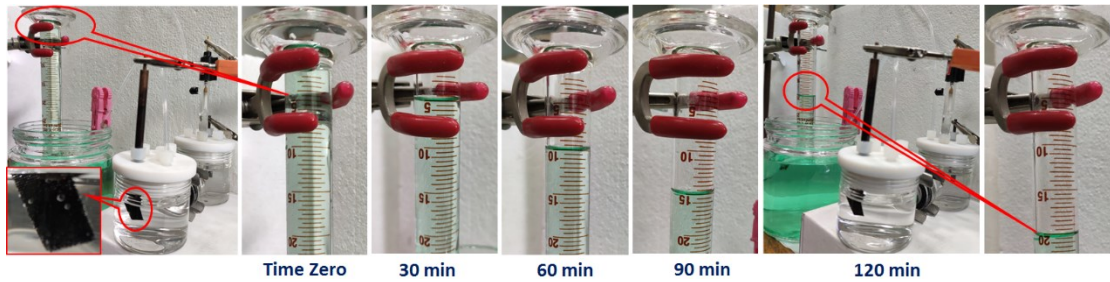
$$\text{Faradaic Efficiency} = \frac{V/V_m}{n_{O_2}} \times 100 \quad \dots\dots\dots S_1$$

Where, ‘V’ is the volume of gas experimentally evolved during the reaction, ‘V<sub>m</sub>’ is the molar volume (22.4 L mol<sup>-1</sup>, at STP) and ‘n<sub>O<sub>2</sub></sub>’ is the theoretical number of moles of O<sub>2</sub> produced.

The theoretical number of moles of O<sub>2</sub> evolved can be obtained by the following equation;

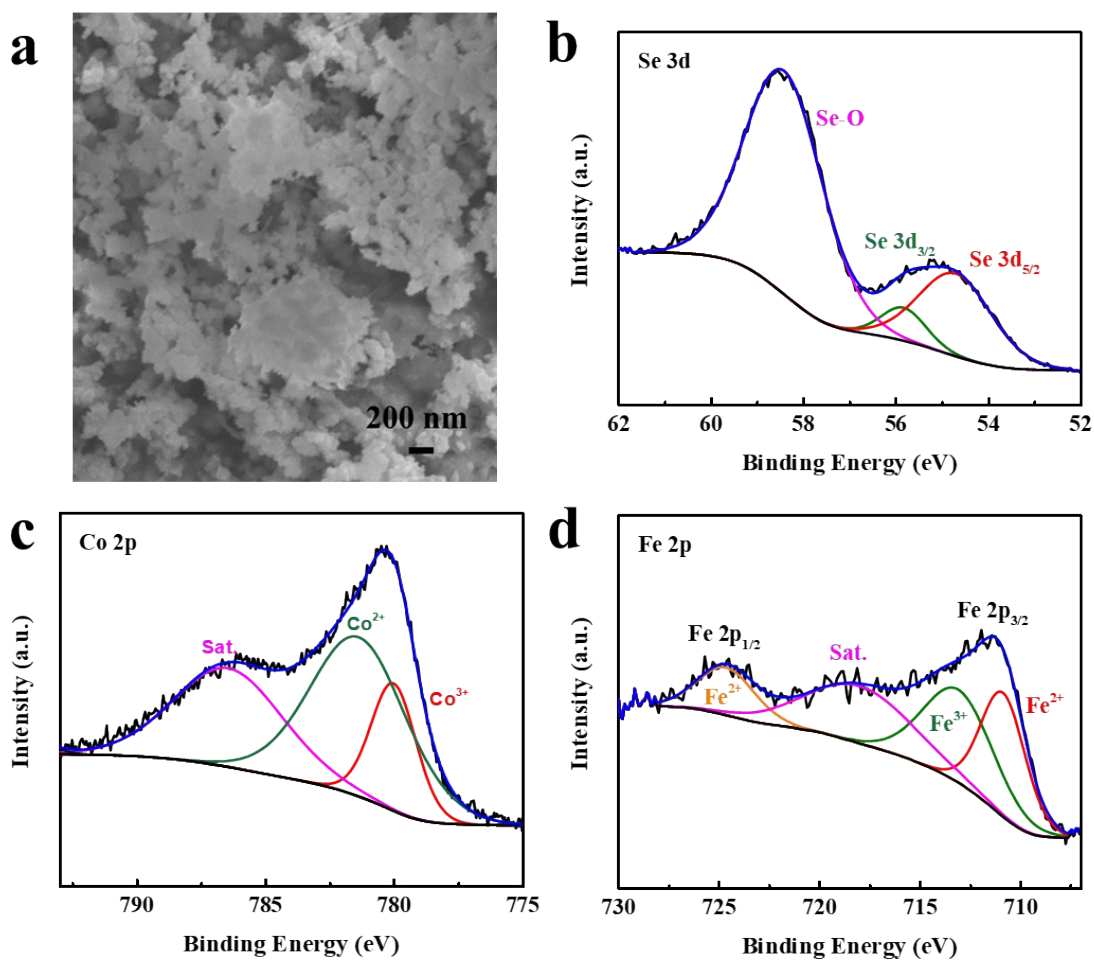
$$n_{O_2} = \frac{Q}{nF} \quad \dots\dots\dots S_2$$

Where, ‘Q’ is the total quantity of electric charge (C), ‘F’ is the Faradaic constant (96485 C mol<sup>-1</sup>), and ‘n’ is the number of electrons transferred during the process (OER; 4 moles of electrons per mole of O<sub>2</sub>)

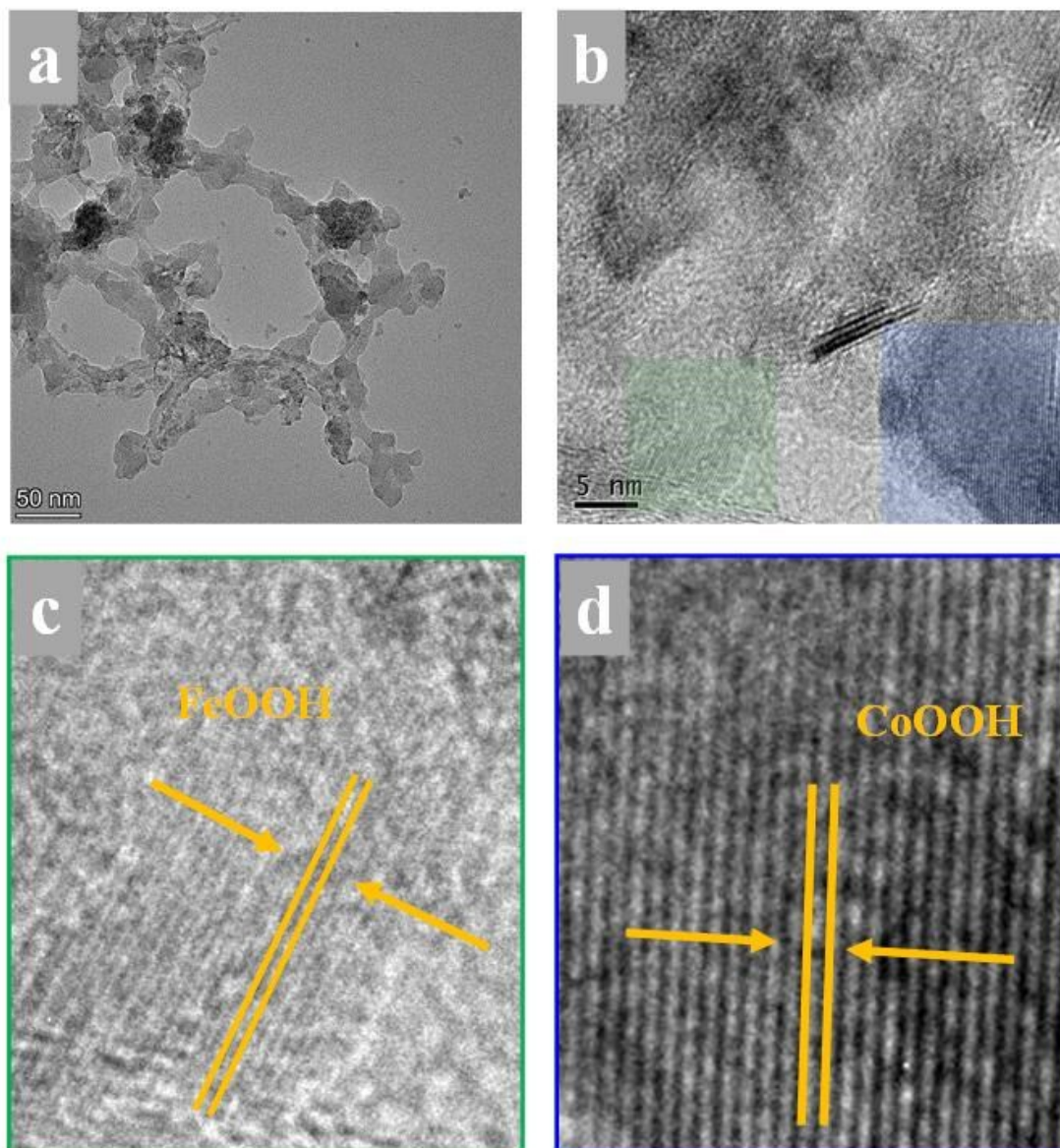


**Fig. S10** Water oxidation process of the electrolyzer derived from CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT material in 1.0 M KOH at a constant current density of 50 mA cm<sup>-2</sup> for 2 h (photographs of lab-made gas collectors at the beginning (Time = Zero minutes) and up to 2 h with 30 minutes interval).





**Fig. S11** The SEM image (a), detailed Se 3d (b), Co 2p (c), and Fe 2p (d) XPS core-level spectra of CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT after 20 h *i-t* test in 1.0 M KOH electrolyte.



**Fig. S12** TEM (a), HRTEM images of CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT after the chronoamperometry test (b-d).

**Table S1.** Comparative overpotential and Tafel slope values of TMDs materials fabricated recently with CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT.

<b>Materials</b>	<b><math>\eta</math> mV@10 mA/cm<sup>2</sup></b>	<b>Tafel slope (mV/ dec)</b>	<b>(<i>i-t</i>) (h)</b>	<b>Ref.</b>
<b>CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT</b>	<b>248 (OER)</b> <b>1.59 V (OWS)</b>	<b>36.0</b>	<b>20</b>	<b>This work</b>
CoSe <sub>2</sub> /FeSe <sub>2</sub> @C	291	62.0	~25	<b>1</b>
CoSe@FeSe <sub>2</sub>	281	34.3	12	<b>2</b>
FeSe <sub>2</sub> @CoSe <sub>2</sub> /rGO	260	38.2	6	<b>3</b>
CoSe <sub>2</sub> /FeSe <sub>2</sub> DS-HNCs	240	44	50	<b>4</b>
CoSe <sub>2</sub>	320	40.1	15	<b>5</b>
Co <sub>0.75</sub> Fe <sub>0.25</sub> -Se/NF	246	41.4	40	<b>6</b>
FeCoMo-Se	264	33	100	<b>7</b>
O-CoSe <sub>2</sub> -HNT	252	62	35	<b>8</b>
CoSe <sub>2</sub> @NC	310	59	-	<b>9</b>
FeCoSe <sub>2</sub> /Co <sub>0.85</sub> Se	330	50.8	12	<b>10</b>
(Ni,Co)Se <sub>2</sub>	278	65	20	<b>11</b>
Co <sub>3</sub> Se <sub>4</sub> /FeSe <sub>2</sub>	280	51	24	<b>12</b>

C = carbon nanorod; DS-HNCs = double-shelled hollow nanocuboids; rGO = reduced Graphene Oxide; NF = Nickel Foam; O- = Oxygen rich; HNT = hierarchical nanotubes; NC = N-Doped-carbon.

**Table S2.** The comparison of the XPS characteristic peaks of CoSe<sub>2</sub>-FeSe<sub>2</sub>/CNT, CoSe<sub>2</sub>/CNT and FeSe<sub>2</sub>/CNT.

CoSe <sub>2</sub> -FeSe <sub>2</sub> /CNT		CoSe <sub>2</sub> -FeSe <sub>2</sub> /CNT	
778.91	781.39	713.58	716.61
Co-Se	Co <sup>2+</sup>	Fe <sup>2+</sup>	Fe <sup>3+</sup>
779.84	781.60	713.20	715.56
CoSe <sub>2</sub> /CNT		FeSe <sub>2</sub> /CNT	

Note: the values in the table are unified in eV.

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