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Pulse-Flow-Assisted Chemical Vapor Deposition Growth of Large-Area Two-Dimensional $MoSe_2$

MUKESH KUMAR[1], SUMANTA SEKHAR SAMAL[1] and PRAMODA K. NAYAK[1,2, †]

[1]*Centre for Nano and Material Sciences, Jain (Deemed-to-be University), Jain Global Campus, Kanakapura, Bangalore 562112, Karnataka, India.*
[2]*Department of Physics, 2D Materials Research and Innovation Group, Indian Institute of Technology Madras, Chennai 600036, India.*

[†]*Email: pramoda.nayak@jainuniversity.ac.in*

Abstract: Scalable synthesis of large-area, high-quality two-dimensional (2D) transition metal dichalcogenides (TMDs) remains a critical challenge for their integration into next-generation electronic and optoelectronic devices. In this work, we demonstrate a pulse-flow-assisted chemical vapor deposition (CVD) strategy for the controlled growth of large-area monolayer MoSe□. The approach involves periodic switching of the carrier gas between ON and OFF states during growth, which enables precise regulation of precursor supersaturation, mass transport, and surface reaction kinetics. This dynamic control effectively suppresses excessive nucleation while promoting sustained lateral domain growth, in contrast to conventional continuous-flow CVD conditions. Optical microscopy analysis reveals a significant enhancement in monolayer domain sizes, reaching lateral dimensions of approximately 50–100 µm on various substrates, indicating the robustness and substrate versatility of the method. Raman spectroscopy confirms the characteristic vibrational modes of monolayer MoSe□ with minimal strain and disorder, while photoluminescence measurements exhibit strong and uniform emission, further validating the high crystalline and optical quality of the as-grown films. Importantly, the pulse-flow technique is promoter-free, simple to implement, and readily scalable, making it highly compatible with existing CVD systems. This work provides an effective and general pathway for the controlled synthesis of large-area MoSe□, advancing the scalable fabrication of high-performance 2D materials for electronic and optoelectronic applications.

Keywords: Pulse flow, **C**hemical vapor deposition, Large area growth, 2D $MoSe_2$.

1. INTRODUCTION

Two-dimensional (2D) transition metal dichalcogenides (TMDs) have attracted extensive attention due to their atomically thin nature, tunable electronic structure, strong light–matter interaction, and excellent mechanical flexibility. Unlike graphene, many semiconducting TMDs exhibit intrinsic band gaps,

making them promising candidates for applications in nanoelectronics, optoelectronics, valleytronics, and energy-related devices. Among them, molybdenum diselenide ($MoSe_2$) is particularly appealing owing to its direct bandgap in the monolayer limit, high carrier mobility, and strong excitonic response, which enable efficient charge transport and light emission[1].

To realise practical applications, scalable synthesis of high-quality, large-area $MoSe_2$ with controlled thickness and uniformity is essential. CVD has emerged as one of the most effective and widely adopted techniques for the growth of 2D TMDs, including $MoSe_2$, due to its compatibility with wafer-scale production and precise control over growth parameters. Significant progress has been made in CVD synthesis of $MoSe_2$ by optimizing precursor chemistry, growth temperature, carrier gas composition, substrate selection, and growth promoters. However, conventional continuous-flow CVD often suffers from high nucleation density, limited lateral domain size, and non-uniform growth, primarily due to uncontrolled precursor supersaturation and rapid nucleation during the growth process. These challenges restrict the realization of large single-crystal domains and hinder scalable integration[2,3].

Recent studies show that growth kinetics and vapor-phase transport strongly influence nucleation and lateral growth in CVD-grown TMDs; however, **carrier gas flow modulation remains largely unexplored**, despite being one of the most direct parameters controlling precursor flux and surface supersaturation, particularly for $MoSe_2$. This approach offers a simple route to suppress excessive nucleation without the use of chemical additives[4]. In this work, we introduce a **pulse-flow–assisted CVD strategy** for the growth of large-area monolayer $MoSe_2$. Periodic ON–OFF switching of the carrier gas regulates precursor supersaturation and surface reaction kinetics, resulting in reduced nucleation density and enhanced lateral domain expansion compared to conventional continuous-flow growth. Optical microscopy, Raman, and photoluminescence analyses confirm a pronounced increase in domain size and improved uniformity.

2. EXPERIMENTAL DETAILS

2.1. Synthesis of Monolayer $MoSe_2$ by Pulse-flow method

Monolayer $MoSe_2$ was synthesized by CVD in a horizontal dual-zone quartz tube furnace (55 mm inner diameter). High-purity MoO_3 (99.97%) and Se (99.998%) powders were used as molybdenum and selenium precursors. Approximately 5 mg of MoO_3 was placed in the high-temperature zone, while 50 mg of Se was positioned in the low-temperature zone, maintaining a source separation of ~30 cm. Pre-cleaned SiO_2/Si substrates (300 nm oxide) and polished sapphire substrates were placed face-down ~2 cm downstream of the MoO_3 source (Figure 1(a)).

The MoO_3 zone was heated to 830 °C at a ramp rate of 15 °C min^{-1}, while the Se zone reached 330 °C approximately 2 min earlier. The growth was carried out for 30 min at the target temperatures (Figure 1b). During synthesis, Ar and H_2 were used as carrier and reducing gases, with flow rates precisely controlled using mass flow controllers. To investigate the effect of precursor delivery dynamics, a pulse-modulated gas flow strategy was employed by periodically switching the carrier gas 5 min ON/5 min OFF during the growth stage (Figure 1(c)). For comparison, $MoSe_2$ was also grown under continuous gas flow conditions with all other parameters kept identical. After growth, the furnace was naturally cooled to room temperature under Ar flow.

2.2. Characterization techniques

The surface morphology and lateral domain size of the as-grown $MoSe_2$ were examined using Olympus optical microscope. Photoluminescence (PL) and Raman spectroscopy measurements were performed using a HORIBA LabRAM in a confocal microscopy backscattering geometry. Continuous wave (CW) laser of energy 2.33 eV and optical power 1mW were used as excitation source. The spot size of the laser beam was 1 μm.

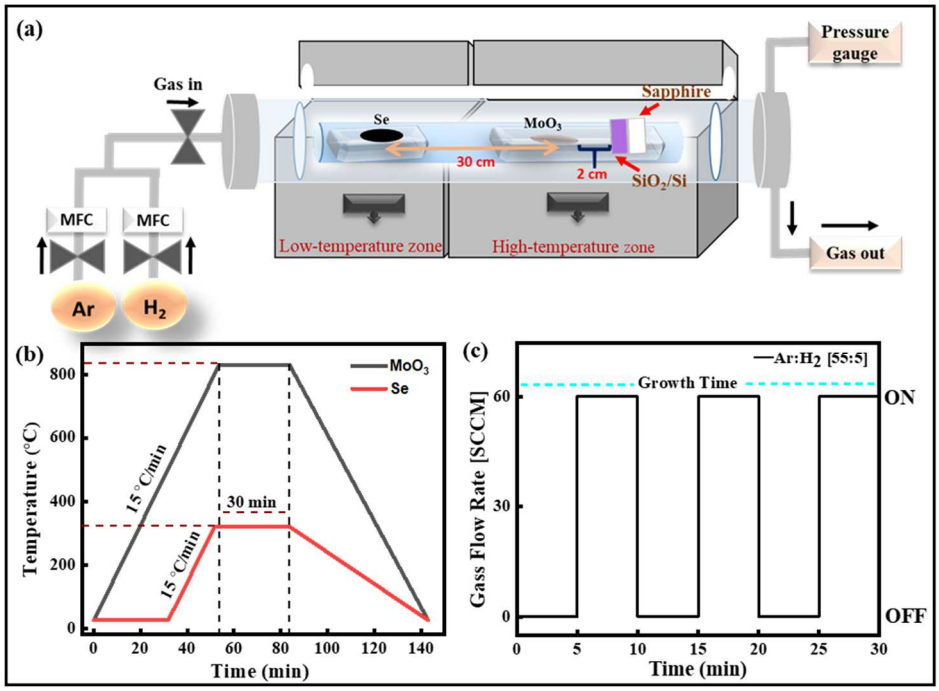

Figure 1 (a) Schematic illustration of the dual-zone APCVD setup for $MoSe_2$ growth. (b) Temperature vs Time profile for the growth of $MoSe_2$. (c) Schematic of Ar/ H_2 pulse-modulated gas flow during the CVD process.

3. RESULTS AND DISCUSSION

2D $MoSe_2$ flakes were synthesized using CVD without and with pulse-modulated gas flow as stated in experimental section. Figure 2 compares the surface morphology of CVD-grown $MoSe_2$ flakes synthesized without and with pulse-modulated gas flow. As shown in Figure 2(a), $MoSe_2$ grown on sapphire under continuous (non-pulsed) gas flow exhibits triangular domains with relatively small lateral sizes and high nucleation density, indicating rapid precursor supersaturation and uncontrolled nucleation during growth. In contrast, Figure 2(b) shows $MoSe_2$ grown on sapphire using pulse-modulated gas flow with 5 min interval, where a clear reduction in nucleation density and a significant increase in lateral domain size are observed. Similar improvements are seen on SiO_2/Si substrates under pulse-flow conditions (Figure 2(c)), demonstrating that the pulse-growth strategy is effective across different substrates. $MoSe_2$ domains in Figure 2(d) exhibit well-defined edges and substantially larger lateral sizes; notably, these domains were formed near the edge of the sapphire substrate, where modified mass-transport conditions and reduced effective nucleation density favor lateral growth. These observations indicate that periodic ON–OFF modulation of the carrier gas effectively regulates precursor delivery, suppresses secondary nucleation, and promotes kinetically controlled lateral growth, consistent with previous reports on temporally regulated CVD growth of 2D systems[5,6].

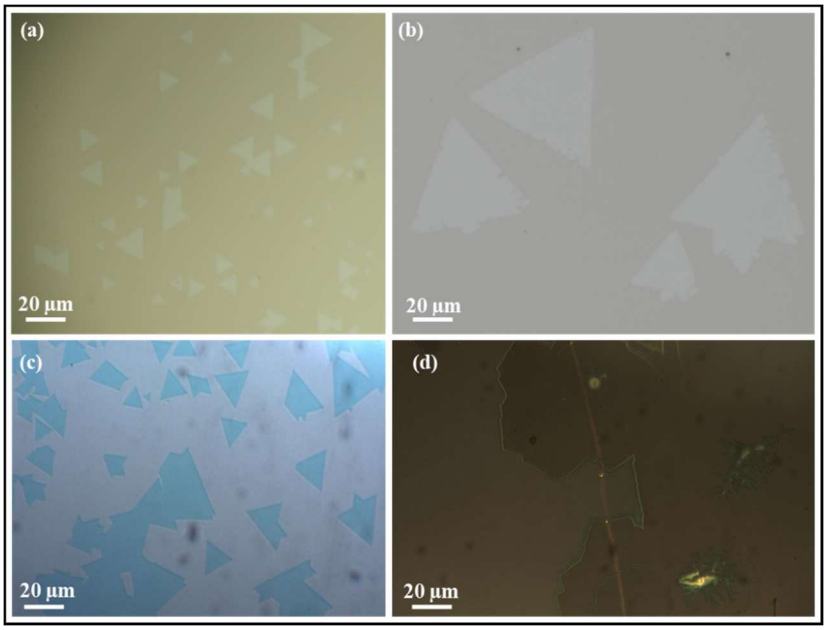

Figure 2. Optical microscopy (OM) images of CVD-grown $MoSe_2$ (a) on sapphire under continuous gas flow, (b) on sapphire under pulse-modulated gas

flow, (c) on SiO_2/Si with pulse-modulated gas flow, and (d) on sapphire with pulse-modulated gas flow.

The material quality of the pulse-grown $MoSe_2$ was evaluated using Raman and PL spectroscopy as non-destructive probes (Figure 3). Raman and PL measurements were performed on a representative triangular domain with an edge length of ~50 μm (inset of Figure 3(a)) using a 532 nm excitation laser. The Raman spectrum (Figure 3(a)) exhibits two characteristic vibrational modes at approximately 240 cm^{-1} (A_{1g}) and 287 cm^{-1} (E^1_{2g}), corresponding to out-of-plane and in-plane phonon vibrations, respectively. The frequency separation of ~47 cm^{-1} is consistent with reported values for monolayer $MoSe_2$, confirming the monolayer nature of the grown films[7].

The PL spectrum (Figure 3(b)) shows a strong emission peak centered at ~1.57 eV, attributed to the direct bandgap transition of A excitons in monolayer $MoSe_2$. The sharp Raman features and intense PL emission indicate good crystallinity and low defect density[8], demonstrating that pulse-modulated gas flow enhances domain size without compromising optical quality. Overall, these results establish pulse-modulated gas flow as a simple and scalable strategy to control nucleation density and achieve large-area monolayer $MoSe_2$ growth, offering a broadly applicable route for high-quality 2D TMD synthesis.

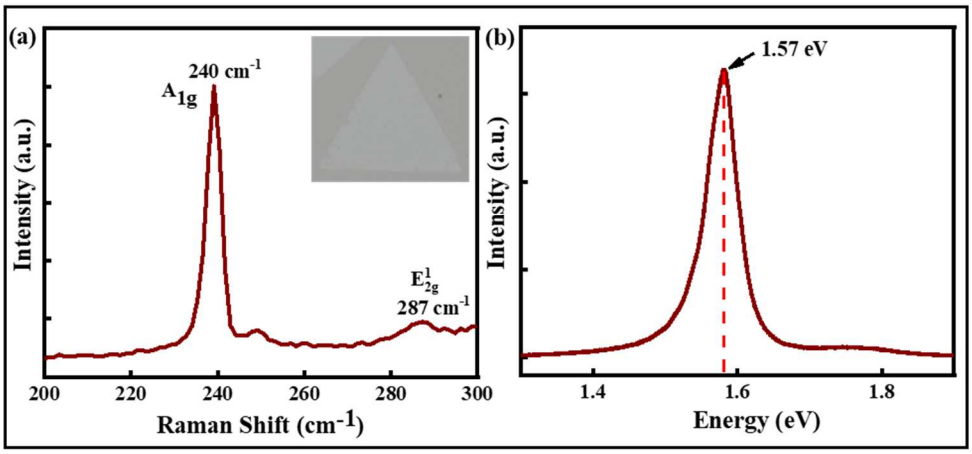

Figure 3. (a) Raman spectrum of the synthesized $MoSe_2$ layer, displaying characteristic peaks at ~240 cm^{-1} (A_{1g}) and ~287 cm^{-1} (E^1_{2g}). (b) PL spectrum of the grown $MoSe_2$ layer that exhibits a strong emission peak at ~1.57 eV.

4. CONCLUSION

In summary, we have successfully demonstrated a pulse-flow-assisted CVD strategy for the controlled growth of large-area monolayer $MoSe_2$ on multiple substrates. By periodically modulating the precursor and carrier gas flow, the pulse-flow technique effectively suppresses excessive nucleation density while promoting lateral domain expansion, enabling the formation of monolayer $MoSe_2$ domains with lateral sizes in the range of 50–100 μm. Compared to conventional continuous-flow CVD growth, this approach yields significantly larger and more uniform domains without compromising structural integrity or optical quality. Owing to its simplicity and effectiveness, the pulse-flow-assisted CVD approach can be readily extended to other TMDs and layered van der Waals materials. Overall, this work provides a practical pathway toward the scalable production of high-quality, large-area 2D TMDs, paving the way for their reliable implementation in next-generation electronic, optoelectronic, and flexible device applications.

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