OSS-CVD for MoS_2 Monolayers: A New Approach Utilizing Oxide-Scale Sublimation of Molybdenum

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Two-dimensional (2D) transition metal dichalcogenides (TMDCs) have shown tremendous potential for engineering next-generation electronics and optoelectronics due to their unique properties. Synthesis of single-crystal TMDC layers with desired properties is highly required but remains very challenging. Diverse techniques have been developed to form TMDCs layers, such as exfoliation from bulk crystals, molecular beam epitaxy, chemical vapor deposition (CVD). Amongst them, the powder vaporization CVD is widely used because of the low-cost and straightforward equipment requests. However, due to the limited controllability of precursor supply during the process, it is difficult to achieve uniform films and versatile heterostructures reproducibly, leading to significant hurdles for future industrial large-scale production.

In this study, we propose and demonstrate an oxide scale sublimation CVD (OSS-CVD) technique to synthesize MoS_2 monolayer. Figure 1(a) shows a schematic of the OSS-CVD system, where volatile MoO_3 is generated in a separated source zone by feeding mixed oxygen and nitrogen gases over Mo metal placed in the upstream chamber. After that, MoO_3 and H_2S are separately transported to substrate surface for MoS_2 deposition. Compared with the conventional powder source method, the OSS-CVD enables (1) to rapidly switch and adjust the supply of MoO_3 by managing the injection of O_2, resulting in good controllability of precursor's partial pressure and ratio, and (2) to ensure the successive MoS_2 deposition by preventing Mo precursor from sulfurization for steady sublimation.

Proof-of-concept growth experiments were carried out on 2 × 2 cm^2 glass substrates, similar to our previous study [1]. During the process, the temperature of 700˚C and the pressure of 20 Torr were maintained. After 3 min deposition, optical microscopy (OM) image clearly showed triangular MoS_2 domains with arbitrary rotation. At longer growth durations the enlarged triangular domains with an edge length of around 10 μm were visible (Fig. 1(b)), and they ultimately coalesced forming continuous MoS_2 layer for 15 min growth. Representative spectra from Raman and photoluminescence measurements (Fig. 1(c)) revealed that high-quality MoS_2 monolayers were successfully achieved. The present results clearly indicate this developed OSS-CVD technique has a great potential in yielding crystalline MoS_2 monolayer and even growing other 2D TMDCs layers with markedly enhanced controllability of precursor supply.

Reference: [1] Y. Sakuma *et al.*, The 80th JSAP Autumn Meeting 2019, 21a-PB1-68.
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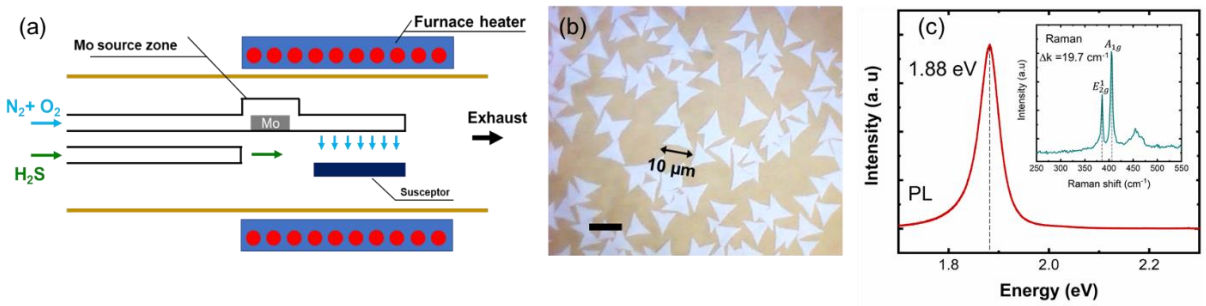

Fig. 1. (a) Schematic of OSS-CVD. (b) OM image of triangular MoS_2 domains on glass substrate. Scale bar is 10 μm. (c) PL and Raman (inset) spectra of the as-grown MoS_2 at 300 K, confirming its monolayer feature.