# **Supplementary Information**

# Amorphous mixed-metal hydroxide nanostructures for advanced water

# oxidation catalysts

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#### S1 Detailed description of the preparation of amorphous Ni-Fe hydroxides

## S1-1 Method

The amorphous Ni-Fe hydroxides electrodes were prepared by a electrochemistry technique which is simple, facile and green (Supplementary Information S1). The targets of Ni-Fe mixture with an area of  $25 \times 25 \text{ mm}^2$  and different atomic ratios (1:4,1:2,1:1,2:1) were placed at the center of a quartz chamber and immersed in highly pure de-ionized water (18.2 M $\Omega$  cm<sup>-1</sup>) without any chemical additives, respectively. The fabrication was carried out under a constant potential voltage of 100 V for 10 h and the final products were deposited on the surface of the graphite sheet (electrode).

## S1-2 Formation mechanism of amorphous Ni-Fe hydroxides

The basic physics and chemistry involved in the preparation of the amorphous Ni-Fe hydroxides are discussed as follows. We propose a formation mechanism in our case as shown in Figure S1. Simply, the formation of the amorphous Ni-Fe hydroxides can be divided into two processes below. (i) When the Ni-Fe mixture target is immersed in pure de-ionized water (18.2 M $\Omega$  cm) with an extra electric field, it can be polarized and releases a small quantity of transitional metal ion (M<sup>++</sup>) and e<sup>+</sup>, adhered on the surface of alloy target. (ii) Under the influence of an applied electric field, M<sup>++</sup> moves toward to the cathode, and then the electrochemical reactions take place on the cathode as follows:

$$M^{x+} + xOH \to M(OH)_x \tag{S1}$$

$$2H^{+} + 2e \rightarrow H^{2} \tag{S2}$$

At the same time, H moves toward to the surface of the alloy target and OH moves toward to the anode under the influence of an applied electric field. Finally, the electrochemical reactions take place as follows:

$$2H^{+} + 2e \to H^2 \tag{S3}$$

$$4OH - 4e \rightarrow O_2 + 2H_2O \tag{S4}$$

#### S1-3 Advantage of the developed method

Compared to the conventional electrochemistry method, the developed technique in this study has the following merits. Firstly, this method is a simple, facile and green electrochemistry technique. Secondly, the highly pure de-ionized water, as the electrolyte instead of electrolytic solution having well electric conductivity, provides a chemically clean reaction environment without any chemical additives, which ensures highly clean, pure and active surface of productions. Finally, this method is controllable for the preparation of amorphous phases of mixed metal hydroxides. For example, we can easily control the composition of the mixed metal hydroxides by designing the component of the alloy target.



Figure S1. A schematic illustration of our electrochemistry experiment setup



**Figure S2.** XRD patterns of the as-synthesized amorphous  $Ni_{0.71}Fe_{0.29}(OH)_x$  sample on the graphite sheet and the bare graphite sheet. Clearly, there are no additional peaks in the XRD pattern, which confirms the amorphous nature of the sample.



**Figure S3.** (a-e) LSV curves for the amorphous  $Ni_{0.26}Fe_{0.74}(OH)_x$ ,  $Ni_{0.48}Fe_{0.52}(OH)_x$ ,  $Ni_{0.71}Fe_{0.29}(OH)_x$ ,  $Ni_{0.89}Fe_{0.11}(OH)_x$  and commercial RuO<sub>2</sub>, respectively with (red) and without (black) iR correction. The ionic resistance (~10  $\Omega$ ) from the solution is determined by the EIS technique.



**Figure S4.** XRD patterns of the amorphous  $Ni_{0.71}Fe_{0.29}(OH)_x$  after OER and the bare graphite sheet. Clearly, there are no additional peaks in the XRD pattern, which confirms the amorphous nature of the intermediate.



**Figure S5.** LSV curve measured in O<sub>2</sub>-saturated 0.1 M KOH solution for amorphous  $Ni_{0.71}Fe_{0.29}(OH)_x$  in the region of ORR. The amorphous  $Ni_{0.71}Fe_{0.29}(OH)_x$  shows negative onset potential and small cathodic currents, suggesting poor ORR catalytic activity of it.