Supporting Information

Improve Charge Injection of the Edge Aligned MoS_2/MoO_2 Hybrid Nanosheets for Highly Robust and Efficient Electrocatalysis of H_2 Production

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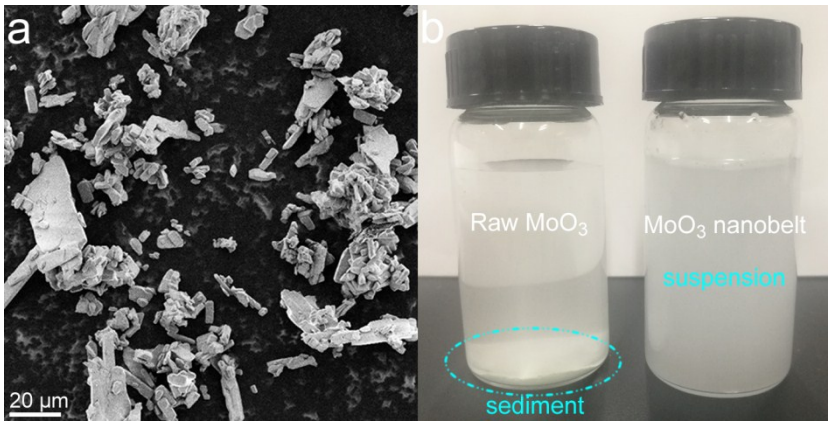

Fig. S1 (a) SEM image of the raw MoO_3 material; (b) Photographs of the suspension of the raw MoO_3 material and MoO_3 nanobelts. The raw MoO_3 material cannot be dispersed well in the aqueous solution, *i.e*. sediment formed with seconds. By contrast, the MoO_3 nanobelt can be dispersed well and stable (no visible sediment formed in half hour) in the aqueous solution, likely owing to the small size.

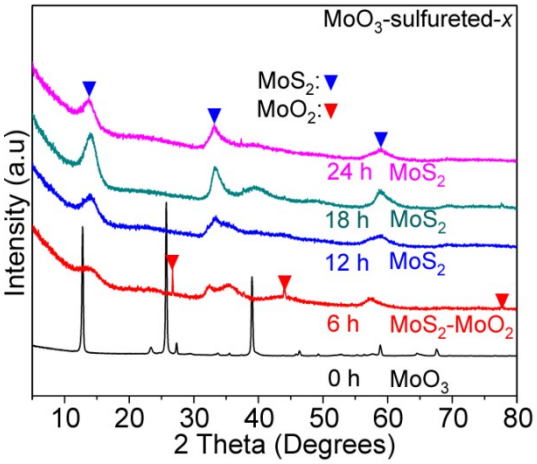

Fig. S2 XRD patterns of the MoO_3 nanobelts (orthorhombic *a*-MoO_3, ICDD card: 05-0508) and MoO_3-sulfureted-*x* samples (*x* = 6, 12, 18, 24 h). Diffraction peaks at 13.9, 33.4, and 58.9° are attributed from the (002), (100) and (110) crystal planes of 2H-MoS_2 (ICDD card: 87-2416). Diffraction peaks at around 26.7, 44.1, and 77.7° can be ascribed to the metallic monoclinic MoO_2 phase (ICDD card: 32-0671).

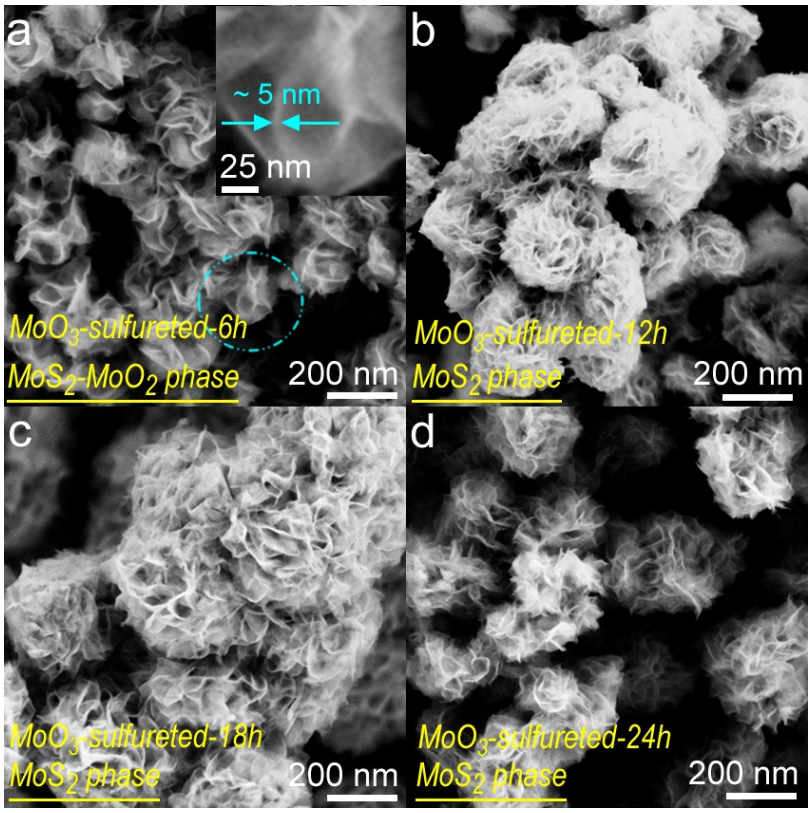

Fig. S3 SEM images of MoO_3-sulfureted-6h (MoS_2-MoO_2 composite) (a), MoO_3-sulfureted-12h (pure MoS_2) (b) MoO_3-sulfureted-18h (pure MoS_2) (c) and MoO_3-sulfureted-24h (pure MoS_2) (d). Inset in figure a is the magnified local area (as labeld).

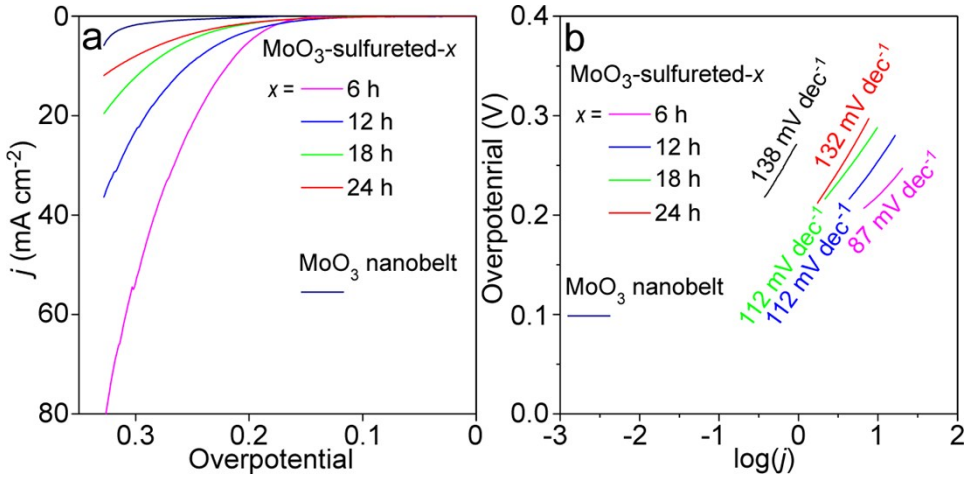

Fig. S4 LSV (a) and Tafel (b) of MoO_3-sulfureted-x (x = 6, 12, 18, 24 h) and MoO_3 nanobelt. The MoO_3-sulfureted-6h exhibited the best catalytic performance among these investigated samples.

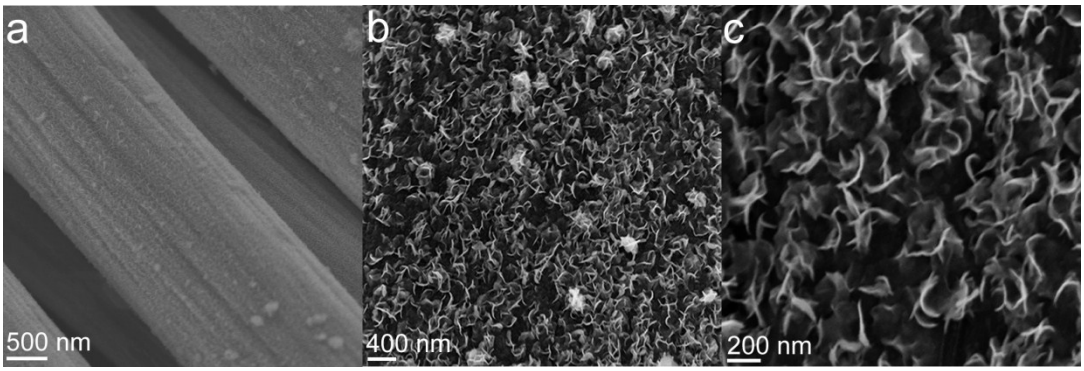

Fig. S5 SEM images of MoS_2@MoO_2/CC-6h.

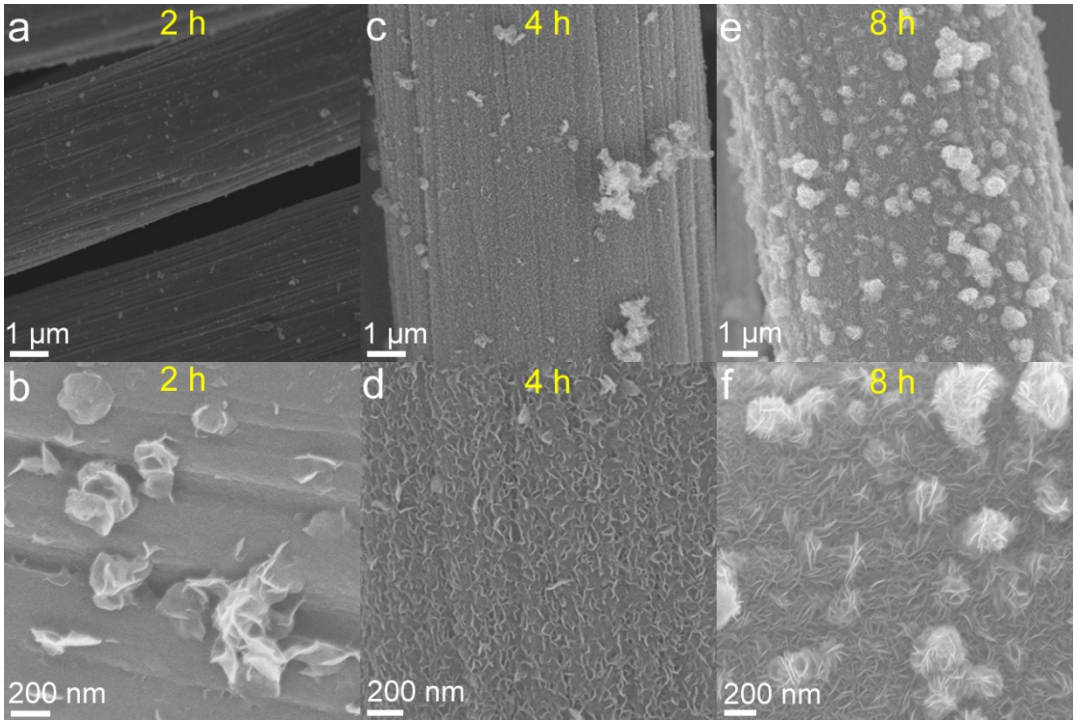

Fig. S6 SEM images of MoS_2@MoO_2/CC-*x* obtained with different reaction time. a-b) 2h. c-d) 4h. e-f) 8h. In MoS_2@MoO_2/CC-2h, the particle on the surface are really few. Figure b shows some particles found occasionlly.

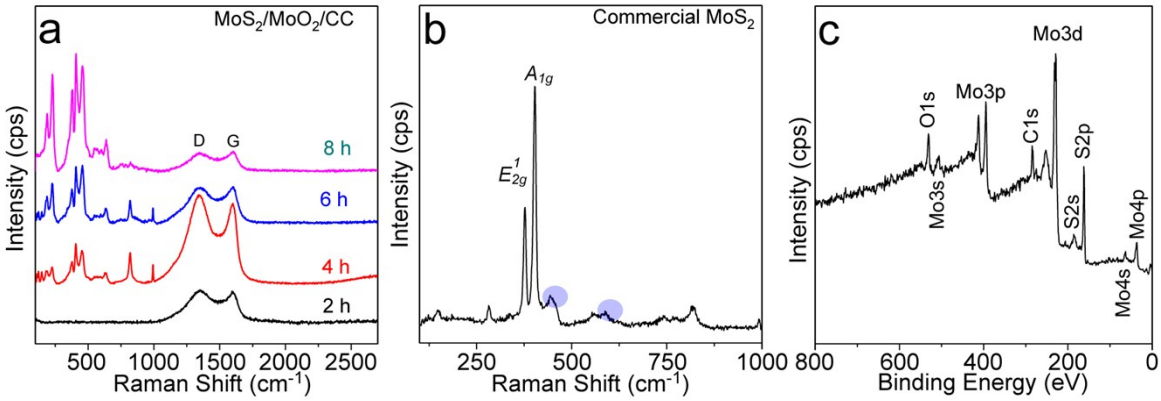

Fig. S7 (a) Raman spectra of MoS_2@MoO_2/CC-2h, MoS_2@MoO_2/CC-4h, MoS_2@MoO_2/CC-6h and MoS_2@MoO_2/CC-8h; (b) Raman spectrum of the commercial MoS_2 flakes (Sigma-Aldrich, product No: 234842). The highlighted peaks are around 457 and 590 cm^{-1} which were also found in our prepared samples; (c) XPS full survery of the MoS_2@MoO_2/CC-6h.

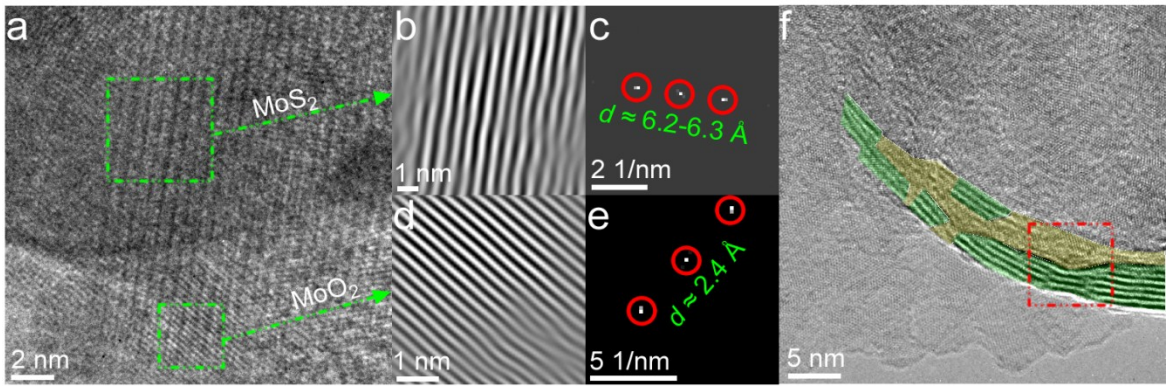

Fig. S8 (a-f) HRTEM image of one sheet show both the 2H-MoS_2 and monoclinic MoO_2 cryatal domains (a), which are highlighted as the corresponding simulated HRTEM images (b, d) and FFT patterns (c, f); f) One edge composed by the MoO_2 and MoS_2, with selected area magnified in Fig. 3l.

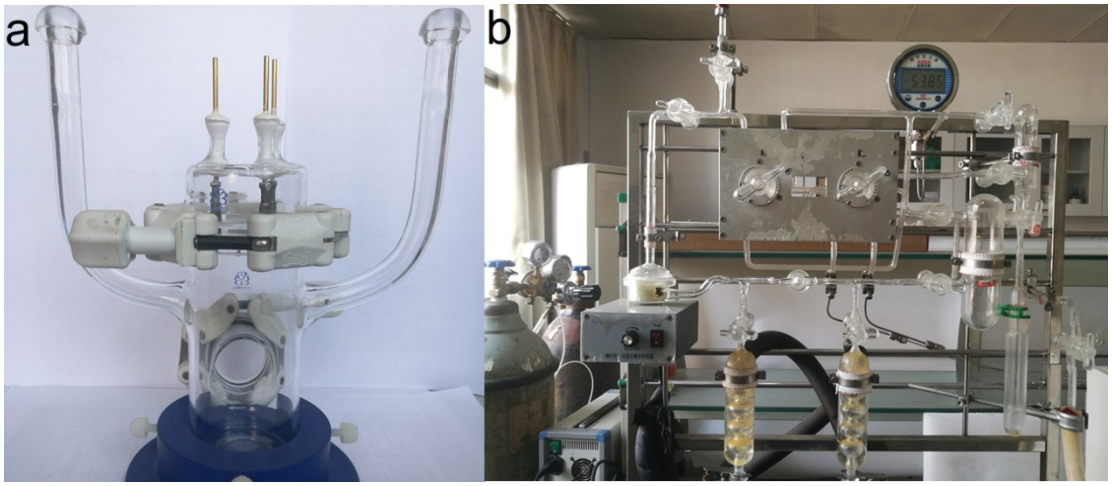

Fig. S9 (a) the photograph of the sealable three-electrode cell for the evaluation of the Faradaic efficiency. In this case, the reference electrode of the Ag/AgCl with saturated KCl was used; (b) The photograph of the full set up used for the estimation of the Faradaic efficiency.

Table S1 Theoretical H_2 production *versus* experimental measured H_2 amount, and Faradaic efficiencies at different period for MoS_2@MoO_2-CC-6h (constant *j* of 30 mA cm^{-2}, 1.0×1.0 cm^2 electrode).

Time (min)	20	40	60	80	100	120
Calculated (mmol)	0.1866	0.3732	0.5598	0.7464	0.933	1.116
Measured (mmol)	0.1824	0.3671	0.5516	0.7396	0.9294	1.1105
Faradaic efficiency	97.7%	98.3%	98.5%	99.1%	99.6%	99.2%
Average	98.7%					

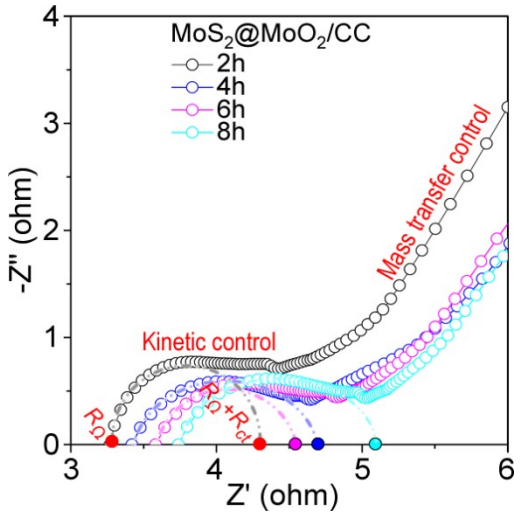

Fig. S10 EIS Nyquist plots of various catalysts modified electrodes in a 0.5 M H_2SO_4 solution at 5 mV s^{-1}. The (R_Ω, R_Ω+R_{ct}) of MoS_2@MoO_2/CC-2h, MoS_2@MoO_2/CC-4h, MoS_2@MoO_2/CC-6h, MoS_2@MoO_2/CC-8h are (3.27 Ω, 4.30 Ω), (3.42 Ω, 4.69 Ω), (3.58 Ω, 4.54 Ω) and (3.74 Ω, 5.10 Ω), respectively.

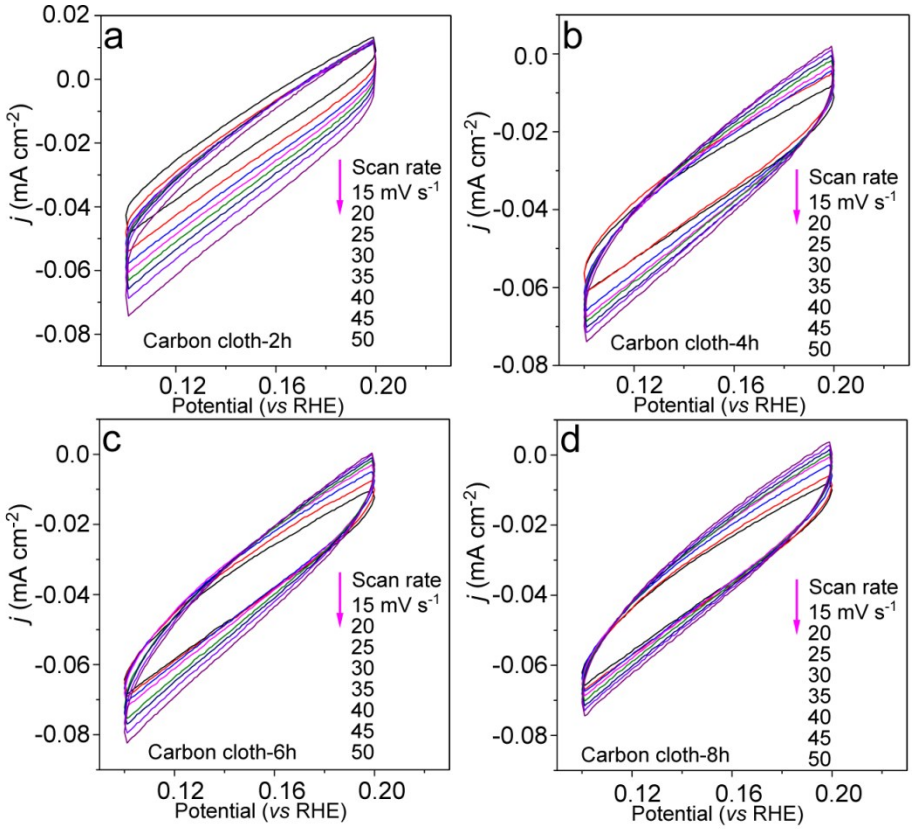

Fig. S11 CV curves of carbon colth after solvothermal reactions with different reaction time: (a) carbon cloth-2h; (b) carbon cloth-4h; (c) carbon cloth-6h; (d) carbon cloth-8h. The C_{dl} calculated at the potential of 0.15 V of these four samples are nearly identical.

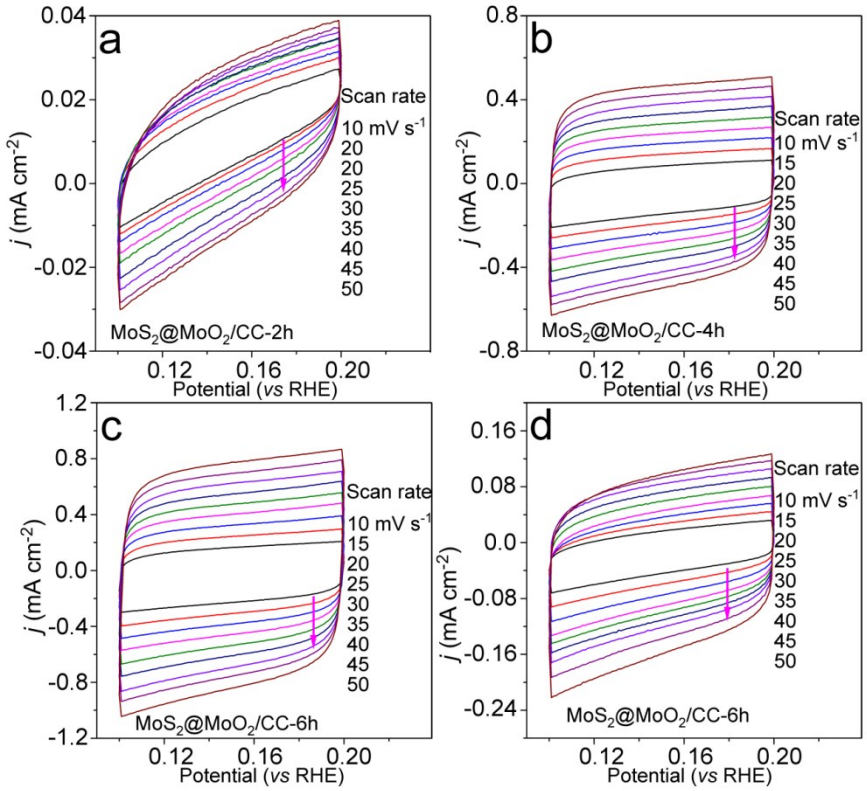

Fig. S12 CV curves of different samples: (a) MoS_2@MoO_2/CC-2h; (b) MoS_2@MoO_2/CC-4h; (c) MoS_2@MoO_2/CC-6h; (d) MoS_2@MoO_2/CC-8h.