Supplementary Information (SI) for Chemical Science.
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Supplementary Information

Enhancing Hydrogen Evolution Reaction Activity Through Defects and Strain Engineering in Monolayer MoS_2

Renjith Nadarajan, Sraboni Dey, Arijit Kayal, Joy Mitra, Manikoth M. Shaijumon*

School of Physics, Indian Institute of Science Education and Research Thiruvananthapuram, Maruthamala PO, Thiruvananthapuram, Kerala 695551, India.

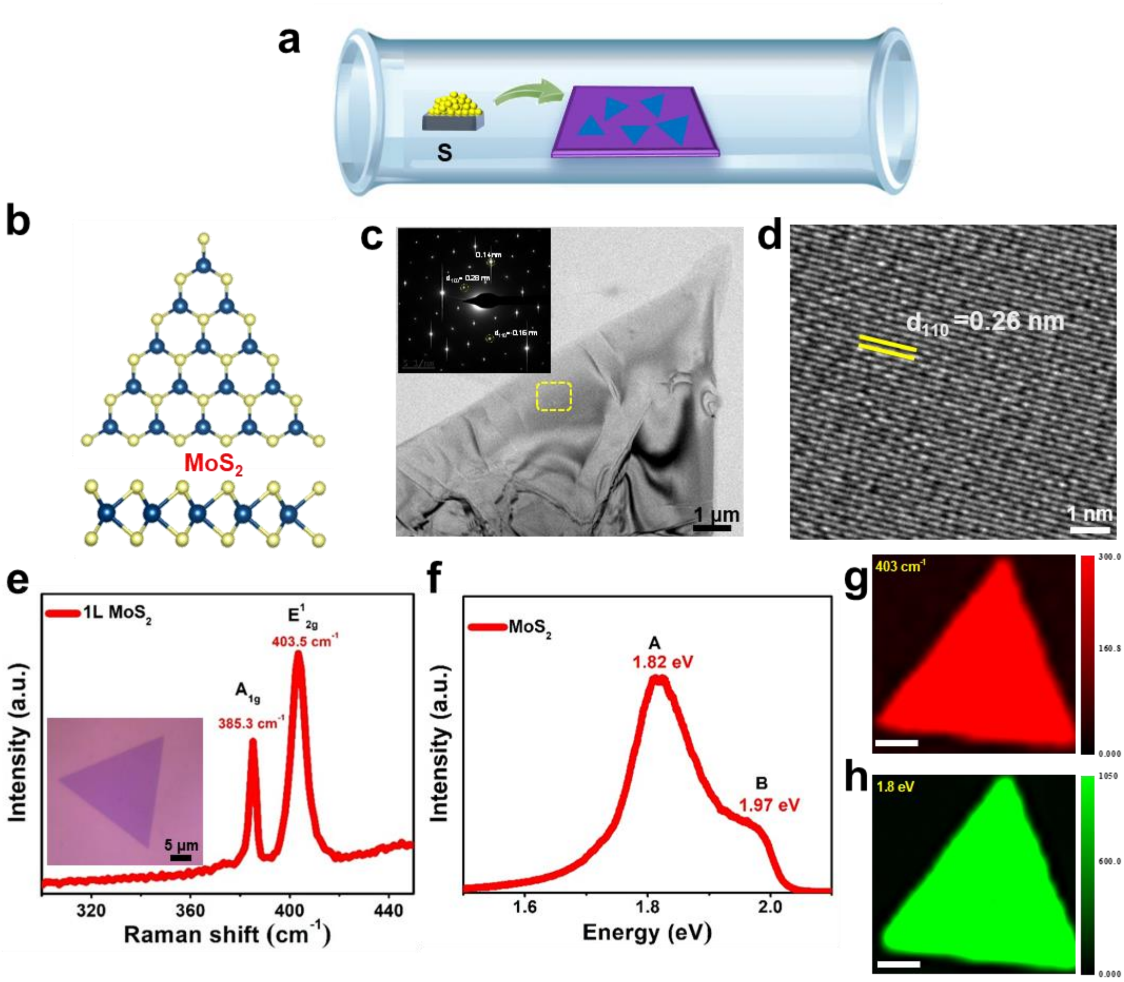

Fig. S1 (a) Schematic illustration of the CVD growth of MoS_2 (b) Schematic crystal configuration of monolayer MoS_2 (c) TEM and (d) HRTEM images of MoS_2 inset of (c) shows the SAED pattern (e) Raman and (f) PL spectra of MoS_2 monolayer, inset of S1e shows the optical image (g) Raman map image of A_{1g} mode (403 cm^{-1}) and (h) PL map image (A exciton) at 1.82 eV of MoS_2 monolayer (scale bar shows 5 µm)

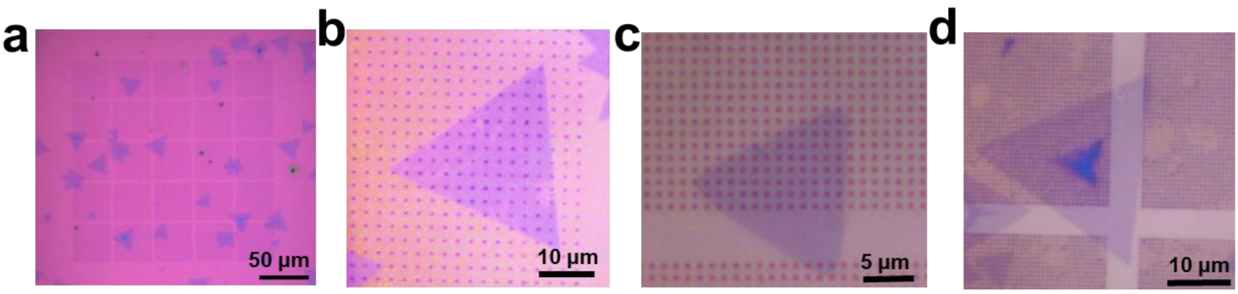

Fig. S2 Optical images of (a, b) $S_{2\mu m}$-MoS_2 (c) $S_{1\mu m}$-MoS_2 (d) $S_{0.5\mu m}$-MoS_2

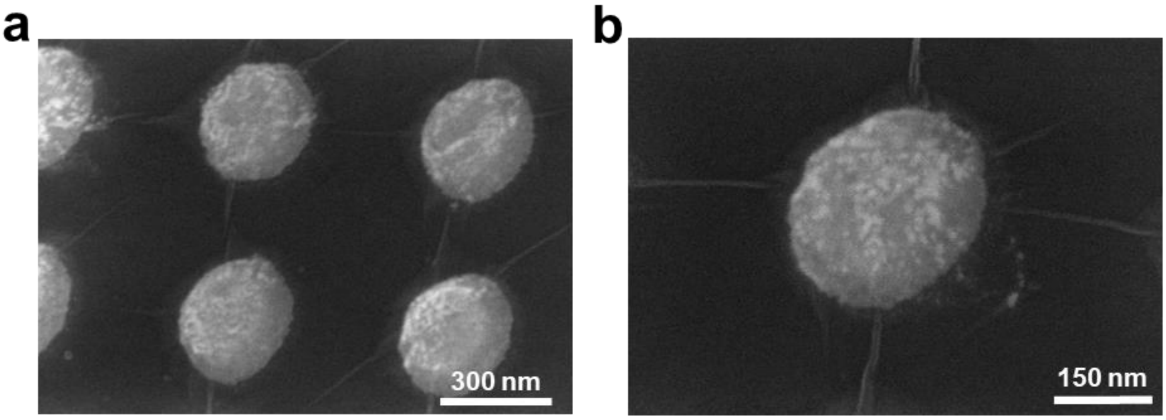

Fig. S3 (a, b) SEM image of $S_{0.5\mu m}$-MoS_2

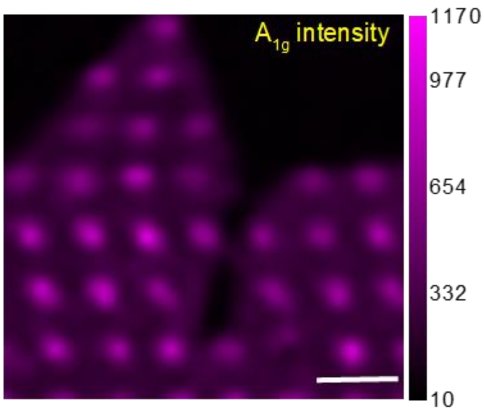

Fig. S4 Raman A_{1g} intensity map image (scale bar 2 µm)

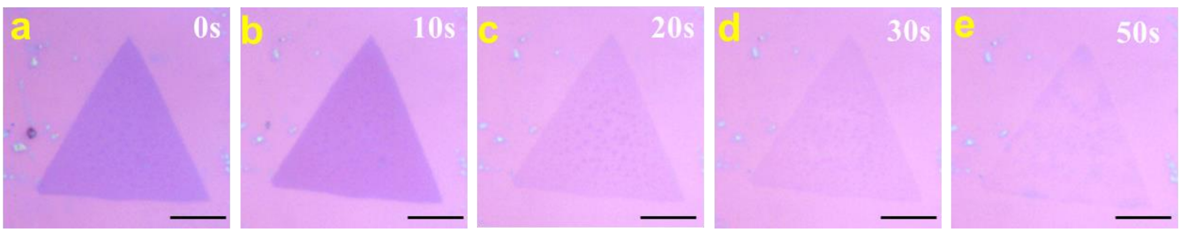

Fig. S5 (a-e) Optical images of MoS_2 monolayer flakes under plasma treatment (scale bar 10 µm)

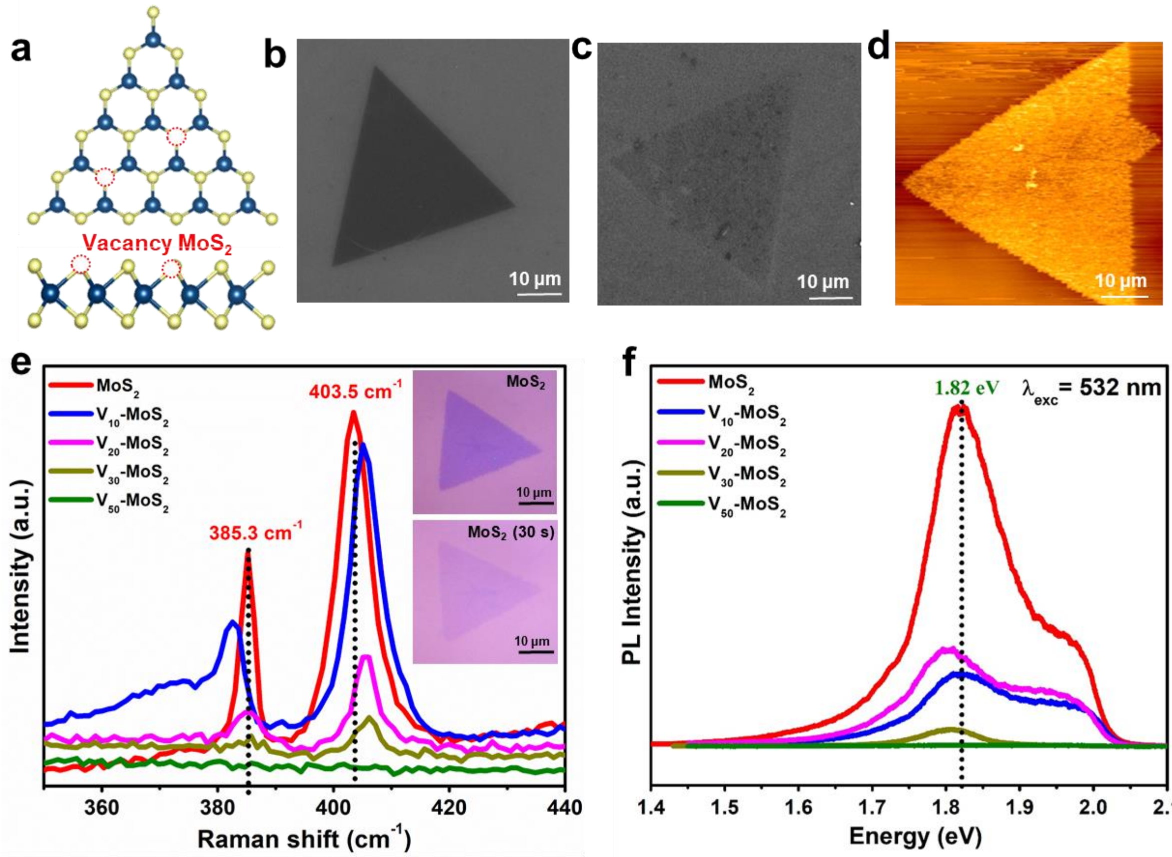

Fig. S6 (a) Schematic crystal configuration of vacancy MoS_2. SEM images of (b) MoS_2 monolayer and (c)V-MoS_2, respectively (d) AFM image of MoS_2 monolayer under plasma treatment for 30 s (V_{30}-MoS_2) (e) Raman and (f) PL spectra of MoS_2 monolayer under different time of plasma treatment inset of (e) showing the optical images of MoS_2 and V_{30}-MoS_2.

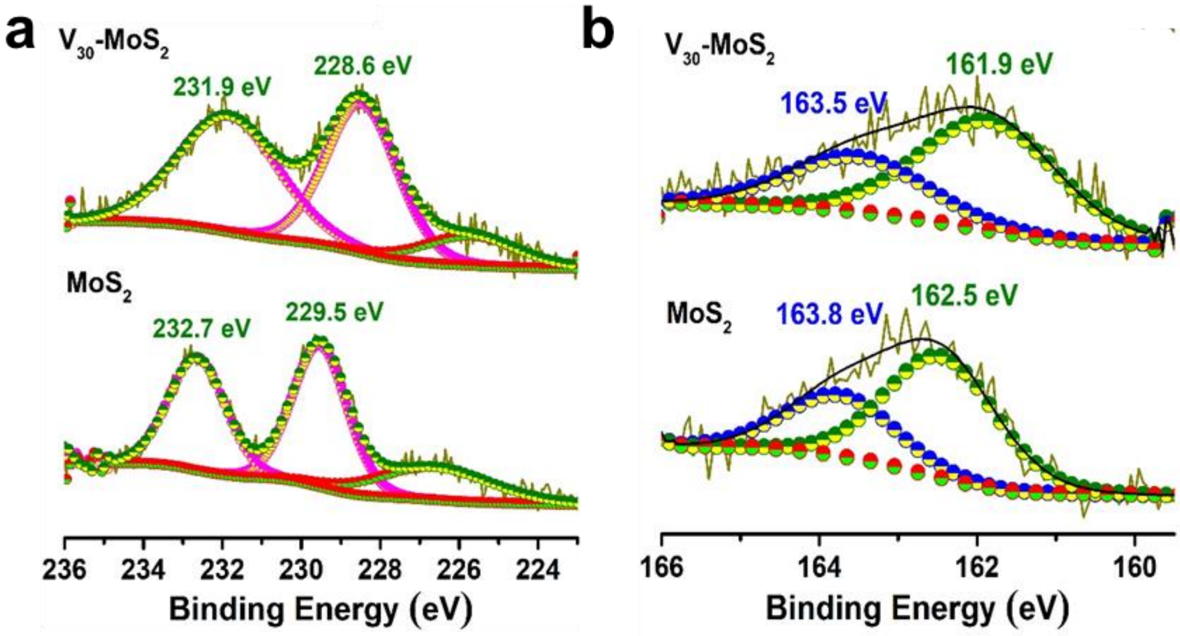

Fig. S7 XPS spectra (a) Mo 3d (b) S 2p of MoS_2 and V_{30}-MoS_2

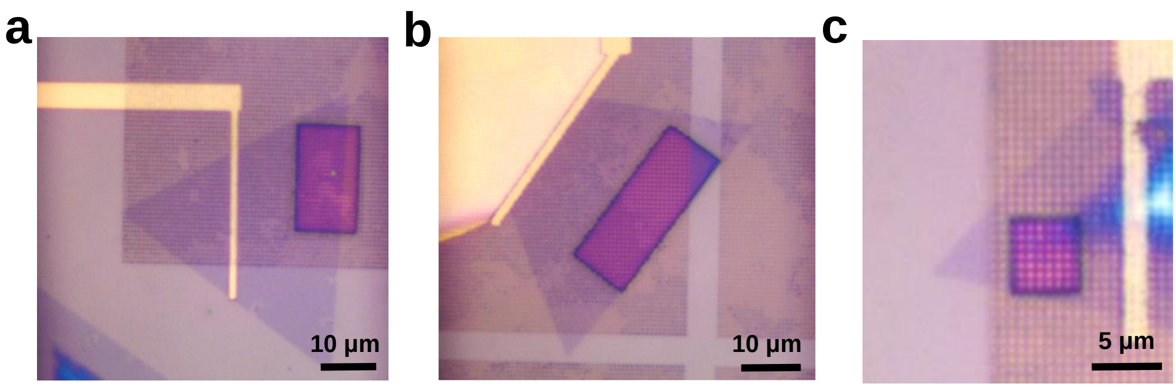

Fig. S8 (a) photograph of the microcell assembly (b-d) Optical images of various $S_{0.5\mu m}MoS_2$ devices

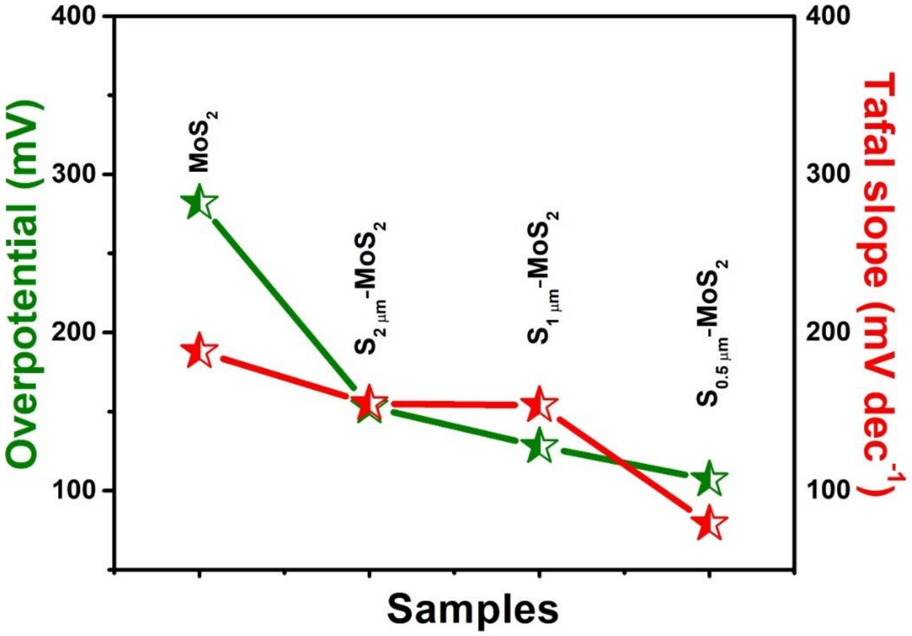

Fig. S9 The overpotential and the Tafel slope for the different strained samples (S-MoS_2)

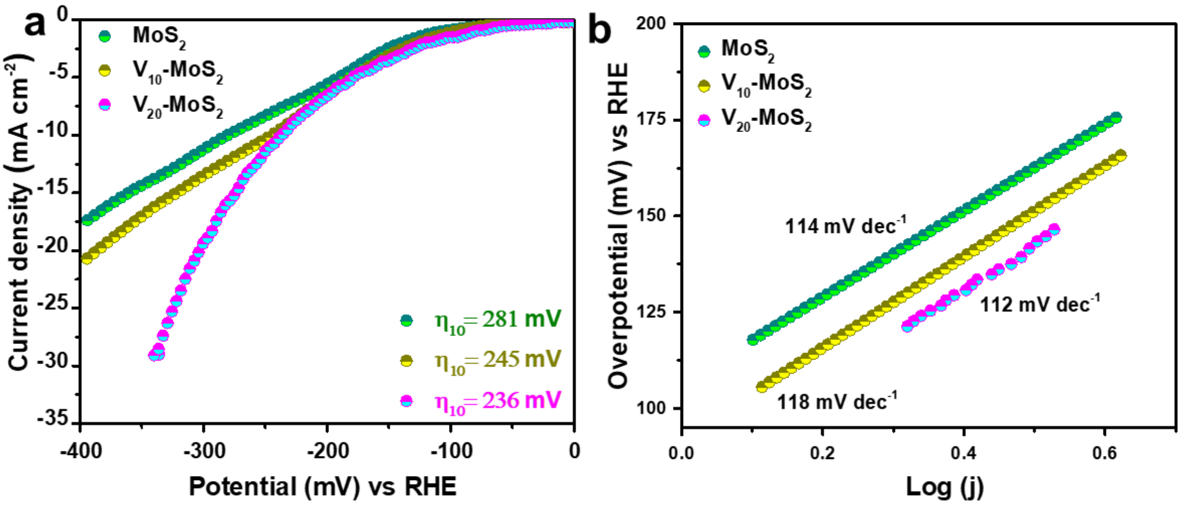

Fig. S10 a) LSV and (b) Tafel curve of MoS_2, V_{10}-MoS_2 and V_{20}-MoS_2.

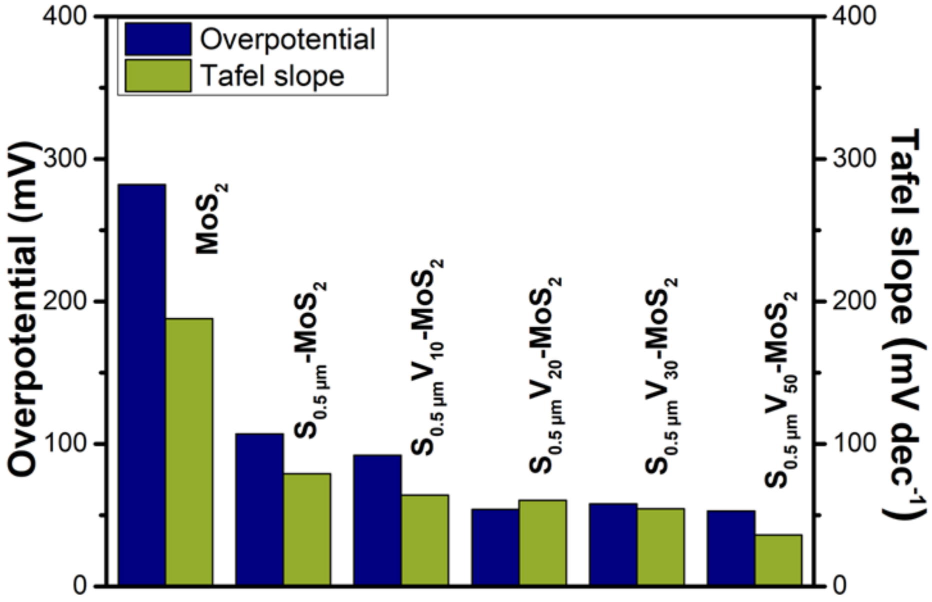

Fig. S11 Comparison bar plots of overpotential and the Tafel slope for the different SV-MoS_2 samples

Table S1 Comparison of HER performance of engineered TMDs-based electrocatalysts.

Modification method	Material	Before modification	After modification	References
Strained and vacancy	MoS2	382 mV 186 mV dec^{-1}	53 mV 118 mV dec^{-1}	This work
	WS_2@ C	-- 98 mV dec^{-1}	117 mV 56 mV dec^{-1}	1
	SV-MoS_2	-- 98 mV dec^{-1}	170 mV 60 mV dec^{-1}	2
	Vs-MoS_2 on 90 nm NCs	498 mV 156.4 mV dec^{-1}	234 mV 79.7 mV dec^{-1}	3
Gating	ReS_2/WS_2	210 mV 115 mV dec^{-1}	49 mV 35 mV dec^{-1}	4
	Treated VSe_2	126 mV	70 mV	5

		70 mV dec^{-1}	59 mV dec^{-1}	
	MoS_2	240 mV 200 mV dec^{-1}	38 mV 110 mV dec^{-1}	6
Interface	MoS_2/WTe_2	-- --	140 mV 40 mV dec^{-1}	7
Defects/ Vacancies	3CoMo−Vs MoS_2	317 mV 175 mV dec^{-1}	75 mV 57 mV dec^{-1}	8
	MoS_2	384 mV 139 mV dec^{-1}	266 mV 90 mV dec^{-1}	9
	FD-MoS_2	358 mV 165 mV dec^{-1}	164 mV 36 mV dec^{-1}	10
	1T'–$MoTe_2$	280 mV 185 mV dec^{-1}	140 mV 160 mV dec^{-1}	11
Phase engineering	1T–MoS_2	— 180 mV dec^{-1}	175 mV 100 mV dec^{-1}	12
	1T′-$MoTe_2$	650 mV 184 mV dec^{-1}	356 mV 127 mV dec^{-1}	13
	1T′-MoS_2	286 mV 70 mV dec^{-1}	205 mV 51 mV dec^{-1}	14
Doping	Co–MoS_2	345 mV 143 mV dec^{-1}	137 mV 59 mV dec^{-1}	15
	Mn-doped MoS_2	369 mV 105 mV dec^{-1}	318 mV 82 mV dec^{-1}	16

Reference

1 W. Han, Z. Liu, Y. Pan, G. Guo, J. Zou, Y. Xia, Z. Peng, W. Li and A. Dong, *Adv. Mater.*, 2020, **32**, 1–9.

2 H. Li, C. Tsai, A. L. Koh, L. Cai, A. W. Contryman, A. H. Fragapane, J. Zhao, H. S. Han, H. C. Manoharan, F. Abild-Pedersen, J. K. Nørskov and X. Zheng, *Nat. Mater.*, 2016, **15**, 48–53.

3 X. Liu, Z. Li, H. Jiang, X. Wang, P. Xia, Z. Duan, Y. Ren, H. Xiang, H. Li, J. Zeng, Y. Zhou and S. Liu, *Small*, 2024, **20**, 2307293.

4 X. Zhu, C. Wang, T. Wang, H. Lan, Y. Ding, H. Shi, L. Liu, H. Shi, L. Wang, H. Wang, Y. Ding, Y. Fu, M. Zeng and L. Fu, *Adv. Mater.*, 2022, **34**, 2202479.

5 M. Yan, X. Pan, P. Wang, F. Chen, L. He, G. Jiang, J. Wang, J. Z. Liu, X. Xu, X. Liao, J. Yang and L. Mai, *Nano Lett.*, 2017, **17**, 4109–4115.

6 J. Wang, M. Yan, K. Zhao, X. Liao, P. Wang, X. Pan, W. Yang and L. Mai, *Adv. Mater.*, 2017, **29**, 1604464.

7 Y. Zhou, J. V. Pondick, J. L. Silva, J. M. Woods, D. J. Hynek, G. Matthews, X. Shen, Q. Feng, W. Liu, Z. Lu, Z. Liang, B. Brena, Z. Cai, M. Wu, L. Jiao, S. Hu, H. Wang, C. M. Araujo and J. J. Cha, *Small*, 2019, **15**, 1–11.

8 Y. Zhou, J. Zhang, E. Song, J. Lin, J. Zhou, K. Suenaga, W. Zhou, Z. Liu, J. Liu, J. Lou and H. J. Fan, *Nat. Commun.*, 2020, **11**, 2253.

9 C. Zhu, M. Yu, J. Zhou, Y. He, Q. Zeng, Y. Deng, S. Guo, M. Xu, J. Shi, W. Zhou, L. Sun, L. Wang, Z. Hu, Z. Zhang, W. Guo and Z. Liu, *Nat. Commun.*, 2020, **11**, 772.

10 J. Xu, G. Shao, X. Tang, F. Lv, H. Xiang, C. Jing, S. Liu, S. Dai, Y. Li, J. Luo and Z. Zhou, *Nat. Commun.*, 2022, **13**, 2193.

11 H. You, Z. Zhuo, X. Lu, Y. Liu, Y. Guo, W. Wang, H. Yang, X. Wu, H. Li and T. Zhai, *CCS Chem.*, 2019, **1**, 396–406.

12 Y. Yu, G. H. Nam, Q. He, X. J. Wu, K. Zhang, Z. Yang, J. Chen, Q. Ma, M. Zhao, Z. Liu, F. R. Ran, X. Wang, H. Li, X. Huang, B. Li, Q. Xiong, Q. Zhang, Z. Liu, L. Gu, Y. Du, W. Huang and H. Zhang, *Nat. Chem.*, 2018, **10**, 638–643.

13 J. Seok, J.-H. Lee, S. Cho, B. Ji, H. W. Kim, M. Kwon, D. Kim, Y.-M. Kim, S. H. Oh, S. W. Kim, Y. H. Lee, Y.-W. Son and H. Yang, *2D Mater.*, 2017, **4**, 025061.

14 L. Liu, J. Wu, L. Wu, M. Ye, X. Liu, Q. Wang, S. Hou, P. Lu, L. Sun, J. Zheng, L. Xing, L. Gu, X. Jiang, L. Xie and L. Jiao, *Nat. Mater.*, 2018, **17**, 1108–1114.

15 H. Duan, C. Wang, G. Li, H. Tan, W. Hu, L. Cai, W. Liu, N. Li, Q. Ji, Y. Wang, Y. Lu, W. Yan, F. Hu, W. Zhang, Z. Sun, Z. Qi, L. Song and S. Wei, *Angew. Chemie Int. Ed.*, 2021, **60**, 7251–7258.

16 Z. Cai, T. Shen, Q. Zhu, S. Feng, Q. Yu, J. Liu, L. Tang, Y. Zhao, J. Wang, B. Liu and H. Cheng, *Small*, 2020, **16**, 1–9.