## Addressable surface engineering for N doped WS<sub>2</sub> nanosheet arrays with abundant active sites and optimal local electronic structure for enhanced hydrogen evolution reaction

Haiqing Wang,<sup>\*, #, a</sup> Zhongfei Xu,<sup>#, b</sup> Zengfu Zhang,<sup>a</sup> Shuxian Hu,<sup>\*, b</sup> Mingjun Ma,<sup>a</sup> Zhicheng Zhang,<sup>\*,c</sup> Weijia Zhou,<sup>a</sup> Hong Liu,<sup>\*,a,d</sup>

 <sup>a</sup> Collaborative Innovation Center of Technology and Equipment for Biological Diagnosis and Therapy in Universities of Shandong, Institute for Advanced Interdisciplinary Research (iAIR), University of Jinan, Jinan 250022, China
<sup>b</sup> Beijing Computational Science Research Center, Beijing 100193, China

<sup>c</sup> Tianjin Key Laboratory of Molecular Optoelectronic Sciences, Department of Chemistry, School of Science, Tianjin University, Tianjin 300072, China.

<sup>d</sup> State Key Laboratory of Crystal Materials, Shandong University, Jinan 250100, PR

China.

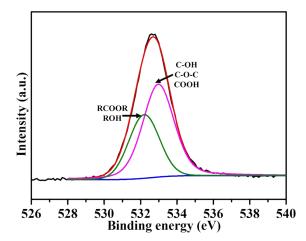


Figure S1 High resolution O 1s of CC-60 sample.

The results is indicative of -O functional groups in hydroxyls, ethers, lactones, and/or carboxyls.

(J. Sanchez, T. R. Hellstern, L. A. King and T. F. Jaramillo, Adv. Energy Mater., 2019, 9,

1901824.)

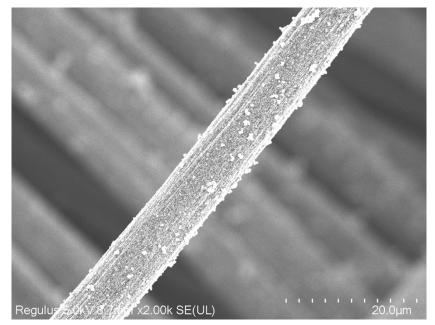


Figure S2. SEM image of WS<sub>2</sub>-CC-60 with low magnification.

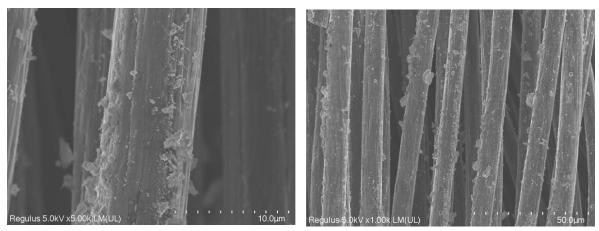


Figure S3. SEM images of  $WS_2@$ mixed acid (HNO<sub>3</sub> and  $H_2SO_4$ ) treated CC.

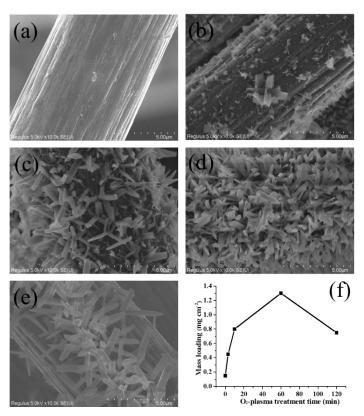


Figure S4. SEM images of Co-MOF-0/3/10/60/120 from a to e.

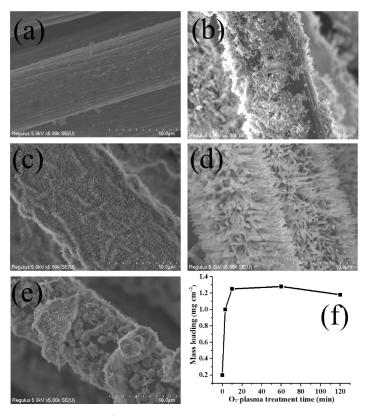


Figure S5. SEM images of NiCoOH/O -0/3/10/60/120 from a to e.

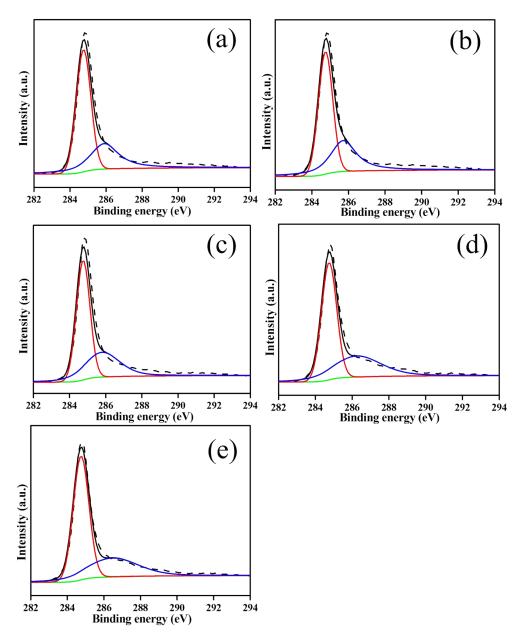


Figure S6. XPS spectra of C 1s peaks of CC-0/3/10/60/120 from a to e, where blue line represents C sp<sup>3</sup> and red line represents C sp<sup>2</sup>.

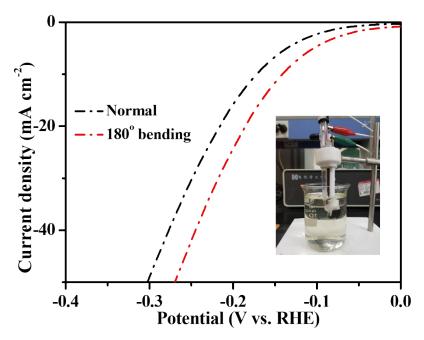


Figure S7. HER performance of N-WS2 under different bending conditions of 180°

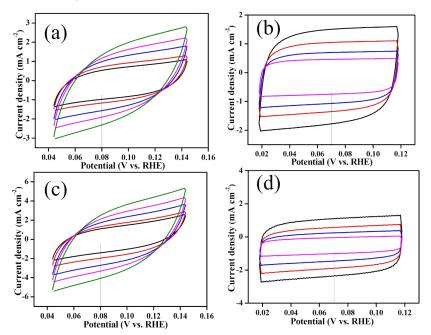


Figure S8 Non-Faradaic scan for double-layer capacitance. (a and c) and (b and d) cyclic voltammetry for acid and alkaline HER, respectively, for measuring  $C_{dl}$  of WS<sub>2</sub> and N-WS<sub>2</sub>.

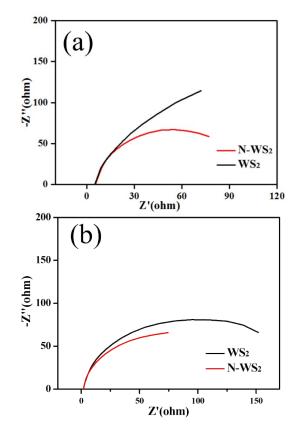


Figure S9 EIS Nyquist plots (a) and (b) of  $WS_2$  and  $N-WS_2$  in alkaline and acid conditions, respectively.

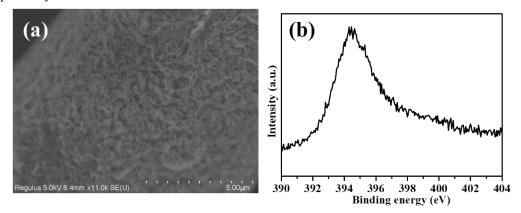


Figure S10 SEM image (a) and high resolution N 1s (b) of N-WS $_2$  after stability test.