Abstract

Two-dimensional molybdenum disulfide (2D-MoS_2) is considered a potential candidate for futuristic electronic and optoelectronic applications owing to its fascinating properties such as high mobility, tunable bandgap, high mechanical strength high on/off ratio and flexibility. In order to meet the demand for fundamental research and future industrial production, high-quality wafer-scale MoS_2 films are required. In view of this, chemical vapor deposition (CVD) has been used to produce high-quality large-area monolayer MoS_2 films in a controlled manner. The CVD process involves many tunable parameters such as temperature, pressure, gas flow rate, etc. This study is mainly focused on understanding the growth parameters in order to grow high-quality (low defect density with low impurity concentrations) large area (wafer scale range) MoS_2 films. In addition, the as-grown films were characterized to determine the properties of the as-grown films.

Large area high-quality, ultrathin 2D-MoS_2 films were grown on SiO_2/Si or Si substrates using atmospheric pressure chemical vapor deposition (APCVD) at elevated temperatures. The growth precursors (MoO_3 and S) were placed separately inside a double zone furnace to control the growth parameters individually for better flexibility during the growth process. The study revealed that the change in distance from the precursor to the substrate significantly influenced the precursor concentration on the substrate surface. The change in chemical potential of the Mo and S precursor concentration affects the shape, edge structure and final composition of the evolved structures. Auger electron spectroscopy (AES) analysis and scanning electron microscopy (SEM) confirmed that the equilibrium crystal shape of the MoS_2 was hexagonal under Mo-rich conditions. On the other hand, the shape of the MoS_2 crystal changed to a triangle under S-rich conditions. This study also showed that when the concentration of the Mo-based precursor was high on the substrate surface, it tended to form MoO_2 crystals instead of MoS_2 films. The growth mechanism of MoO_2 crystals on different substrates was quite similar. However, the arrangement of the as-grown crystals on the different substrates was not the same. Density functional theory (DFT) based calculations and experimental results suggest that surface energy plays a crucial role in the alignment of the as-grown MoO_2 crystals on the three substrates, which combined with substrate effects, altered the stacking arrangements of the grown crystals.

In addition to the growth of the MoS_2 films, a study was also performed using air plasma treatment of the as-grown MoS_2 films (monolayer and bilayer) at different time intervals with fixed power of 240 W to modulate the electronic and photonic properties. On increasing plasma irradiation time, a significant shift (blue shift) was observed in the Raman spectra of both the monolayer and bilayer MoS_2, which was attributed to the phonon hardening by air molecule insertion in the 2H-MoS_2 crystal cell. The polymorphic transformation from 2H to 1T'-MoS_2 was observed after a few minutes of plasma irradiation. The optical bandgap of both mono and bilayers MoS_2 films initially increased up to some extent on plasma irradiation. However, long-time plasma exposure led to the reduction in the PL intensity due to metallization by polymorphic transformation. The studies also reveal that MoS_2 films are etched by air plasma upon long-time irradiation.

Keywords: Two-dimensional molybdenum disulfide (2D-MoS_2), Chemical vapor deposition (CVD), Shape evolution, Surface energy, Density functional theory (DFT), Plasma treatment