

## Supporting Information

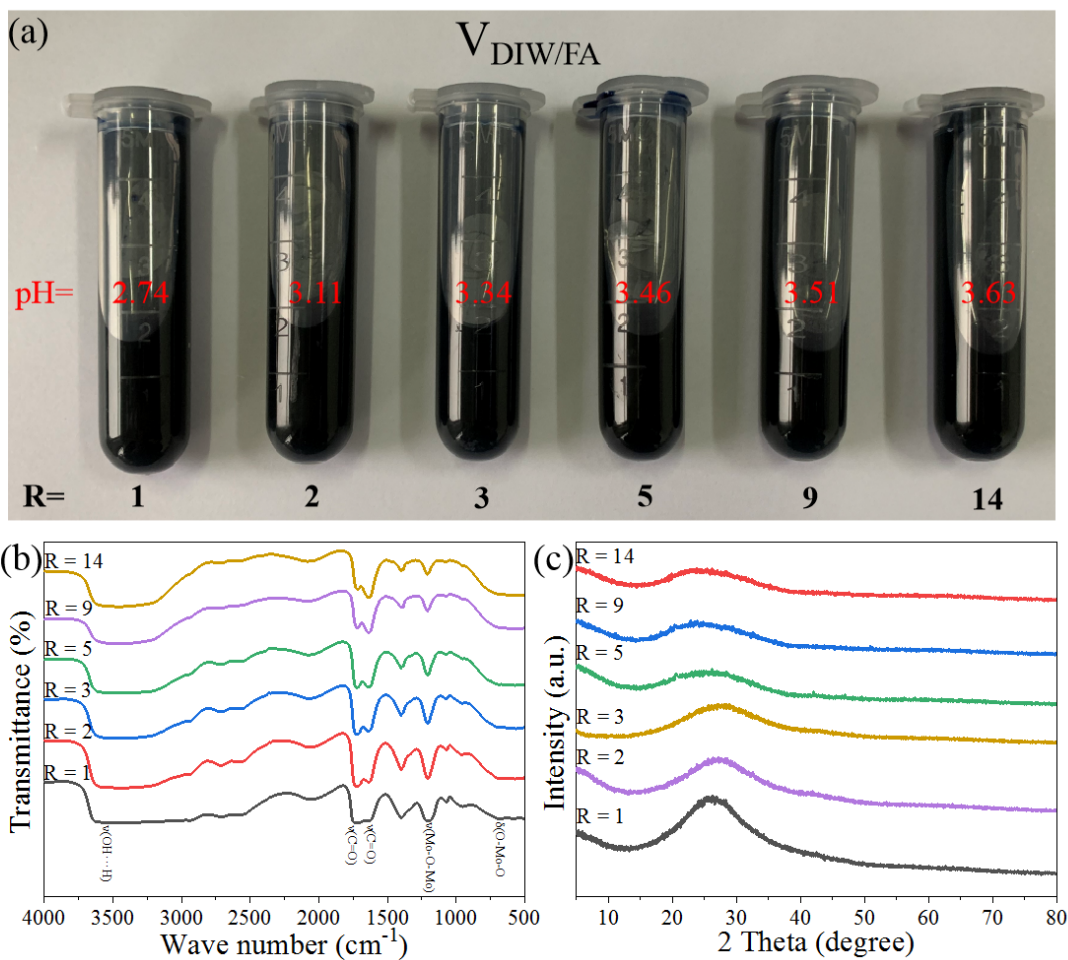
### **Two-dimensional metal phase layered molybdenum disulfide for electrocatalytic hydrogen evolution reaction**

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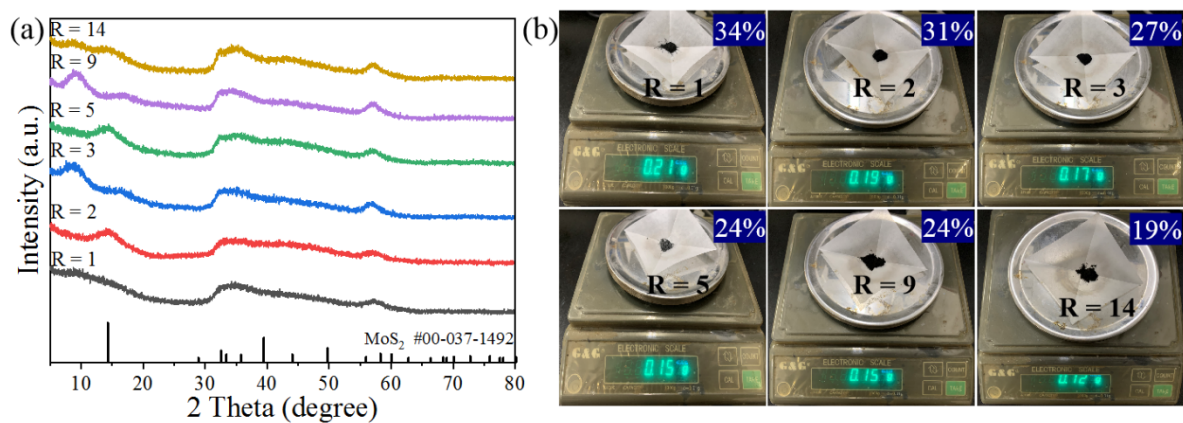
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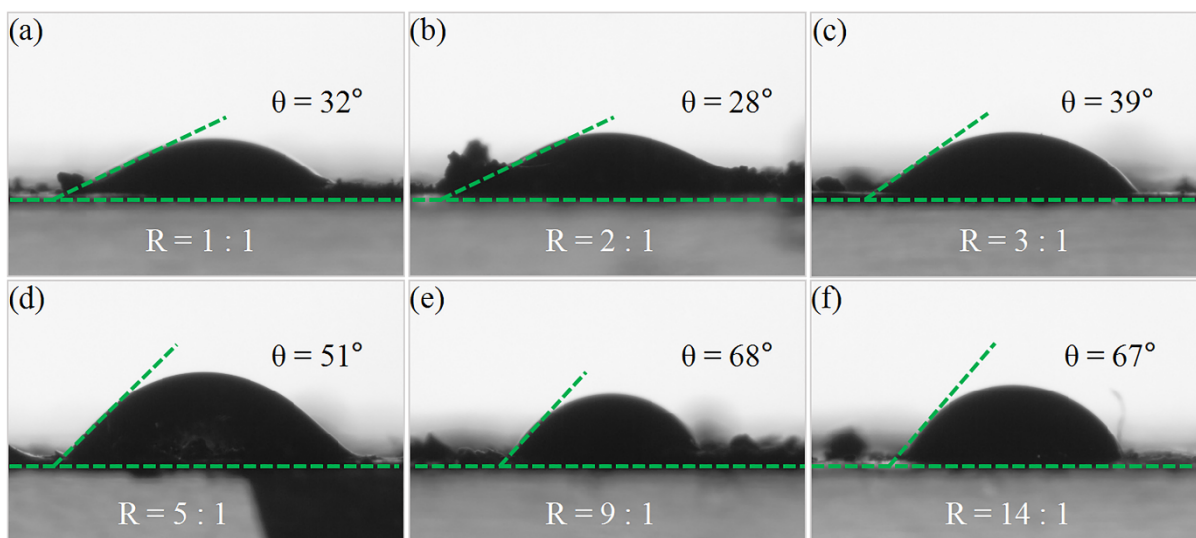
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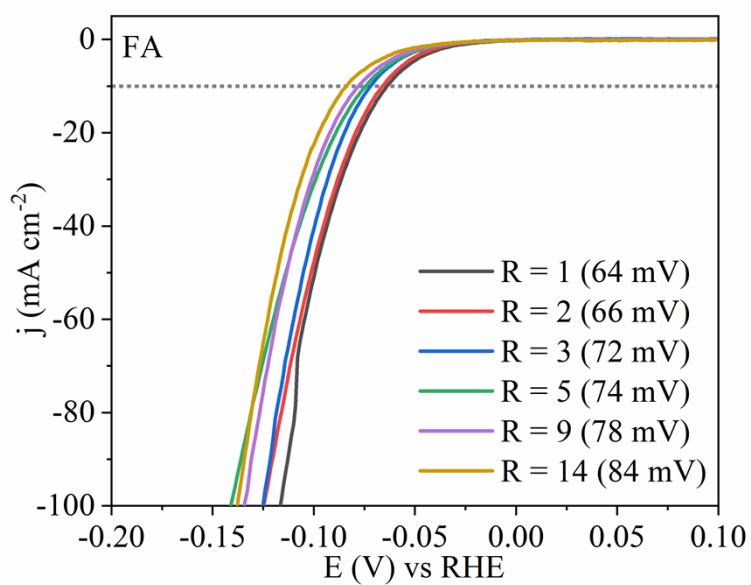
**Fig. S1.** Photos of MBS precursor solutions (a), FT-IR pattern (b) and XRD pattern (c) after 6 h at room temperature (25 °C) with different  $V_{\text{DIW/FA}}$  ( $R = 1, 2, 3, 5, 9$  and  $14$ ).



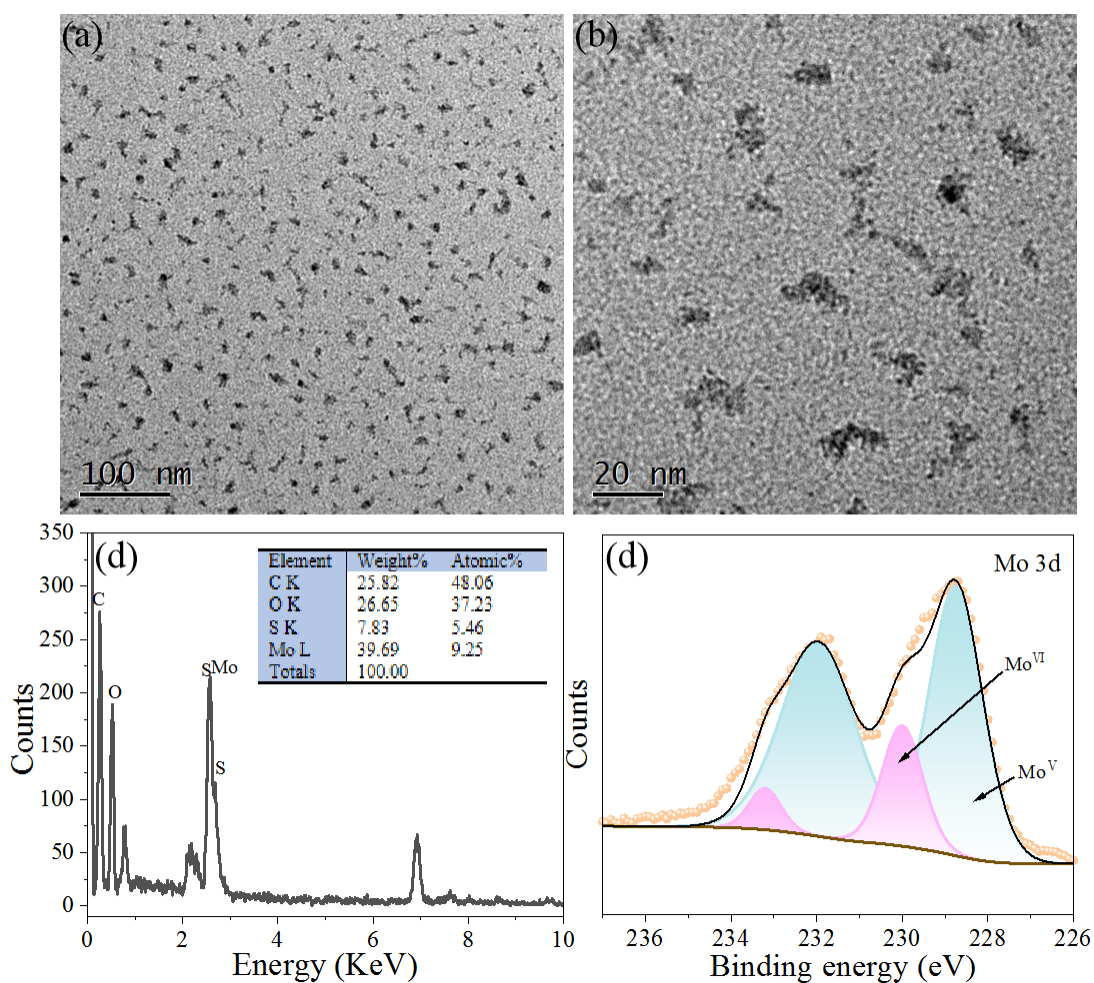
**Fig. S2.** XRD pattern (a) and yield (b) of products with different  $V_{\text{DIW/FA}}$  ( $R = 1, 2, 3, 5, 9$  and  $14$ ).



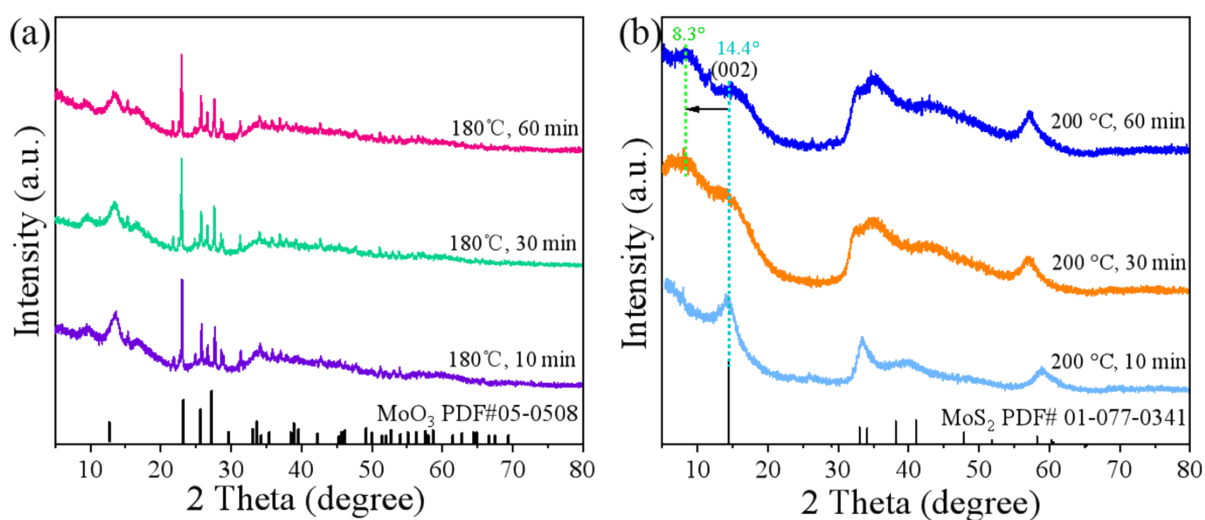
**Fig. S3.** Contact angle of products with different  $V_{DIW/FA}$  ( $R = 1, 2, 3, 5, 9$  and  $14$ ).



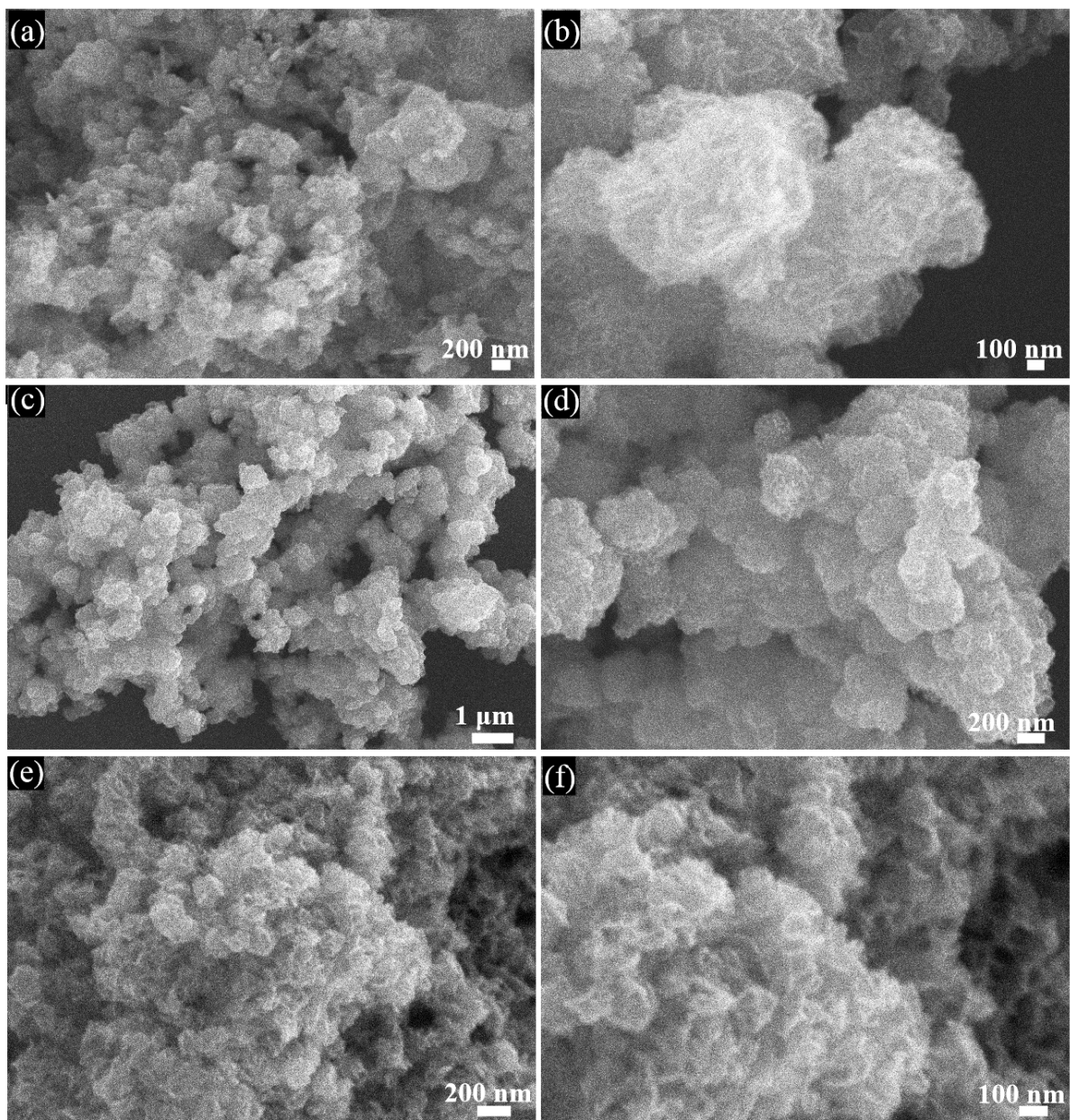
**Fig. S4.** LSV curves of MW-MoS<sub>2</sub> prepared with different  $V_{DIW/FA}$  ( $R = 1, 2, 3, 5, 9$  and  $14$ ).



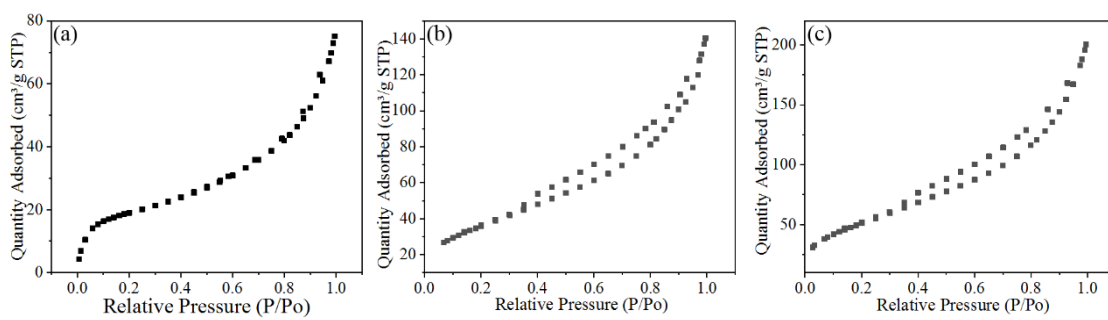
**Fig. S5.** TEM images (a-b), EDX pattern (c) and XPS spectra of Mo 3d (d) of MBS.



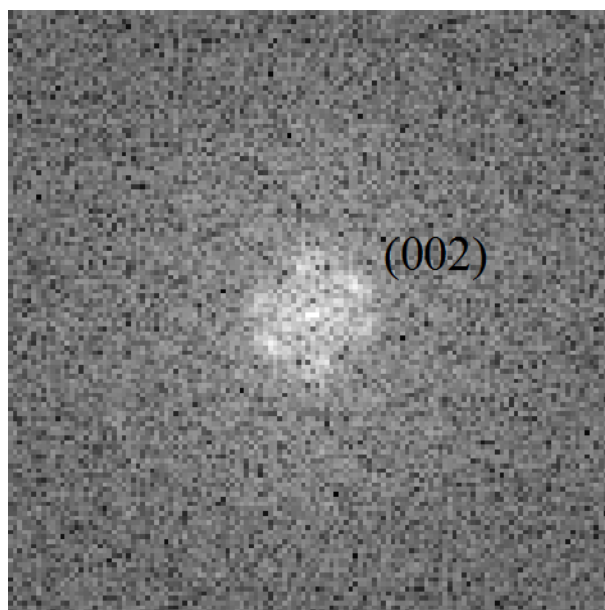
**Fig. S6.** XRD pattern of the corresponding products prepared at different reaction times and reaction temperatures.



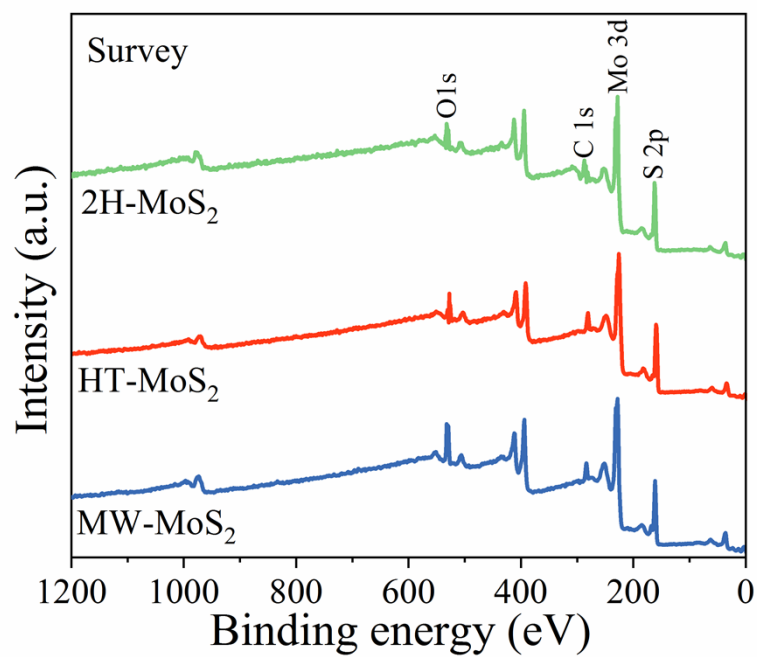
**Fig. S7.** SEM images of 2H-MoS<sub>2</sub> (a-b), HT-MoS<sub>2</sub> (c-d) and MW-MoS<sub>2</sub> (e-f).



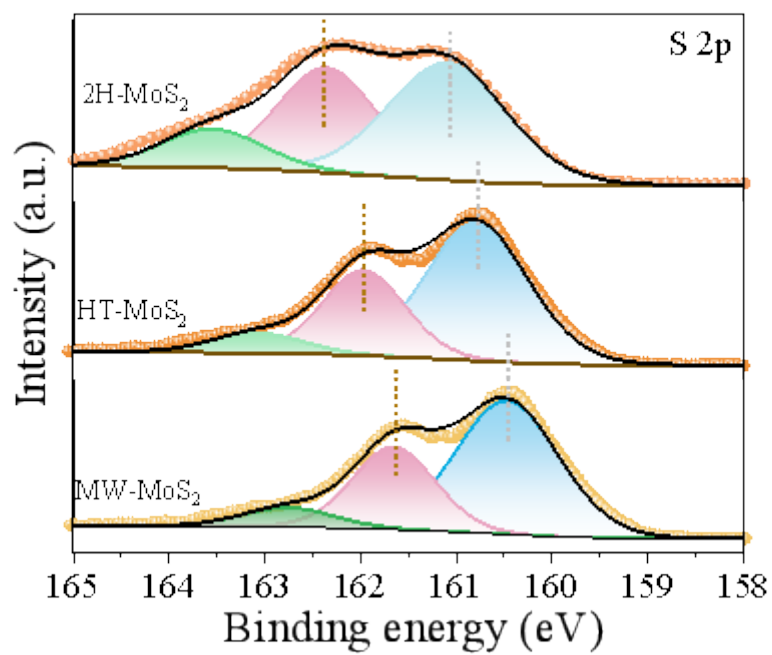
**Fig. S8.** S<sub>BET</sub> of 2H-MoS<sub>2</sub> (a), HT-MoS<sub>2</sub> (b) and MW-MoS<sub>2</sub> (c).



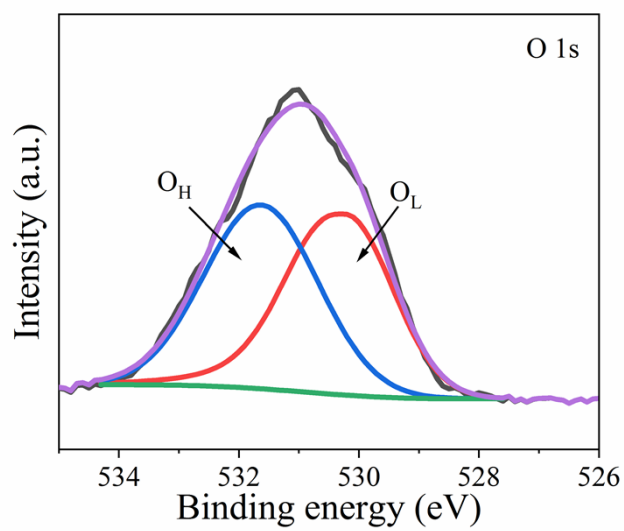
**Fig. S9.** SAED pattern of MW-MoS<sub>2</sub>.



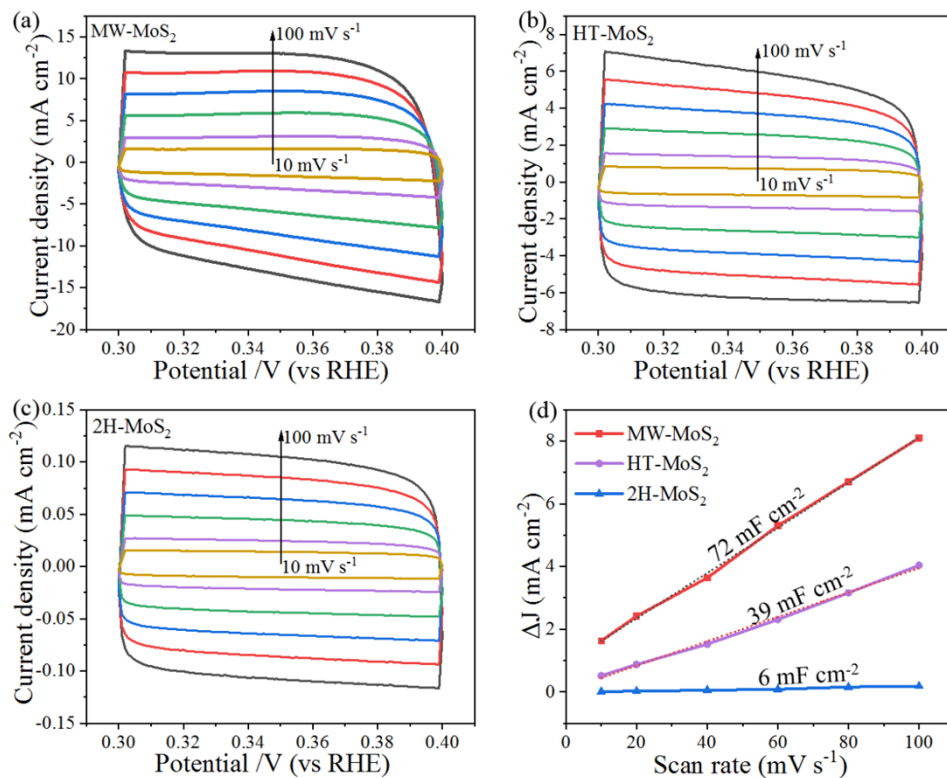
**Fig. S10.** Survey XPS patterns of 2H-MoS<sub>2</sub>, HT-MoS<sub>2</sub>, and MW-MoS<sub>2</sub>.



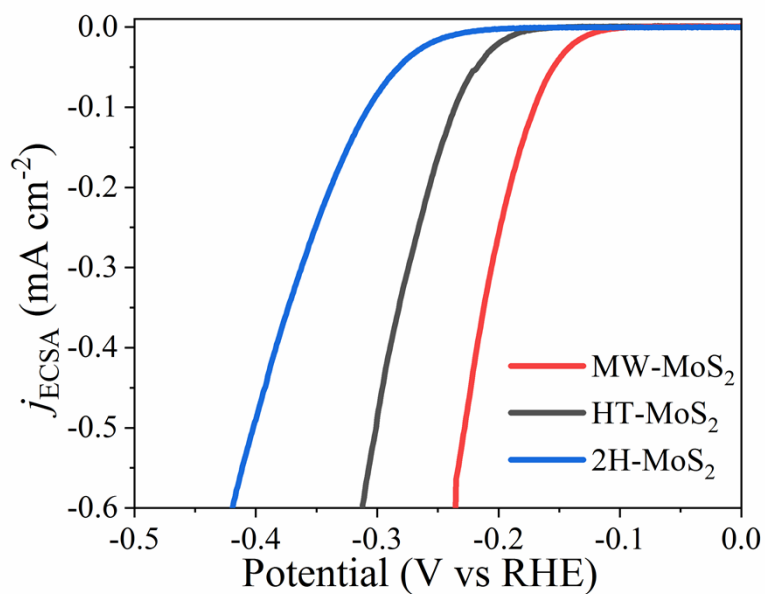
**Fig. S11.** XPS spectra of S 2p of 2H-MoS<sub>2</sub>, HT-MoS<sub>2</sub> and MW-MoS<sub>2</sub>.



**Fig. S12.** XPS spectra of O 1s of MW-MoS<sub>2</sub>.

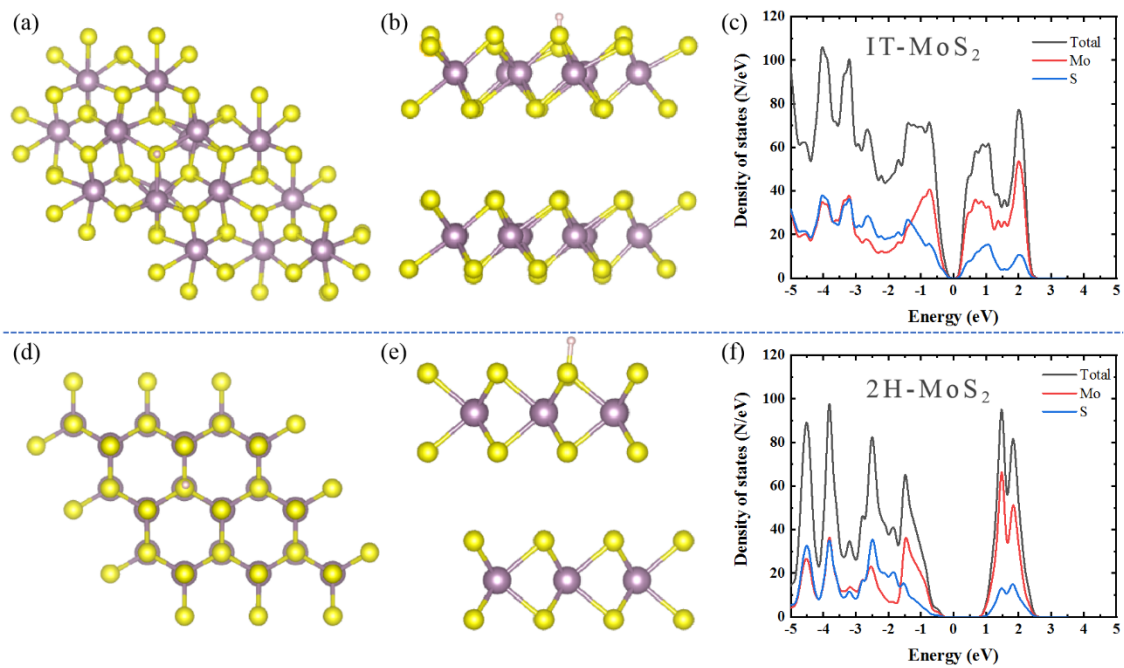


**Fig. S13.** CV plots of non-Faraday regions for MW-MoS<sub>2</sub> (a), HT-MoS<sub>2</sub> (b) and 2H-MoS<sub>2</sub> (c); the corresponding  $C_{dl}$  values were obtained at 0.35 V (vs RHE) and at different scan rates for the current density ( $\Delta j$ ) (d).



**Fig. S14.** HER polarization curves of MW-MoS<sub>2</sub>, HT-MoS<sub>2</sub> and 2H-MoS<sub>2</sub> catalysts normalized by the ECSA.





**Fig. S15.** Hydrogen absorption site model of 1T-MoS<sub>2</sub>: a: Top, b: Side, c: DOS; Hydrogen absorption site model of 2H-MoS<sub>2</sub>: d: Top, e: Side, f: DOS.

**Table S1.** Comparison of several existing methods for the preparation of 1T-MoS<sub>2</sub>.

Material	Preparation method	Reaction temperature (°C)	Reaction time (h)	Reference
MW-MoS <sub>2</sub>	microwave method	200	0.5	This work
1T-MoS <sub>2</sub> /CC	hydrothermal route	220	24	<i>Appl. Catal. B: Environ.</i> <b>2019</b> , <i>246</i> , 296-302.[1]
1T-MoS <sub>2</sub>	electrochemically intercalate	—	48	<i>Nature Nanotech.</i> <b>2015</b> , <i>10</i> , 313-318.[2]
M-MoS <sub>2</sub>	hydrothermal process	200	12	<i>Nat. Commun.</i> <b>2016</b> , <i>7</i> , 10672.[3]
Li <sub>x</sub> MoS <sub>2</sub>	chemically embedded and exfoliated	—	48	<i>Nano Lett.</i> <b>2011</b> , <i>11</i> , 5111-5116.[4]
1T-MoS <sub>2</sub>	chemically exfoliate	25	168	<i>Nat. Chem.</i> <b>2015</b> , <i>7</i> , 45-9.[5]
1T@2H MoS <sub>2</sub>	hydrothermal route	200/220	24	<i>Catal. Sci. Technol.</i> <b>2017</b> , <i>7</i> , 5635-5643.[6]
1T-MoS <sub>2</sub>	chemically embedded and exfoliated	—	49	<i>ACS Energy Lett.</i> <b>2018</b> , <i>3</i> , 7-13.[7]

**Table S2.** The content of elements by XPS.

Sample	Percent of Mo (%)	Percent of S (%)	Percent of O (%)
MW-MoS <sub>2</sub>	33.12	62.58	4.30
HT-MoS <sub>2</sub>	30.40	66.23	3.37
2H-MoS <sub>2</sub>	33.84	63.53	2.63

## Reference

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- [2] M. Acerce, D. Voiry, M. Chhowalla, *Nature Nanotech.* **10** (2015) 313.
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- [4] G. Eda, H. Yamaguchi, D. Voiry, T. Fujita, M. Chen, M. Chhowalla, *Nano Lett.* **11** (2011) 5111.

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- [6] Y. Liu, Y. Xie, L. Liu, J. Jiao, *Catal. Sci. Technol.* 7 (2017) 5635.
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