Supporting information

High-Density Nickel–Cobalt Alloy Embedded in Nitrogen-Doped Carbon Nanosheets for Hydrogen Evolution Reaction

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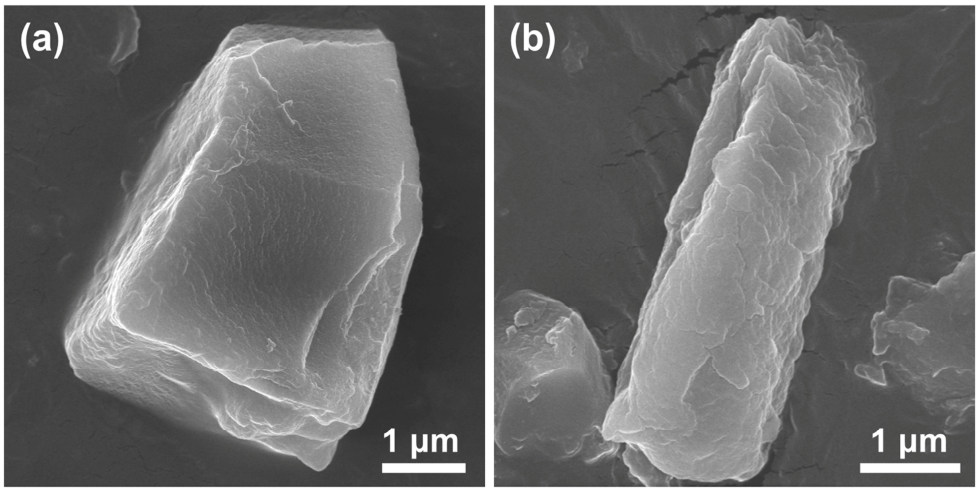


Figure S1. SEM images of the Ni_7Co_3–CS precursor.

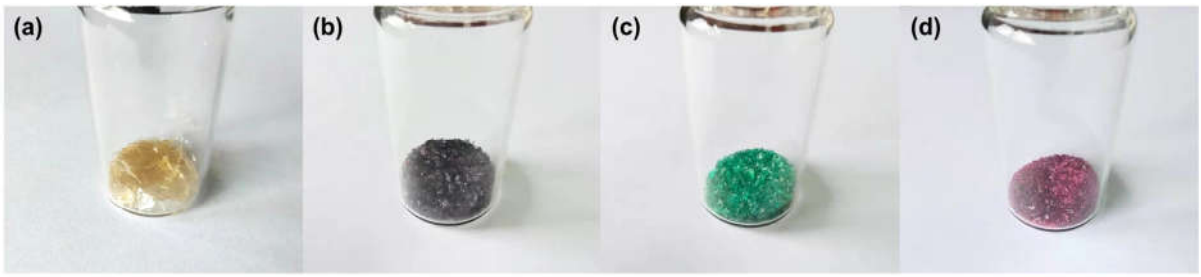


Figure S2. Photographs of as-prepared **(a)** chitosan, **(b)** Ni_7Co_3–CS, **(c)** Ni–CS, and **(d)** Co–CS precursors.

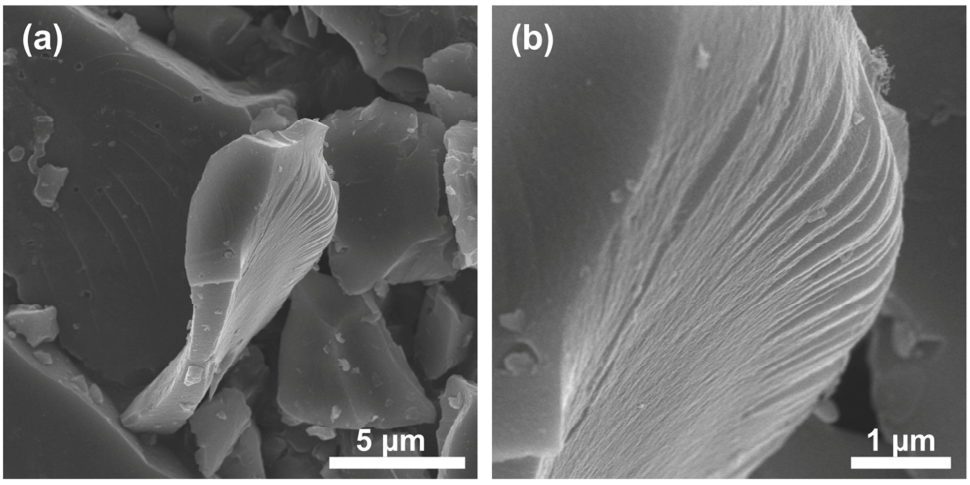


Figure S3. SEM images of NC.

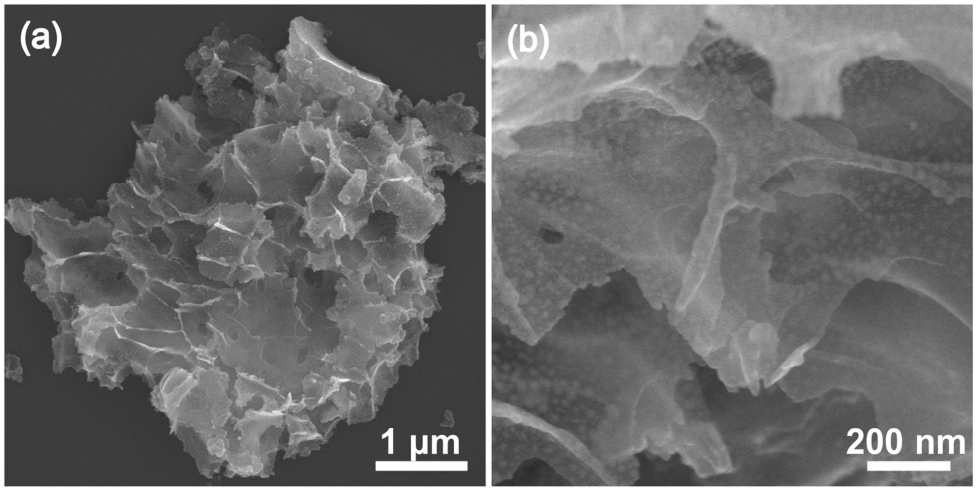


Figure S4. SEM images of Ni/NC.

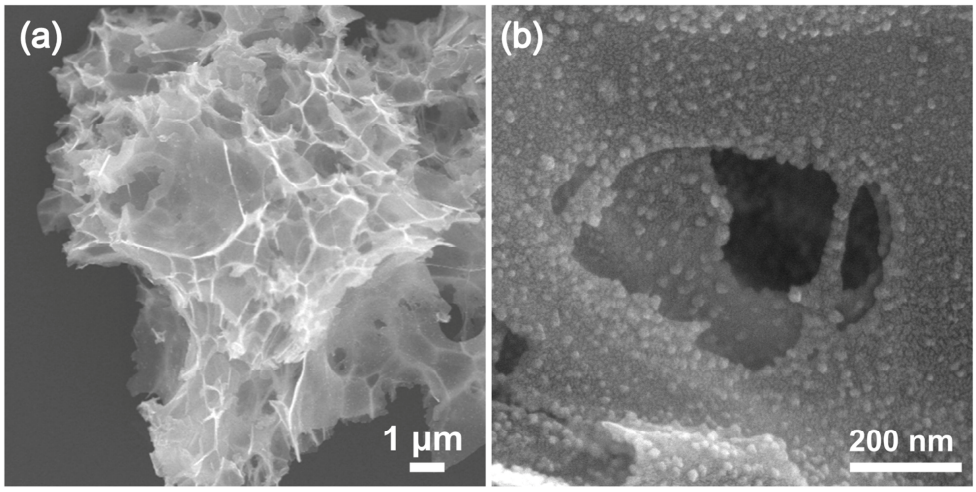


Figure S5. SEM images of Co/NC.

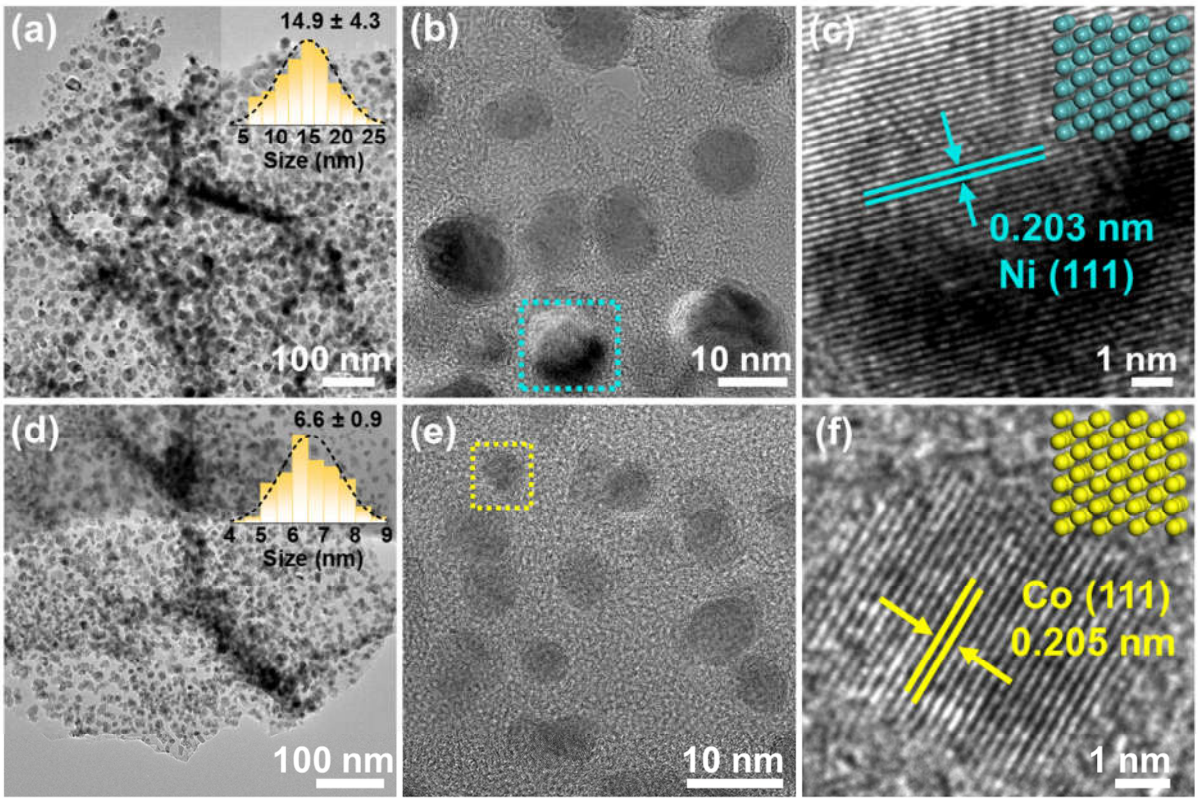


Figure S6. (a) TEM image (inset shows the particle size distribution), and **(b-c)** HR-TEM images of Ni/NC; **(d)** TEM image (inset shows the particle size distribution), and **(e-f)** HR-TEM images of Co/NC.

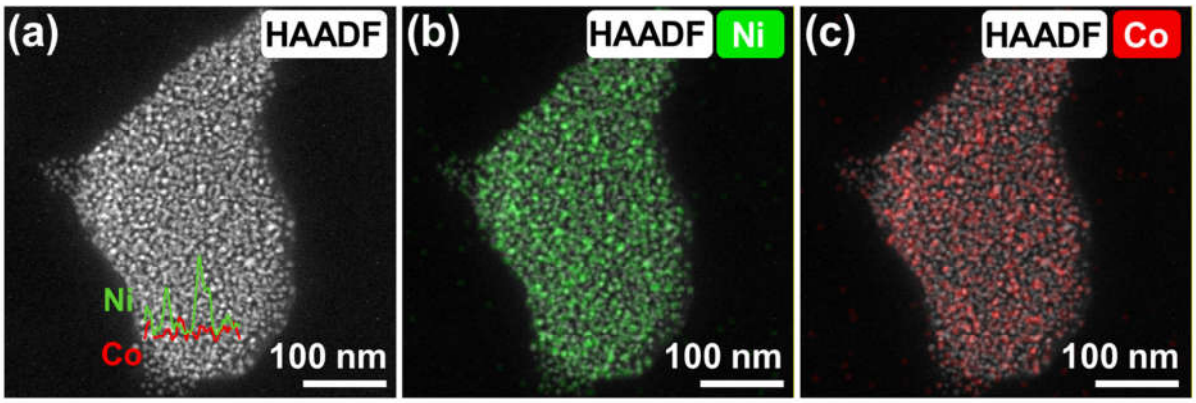


Figure S7. High annular dark-field scanning TEM (HAADF-STEM) and corresponding elemental mapping images of Ni_7Co_3/NC-500.

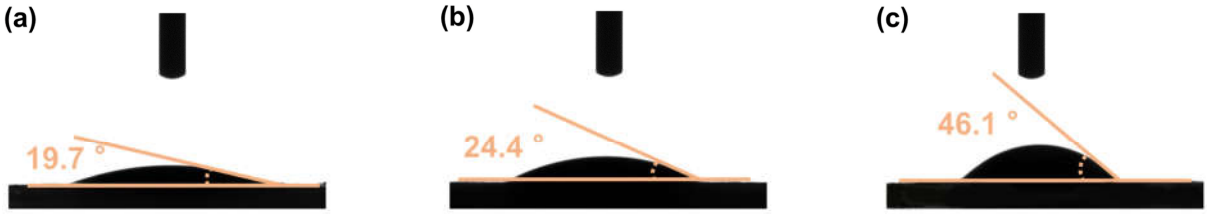


Figure S8. Water contact angle of **(a)** Ni_7Co_3/NC-500, **(b)** Ni/NC, and **(c)** Co/NC.

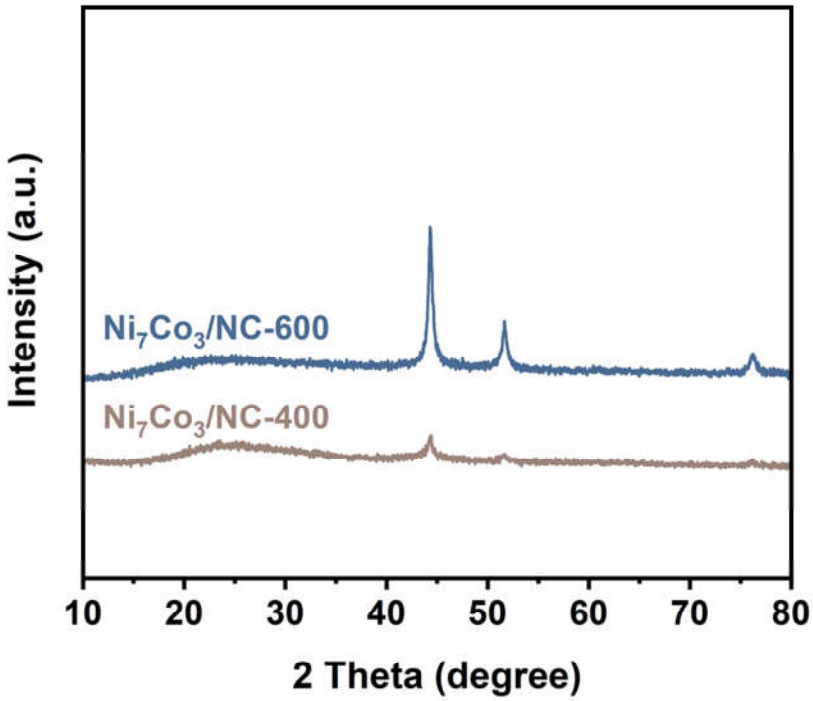


Figure S9. XRD patterns of Ni_7Co_3/NC-600 and Ni_7Co_3/NC-400.

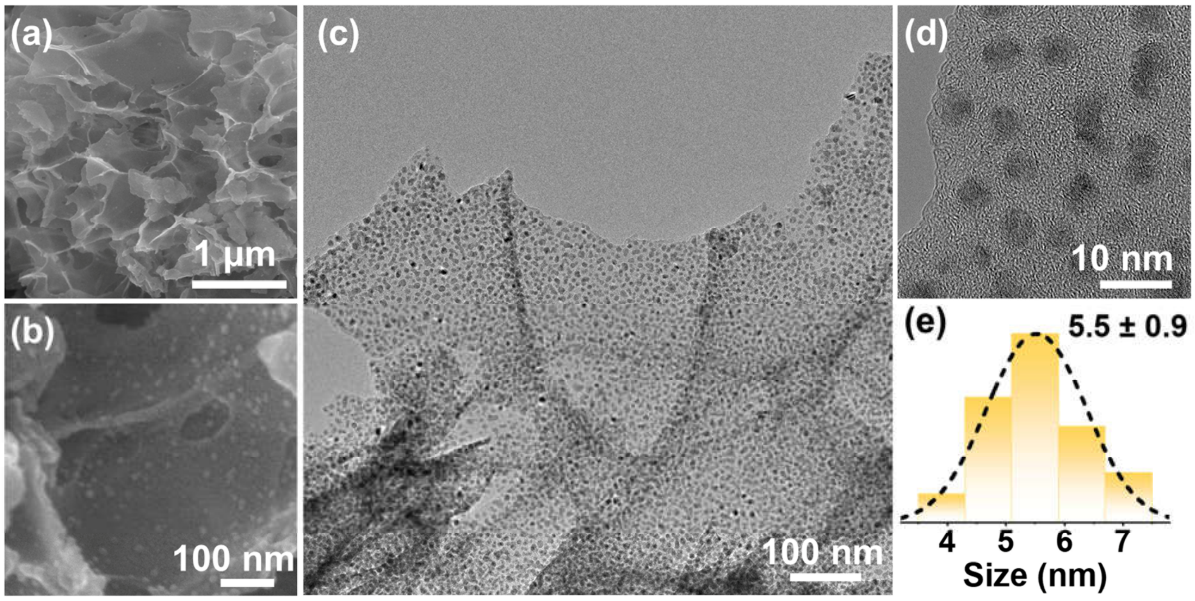


Figure S10. (a-b) SEM images, **(c)** TEM image, and **(d)** HR-TEM image of Ni_7Co_3/NC-400. **(e)** The particle size distribution in c.

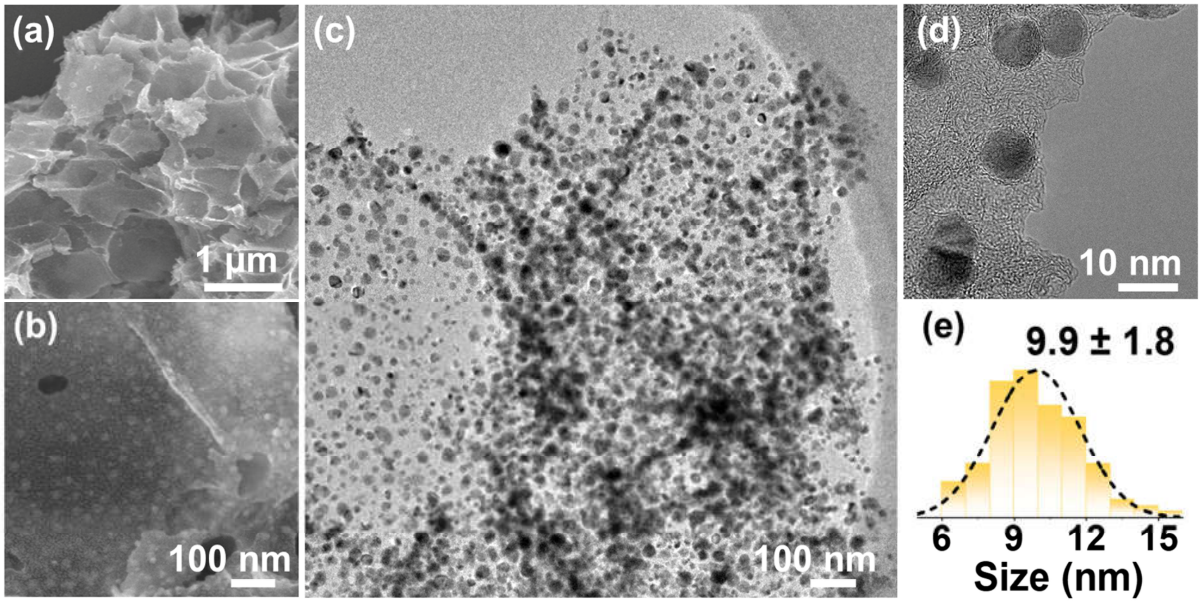


Figure S11. (a-b) SEM images, **(c)** TEM image, and **(d)** HR-TEM image of Ni_7Co_3/NC-600. **(e)** The particle size distribution in c.

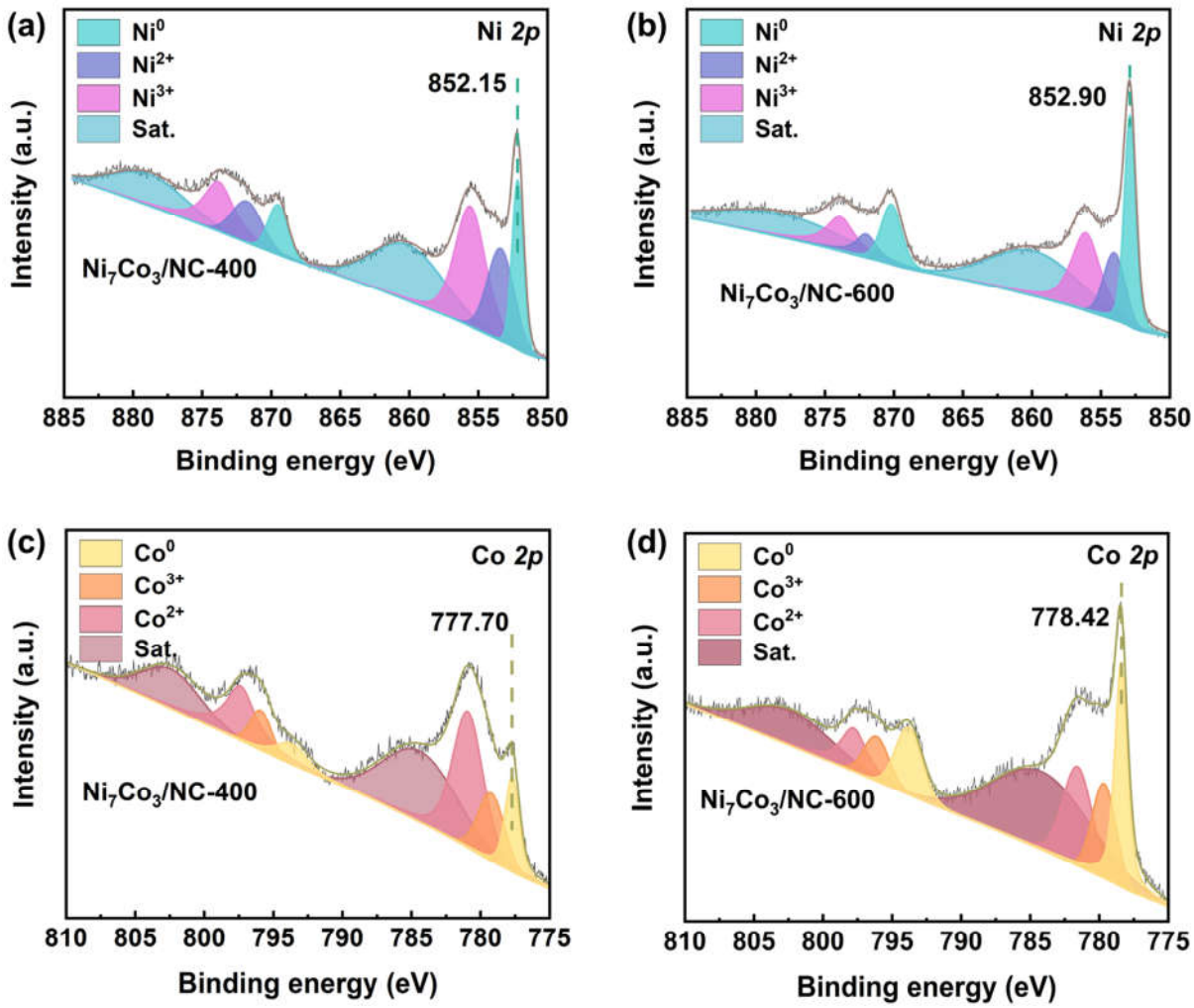


Figure S12. High resolution XPS spectra of Ni 2*p* of **(a)** Ni_7Co_3/NC-400 and **(b)** Ni_7Co_3/NC-600. High resolution XPS spectra of Co 2*p* of **(c)** Ni_7Co_3/NC-400 and **(d)** Ni_7Co_3/NC-600.

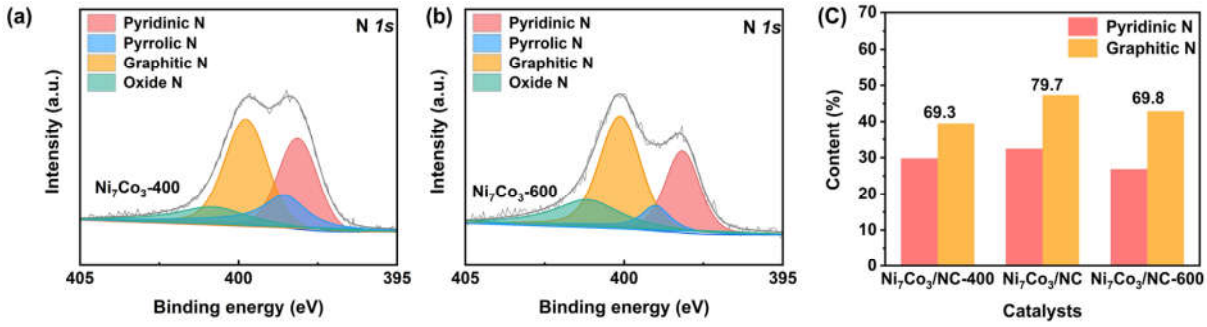


Figure S13. High resolution XPS spectra of N 1*s* of **(a)** Ni_7Co_3/NC-400 and **(b)** Ni_7Co_3/NC-600. **(c)** Surface pyridinic N and graphitic N percentages of Ni_7Co_3/NC-T determined by N 1*s* of XPS profile.

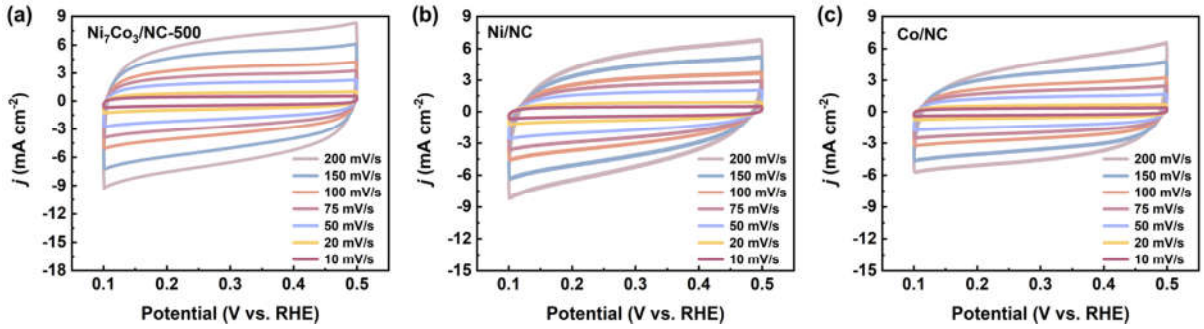


Figure S14. Cyclic voltammograms of **(a)** Ni_7Co_3/NC-500, **(b)** Ni/NC, and **(c)** Co/NC with various scan rates in 1 M KOH solution.

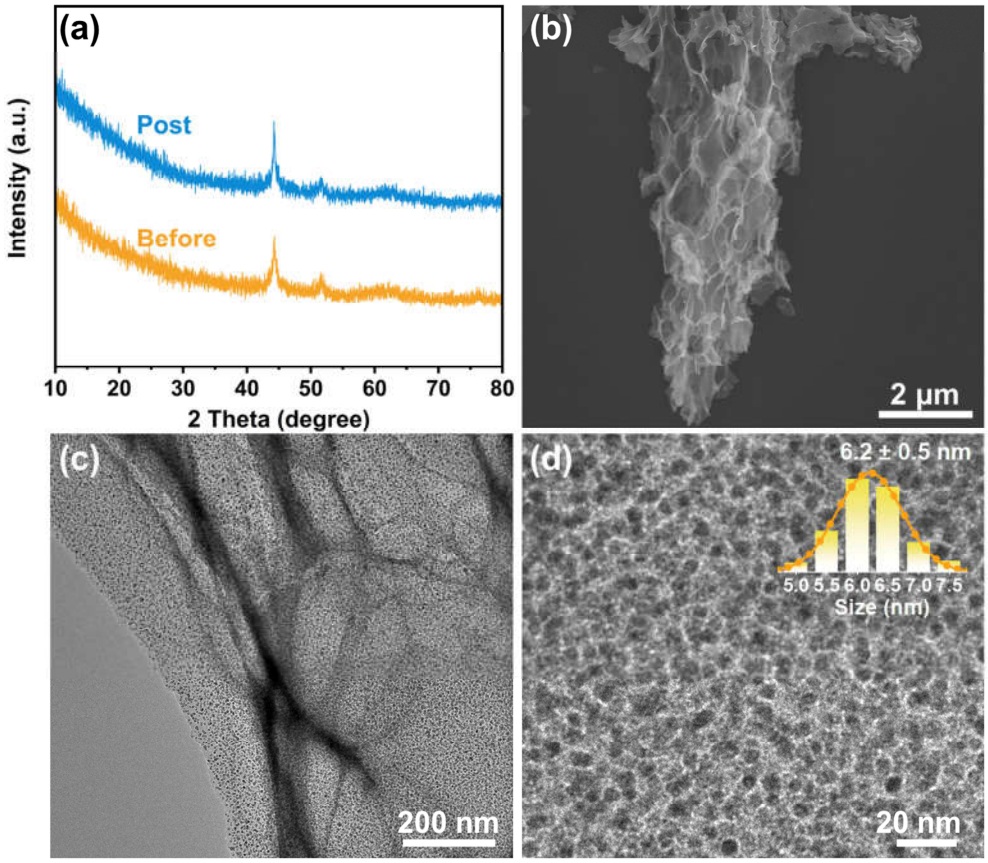


Figure S15. (a) XRD patterns of Ni_7Co_3/NC-500 before and after reaction 12 h; **(b)** SEM image and **(c-d)** TEM images of Ni_7Co_3/NC-500 after 12 h stability test.

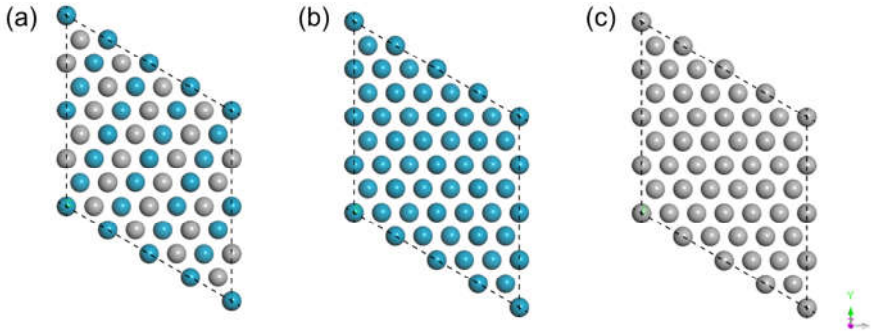
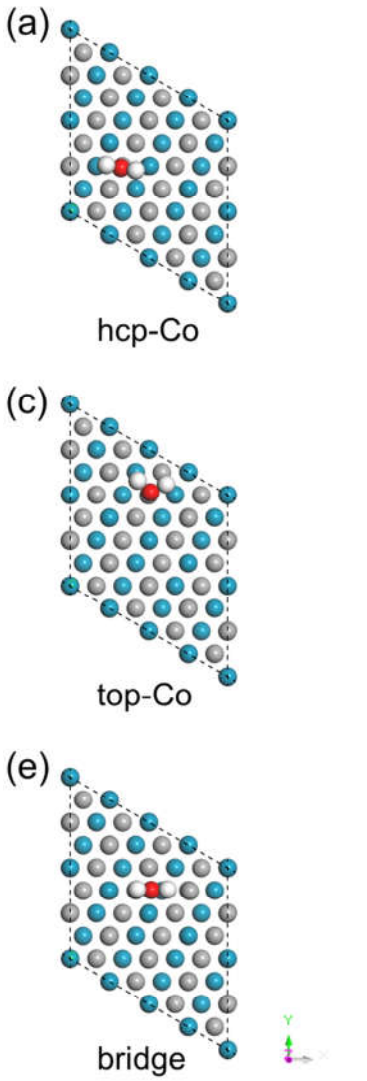


Figure S16. The structure of **(a)** NiCo (111), **(b)** Ni (111), and **(c)** Co (111) (Ni: blue, Co: grey).



H_2O adsorption site	*E* (eV)
hcp-Co	0.30
hcp-Ni	0.23
top-Co	-0.26
top-Ni	-0.15
bridge	0.31

Figure S17. Determination of the possible adsorption site of water on NiCo(111) surface. The corresponding water adsorption energy are listed in the table on right (Ni: blue, Co: grey, O: red and H: white).

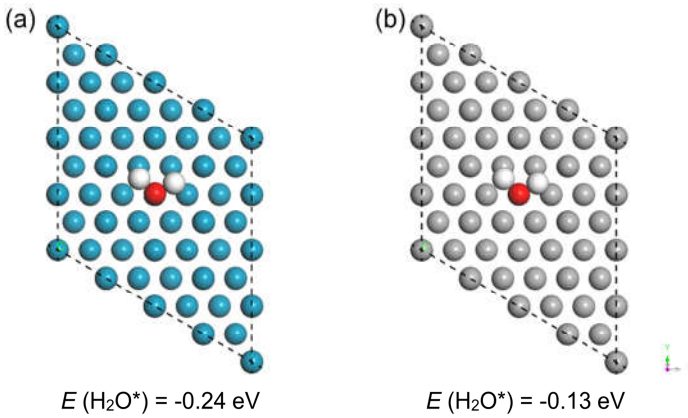


Figure S18. The water adsorption energy on **(a)** Ni (111) and **(b)** Co (111) (Ni: blue, Co: grey, O: red and H: white).

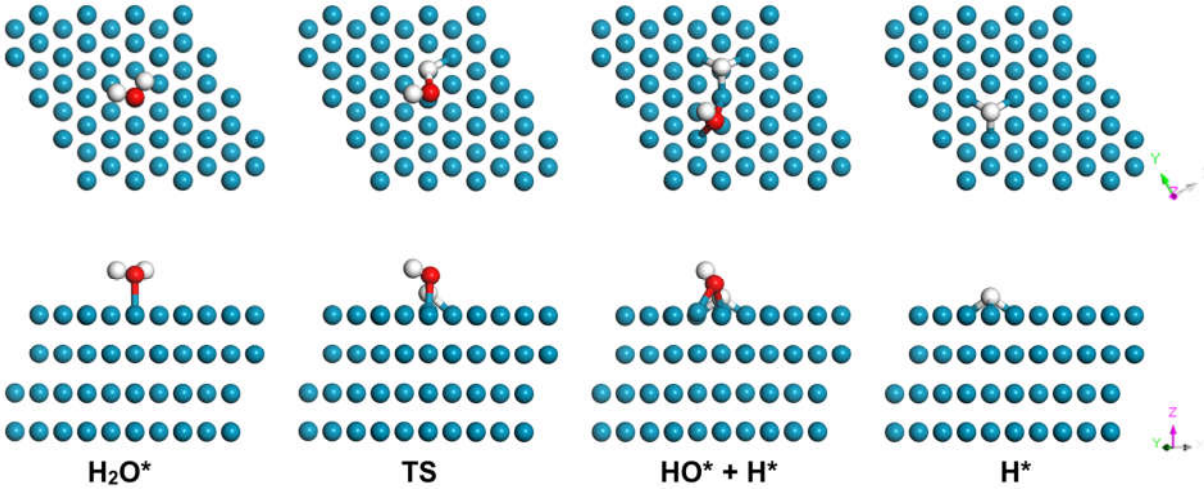


Figure S19. Top and side views of structures of the Ni (111) during alkaline HER process (Ni: blue, O: red and H: white).

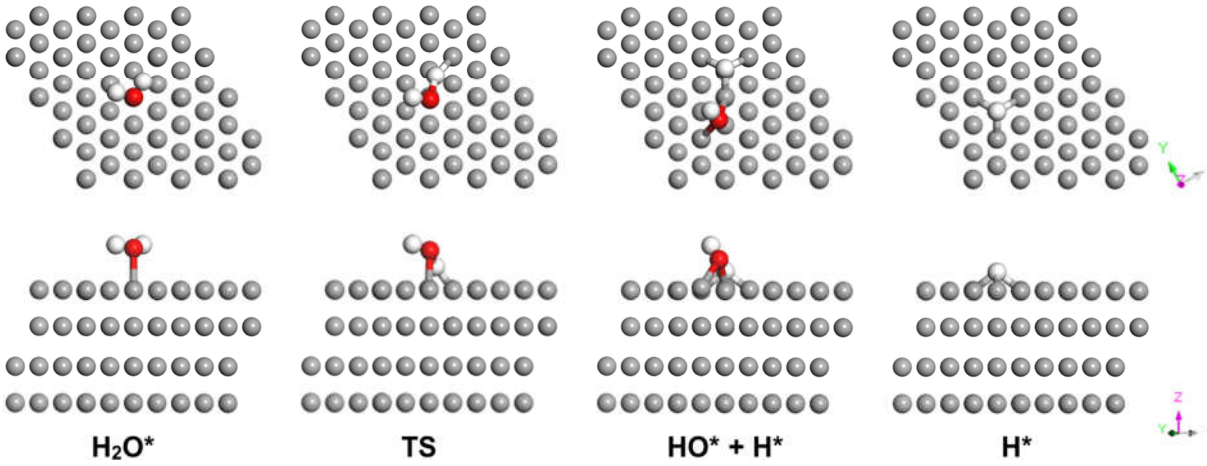
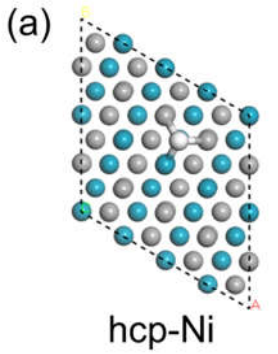
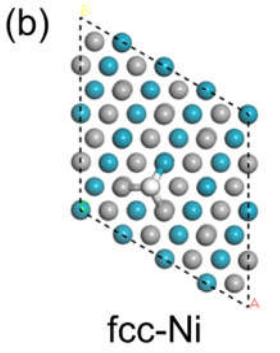
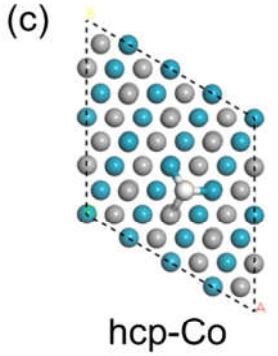
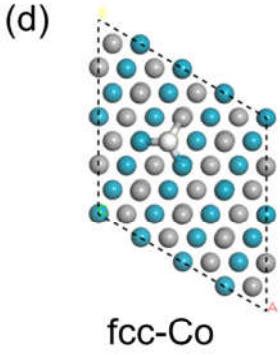


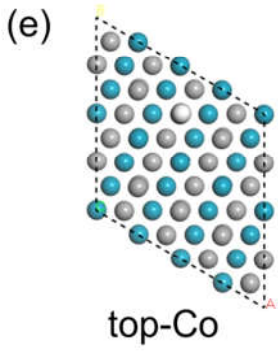
Figure S20. Top and side views of structures of the Co (111) during alkaline HER process (Co: grey, O: red and H: white).











H adsorption site	*E* (eV)
hcp-Ni	-0.50
fcc-Ni	-0.52
hcp-Co	-0.50
fcc-Co	-0.49
top-Co	0.10

Figure S21. Determination of the possible adsorption site of H on NiCo(111) surface; the corresponding H adsorption energy are listed in the table on right (Ni: blue, Co: grey, and H: white).

Table S1. Metal content in Ni_7Co_3/NC-500, Ni /NC, and Co /NC determined by ICP-MS.

Catalysts	Ni (wt%)	Co (wt%)	n(Ni): n(Co)
Ni_7Co_3/NC-500	47	21	2.24
Ni/NC	72	-	-
Co/NC	-	57	-

Table S2. Metal content in Ni_7Co_3/NC-400 and Ni_7Co_3/NC-600 determined by ICP-MS.

Catalysts	Ni (wt%)	Co (wt%)	n(Ni): n(Co)
Ni_7Co_3/NC-400	37	17	2.17
Ni_7Co_3/NC-600	54	24	2.25

Table S3. Summary of the HER performances of Ni_7Co_3/NC-500, Ni/NC, and Co/NC in 1 M KOH solution.

Catalysts	η_{10} (mV)	η_{50} (mV)	Tafel Slope (mV dec^{-1})	C_{dl} [a] (mF cm^{-2})	R_{ct} [b] (ohm)	j_0 [c] (mA cm^{-2})
Ni_7Co_3/NC-500	90	175	64	32.5	11.19	7.4×10^{-1}
Ni/NC	130	230	82	25.8	14.54	4.4×10^{-1}
Co/NC	210	330	150	22.5	37.69	1.5×10^{-1}

[a] Data were calculated according to the CV results;

[b] Data were measured at η = 200 mV;

[c] Exchange current densities (j_0) were obtained from Tafel curves using extrapolation methods.