Suppression of WSe_2 Bilayer Formation by Water-promoted Multi-step Metal-Organic Chemical Vapor Deposition (MOCVD)

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Abstract
In recent years, transition metal dichalcogenides (TMDC), a mainstream of two-dimensional materials, have garnered substantial attention in global research due to their ultra-thin geometry (few atomic layers) as well as exceptional electrical, optical, and mechanical properties. MOCVD stands out for its high repeatability and scalability and has become an established method for the deposition of two-dimensional materials. However, preparing high-quality large-scale TMDC materials with MOCVD is still challenging. Fundamental limitations are set by a complex trade-off between monolayer (ML) growth time, parasitic bilayer (BL) nucleation before coalescence of the first ML and C incorporation for too high growth temperatures. This presentation will focus on the MOCVD of coalesced WSe_2 ML and the suppression of BL nucleation assisted by water injection. All MOCVD processes are conducted in a commercial AIXTRON MOCVD CCS reactor on standard c-plane sapphire substrates (0.2° off towards m-plane). WCO (W hexacarbonyl, 99.99% purity) and DiPSe (di-isolpropyl Se, 99.999% purity) are used as precursors. The carrier gas is H_2. In our research, we devised a multi-step MOCVD process for enhancing metal adatom migration on WSe_2 ML domains, mitigating parasitic BL nucleation. The process generally starts with a desorption step for 30 min under 150 hPa at 1050 °C in H_2, followed by nucleation stage at 720 °C for 10 min. The second stage is lateral growth, in which the temperature is increased to 820 °C and the flux of WCO is reduced from 100 nmol/min to 50 nmol/min. The WCO flux is gradually decreased in the final stage for achieving a coalesced sample. In some experiments, ultra-pure water is introduced (molar ratio H_2O:W =1:1) as an additive to further enhance the effective migration of W adatoms. A WSe_2 ML sample prepared using the multi-step MOCVD recipe (growth time=45 min) w/ added H_2O exhibited a notable reduction in BL coverage from 15% to 6% compared to a reference run w/o H_2O. ML coverage is even increased by species formerly consumed for parasitic BL nucleation **(Figure 1)**. As a preliminary explanation, we speculate that by adding H_2O, a reverse reaction path is introduced to WCO decomposition/W adatom adsorption, increasing the effective W migration length by contributions from gas phase diffusion. In-situ reflectance, SEM, AFM, Raman/PL spectroscopy and XPS are used for further analysis.
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Figures

	Multi-step process without Water	Multi-step process with Water
Monolayer Coverage [%]	99	99.9
Bilayer Coverage [%]	15	6

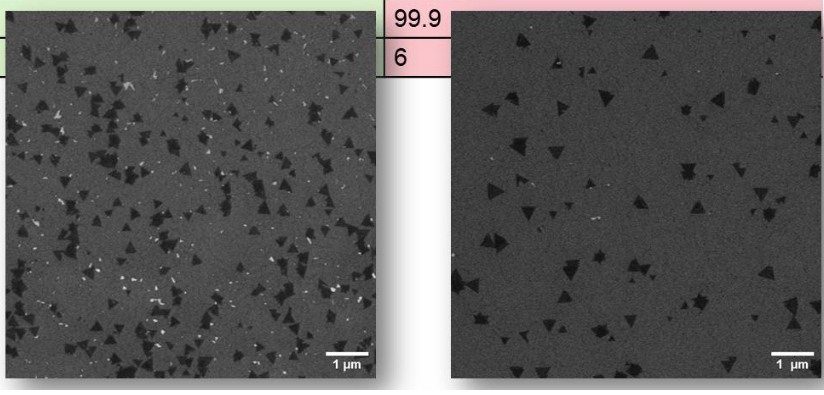

Figure 1: SEM images and analysis of WSe_2 samples w/o and w/ water.