Supporting Information

3D Hierarchical Network Derived from 2D Fe-doped NiSe Nanosheets/Carbon Nanotubes with Enhanced OER Perfromance for Overall Water Splitting

Kai Chang[1], Duy Thanh Tran[1], Jingqiang Wang[1], Nam Hoon Kim[1], Joong Hee Lee[1,2]*
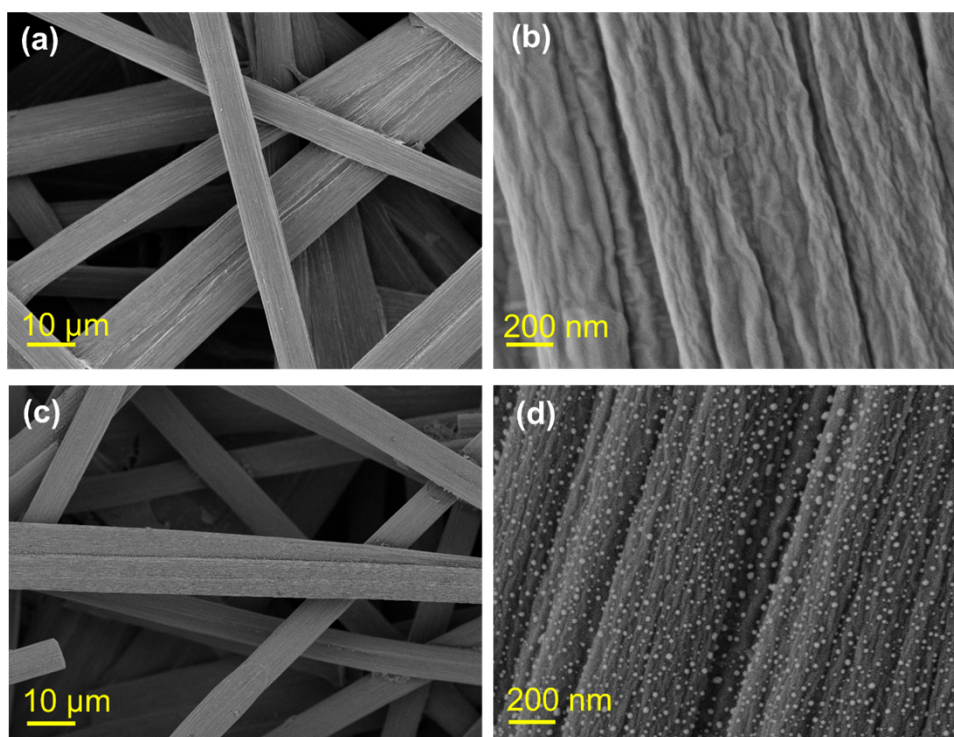

Figure S1. (a and b) SEM image of CP surface; (c and d) distribution of Ni NPs on CP surface.
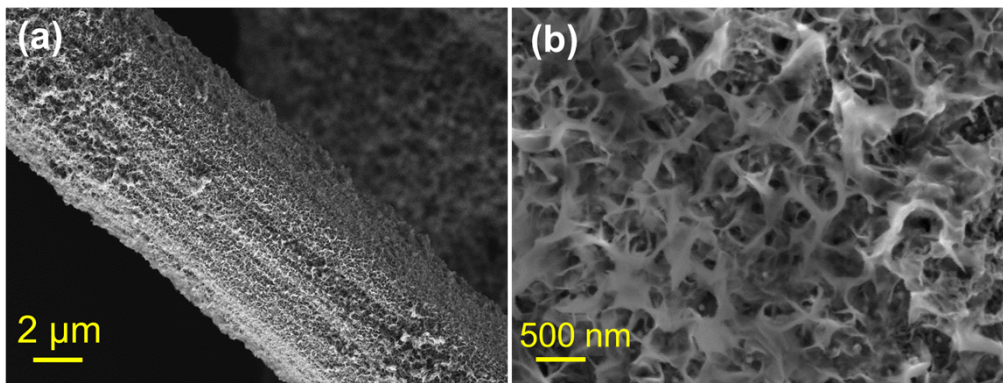
Figure S2. (a and b) SEM images of Fe-doped NiSe NSs on CP at different magnifications.
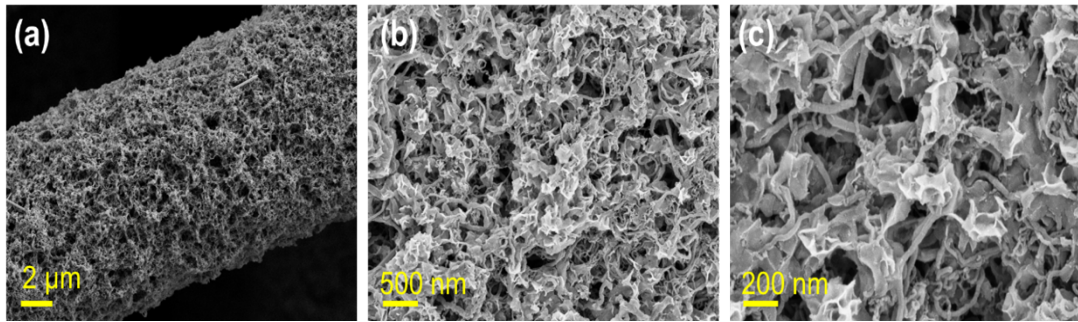
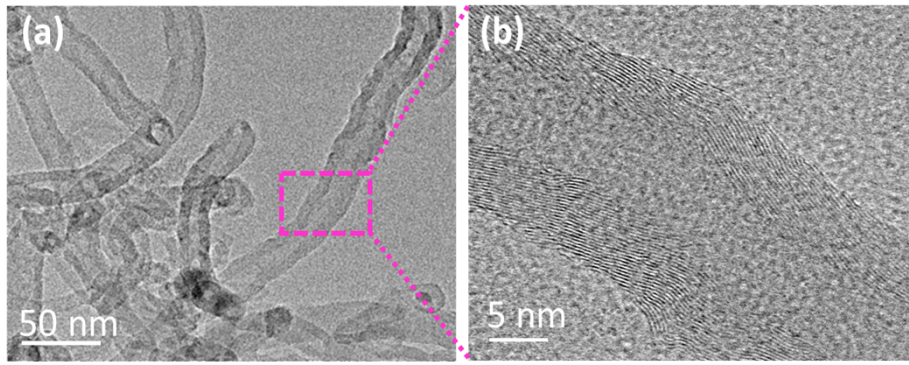**Figure S3.** (a-c) SEM images of NiSe NSs/CNTs at different magnifications.

Figure S4. (a) TEM and (b) HR-TEM images of CNTs grown on CP.
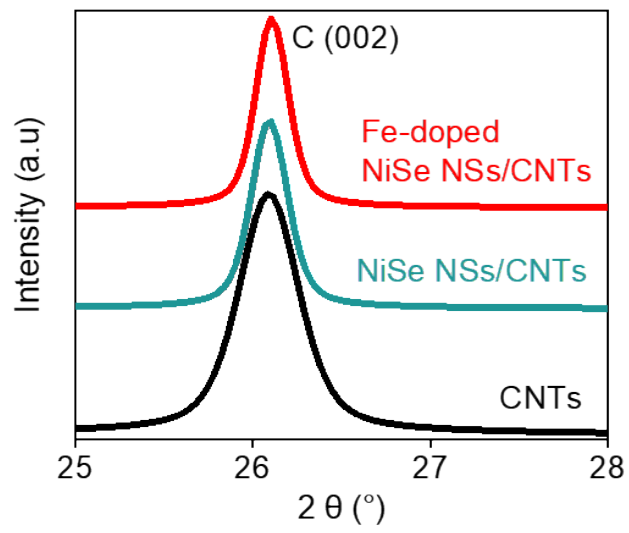
Figure S5.Magnified XRD spectra of CNTs, NiSe NSs/CNTs and Fe-doped NiSe NSs/CNTs
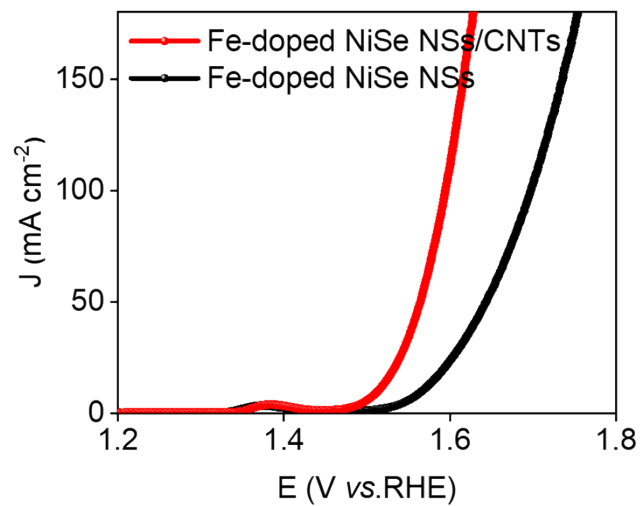
on CP

Figure S6. Comparison of OER activities between Fe-doped NiSe NSs/CNTs and Fe-doped NiSe NSs.
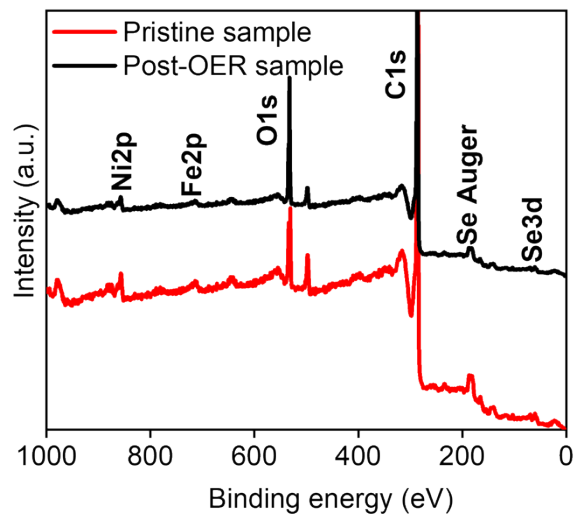
Figure S7. Comparison of XPS survey spectra between the pristine and post-OER Fe-doped NiSe NSs/CNTs.
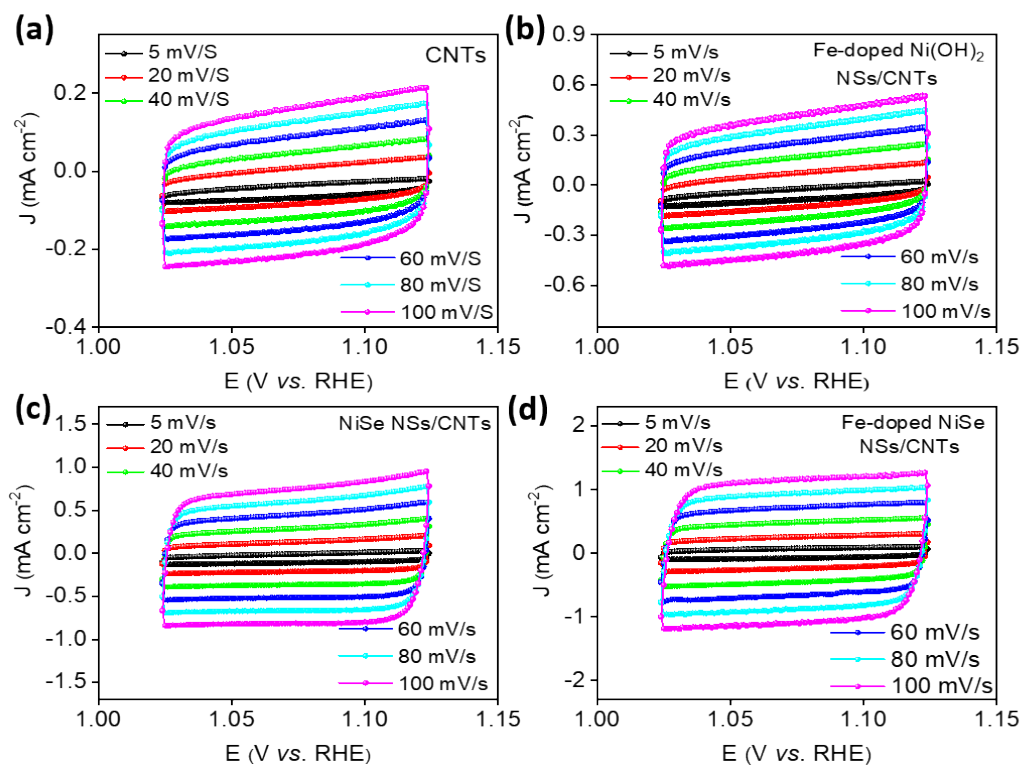
Figure S8. Typical CV curves of (a) CNTs, (b) Fe-doped Ni(OH)₂ NSs/CNTs, (c) NiSe NSs/CNTs, and (d) Fe-doped NiSe NSs/CNTs at different scan rates in 1.0 M KOH.

Turnover frequency (TOF) calculations

The calculation of TOF was refered from a method reported by Wang et al.[1] In a specific procedure, CV curves were initially measured at 50 mV s⁻¹ in PBS solution (pH = 7.0). Then the absolute components of the voltammetric charges (cathodic and anodic) reported during a single measurement was added. The quantity of active species (n) is calculated according to Equation S1.

$$n = \frac{Q}{2F} = \frac{It}{2F} = \frac{IV}{2Fv} \qquad (S1)$$

where Q is the voltammetric charge, F is the Faraday constant (C mol⁻¹), I is the current (A), t is the time (s), V is the voltage (V) and v is the scanning rate (V s⁻¹).
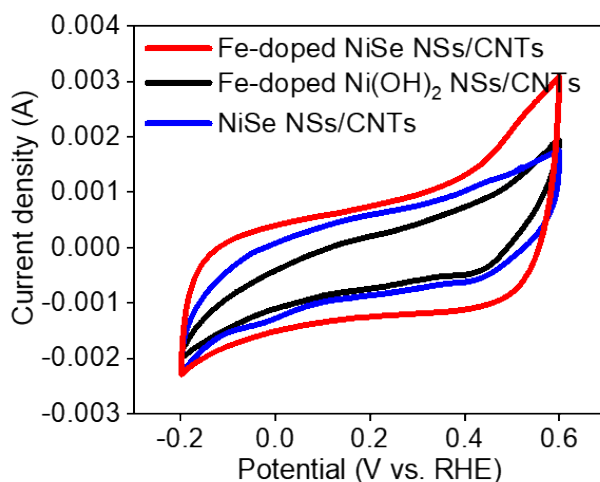

Figure S9. CV curves of Fe-doped NiSe NSs/CNTs, NiSe NSs/CNTs and Fe-doped Ni(OH)$_2$ NSs/CNTs in PBS solution (pH = 7.0) at a scan rate of 50 mV s⁻¹.

The active site number of Fe-doped NiSe NSs/CNTs, NiSe NSs/CNTs and Fe-doped Ni(OH)$_2$ NSs/CNTs are calculated to be 1.7448259e-7, 1.12361414e-7, and 6.96084676e-8 mol, respectively. Therefore, the TOF can be calculated by Equation S2:

$$TOF = \frac{|j| A}{nmF}$$

(S2)

j: Current (A) during the linear sweep voltammetry (LSV) tests.

A: The area of the electrode (1 cm^{-2})

F: Faraday constant (96485 C mol^{-1}).

n: Number of active sites (mol).

m: The factor 1/m represents that m electrons are required to form one H_2/O_2 molecule from water, which means that the m values for hydrogen evolution and oxygen evolution reactions are 2 and 4, respectively.
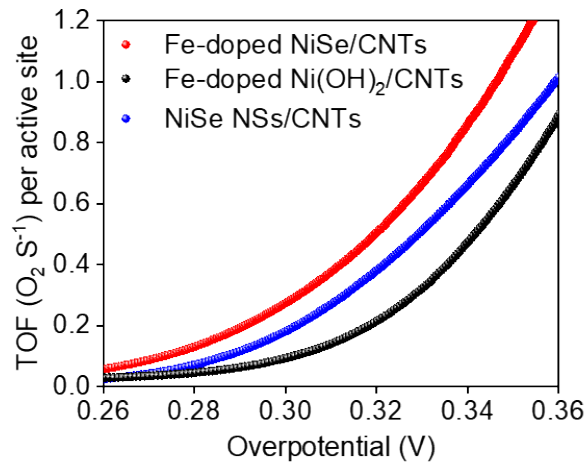

Figure S10. Calculated O_2 TOF values of Fe-doped NiSe NSs/CNTs, NiSe NSs/CNTs, and Fe-doped Ni(OH)$_2$ NSs/CNTs.
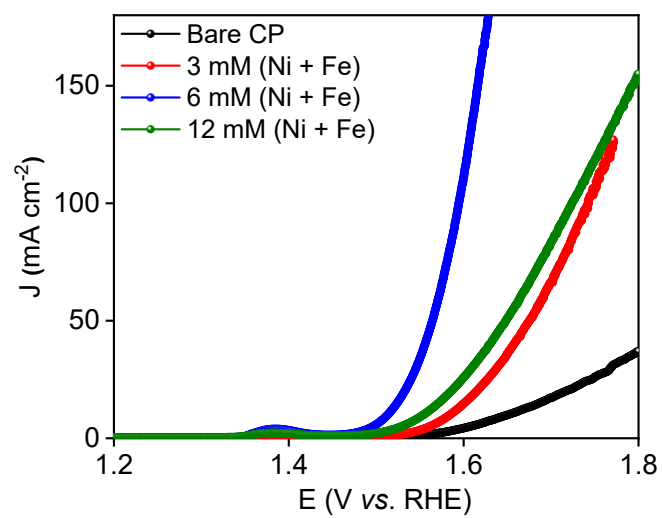
Figure S11. OER performance of the Fe-doped NiSe NSs/CNTs on CP derived from different

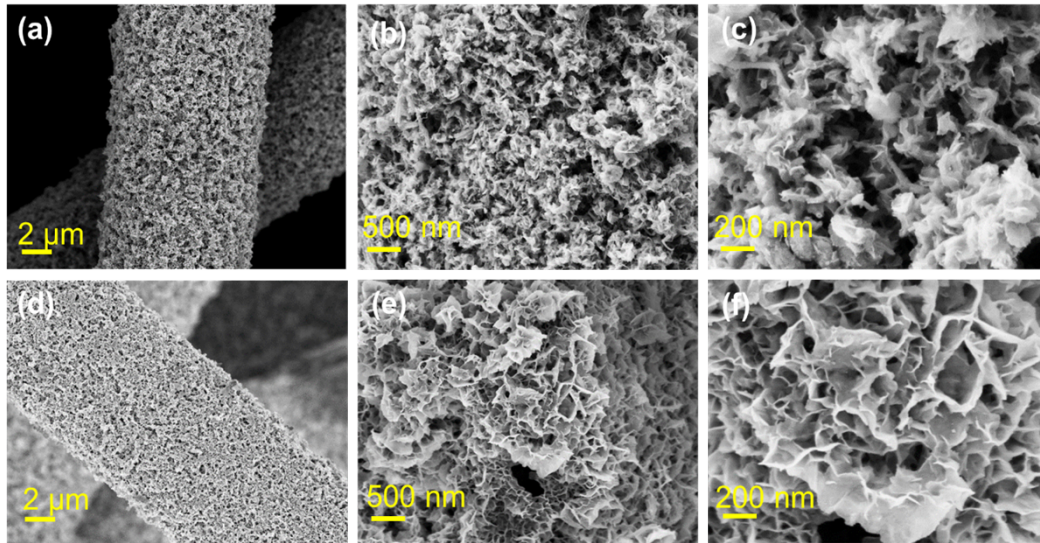total concentrations of (Ni + Fe) precursors.

Figure S12. SEM images of the Fe-doped NiSe NSs/CNTs materials derived from different total (Ni + Fe) precursors: (a-c) 3.0 mM, and (d-f) 12.0 mM.

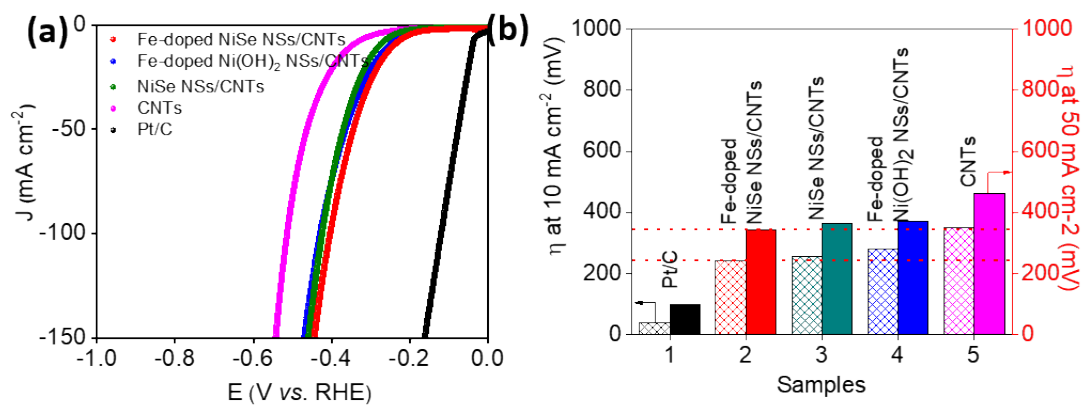**Figure S13. (a)** iR-corrected LSV curves of materials towards HER in 1.0 M KOH medium; **(b)** A comparison of η value at 10 mA cm^{-2} between materials.
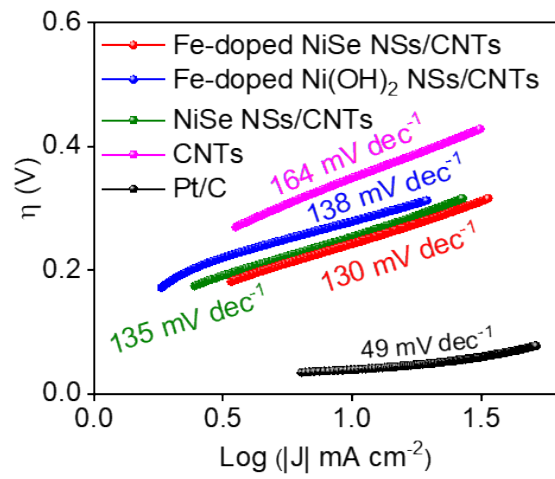
Figure S14. Tafel slope values of materials towards HER in 1.0 M KOH medium.

Table S1. Comparison of O_2 TOF between the Fe-doped NiSe NSs/CNTs with recently reported electrocatalysts.

Catalysts	O_2 TOF at $\eta_{350\,mV}$ (s^{-1})	References
Fe-doped NiSe NSs/CNTs	**1.097**	**This work**
CoO@NiFe LDH/NF	0.59	*Chem. Eng. J.*, 2021, 410, 128366
FeNi$_3$@NC	0.149	*Appl. Catal. B.*, 2020, 268, 118729
Ni-doped FeF$_2$	0.05	*Chem. Commun.*, 2020, 56, 7889
Ni$_{2.2}$Fe(OH)$_x$ HNAs	0.15	*Sci. Rep.*, 2017, 7, 46154
FeP/Ni$_2$P@CNT	0.979	*J. Alloys Compd.*, 2021, 883, 160926
Ni-MOF/NF	0.24	*Inorg. Chem. Front.*, 2021, 8, 3007-3011
Co-Fe-S NFs@MS/NF	0.441	*Electrochim. Acta*, 2020, 361, 137038
NiFe LDH-Ci/CC	0.59	*Chem. Eng. J.*, 2021, 410, 128366
Vo-(Co, Fe)$_3$O$_4$/CC	0.43	*Appl. Surf. Sci.*, 2020, 529, 147125

References

1. P. Wang, R. Qin, P. Ji, Z. Pu, J. Zhu, C. Lin, Y. Zhao, H. Tang, W. Li, S. Mu , *Small*, 2020, **16**, 2001642