Scalable Synthesis of Self-assembled Bimetallic Phosphide/N-doped Graphene Nanoflakes as Efficient Electrocatalyst for Overall Water Splitting

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Supplementary Information

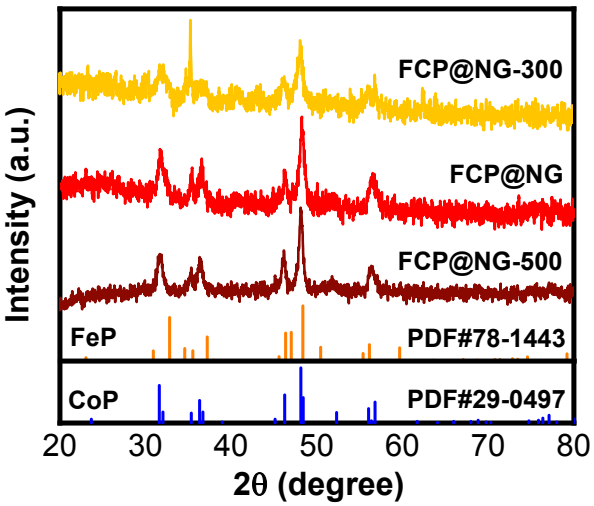

Fig. S1 XRD spectra of FCP@NG-300, FCP@NG and FCP@NG-500 (corresponding phosphorization temperature of 300 °C, 400 °C and 500 °C, respectively), as well as the reference XRD peaks of CoP and FeP.

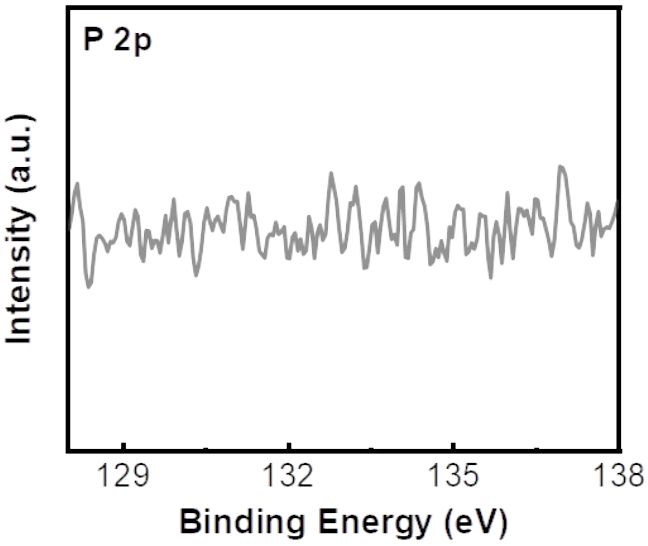

Fig. S2 XPS P 2p spectrum of rGO after phosphorization process

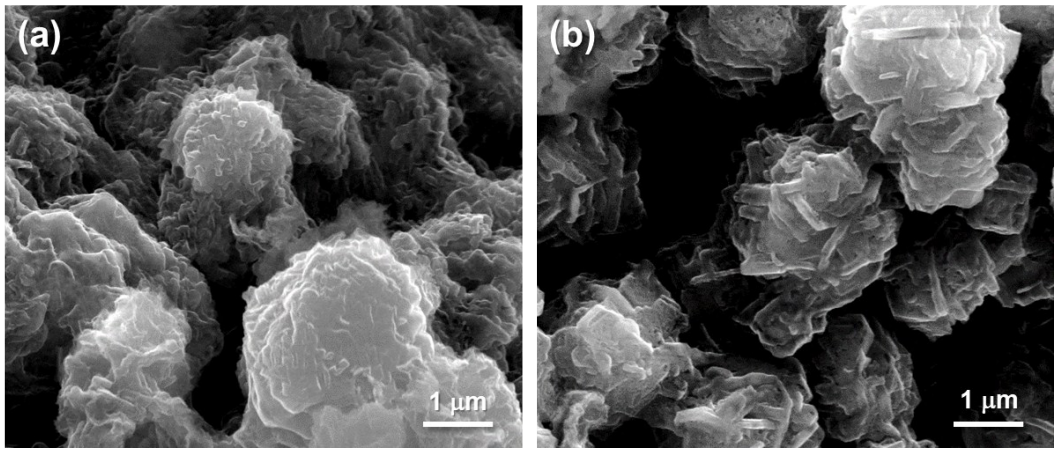

Fig. S3 SEM images of (a) FCP@NG-300 and (b) FCP@NG-500

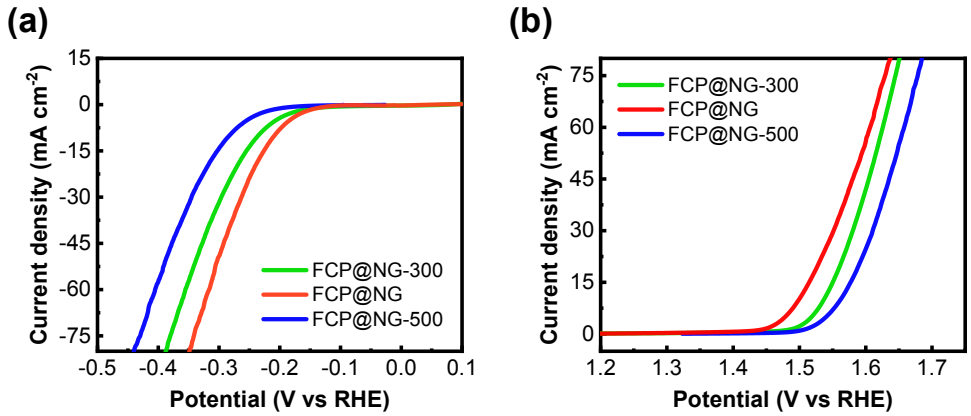

Fig. S4 HER (a) and OER (b) performance of FCP@NG-300, FCP@NG and FCP@NG-500

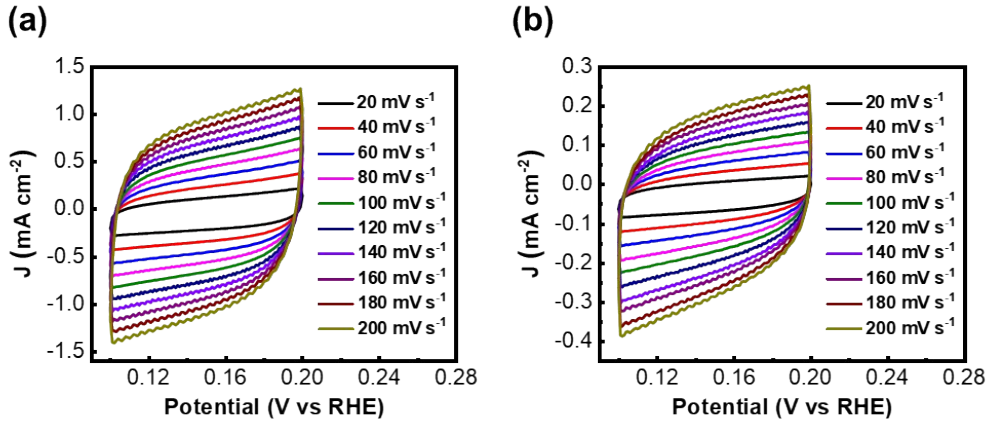

Fig. S5 Cyclic voltammogram curves of (a) FCP@NG and (b) FCP with various scan rate (20-200 mV s^{-1}) in HER scanning range.

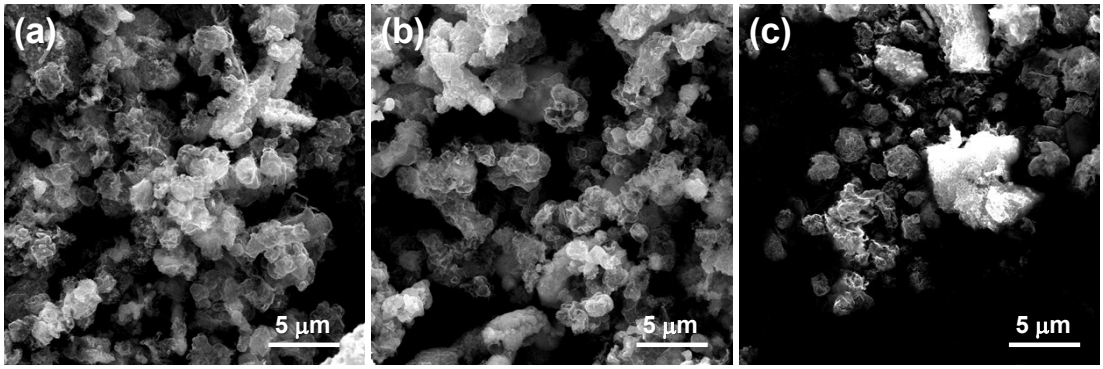

Fig. S6 SEM images of FCP@NG catalyst (a) as prepared on electrode; (b) after 1000 cycles in HER; and (c) after 1000 cycles in OER

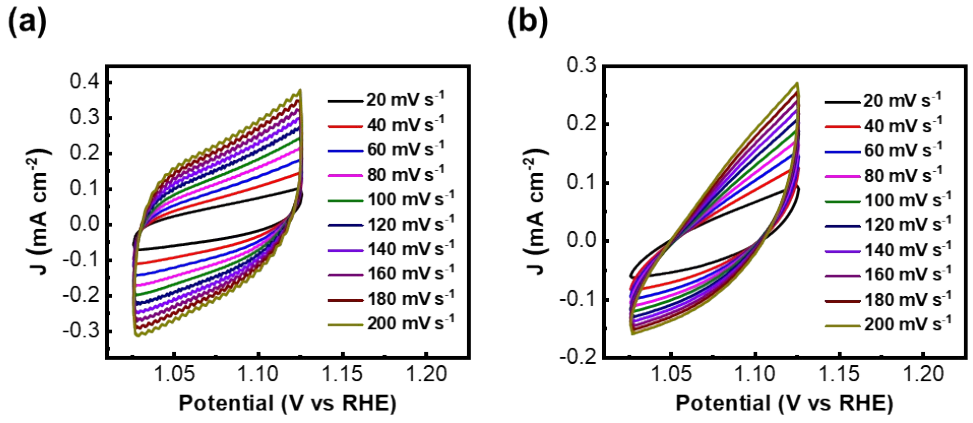

Fig. S7 Cyclic voltammogram curves of (a) FCP@NG and (b) FCP with various scan rate (20-200 mV s^{-1}) in OER scanning range.

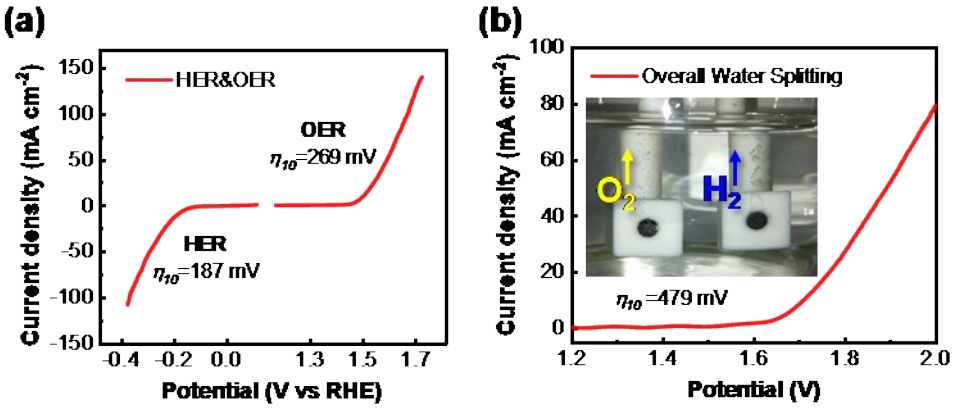

Fig. S8 LSV curves of FCP@NG in (a) HER and OER and (b) overall water splitting in 1.0 M KOH on glassy carbon electrodes (with catalyst loading of 0.27 mg cm^{-2}); with the inset of the digital photo of the two-electrode during overall water splitting.

Catalyst	Type	Electrode	Loading (mg cm^{-2})	Tafel Slope (mV dec^{-1})	η_{10} (mV)	Ref
FCP@NG	HER	GCE	0.27	76	187	This work
	OER	GCE	0.27	58	269	
	OWS	Ni foam	2.0	-	400	
FeNi–P/NCN	HER	GCE	0.48	69	190	[1]
	OER	GCE	0.48	72	240	
	OWS	Ni foam	3.0	-	270	
CoP@NPCSs	HER	GCE	0.78	109	115	[2]
	OER	GCE	0.78	103	350	
	OWS	carbon paper	2.0	-	413	

	HER	GCE	0.39	66	115	
CoP/NCNHP	OER	GCE	0.39	70	310	[3]
	OWS	carbon paper	2.0	-	420	
	HER	GCE	0.285	83	208	
$CoTe_2$@NCNTFs	OER	GCE	0.285	58	330	[4]
	OWS	Ni foam	1.0	-	440	
	HER	GCE	0.25	67	102	
FeCoNi@FeNC	OER	GCE	0.25	57	330	[5]
	OWS	Ni foam	2.0	-	~400	
	HER	Ni foam	2.0	46	107	
Ni–Co–P HNBs	OER	Ni foam	2.0	76	270	[6]
	OWS	Ni foam	2.0	-	390	

Table S1 Comparison of HER, OER and overall water splitting (OWS) performance (in 1.0 M KOH) of recently reported transition-metal-based catalysts

References:

1. Wu, Y., Tao, X., Qing, Y., Xu, H., Yang, F., Luo, S., Tian, C., Liu, M. and Lu, X., *Adv. Mater.*, 2019, **31**, 1900178.
2. Wu, K., Chen, Z., Cheong, W., Liu, S., Zhu, W., Cao, X., Sun, K., Lin, Y., Zheng, L., Yan, W., Pan, Y., Wang, D., Peng, Q., Chen, C. and Li, Y., *ACS Appl. Mater. Inter.*, 2018, **51**, 44201-44208.
3. Pan, Y., Sun, K., Liu, S., Cao, X., Wu, K., Cheong, W., Chen, Z., Wang, Y., Li, Y., Liu, Y., Wang, D., Peng, Q., Chen, C. and Li, Y., *J. Am. Chem. Soc.*, 2018, **140**, 2610-2618.
4. Wang, X., Huang, X., Gao, W., Tang, Y., Jiang, P., Lan, K., Yang, R., Wang, B. and Li, R., *J. Mater. Chem. A*, 2018, **6**, 3684-3691.
5. Zhang, Q., Webster, R.F., Cheong, S., Tilley, R.D., Lu, X. and Amal, R., *Part. Part. Syst. Char.*, 2018, **36**, 252-261.
6. Hu, E., Feng, Y., Nai, J., Zhao, D., Hu, Y. and Lou, X.W.D., *Energ. Environ. Sci.*, 2018, **11**, 872-880.