Oxygen-Assisted Vapor-Liquid-Solid Growth of 2D Materials

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The vapor–liquid–solid (VLS) method has recently emerged as an advanced synthesis strategy for the rapid and large-scale synthesis of two-dimensional transition metal dichalcogenides (TMDCs)[1][2]. By utilizing an alkali salt to form a eutectic molten flux with metal oxide precursors, the VLS process facilitates the efficient uptake of chalcogenide species, reaching a critical supersaturation point that drives the precipitation of high-quality, solid-phase (M = Mo, W; X = S, Se)[1]. Despite its potential, conventional VLS growth has been largely restricted to the use of solid powder precursors, such as MoO_3(s) or S(s)[2]. These powder-based systems inherently suffer from poor reproducibility, severe reaction chamber contamination, and a lack of compatibility with continuous industrial processes, thereby hindering their practical implementation.

In this study, we report a highly reproducible gas-phase precursor-based VLS synthesis route that overcomes the limitations of solid-source methods. Our approach utilizes metal-organic gas precursors, $Mo(CO)_6$, which is constrained by the production of small-grain films and the requirement for prolonged reaction times [3]. To facilitate the VLS mechanism, O_2(g) is introduced into the system to react with the metal-organic gas, converting $Mo(CO)_6$ into MoO_3. This resulting metal oxide then reacts with NaCl(s) to form a stable sodium molybdate liquid flux. The subsequent dissolution of chalcogenide precursors, H_2S(g), into this liquid phase leads to rapid supersaturation and the precipitation of TMDCs with significantly enhanced crystallinity. Using this method, we successfully synthesized TMDC films with large-domain grain sizes reaching up to ~200μm in 20 min . By establishing a controllable and scalable gas-phase synthesis strategy, this work provides a critical foundation for the industrial-scale production of high-performance 2D materials.

References

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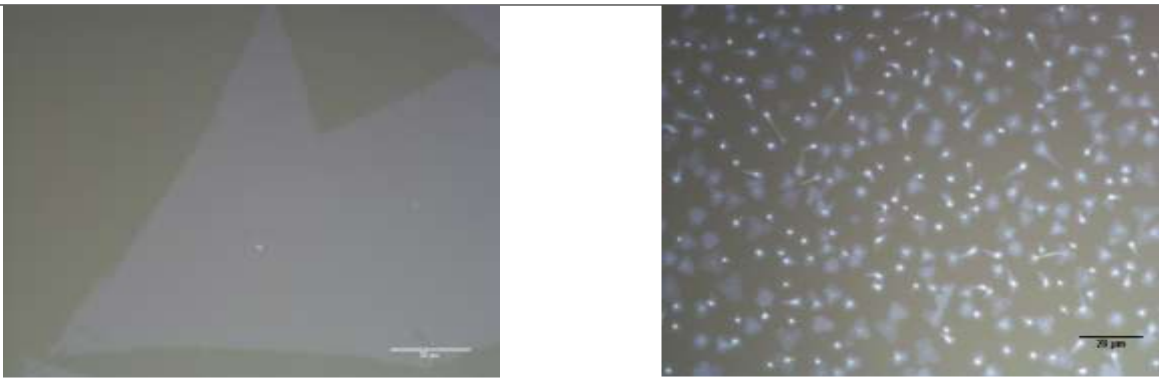
Figures



a. VLS growth with oxygen   b. VLS growth without oxygen

Figure 1: MoS_2 synthesized via (a) oxygen-assisted VLS growth Scale bar 50 μm and (b) VLS growth without oxygen Scale bar 20 μm