Supporting Information

Facile Synthesis of Nanoparticle-stacked Tungsten-doped Nickel Iron Layered Double Hydroxide Nanosheets for Boosting Oxygen Evolution Reaction

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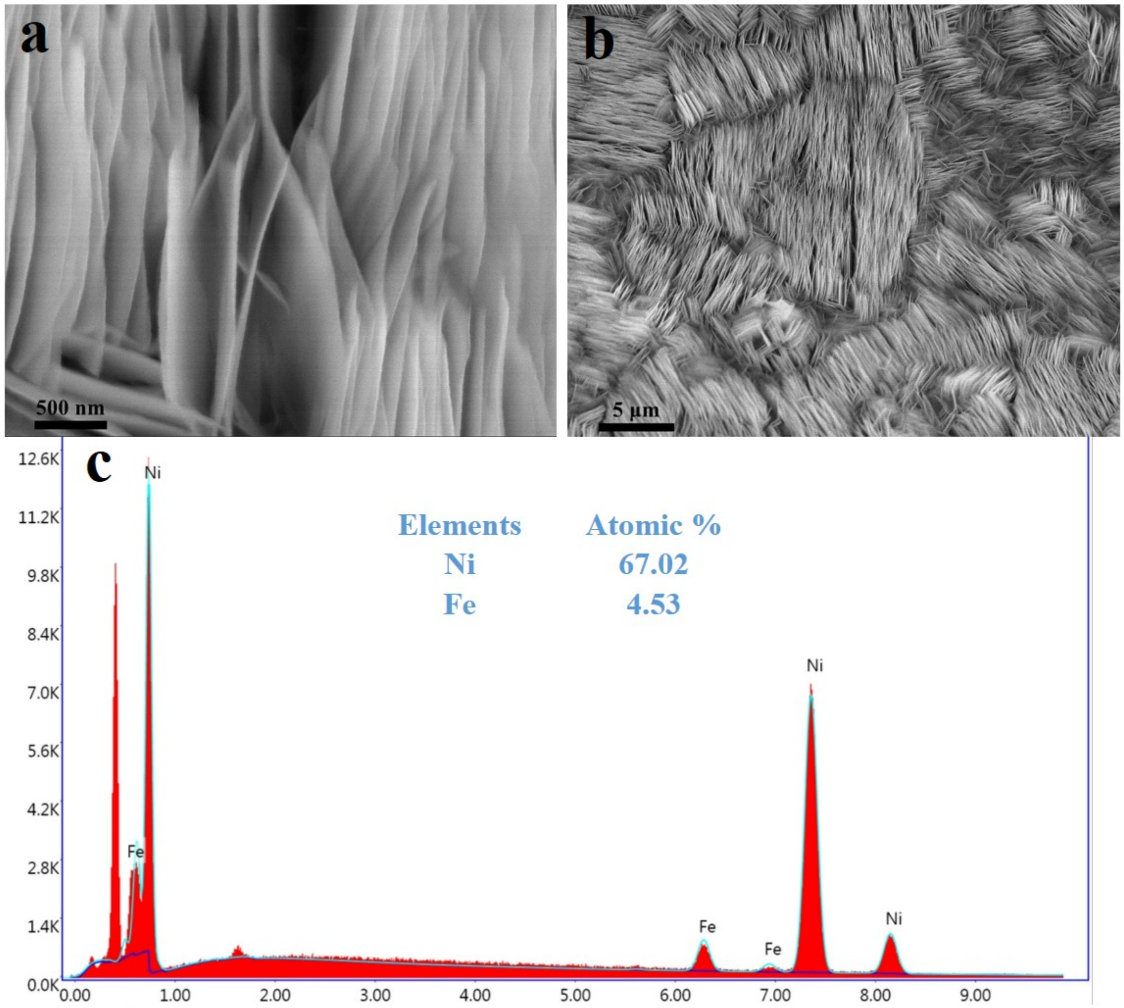

Figure S1. (a, b) SEM images and (c) the corresponding EDS spectrum of Ni_3Fe LDH/NF.

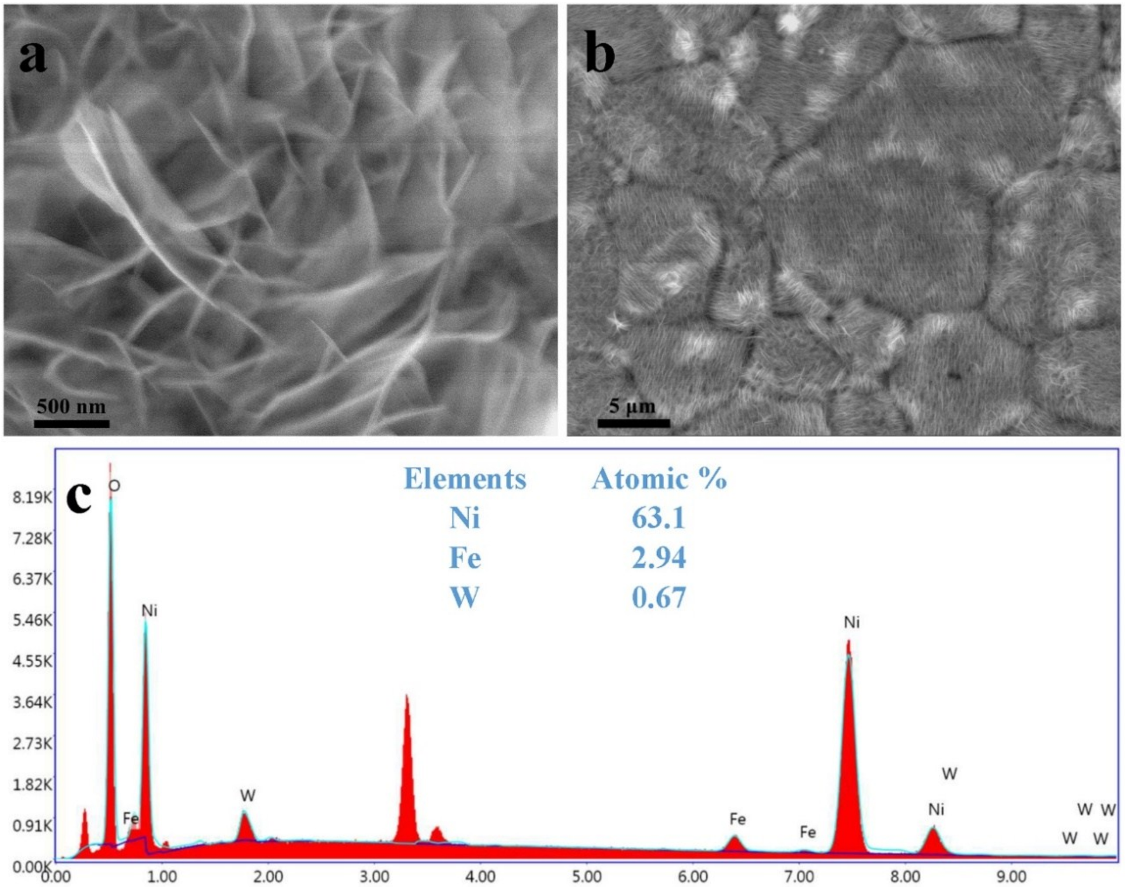

Figure S2. (a, b) SEM images and (c) the corresponding EDS spectrum of $Ni_3FeW_{0.5}$ LDH/NF.

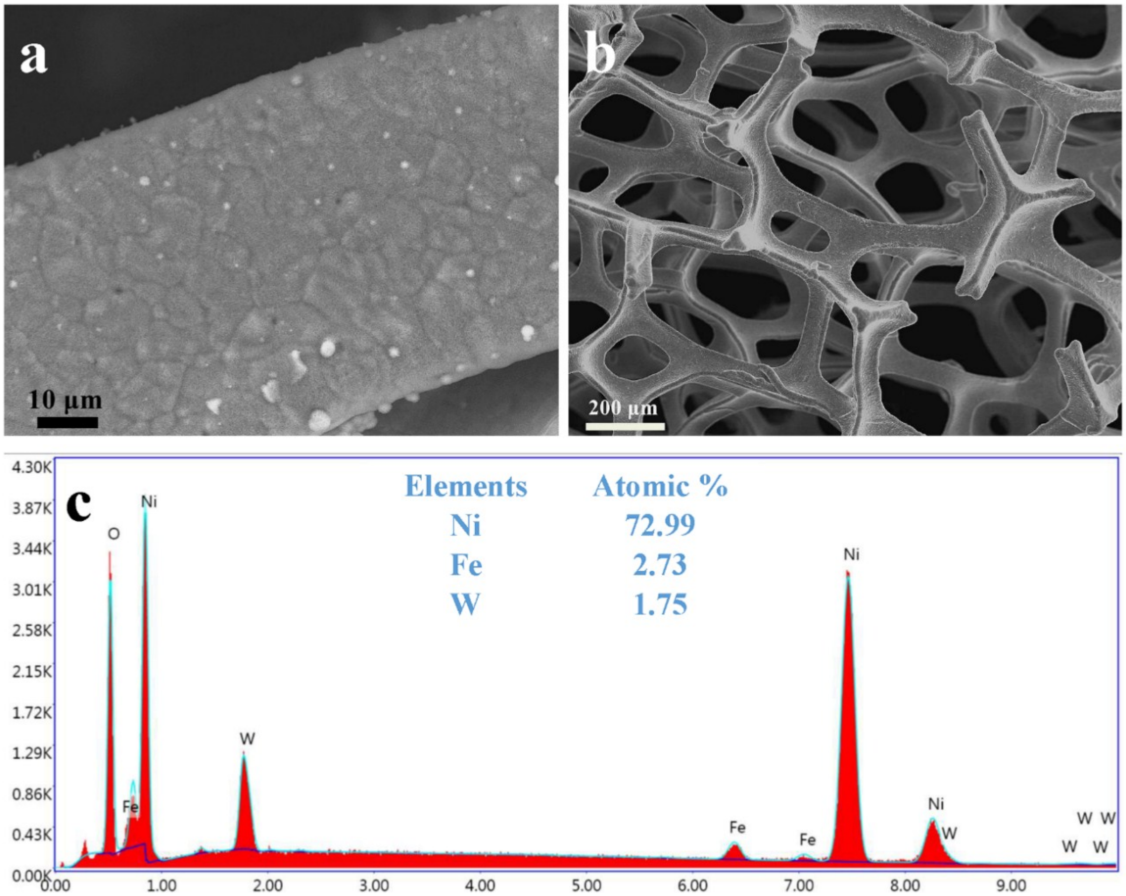

Figure S3. (a, b) SEM images and (c) the corresponding EDS spectrum of the Ni_3FeW LDH/NF.

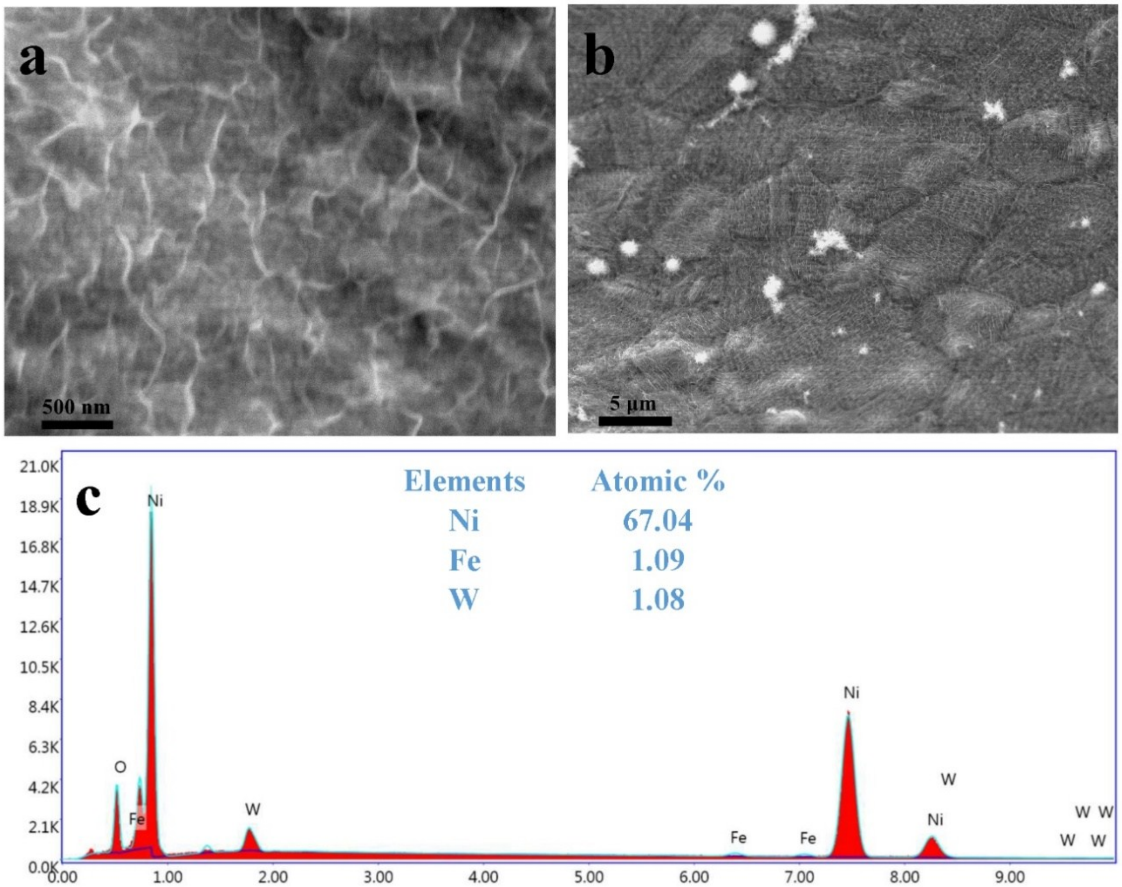

Figure S4. (a, b) SEM images and (c) the corresponding EDS spectrum of Ni_3FeW_2 LDH/NF.

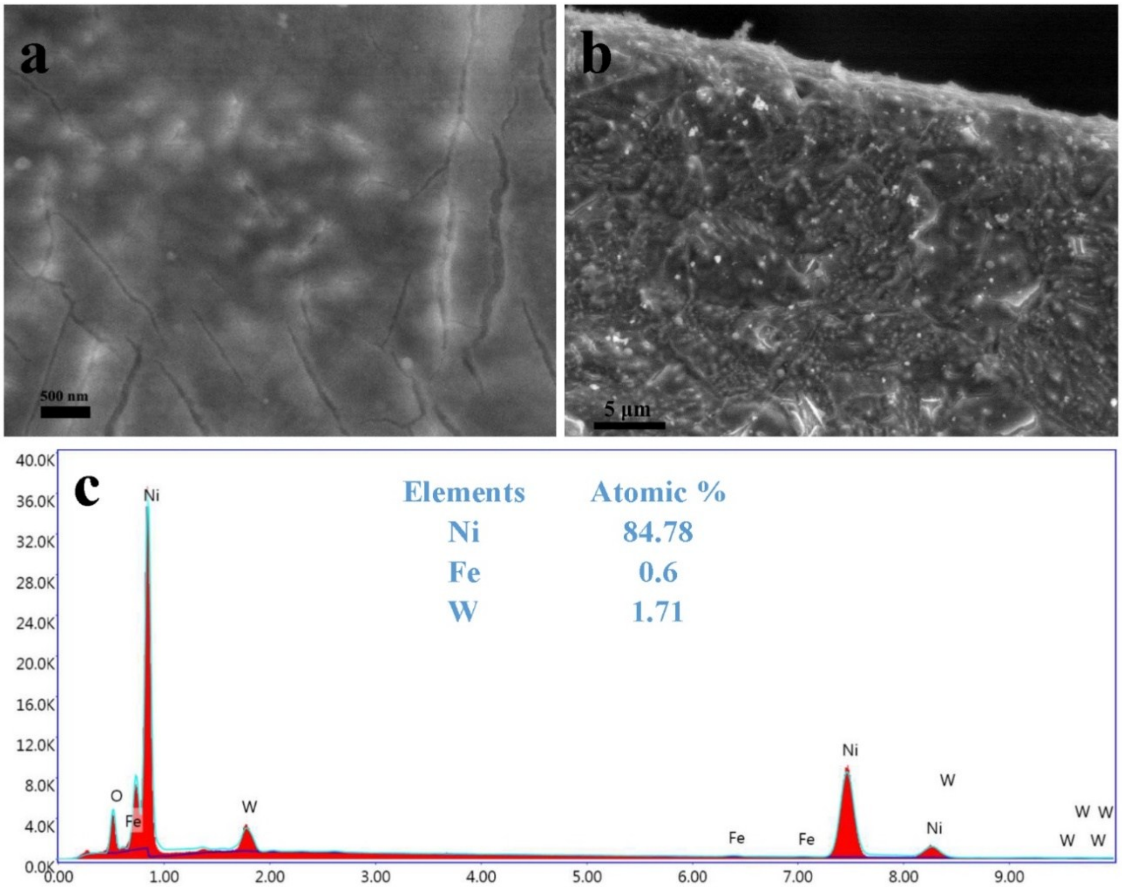

Figure S5. (a, b) SEM images and (c) the corresponding EDS spectrum of Ni_3FeW_3 LDH/NF.

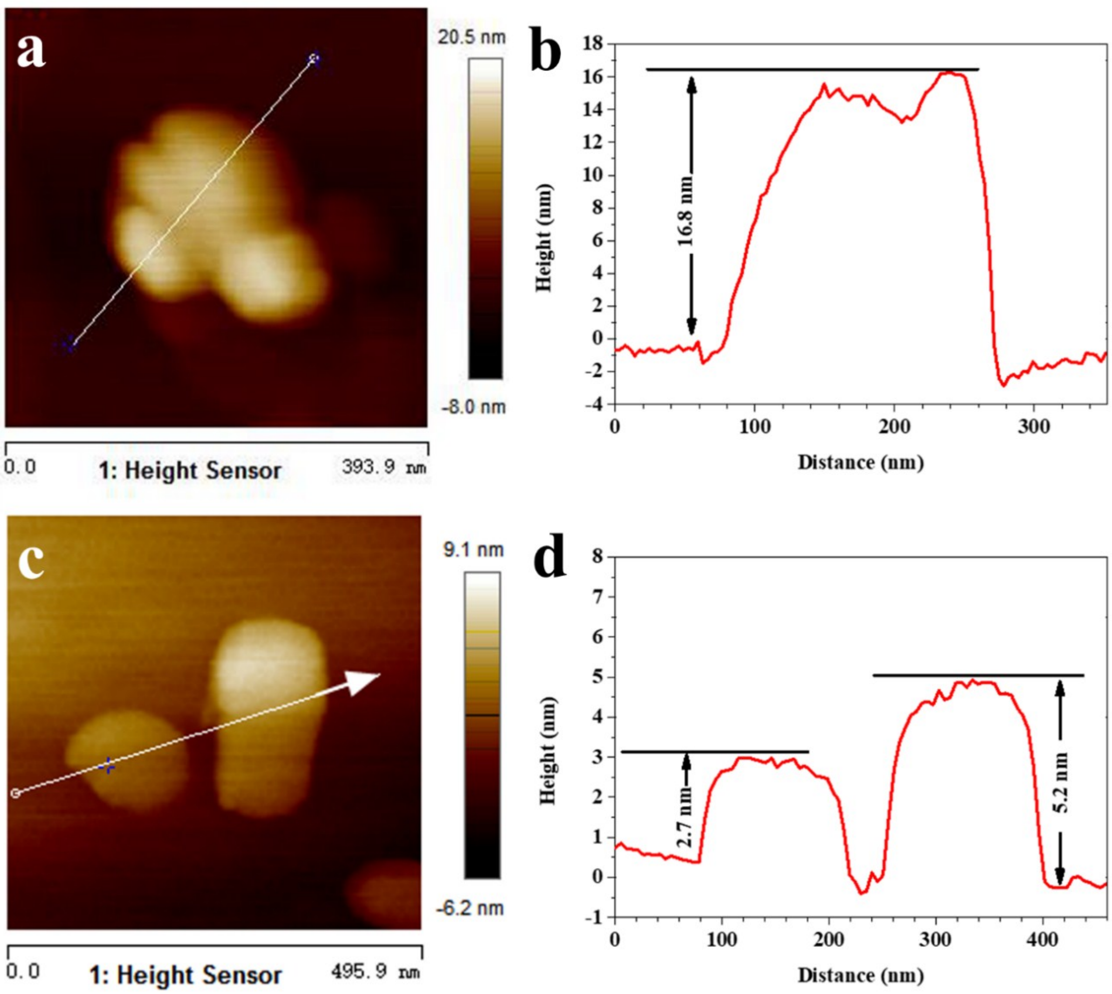

Figure S6. AFM images and corresponding height profiles of (a, b) Ni_3Fe LDH and (c, d) Ni_3FeW LDH nanosheets.

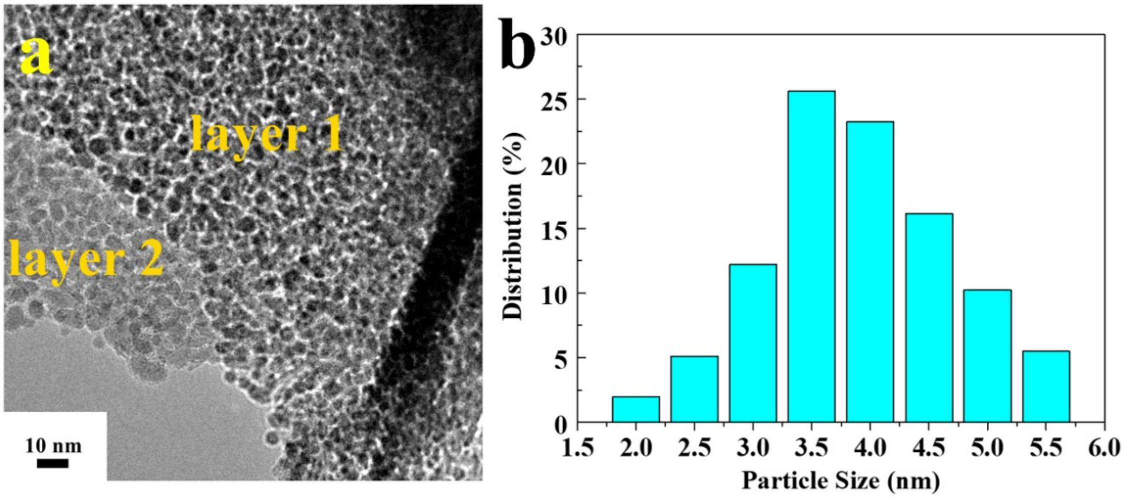

Figure S7. (a) TEM image of two layers of Ni_3FeW LDH nanosheets and (b) the corresponding nanoparticle diameter distribution calculated from (a).

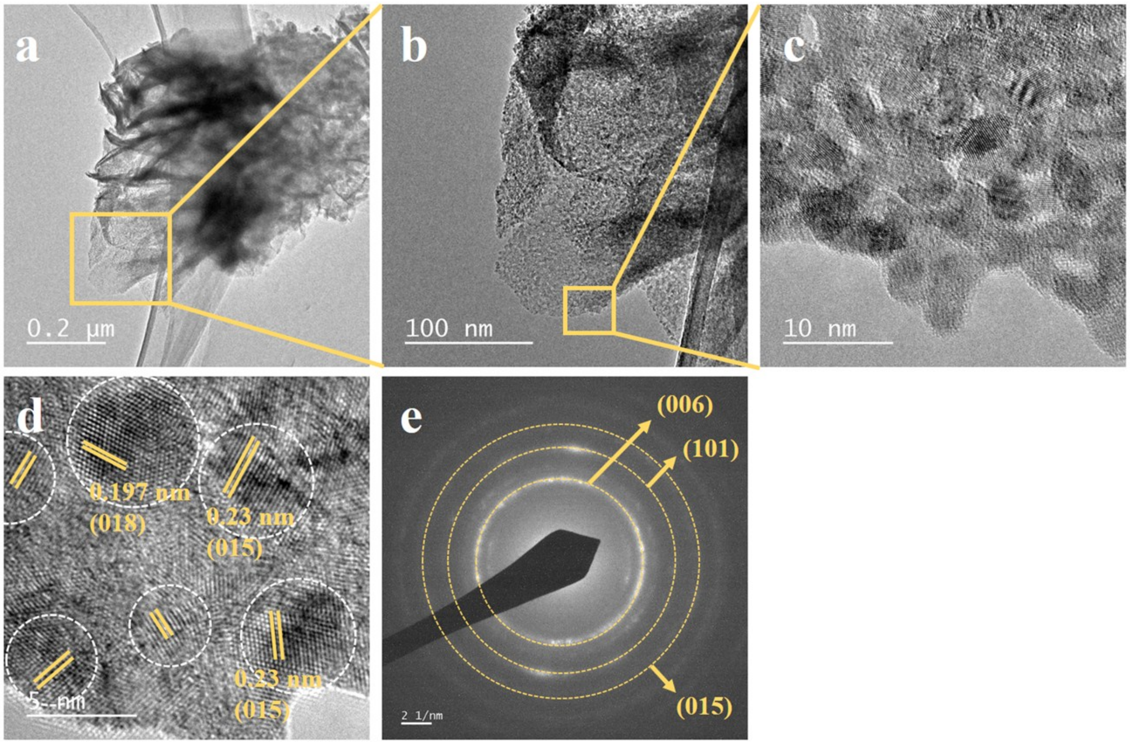

Figure S8. (a, b) TEM and (c, d) HRTEM images and (e) SAED pattern of Ni_3FeW LDH nanosheets.

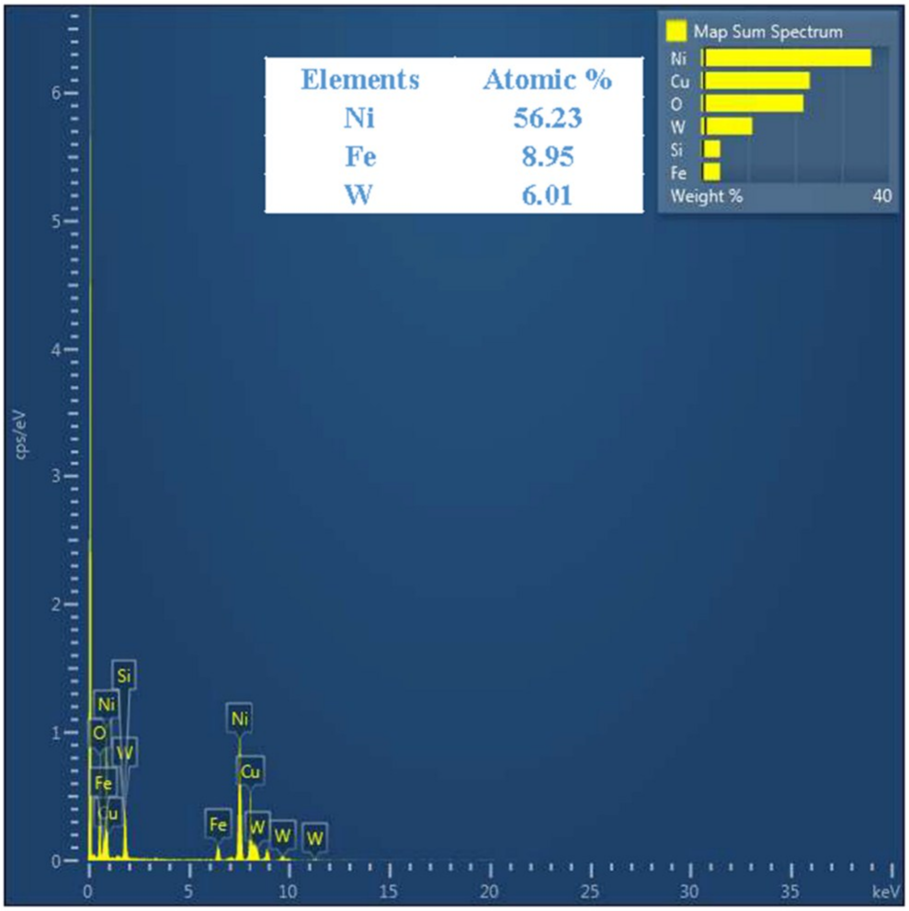

Figure S9. TEM EDX spectrum of Ni_3FeW LDH nanosheets.

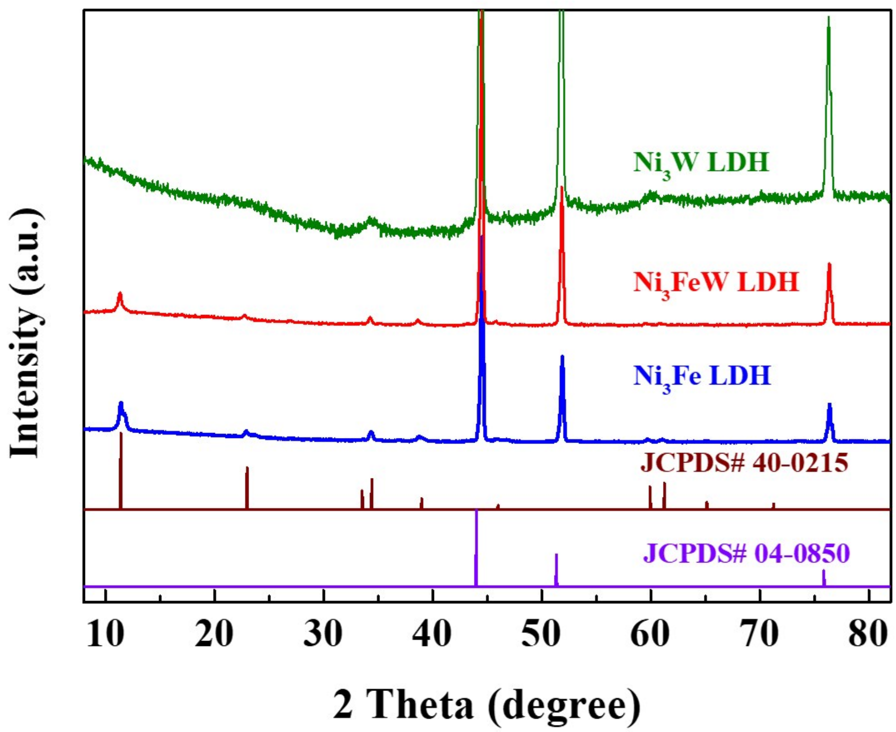

Figure S10. XRD patterns of Ni_3Fe, Ni_3FeW, and Ni_3W LDH/NF and the standard PDF cards of iron nickel carbonate hydroxide (JCPDS#40-0215) and Ni (JCPDS#04-0850).

The introduction of Fe or W does not change the phase of the LDH.

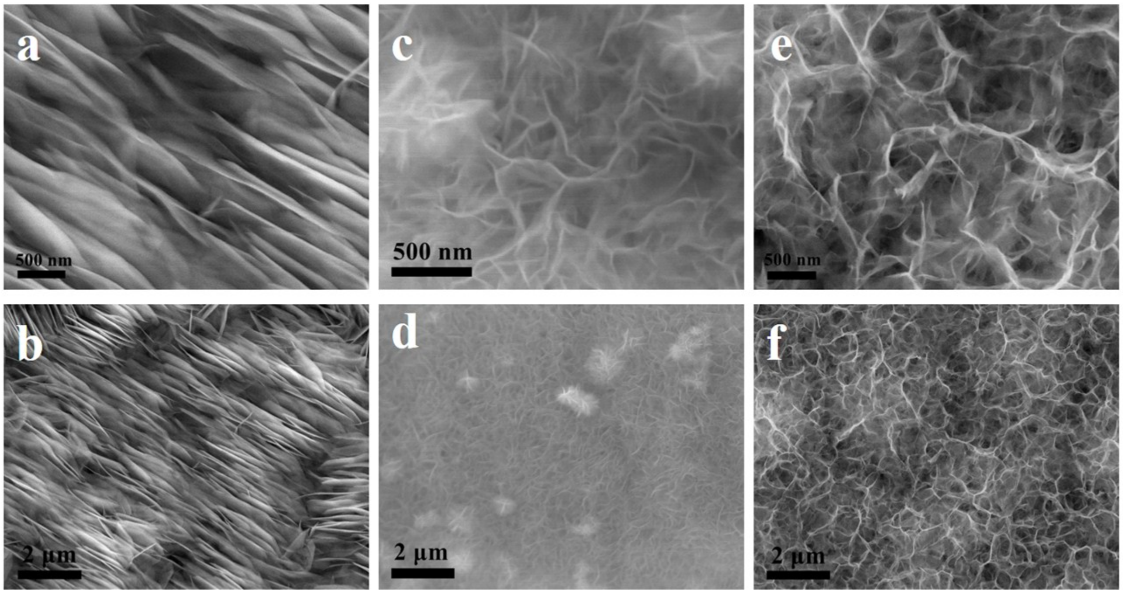

Figure S11. SEM images of (a, b) Ni_3Fe, (c,d) Ni_3FeW, and (e,f) Ni_3W LDH/NF.

After introducing W, the thickness of Ni_3FeW LDH nanosheets decreases. Ni_3W LDH nanosheets are more porous and thinner than either Ni_3Fe or Ni_3FeW LDH. It can be concluded that either Fe or W can affect the nanosheet structure of the final LDH, especially W.

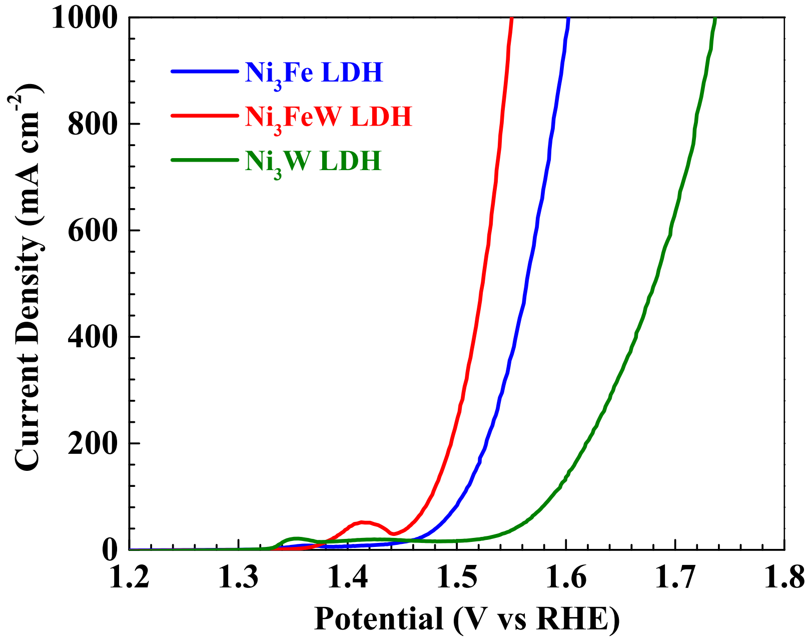

Figure S12. OER LSV curves of Ni_3Fe, Ni_3FeW, and Ni_3W LDH samples in 1.0 M KOH electrolyte.

In terms of OER performance, Ni_3FeW LDH is the best, Ni_3Fe LDH is the second best, and Ni_3W LDH is the worst. The performance difference between the Ni-Fe-W and Ni-Fe LDHs is much smaller than that between the Ni-Fe and Ni-W LDHs, indicating that Fe plays a critical role in the OER activity of LDH-based catalysts. The excellent OER activity of both the Ni_3Fe and Ni_3FeW LDHs should be attributed to the intrinsically high catalytic activity of Ni-Fe LDH.

The exact role of iron and the mechanism underlying its effect on the OER performance of Ni-Fe LDH catalysts are still under debate. Previous reports have shown that a layer of NiOOH can be developed on the Ni-Fe LDH catalyst surface at potentials approaching the evolution of oxygen.[1] Thus, NiOOH has been demonstrated to be the active phase. Normally, the introduction of Fe could create more structural defects and alter the local environment of Ni-O and NiOOH, thus enhancing the OER activity.[2] Here we mainly focus on the effects of W in enhancing the OER performance of Ni-Fe LDH.

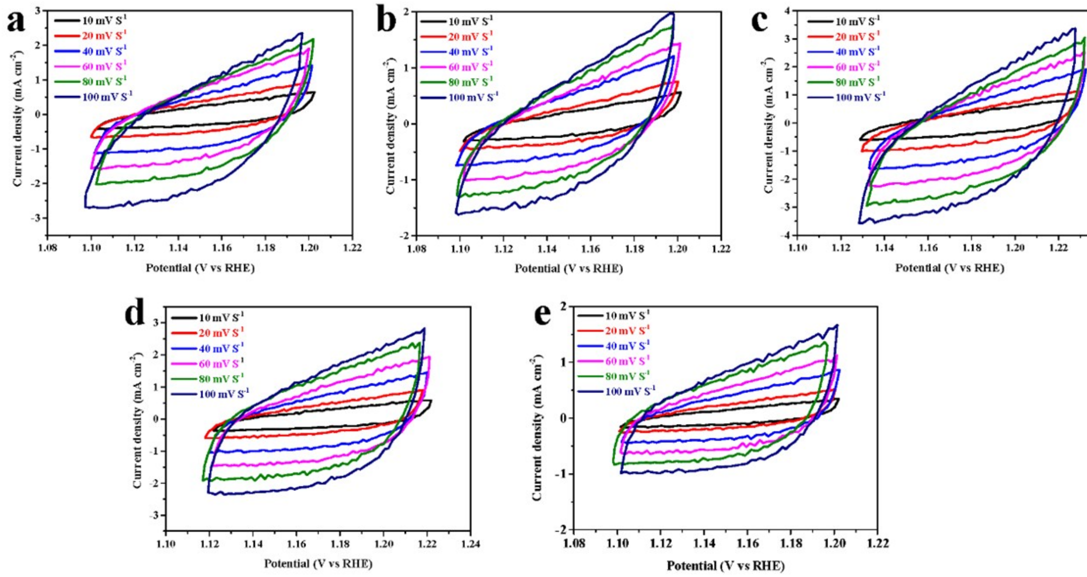

Figure S13. CV curves of (a) Ni_3Fe LDH/NF, (b) $Ni_3FeW_{0.5}$ LDH/NF, (c) Ni_3FeW LDH/NF, (d) Ni_3FeW_2 LDH/NF, and (e) Ni_3FeW_3 LDH/NF at different scan rates of 10, 20, 40, 60, 80, and 100 mV S^{-1} in a non-faradaic region.

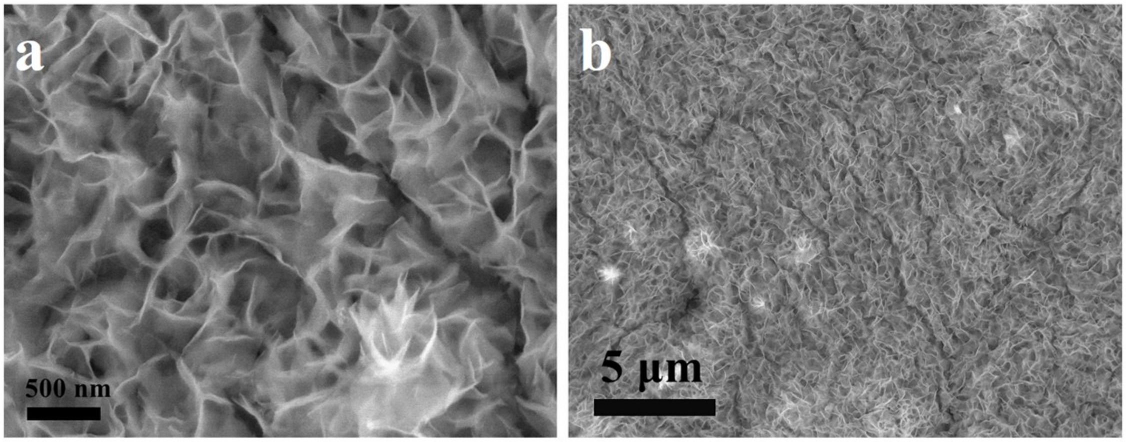

Figure S14. (a, b) SEM images of Ni_3FeW LDH/NF after 5000 CV scans.

Reference:

1. H. Liu, Y. Wang, X. Lu, Y. Hu, G. Zhu, R. Chen, L. Ma, H. Zhu, Z. Tie, J. Liu and Z. Jin, *Nano Energy*, 2017, **35**, 350-357.
2. Z. Lu, W. Xu, W. Zhu, Q. Yang, X. Lei, J. Liu, Y. Li, X. Sun and X. Duan, *Chem. Commun. (Camb)*, 2014, **50**, 6479-6482.