Electronic Supplementary Material (ESI) for RSC Advances.


Supplementary Information

# Ultrathin Co-Fe Hydroxide Nanosheet Arrays for Improved Oxygen Evolution during Water Splitting

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**Table S1**. ICP results of $Co_yFe_{1-y}(OH)_x$ NSAs.

| Number | Feed ratio of Co:Fe | Content of Co ($mg/cm^2$) | Content of Fe ($mg/cm^2$) | Atom ratio (Co : Fe) | y |
|---|---|---|---|---|---|
| **1** | 1:0 | 0.37 | 0 | ---- | 1 |
| **2** | 3:1 | 0.38 | 0.13 | 1:0.34 | 0.75 |
| **3** | 2:1 | 0.39 | 0.16 | 1:0.41 | 0.7 |
| **4** | 1:1 | 0.28 | 0.23 | 1:0.82 | 0.55 |
| **5** | 1:2 | 0.22 | 0.31 | 1:1.40 | 0.41 |
| **6** | 0:1 | 0 | 0.39 | ---- | 0 |

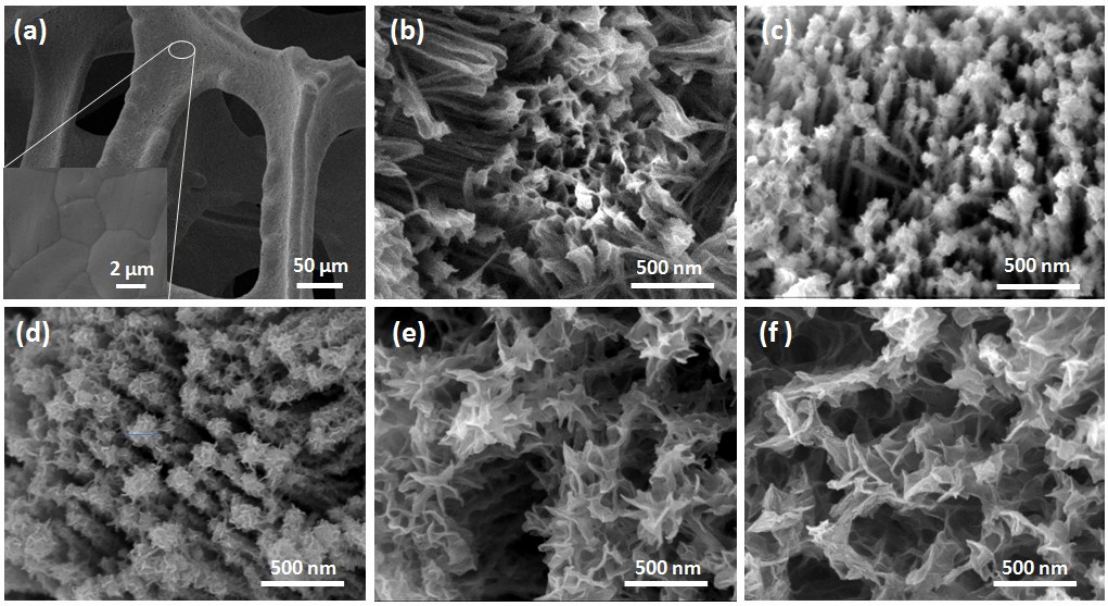


**Fig. S1.** SEM images of (a) foam Cu, (b) $Co(OH)_2$ NSAs, (c) $Co_{0.75}Fe_{0.25}(OH)_x$ NSAs, (d) $Co_{0.55}Fe_{0.45}(OH)_x$ NSAs, (e) $Co_{0.41}Fe_{0.59}(OH)_x$ NSAs and (f) $Fe(OH)_x$ NSAs.

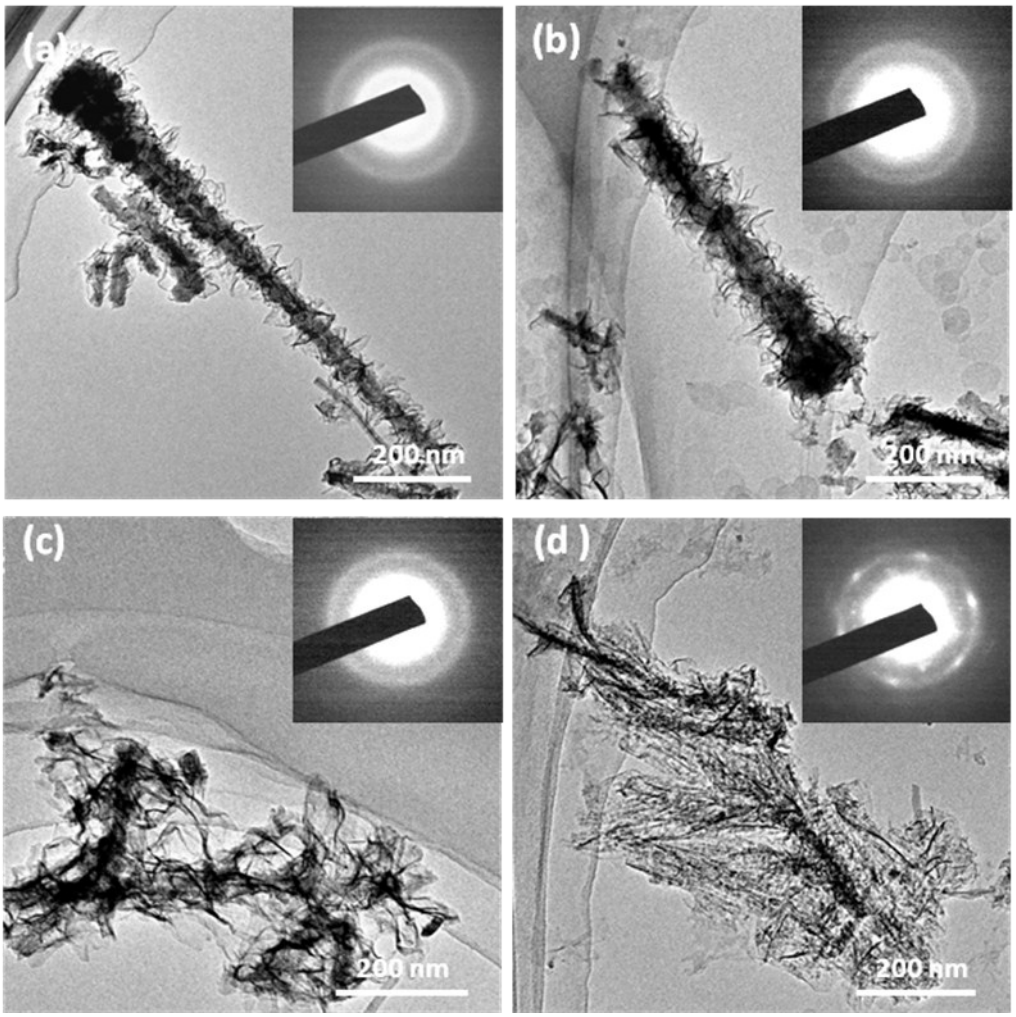


**Fig. S2.** TEM images and (inset) SAED patterns of (a) $Co_{0.75}Fe_{0.25}(OH)_x$ NSAs, (b) $Co_{0.55}Fe_{0.45}(OH)_x$ NSAs, (c) $Co_{0.41}Fe_{0.59}(OH)_x$ NSAs and (d) $Fe_2O_3$ NSAs.

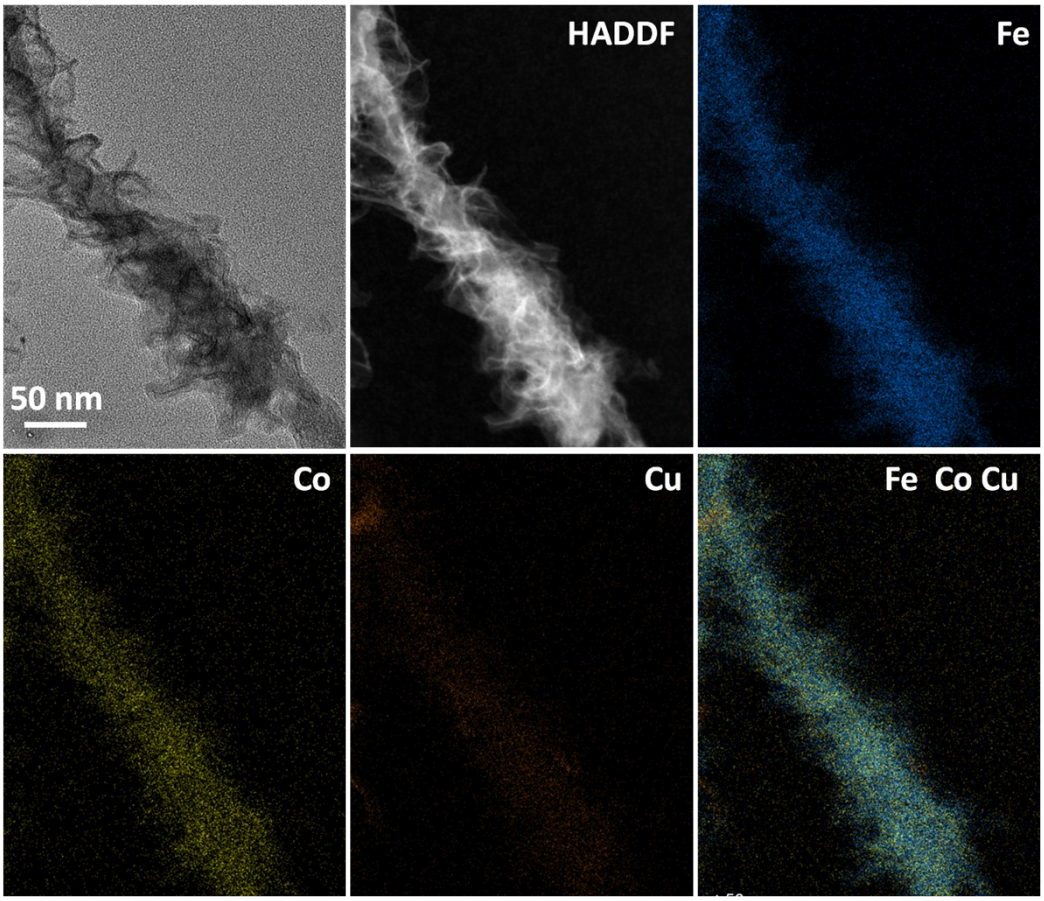


**Fig. S3.** The TEM image and corresponding Co, Fe and Cu EDS mapping images of $Co_{0.70}Fe_{0.30}(OH)_x$ NSAs.

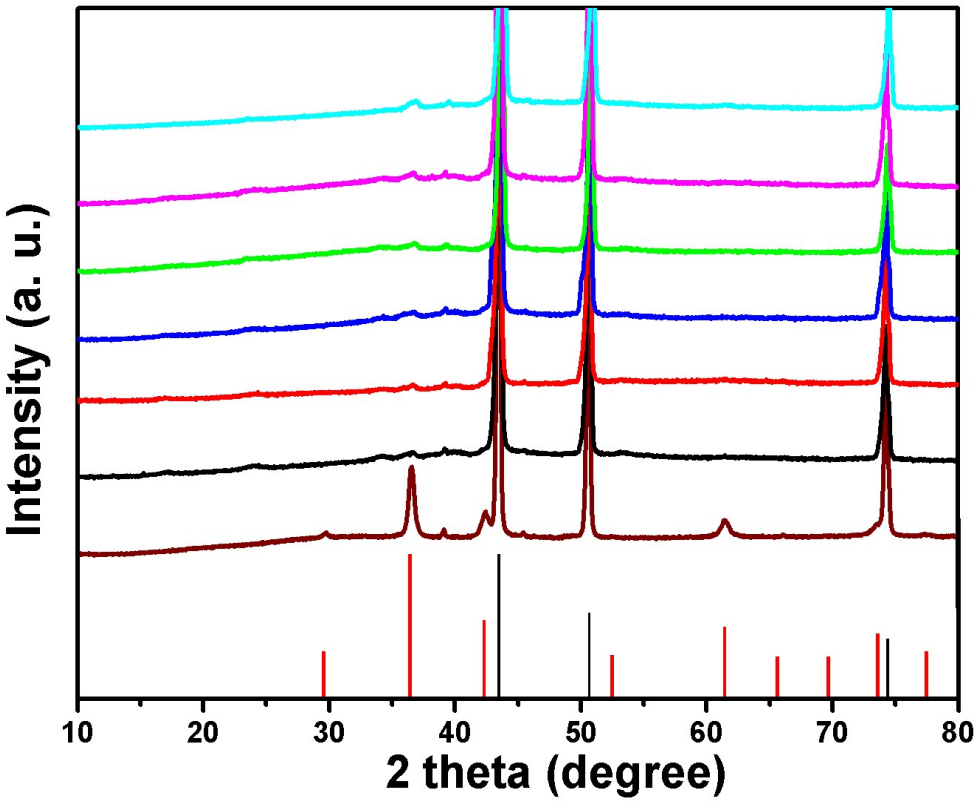


**Fig. S4.** XRD patterns for $Cu_2O$ template, $Co(OH)_2$, $Co_{0.75}Fe_{0.25}(OH)_x$, $Co_{0.70}Fe_{0.30}(OH)_x$, $Co_{0.55}Fe_{0.45}(OH)_x$, $Co_{0.41}Fe_{0.59}(OH)_x$ and $Fe_2O_3$ NSAs (lines from bottom to top). The peaks are indexed using reference peaks from the appropriate PDF cards: $Cu_2O$ phases (red, PDF#65-3288) and Cu phases (black, PDF# 65-9743).

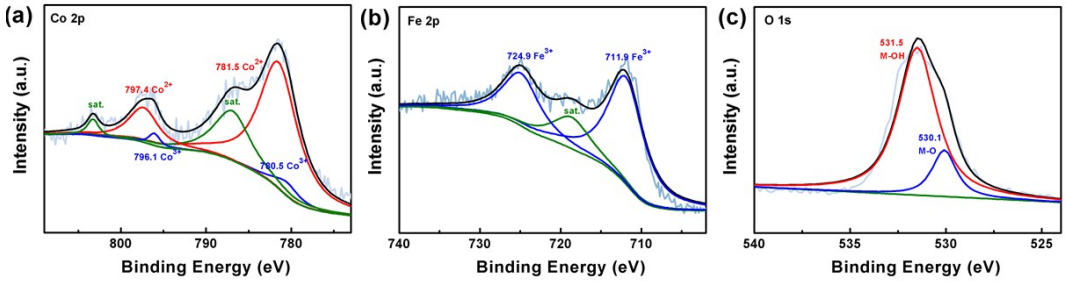


**Fig. S5.** High-resolution XPS spectra of (a) Co 2p, (b) Fe 2p and (c) O 1s of $Co_{0.75}Fe_{0.25}(OH)_x$ NSAs.

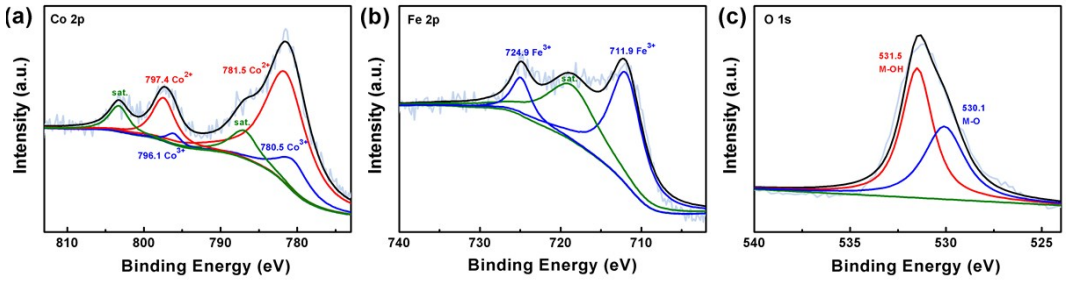


**Fig. S6.** High-resolution XPS spectra of (a) Co 2p, (b) Fe 2p and (c) O 1s of $Co_{0.55}Fe_{0.45}(OH)_x$ NSAs.

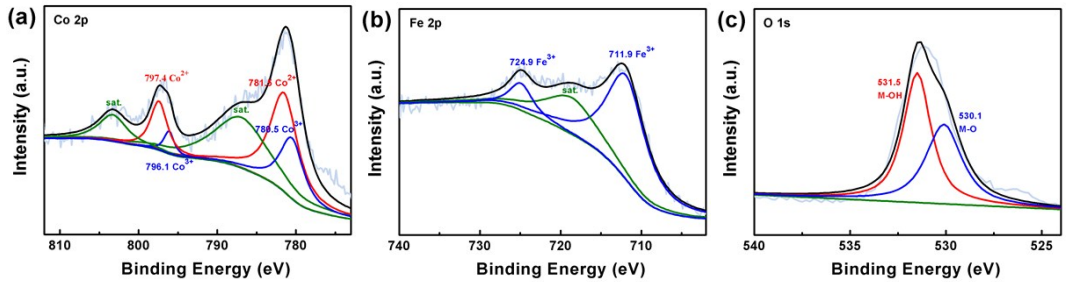


**Fig. S7**. High-resolution XPS spectra of (a) Co 2p, (b) Fe 2p and (c) O 1s of $Co_{0.41}Fe_{0.59}(OH)_x$ NSAs.

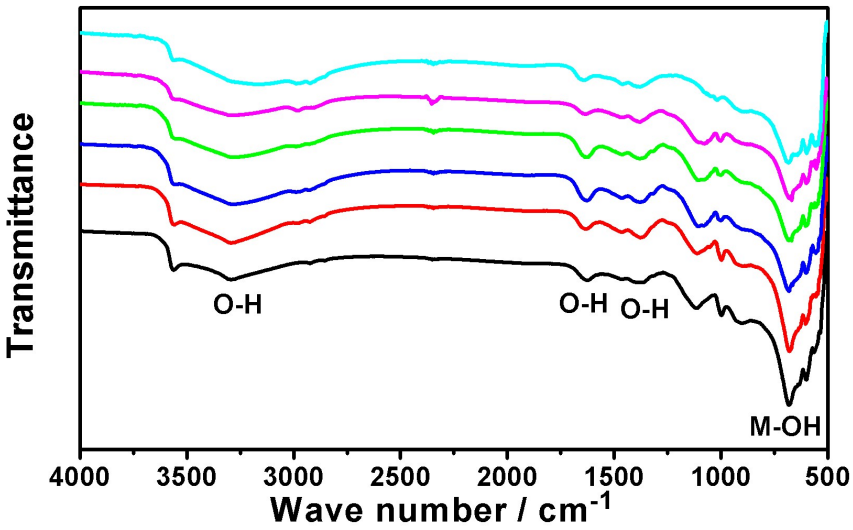


**Fig. S8.** Infrared spectra of $Co(OH)_2$, $Co_{0.75}Fe_{0.25}(OH)_x$, $Co_{0.70}Fe_{0.30}(OH)_x$, $Co_{0.55}Fe_{0.45}(OH)_x$, $Co_{0.41}Fe_{0.59}(OH)_x$ and $Fe_2O_3$ NSAs (lines from bottom to top).

**Table S2.** OER catalytic performances of $Co_yFe_{1-y}(OH)_x$ NSAs in 1 M KOH.

| Catalyst | Onset η (mV) | η at 10 mA $cm^{-2}$ (mV) | η at 100 mA $cm^{-2}$ (mV) | Tafel Slope (mV/dec) | TOF at η=380 mV ($s^{-1}$) |
|---|---|---|---|---|---|
| $Co(OH)_2$ | 250 | 270 | 347 | 74.9 | 0.081 |
| $Co_{0.75}Fe_{0.25}(OH)_x$ | 235 | 250 | 320 | 67.3 | 0.103 |
| $Co_{0.70}Fe_{0.30}(OH)_x$ | 220 | 245 | 310 | 62.4 | 0.172 |
| $Co_{0.55}Fe_{0.45}(OH)_x$ | 220 | 248 | 315 | 65.4 | 0.126 |
| $Co_{0.41}Fe_{0.59}(OH)_x$ | 225 | 262 | 330 | 68.9 | 0.076 |
| $Fe_2O_3$ | 270 | 325 | 375 | 50.9 | 0.045 |

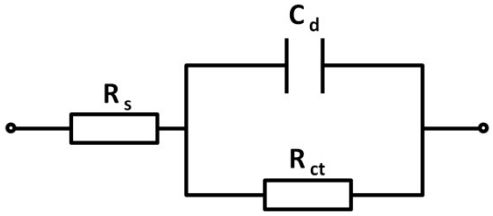


**Fig. S9.** Equivalent circuit used for fitting the EIS data.

**Table S3.** Fitting results of $Co_yFe_{1-y}(OH)_x$ NSAs.

| Material | Rs (Ω) | Rct (Ω) | Rcp (Ω) |
|---|---|---|---|
| $Co(OH)_2$ | 2.22 | 3.57 | 0.21 |
| $Co_{0.75}Fe_{0.25}(OH)_x$ | 3.62 | 4.08 | 0.09 |
| $Co_{0.70}Fe_{0.30}(OH)_x$ | 3.20 | 2.91 | 0.16 |
| $Co_{0.55}Fe_{0.45}(OH)_x$ | 2.74 | 3.53 | 8.12 |
| $Co_{0.41}Fe_{0.59}(OH)_x$ | 3.18 | 9.64 | 3.68 |
| $Fe_2O_3$ | 3.74 | 24.4 | 0.089 |

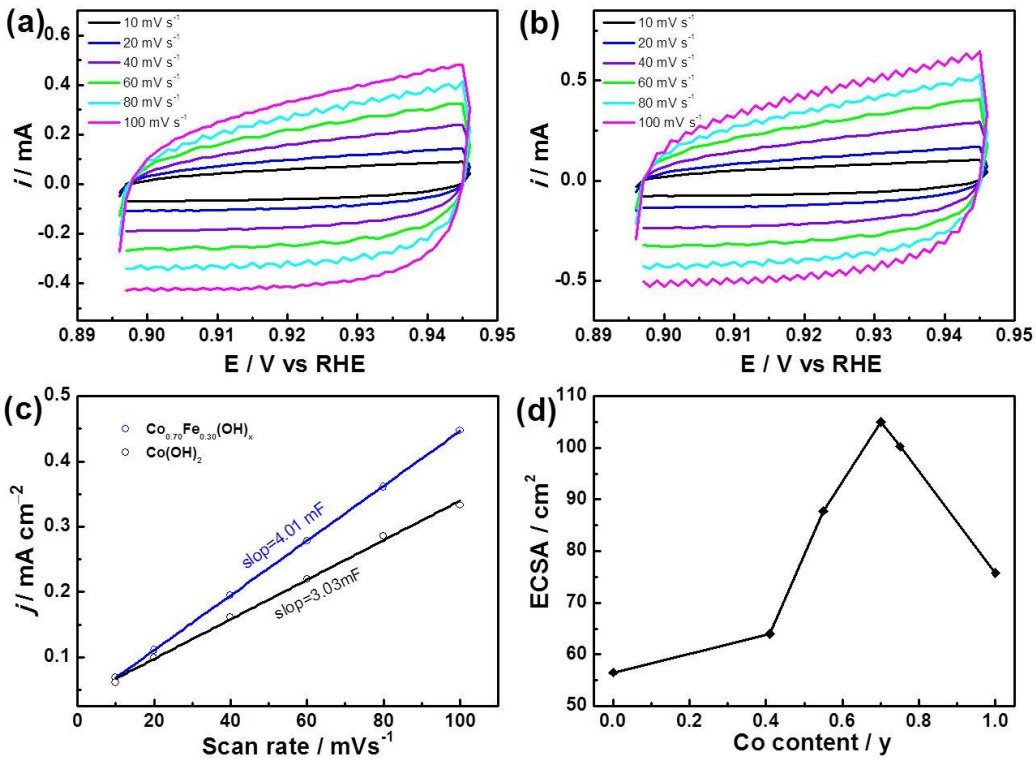


**Fig. S10.** Cyclic voltammetry curves of (a) $Co(OH)_2$ and (b) $Co_{0.70}Fe_{0.30}(OH)_x$ NSAs, (c) the cathodic charging currents measured at 0.92 V vs RHE plotted as a function of scan rate for $Co(OH)_2$ and $Co_{0.70}Fe_{0.30}(OH)_x$ NSAs, (d) ECSA of $Co_yFe_{1-y}(OH)_x$ NSAs.

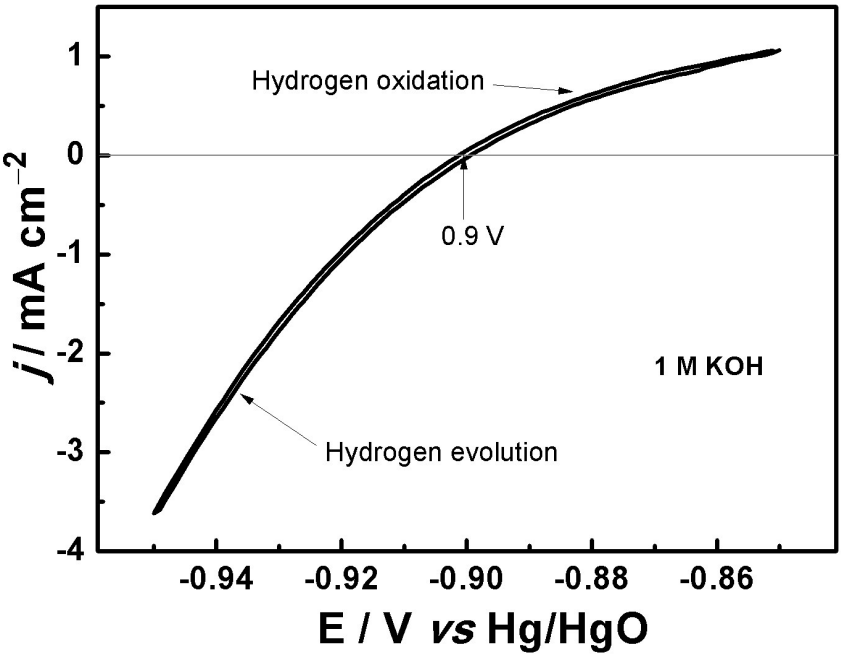


**Fig. S11.** Hg/HgO (1 M KOH) electrode calibrate with a 1M KOH solution.

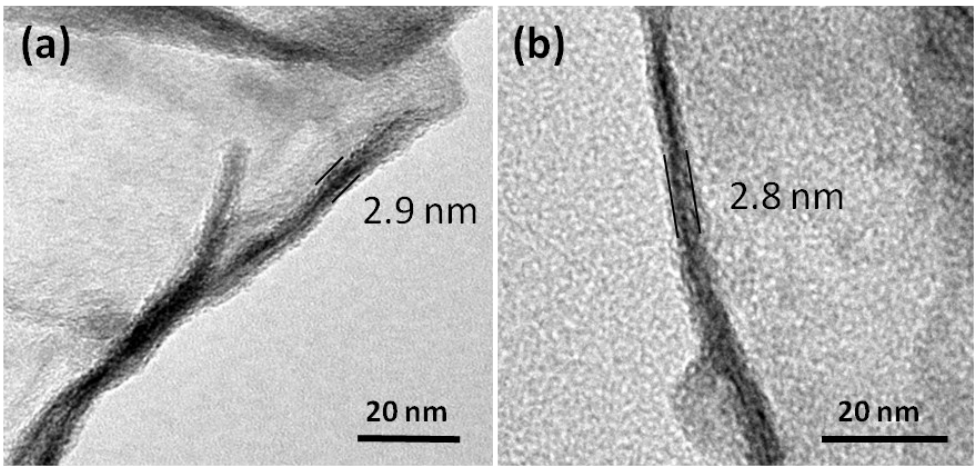


**Fig. S12** TEM images of $Co_{0.70}Fe_{0.30}(OH)_x$ NSAs edge curled nanosheets.

**Table S4.** The OER activities of some Co-based electrocatalysts for water oxidation under alkaline solution.

| Catalyst | Electrolyte | Onset η (mV) | η at 10 mA $cm^{-2}$ (mV) | Tafel Slope (mV $dec^{-1}$) | TOF ($s^{-1}$) | Mass loading (mg $cm^{-2}$) | Ref. |
|---|---|---|---|---|---|---|---|
| **$NiCo_2O_4$ core-shell nanowire** | 1 M NaOH | 270 | 320 | 63.1 | NA | NA | [1] |
| **Mesoporous Ni sphere array** | 0.1 M KOH | 190 | 254 | 39 | 0.0281 (η=450 mV) | 0.2 | [2] |
| **$Zn_xCo_{3-x}O_4$ nanoarrays** | 0.1 M KOH | NA | 320 | 51 | NA | 0.2 | [3] |
| **$Ni_2P$ nanowires** | 1.0 M KOH | 310 | 400 | 60 | NA | 0.1 | [4] |
| **$Co_3O_4$ Nanoparticles** | 1 M KOH | 320 | 370 | 62 | NA | 0.325 | [5] |
| **$Co_3O_4$ nanocube/CoO** | 1 M KOH | NA | 430 | 89 | NA | 56.5 | [6] |
| **$CoO_x$ nanotube** | 1 M KOH | 230 | NA | 75 | NA | 0.136 | [7] |
| **Ni-V LDH** | 1 M KOH | 250 | 300 | 50 | 0.054 (η=350 mV) | 0.143 | [8] |
| **$NiCoO_x$ hollow nanosponges** | 0.1 M KOH | 271 | 362 | 73.2 | NA | 0.2 | [9] |
| **Screw CoNi LDH/C** | 1M KOH | 330 | 360 | 38.5 | NA | 2 | [10] |
| **NiCo/NF or NiCo/CP** | 1 M KOH | NA | 360 | 50-60 | NA | 0.4 | [11] |
| **$NiCoO_x$ nanoarray** | 1 M KOH | 280 | 290 | 79 | NA | 3 | [12] |
| **Co-S nanosheets film** | 1 M KOH | 320 | 361 | 64 | NA | NA | [13] |
| **NiCo hydroxide** | 0.1 M KOH | 310 | 460 | 65 | NA | NA | [14] |
| **Hollow fluffy cages** | 1 M KOH | NA | 409 | 70 | 0.0167 (η=400 mV) | 0.14 | [15] |
| **$Ni_{2/3}Fe_{1/3}$-rGO** | 1 M KOH | 210 | 230 | 40 | 0.1 (η=300 mV) | 0.25 | [16] |
| **$Ni_3S_2$ nanorods/NF** | 0.1 M KOH | 157 | 187 | 153 | NA | NA | [17] |
| **Fe-Ni/NF** | 1MKOH | 200 | NA | 32 | 0.075 (η=400 mV) | NA | [18] |
| **$CoFe_2O_4$ nanoparticles** | 1M NaOH | 270 | 378 | 73 | NA | 1.031 | [19] |
| **CoFe-LDH** | 0.1 M KOH | 260 | 325 | 43 | 0.12 (η=350 mV) | 0.2 | [20] |
| **FeOOH/Co/FeOOH Hybrid Arrays** | 1M NaOH | 230 | NA | 32 | 5.6 (η=300 mV) | 0.5 | [21] |

**Notes and references**


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