## Electronic Supplementary Information

Controlled self-assembly of Ni foam supported poly(ethyleneimine)/reduced graphene oxide three-dimensional composite electrodes with remarkable synergistic effects for efficient oxygen evolution

Ming Zhao, ab Weiyong Yuan\*ab and Chang Ming Liab

<sup>a</sup>Institute for Clean energy & Advanced Materials, Faculty of Materials & Energy, Southwest University, Chongqing 400715, China.

<sup>b</sup>Chongqing Key Laboratory for Advanced Materials and Technologies of Clean Energies, Chongqing 400715, China.

\*Corresponding author. E-mail address: yuanweiyong@swu.edu.cn

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The catalytic performance of NF@(PEI/RGO)<sub>1</sub> (pH=7), NF@(PEI/RGO)<sub>1</sub>, NF@(PEI/RGO)<sub>2</sub> (pH=7), and NF@(PEI/RGO)<sub>2</sub> measured in 0.1 M KOH (Table S2).

FESEM images of NF after chronopotentiometry at a current density of 10 mA cm<sup>-2</sup> for 18.8 h and 350 cycles of CV measurement (Fig. S5).



Figure S1. Large-area FESEM image of pristine NF.



Figure S2. Carbon contents of multilayers versus the number of bilayers.



**Figure S3.** EDX mapping of representative areas in NF@(PEI/RGO)<sub>2</sub> (A, B and C) and NF@(PEI/RGO)<sub>3</sub> (D, E and F) obtained during FESEM imaging.

Table S1. Calculated ratios of C=O to C-O for RGO reduced in multilayers and in solution.

Catalyst	NF@(PEI/RGO)1	NF@(PEI/RGO) <sub>2</sub>	NF@(PEI/RGO) <sub>3</sub>	RGO in solution
C=O/C-O	1.49	1.69	2.00	0.55



**Figure S4.** Polarization curves (without iR correction) (A) and Tafel plots (B) of  $NF@(PEI/RGO)_1$  (pH=7),  $NF@(PEI/RGO)_1$ ,  $NF@(PEI/RGO)_2$  (pH=7), and  $NF@(PEI/RGO)_2$  in 0.1 M KOH.

**Table S2.** The catalytic performance of NF@(PEI/RGO)<sub>1</sub> (pH=7), NF@(PEI/RGO)<sub>1</sub>, NF@(PEI/RGO)<sub>2</sub> (pH=7), and NF@(PEI/RGO)<sub>2</sub> measured in 0.1 M KOH.

Electrode	Onset potential (V)	Overpotential@10.0 mA cm <sup>-2</sup> (mV)	Tafel slope (mV dec <sup>-1</sup> )
NF@(PEI/RGO) <sub>1</sub> (pH=7)	0.62	448	195
NF@(PEI/RGO) <sub>1</sub>	0.60	402	126
NF@(PEI/RGO) <sub>2</sub> (pH=7)	0.62	447	167
NF@(PEI/RGO) <sub>2</sub>	0.52	322	78



**Figure S5.** FESEM images of NF after chronopotentiometry at a current density of 10 mA cm<sup>-2</sup> for 18.8 h (A) and 350 cycles of CV measurement (B).

## REFERENCES

(1) Hummers, W. S.; Offeman, R. E. Preparation of Graphitic Oxide. *J. Am. Chem. Soc.* **1958**, *80*, 1339-1339.