Supplemental Materials for

Fe stabilized metallic phase of NiS_2 for highly efficient oxygen evolution reaction

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Table S1 The actual doped Fe concentrations in $Fe_xNi_{1-x}S_2$ measured by inductively coupled plasma mass spectrometry (ICP-MS). The actual x values are calculated using the following equation: $x = \frac{\rho_{Fe}}{M_{Fe}} \div (\frac{\rho_{Fe}}{M_{Fe}} + \frac{\rho_{Ni}}{M_{Ni}})$, where ρ represents the concentration of metal element in the solution obtained from ICP-MS, M_{Fe} = 55.85 g mol^{-1}, M_{Ni} = 58.69 g mol^{-1}.

x in $Fe_xNi_{1-x}S_2$	The concentrations of metal element from ICP-MS		x in $Fe_xNi_{1-x}S_2$ from ICP-MS
	ρ_{Fe} (mg L^{-1})	ρ_{Ni} (mg L^{-1})	
0	0	6.31	0
0.05	0.29	6.23	0.046
0.10	0.48	4.72	0.097
0.15	0.66	3.90	0.151
0.20	1.22	5.34	0.194

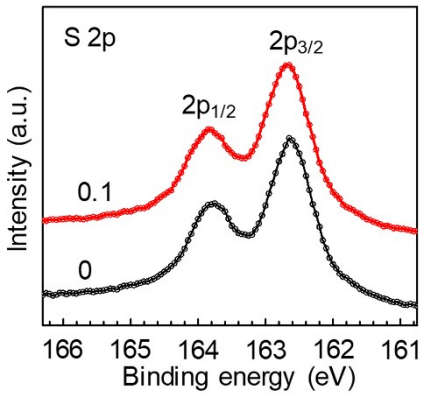

Fig. S1 S 2p XPS spectra of as-synthesized NiS_2 and $Fe_{0.1}Ni_{0.9}S_2$ nanosheets.

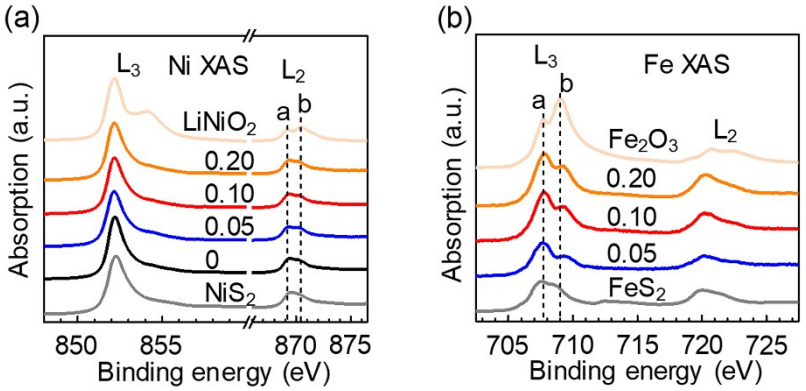

Fig. S2 (a) Ni L-edge XAS spectra of as-synthesized $Fe_xNi_{1-x}S_2$ (x=0, 0.05, 0.10 and 0.20) along with reference spectra of single crystal NiS_2 representing Ni^{2+} and $LiNiO_2$ representing Ni^{3+}. (b) Fe L-edge XAS spectra of as-synthesized $Fe_xNi_{1-x}S_2$ (x=0.05, 0.10 and 0.20) along with reference spectra of FeS_2 representing Fe^{2+} and Fe_2O_3 representing Fe^{3+}. The results suggest oxidization state of Ni and Fe in $Fe_xNi_{1-x}S_2$ are consistent with +2.

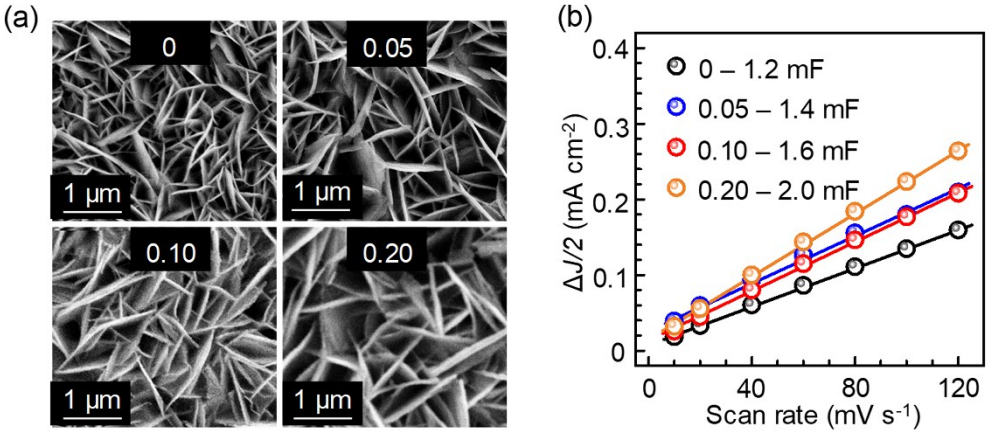

Fig. S3 (a) SEM images of as-synthesized $Fe_xNi_{1-x}S_2$ (x=0, 0.05, 0.10 and 0.20). (b) Plot of current density v.s. scan rates extracted from Fig. S4; the slope is double layer capacitance (C_{dl}) and is proportional to electrochemical active surface area (ECSA). This data suggests all samples have similar ECSA.

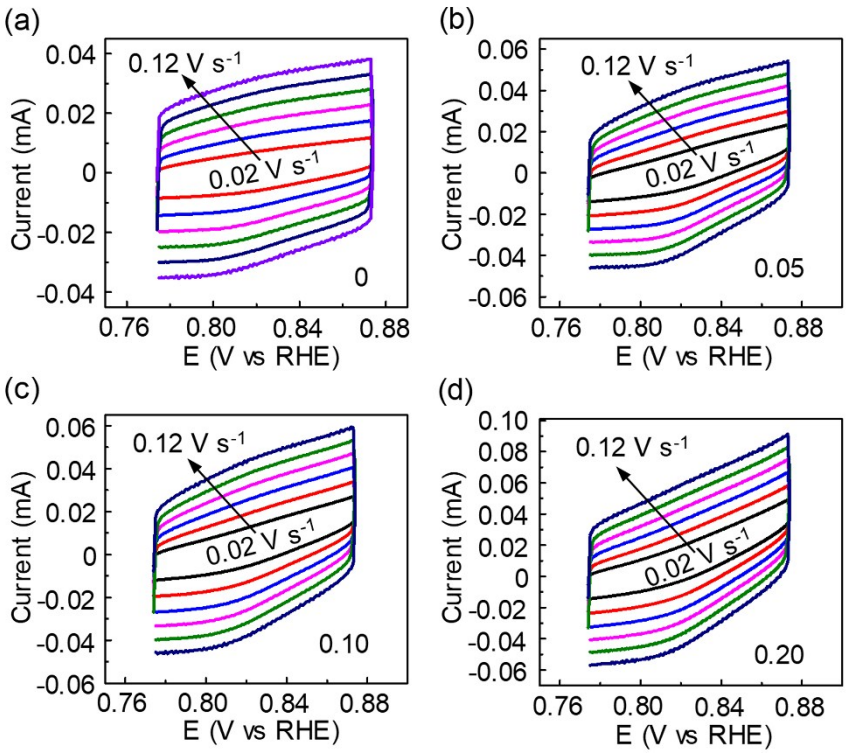

Fig. S4 Cyclic voltammogram of curves of $Fe_xNi_{1-x}S_2$ with different scan rates (0.02, 0.04, 0.06, 0.08, 0.10, 0.12 V s^{-1}): (a) x = 0; (b) x = 0.05; (c) x = 0.10; (d) x = 0.20.

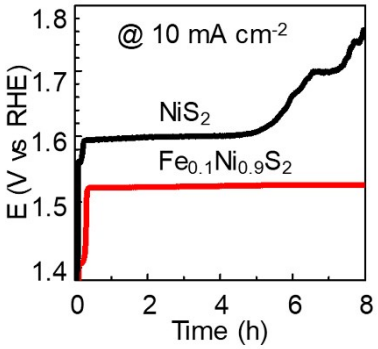

Fig. S5 Chronopotentiometric curves for first 8 hours of NiS_2 and $Fe_{0.1}Ni_{0.9}S_2$ extracted from Fig. 3d.

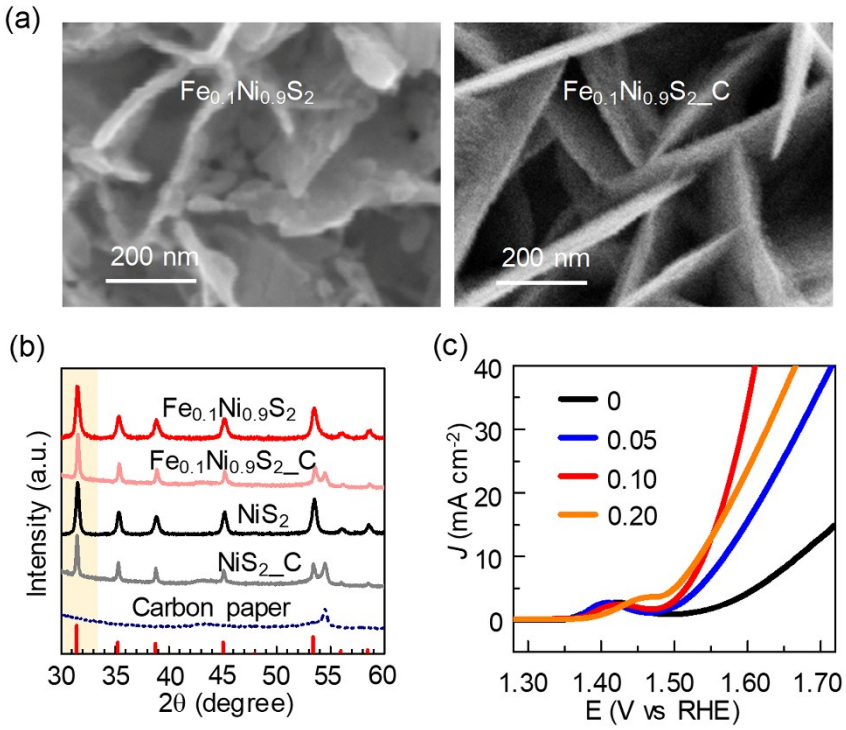

Fig. S6 (a) SEM image of as-synthesized $Fe_{0.1}Ni_{0.9}S_2$ powder (denoted as $Fe_{0.1}Ni_{0.9}S_2$) and $Fe_{0.1}Ni_{0.9}S_2$ prepared on carbon paper (denoted as $Fe_{0.1}Ni_{0.9}S_2$_C). (b) XRD patterns of the catalysts powder (denoted as NiS_2 and $Fe_{0.1}Ni_{0.9}S_2$) and the catalysts prepared on carbon paper (denoted as NiS_2_C and $Fe_{0.1}Ni_{0.9}S_2$_C). (c) Linear sweep voltammetry (LSV) polarization curves of $Fe_xNi_{1-x}S_2$ (x=0, 0.05, 0.10 and 0.20) prepared on carbon paper.

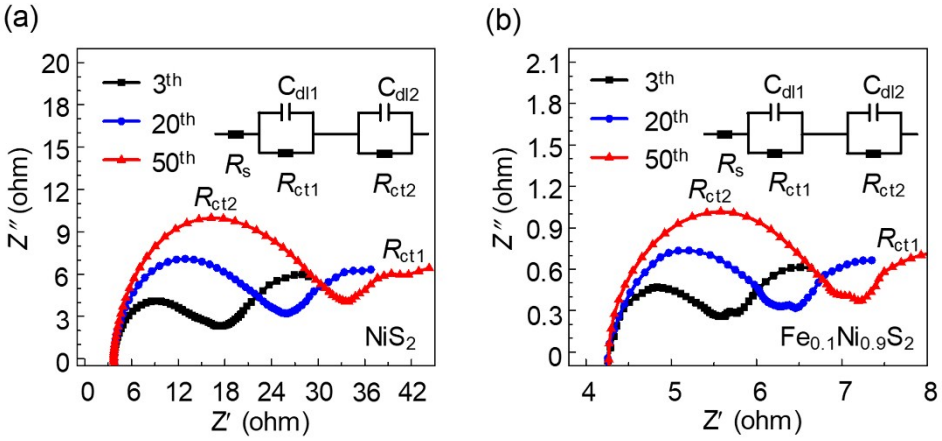

Fig. S7 Nyquist plots of the EIS at potential of +1.63 V vs. RHE for (a) NiS_2 after 3, 20 and 50 cycles of OER measurements; (b) $Fe_{0.1}Ni_{0.9}S_2$ after 3, 20 and 50 cycles of OER measurements; Inset show the equivalent circuit for fitting the data, consisting of an electrolyte resistance (R_s), a charge transfer resistance (R_{ct1}) caused by electron transfer from electrolyte to the catalyst, and a resistance (R_{ct2}) caused by electron transfer from $Fe_xNi_{1-x}OOH$ layer to $Fe_xNi_{1-x}S_2$ layer.

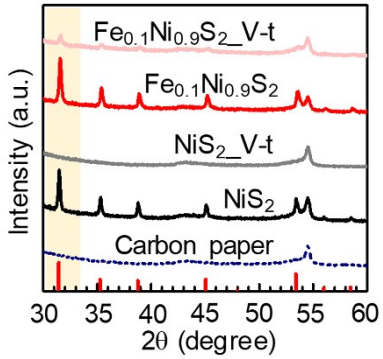

Fig. S8 XRD patterns of the as-synthesized samples (denoted as NiS_2 and $Fe_{0.1}Ni_{0.9}S_2$) and those of the samples after V-t test (denoted as NiS_2_V-t and $Fe_{0.1}Ni_{0.9}S_2$_V-t).

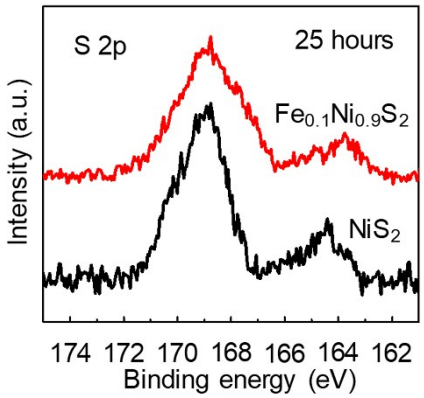

Fig. S9 S 2p XPS spectra for NiS_2 and $Fe_{0.1}Ni_{0.9}S_2$ after 25 hours stability test.

Table S2 Lattice parameters and the bond length of $Fe_xNi_{1-x}S_2$ (x=0, 0.05, 0.10 and 0.20) and FeS_2. The shorter Ni/Fe-S bond lengths suggest larger Ni/Fe-S binding energies, and thereby higher energy barrier to break Ni/Fe-S bonds for transformation of $Fe_xNi_{1-x}S_2$ into $Fe_xNi_{1-x}OOH$.

x in $Fe_xNi_{1-x}S_2$	*a* (Å)	Ni-S (Å)	S-S (Å)	Fe-S (Å)
0.00	5.679	2.400	2.038	-
0.05	5.677	2.398	2.046	-
0.10	5.674	2.396	2.048	-
0.20	5.667	2.390	2.077	-
FeS_2[1]	5.404	-	2.171	2.259

Reference

1. M. Elliott, *J. Chem. Phys.*, 1960, **33**, 903-905.