Electronic Supplementary Information

Direct conversion of coordination compounds into  $Ni_2P$ nanoparticles entrapped in 3D mesoporous graphene for efficient hydrogen evolution reaction

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## 1. Supplementary Table S1 and Figures S1-S12

**Table S1.** Activity comparison table for HER in acidic media by nickel phosphide based catalysts.  $\eta_{10}$  indicates the overpotential required to drive a current density of -10 mA cm<sup>-2</sup>.

Reference	Catalyst	Loading (mg cm <sup>-2</sup> )	Electrolyte	η <sub>10</sub> (mV)	Tafel slope (mV dec <sup>-1</sup> )
This work	Ni <sub>2</sub> P@mesoG	1	0.5 M H <sub>2</sub> SO <sub>4</sub>	98	56
		3	$0.5 \text{ M} \text{H}_2 \text{SO}_4$	79	83
[S1]	Ni <sub>2</sub> P NPs/Ti	~1	$0.5 \text{ M} \text{H}_2 \text{SO}_4$	~118	~46
[S2]	Ni <sub>2</sub> P NPs	0.38	$1 \text{ M H}_2\text{SO}_4$	122	87
[S3]	Ni <sub>12</sub> P <sub>5</sub> NPs/Ti	3	$0.5 \text{ M} \text{H}_2 \text{SO}_4$	107	63
[S4]	Ni <sub>2</sub> P/Ti	2	1 M H <sub>2</sub> SO <sub>4</sub>	122	60
[85]	NiP <sub>2</sub> NS/CC	2.6	0.5 M H <sub>2</sub> SO <sub>4</sub>	75	~51
[S6]	Ni <sub>5</sub> P <sub>4</sub>	177	1 M H <sub>2</sub> SO <sub>4</sub>	23	33
[S7]	Ni <sub>5</sub> P <sub>4</sub> -Ni <sub>2</sub> P NS/Ni foam	-	0.5 M H <sub>2</sub> SO <sub>4</sub>	120	79.1
[S8]	Ni <sub>2</sub> P-G@Ni foam	-	0.5 M H <sub>2</sub> SO <sub>4</sub>	55	~30/107
[89]	Ni <sub>5</sub> P <sub>4</sub>	1.99	$0.5 \text{ M} \text{H}_2 \text{SO}_4$	118	42



Figure S1. TEM images of mesoG before (a) and after (b) acid etching.



**Figure S2.** (a) STEM image of Ni<sub>2</sub>P@mesoG and (b-d) corresponding EDS mapping images for (b) nickel, (c) phosphorus, and (d) nitrogen.



Figure S3. N<sub>2</sub> adsorption-desorption isotherms of Ni@mesoG and Ni<sub>2</sub>P@mesoG.



**Figure S4.** XRPD patterns of Ni@mesoG samples phosphidated at 300, 400 (Ni<sub>2</sub>P@mesoG), 500, and 600 °C.



Figure S5. TEM images of Ni@mesoG samples phosphidated at 300, 400 (Ni<sub>2</sub>P@mesoG), 500, and 600 °C.



Figure S6. N<sub>2</sub> adsorption-desorption isotherms of Ni@mesoG samples phosphidated at 300, 400 (Ni<sub>2</sub>P@mesoG), 500, 600 °C.



**Figure S7.** Ni  $2p_{3/2}$  XPS spectra of Ni@mesoG samples phosphidated at (a) 300, (b) 500, and (c) 600 °C.



**Figure S8.** P  $2p_{3/2}$  XPS spectra of Ni@mesoG samples phosphidated at (a) 300, (b) 500, and (c) 600 °C.



**Figure S9.** C 1*s* XPS spectra of Ni@mesoG samples phosphidated at (a) 300, (b) 500, and (c) 600 °C.



Figure S10. (a) TEM image and (b) XRPD pattern of  $Ni_2P/mesoG$ .



**Figure S11.** Polarization curves for  $Ni_2P@mesoG$  with different catalyst loadings in (a) 0.5 M H<sub>2</sub>SO<sub>4</sub>, and (b) 1 M KOH.



Figure S12. (a) Nyquist plots for the impedance spectra obtained at  $\eta = 100 \text{ mV}$  (vs RHE) in 1 M KOH. The empty triangles and the solid lines represent numerical raw data and fitting results, respectively. (b) Tafel plots in 1 M KOH. The Tafel plots were depicted with error bar and without correction for ohmic drop.

## 2. References for Electronic Supplementary Information

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