

## Electronic Supplementary Information

### Interface Modifications for RuO<sub>2</sub>-decorated MoS<sub>2</sub> Nanosheets as Excellent Electrocatalyst toward Alkaline Hydrogen Evolution

Changjiang Zuo<sup>a</sup>, Jiehua Bao<sup>b</sup>, Xiwang Zhao<sup>a</sup>, Chunfeng Mao<sup>a</sup>, Bo Wu<sup>c</sup>, Yanyun Wang<sup>a</sup>, Yiwei Zhang<sup>a</sup>, Zewu Zhang<sup>b\*</sup>, Yuming Zhou<sup>a\*</sup>

<sup>a</sup>Jiangsu Optoelectronic Functional Materials and Engineering Laboratory, School of Chemistry and Chemical Engineering, Southeast University, Nanjing 211189, P. R. China.

<sup>b</sup>School of Materials Engineering, Nanjing Institute of Technology, Nanjing 211167, P. R. China

<sup>c</sup>Multiscale Computational Materials Facility, Key Laboratory of Eco-Materials Advanced Technology, College of Materials Science and Engineering, Fuzhou University, Fuzhou 350100, P. R. China.

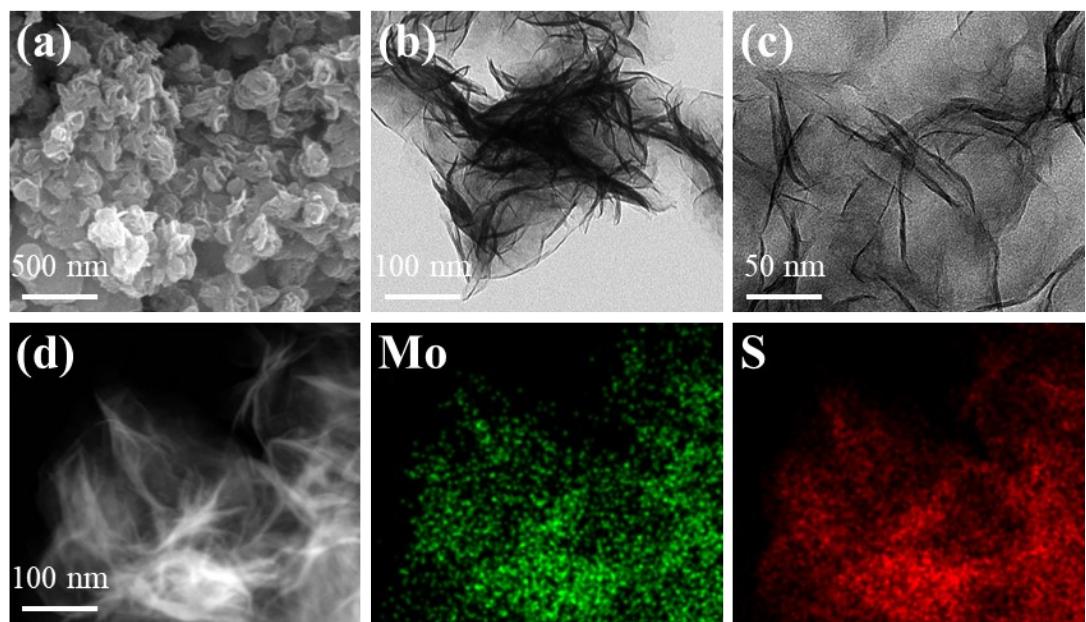
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\* Corresponding authors:

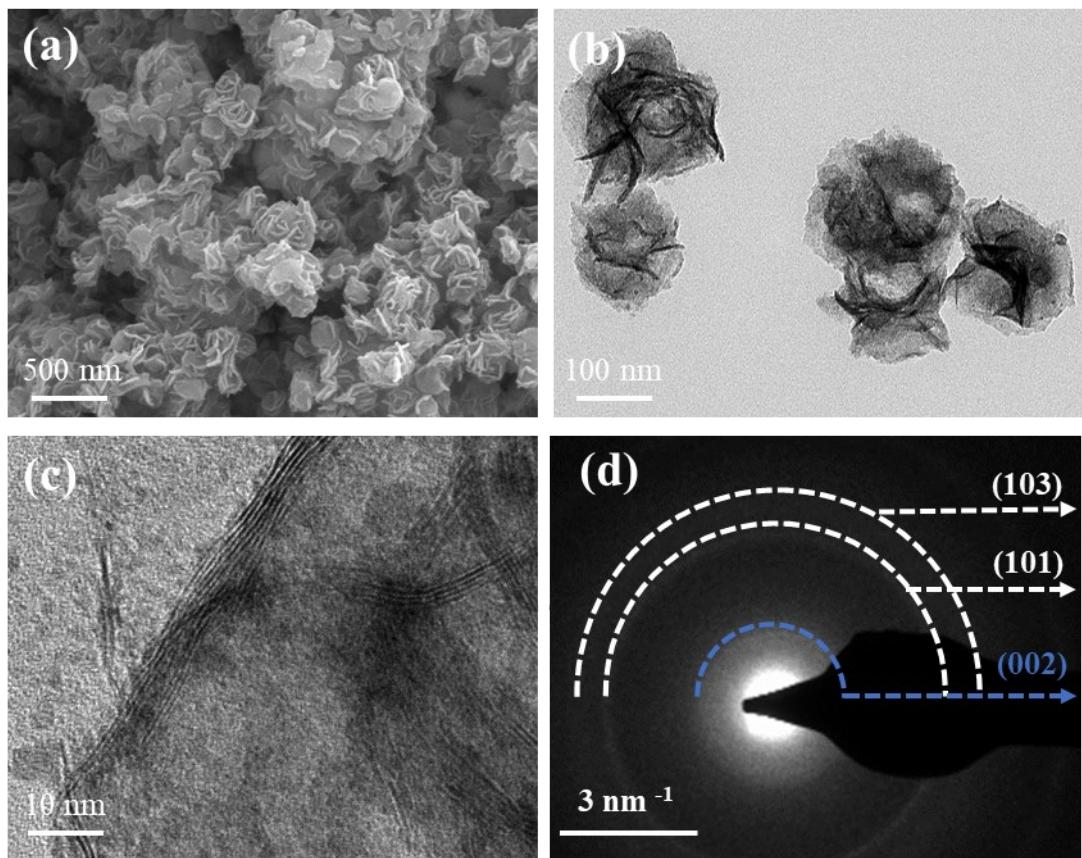
E-mail: [ymzhou@seu.edu.cn](mailto:ymzhou@seu.edu.cn) (Y, Zhou)

[seuzzw06@163.com](mailto:seuzzw06@163.com) (Z, Zhang)

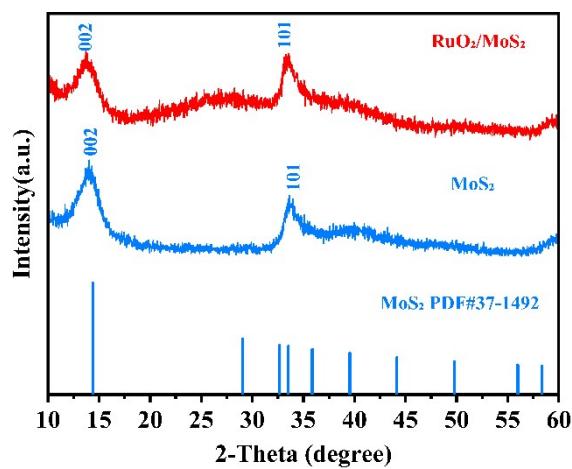
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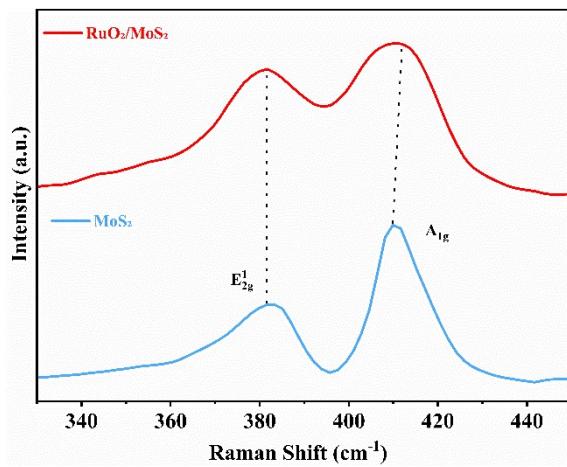
**Fig. S1.** (a) SEM image of MoS<sub>2</sub>. (b-c) TEM images of MoS<sub>2</sub> with different amplifications. (d) TEM-EDS elemental mapping of Mo, S in MoS<sub>2</sub>.



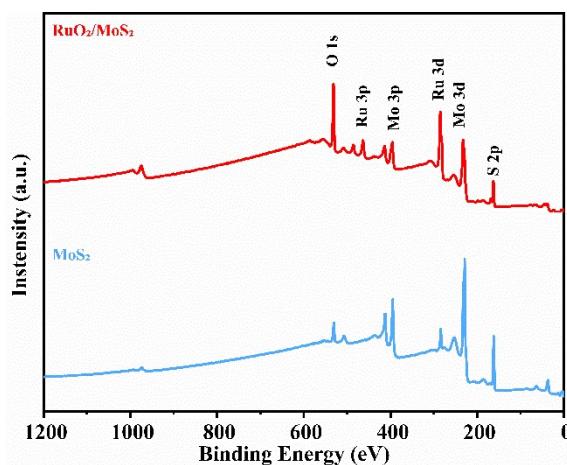
**Fig. S2.** (a) SEM image of RuO<sub>2</sub>/MoS<sub>2</sub> catalyst. (b) TEM image of RuO<sub>2</sub>/MoS<sub>2</sub> catalyst. (c) HRTEM image of RuO<sub>2</sub>/MoS<sub>2</sub> catalyst. (d) SAED pattern of RuO<sub>2</sub>/MoS<sub>2</sub> catalyst.



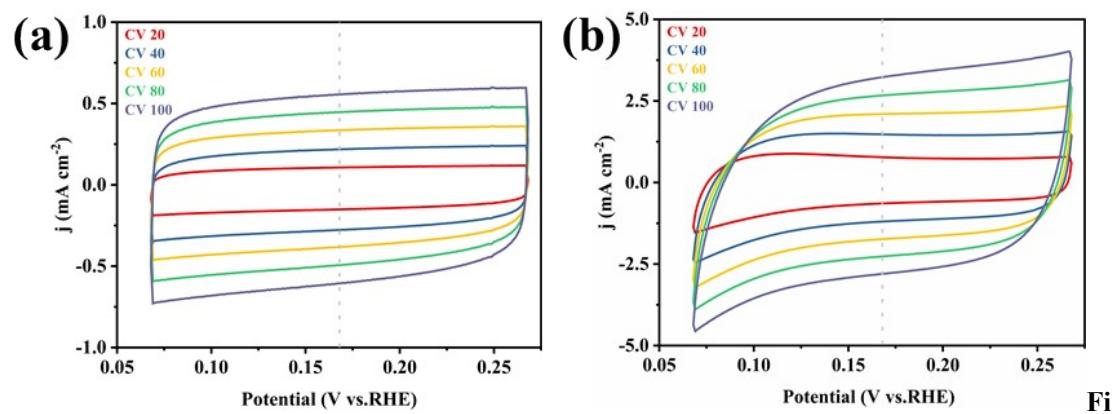
**Fig. S3.** The XRD patterns of RuO<sub>2</sub>/MoS<sub>2</sub> catalyst and MoS<sub>2</sub>.



**Fig. S4.** Raman spectra of RuO<sub>2</sub>/MoS<sub>2</sub> catalyst and MoS<sub>2</sub>.

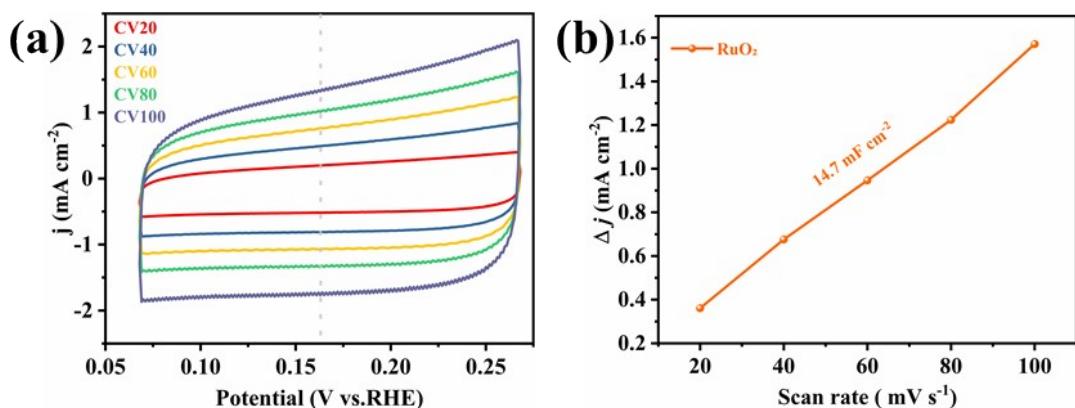


**Fig. S5.** XPS survey spectra of RuO<sub>2</sub>/MoS<sub>2</sub> catalyst and MoS<sub>2</sub>.

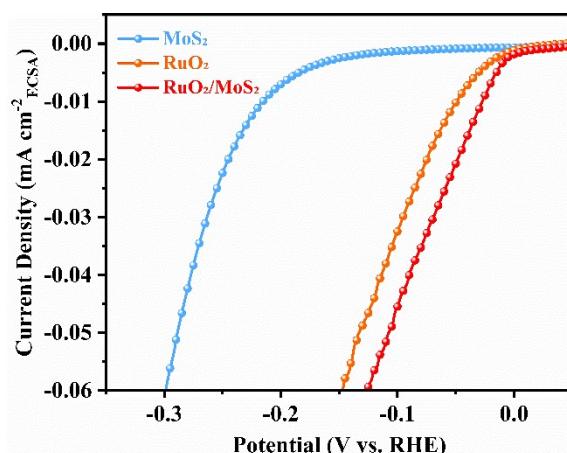


**Fig. S6.** CV curves of (a) MoS<sub>2</sub>, (b) RuO<sub>2</sub>/MoS<sub>2</sub> catalyst. at scan rates of 20, 40, 60, 80, 100 mV s<sup>-1</sup>

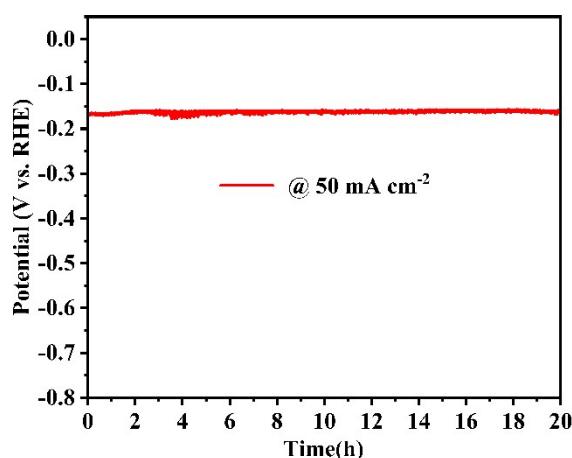
in 1.0 M KOH, respectively.



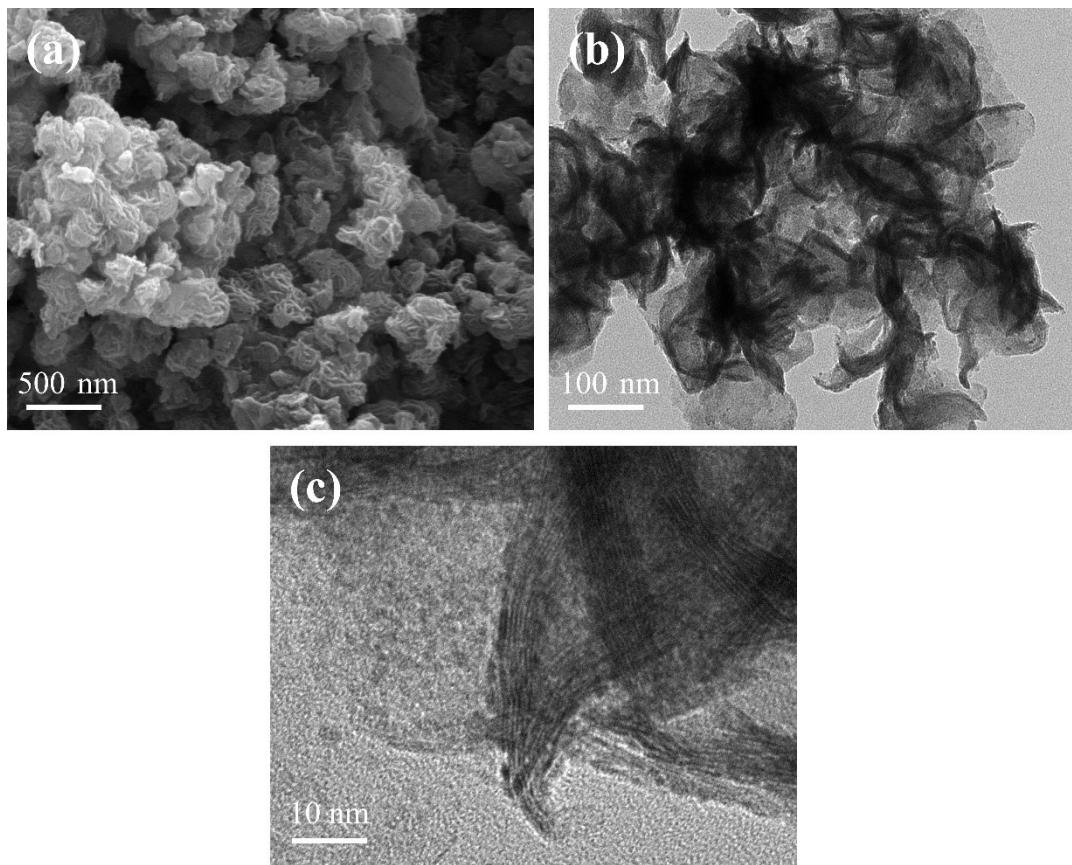
**Fig. S7.** (a) CV curves of RuO<sub>2</sub> catalyst at scan rates of 20, 40, 60, 80, 100 mV s<sup>-1</sup> in 1.0 M KOH, (b) calculated electrochemical double-layer capacitance for RuO<sub>2</sub>.



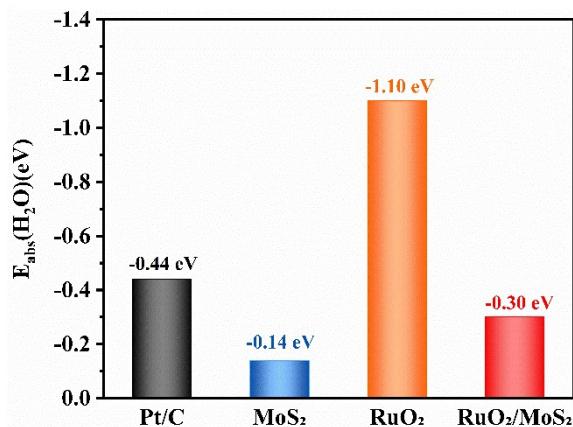
**Fig. S8.** Polarization curves of MoS<sub>2</sub>, RuO<sub>2</sub> and RuO<sub>2</sub>/MoS<sub>2</sub> catalyst normalized ECSAs.



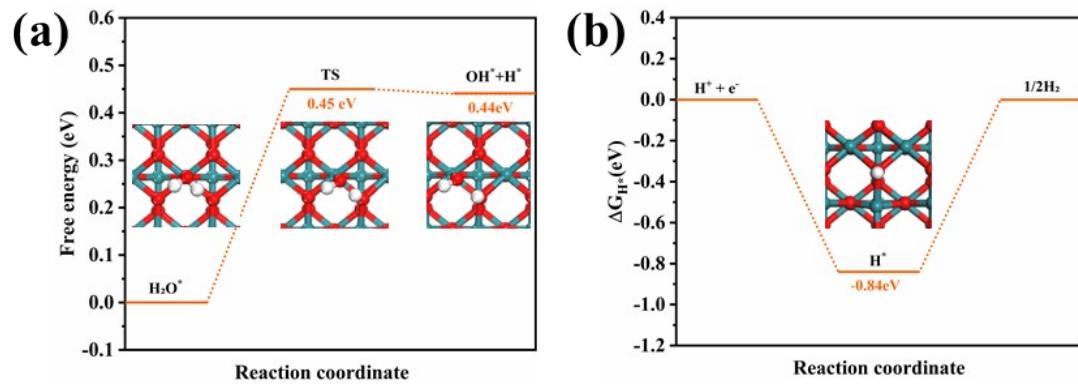
**Fig. S9.** The long-time chronopotentiometry curve of RuO<sub>2</sub>/MoS<sub>2</sub> catalyst at 50 mA cm<sup>-2</sup>.



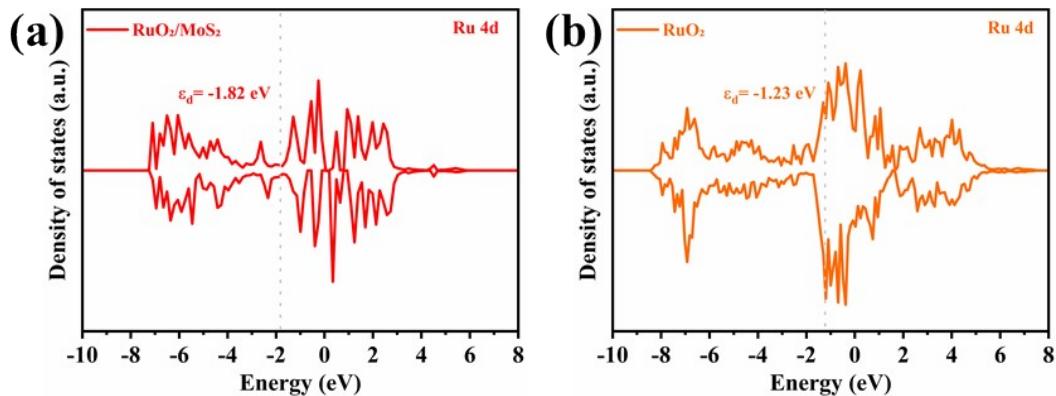
**Fig. S10.** (a) SEM image of RuO<sub>2</sub>/MoS<sub>2</sub> catalyst after long-time HER test. (b-c) TEM and HRTEM images of RuO<sub>2</sub>/MoS<sub>2</sub> catalyst.



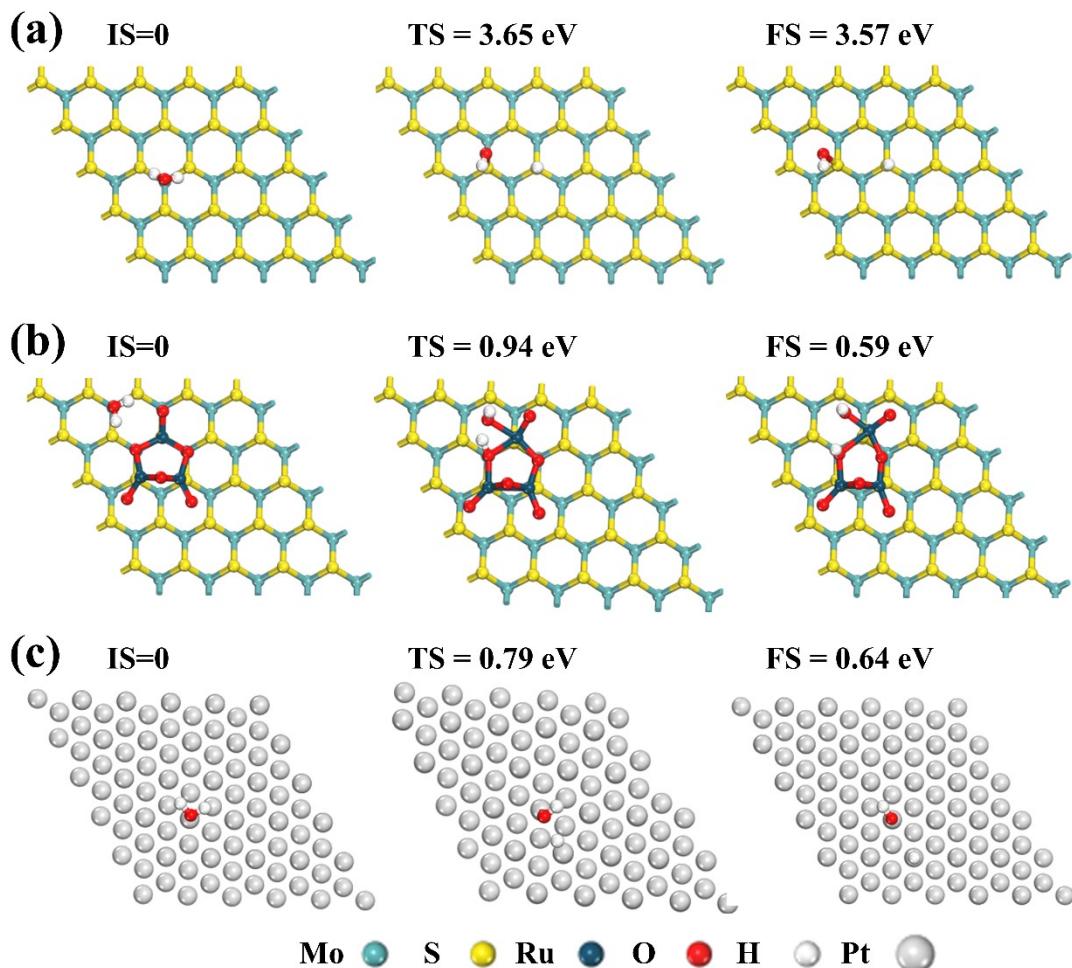
**Fig. S11.** The adsorption energies of H<sub>2</sub>O on Pt/C, MoS<sub>2</sub>, RuO<sub>2</sub> and RuO<sub>2</sub>/MoS<sub>2</sub> catalyst.



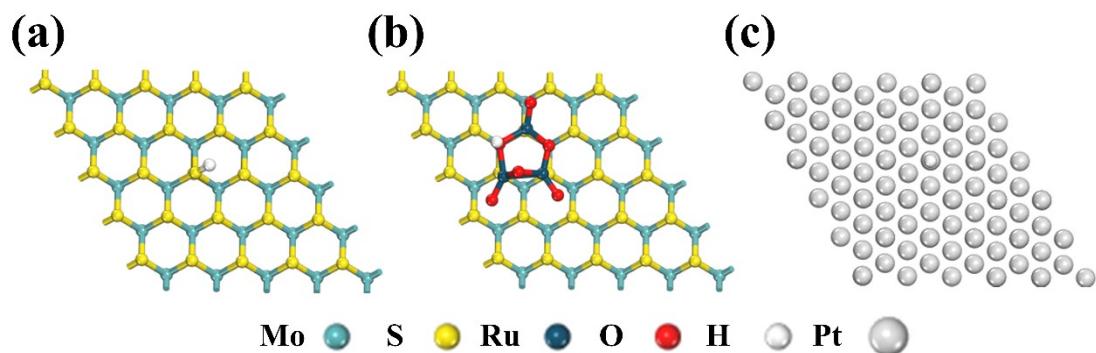
**Fig. S12.** (a) The free energy diagrams for  $\text{H}_2\text{O}$  dissociation on  $\text{RuO}_2$  (110). (b) The hydrogen adsorption free energy of  $\text{RuO}_2$  (110).



**Fig. S13.** Projected density states of Ru 4d on (a)  $\text{RuO}_2/\text{MoS}_2$  catalyst and (b)  $\text{RuO}_2$  (110).



**Fig. S14.** Optimized adsorption configurations and free energy changes in the water dissociation pathways on (a) MoS<sub>2</sub>(002), (b) RuO<sub>2</sub>/MoS<sub>2</sub>, and (c) Pt(111).



**Fig. S15.** Optimized H atom adsorb on different DFT models. (a) MoS<sub>2</sub>(002), (b) RuO<sub>2</sub>/MoS<sub>2</sub>, and (c) Pt(111).

**Table S1.** Comparison of HER performance of electrocatalysts reported in different literatures in alkaline or acidic solution.

Samples	$\eta_{10}$ (mV)	Tafel (mV dec <sup>-1</sup> )	solution	Ref.
1T-2H Cr <sub>x</sub> -MoS <sub>2</sub> Ultrathin Nanosheets	200	41.6	alkaline	1
Al-MoS <sub>2</sub>	198	134	alkaline	2
1T-2H MoS <sub>2</sub>	320	65	alkaline	3
Co@MoS <sub>2</sub> -S <sub>V</sub>	36	33	alkaline	4
RuO <sub>2</sub> /F-graphene	49	31	alkaline	5
RuO <sub>2</sub> /N-C	49	44	alkaline	6
Ru-MoS <sub>2</sub>	98	65	alkaline	7
Ru-MoS <sub>2</sub> /CNT	50	62	alkaline	8
Ni <sub>2</sub> P/MoS <sub>2</sub>	149	69.5	alkaline	9
MoS <sub>2</sub> /Ni <sub>3</sub> S <sub>2</sub> heterostructures	110	83	alkaline	10
2D-MoS <sub>2</sub> /Co(OH) <sub>2</sub>	125	76	alkaline	11
Ni-1T-MoS <sub>2</sub>	112	48	alkaline	12
NiO@1T-MoS <sub>2</sub>	46	52	alkaline	13
NiS microsphere	96	52	acidic	14
2H-TaS <sub>2</sub>	145	121	acidic	15
RuSi	42	23.6	acidic	16
Edge-rich 1T-MoS <sub>2</sub> /Ni(OH) <sub>2</sub>	57	30	alkaline	17
<b>RuO<sub>2</sub>/MoS<sub>2</sub></b>	<b>36</b>	<b>34.8</b>	<b>alkaline</b>	<b>This work</b>

## References

1. J. Jian, H. Li, X. Sun, D. Kong, X. Zhang, L. Zhang, H. Yuan and S. Feng, *ACS Sustain. Chem. Eng.*, 2019, **7**, 7227-7232.
2. J. Jian, Y. Li, H. Bi, X. Wang, X. Wu and W. Qin, *ACS Sustain. Chem. Eng.*, 2020, **8**, 4547-4554.
3. S. Wang, D. Zhang, B. Li, C. Zhang, Z. Du, H. Yin, X. Bi and S. Yang, *Adv. Energy Mater.*, 2018, **8**, 1801345.
4. P. A. Koudakan, C. Wei, A. Mosallanezhad, B. Liu, Y. Fang, X. Hao, Y. Qian and G. Wang, *Small*, 2022, **18**, 2107974.
5. Z. Fan, F. Liao, H. Shi, Y. Liu, M. Shao and Z. Kang, *Inorg. Chem. Front.*, 2020, **7**, 2188-2194.
6. C. Z. Yuan, Y. F. Jiang, Z. W. Zhao, S. J. Zhao, X. Zhou, T. Y. Cheang and A.W. Xu, *ACS Sustain. Chem. Eng.*, 2018, **6**, 11529-11535
7. S. Geng, F. Tian, M. Li, Y. Liu, J. Sheng, W. Yang, Y. Yu and Y. Hou, *Nano Res.*, 2022, **15**, 1809-1816
8. X. Zhang, F. Zhou, S. Zhang, Y. Liang and R. Wang, *Adv. Sci.*, 2019, **6**, 1900090.
9. M. Kim, M. A. R. Anjum, M. Lee, B. J. Lee and J. S. Lee, *Adv. Funct. Mater.*, 2019, **29**, 1809151.
10. J. Zhang, T. Wang, D. Pohl, B. Rellinghaus, R. Dong, S. Liu, X. Zhuang and X. Feng, *Angew. Chem. Int. Ed.*, 2016, **55**, 6702-6707.
11. Z. Zhu, H. Yin, C. T. He, M. Al Mamun, P. Liu, L. Jiang, Y. Zhao, Y. Wang, H. G. Yang, Z. Tang, D. Wang, X. M. Chen and H. Zhao, *Adv. Mater.*, 2018, **30**, 1801171.
12. G. Wang, G. Zhang, X. Ke, X. Chen, X. Chen, Y. Wang, G. Huang, J. Dong, S. Chu and M. Sui, *Small*, 2022, **18**, 2107238.
13. Y. Huang, Y. Sun, X. Zheng, T. Aoki, B. Pattengale, J. Huang, X. He, W. Bian, S. Younan, N. Williams, J. Hu, J. Ge, N. Pu, X. Yan, X. Pan, L. Zhang, Y. Wei and J. Gu, *Nat. Commun.*, 2019, **10**, 982.
14. A. Wang, H. Li, J. Xiao, Y. Lu, M. Zhang, K. Hu and K. Yan, *ACS Sustain. Chem. Eng.*, 2018, **6**, 15995-16000.
15. M. Zhang, Y. He, D. Yan, H. Xu, A. Wang, Z. Chen, S. Wang, H. Luo and K. Yan, *Nanoscale*, 2019, **11**, 22255-22260.
16. Y. He, T. L. Wang, M. Zhang, T. W. Wang, L. F. Wu, L. Zeng, X. Wang, M. Boubeche, S. Wang, K. Yan, S. H. Lin and H. Luo, *Small*, 2021, **17**, 2006153
17. W. Chen, J. Gu, Y. Du, F. Song, F. Bu, J. Li, Y. Yuan, R. Luo, Q. Liu and D. Zhang, *Adv. Funct. Mater.*, 2020, **30**, 2000551.