In situ evolution of active phase on stainless steel mesh toward cost-effective bifunctional electrode for energy storage and conversion

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Experimental Section

Preparation of EOSSP. At first, a piece of SS (304, 1000 mesh, 1.0 cm × 1.2 cm)) was cleaned by alcohol, acetone, and distilled water in turn. And then, all the clean SS was calcined in air for 2 h with the temperature of 300 °C. After the heating period, the asobtained OSS was immersed into 1 M HCl for 10 s at room temperature. For the process of phosphorization, the obtained EOSS and NaH₂PO₂ were put at two separate positions in a porcelain-boat with 1 g NaH₂PO₂ at the upstream side of a furnace, and then heated at 250°C for 2 h in static N₂ atmosphere.

Physicochemical Characterizations. The surface morphology of the as-prepared samples were analyzed using field emission scanning electron microscopy (FE-SEM, JEOL JSM-7001F). The structure of the samples was analyzed by powder X-ray diffraction (XRD, Bruker , D8 ADVANCE) with K α radiation (λ = 1.5418 Å). The chemical state and compositions of the samples were analyzed using X-ray Photoelectron Spectroscopy (XPS, ESCALab250).

Electrochemical Measurements.

All the electrochemical measurements were carried out in a three-electrode system on an electrochemical workstation (CHI760E). The samples prepared above were used directly as working electrodes, graphite carbon rod as counter electrode and saturated calomel electrode (SCE) as reference electrode, in 1 M KOH electrolyte. Before measurements, the electrodes were electrochemically pretreated with repetitive cyclic voltammetry (CV) for 50 scan cycles from -1.5 to 0 V vs. SCE in 1.0 M KOH. This was performed to increase the oxygenated surface functional groups and further improve the hydrophilicity and wettability of the surface.

Supplementary Figures



Figure S1 Digital photo of the EOSSP sample (20 cm × 20 cm).



Figure S2 Raman spectra of OSS. The spectrum of OSS showed eight Raman bands with peaks at 237, 257, 303, 422, 510, 621, 670 and 830 cm⁻¹, which were the characteristic peaks of the Fe_2O_3 phase.^[1-2]



Figure S3 XRD pattern of SS, OSS, and EOSSP samples, respectively.



Figure S4 The XPS survey spectra of EOSSP.



Figure S5 CV curves for (a) EOSSP; (b) OSS; (c) EOSS samples in the double layer region with scan rates ranging from 2 to 10 mV s⁻¹. (d) Charging current density with different scan rates, indicating the largest electrochemical active area of EOSSP than that of OSS and EOSS.



Figure S6 Nyquist plots of EOSSP, EOSS and OSS with a potential of -0.3 V versus RHE.



Figure S7 XPS spectra of (a) Fe 2p and (b) P 2p regions of EOSSP after HER.



Figure S8 SEM images of EOSSP before (a-b) and after (c-d) electrolyzing HER.



Figure S9 Nyquist plots of EOSSP with the potential of -0.3 V versus RHE before and after long-term HER tests.



Figure S10 (a) Polarization curve and (b) time-dependence of the current density of Pt/C catalyst.



Figure S11 CV curves of the OSS, EOSS and EOSSP electrodes at a scan rate of 100 mV s⁻¹.



Figure S12 GCD curves of EOSSP electrode under various current densities.



Figure S13 Areal capacitance versus current density of the EOSSP electrode.



Figure S14 GCD curves of bare SS, OSS and EOSS electrodes at current density of 1 mA cm⁻².



Figure S15 SEM images of EOSSP before (a-b) and after (c-d) long-term cyclic tests.

Catalysts	Electrolyte	η (mV)	Current	Referen
			retention	ce
Fe_2O_3 nanoparticles	1 M KOH	321 mV at 10 mA cm ⁻²	—	3
FeP nanorod arrays	1 М КОН	218 mV at 10 mA cm ⁻²	81% after 20 h	4
FeP nanoparticles@N,P-	1 M KOH	214 mV at 10 mA cm ⁻²	~70% after 10 h	5
carbon				5
FeP arrays	1 M KOH	194 mV at 10 mA cm ⁻²	70 % after 9 h	6
FeP/C	1 М КОН	185 mV at 10 mA cm ⁻²	~100% after 24 h	7
EOSSP	1 M KOH	238 mV at 30 mA cm ⁻²	93% after 200 h	This
				work

Table S1 Comparison of the HER performance of Fe-based catalysts.

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