

## Supporting Information

### High-Entropy FeCoMnCuNi diselenide Self-Standing Electrode with Outstanding Water-Electrolysis Performance in Alkaline Medium

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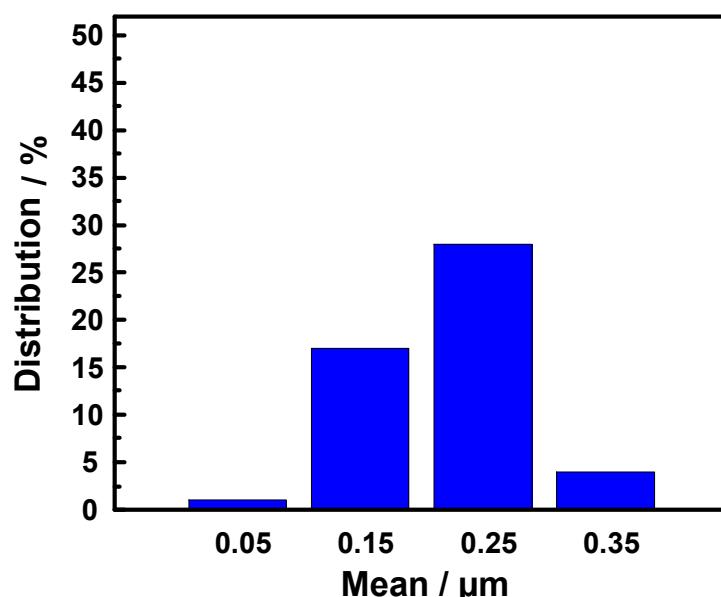
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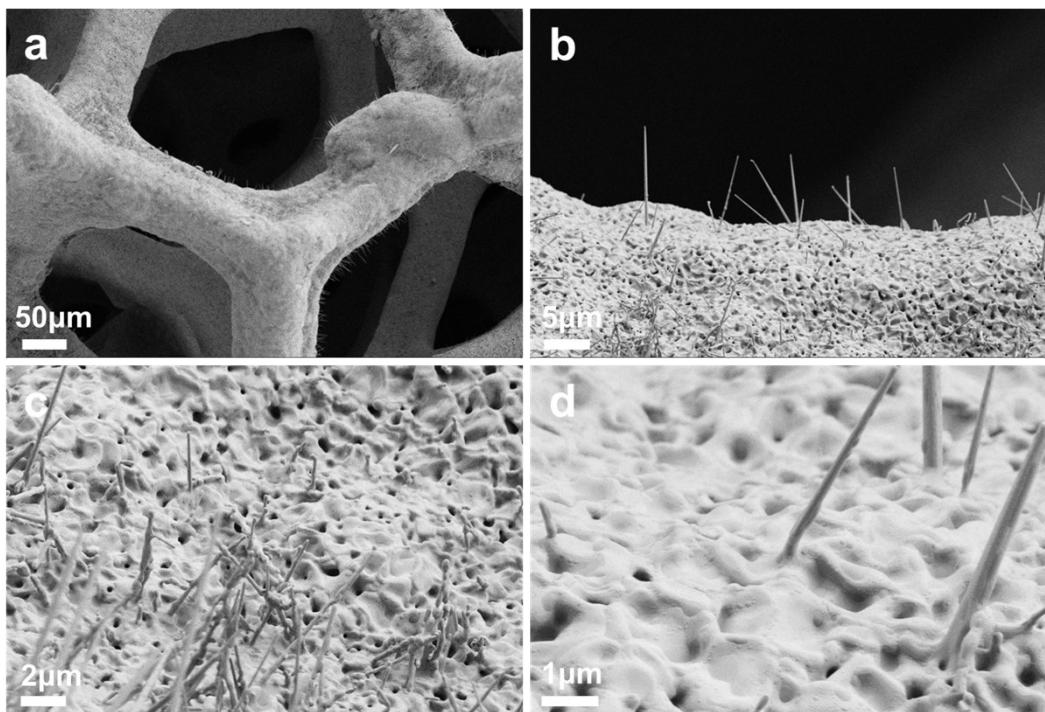
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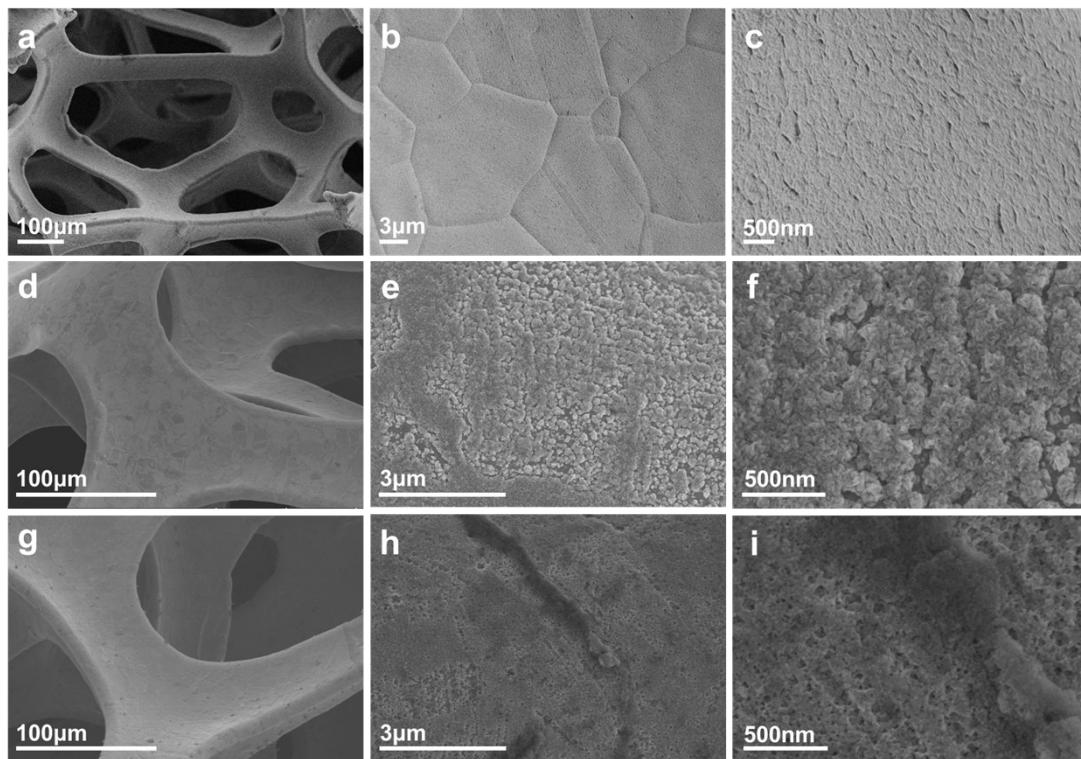


**Fig. S1** The thickness-distribution histograms of petunia leaves on the surface of

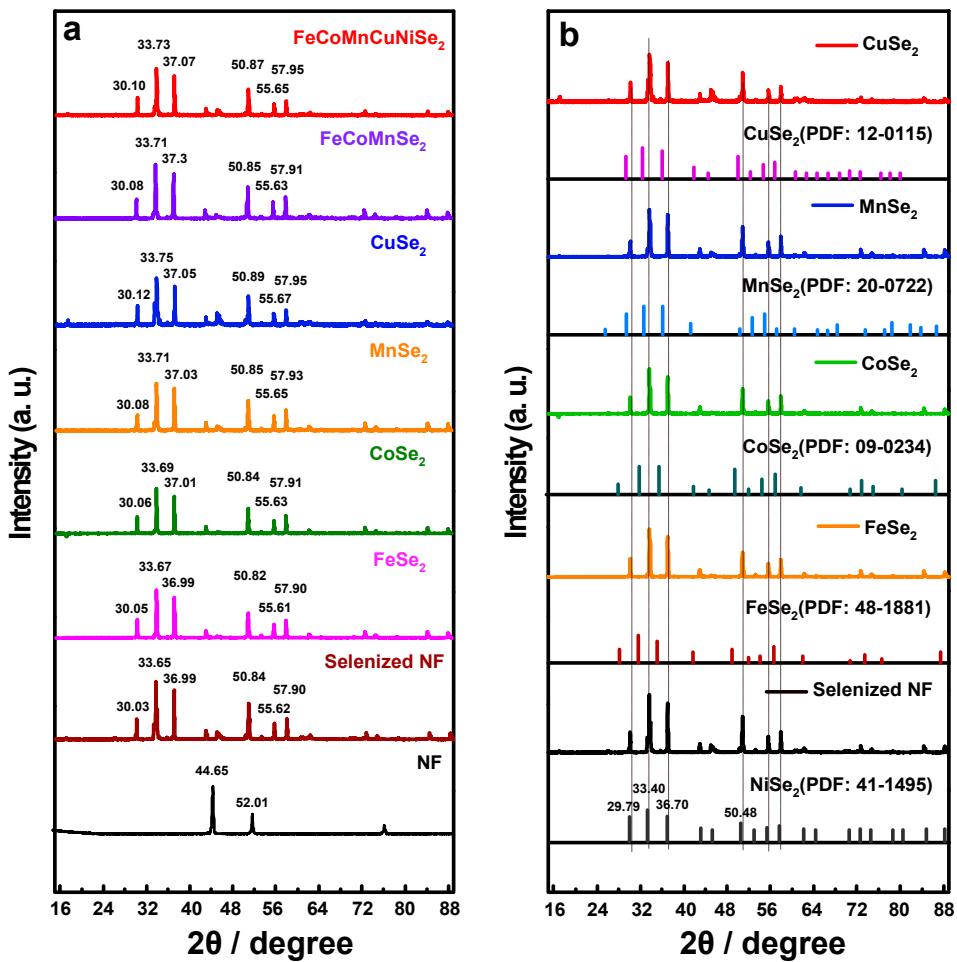
FeCoMnCuNiSe<sub>2</sub>.



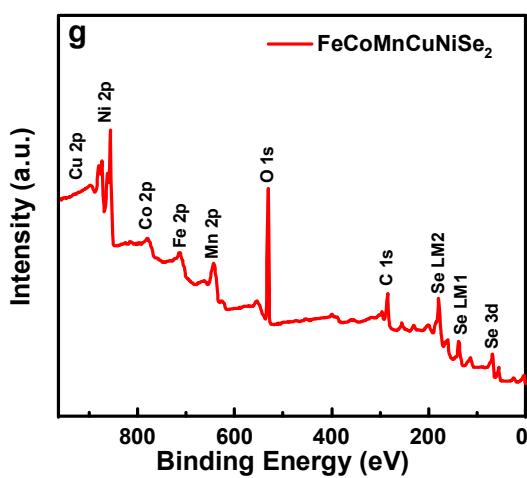
**Fig. S2** The SEM images of selenized bare NF (a-d).



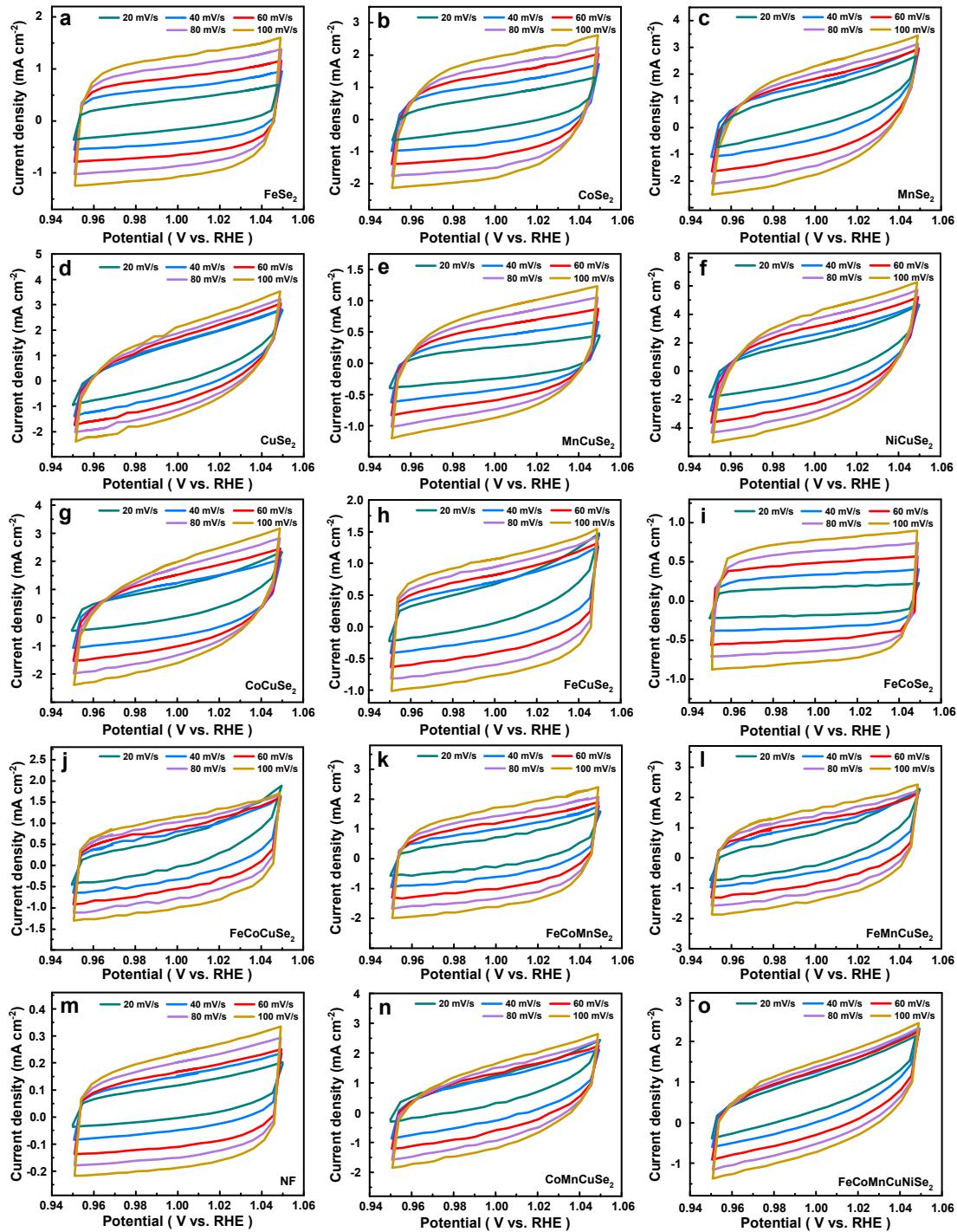
**Fig. S3** The SEM images of bare NF (a, b, c), bare NF heated at 450 °C for 3 h with argon protection (d, e, f) and FeCoMnCuOOH before selenization (g, h, i).



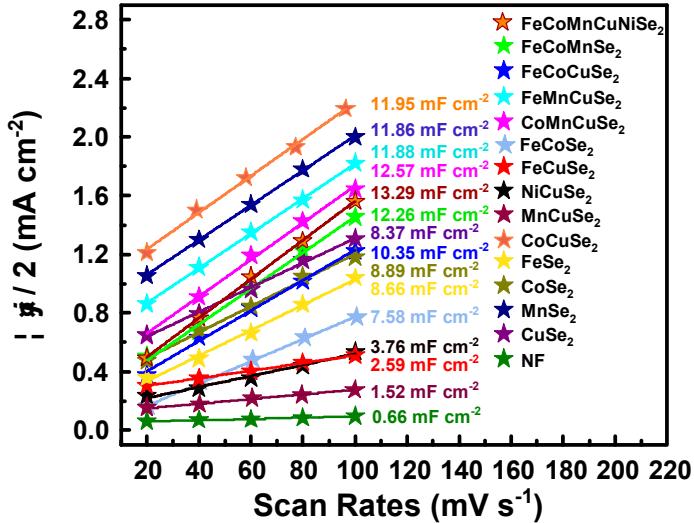
**Fig. S4** XRD patterns of FeCoMnCuNiSe<sub>2</sub>, FeCoMnSe<sub>2</sub>, CuNiSe<sub>2</sub>, FeSe<sub>2</sub>, CoSe<sub>2</sub>, MnSe<sub>2</sub>, CuSe<sub>2</sub>, NFSe<sub>2</sub> and NF (a); XRD patterns of FeSe<sub>2</sub>, CoSe<sub>2</sub>, MnSe<sub>2</sub>, CuSe<sub>2</sub>, selenized NF and JCPDS cards of CoSe<sub>2</sub>, FeSe<sub>2</sub>, CuSe<sub>2</sub>, MnSe<sub>2</sub> and NiSe<sub>2</sub> (b).



**Fig. S5** XPS survey of the FeCoMnCuNiSe<sub>2</sub>.



**Fig. S6** Cyclic voltammograms for FeSe<sub>2</sub> (a), CoSe<sub>2</sub> (b), MnSe<sub>2</sub> (c), CuSe<sub>2</sub> (d), MnCuSe<sub>2</sub> (e), NiCuSe<sub>2</sub> (f), CoCuSe<sub>2</sub> (g), FeCuSe<sub>2</sub> (h), FeCoSe<sub>2</sub> (i), FeCoCuSe<sub>2</sub> (j), FeCoMnSe<sub>2</sub> (k), FeMnCuSe<sub>2</sub> (l), NF (m), CoMnCuSe<sub>2</sub> (n), and FeCoMnCuNiSe<sub>2</sub> (o) at the scan rate of 20, 40, 60, 80 and 100  $\text{mV s}^{-1}$  in 1.0 M KOH.



**Fig. S7**  $C_{dl}$  values of  $\text{FeSe}_2$ ,  $\text{CoSe}_2$ ,  $\text{MnSe}_2$ ,  $\text{CuSe}_2$ ,  $\text{MnCuSe}_2$ ,  $\text{NiCuSe}_2$ ,  $\text{CoCuSe}_2$ ,  $\text{FeCoCuSe}_2$ ,  $\text{FeCuSe}_2$ ,  $\text{FeCoSe}_2$ ,  $\text{FeCoMnSe}_2$ ,  $\text{FeMnCuSe}_2$ ,  $\text{NF}$ ,  $\text{CoMnCuSe}_2$  and  $\text{FeCoMnCuNiSe}_2$  at -0.20 V.

In the process of linear scanning, the current displayed by the instrument consists of two parts:

$$i = i_f + i_c \quad (1-1)$$

$i_f$  is faradaic current from electrochemical reaction,  $i_c$  is charge and discharge current of the electric double layer, and:

$$i_c = \frac{dq}{dt} = \frac{d(C_{dl}\psi)}{dt} = C_{dl} \frac{d\psi}{dt} + \psi \frac{dC_{dl}}{dt} \quad (1-2)$$

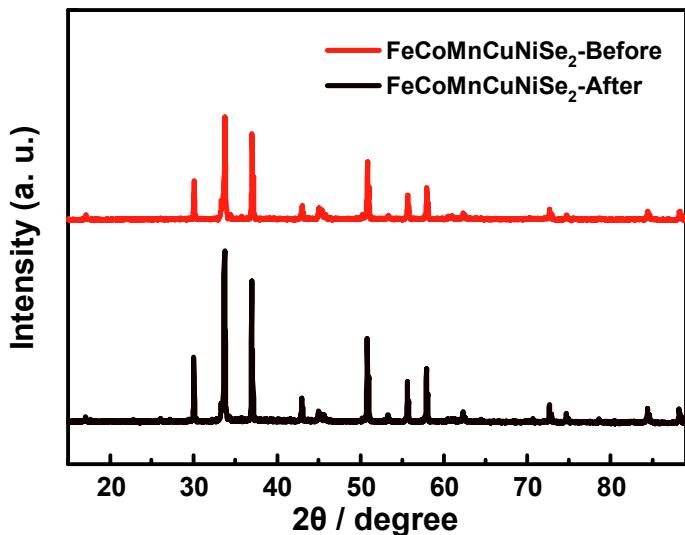
When the potential window in the Faraday region is selected to be smaller, the electric double layer capacitance  $C_{dl}$  remains almost unchanged, Then (1-2) can be simplified as:

$$i_c = C_{dl} \frac{d\psi}{dt} \quad (1-3)$$

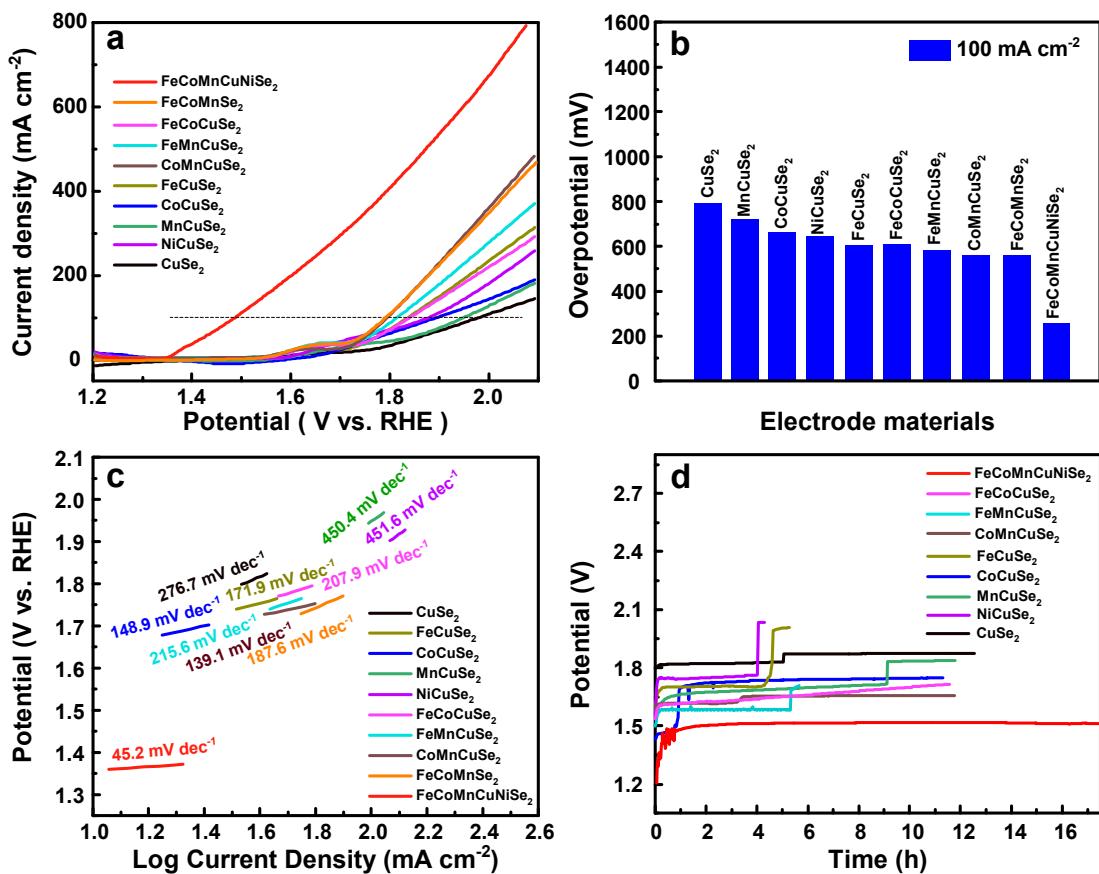
At the same time, there is a linear relationship between the electrochemical active area and the electric double layer capacitance:

$$ECSA = \frac{C_{dl}}{R} \quad (1-4)$$

where R is a constant in (1-4),  $C_{dl}$  is directly used to measure the ECSA of the catalyst.



**Fig. S8** The XRD spectra of FeCoMnCuNiSe<sub>2</sub> before and after 16 h stability.



**Fig. S9** The OER LSV curves (a), overpotentials at current density of 100 mA cm<sup>-2</sup> (b), Tafel plots (c), OER durability curves (d) of, FeCoMnCuNiSe<sub>2</sub>, FeCoCuSe<sub>2</sub>, CoMnCuSe<sub>2</sub>, FeMnCuSe<sub>2</sub>, FeCuSe<sub>2</sub>, CoCuSe<sub>2</sub>, MnCuSe<sub>2</sub>, NiCuSe<sub>2</sub> and CuSe<sub>2</sub> at 10 mA cm<sup>-2</sup>.

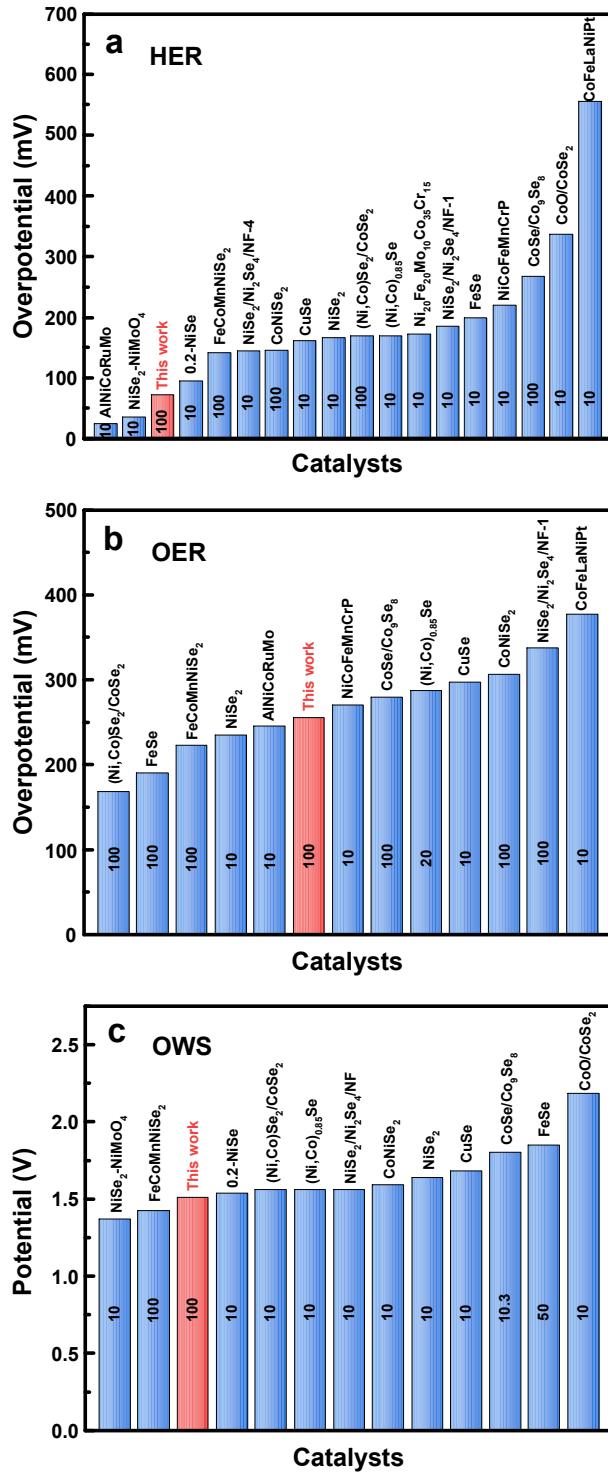
**Table S1** Atomic percentages of Fe, Co, Mn, Cu, Ni in the corresponding solutions of dissolved FeCoMnCuNiSe<sub>2</sub> in 6.0 M HCl for 1, 2, 4, 10 min at room temperature and in aqua regia at 170 °C for 36 h.

Time	Fe	Co	Mn	Cu	Ni
1 min	33.0	14.0	24.0	3.0	25.0
2 min	34.0	15.0	28.0	1.0	21.0
4 min	24.0	9.0	15.0	4.0	47.0
10 min	8.0	5.0	10.0	1.0	76.0
36 h	0.54	0.38	0.38	0.34	98.35

**Table S2** The HER, OER and OWS performance comparisons of prepared FeCoMnCuNiSe<sub>2</sub> to recently reported advanced electrocatalysts.

Catalyst	Support	Overpotential for HER (mV / mA cm <sup>-2</sup> )	Overpotential for OER (mV / mA cm <sup>-2</sup> )	Cell voltage (mA cm <sup>-2</sup> / V)	Reference
<b>FeCoMnCuNiSe<sub>2</sub></b>	<b>NF</b>	<b>71.6 / 100</b>	<b>254.8 / 100</b>	<b>100 / 1.51</b>	<b>This work</b>
0.2-NiSe	NF	95 / 10	-	10 / 1.54	[1]
NiSe <sub>2</sub>	NF	166 / 10	235/10	10 / 1.64	[2]
CoO/CoSe <sub>2</sub>	Ti-mesh	337 / 10	-	10 / 2.18	[3]
FeSe	NF	200 / 10	190/100	50 / 1.85	[4]
CoSe/Co <sub>9</sub> Se <sub>8</sub>	NF	268 / 100	280/100	10.3 / 1.8	[5]
CuSe	NF	162 / 10	297/10	10 / 1.68	[6]
CoNiSe <sub>2</sub>	NF	146 / 100	307/100	10 / 1.59	[7]
(Ni,Co)Se <sub>2</sub> /CoSe <sub>2</sub>	NF	169 / 100	169/100	10 / 1.56	[8]
(Ni,Co) <sub>0.85</sub> Se	NF	169 / 10	287/20	10 / 1.56	[9]
NiSe <sub>2</sub> /Ni <sub>2</sub> Se <sub>4</sub> /NF-4	NF	145 / 10	-	10 / 1.56	[10]
NiSe <sub>2</sub> /Ni <sub>2</sub> Se <sub>4</sub> /NF-1	NF	185 / 10	337/100	-	[10]
NiSe <sub>2</sub> -NiMoO <sub>4</sub>	NF	-35.6 / -10	-	10 / 1.37	[11]
Ni <sub>20</sub> Fe <sub>20</sub> Mo <sub>10</sub> Co <sub>35</sub> Cr <sub>15</sub>	-	172 / 10	-	-	[12]
AlNiCoRuMo	-	-24.5 / -10	245/10	-	[13]

CoFeLaNiPt	-	555 / 10	377/10	-	[14]
NiCoFeMnCrP	-	220 / 10	270/10	-	[15]
FeCoMnNiSe <sub>2</sub>	NF	142.2 / 100	223.1/100	100 / 1.42	[16]



**Fig. S10** The HER, OER and OWS performance comparisons of prepared FeCoMnCuNiSe<sub>2</sub> to recently reported advanced electrocatalysts.

Video of overall water splitting based on prepared self-standing high-entropy electrode



OWS Video.mp4

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