Supporting Information


Libo Wu,a,b Luo Yu,a Fanghao Zhang,a,c Dezhi Wang,a Dan Luo,a Shaowei Song,a,b Chuqing Yuan,b Alamgir Karim,b,d Shuo Chen,a, and Zhifeng Ren,a,*

a Department of Physics and Texas Center for Superconductivity at the University of Houston (TcSUH), University of Houston, Houston, Texas 77204, USA
b Materials Science and Engineering Program, University of Houston, Houston, Texas 77204, USA
c Department of Chemistry, University of Houston, Houston, Texas 77204, USA
d Department of Chemical and Biomolecular Engineering, University of Houston, Houston, Texas 77204, USA

*Corresponding author: Zhifeng Ren, E-mail address: zren@uh.edu
Figure S1. (a, b) SEM images and (c) the corresponding EDS spectrum of Ni$_3$Fe LDH/NF.
Figure S2. (a, b) SEM images and (c) the corresponding EDS spectrum of Ni$_3$FeW$_{0.5}$ LDH/NF.
Figure S3. (a, b) SEM images and (c) the corresponding EDS spectrum of the Ni$_3$FeW LDH/NF.
Figure S4. (a, b) SEM images and (c) the corresponding EDS spectrum of $\text{Ni}_3\text{FeW}_2$ LDH/NF.
Figure S5. (a, b) SEM images and (c) the corresponding EDS spectrum of Ni$_3$FeW$_3$ LDH/NF.
Figure S6. AFM images and corresponding height profiles of (a, b) Ni$_3$Fe LDH and (c, d) Ni$_3$FeW LDH nanosheets.
Figure S7. (a) TEM image of two layers of Ni$_3$FeW 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$_3$FeW LDH nanosheets.
Figure S9. TEM EDX spectrum of Ni$_3$FeW LDH nanosheets.
Figure S10. XRD patterns of Ni$_3$Fe, Ni$_3$FeW, and Ni$_3$W 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$_3$Fe, (c,d) Ni$_3$FeW, and (e,f) Ni$_3$W LDH/NF.

After introducing W, the thickness of Ni$_3$FeW LDH nanosheets decreases. Ni$_3$W LDH nanosheets are more porous and thinner than either Ni$_3$Fe or Ni$_3$FeW LDH. It can be concluded that either Fe or W can affect the nanosheet structure of the final LDH, especially W.
In terms of OER performance, Ni$_3$FeW LDH is the best, Ni$_3$Fe LDH is the second best, and Ni$_3$W 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$_3$Fe and Ni$_3$FeW 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. 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. 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$_3$Fe LDH/NF, (b) Ni$_3$FeW$_{0.5}$ LDH/NF, (c) Ni$_3$FeW LDH/NF, (d) Ni$_3$FeW$_2$ LDH/NF, and (e) Ni$_3$FeW$_3$ 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$_3$FeW LDH/NF after 5000 CV scans.

Reference: