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Minimizing the Water Effect in Synthesis of High-Quality Monolayer MoS₂ Nanosheets: Implications for Electronic and Optoelectronic Devices

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Cite This: ACS	5 Appl. Nano Mater. 2021, 4, 8094	-8100	Read Online	
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ABSTRACT: Tw information techn	o-dimensional materials are nology, energy, and sensing	promising building bloc g devices. However, cor	cks for ntrolled	Minimize water effect

information technology, energy, and sensing devices. However, controlled synthesis of high crystallinity materials is still a challenge, limiting their further developments. Here, we report the effect of water in synthesizing monolayer MoS_2 by chemical vapor deposition. We show that, from both experiments and simulation, the presence of water can reduce the precursor flux and diffusivity, resulting in altered growth kinetics and different morphologies. By using an unglazed crucible approach, the growth can be restored to the kinetic Wulff shape regime, thereby providing high crystallinity monolayer crystals with enhanced reproducibility.



KEYWORDS: two-dimensional materials, MoS₂, flux, diffusivity, phase-field simulation

INTRODUCTION

The discovery of monolayer graphene in 2004 opened the door for studying materials in a new dimensionality,¹ namely, two-dimensional (2D) materials. Since then, many new 2D materials have been discovered and the previously overlooked ones have been re-examined.² In particular, because of the rich chemical composition and the unique physical and chemical properties, such as indirect to direct band gap transition from the bulk to monolayer,³ inversion symmetry breaking-induced valley selectivity,⁴ and abundant surface-active sites for chemical reactions,⁵ transition metal dichalcogenides (TMDs) and related heterostructures have recently attracted much attention in 2D materials research. Consequently, TMDs have inspired countless possibilities in enabling high-efficiency photovoltaic devices,^{6,7} building next-generation transistors,⁸⁻¹⁰ and serving as catalysts for generating hydrogen as fuels,¹¹ thus becoming the new frontier for different research fields.

Despite the great promise of 2D TMDs, the preparation of high-quality materials on a large scale with a controlled layer number, however, is still at its infancy. This immaturity of materials preparation has been a bottleneck limiting the deployment of 2D TMDs in technological valuable areas and the continuous explorations in fundamental research. Among the techniques developed to address this challenge, chemical vapor deposition (CVD), including metal–organic CVD (MOCVD), is widely regarded as an important method because it strikes a better balance between crystal quality and cost, as evidenced by its success in producing large-scale graphene and conventional semiconductors such as Si, GaAs, etc. Much effort has been put into the controllable synthesis of 2D TMDs. For instance, employing different Mo and S sources in powder-based CVD^{12,13} and precursors in gas-based MOCVD, controlling the nucleation density and nucleation location,¹⁴ understanding the temperature and pressure effects,¹⁵ and applying different substrates^{12,16} are all important progress. While the advancements from different approaches have deepened our understanding of 2D TMD synthesis by CVD, they have also revealed the complexities involved in the synthesis process. Slight alternations in growth conditions can drastically change the final growth outcome. As a result, there are substantial variations in the crystal morphology, monolayer coverage, and crystal quality from lab to lab and throughout the literature. Therefore, it is highly desirable to understand the critical synthesis parameters involved in the growth and achieve better control over these parameters.

Herein, we show that water has a drastic effect on the growth of monolayer MoS_2 . Water present in MoO_3 will not only hinder the evaporation of MoO_3 , decreasing the overall coverage of MoS_2 on the substrate, but also reduce the diffusion rate of Mo species on the growth substrate and change the growth kinetics. More importantly, we discovered that by adopting an unglazed crucible, water can be effectively absorbed by capillary action, thereby achieving highly

 Received:
 May 24, 2021

 Accepted:
 July 29, 2021

 Published:
 August 6, 2021





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reproducible monolayer MoS₂ single crystals with high quality and a large crystal size.

RESULTS AND DISCUSSION

In our experiment, MoO₃ powder (99.97%, see the Methods section in the Supporting Information) was used as a source of molybdenum to synthesize monolayer MoS_2 (Scheme 1). We

Scheme 1. Schematic Illustration of the CVD System to Produce 2D MoS₂ Crystals^a



find that MoO₃ powder absorbs water strongly even at room temperature. Figure S1 shows the thermogravimetric analysis (TGA) result of the MoO₃ powder stored under ambient conditions. The total weight loss measured by TGA is about 0.028 mg, 0.175% of the initial weight of the MoO_3 powder. Being ultrahigh pure, this 0.175% weight loss is likely due to the water absorbed in the MoO₃ powder. In our subsequent

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monolayer MoS₂. Figure 1 presents the results of MoS₂ growth using MoO₃ powder with different water concentrations. In the control group, we used MoO₃ stored under ambient conditions with 0.175% water concentration. After conducting a normal growth, no noticeable material on the growth substrate was observed in a representative optical image in Figure 1a. Next, in the first experimental group, we decreased the water concentration by heating the MoO₃ powder in a vacuum oven at 120 °C for 6 h prior to the growth, while all of the other parameters were kept unchanged. The water concentration in this case was estimated to be $\sim 0.15\%$ (see Figure S2 for more details). This experiment resulted in many micrometer-size particles on the growth substrate, marked by the white circles in Figure 1b. Characterization of these particles by Raman microscopy reveals that they are multilayer MoS₂, with two characteristic peaks located at 379 \mbox{cm}^{-1} for E_{α} in-plane vibration mode and 405 cm^{-1} for A_g out-of-plane vibration mode (Figure 1c). The spacing between these two peaks is ~26 cm⁻¹, in agreement with the multilayer MoS₂ observed by Raman microscopy.^{14,17} The atomic force microscopy (AFM) image in Figure 1d further confirms the multilayer nature, with thicknesses ranging from 3 to over 30 nm. Compared to the control group, the results of the first experimental group indicate that the water concentration in the MoO₃ powder



Figure 1. (a) Optical image of the growth result using MoO_3 powder with a 0.175% water content. There is no noticeable MoS_2 growth in this experiment. Scale bar: 200 μ m. (b) Particle-like growth can be observed after heating the MoO₃ powder at 120 °C for 6 h. The white circles show the MoS₂ particles in the optical image. Scale bar: 10 μ m. (c) Raman spectrum of the particle-like MoS₂ in (b). The spacing between E_g in-plane vibration mode and A_x out-of-plane vibration mode is 26 cm⁻¹, indicating multilayer growth. (d) AFM image of the particles in (b). The heights of the particles vary from several nanometers to over 30 nm. Scale bar: 5 μ m.





Figure 2. (a) Optical and (c) SEM image of the MoS₂ growth using MoO₃ powder directly heated in the growth tube for 2 h at 200 °C without exposure to the ambient air. Many truncated triangular monolayer single crystals can be observed in this experiment. The black crystals are MoS₂ grown vertical to the substrate. Scale bar in (a): 10 μ m. Scale bar in (c): 5 μ m. Scale bar in the inset: 1 μ m. (b) Overall MoS₂ coverages and moisture levels under different experimental conditions. The coverage increases from 0% to over 60% as we decrease the moisture level from 0.175% to 0.018%. (d) Normalized PL spectrum of the monolayer single crystals with intensity over 80, indicating the direct band gap nature of the material.

does play a role in the synthesis of MoS_2 . By decreasing the water concentration in the MoO_3 powder precursor from 0.175% to 0.15%, the yield and coverage of MoS_2 rise from nearly 0 to 0.9%. However, no monolayer MoS_2 was detected in the first experimental group.

To further decrease the water concentration, in the second experimental group, we heated the MoO₃ powder directly in the growth tube furnace without exposing it to air during the sample loading procedure (in contrast to the first experimental group). This practice led to a much lower water concentration, estimated to be 0.018% from TGA data (Figure S3), after heating for 2 h at 200 °C. We note that higher temperature should be a more significant factor than air exposure in this experiment. As a result, a significant enhancement of the MoS_2 yield was observed from the optical image (Figure 2a). The overall coverage changes from 0.9% in the first experimental group to $\sim 61.3\%$ in the second experimental group, a 67-fold increase. Figure 2b summarizes the MoS₂ coverage at different water concentrations. Reducing the water concentration in the MoO_3 precursor from 0.175% to 0.018%, the coverage of MoS_2 on the growth substrate increases from 0% to over 61% accordingly.

Not only the overall coverage increase was observed, but many monolayer crystals started to appear in the second experimental group as well. The scanning electron microscopy (SEM) image in Figure 2c shows that all crystals adopted truncated triangular shapes, with the size of each crystal being about 1 μ m (Figure 2c, inset). Very high photoluminescence (PL) intensity with normalized intensity (using silicon peak intensity as a reference) over 80 further confirms that these crystals are direct band gap monolayer MoS₂. The emergence of monolayer MoS₂ is in sharp contrast to the multilayer particle growth alone in the first experimental group, suggesting that the reduction in the water concentration simultaneously affects the MoO₃ precursor evaporation and the following monolayer formation process.

The above experiments demonstrate that the presence of a small amount of water can drastically affect the growth of monolayer MoS_2 . Lengthy heating at higher temperatures prior to growth is a potential way to further lower the water effect, but it requires increased energy consumption. In our experiment, we found that by modifying the crucible used to carry MoO_3 powder, from a glazed surface to an unglazed surface, the effect of water can be dramatically minimized. Figure 3a displays the optical image of the growth result using an unglazed surface crucible. The MoO_3 powder has the same amount of water concentration, i.e., 0.175%, as in the control group. Triangular MoS_2 crystals with sharp edges are observed



Figure 3. (a) Optical image of the MoS₂ growth using an unglazed crucible as a MoO₃ holder. The MoO₃ powder is the same as in the control group without any further heat treatment. Triangular, sharp single crystal MoS₂ can be observed all over the substrate. Scale bar: 100 μ m. (b) PL map of an individual triangle, showing very high uniformity and intensity. Scale bar: 5 μ m. (c) Raman spectrum with a peak spacing of about 21.2 cm⁻¹ and (d) PL spectrum of the crystals with very high intensity confirm that they are monolayers. SEM surface morphology images of (e) unglazed crucible and (f) glazed crucible. The unglazed crucible has a rougher surface with microsize holes, while there is no noticeable feature in the glazed crucible. Scale bar: 2 μ m.

all over the substrate. The edge length varies from tens of microns to as large as over 100 μ m. Raman spectroscopy and PL confirm that the crystals are monolayers (Figure 3b). Individual PL spectra and PL mapping in Figure 3c,d show strong intensity and high uniformity, with normalized PL intensity over 100, indicative of high crystal quality of the asgrown monolayer MoS₂. X-ray photoelectron spectroscopy of the Mo 3d spectrum of the sample (Figure S4) shows two doublets with a Mo $3d_{5/2}$ binding energy of 230.0 eV, the expected value for the Mo4+ centers in MoS2. The corresponding S 2p spectrum consists of a single doublet with a S $2p_{3/2}$ at a binding energy of 163.0 eV, which is also in agreement with the previous literature.¹⁸ It is worth noting that after adopting an unglazed crucible, the reproducibility of monolayer MoS₂ growth with a similar coverage and size is nearly 100%.

To understand why the significant improvement of MoS_2 growth occurred in the unglazed crucible, we used SEM to examine the surface of the crucibles. The surface textures of these two crucibles are distinctively different: the unglazed crucible has a much rougher surface made of micrometer-sized pores, while the surface of the glazed crucible is very smooth and does not have any noticeable features (Figure 3e,f). It has been reported that porous ceramic structures can be used to extract moisture from soil¹⁹ and as moisture sensors, taking advantage of the easiness of absorbing moisture through their

micropores and capillary tubes.^{20,21} Water absorption by porous ceramics is also very common in real-life experience. For example, the unglazed floor tile has better slip resistance performance than glazed floor tile in a high moisture environment. It is highly plausible that the same principle applies in our experiment: the porous structure of the unglazed crucible absorbs the trace amount of water during the synthesis, and as a result, a large crystal size with sharp edges and highly reproducible monolayer MoS₂ growth can be achieved. The previous study suggested that water could intercalate between MoS₂ and SiO₂ substrates and changed the morphology and friction properties of MoS₂.²² By reducing the water content during the synthesis, our approach could achieve a much cleaner interface between MoS₂ and SiO₂ substrates, mitigating the undesired impact on the crystal morphology and quality.

The drastic changes in the MoS₂ crystal morphology and size at different moisture levels suggest that the rate-limiting step of crystal growth is potentially altered during the CVD synthesis. Generally, the growth of MoS₂ involves several kinetic steps: (i) MoO₃ evaporation into the vapor phase; (ii) deposition of Mo species such as oxysulfides of Mo, MoO_{2-x}S_x (x = 0-2) on the substrate; (iii) surface diffusion of Mo species; and (iv) Mo and sulfur attachment to MoS₂ edges and incorporation into the MoS₂ lattice.^{23,24} When the deposition flux and surface diffusion coefficient of Mo precursors are large



Figure 4. Phase field simulation of the monolayer MoS_2 morphology under different growth conditions. (a) Sharp triangles result from conditions where both flux and diffusivity are large. In this case, the growth is in a thermodynamic equilibrium state. Crystals adopt the most energetically favorable Wulff morphology, i.e., triangles. (b) Crystal morphology when diffusivity is reduced by 10 times. Because it deviates from thermodynamic equilibrium, the morphology becomes a truncated triangle. (c) Similar to (b), the truncated triangle is the result if flux is decreased by about 7 times. (d) Crystal size vs growth time under different growth conditions. At high diffusivity and flux, the final crystal size can be 2 orders of magnitude larger given enough growth time.

enough, the morphology of the MoS₂ crystal reflects the anisotropy of the rate-limiting edge attachment kinetics, which is referred to as the kinetic Wulff shape. The compact triangular MoS₂ obtained at a low moisture level (Figure 3a) is consistent with the Wulff shape when the three (1010) Szigzag or $(10\overline{1}0)$ Mo-zigzag edges grow much slower than other facets.²⁵ However, the development of rounded truncated crystals shown in Figure 2a,c at a higher moisture level suggests that water promotes a more isotropic growth process. It has been reported that MoO₃ can have strong interactions with moisture at certain temperature ($\sim 300 \,^{\circ}$ C), forming hydroxyl groups on the surface of MoO₃ even at very low water partial pressure. The formation of hydroxyl groups likely hinders the reaction between MoO3 and S to form oxysulfides, thereby decreasing the amount of reactive Mo species deposited onto the growth substrate in steps (i) and (ii), which may explain the reduced MoS_2 surface coverage on the substrate. Furthermore, it is found that the hydroxyl groups can be thermally stable on a flat silica surface up to 900 $^{\circ}C$.² The interaction of hydroxyl groups with both Mo species and the growth substrate may slow down the surface diffusion of Mo species with the presence of moisture in step (iii). Since both precursor deposition and surface diffusion on the

amorphous silica substrate have no orientation preference, the effects of water on these two kinetic steps may lead to weaker growth anisotropy as observed in experiments.

Although the deposition flux and the surface diffusivity of Mo species are difficult to assess to make quantitative comparison with experiments, their influence on crystal growth can be qualitatively examined by phase field simulations (see the description in the Supporting Information). Figure 4a-c shows the simulated crystal morphologies under three different conditions, in which MoS₂ grows with large surface diffusivity D and deposition flux F of Mo precursors relative to edge attachment (Figure 4a), with 10 times smaller D (Figure 4b) and \sim 7 times smaller F (Figure 4c). Given sufficient supply of the reactive Mo species at large D and F_1 , MoS₂ growth is controlled by the precursor attachment to crystal edges. This results in triangular crystals as shown in Figure 4a, which require the (1010) S-zigzag edges to grow more than twice as fast as the (1010) Mo-zigzag edges or vice versa. However, such conditions are no longer satisfied at lower D or F because sluggish deposition and surface diffusion reduce not only the overall crystal growth rate but also the relative difference in the growth speed of different facets due to their independence from crystal orientation. Accordingly, the crystal in simulation

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forms truncated triangular shapes with rounded edges, as shown in Figure 4b,c. The simulation results reproduce the shape change with the increasing moisture level as observed in experiments (i.e., Figures 3a and 2c). The smaller D and F also reduce the crystal growth rate by 1 and 2 orders of magnitude, respectively (Figure 4d), which is consistent with the lower surface coverage of monolayer MoS₂ (Figure 2) and the 67fold decrease of the crystal size at the increased moisture level (Figure S5). The similar trends of crystal size reduction and morphology transition observed in experiments when increasing the moisture amount and in simulations when reducing Dand F indicate that the main effect of moisture is likely to alter the rate-limiting steps in MoS₂ growth from the edge reaction to feedstock transport, potentially by impeding surface diffusion or deposition of Mo species on the substrate.

CONCLUSIONS

In conclusion, we find that water present in the MoO_3 powder precursor has a significant effect on the synthesis of monolayer MoS_2 . Not only the deposition flux of the Mo species precursor decreases, but the diffusion rate of Mo species on the growth substrate also reduces. These factors impose strong limitations preventing monolayer MoS_2 crystals from following the kinetic Wulff shape. Decreasing water concentrations in the MoO_3 powder precursor led to MoS_2 crystal morphology evolution from particle-like multilayers to monolayer truncated triangle crystals and finally large monolayer triangular crystals. By adopting an unglazed surface crucible to minimize the water effect and to restore to the kinetic Wulff shape regime, we have achieved highly reproducible, high-yield, and high-quality monolayer MoS_2 growth, providing important insight into well-controlled scalable CVD synthesis of monolayer TMDs.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsanm.1c01361.

Details of experimental methods including material growth, characterization, and phase-field simulation and thermogravimetric analysis, estimation of weight change, and X-ray photoelectron spectroscopy (Figures S1-S5) (PDF)

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Author Contributions

J.Y. and J.L. conceived the experiment. J.Y. synthesized and characterized the MoS_2 crystal by Raman microscopy, PL, and SEM. F.W. and M.T. performed the phase-field simulation and analyzed the data. S.P. contributed to the materials growth. Z.-L.H. contributed to the simulation. J.Y., F.W., and J.L. prepared the manuscript. All authors discussed the results and commented on this manuscript. This manuscript was written through contributions of all authors. All authors have given approval to the final version of this manuscript.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

J.Y. and J.L. acknowledge the financial support from the Welch Foundation (C-1716) and the NSF I/UCRC Center for Atomically Thin Multifunctional Coatings (ATOMIC) under award no. IIP-1539999. J.Y. also acknowledges the support from the Lodieska Stockbridge Vaughn Fellowship from Rice University. F.W. and M.T. were supported by the US Department of Energy (DOE) under project no. DE-SC0019111. Z.H. was supported by the National Natural Science Foundation of China (11802121) and the Natural Science Foundation of Jiangsu Province (BK20180416).

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