Supplementary Materials

Strong and Hierarchical $Ni(OH)_2$/Ni/rGO Composites as Multifunctional Catalysts for Excellent Water Splitting

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Preparation of Ni/rGO composite catalyst

In a classic experiment, GO was prepared by modified Hummers method. In this work, the Ni/rGO composite electrodes were prepared by supergravity electrodeposition, and the configuration of equipment is presented in our published paper [1]. Firstly, a ø10 cm × 2 cm Ni foam circle was cleaned sequentially in HCl solution (15%), acetone and DI water for 5 min with ultrasonication to use as cathode. And a pure nickel pipe was used as anode. The Ni/rGO composite coatings were electrodeposited in a blackish green plating bath which contained 350 g L^{-1} $Ni(NH_2SO_3)_2 \cdot 6H_2O$, 10g L^{-1} $NiCl_2 \cdot 6H_2O$, 30 g L^{-1} NH_4Cl with 1.0 g L^{-1} GO, respectively. The pH value of the plating bath is 3.5-3.8. All composite electrodes were performed under the strength of the supergravity with G=350 g at a current density of 3 A dm^{-2} for 60 min at 318 K. Afterwards the Ni foam coated with Ni/rGO hybrid was taken out of the reaction vessel, followed by washed with deionized water to remove physical adsorption residua, and then dried at 80 °C.

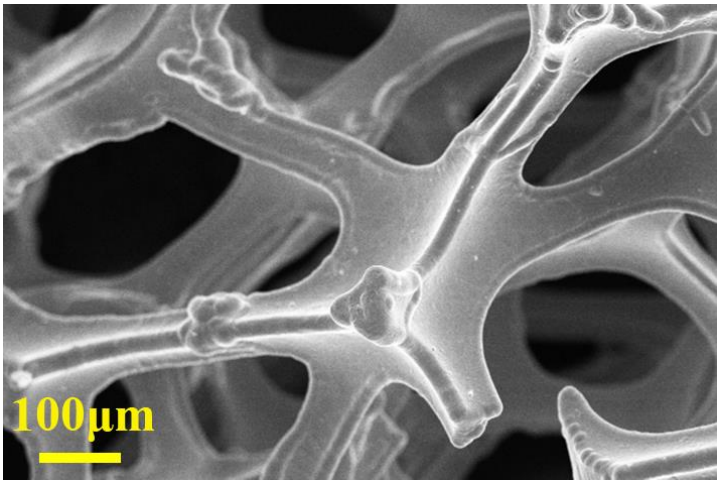

Figure S1. Low-magnification SEM images of Ni foam.

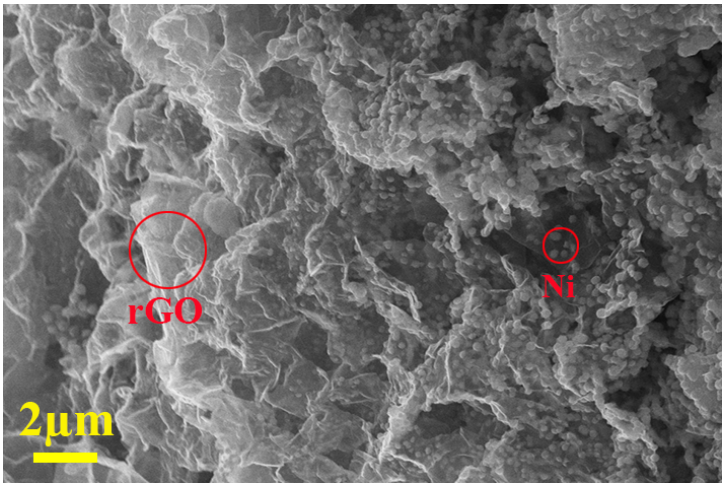

Figure S2. High-magnification SEM image of Ni/rGO composite catalyst.

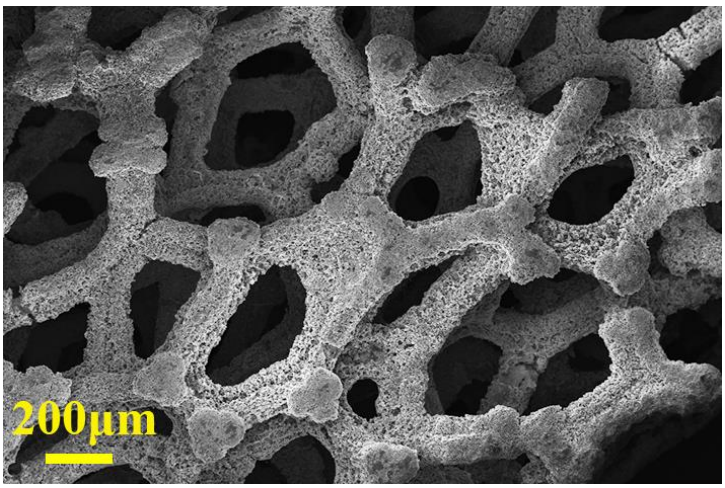

Figure S3. Low-magnification SEM images of $Ni(OH)_2$/Ni/rGO-1 composite catalyst.

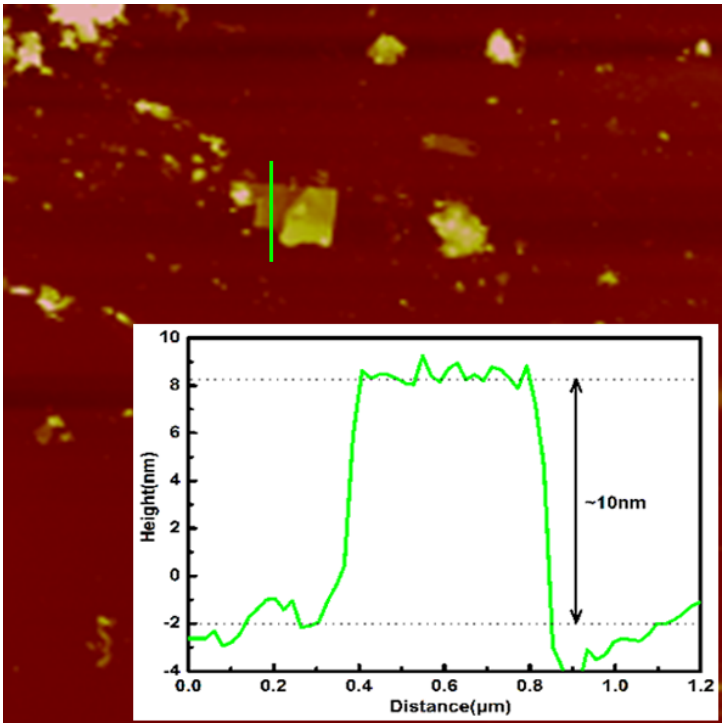

Figure S4. AFM image of $Ni(OH)_2$ nanosheets spalled off $Ni(OH)_2$/Ni/rGO-1 catalyst (with 1 h sonication), and inset image is corresponding AFM height image.

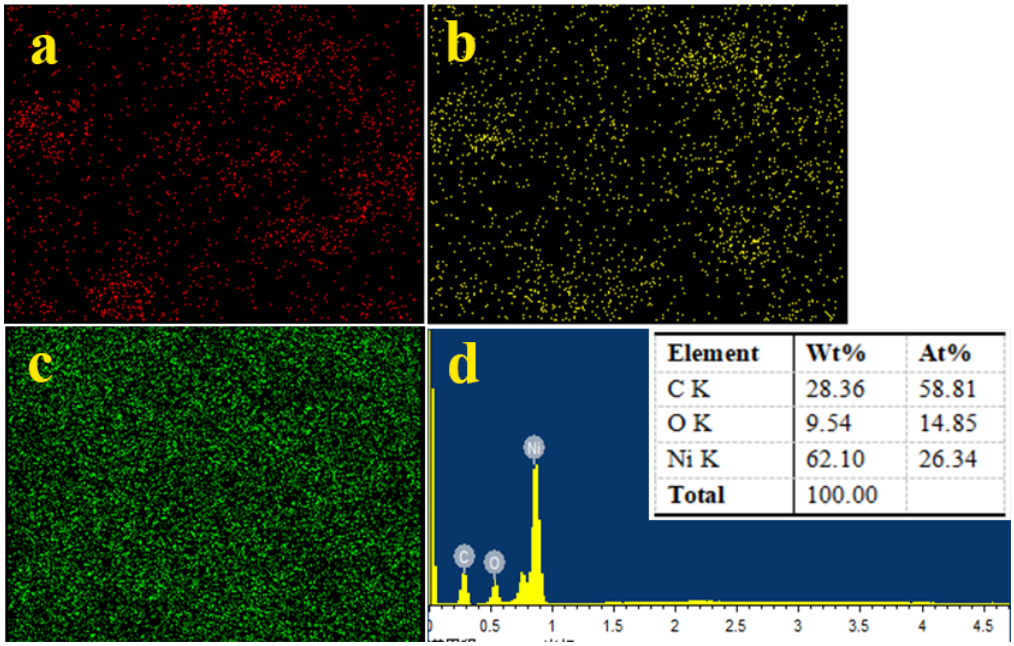

Figure S5. (a-c) Elemental mapping of individual elements (C, O, Ni) of image Figure 1,a. (d) EDS analysis of $Ni(OH)_2$/Ni/rGO-1 composite coating.

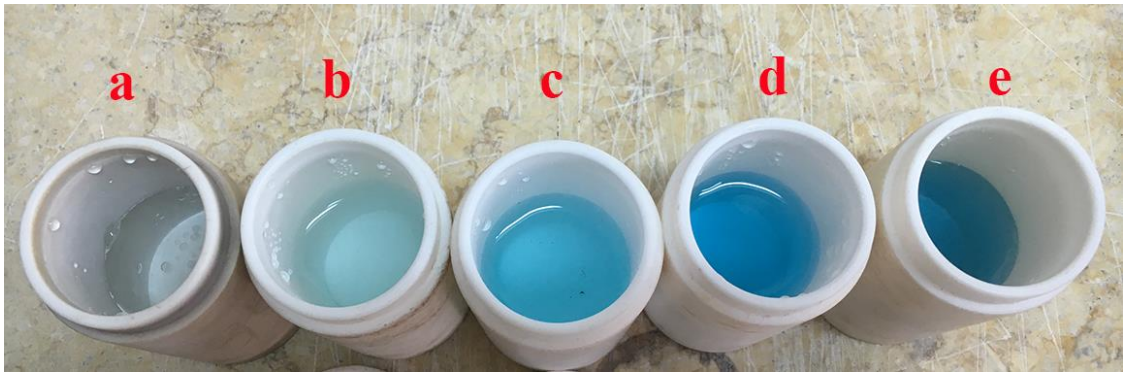

Figure S6. The digital photos of the color of the urea solution after hydrothermal process. The color of the urea solution changes obviously from colorless to dark blue. (a) $Ni(OH)_2$/Ni/rGO-1, (b) $Ni(OH)_2$/Ni/rGO-2, (c) $Ni(OH)_2$/Ni/rGO-3, (d) $Ni(OH)_2$/Ni/rGO-4, and (e) $Ni(OH)_2$/Ni/rGO-5 composite catalysts.

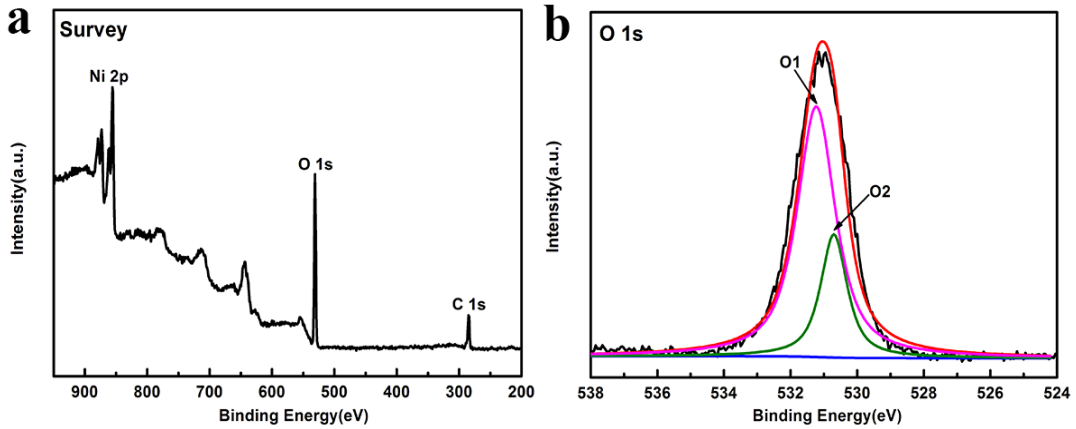

Figure S7. (a) XPS survey spectrum for $Ni(OH)_2$/Ni/rGO-1. (b) XPS spectrum of O 1s of $Ni(OH)_2$/Ni/rGO-1 catalyst.

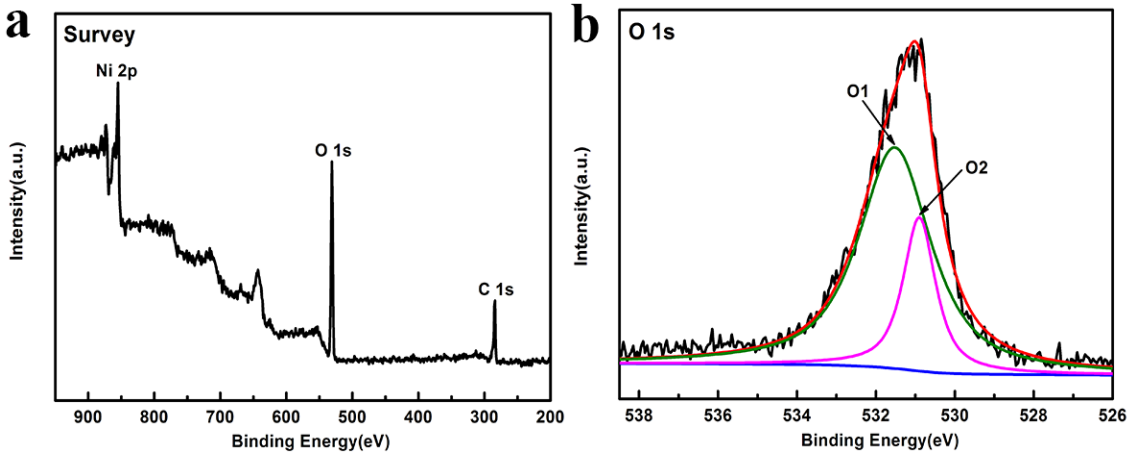

Figure S8. (a) XPS survey spectrum for $Ni(OH)_2$/Ni/rGO-3. (b) XPS spectrum of O 1s of $Ni(OH)_2$/Ni/rGO-3 catalyst.

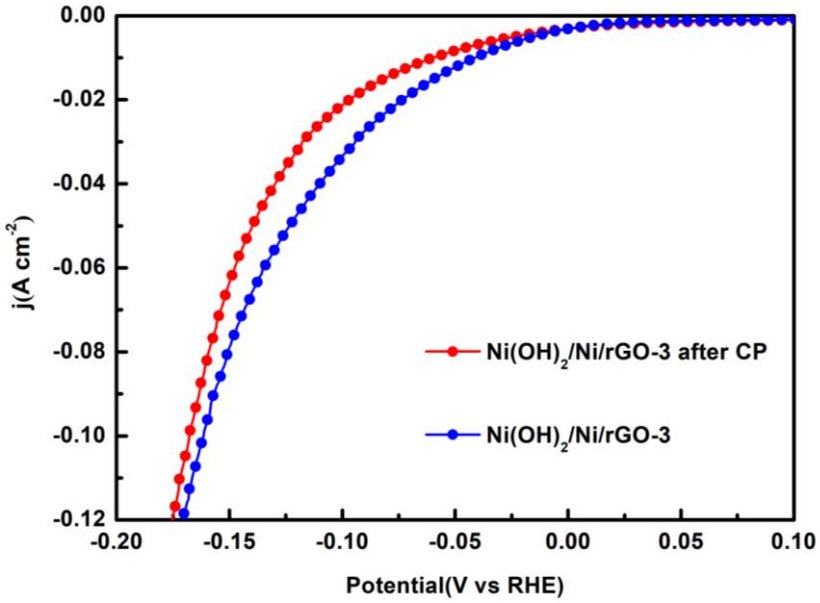

Figure S9. LSV curves for $Ni(OH)_2$/Ni/rGO-3 before and after chronopotentiometry test.

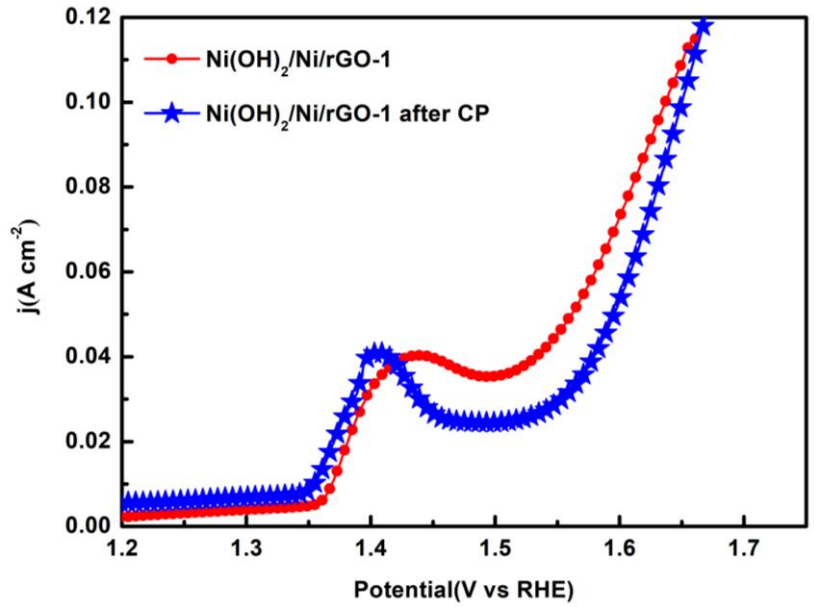

Figure S10. LSV curves for $Ni(OH)_2$/Ni/rGO-1 before and after chronopotentiometry test.

Table S1. Comparison of electrocatalytic activity for HER, OER and EWS of diverse Ni-based catalysts in alkaline solution.

Catalyst	Water electrolysis	Electrolyte	Current density	Overpotential	Reference
NiFe LDH-NS	HER	1 M KOH	20 mA cm^{-2}	115 mV	[2]
	OER		10 mA cm^{-2}	210 mV	
	EWS		20 mA cm^{-2}	1.5 V	
	EWS		10 mA cm^{-2}	1.43 V	
FNHNs/NF	HER	1 M KOH	10 mA cm^{-2}	140 mV	[3]
	OER		10 mA cm^{-2}	290 mV	
	EWS		10 mA cm^{-2}	1.55 V	
$Ni(OH)_2$/NF	HER	1 M KOH	10 mA cm^{-2}	127 mV	[4]
	HER		20 mA cm^{-2}	172 mV	
	OER		50 mA cm^{-2}	330 mV	
	EWS		10 mA cm^{-2}	1.68V	
$NiP_2/NiSe_2$	HER	1 M KOH	10 mA cm^{-2}	93mV	[5]
	HER		100 mA cm^{-2}	160 mV	
	OER		10 mA cm^{-2}	253 mV	
	EWS		10 mA cm^{-2}	1.56 V	
Mo_2S_3@$NiMo_3S_4$	HER	1 M KOH	10 mA cm^{-2}	32mV	[6]
	HER		100 mA cm^{-2}	124 mV	
	OER		10 mA cm^{-2}	173 mV	
	OER		100 mA cm^{-2}	256 mV	
	EWS		500 mA cm^{-2}	1.639 V	
$NiCo_2S_4$ NA/CC	HER	1 M KOH	100 mA cm^{-2}	305 mV	[7]
	OER		100 mA cm^{-2}	340 mV	
	EWS		10 mA cm^{-2}	1.68 V	

NiS/Ni foam	HER	1M KOH	20 mA cm^{-2}	158 mV	[8]
	OER		50 mA cm^{-2}	335 mV	
	EWS		10 mA cm^{-2}	1.64 V	
Ni_2P	HER	1M KOH	20 mA cm^{-2}	255 mV	[9]
	OER		10 mA cm^{-2}	290 mV	
	EWS		10 mA cm^{-2}	1.63 V	
NiMo HNRs/TiM	HER	1M KOH	10 mA cm^{-2}	92 mV	[10]
	OER		50 mA cm^{-2}	344 mV	
	EWS		10 mA cm^{-2}	1.64 V	
$Ni(OH)_2$/Ni/rGO-1	OER	1M KOH	10 mA cm^{-2}	130 mV	This work
	OER		50 mA cm^{-2}	331 mV	
	OER		100 mA cm^{-2}	407 mV	
$Ni(OH)_2$/Ni/rGO-3	HER	1M KOH	10 mA cm^{-2}	41 mV	This work
	HER		100 mA cm^{-2}	161.7 mV	
$Ni(OH)_2$/Ni/rGO	EWS	1M KOH	10 mA cm^{-2}	1.43 V	This work
			50 mA cm^{-2}	1.48 V	

References

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