Supporting Information

## Chemical Vapor Deposition of Clean and Pure MoS<sub>2</sub> Crystals by Inhibition of MoO<sub>3-x</sub> Intermediates

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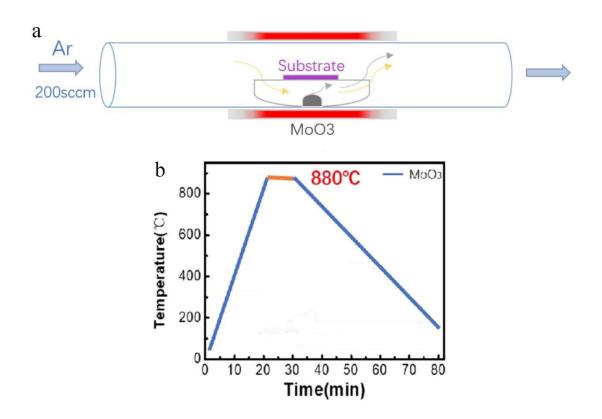
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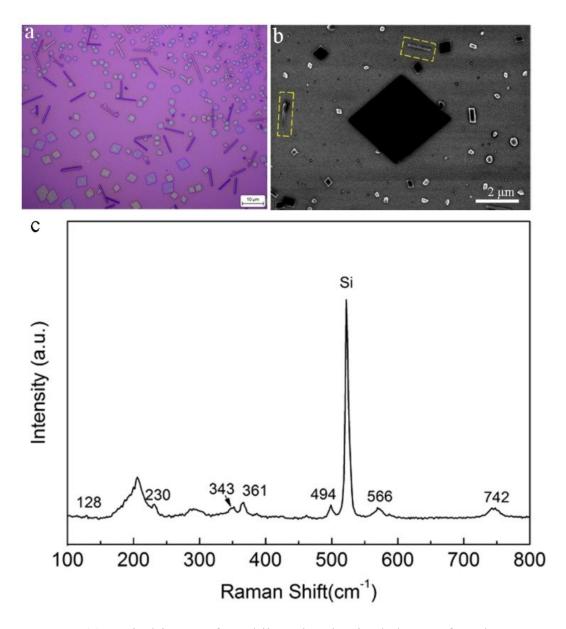
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Sample No.	Heating rate of S	Raman peaks for MoS <sub>2</sub> (cm <sup>-1</sup> )	$\begin{array}{c} A_{1g}-E^{1}{}_{2g}\\ (\Delta k)\end{array}$	Raman peaks for MoO <sub>2</sub> and/or MoOS <sub>2</sub> (cm <sup>-1</sup> )
	(°C/min)		$(cm^{-1})$	
1	1	E <sup>1</sup> <sub>2g</sub> : 381	27	128, 204, 230, 344, 361, 494,
		A1g: 408		566, 741
2	5	E <sup>1</sup> <sub>2g</sub> : 382 A <sub>1g</sub> : 405	23	124, 204, 227, 494, 566, 742
3	8	E <sup>1</sup> <sub>2g</sub> : 381 A <sub>1g</sub> : 402	21	

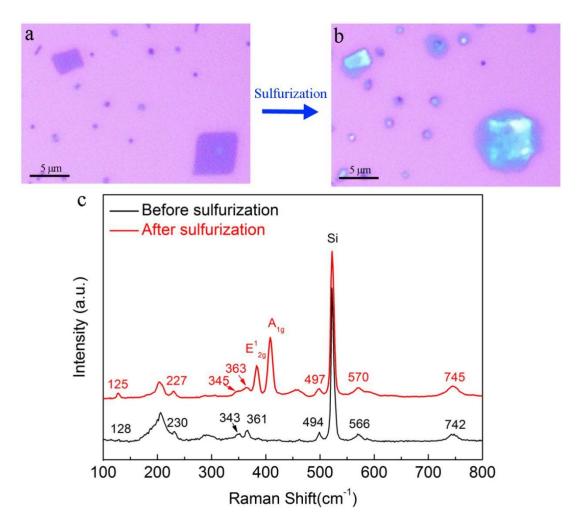
 $\textbf{Table S1}. \ Summary \ of \ Raman \ characterization \ of \ MoS_2 \ and \ MoO_{3-x}/MoS_2 \ composites.$ 



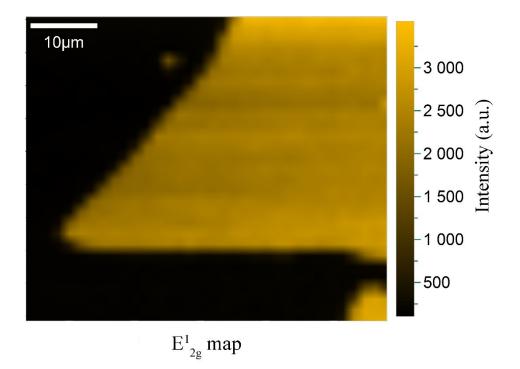
**Figure S1.** (a) Schematic diagram of the CVD setup for the S-free growth which has only high temperature zone. (b) Temperature ramping processes of the MoO<sub>3</sub> precursor as a function of time.



**Figure S2.** (a) Optical image of quadrilateral and striped shapes of products grown without S precursor. (b) Enlarged SEM image of quadrilateral and striped shapes of products grown without S precursor. (d) Raman spectra of a typical quadrilateral product grown without S precursor, showing obvious Raman characteristics of MoO<sub>2</sub>.



**Figure S3.** (a) Optical image of quadrilateral CVD products grown before S sulfurization and (b) after sulfurization. (c) The corresponding Raman spectra of quadrilateral CVD products grown before sulfurization and after sulfurization.



**Figure S4.** The  $E_{2g}^1$  intensity Raman map corresponding to the corner area of the clean and pure MoS<sub>2</sub> grain shown in the up right of Figure 4a.

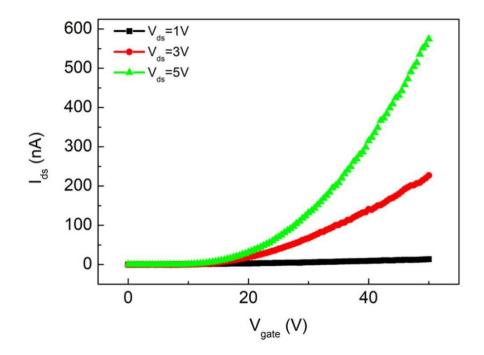


Figure S5. Transfer characteristics curves ( $I_{ds}$ - $V_{gate}$ ) of the FET device based on clean and pure MoS<sub>2</sub> at 1, 3, and 5V S-D voltage, respectively.