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Impact of Glass Substrates on MoS_2 Monolayers Grown by Novel Oxychloride CVD

We have developed a scalable MOCVD-type growth technique for MoS_2 and WS_2 monolayers with a combination of novel oxychlorides (MoO_2Cl_2 and $WOCl_4$) as gaseous molybdenum and tungsten precursors, and H_2S as sulpur source. Although these oxychloride precursors are solid at room temperature, they show moderate volatility and therefore can be transported into reactor by N_2 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 MoS_2 and WS_2 monolayers were successfully grown over 2-inch SiO_2/Si substrates in our horizontal-flow reactor. However, one of the drawbacks is smaller single crystal grains (typically less than 0.3 μm) on SiO_2 substrates, probably due to higher nucleation density.

Recently, however, there are several studies which report the grain size of MoS_2 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 Na_2O in composition. We found that the usage of sodium-aluminosilicate glass has huge impact on the grain size and nuleation density of MoS_2 grown by our oxychloride CVD.

Figure 1 shows a series of SEM images of MoS_2 samples after 60 min growth under a various N_2 flow rates through MoO_2Cl_2 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 N_2 flow rate of 50 sccm through MoO_2Cl_2 container, MoS_2 islands were formed with an average spacing of several μm in between (fig. 1 (a)). With increasing the N_2 flow rate for MoO_2Cl_2, 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 ten times larger than on SiO_2/Si substrates in our previous experiments. When the supply of MoO_2Cl_2 is further increased, continuous MoS_2 monolayer with full coverage was completed (fig. 1(d)), and the second layer of MoS_2 starts to grow (fig. 1 (e)). These results suggest the sodium catalyst from glass substrates affect the MoS_2 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, E^1_{2g} and A_{1g}, shows that the MoS_2 film is monolayer. Strong PL emission was observed around 1.89 eV, indicating that the MoS_2 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 MoS_2 atomic layers in a scalable manner.

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References

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Figures

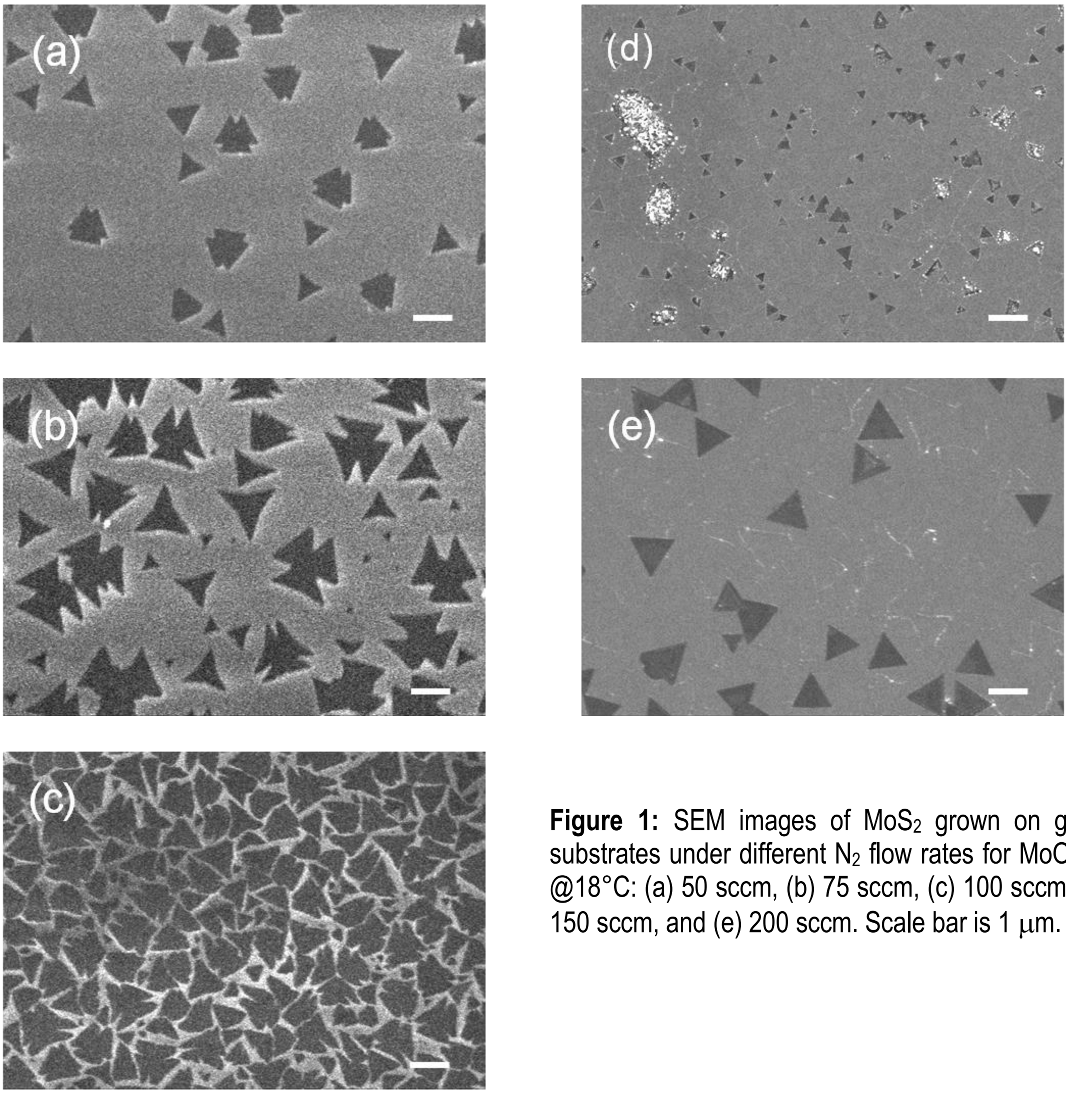

Figure 1: SEM images of MoS_2 grown on glass substrates under different N_2 flow rates for MoO_2Cl_2 @18°C: (a) 50 sccm, (b) 75 sccm, (c) 100 sccm, (d) 150 sccm, and (e) 200 sccm. Scale bar is 1 μm.

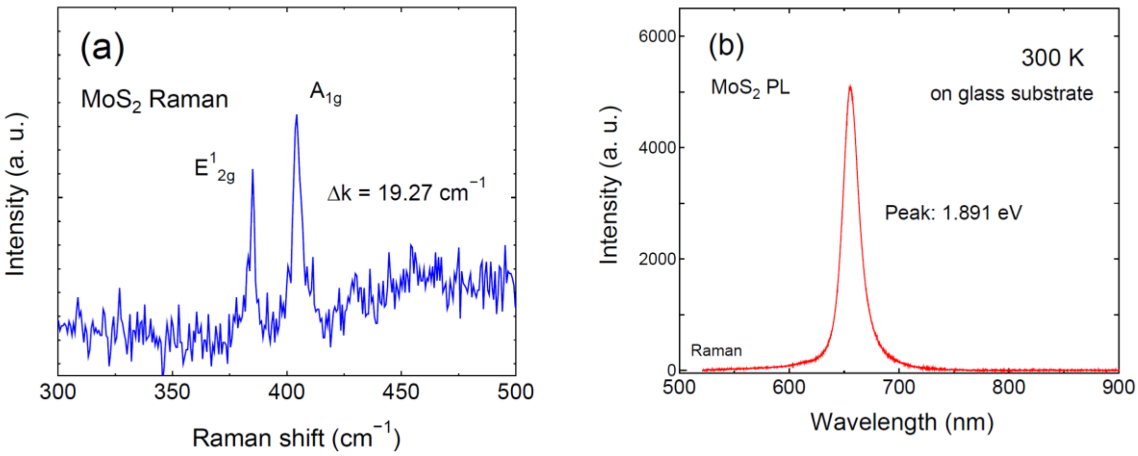

Figure 2: (a) Raman and (b) PL spectrum from fully-covered MoS_2 monolayer on glass substrate.