Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting information

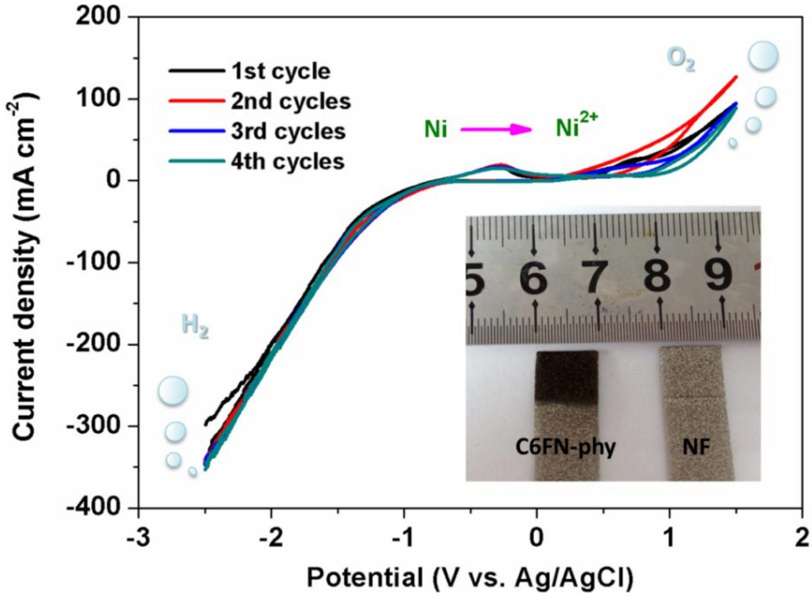

Fig. S1 The electrochemical processes of electrode preparation and the photograph of bare NF and C6FN-phy.

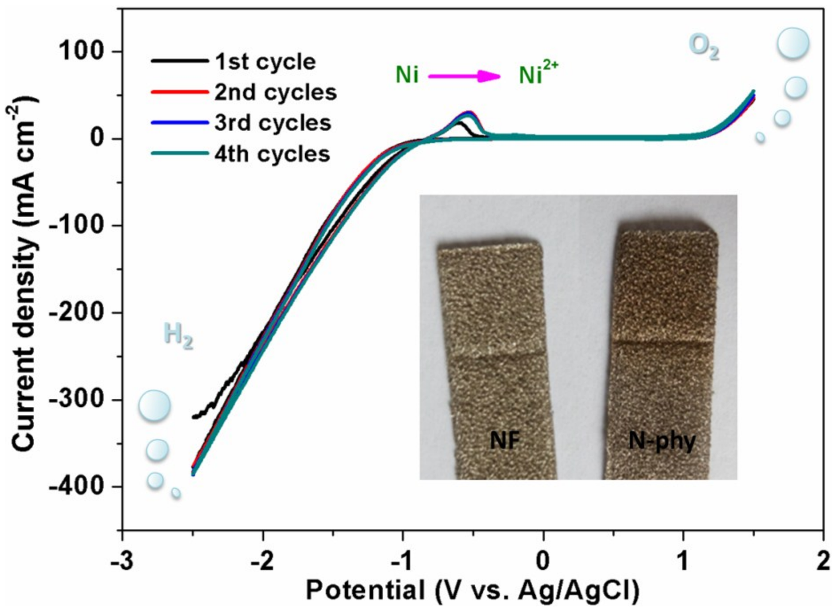

Fig. S2 The electrochemical processes of electrode preparation and the photograph of bare NF and N-phy.

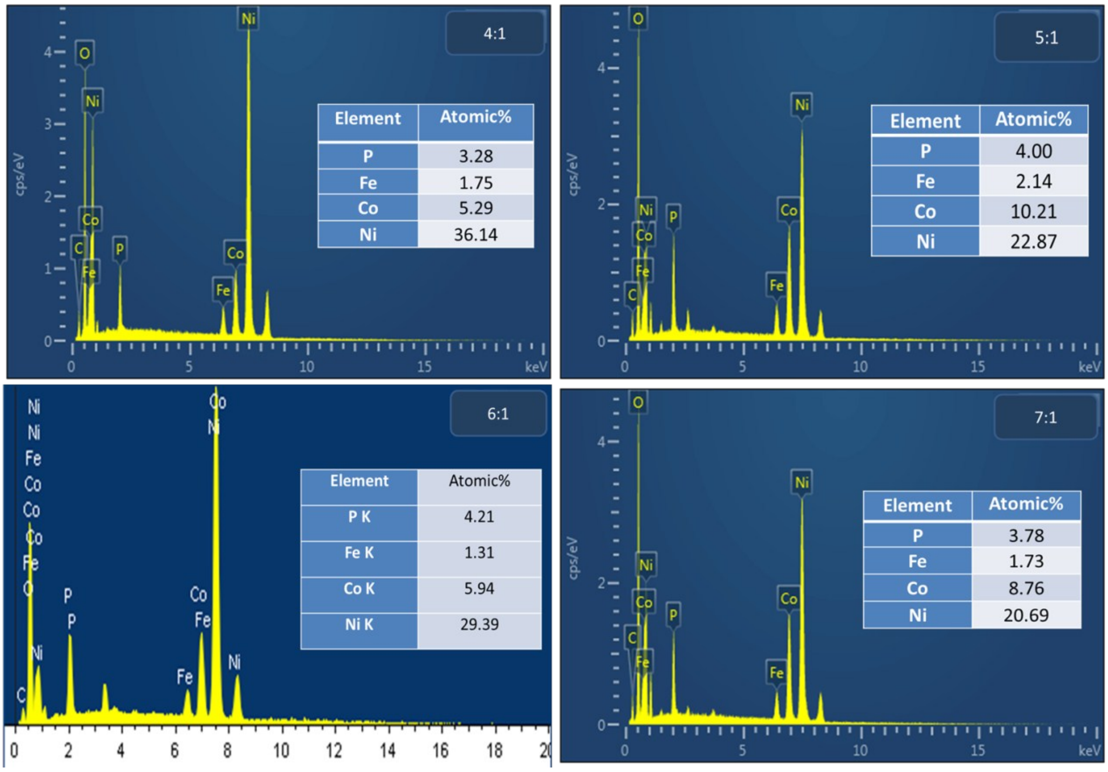

Fig. S3 EDS images of C4FN-phy , C5FN-phy , C6FN-phy and C7FN-phy.

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.

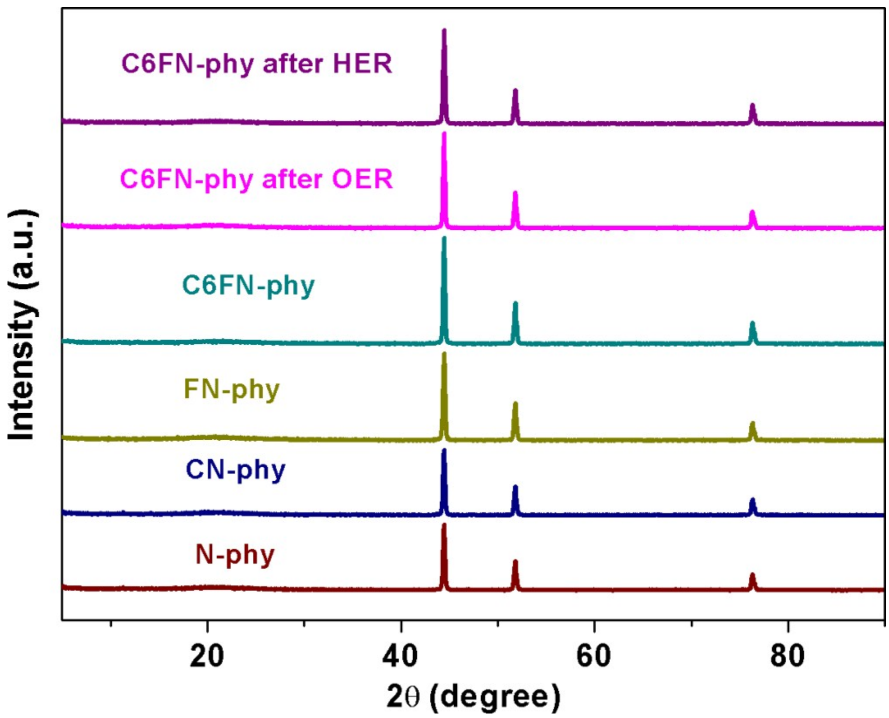

Fig. S4 XRD images of the electrodes.

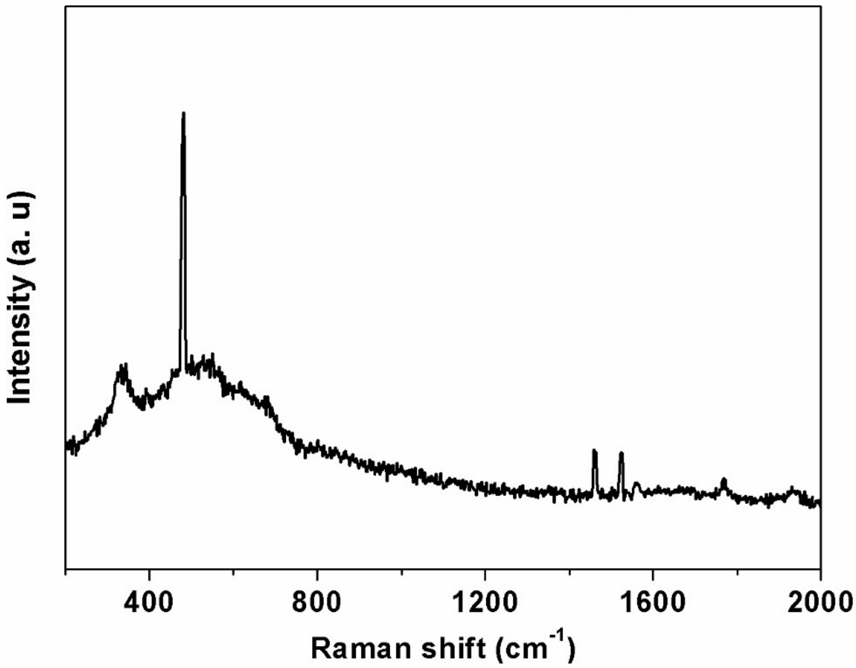

Fig. S5 RM spectra of C6FN-phy.

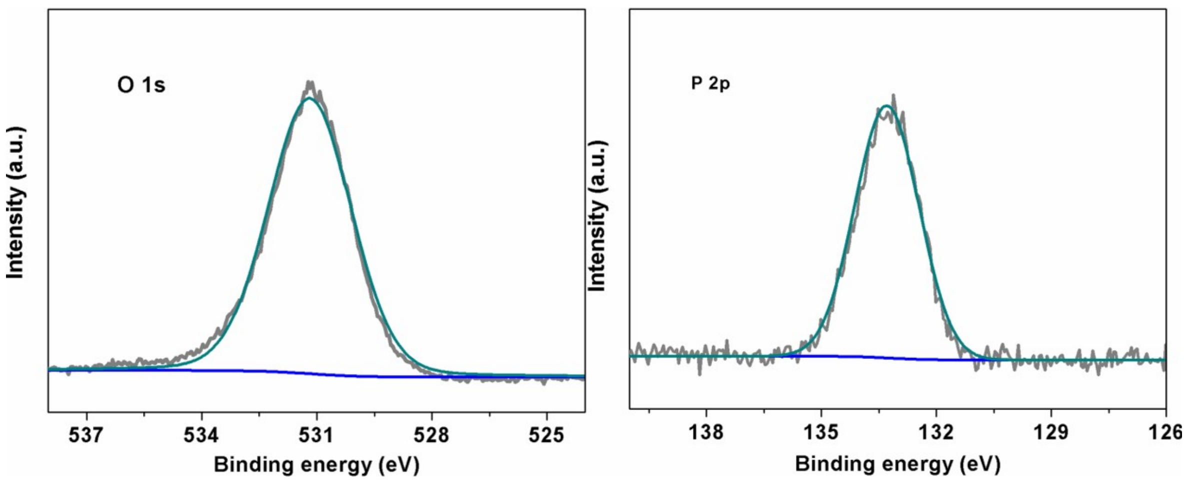

Fig. S6 XPS images of C6FN-phy.

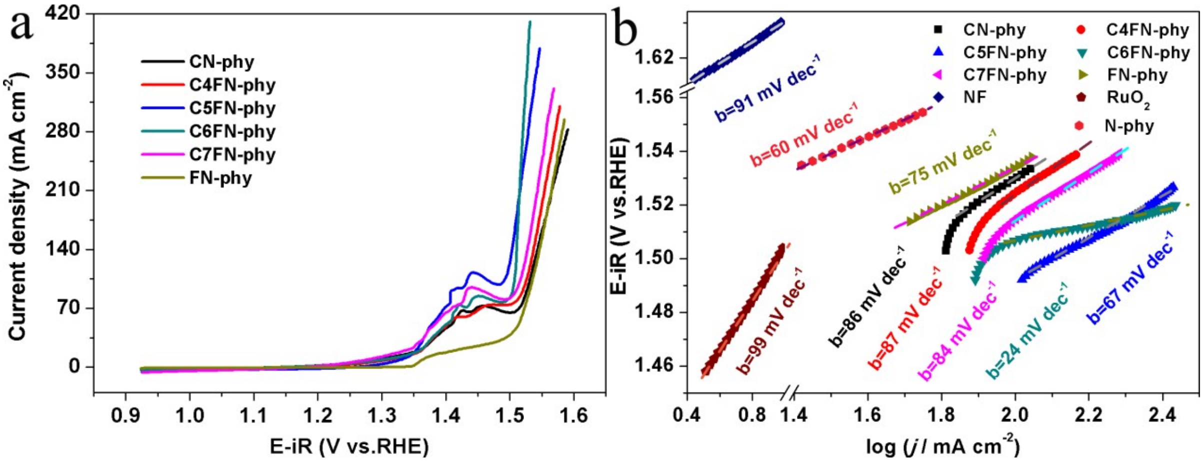

Fig. S7 The OER performances of electrodes and corresponding Tafel slopes.

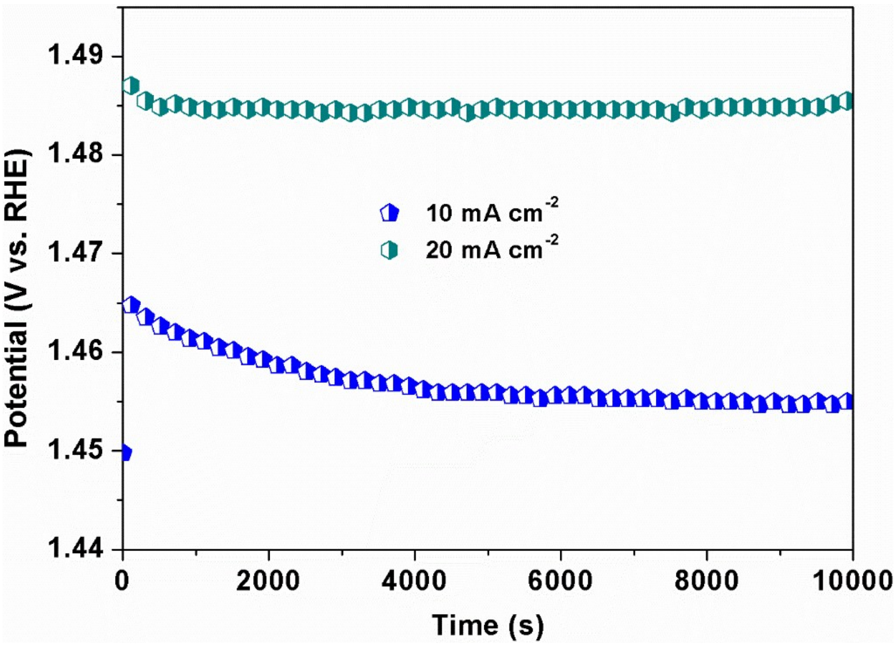

Fig. S8 Chronopotentiometric curves of C6FN-phy for OER at 10 and 20 mA cm^{-2}.

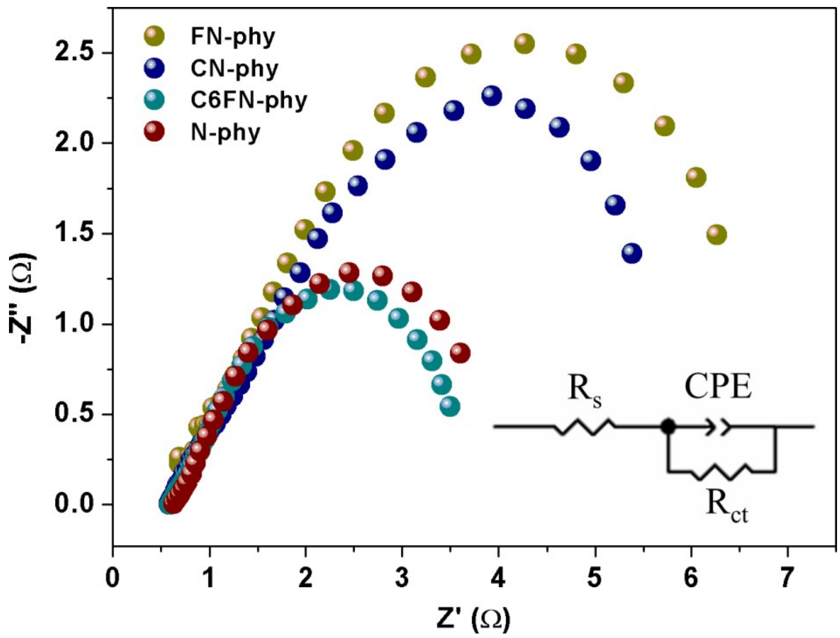

Fig. S9 EIS of the electrodes at the applied potential of 1.58 V vs RHE and the equivalent circuit diagram.

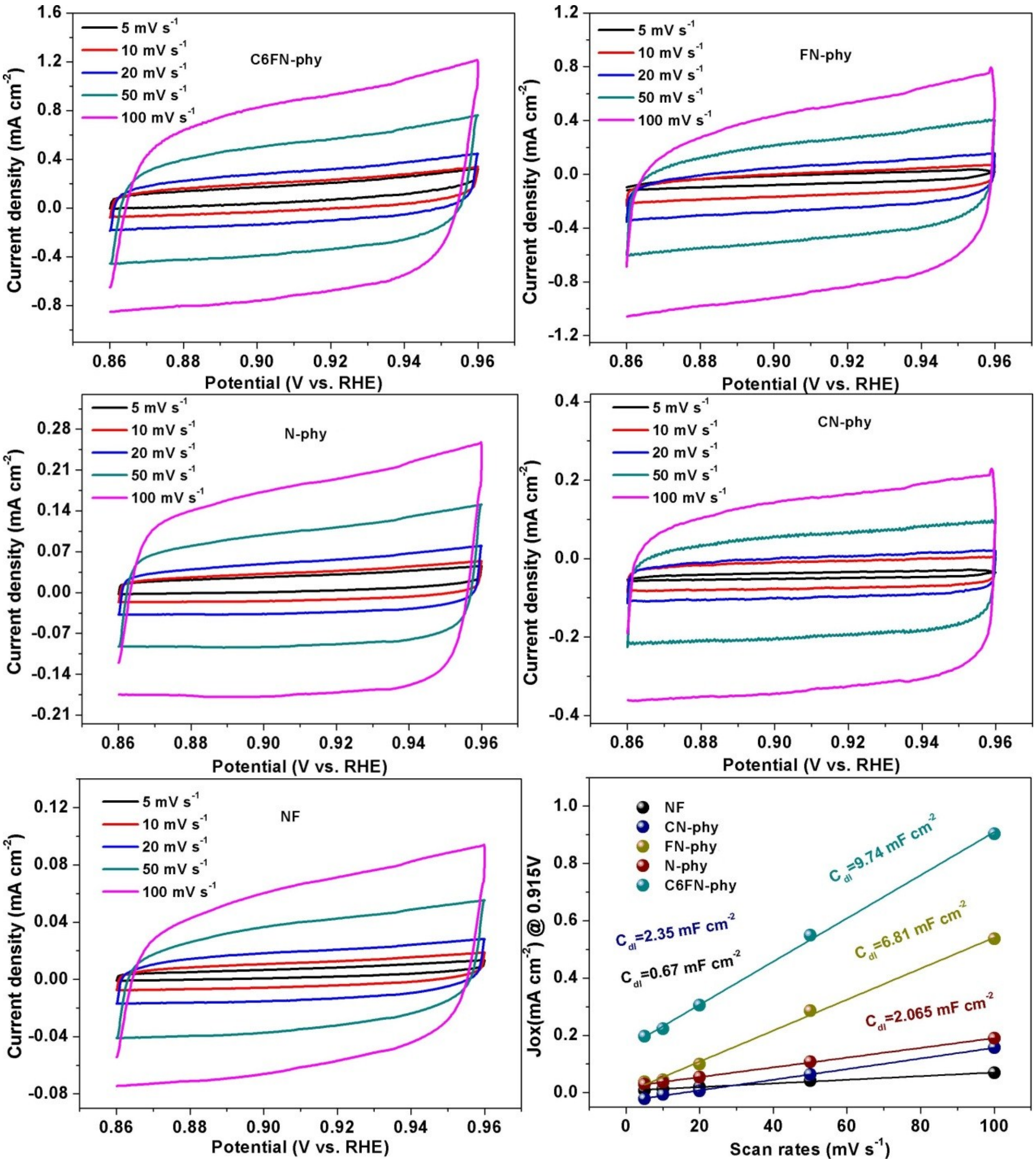

Fig. S10 Electrochemical double-layer capacitance measurements of various electrodes at the scan rates of 5, 10, 20, 50 and 100 mV s^{-1} in 1 M KOH and the linear fitting curves of the charged currents at 0.915 V of each electrode vs. scan rates.

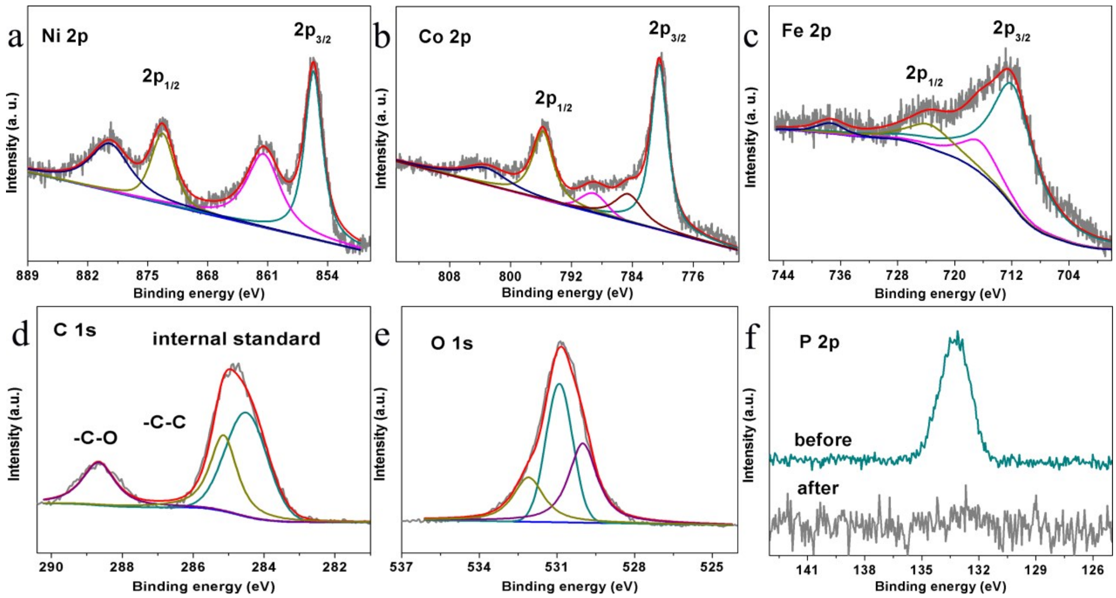

Fig. S11 (a-g) XPS images of C6FN-phy after OER stability measurement.

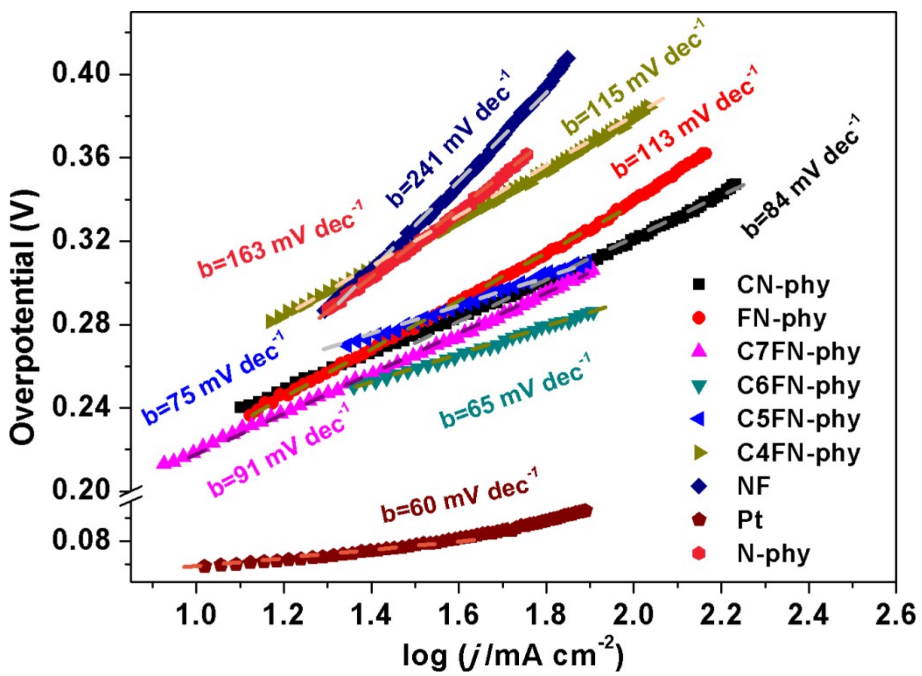

Fig. S12 The corresponding Tafel slopes of electrodes in Figure 4a.

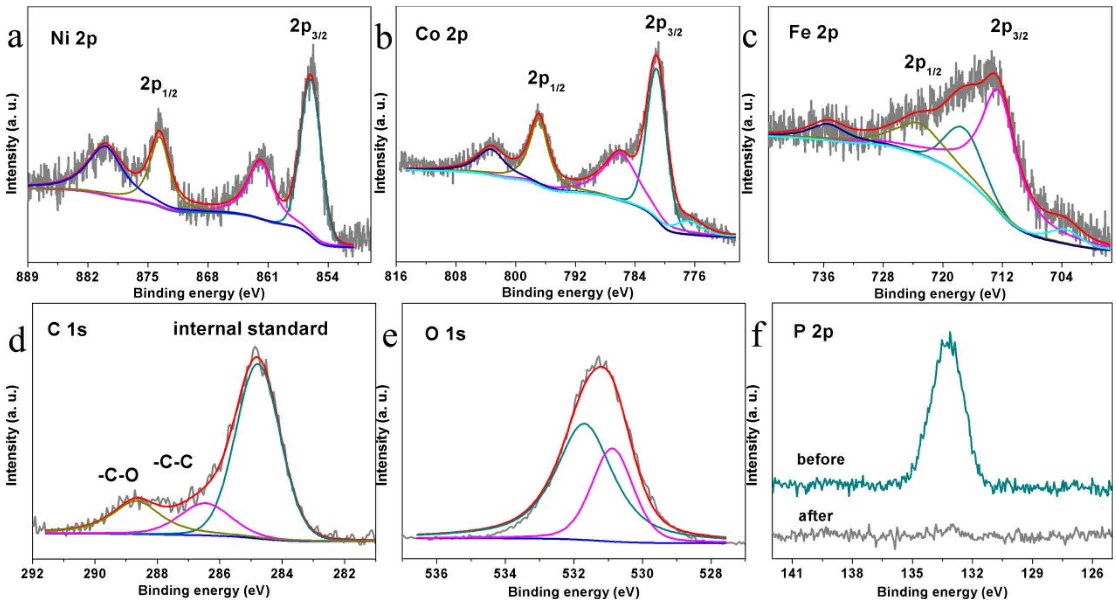

Fig. S13 (a-g) XPS images of C6FN-phy after HER stability measurement.

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_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_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.

Catalysts	Substrate	Tafel slope	Current density (J, mA cm^{-2})	η at corresponding J (mV)	Stability	Ref
$Ni_{0.9}Fe_{0.1}$/NC, 1 M KOH	Glass carbon	45	10	330	An increase of 25 mV after 10000 cycles at 10 mA cm-2	1
$CoNi(OH)_x$, 1 M KOH	Cu film	77	10	280	64% current density retention after 24 hours' electrocatalysis at η = 320 mV	2
NiCo-LDH, 1 M KOH	Carbon paper	40	10	367	An increase of 22 mV after 6 hours' electrocatalysis at 10 mA	3
$NiCo_2O_4$ nanowires, 1 M KOH	Ti foil	60	10	370	~ 93% current density retention after 20 hours' electrocatalysis at η = 420 mV	4
3D-NA/Ni/NiNPAs, 1 M KOH	Cu-coated polyimide film	96	10	285	94 % current density retention after 175 hours' electrocatalysis at η = 340 mV	5
Ni/Ni_3N, 1 M KOH	NF	60	10	~322	~ 96% current density retention after 12 hours' electrocatalysis at 100 mA cm^{-2}	6
Ni_2P, 1 M KOH	GC	59	10	290	An increase of 10 mV after 10 hours' electrocatalysis at 10 mA cm^{-2}	7
ECT-$Co_{0.37}Ni_{0.26}Fe_{0.37}O$, 1 M KOH	carbon fiber cloth	37.6	200	293	~ 95% current density retention after 100 hours' electrocatalysis at 20 mA cm^{-2}	8
NiFe/NF, 1 M KOH	NF	28	80 100	270 370	~ 100% current density retention after 10 hours' electrocatalysis at 100 mA cm^{-2}	9
NiFe films, 1 M NaOH	NF	-	100	300	~ 100% current density retention after 72 hours' electrocatalysis at 100 mA cm^{-2}	10
NiS 1 M KOH	NF	89	50	335	~ 100% current density retention after 35 hours' electrocatalysis at 13 mA cm^{-2}	11
NiSe 1 M KOH	NF	64	100	314	~ 99% current density retention after 12 hours' electrocatalysis at 100 mA cm^{-2}	12
FeOOH/Co/FeOOH HNTAs 1 M NaOH	NF	32	21 91 199	250 300 350	anodic polarzation tests at current densities of 20, 50, 100, and 200 mA cm^{-2} for 50 h, showing that the overpotentials remain unchange	13
NiP	NF	23	191	350	From 1.33V gradually increases to	14

					1.45 V vs. RHE after 0.2 h, and then remains fairly stable at this potential up to 26 h	
CN-phy	NF	86	150 200 250	316 332 349	98% current density retention after 10 hours' electrocatalysis at 100 mA cm^{-2}	This work
C6FN-phy	NF	24	150 200 250 300	281 285 288 291	~ 96% and 100%current density retention after 70 and 32 hours' electrocatalysis at 200 and 100mA cm^{-2}	This work
FN-phy	NF	75	150 200 250	319 332 344	~ 99% current density retention after 10 hours' electrocatalysis at 100 mA cm^{-2}	This work
N-phy	NF	60	150 200	343 354	~ 99% current density retention after 10 hours' electrocatalysis at 100 mA cm^{-2}	This work

Table S2. Comparisons of the two-electrode configuration performance according to the reports and this paper in 1 M KOH.

Catalyst	Substrate	η@10 mA cm^{-2} (mV)	Current density (J, mA cm^{-2})	η at correspondin g J (mV)	Reference
$FeNi_3N$/NF	NF	390	～90	770	15
Ni–P foam	NF	410	100	820	14
NiSe/NF	NF	400	～60	770	12
$NiCo_2S_4$ NW	NF	400	～70	770	16
$Ni_{2.3\%}$-CoS_2/CC	Carbon cloth	430	60	770	17
NiCoP/rGO	-	360	60	670	18
C6FN-phy	NF	460	100	660	This work

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