

Non-equilibrium fractal growth of MoS₂ for electrocatalytic hydrogen evolution

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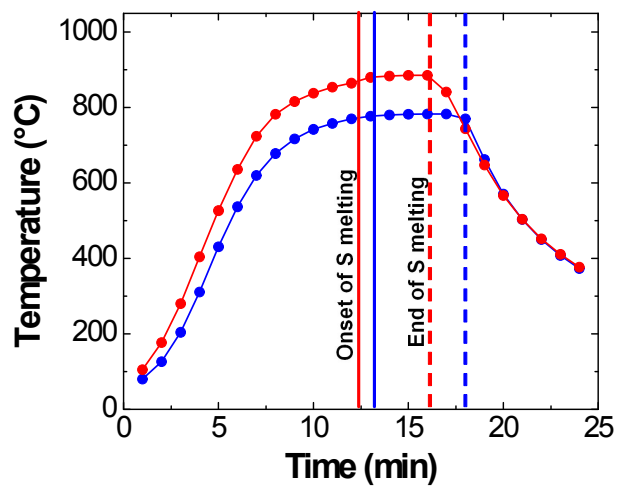


Fig. S1 CVD temperature profiles for the 780 °C (blue) and 890 °C (red) experiments. The onset and end points for sulfur melting are indicated with solid and dashed lines, respectively.

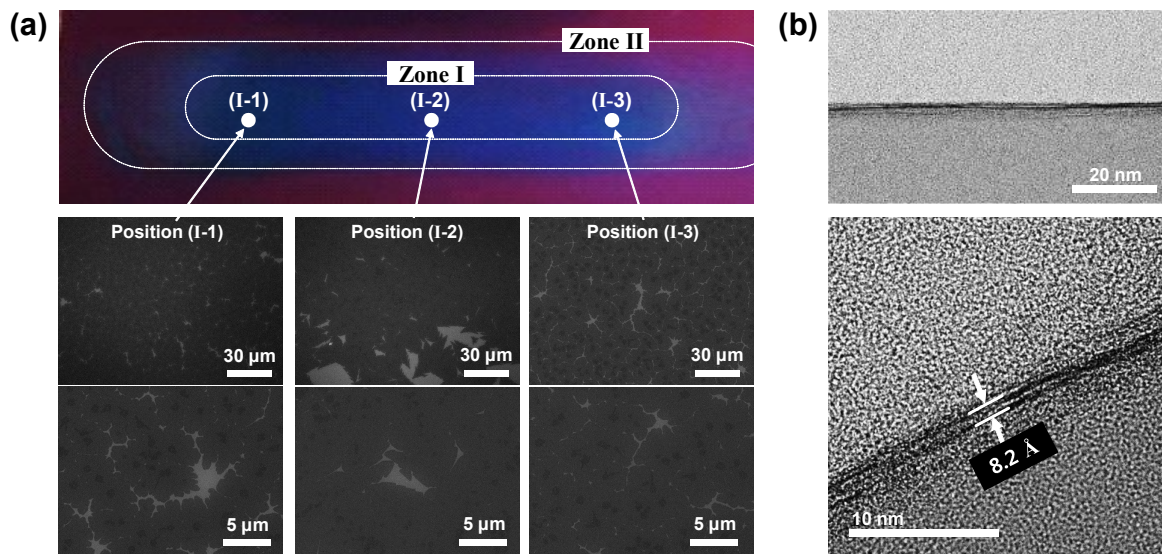


Fig. S2 (a) SEM images of the coalesced MoS₂ films grown on different positions in Zone I. (b) TEM analysis results of a monolayer MoS₂ film obtained from position I-2 in Zone I.

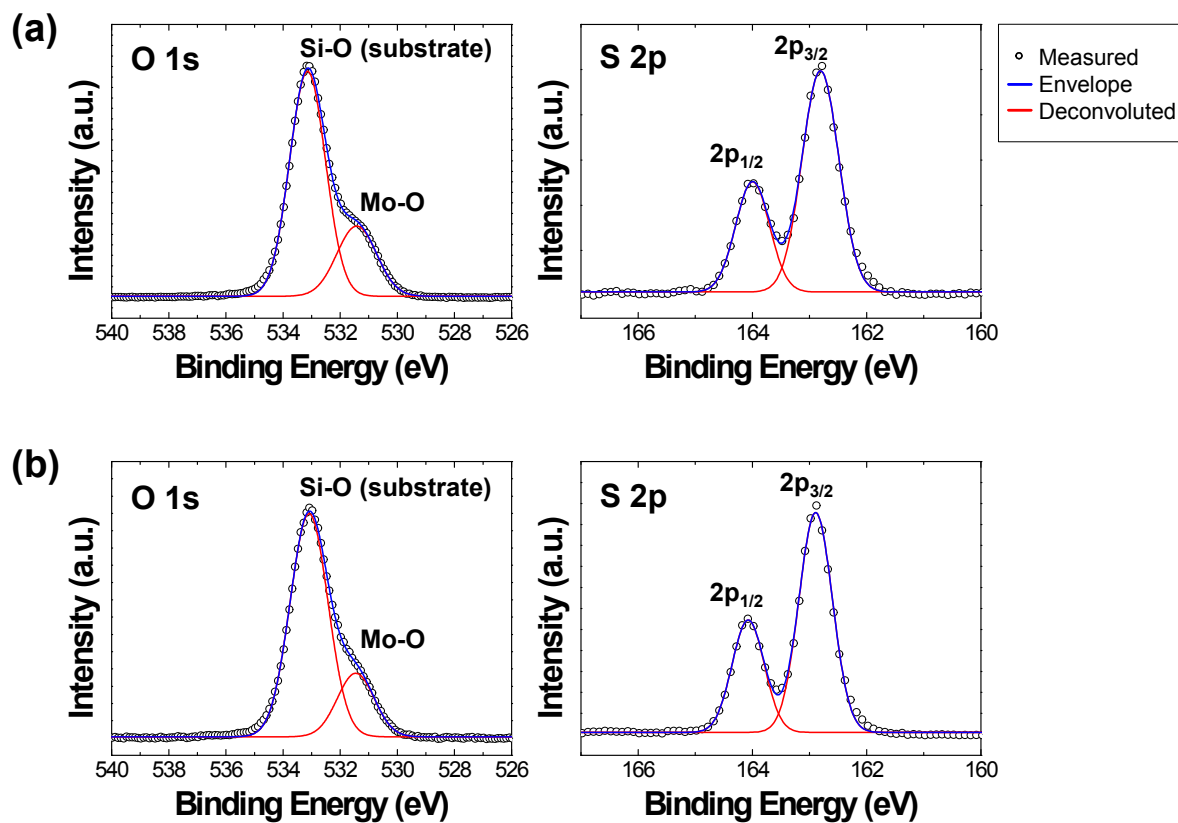


Fig. S3 XPS spectra of O 1s and S 2p peaks measured from (a) center and (b) tail regions marked in Fig. 1(b).

Table S1. Peak information and quantification results of XPS.

Position	Peak	Binding energy (eV)	FWHM (eV)	Area (normalized)	Atomic %
Confined center Mo : S = 0.95 : 1.00	Mo 3d _{5/2} (Mo ⁴⁺)	229.93	0.79	25.82	7.1
	Mo 3d _{5/2} (Mo ⁵⁺)	232.46	1.6	11.54	3.2
	Mo 3d _{5/2} (Mo ⁶⁺)	233.38	1.27	14.93	4.1
	O 1s	533.11	1.62	256.73	70.5
	S 2p	162.81	0.74	55	15.1
Confined tail Mo : S = 0.63 : 1.00	Mo 3d _{5/2} (Mo ⁴⁺)	230.07	0.69	47.63	14.7
	Mo 3d _{5/2} (Mo ⁵⁺)	232.75	1.81	11.51	3.6
	Mo 3d _{5/2} (Mo ⁶⁺)	233.75	1.1	5.52	1.7
	O 1s	533.06	1.76	156	48.2
	S 2p	162.9	0.67	102.98	31.8

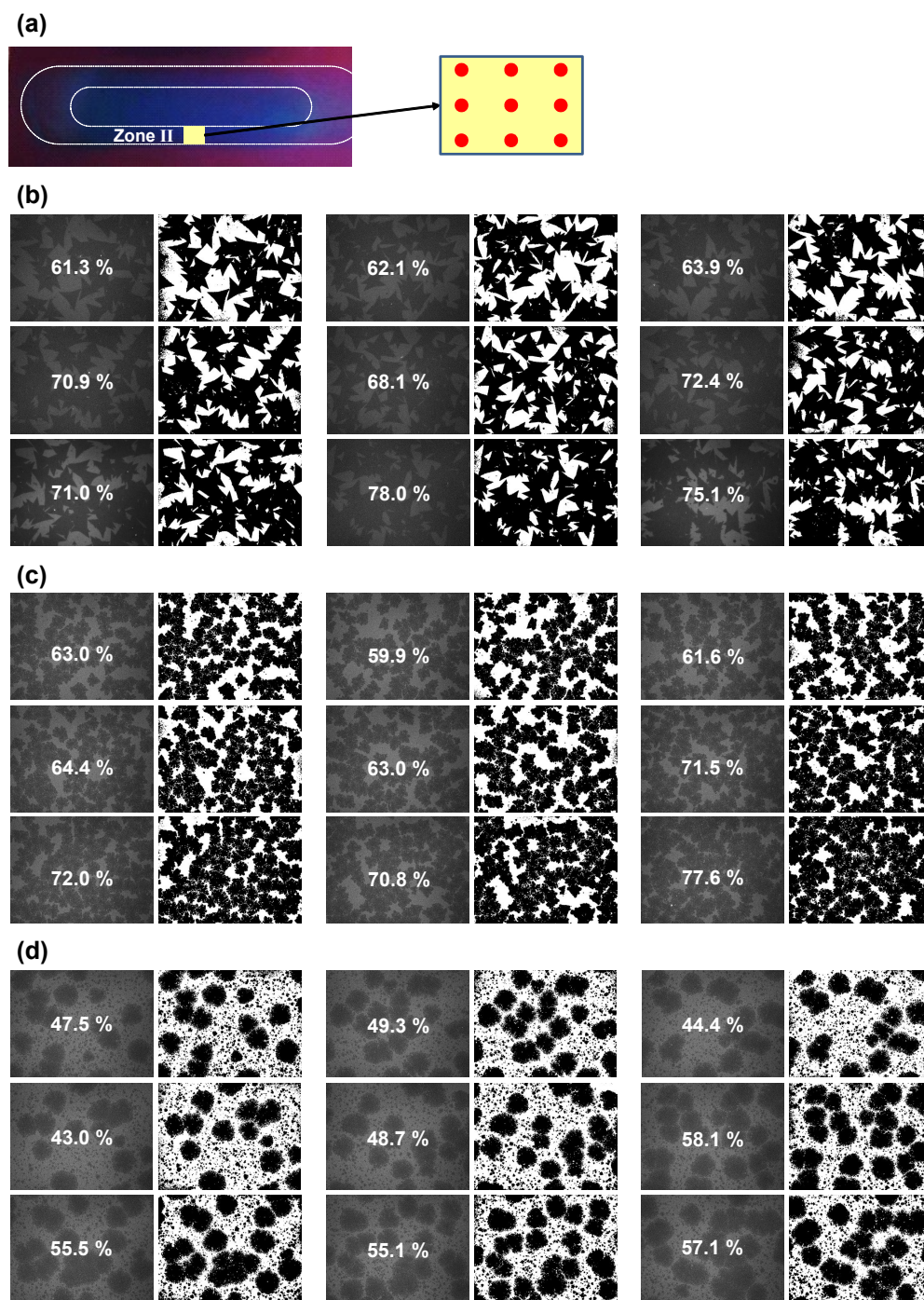


Fig. S4 (a) Diagram showing the sample area (yellow box) used for the LSV measurement and also the nine positions (red dots) taken for SEM imaging. (b)–(d) SEM images and the corresponding converted binary contrast images of (b) T-MoS₂, (c) S-MoS₂, and (d) D-MoS₂ samples. All the SEM images are taken after transferring the MoS₂ flakes on the Au/Si substrates and the estimated projected areal portions are presented in each SEM image.

Table S2. Estimated dimensions and corresponding HER performance of T-, S-, and D-MoS₂.

Flake	Flake area exposed to LSV (μm^2)	Flake edge length exposed to LSV (μm)	Current density at -4.0 V (mA/cm ²)	HER in the unit area* (mA/cm ²)	HER in the unit length** (mA/cm ²)
T-MoS ₂	41520 \pm 3480	11061 \pm 409	15.8	3.8 \times 10 ⁻⁴	1.4 \times 10 ⁻³
S-MoS ₂	40260 \pm 3600	20683 \pm 770	25.2	6.3 \times 10 ⁻⁴	1.2 \times 10 ⁻³
D-MoS ₂	30600 \pm 3360	29611 \pm 1065	58.5	1.9 \times 10 ⁻³	2.0 \times 10 ⁻³

* HER in the unit area was calculated by dividing the current density with the flake area.

** HER in the unit length was calculated by dividing the current density with the flake edge length. The similar values for T- and S-MoS₂ suggested their similar crystalline edge qualities. In the case of D-MoS₂, the value was believed to be overestimated because the additional edge length from the overlapping crystals could not be determined from the binary contrast images (see Fig. S4d).

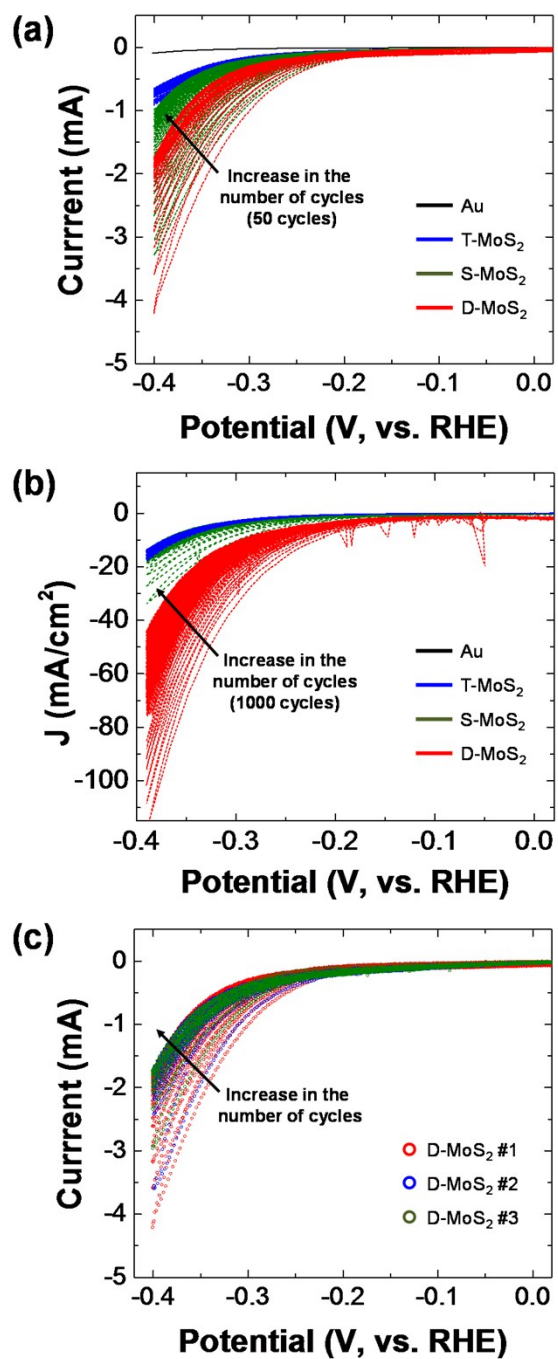


Fig. S5 (a) LSV measurements for 50 cycles and (b) HER polarization curves during 1000 measurement cycles for the T-, S-, and D-MoS₂ samples. (c) LSV measurements of the D-MoS₂ obtained from three different batches.