**Fig. S1** The electrochemical processes of electrode preparation and the photograph of bare NF and C6FN-phy.

**Fig. S2** The electrochemical processes of electrode preparation and the photograph of bare NF and N-phy.
Fig. S3 EDS images of C4FN-phy, C5FN-phy, C6FN-phy and C7FN-phy. The EDS analyses were carried out to verify the actual metal proportions deposited on NF surface. As a result, the actual metal atomic ratios almost agree with the original feeding ratios.

Fig. S4 XRD images of the electrodes.
**Fig. S5** RM spectra of C6FN-phy.

**Fig. S6** XPS images of C6FN-phy.
Fig. S7 The OER performances of electrodes and corresponding Tafel slopes.

Fig. S8 Chronopotentiometric curves of C6FN-phy for OER at 10 and 20 mA cm$^{-2}$. 
**Fig. S9** EIS of the electrodes at the applied potential of 1.58 V vs RHE and the equivalent circuit diagram.
Fig. S10 Electrochemical double-layer capacitance measurements of various electrodes at the scan rates of 5, 10, 20, 50 and 100 mV s\(^{-1}\) in 1 M KOH and the linear fitting curves of the charged currents at 0.915 V of each electrode vs. scan rates.
Fig. S11 (a-g) XPS images of C6FN-phy after OER stability measurement.

Fig. S12 The corresponding Tafel slopes of electrodes in Figure 4a.
Fig. S13 (a-g) XPS images of C6FN-phy after HER stability measurement.

The calculation of faradaic efficient:
At first, the chronopotentiometric measurement was applied to the three-electrode system, with the gas-collecting method to gather the volume of the generated O\textsubscript{2} in saturated solution of oxygen. The state equation of gas (PV=nRT, normal temperature and pressure) was used to obtain the molar of actual gas. Comparing to the theoretical O\textsubscript{2} yield, the corresponding faradaic efficient image is shown in Fig. 3d:
### Table S1. Comparisons of the various OER catalysts in alkaline electrolyte according to the reports and this paper.

<table>
<thead>
<tr>
<th>Catalysts</th>
<th>Substrate</th>
<th>Tafel slope</th>
<th>Current density (J, mA cm(^{-2}))</th>
<th>(\eta) at corresponding J (mV)</th>
<th>Stability</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni(<em>{0.9})Fe(</em>{0.1})/NC, 1 M KOH</td>
<td>Glass carbon</td>
<td>45</td>
<td>10</td>
<td>330</td>
<td>An increase of 25 mV after 10000 cycles at 10 mA cm(^{-2})</td>
<td>1</td>
</tr>
<tr>
<td>CoNi(OH)(_x), 1 M KOH</td>
<td>Cu film</td>
<td>77</td>
<td>10</td>
<td>280</td>
<td>64% current density retention after 24 hours' electrocatalysis at (\eta = 320) mV</td>
<td>2</td>
</tr>
<tr>
<td>NiCo-LDH, 1 M KOH</td>
<td>Carbon paper</td>
<td>40</td>
<td>10</td>
<td>367</td>
<td>An increase of 22 mV after 6 hours' electrocatalysis at 10 mA</td>
<td>3</td>
</tr>
<tr>
<td>NiCo(_2)O(_4) nanowires, 1 M KOH</td>
<td>Ti foil</td>
<td>60</td>
<td>10</td>
<td>370</td>
<td>~ 93% current density retention after 20 hours' electrocatalysis at (\eta = 420) mV</td>
<td>4</td>
</tr>
<tr>
<td>3D-NA/Ni/NiNPA(_s), 1 M KOH</td>
<td>Cu-coated polyimide film</td>
<td>96</td>
<td>10</td>
<td>285</td>
<td>94 % current density retention after 175 hours' electrocatalysis at (\eta = 340) mV</td>
<td>5</td>
</tr>
<tr>
<td>Ni/Ni(_3)N, 1 M KOH</td>
<td>NF</td>
<td>60</td>
<td>10</td>
<td>~322</td>
<td>~ 96% current density retention after 12 hours' electrocatalysis at 100 mA cm(^{-2})</td>
<td>6</td>
</tr>
<tr>
<td>Ni(_2)P, 1 M KOH</td>
<td>GC</td>
<td>59</td>
<td>10</td>
<td>290</td>
<td>An increase of 10 mV after 10 hours' electrocatalysis at 10 mA cm(^{-2})</td>
<td>7</td>
</tr>
<tr>
<td>ECT-Co(<em>{0.37})Ni(</em>{0.26})Fe(<em>{0.37})O(</em>{4}), 1 M KOH</td>
<td>carbon fiber cloth</td>
<td>37.6</td>
<td>200</td>
<td>293</td>
<td>~ 95% current density retention after 100 hours' electrocatalysis at 20 mA cm(^{-2})</td>
<td>8</td>
</tr>
<tr>
<td>NiFe/NF, 1 M KOH</td>
<td>NF</td>
<td>28</td>
<td>80</td>
<td>270</td>
<td>~ 100% current density retention after 10 hours' electrocatalysis at 100 mA cm(^{-2})</td>
<td>9</td>
</tr>
<tr>
<td>NiFe films, 1 M NaOH</td>
<td>NF</td>
<td>-</td>
<td>100</td>
<td>300</td>
<td>~ 100% current density retention after 72 hours' electrocatalysis at 100 mA cm(^{-2})</td>
<td>10</td>
</tr>
<tr>
<td>NiS, 1 M KOH</td>
<td>NF</td>
<td>89</td>
<td>50</td>
<td>335</td>
<td>~ 100% current density retention after 35 hours' electrocatalysis at 13 mA cm(^{-2})</td>
<td>11</td>
</tr>
<tr>
<td>NiSe, 1 M KOH</td>
<td>NF</td>
<td>64</td>
<td>100</td>
<td>314</td>
<td>~ 99% current density retention after 12 hours' electrocatalysis at 100 mA cm(^{-2})</td>
<td>12</td>
</tr>
<tr>
<td>FeOOH/Co/OOH HNTAs, 1 M NaOH</td>
<td>NF</td>
<td>32</td>
<td>21</td>
<td>250</td>
<td>Anodic polarization tests at current densities of 20, 50, 100, and 200 mA cm(^{-2}) for 50 h, showing that the overpotentials remain unchanged</td>
<td>13</td>
</tr>
<tr>
<td>NiP, 1 M KOH</td>
<td>NF</td>
<td>23</td>
<td>191</td>
<td>350</td>
<td>From 1.33V gradually increases to</td>
<td>14</td>
</tr>
</tbody>
</table>
1.45 V vs. RHE after 0.2 h, and then remains fairly stable at this potential up to 26 h

<table>
<thead>
<tr>
<th>Material</th>
<th>NF</th>
<th>Percent</th>
<th>Current Density Retention</th>
</tr>
</thead>
<tbody>
<tr>
<td>CN-phy</td>
<td>86</td>
<td>98%</td>
<td>98% current density retention after 10 hours’ electrocatalysis at 100 mA cm$^{-2}$</td>
</tr>
<tr>
<td>C6FN-phy</td>
<td>24</td>
<td>~ 96% and 100%</td>
<td>~ 96% and 100% current density retention after 70 and 32 hours’ electrocatalysis at 200 and 100 mA cm$^{-2}$</td>
</tr>
<tr>
<td>FN-phy</td>
<td>75</td>
<td>~ 99%</td>
<td>~ 99% current density retention after 10 hours’ electrocatalysis at 100 mA cm$^{-2}$</td>
</tr>
<tr>
<td>N-phy</td>
<td>60</td>
<td>~ 99%</td>
<td>~ 99% current density retention after 10 hours’ electrocatalysis at 100 mA cm$^{-2}$</td>
</tr>
</tbody>
</table>
### Table S2. Comparisons of the two-electrode configuration performance according to the reports and this paper in 1 M KOH.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Substrate</th>
<th>$\eta$@10 mA cm$^{-2}$ (mV)</th>
<th>Current density (J, mA cm$^{-2}$)</th>
<th>$\eta$ at corresponding J (mV)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>FeNi$_3$N/NF</td>
<td>NF</td>
<td>390</td>
<td>~90</td>
<td>770</td>
<td>15</td>
</tr>
<tr>
<td>Ni–P foam</td>
<td>NF</td>
<td>410</td>
<td>100</td>
<td>820</td>
<td>14</td>
</tr>
<tr>
<td>NiSe/NF</td>
<td>NF</td>
<td>400</td>
<td>~60</td>
<td>770</td>
<td>12</td>
</tr>
<tr>
<td>NiCo$_3$S$_4$ NW</td>
<td>NF</td>
<td>400</td>
<td>~70</td>
<td>770</td>
<td>16</td>
</tr>
<tr>
<td>Ni$_{2.3%}$CoS$_2$/CC</td>
<td>Carbon cloth</td>
<td>430</td>
<td>60</td>
<td>770</td>
<td>17</td>
</tr>
<tr>
<td>NiCoP/rGO</td>
<td>-</td>
<td>360</td>
<td>60</td>
<td>670</td>
<td>18</td>
</tr>
<tr>
<td>C6FN-phy</td>
<td>NF</td>
<td>460</td>
<td>100</td>
<td>660</td>
<td>This work</td>
</tr>
</tbody>
</table>

**References:**
