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Electronic Supplementary Information

Hierachical Nickel-Carbon Structure Templated by Metal-Organic Frameworks for Efficient Overall Water Splitting

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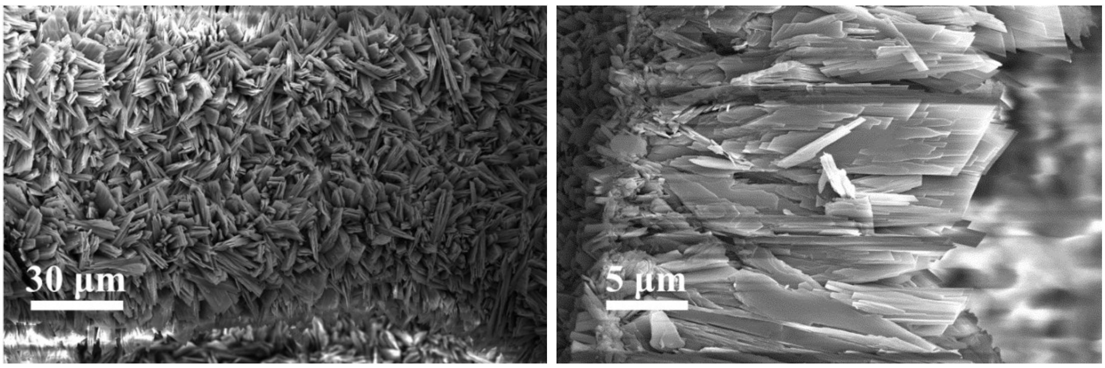


Fig. S1 SEM images of NF@Ni(BDC). A full coverage of Ni(BDC) with an average thickness about 20 μm was obtained for all NF@Ni(BDC) samples.

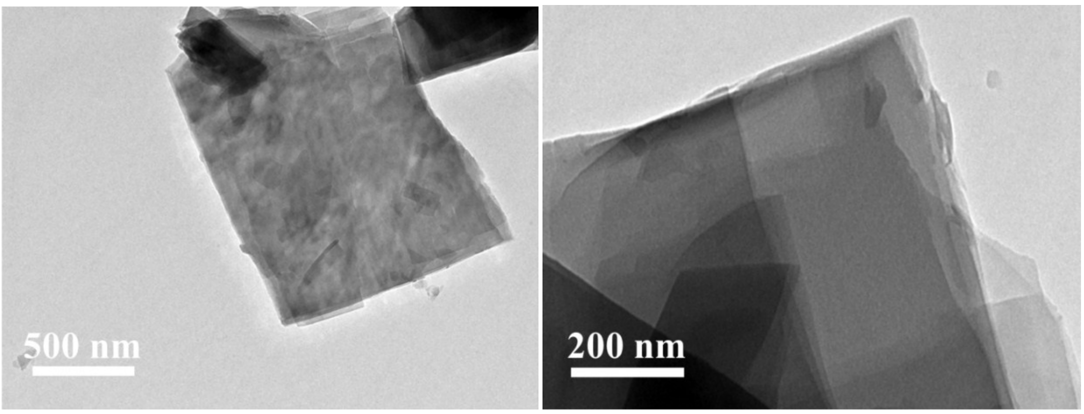


Fig. S2 TEM images of Ni(BDC).

Table S1. The weight of the carbonized product can be estimated by measuring the difference in mass after stripping it off from the nickel foam through extensive high-power sonication. The area of NF@Ni/C-600 is 2.3 cm^{-2}, so the average mass density is 7.3 ± 0.3 mg cm^{-2}.

Weight (NF@Ni/C, mg)	Weight (NF, mg)	Loss weight (mg)	Weight density (mg/cm^{-2})
62.3	**46.5**	**15.8**	**6.87**
63.1	**46.7**	**16.4**	**7.13**
63.9	**46.6**	**17.3**	**7.52**
66.6	**48.8**	**17.8**	**7.74**
63.6	**46.3**	**17.3**	**7.50**
63.5	**47.1**	**16.4**	**7.14**

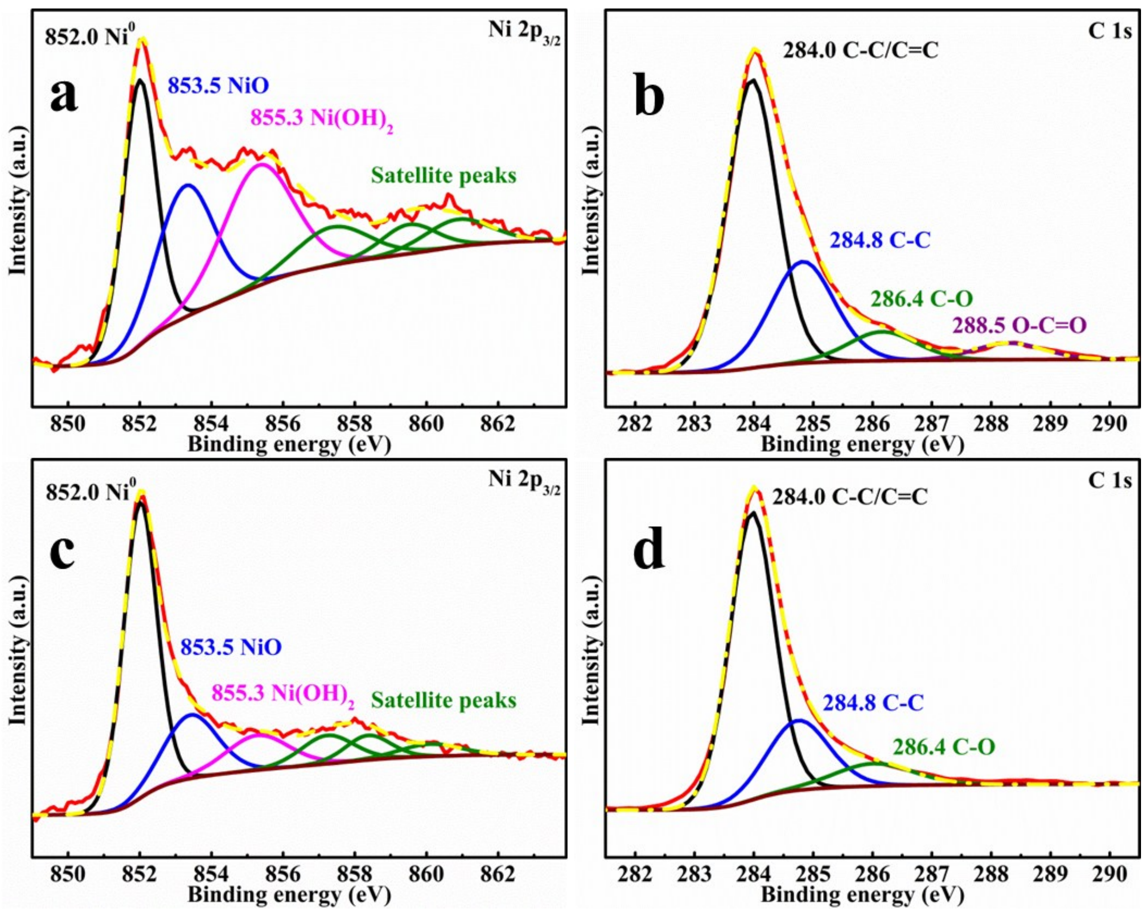


Fig. S3 (a) Ni $2p_{3/2}$ and (b) C 1s XPS spectra of Ni/C-500. (c) Ni $2p_{3/2}$ and C 1s (d) XPS spectra of Ni/C-700.

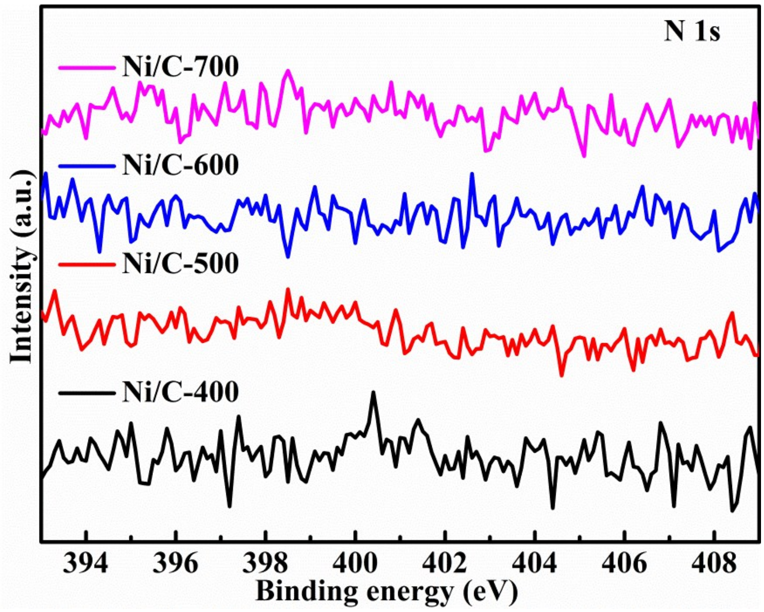
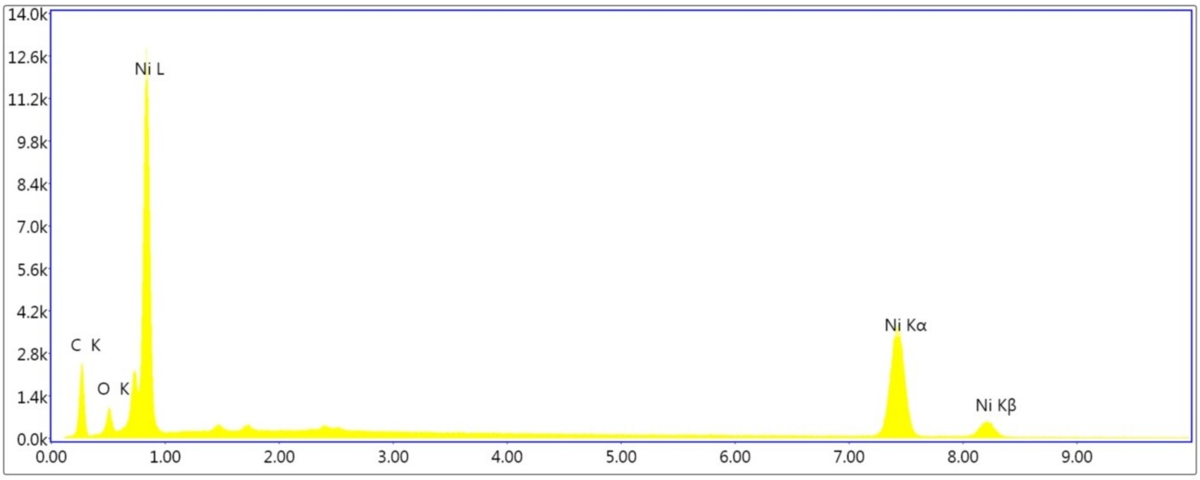


Fig. S4 XPS N 1s spectra of Ni/C-400, Ni/C-500, Ni/C-600 and Ni/C-700



Element	Weight%	Atom %	Net strength	Error
C K	23.71	57.07	39.7	0.01
O K	4.08	7.37	19.2	0.01
NiK	72.22	35.57	163.9	0

Fig. S5 EDX spectrum of the Ni/C-600.

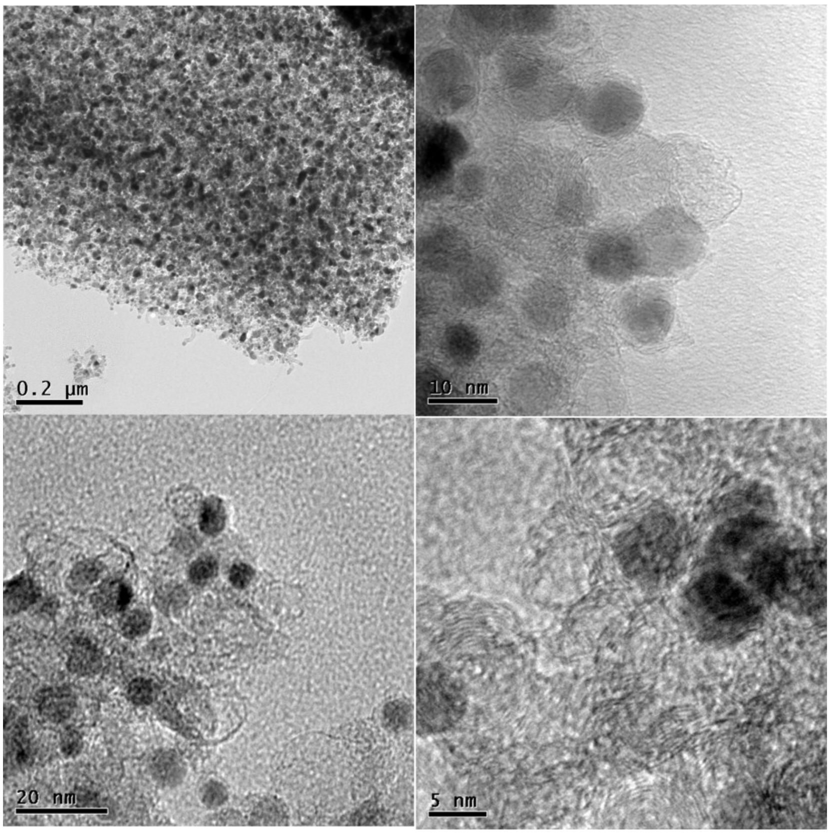


Fig. S6 TEM images of Ni/C-600.

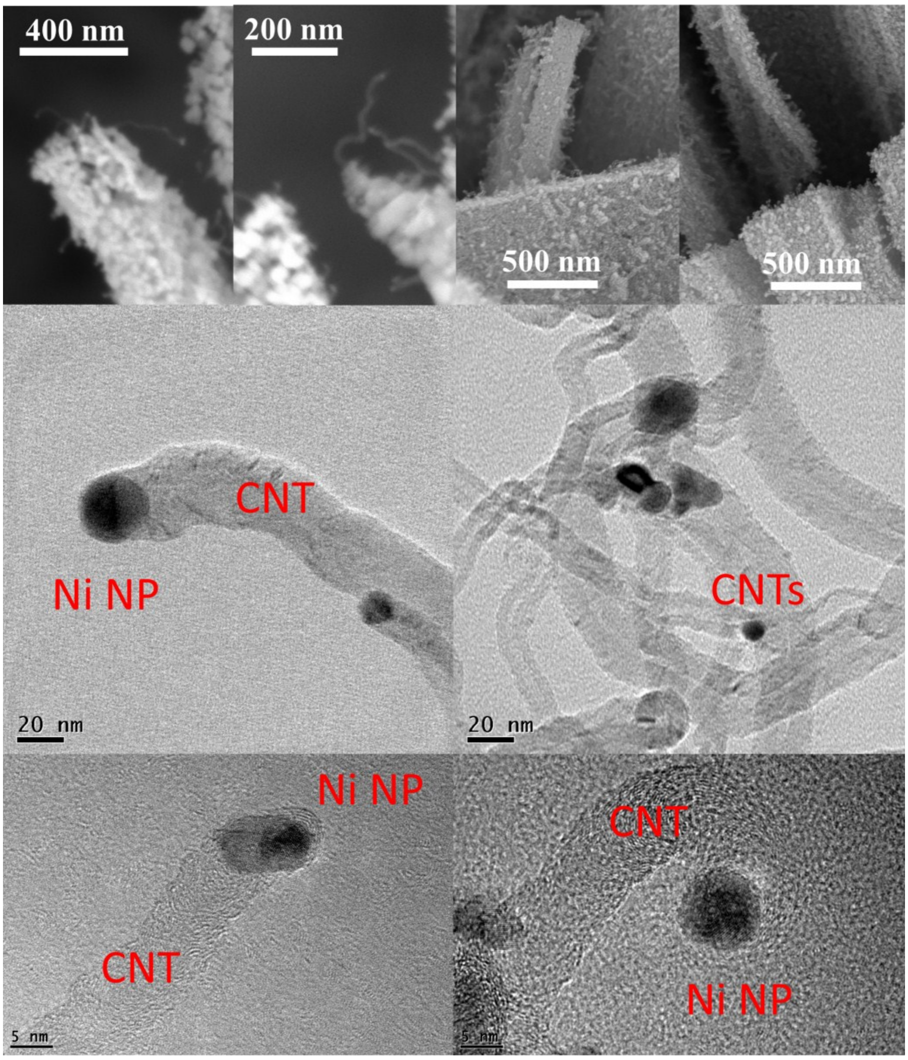


Fig. S7 SEM and TEM images of CNTs in Ni/C samples.

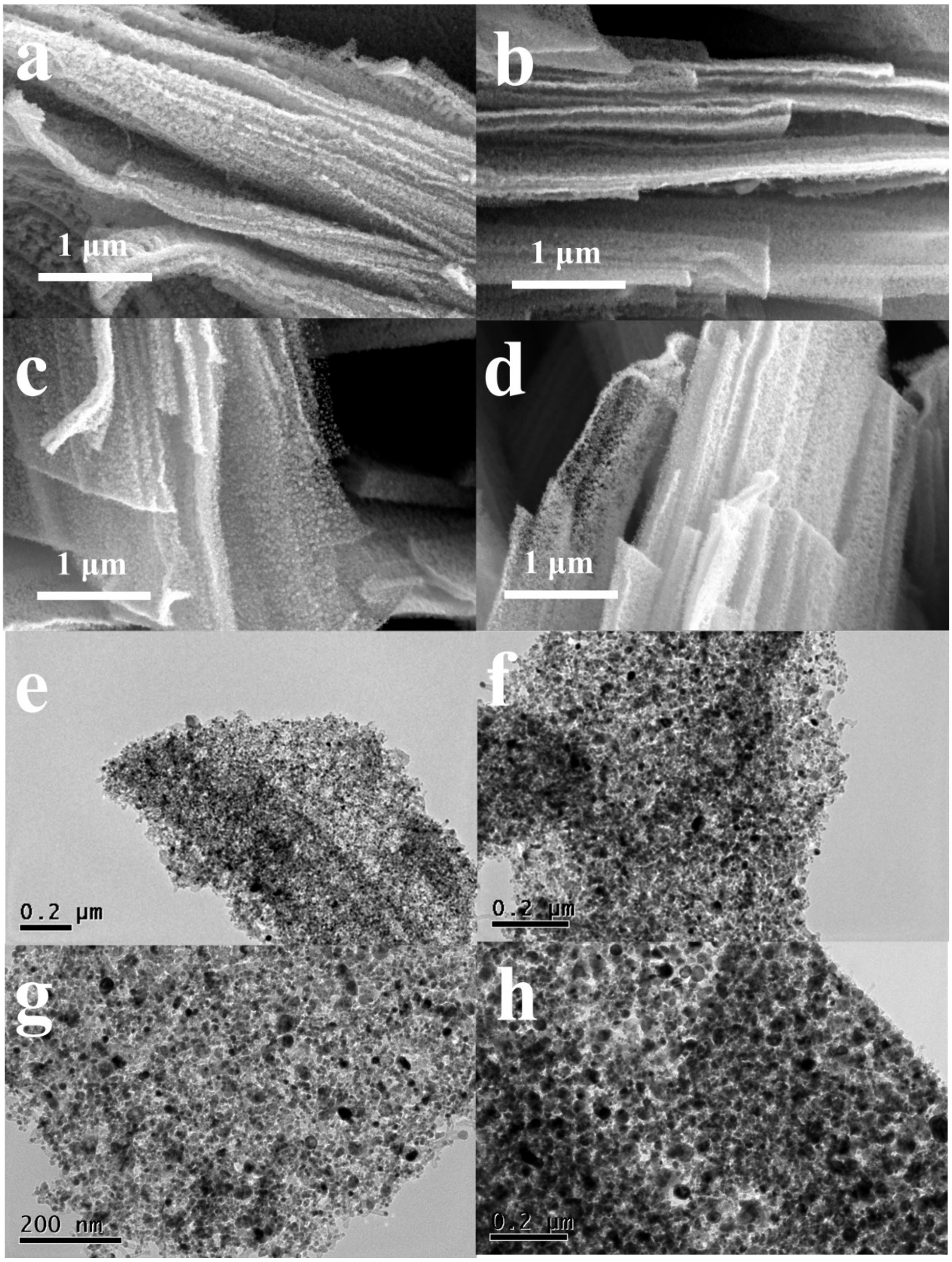


Fig. S8 (a) SEM and (e) TEM images of Ni/C-400. (b) SEM and (f) TEM images of Ni/C-500. (c) SEM and (g) TEM images of Ni/C-600. (d) SEM and (h) TEM images of Ni/C-700.

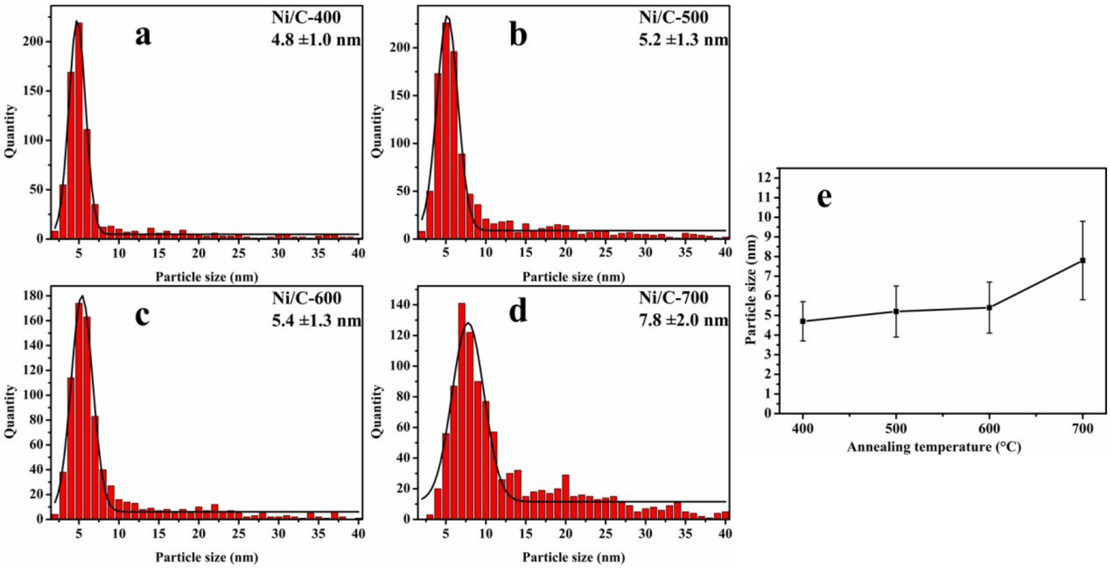


Fig. S9 Ni particle size distribution of (a) Ni/C-400, (b) Ni/C-500, (c) Ni/C-600 and (d) Ni/C-700. (e) Ni particle size as a function of the annealing temperature.

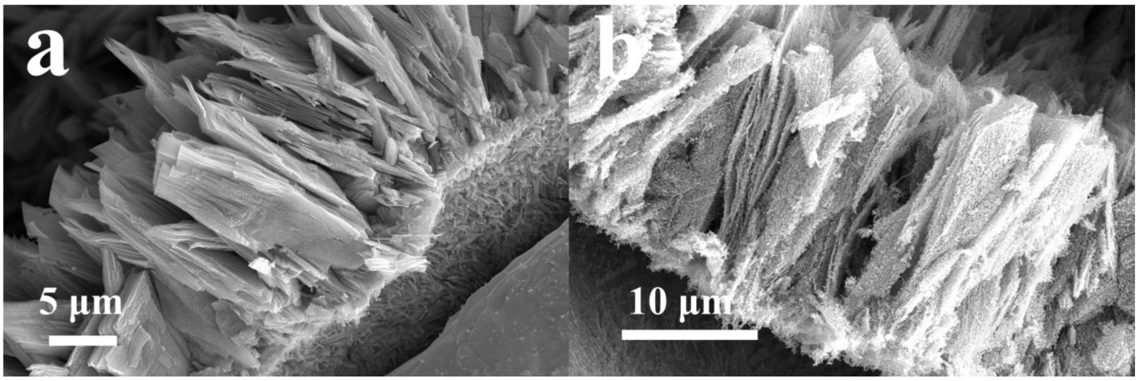


Fig. S10 Cross-sectional SEM images of (a) NF@Ni/C-600 and (b) NF@Ni/C-700.

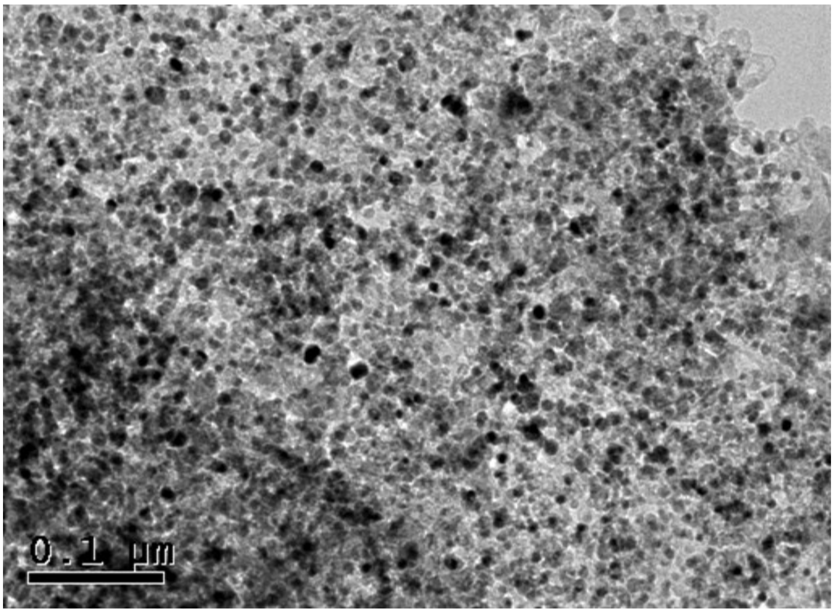


Fig. S11 TEM images of the Ni/C-600 sample after 24 hours of excessive soaking in dilute hydrochloric acid.

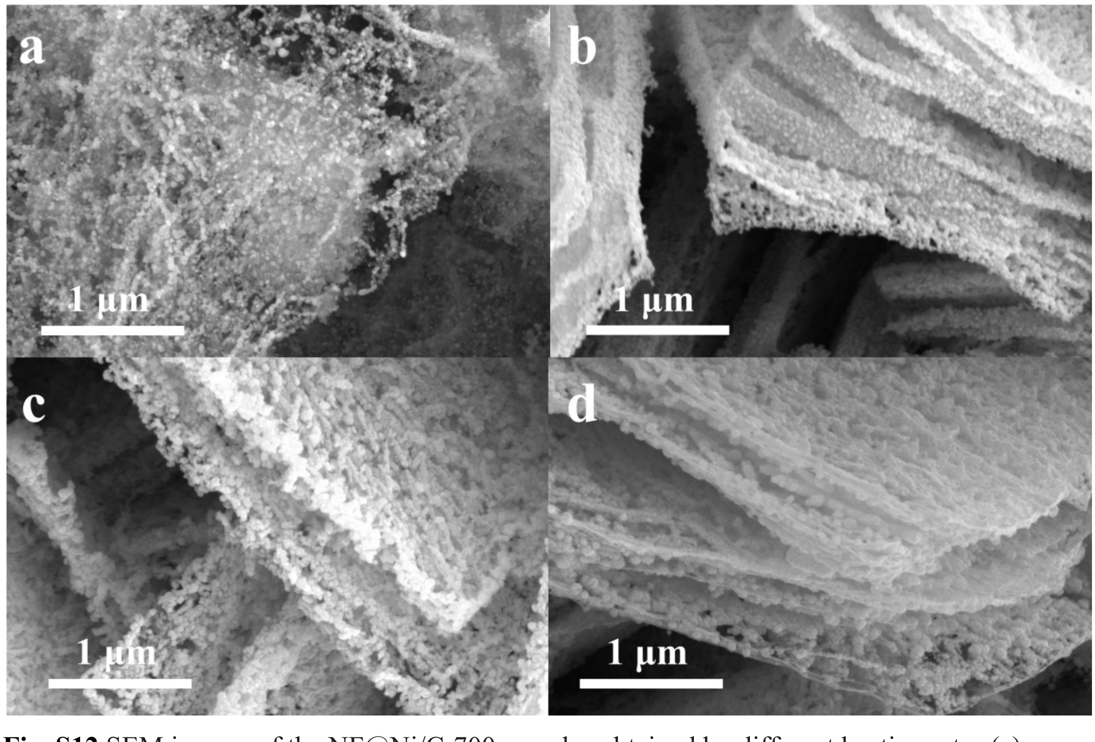


Fig. S12 SEM images of the NF@Ni/C-700 samples obtained by different heating rates (a) 12.8, (b) 6.4, (c) 3.2, (d) 1.6 °C min^{-1}.

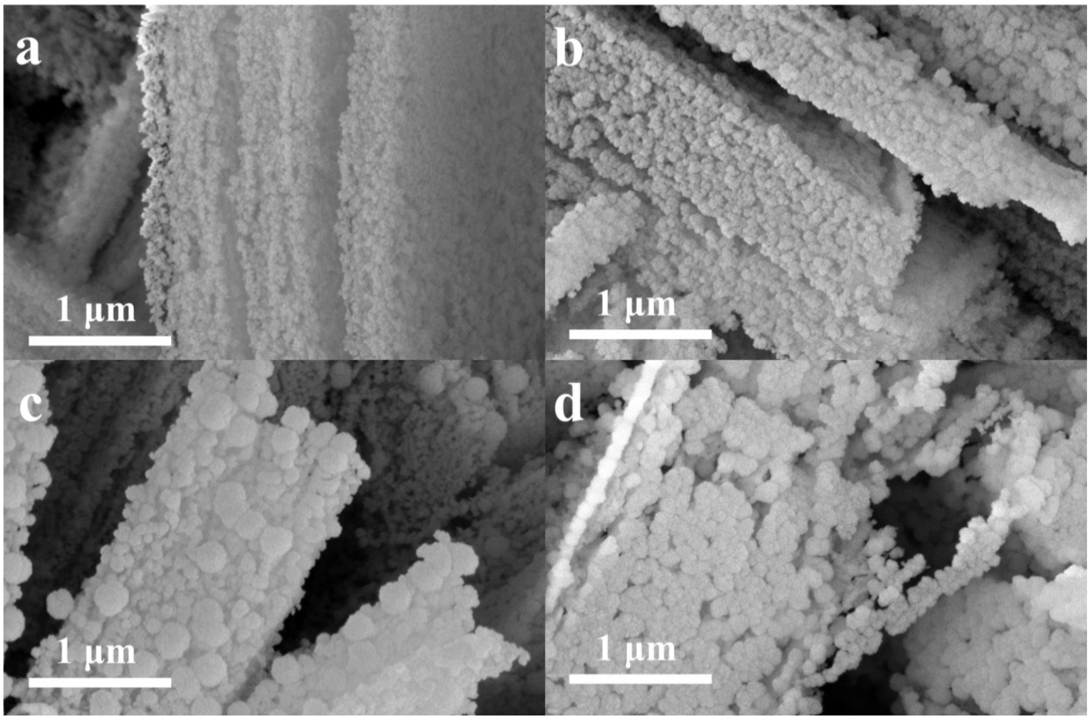


Fig. S13 SEM images of the NF@Ni/C-700 samples obtained from different annealing holding times (a) 1, (b) 2, (c) 4, (d) 8h.

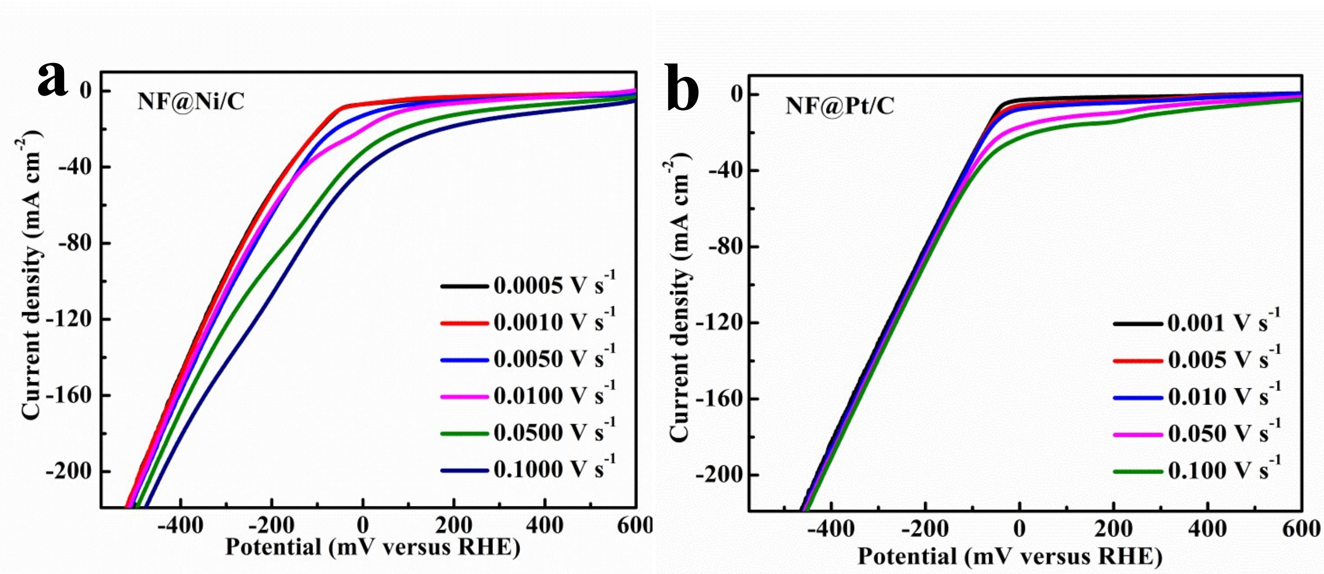


Fig. S14 (a) The HER polarization curves of NF@Ni/C at different scanning speeds (0.5-100 mV s^{-1}). (b) The HER polarization curves of NF@Pt/C at different scanning speeds (1-100 mV s^{-1}). These two figures show that low scanning speed can effectively reduce the background current and we obtain the same curve which has the lowest background current at two different sweep speeds, 0.5 and 1 mV s^{-1}, so we chose the sweep speed 1 mV s^{-1}.

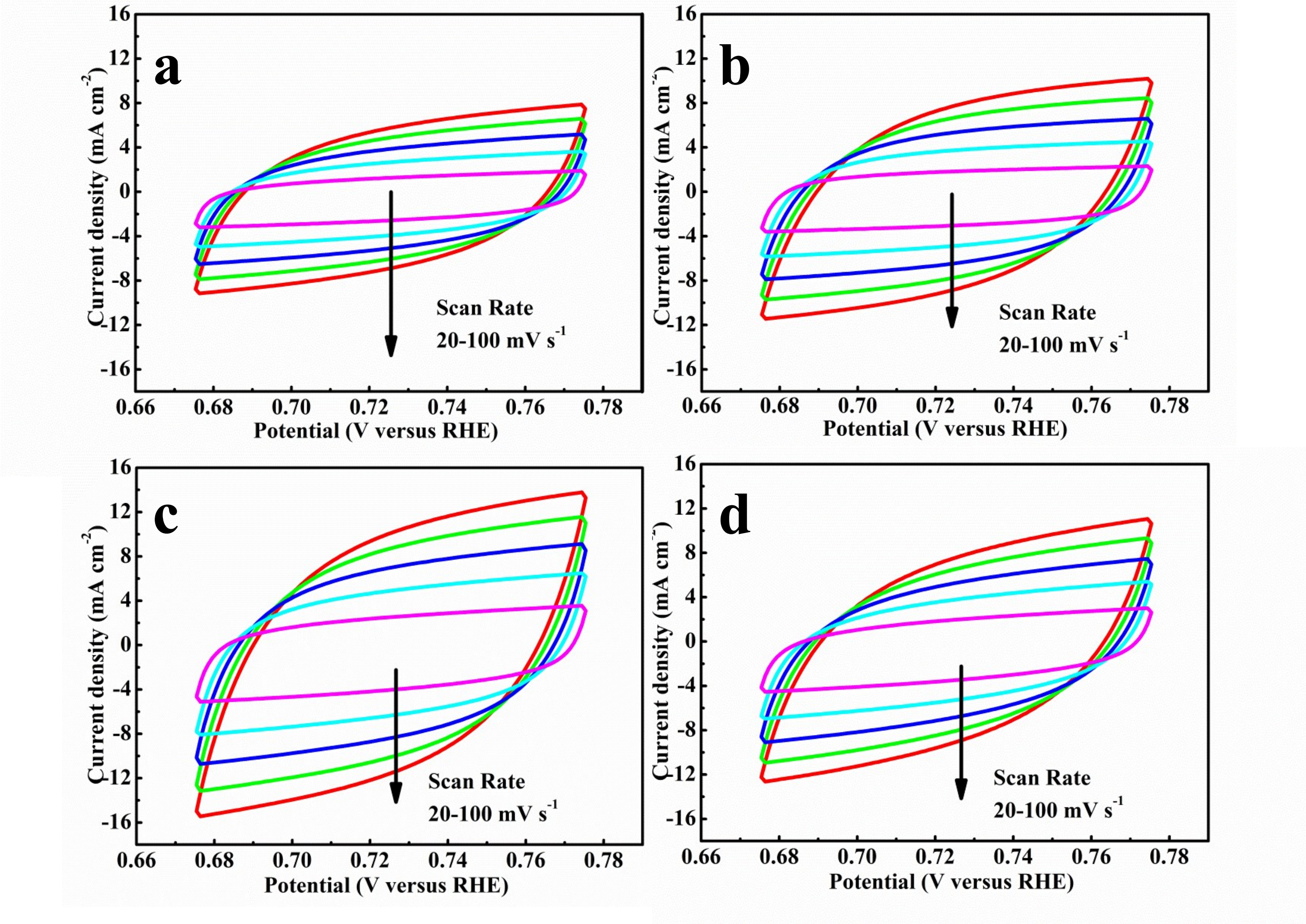


Fig. S15 Electrochemical cyclic voltammetry curves of a) NF@Ni/C-400, b) NF@Ni/C-500, c) NF@Ni/C-600, and d) NF@Ni/C-700 at different potential scanning rates.

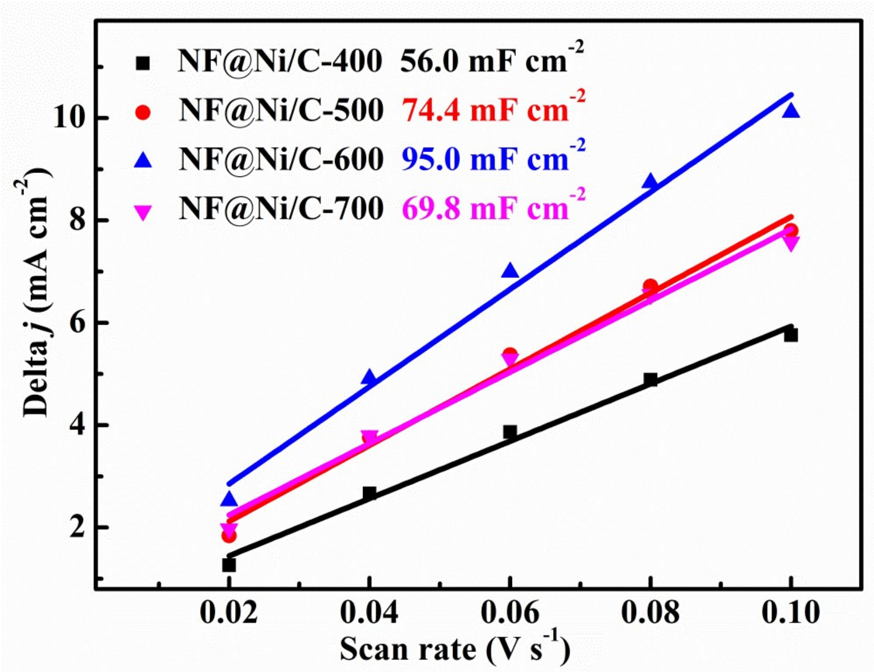


Fig. S16 Plots of the capacitive currents as a function of scan rate of various NF@Ni/C samples.

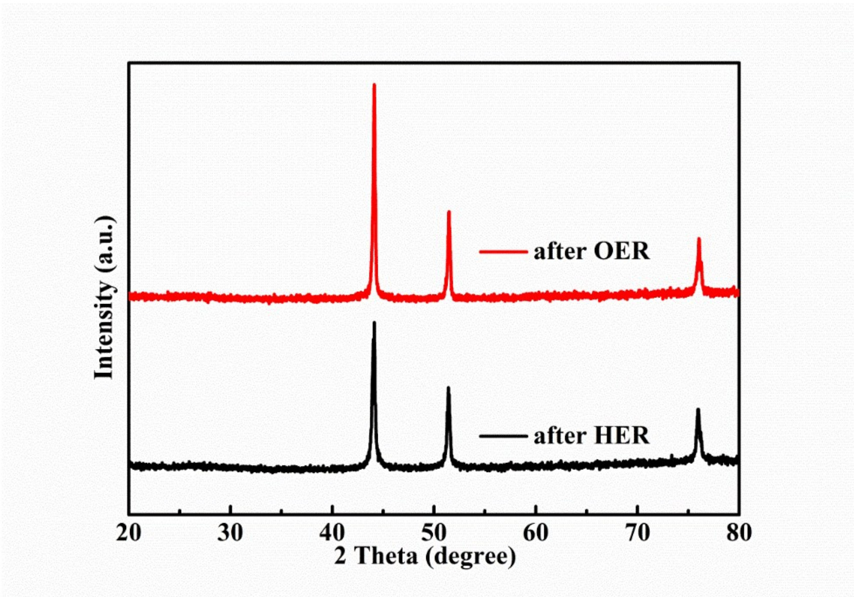


Fig. S17 XRD patterns of the NF@Ni/C-600 after continuous water-splitting reaction in alkaline electrolyte solution.

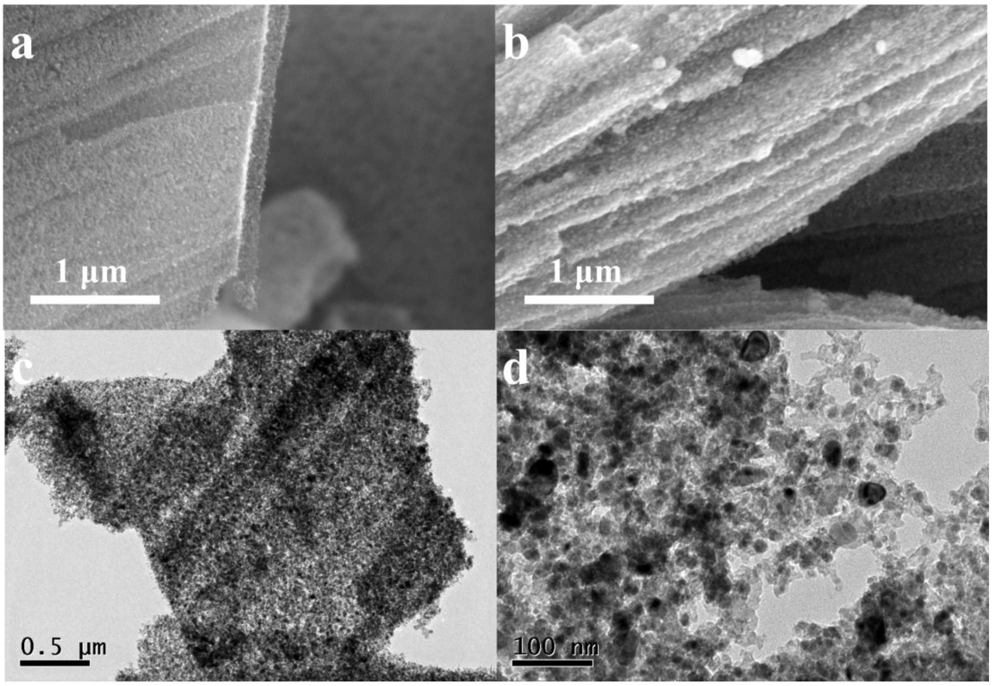


Fig. S18 SEM and TEM images of the Ni/C-600 after continuous water-splitting reaction in alkaline electrolyte solution: (a) and (c) post-HER, (b) and (d) post-OER.

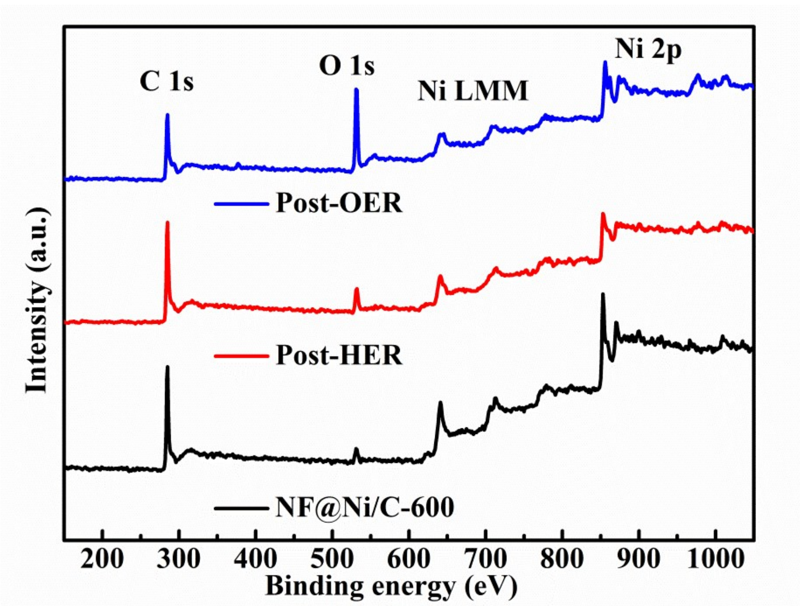


Fig. S19 XPS survey spectra of the NF@Ni/C-600, post-HER NF@Ni/C-600 and post-OER NF@Ni/C-600.

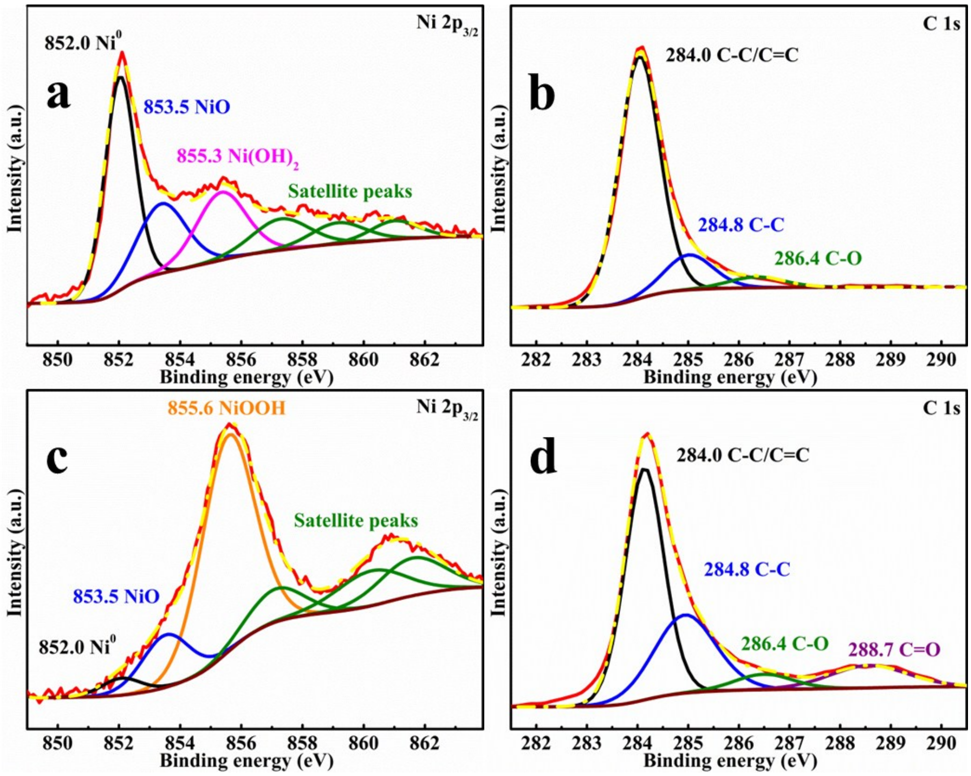


Fig. S20 (a) Ni $2p_{3/2}$ and (b) C 1s XPS spectra of post-HER Ni/C-600. (c) Ni $2p_{3/2}$ and (d) C 1s XPS spectra of post-OER Ni/C-600.

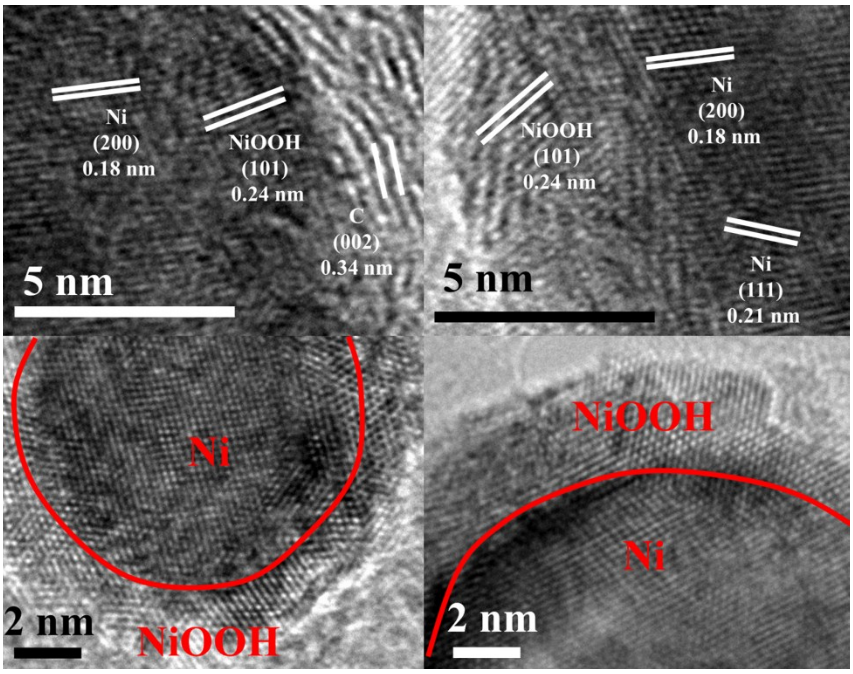


Fig. S21, HR-TEM images of post-OER Ni/C-600 showing explicit lattice fringes of Ni particles and the oxidized surface layer containing NiOOH.

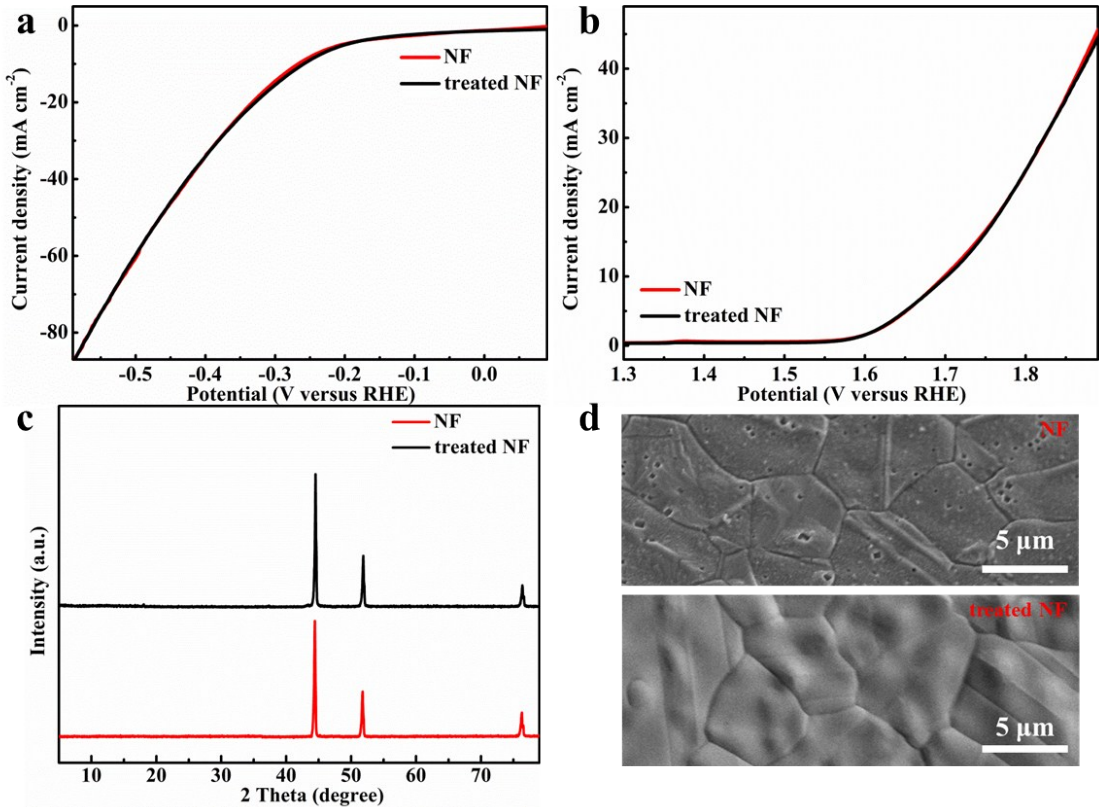


Fig. S22, Comparison of NFs with and without going through the hydrothermal and annealing process for the control study. (a) HER and (b) OER polarization curves of untreated and treated NFs. (c) XRD patterns and (d) SEM images of untreated and treated NFs.

Table S2. Summary of high-performance bifunctional catalytic electrodes for overall water splitting.

Catalytic Materials	Electrolyte	Activity	Reported Durability	Reference
Ni/C-600/NF	1 M KOH	35 mA cm^{-2} @ 1.6 V	70 h	This work
NiFe-MOF/NF	0.1 M KOH	~12.5 mA cm^{-2} @ 1.5 V	20 h	[1]
NC@$CuCo_2N_x$/CF	1 M KOH	10 mA cm^{-2} @ 1.62 V	60 h	[2]
NiS/Ni_2P/CC	1 M KOH	10 mA cm^{-2} @ ~1.67 V	10 h	[3]
Ni_2P/rGO/NF	1 M KOH	10 mA cm^{-2} @ 1.62 V	50 h	[4]
Ni_3S_2/NF	pH=14	~13 mA cm^{-2} @ ~1.76 V	150 h	[5]
Co_9S_8/WS_2	1 M KOH	<10 mA cm^{-2} @ 1.65 V	24 h	[6]
NC-800/NF	1 M KOH	18 mA cm^{-2} @ 1.62 V	50 h	[7]
Co_4Ni_1P /NF	1 M KOH	10 mA cm^{-2} @ 1.59 V	50 h	[8]
porous MoO_2/NF	1 M KOH	10 mA cm^{-2} @ 1.53 V	24 h	[9]
SNCF-NR	1 M KOH	10 mA cm^{-2} @ 1.68 V	30 h	[10]
NESSP//NESS	1 M KOH	~11 mA cm^{-2} @ 1.74 V	134 h	[11]
Cu@NiFe LDH	1 M KOH	10 mA cm^{-2} @ 1.54 V	48 h	[12]
Co_1Mn_1CH/NF	1 M KOH	10 mA cm^{-2} @ 1.68 V	15 h	[13]
Ni_xCo_{3-x}S4/Ni_3S_2/NF	1 M KOH	10 mA cm^{-2} @ 1.53 V	200 h	[14]
NiCoP/rGO	1 M KOH	10 mA cm^{-2} @ 1.59 V	80 h	[15]
MoS_2/Ni_3S_2	1 M KOH	10 mA cm^{-2} @ 1.56 V	10 h	[16]
2.5H-PHNCMs	1 M KOH	10 mA cm^{-2} @ 1.49 V	24 h	[17]
Ni-Co-P HNBs	1 M KOH	10 mA cm^{-2} @ 1.62 V	20 h	[18]
Ni_2Fe_1-O	1 M KOH	10 mA cm^{-2} @ 1.64 V	10 h	[19]
CoP@NPMG	1 M KOH	10 mA cm^{-2} @ 1.58 V	30 h	[20]
Co_3S_4@MoS_2	1 M KOH	10 mA cm^{-2} @ 1.58 V	10 h	[21]

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