Electronic Supplementary Material (ESI) for Catalysis Science & Technology.

Supplementary data

Cracked monolayer 1T MoS_2 with Abundant Active Sites for Enhanced Electrocatalytic Hydrogen Evolution

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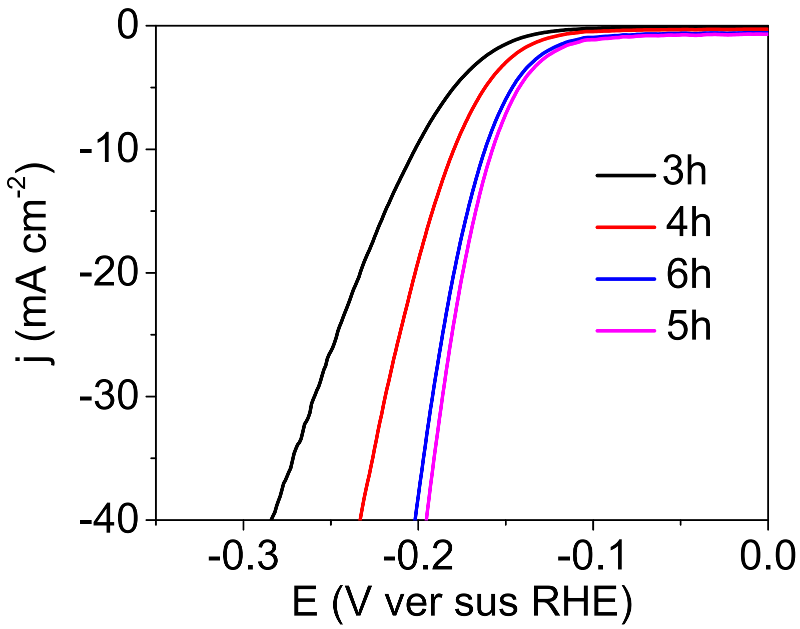

Fig. S1 Polarization curves of C-1T MoS_2 samples prepared under different sonication time: 3 h, 4 h, 5 h, and 6 h.

From this figure, we can see that the sonication time can great influence on the HER performance. The overpotential at a current density of 10 mA cm^{-2} decreases first and then increases with prolonging sonication time. The maximum overpotential vs reversible hydrogen electrode was achieved on 5 h sonication time.

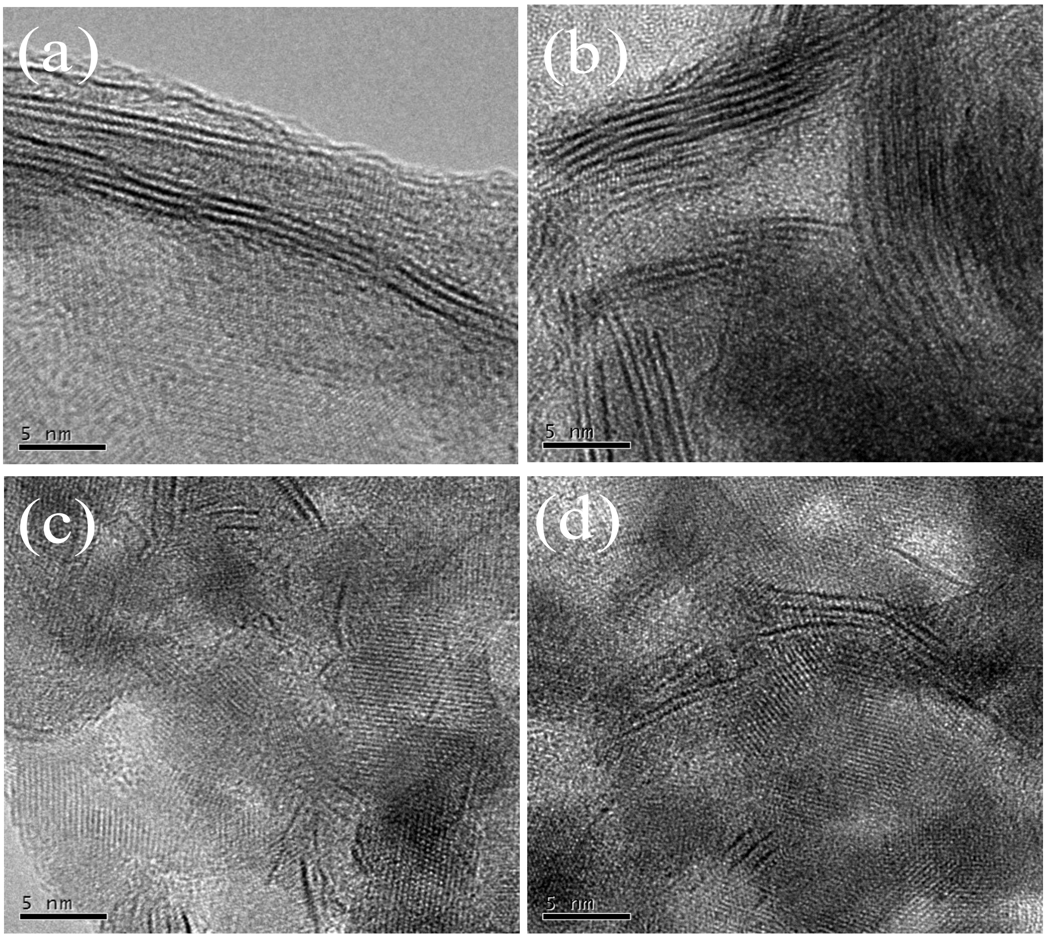

Fig. S2 Additional TEM and HRTEM images of 2H MoS_2 nanosheets (a-b) and C-1T MoS_2 nanosheets (c-d).

Fig. S2a and b reveals the layer of hydrothermal synthesis MoS_2 nanosheets is 3-7 layers, and the edge of the nanosheets is long and continuous. However, after exfoliating the 2H MoS_2 nanosheets, the entire nanosheets was created into some monolayer smaller nanosheets as shown in Fig. S2c and d. As we can see, the edge of C-1T MoS_2 is shortened and discontinuous, confirming the C-1T MoS_2 is consisted of some MoS_2 pieces.

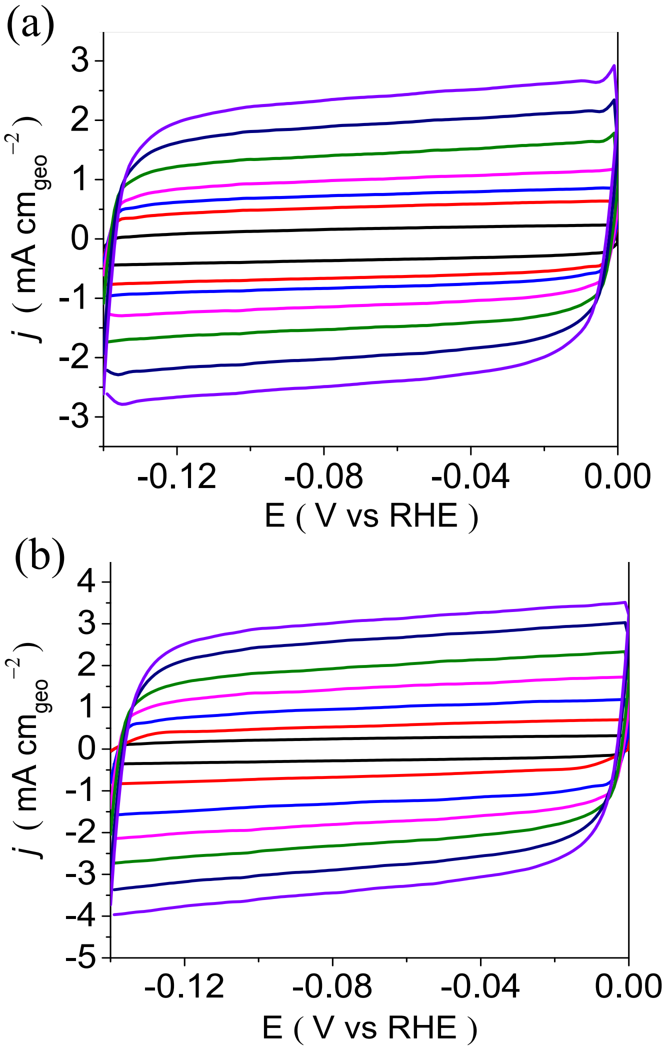

Fig. S3 CV curves in the region of 0.0-0.14 V (V *vs* RHE) for 2H MoS_2 nanosheets (a) and C-1T MoS_2 nanosheets (b).

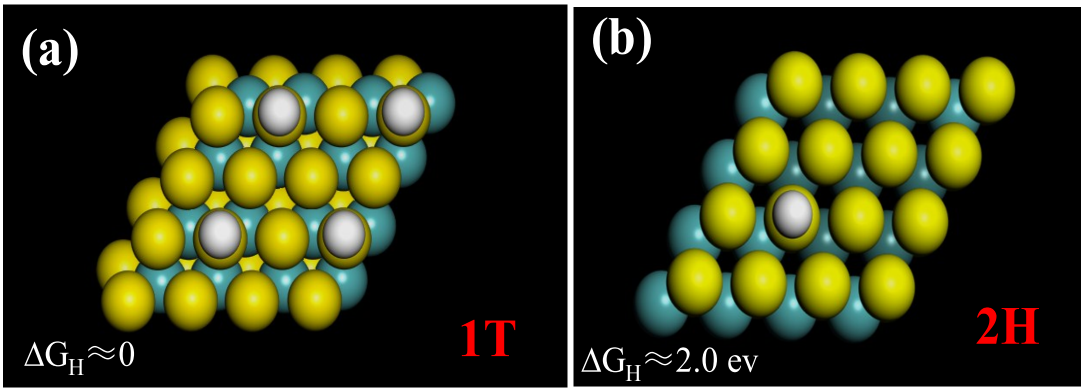

Fig. S4 Gibbs free energy of adsorbed hydrogen (ΔG_H) for (a) 2H MoS_2 basal plane, and (b) 1T MoS_2 basal plane with around 12.5%~25% H coverage. Blue, yellow and white balls indicate Mo, S and H atoms, respectively.

According to the recent reported calculations by Jiang's group, the ΔG_H is between −0.28 and 0.13 eV at the 1T MoS_2 basal plane with around 12.5%~25% H coverage, while the ΔG_H value is about 2.0 ev for 2H MoS_2 basal plane. As we know, the optimal HER catalysts have hydrogen adsorption energies is close to zero ($\Delta G_H \approx 0$), or the binding hydrogen energy is neither too weakly nor too strongly. Fig. S4a shows the model of 1T MoS_2 basal plane with around 25% H coverage. Fig. S4b shows only one hydrogen adsorbed on the 2H MoS_2 basal plane because higher ΔG_H makes the protons bonded too weakly to the catalyst surface, thereby leading to a slower HER kinetics.

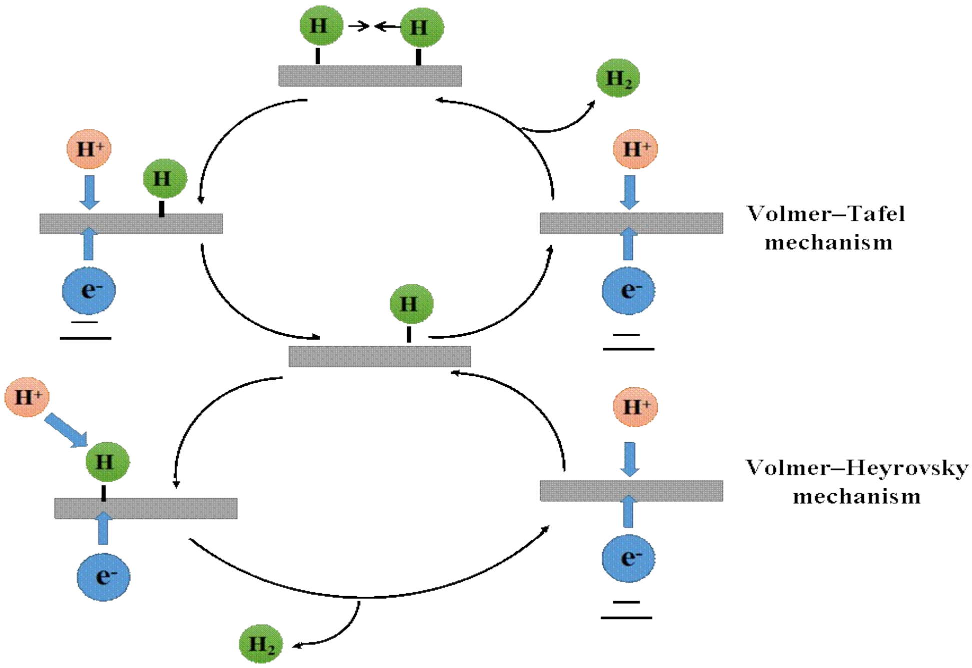

Fig. S5 The mechanism of hydrogen evolution on the surface of an electrode in acidic solutions.

Table S1. Comparison of representative MoS_2 or MoS_2-based catalytic parameters recently.

Photocatalysts	Onset overpotential [mV]	Tafel slopes [mV decade^{-1}]	η@ j = 10 mA cm^{-2} [mV]	Ref.
C-1T MoS_2	113	42.7	156	this work
2H MoS_2	161	75.3	274	this work
Defect-rich/MoS_2	120	50	195	1
1T-MoS_2	135	43	187	2
O-MoS_2	120	55	N/A	3
MoS_2-NR/rGO	N/A	46	N/A-	4
hierarchical MoS_2	50	60	167	5
O-MoS_2	120	58	N/A-	6
O-MoS_2/G	120	51	N/A-	6
plasma exposed MoS_2	300	117	N/A	7
MoS_2 nanodots	90	61	248	8
MoS_2 dots/Au	190	74	N/A-	9
P-1T MoS_2	N/A	43	153	10
MoS_2 hollow spheres	112	74	214	11
edge-terminated MoS_2	103	49	149	12
1T@2H-MoS_2	42	49	64	13
CoS_2/MoS_2/CC	N/A	66	177	14
micro and ground microflakes -MoS_2	N/A	60-70	174	15
MoS_2/Graphene	30	67.4	110	16
Hollow MoS_2	N/A	48	202	17
C/MoS_2@G	165	46	N/A	18
Laser MoS_2/carbon	N/A	64	216	19
DR-MoS_2	166	66	208	20

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