Impact of Glass Substrates on MoS2 Monolayers Grown by Novel Oxychloride CVD

We have developed a scalable MOCVD-type growth technique for MoS2 and WS2 monolayers with a combination of novel oxychlorides (MoO2Cl2 and WOCl4) as gaseous molybdenum and tungsten precursors, and H2S as sulphur source. Although these oxychloride precursors are solid at room temperature, they show moderate volatility and therefore can be transported into reactor by N2 carrier gas through mass-flow controllers. One of the advantages of oxychlorides is carbon-free, which prevents from incorporating carbon contamination into grown films. So far, we found that uniform MoS2 and WS2 monolayers were successfully grown over 2-inch SiO2/Si substrates in our horizontal-flow reactor. However, one of the drawbacks is smaller single crystal grains (typically less than 0.3 μm) on SiO2 substrates, probably due to higher nucleation density.

Recently, however, there are several studies which report the grain size of MoS2 is strongly enlarged by sodium catalyst effect from NaCl powder placed at hot region of the reactor [1-4]. Here we report more facile and reproducible method using sodium-aluminosilicate glass as substrates, which contain a considerable amount of Na2O in composition. We found that the usage of sodium-aluminosilicate glass has huge impact on the grain size and nuleation density of MoS2 grown by our oxychloride CVD.

Figure 1 shows a series of SEM images of MoS2 samples after 60 min growth under a various N2 flow rates through MoO2Cl2 container. Growth temperature and pressure are 700 °C and 50 Torr. Because of the softening temperature of glass as high as over 800 °C, it does not need any special care for the substrates and graphite susceptor. At N2 flow rate of 50 sccm through MoO2Cl2 container, MoS2 islands were formed with an average spacing of several μm in between (fig. 1 (a)). With increasing the N2 flow rate for MoO2Cl2, the size of islands become larger in a layer-by-layer manner and almost coalesce under 100 sccm condition (fig. 1 (b), (c)). The grain size is around 1-2 μm, which is times larger than on SiO2/Si substrates in our previous experiments. When the supply of MoO2Cl2 is further increased, continuous MoS2 monolayer with full coverage was completed (fig. 1(d)), and the second layer of MoS2 starts to grow (fig. 1 (e)). These results suggest the sodium catalyst from glass substrates affect the MoS2 nucleation and subsequent 2D growth.

Figure 2 shows Raman and PL spectra from the sample with full coverage. Peak separation of the two Raman modes, E12g and A1g, shows that the MoS2 film is monolayer. Strong PL emission was observed around 1.89 eV, indicating that the MoS2 layer is high crystalline quality. Thus, experimental results clearly indicate that our novel CVD process using glass substrates is a promising approach to grow high-quality MoS2 atomic layers in a scalable manner.

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References

Figures

Figure 1: SEM images of MoS$_2$ grown on glass substrates under different N$_2$ flow rates for MoO$_2$Cl$_2$ @18°C: (a) 50 sccm, (b) 75 sccm, (c) 100 sccm, (d) 150 sccm, and (e) 200 sccm. Scale bar is 1 µm.

Figure 2: (a) Raman and (b) PL spectrum from fully-covered MoS$_2$ monolayer on glass substrate.