

## Supporting Information

### **Coupling Effect Induced Acceleration of Electron Transfer for $\alpha$ -Ni(OH)<sub>2</sub> with Enhanced Oxygen Evolution Reaction Activity**

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**Synthesis of  $\text{Co(OH)}_2$  nanosheets.** In a typical synthesis, 8.5 mg  $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$  and 100 mg PVP were dissolved in 21.5 mL  $\text{H}_2\text{O}$  in a 100 mL round-bottomed flask. Then the solution was purged with  $\text{N}_2$  for 10 min. After that, 10 mL  $13 \text{ mmol dm}^{-3}$  freshly prepared  $\text{NaBH}_4$  were added dropwise to the mixture under stirring. 10 min later, the solution was exposed to the air and then the products were centrifuged and washed with water and ethanol for several times, the samples were dried at  $40^\circ\text{C}$  under vacuum.

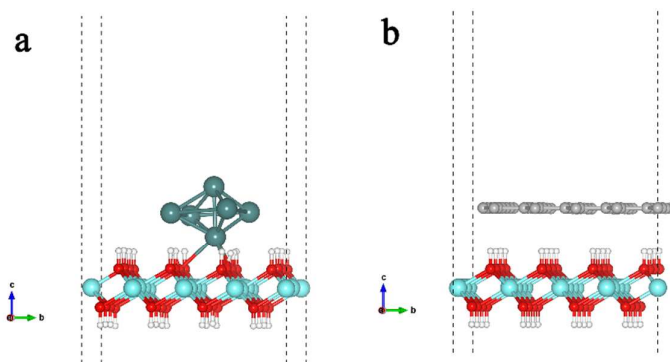
**Synthesis of  $\text{Co(OH)}_2$ -Ag-RGO composite.**  $\text{Co(OH)}_2$  nanosheets (10 mg) prepared above were dispersed in 4 mL ethanol in a 20 mL bottle, then the bottle was sealed and degassed with nitrogen gas for 30 min. After that, the solution was irradiated with 254 nm UV light for another 30 min. Then 150  $\mu\text{L}$   $0.0647 \text{ mol dm}^{-3}$  degassed  $\text{AgNO}_3$  solution was added, the reaction sustained for 12 h under dark with stirring. After that, the obtained sample was washed with ethanol, dissolved in 4 mL ethanol again and degassed, reirradiated once more. Then 0.5 mL GO nanosheets ethanol solution (2 mg/mL) was added into the above solution. After stirring for 5 h, the sample was washed with water and ethanol, dried at  $40^\circ\text{C}$  under vacuum.

**Synthesis of  $\text{FeOOH}$  nanosheets.** 182 mg CTAB was first dissolved in 50 mL of  $\text{H}_2\text{O}$  in a 100 mL round-bottomed flask, then the pH of the solution was adjusted to 3 by  $\text{H}_2\text{SO}_4$  before 41.7 mg  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  was added. Then 2 mL  $0.4 \text{ mol dm}^{-3}$  freshly prepared  $\text{NaBH}_4$  was dropwise to the solution, which had been purged with  $\text{N}_2$  for 10 min. After another 10 min, the solution was exposed to the air. Then the products were collected and washed with water and ethanol, dried at  $40^\circ\text{C}$  under vacuum.

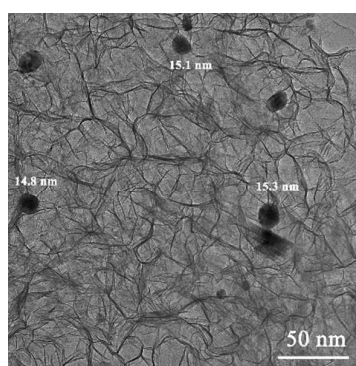
**Synthesis of  $\text{FeOOH}$ -Ag-RGO composite.**  $\text{FeOOH}$  nanosheets (10 mg) prepared above were dispersed in 4 mL ethanol in a 20 mL bottle, then the bottle was sealed and degassed with nitrogen gas for 30 min. After that, the solution was irradiated with 254 nm UV light for another 30 min. Then 150  $\mu\text{L}$   $0.0647 \text{ mol dm}^{-3}$  degassed  $\text{AgNO}_3$  solution was added, the reaction sustained for 12 h under dark with stirring. After that, the obtained sample was washed with ethanol, dissolved in 4 mL ethanol again and degassed, reirradiated once more. Then 0.5 mL GO nanosheets ethanol solution (2 mg/mL) was added into the above solution. After stirring for 5 h, the sample was washed with water and ethanol, dried at  $40^\circ\text{C}$  under vacuum.

**Table S1.** Comparison of OER activity for as-prepared catalysts (NA=Not applicable)

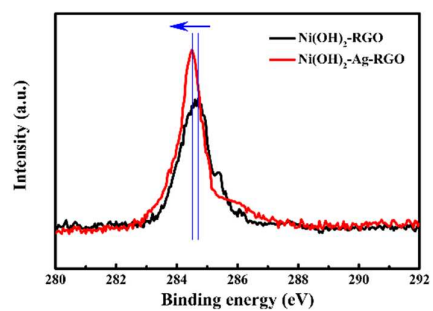
Catalyst	$\eta$ at $j = 10$ $\text{mA cm}^{-2}$ (mV)	$\eta$ at $j = 20$ $\text{mA cm}^{-2}$ (mV)	mass activity at $\eta = 0.30$ V ( $\text{A g}^{-1}$ )	specific activity at $\eta = 0.30$ V ( $\text{mA cm}^{-2}$ )	Tafel slope (mV $\text{dec}^{-1}$ )	TOF at $\eta =$ <b>0.30 V</b> ( $\times 10^{-3} \text{ s}^{-1}$ )
<b>Ni(OH)<sub>2</sub>-Ag-RGO</b>	292	349	90.6	0.061	32	22.3
<b>Ni(OH)<sub>2</sub>-Ag</b>	349	395	67.7	0.030	38	16.4
<b>Ni(OH)<sub>2</sub>-RGO</b>	356	403	66.8	0.029	50	16.3
<b>Ni(OH)<sub>2</sub></b>	387	446	51.3	0.027	53	12.3
<b>IrO<sub>2</sub></b>	340	436	40.3	NA	74	23.4



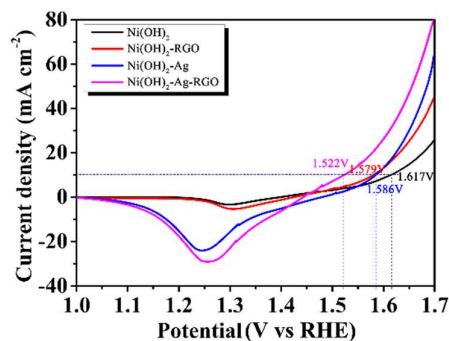
**Figure S1.** Computational models for (a)  $\text{Ni(OH)}_2\text{-Ag}$  and (b)  $\text{Ni(OH)}_2\text{-RGO}$  after optimization.



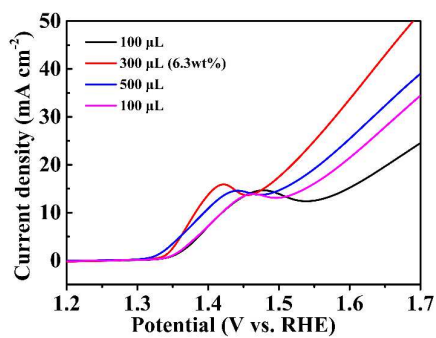
**Figure S2.** HRTEM image of  $\text{Ni(OH)}_2\text{-Ag-RGO}$  electrocatalyst



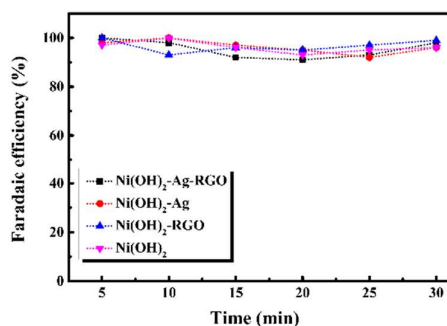
**Figure S3.** XPS spectra of C1s for  $\text{Ni(OH)}_2\text{-RGO}$  and  $\text{Ni(OH)}_2\text{-Ag-RGO}$



**Figure S4** The linear voltammetry curves without  $iR$ -correction reverse scanned from 1.7 V to 1.0 V.



**Figure S5.** OER polarization curves of Ni(OH)<sub>2</sub>-Ag nanocomposite with different loading amounts of Ag.



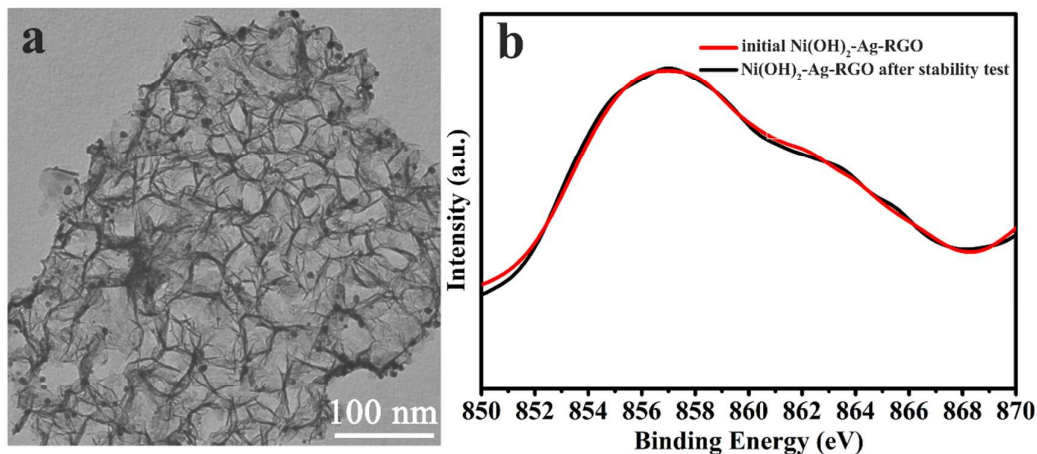
**Figure S6.** Faradic efficiencies of the as-prepared electrocatalysts during OER measurement.

The Faradic efficiencies of the electrocatalytic system are measured on a gas chromatograph at 1.56V

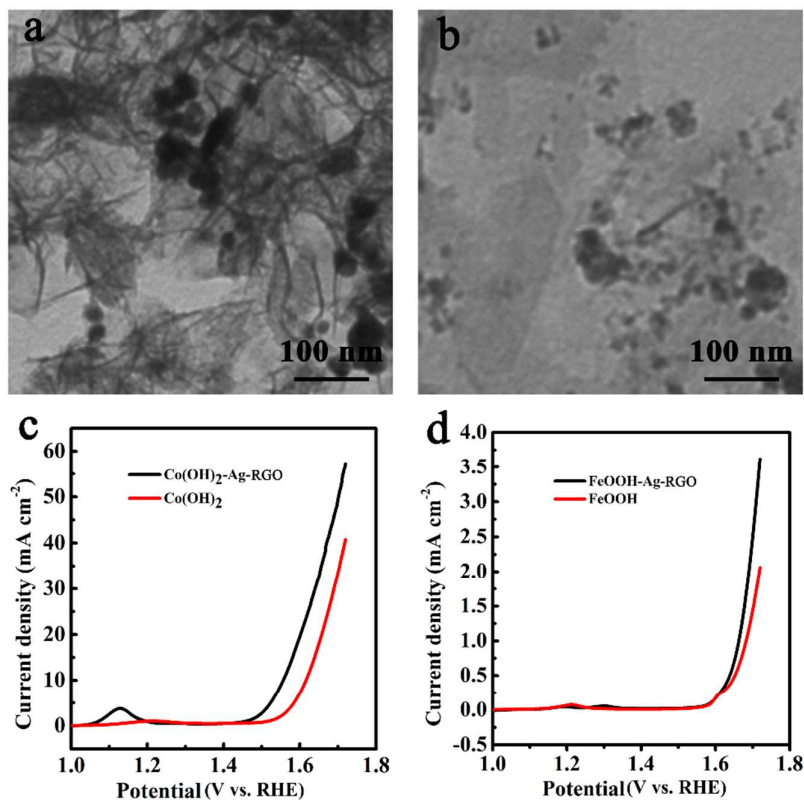
vs. RHE under solar simulated illumination and calculated according to the equation ( $\eta = \frac{mnF}{It}$ ) as

shown in Figure S6, where  $\eta$  is the Faradic efficiency,  $m$  is the actual molar number of O<sub>2</sub>,  $n$  is the

number of reactive electrons( considered as 4 here),  $F$  is Faraday constant ( $96485.3 \text{ C mol}^{-1}$ ),  $I$  is the used current and  $t$  is time.



**Figure S7.** (a) TEM image of  $\text{Ni(OH)}_2\text{-Ag-RGO}$  electrocatalyst after stability test, (b) XPS spectra comparison of Ni 2p.



**Figure S8.** TEM images of  $\text{Co(OH)}_2\text{-Ag-RGO}$  and  $\text{FeOOH-Ag-RGO}$  nanostructures (a, b) and the linear voltammetry curves (no  $iR$ -correction) of  $\text{Co(OH)}_2\text{-Ag-RGO}$ ,  $\text{FeOOH-Ag-RGO}$  (c),  $\text{Co(OH)}_2$

and FeOOH catalysts (d).

***Method for TOF calculation:***

$$TOF = \frac{jS}{4Fn}$$

Where  $j$  is the measured current density ( $\text{mA cm}^{-2}$ ),  $S$  is the surface area of the GC electrode, the number 4 in the TOF calculation means 4 electrons required for one  $\text{O}_2$  molecule evolution,  $F$  is Faraday's constant ( $96485.3 \text{ C mol}^{-1}$ ), and  $n$  is the moles of metal atom on the electrode