Supporting Information


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Materials: Cobalt chloride hexahydrate (CoCl$_2$·6H$_2$O, Aladdin), Iron (III) nitrate nonahydrate (Fe(NO$_3$)$_3$·9H$_2$O, Aladdin), 2-methylimidazole (C$_4$H$_6$N$_2$, Aladdin), sodium hypophosphite (NaH$_2$PO$_2$·H$_2$O, Aladdin), ethanol (Aladdin), Nafion (Sigma-Aldrich), IrO$_2$ (Sigma-Aldrich). All reagents are purchased from commercial companies without further treatment. Deionized water acts as solvent in the experiment.

Electrochemical Measurements: OER and HER tests were carried out at room temperature with an electrochemical workstation (CHI660E, Shanghai, China) with a standard three-electrode system. Comprising a graphite rod as the counter electrode, a saturated Hg/HgO as the reference electrode, and Ni foam coated with catalyst as working electrode. To prepare the working electrode, the as-prepared electrocatalyst (5 mg) and Nafion (20 µL, 5 wt%) were scattered in 980 µL isopropanol-water solution (volume ratio of isopropanol to water is 9:1) and then mixture was ultrasonicated for 1 h to form homogeneous ink. The well-mixed ink was used to coat and cover Ni foam surface to achieve a mass loading of 3 mg cm$^{-2}$. For comparison, IrO$_2$ ink was prepared by the same method. Ni foam substrate was thoroughly cleaned with 0.5 M H$_2$SO$_4$ (15 min), absolute ethanol (20 min) and finally rinsed with deionized water (20 min) in an ultrasound bath. Polarization curves were obtained at room temperature with a scan rate of 5 mV s$^{-1}$ in 1.0 M KOH. All potentials were transformed to the reversible hydrogen electrode (RHE) by following the equation: $E_{\text{RHE}} = E_{\text{Hg/HgO}} + 0.059pH + 0.098V$. The overpotential ($\eta$) of OER was calculated by the formula ($\eta$ (V) = $E_{\text{RHE}} - 1.23$ V) and the overpotential ($\eta$) of HER was calculated by the following formula ($\eta$ (V) = - $E_{\text{RHE}}$). The obtained polarization curves were iR-compensated. Tafel plots were
fitted according to the formula ($\eta = \log j + a$), where $\eta$ is the overpotential, $j$ represents the current density, $a$ and $b$ represent constant and Tafel slope, respectively.

Electrochemical impedance spectroscopy (EIS) was performed at open circuit potential with the frequency range of $10^{-1}$ Hz to $10^5$ Hz. The double layer capacitance ($C_{dl}$) was determined by cyclic voltammetry curves measured by scan rates of 80, 100, 120, 140 and 160 mV s$^{-1}$. 
Figure S1. SEM image of Fe$_{0.27}$Co$_{0.73}$ precursor
Figure S2. SEM images of Fe$_{0.27}$Co$_{0.73}$ precursor that was calcined in nitrogen without adding phosphorus source.
Figure S3. XRD patterns of ZIF-67 and Fe$_{0.27}$Co$_{0.73}$ precursor
Figure S4. XRD patterns of Fe$_{0.22}$Co$_{0.78}$P, Fe$_{0.44}$Co$_{0.56}$P, CoP.
Figure S5. XRD patterns for a slow scan of 1 degree per minute for a specific diffraction angle including CoP, Fe$_{0.22}$Co$_{0.78}$P and Fe$_{0.44}$Co$_{0.56}$P.
Figure S6. Low-resolution TEM image of Fe$_{0.27}$Co$_{0.73}$P
Figure S7. (a) N$_2$ adsorption-desorption isotherms and (b) pore size distributions of Fe$_{0.27}$Co$_{0.73}$P.
Figure S8. XPS spectrum of Fe$_{0.27}$Co$_{0.73}$P and Fe$_{0.22}$Co$_{0.78}$P in Fe 2p region.
**Figure S9.** Multi-step current curve for Fe$_{0.27}$Co$_{0.73}$P/NF without iR correction.
Figure S10. Polarization curves of $\text{Fe}_{0.27}\text{Co}_{0.73}\text{P/NF}$, $\text{Fe}_{0.22}\text{Co}_{0.78}\text{P/NF}$ and $\text{Fe}_{0.44}\text{Co}_{0.56}\text{P/NF}$. 
Figure S11. Tafel slopes of Fe$_{0.27}$Co$_{0.73}$P/NF, Fe$_{0.22}$Co$_{0.78}$P/NF and Fe$_{0.44}$Co$_{0.56}$P/NF.
Figure S12. Polarization curves of Fe$_{0.27}$Co$_{0.73}$P/NF, Fe$_{0.22}$Co$_{0.78}$P/NF and Fe$_{0.44}$Co$_{0.56}$P/NF.
Figure S13. Tafel slopes of Fe$_{0.27}$Co$_{0.73}$P/NF, Fe$_{0.22}$Co$_{0.78}$P/NF and Fe$_{0.44}$Co$_{0.56}$P/NF.
Figure S14. Polarization curves for Fe$_{0.27}$Co$_{0.73}$P/NF initial and after 3000 CV cycles.
Figure S15. XPS spectrum of Fe$_{0.27}$Co$_{0.73}$P before and after OER test in Co 2p region and Fe 2p region.
Figure S16. CVs of (a) CoP/NF and (c) Fe\textsubscript{0.27}Co\textsubscript{0.73}P/NF and corresponding current densities of (b) CoP/NF and (d) Fe\textsubscript{0.27}Co\textsubscript{0.73}P/NF plotted as a function of scan rate.
Figure S17. CVs of (a) Fe$_{0.22}$Co$_{0.78}$P/NF and (c) Fe$_{0.44}$Co$_{0.56}$P/NF and corresponding current densities of (b) Fe$_{0.22}$Co$_{0.78}$P/NF and (d) Fe$_{0.44}$Co$_{0.56}$P/NF plotted as a function of scan rate.
Figure S18. EIS spectra of Fe$_{0.27}$Co$_{0.73}$P/NF, Fe$_{0.22}$Co$_{0.78}$P/NF, CoP/NF and Fe$_{0.44}$Co$_{0.56}$P/NF.
**Table S1.** Comparison of OER performance in 1.0 M KOH for Fe$_{0.27}$Co$_{0.73}$P /NF with other recently reported representative OER electrocatalysts in alkaline solution.

<table>
<thead>
<tr>
<th>Electrocatalsys</th>
<th>Current density (mA cm$^{-2}$)</th>
<th>Overpotential (mV)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe$<em>{0.27}$Co$</em>{0.73}$P/NF</td>
<td>10</td>
<td>251</td>
<td>This work</td>
</tr>
<tr>
<td>NiCo LDH</td>
<td>20</td>
<td>393</td>
<td>Nano Lett. 2015, 15, 1421.</td>
</tr>
<tr>
<td>Ni$<em>{1.85}$Fe$</em>{0.15}$P NSAs/NF</td>
<td>20</td>
<td>270</td>
<td>ACS Appl. Mater. Interfaces 2017, 9, 26001.</td>
</tr>
<tr>
<td>Ni$_3$N/NF</td>
<td>20</td>
<td>399</td>
<td>J. Mater. Chem. 2015, 3, 8171.</td>
</tr>
</tbody>
</table>
Video S1. Video of Fe$_{0.27}$Co$_{0.73}$P/NF electrodes electrolyze water in electrolyzed water reaction tank containing 1.0 M KOH alkaline solution.
Video S2. Video demonstration uses a drainage method to collect hydrogen and oxygen for characterizing the Fe$_{0.27}$Co$_{0.73}$P Faradaic efficiency.