

# CVD Synthesis of MoS<sub>2</sub> Using Direct MoO<sub>2</sub> Precursor: A Study on the Effects of Growth Temperature on Precursor Diffusion and Morphology Evolutions

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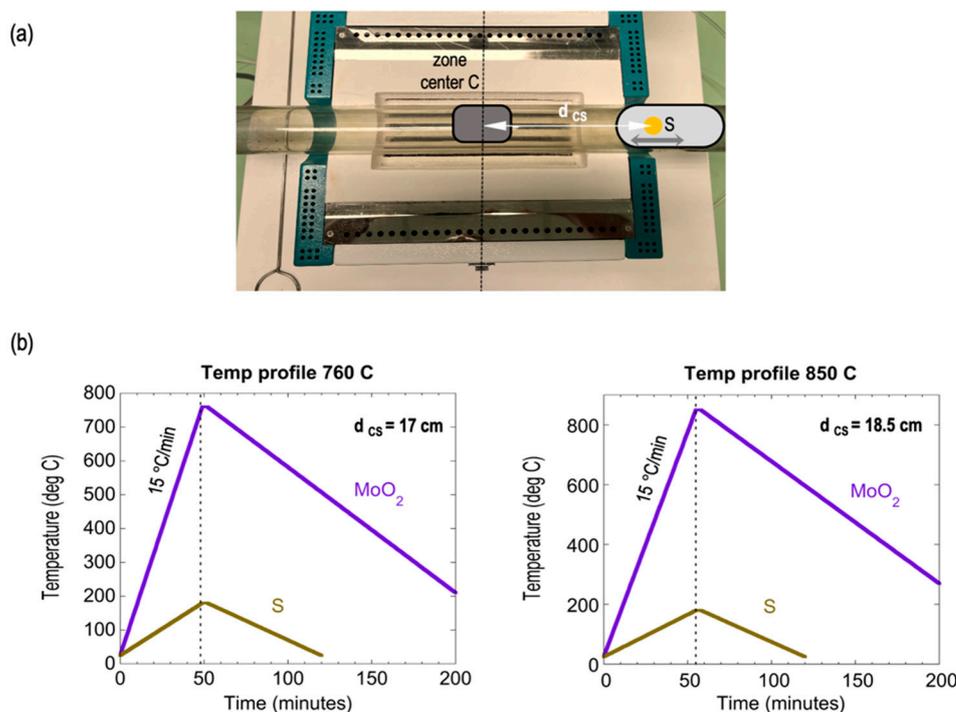
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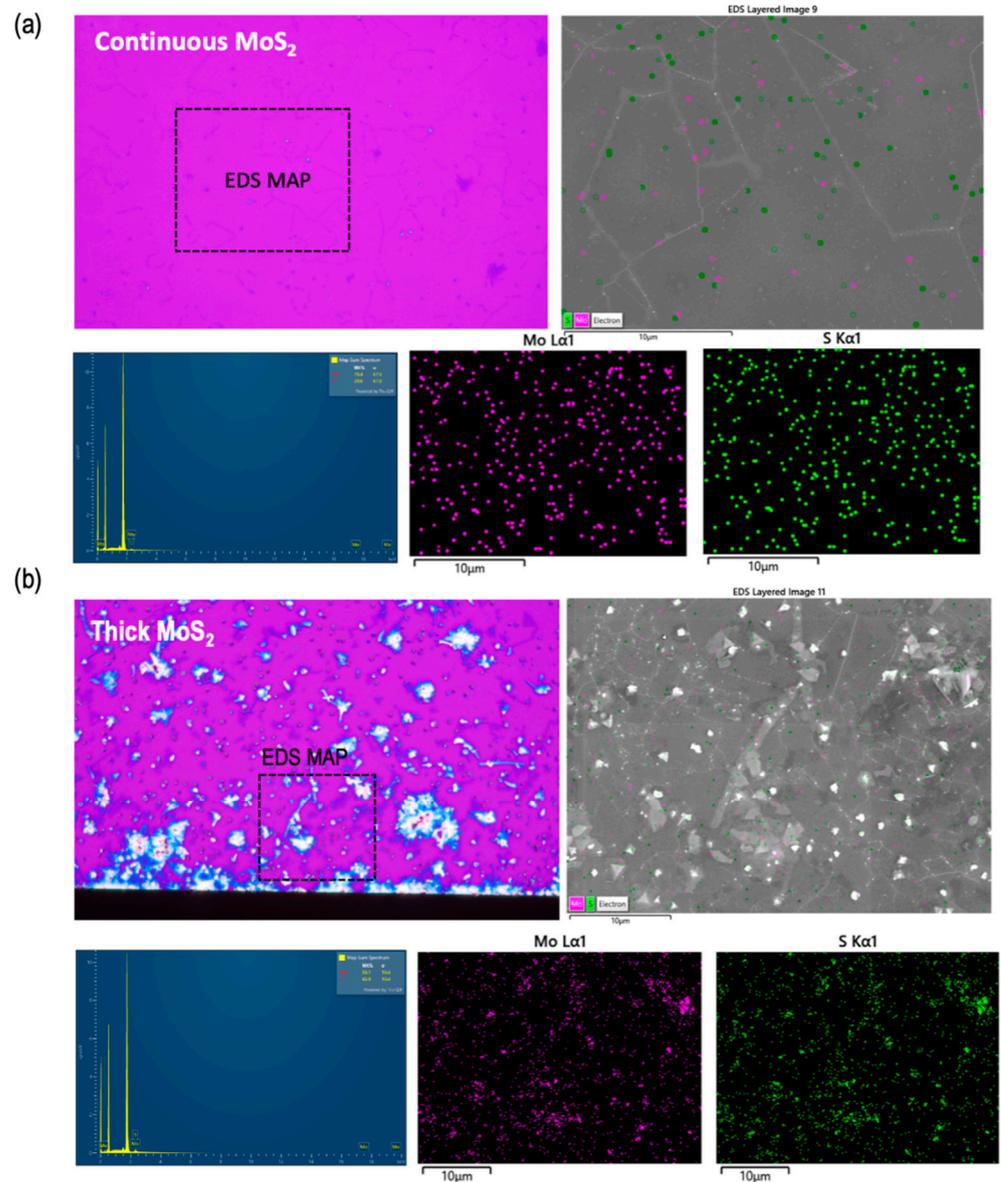
## 1. Growth temperature profiles



**Figure S1.** (a) Photograph of CVD setup. MoO<sub>2</sub> precursors and the growth substrate (separated by ~ 1 cm) are positioned exactly at the center of the heating zone (length 22 cm). The distance  $d_{cs}$  of the Sulphur from the zone center is  $17 \pm 1.5$  cm depending on the set MoS<sub>2</sub> growth temperature, so as to achieve an approximately fixed S zone temperature of 180 °C in all growth experiments. For experiments conducted at 650 °C, 760 °C and 850 °C,  $d_{cs} \sim 15.5$  cm, 17 cm and 18.5 cm, respectively. These values are calibrated prior to conducting the actual growth experiments by externally measuring the crucible temperature using an infrared temperature sensor while the furnace is held at the respective temperatures. (b) Temperature profile of the MoO<sub>2</sub> zone and the expected profiles for the S zone, shown for the cases of growth at 760 °C and 850 °C.

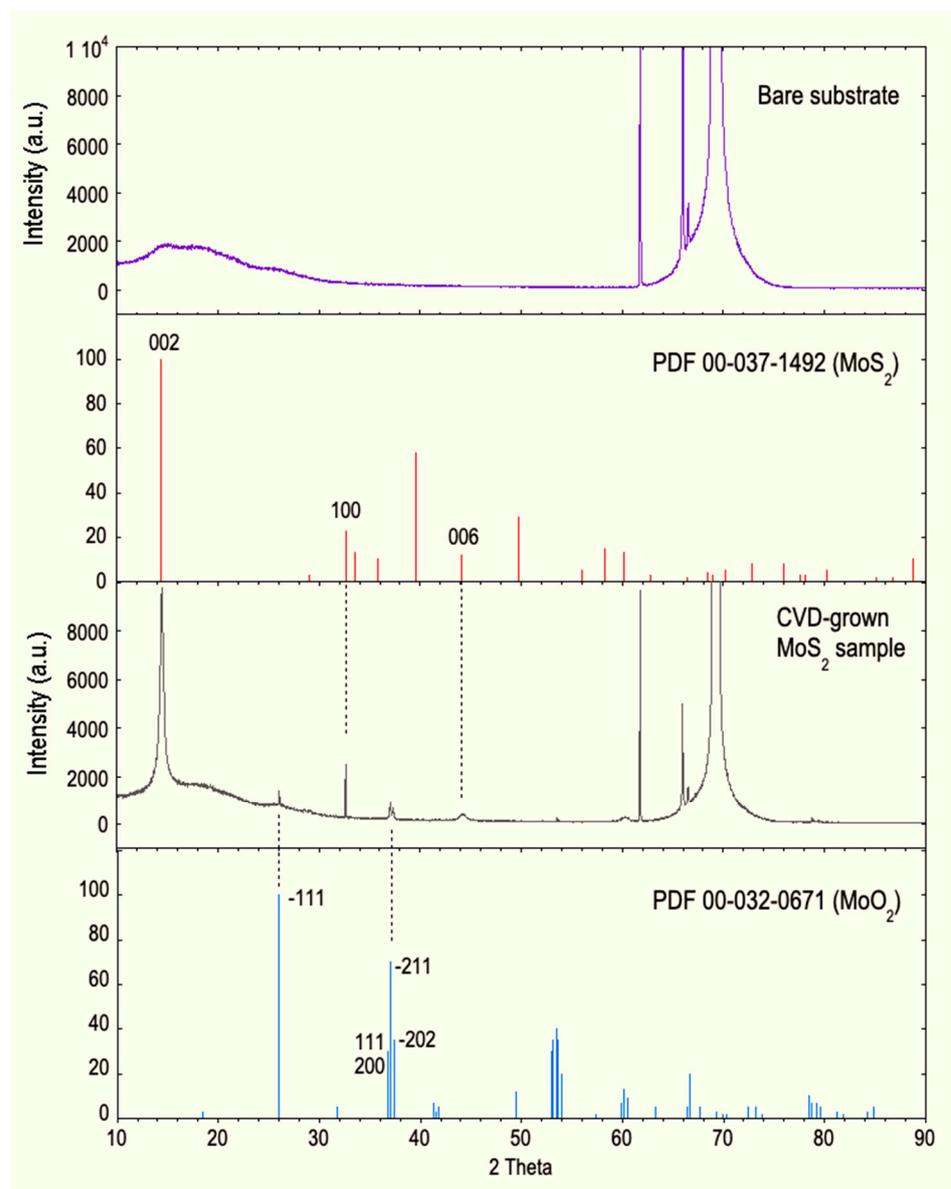


### 3. Elemental Mapping results



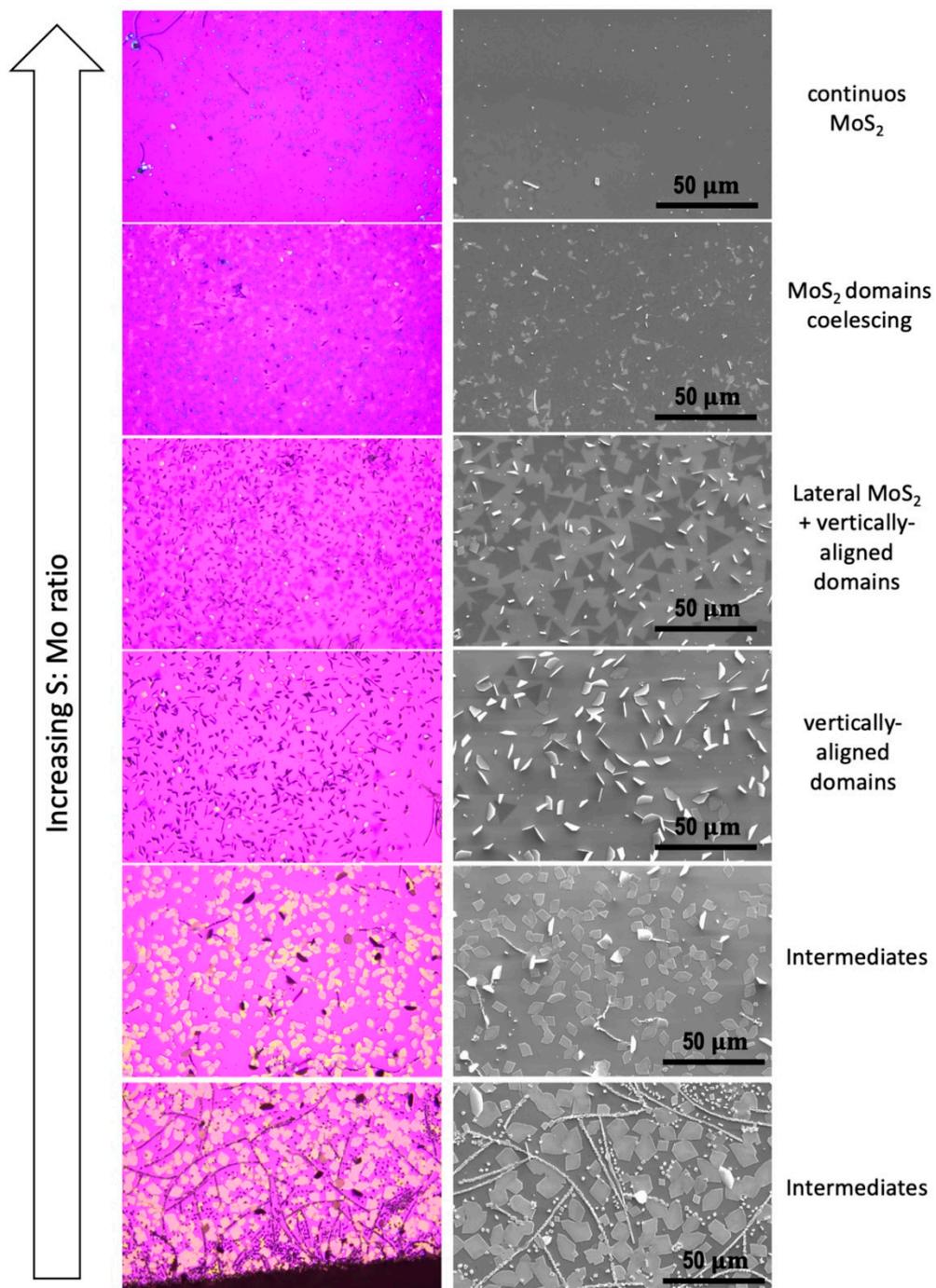
**Figure S3.** Elemental mapping (EDS) results obtained from (a) continuous MoS<sub>2</sub> formed in outer zones and (b) Thick MoS<sub>2</sub> product formed near the wafer edge. Elemental maps of Mo and S confirm the presence of MoS<sub>2</sub>. Stoichiometric ratio from EDS have not been reported in this work as they are erroneous, owing to the very weak signal (in comparison to Si and O) as well as overlapping Mo and S energies, which is a known issue.

#### 4. XRD results



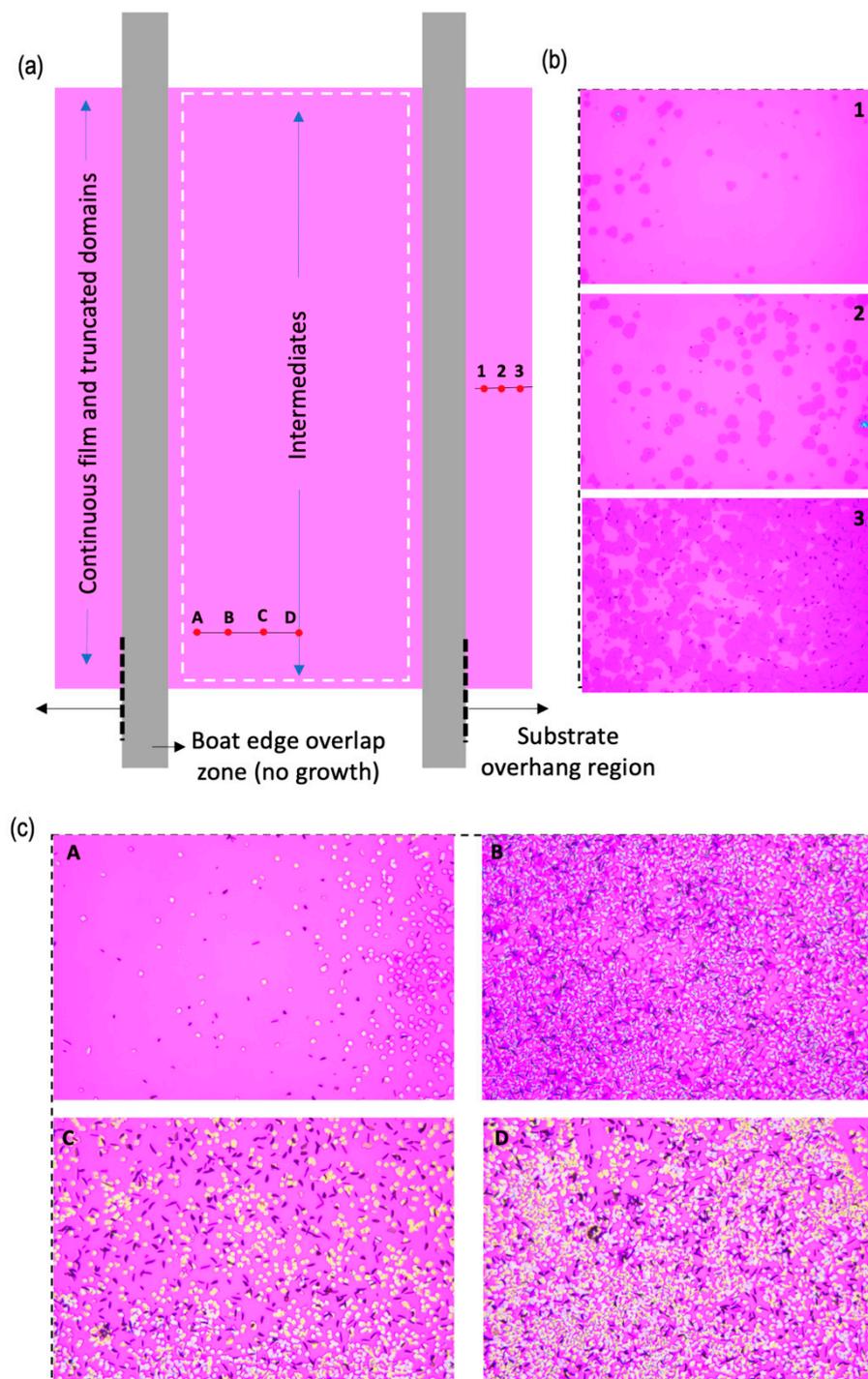
**Figure S4.** XRD spectra obtained from the CVD-grown MoS<sub>2</sub> sample grown at 760 °C. XRD spectrum of the bare substrate (Si/300 nm SiO<sub>2</sub>) was also collected which allowed for a clearer analysis of the MoS<sub>2</sub> diffraction peaks. All observed peaks are compared against XRD database of MoS<sub>2</sub> (PDF # 00-037-1492) and MoO<sub>2</sub> (PDF # 00-032-0671), the precursor used for MoS<sub>2</sub> growth in this study. Two main diffraction peaks are detected at  $2\theta$  positions of 14.378° and 32.677°, and correspond respectively to the (002) and (100) lattice planes of 2H-MoS<sub>2</sub> phase. The 002 orientation is dominant as evidenced by the comparatively large intensity. In addition, weak second-order reflections of the (006) plane is also detected at  $2\theta$  position of 44.152°. Absence of other higher order peaks corresponding to the bulk MoS<sub>2</sub> reference are not detected, which may imply the overall thin film nature of the materials. In addition to the MoS<sub>2</sub> peaks, clear but relatively small intensity reflections that correspond to the reference peaks of MoO<sub>2</sub> are detected at the  $2\theta$  position of 18.45° (-111) and a collection of closely spaced reflections (200, 111, -211, -202) are also detected at a mean position ~37°. We believe these to arise from trace amounts of unreacted precursor left behind upon cessation of CVD growth, as materials freeze at this stage and cannot be driven to complete reaction. The lower value (20:1) of S: Mo loading ratio used in this study in comparison to our previous study (100:1), may also be a contributing factor. The parabolic growth zone pattern suggests that the majority of these products may be present at the wafer edge.

## 5. Morphology evolutions at 800 °C



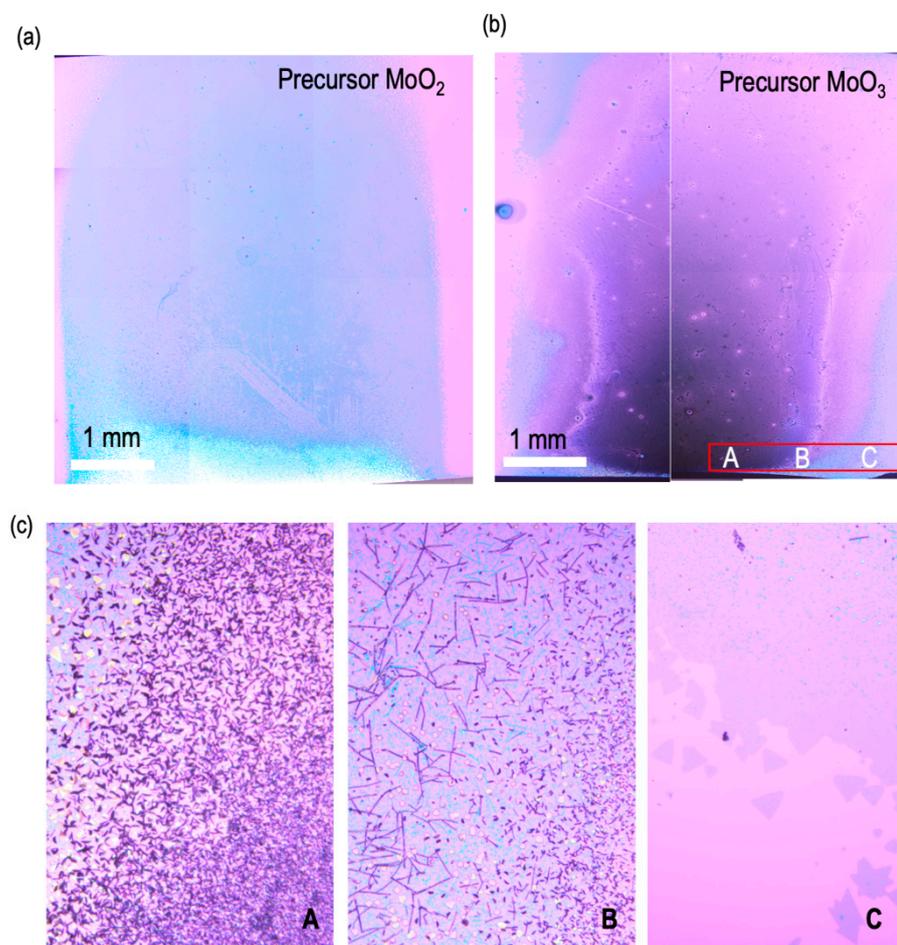
**Figure S5.** SEM images capturing varying growth morphology at 800 °C. The morphology is seen to evolve from intermediates to MoS<sub>2</sub> with transformations in between. Just prior to the formation of lateral MoS<sub>2</sub> domains, the dominance of vertically-oriented structures are also observed in the SEM micrographs.

## 6. Morphology evolutions at 850 °C



**Figure S6.** General results of growth morphologies at the highest investigated growth temperature of 850 °C. Substrate is largely comprised of intermediate states, while the overhanging regions of the substrate show the formation of truncated MoS<sub>2</sub> domains and continuous films forming from the coalescence of the individual domains.

## 7. Precursor comparison



**Figure S7.** A direct comparison of MoS<sub>2</sub> growth obtained from using either (a) MoO<sub>2</sub> or (b) MoO<sub>3</sub> as the precursor. A gas flow rate of 30 sccm is used for this experiment. Clear differences in growth pattern and morphologies are noted, which can be explained as a result of dramatic difference in diffusion behaviour of these two precursor materials. These could arise from the significantly different vaporization temperatures as well as the different reaction pathways followed to form MoS<sub>2</sub>. These differences could imply the reason behind the requirement of large S: Mo loading ratios used for achieving ideal MoS<sub>2</sub> growth from MoO<sub>3</sub>, as is often reported in the literature.