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Synthesis of MoS_2 thin films using the two-step approach

İki adımlı yaklaşım kullanılarak MoS_2 ince filmlerin sentezlenmesi

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Abstract

In this study, MoS_2 thin films were grown using two-step approach, which is based on employing both PVD and CVD techniques. The films were obtained initially by sputtering 1nm Mo film in the PVD system and followed by sulphurization of the film in CVD at 700ºC. The grown films were optimized employing different sulphurization times. The main difference in our study from the current literature is using preheated CVD furnace (700ºC) ahead of sulphurization. The films quality are then investigated using Raman and Photoluminance spectrometer as well as AFM measurements. The Raman spectrums indicate that two characteristic vibration modes of 2H-MoS_2 phase were observed in all samples, however, vibration modes of 1T-MoS_2 phase were also observed in some films at low sulphurization time. These results were also in line with PL measurements that confirm the direct band transition of the MoS_2 films. The surface topography of the films were investigated by AFM for MoS_2 films obtained by the sulfurization of 1 nm-thick Mo film in 15 minutes at 700ºC which shows MoS_2 crystals in triangle shape.

Keywords: MoS_2, Two-dimensional materials, PVD, CVD, Raman spectroscopy

Öz

Bu çalışmada, hem PVD hem de CVD tekniklerinin kullanılmasına dayanan iki aşamalı yaklaşım kullanılarak MoS_2 ince filmler büyütülmüştür. Filmler ilk olarak 1 nm Mo filmin PVD sisteminde saçtırma yöntemiyle ve ardından filmin CVD' de 700ºC' de sülfürlenerek elde edilmiştir. Büyütülmüş filmler, farklı sülfürizasyon süreleri kullanılarak optimize edilmiştir. Çalışmamızın güncel literatürden temel farkı, kükürtleme öncesinde önceden ısıtılmış CVD fırını (700ºC) kullanılmasıdır. Filmlerin kalitesi daha sonra AFM ölçümlerinin yanı sıra Raman ve Fotolüminesans spektrometresi kullanılarak araştırılmıştır. Raman spektrumları, tüm örneklerde 2H-MoS_2 fazının iki karakteristik titreşim modunun gözlemlendiğini, ancak bazı filmlerde düşük sülfürizasyon süresinde 1T-MoS_2 fazının titreşim modlarının da gözlendiğini göstermektedir. Bu sonuçlar ayrıca MoS_2 filmlerinin doğrudan bant geçişini doğrulayan PL ölçümleriyle de uyumludur. Filmlerin yüzey topografyası, 1 nm kalınlığında Mo filminin 700ºC'de 15 dakikada sülfürlenerek elde edilen ve MoS_2 kristallerini üçgen şeklinde gösteren MoS_2 filmleri için AFM ile incelenmiştir.

Anahtar kelimeler: MoS_2, İki-boyutlu malzemeler, PVD, CVD, Raman spektroskopisi

1 Introduction

Two-dimensional (2D) materials usually called one atomic thick materials such as graphene, carbon nanotube, molybdenum disulfide (MoS_2) have attracted the attention of many researchers due to their superior structural and physical properties [1]. Transition metal dichalcogenides (TMDs) are the most popular materials in the family of 2D materials in that they have high transmittance, high durability and direct energy band gap. The generalized formula for TMDs is MX_2, where M is the transition metals (molybdenum, tungsten, vanadium) and X represents chalcogens (sulfur, tellurium and selenium) [2]. MoS_2 stands out among the TMDs due to its remarkable properties. MoS_2 has a hexagonal structure connected to each other (S-Mo-S) with covalent bonds. In addition, MoS_2 has 1.3 eV indirect band transition in its bulk form and 1.85 eV direct band gap transition in the single and multi-layer form [3, 4]. The band gap called "direct" is the top of the valence band and the bottom of the conduction band occur at the same value of the K-point of Brillouin Zone. Therefore, when an electron rises from valance band to the conducting band, it does not need addition of phonon energy. When the conducting and valance bands do not have the same value with the K-point of Brillouin Zone, this is called "indirect" band gap. Thus, in order for an electron to move from valance band to the conducting band, it needs extra energy known as phonon energy to push the electron to the conducting band [5]. This restricts especially opto-electronic applications. In this respect, the transition from indirect to direct band structure when MoS_2 is thinned down creates an important advantage for the opto-electronic applications [6].

MoS_2 has commonly two typical phases called 2H and 1T. The 2H phase represents semiconductor while the 1T addresses metallic behavior. The structural difference between the 2H and 1T phases is due to the geometric arrangement of the S-atoms. Phase change/transformation can be made/done in the MoS_2 structure by applying annealing processes without the need for high pressure or doping [7-9].

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There are a few different approaches regarding MoS_2 synthesis such as magnetron sputtering [10], chemical vapor deposition (CVD) [11, 12], hydrothermal synthesis [13] and electrochemical processes [14], which are considered to be more suitable and applicable techniques. The magnetron sputtering technique provides large-scale synthesis and is compatible with the existing industrial processes with low cost and easy control. However, the magnetron sputtering technique creates an obstacle in synthesizing TMDs, especially for MoS_2 and $MoSe_2$, due to the difficulty of controlling stoichiometric ratio of S or Se [15].

In this study, large scale and homogeneous MoS_2 films were synthesized using both magnetron sputtering and CVD methods. Although, CVD provides many advantages in MoS_2 synthesis, this method includes many parameter optimizations such as; sulphurization time, gas flow ratio, pressure and etc. Mo films were first coated on the glass and Si/SiO_2 substrates by magnetron sputtering and then sulphurization was carried out in the preheated CVD at high temperature to obtain MoS_2 thin films with high homogeneity. We call this approach as a two-step approach, which is based on sputtering Mo films and the annealing them in sulfur atmosphere in the preheated CVD at high temperature to obtain MoS_2 thin films.

2 Method

In this study, the glass and Si/SiO_2 substrates were cleaned using the standard cleaning procedures (5 minutes acetone, 5 minutes isopropanol alcohol and 10 minutes deionize water in ultrasonic bath) ahead of synthesis process. Mo was sputtered on these substrates to achieve 1 nm film thickness using 50 W RF power under $1.6x10^{-6}$ Torr base pressure in a pure Ar gas atmosphere. Then, 1 nm thick Mo films were sulfurized in the CVD system applying different sulphurization times for the MoS_2 film synthesis. Sulphur powder (50 mg) and 50 sccm Ar gas flow ratio were used in the sulphurization process.

In the MoS_2 synthesis procedure, the Mo coated substrate was placed in the quartz tube and exposed to 700 °C directly in the preheated CVD furnace. In other words, samples were not placed in quartz tube during the heating process of the furnace. The temperature of the samples in the furnace was increased rapidly with a ramping rate of 1.67 °C/s, which was measured using thermocouple. As for the sulphurization temperature, it was kept constant at 700 °C for all the investigated samples according our many runs and literature [16, 17].

Sulphurization time is an important parameter to enhance MoS_2 film quality. So different dwell times were applied to determine the optimum sulphurization time, which were 5-, 10-, 15- and 20-minutes. This process was carried out in the CVD furnace at 700 °C. The structural properties of the synthesized MoS_2 films were characterized by Raman and Photoluminescence (PL) Spectrometer and Atomic Force Microscope (AFM) measurements.

The reason why Raman spectroscopy was employed for characterization is because it is one of the essential and straightforward characterization tools to determine MoS_2 film quality. MoS_2 shows two characteristic modes (two peaks) in Raman spectrum, which are A_{1g} and E_{2g}^1; A_{1g} mode is related to out-of-plane vibration of S atoms and E_{2g}^1 mode is associated with in-plane vibration of Mo and S atoms [18]. The frequency difference (peak positions) between these two modes in Raman spectrum is increasing with the increase of the film thickness along with shifting PL emission to longer wavelengths [19, 20]. Additionally, if the frequency difference between the two modes in Raman spectrums is around or below 20 cm^{-1}, the films tend to become thinner towards a single layer [21-24].

3 Result and discussions

Raman spectrum of MoS_2 films obtained with different sulphurization times of 1 nm thick Mo films is presented in Figure 1. MoS_2 structure was not obtained when 5 minutes dwell time was applied. This can be attributed to the S atoms, which do not have enough time to penetrate into Mo film at 700 °C. MoS_2 structure was first obtained for 10 minutes dwell time. Characteristic modes of MoS_2 in the Raman spectrum, which are A_{1g} and E_{2g}^1, were determined around 386 cm^{-1} and 410 cm^{-1}, respectively for the film obtained in 10 minutes dwell time. These peaks also underline the presence of 2H-MoS_2 phase, which is the so-called semiconductor phase exhibiting semiconductor properties. Additionally, this film has another three characteristics Raman modes named J_1 (156 cm^{-1}), J_2 (220 cm^{-1}), J_3 (290-304 cm^{-1}). These modes are related to the 1T-MoS_2 phase, which is metallic. Therefore, the MoS_2 film can also exhibit metallic properties as well as semiconducting properties (2H-MoS_2 phase) at the same time [25, 26]. The 1T-MoS_2 phase occurs due to the failure of S atoms to settle in the structure within a sufficient time phase. This was also applied in our case for the sample obtained in 10 minutes dwell times, which is not sufficient for S atoms to penetrate into Mo structure [7, 27, 28]. When the dwell time increased from 10 to 15 minutes, the 1T-MoS_2 phase evolved to 2H-MoS_2 phase [29-32]. Upon increasing the dwell time to 15 minutes, previously seen J_1, J_2 and J_3 modes disappeared, and the characteristic modes of 2H phases were observed at around 384 cm^{-1} and 409 cm^{-1}, respectively. In brief, S atoms could penetrate to the Mo structure and 1T phase evolved to 2H phase in 15 minutes dwell time. However, higher dwell time (20 minutes) affected the MoS_2 film quality seriously and no MoS_2 film was obtained at all. These results can be ascribed to the defect formation in the MoS_2 film structure by the increase in the dwell time due to the rise in the amount of S atoms in the vicinity [33].

PL characterization is essential to determine band transition in MoS_2 structure. PL spectra of MoS_2 films obtained with different dwell times are given in Figure 2, which also verifies the Raman measurements regarding the formation of metallic and semiconductor phase of MoS_2 structure. PL spectra of the MoS_2 film obtained in 10 minutes dwell time show a peak at 972 nm, which corresponds to a band gap of 1.27 eV. This gap indicates an indirect band gap transition in addition to metallic phase, which is confirmed by Raman measurements. On the other hand, when the dwell time was changed from 10 to 15 minutes, two PL emission peaks were determined at 972 nm and 687 nm and these

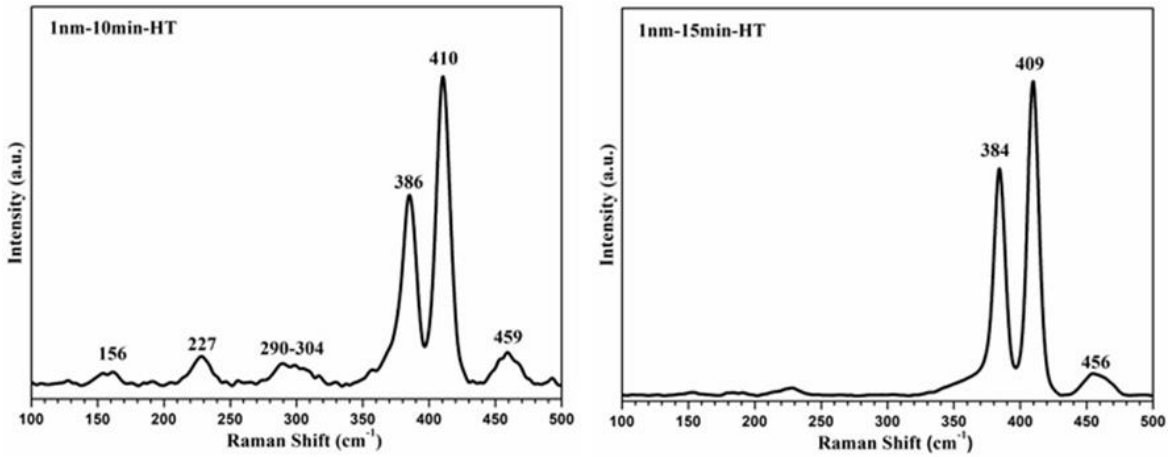

Figure 1. Raman spectrum of MoS_2 film obtained with different sulphurization times (5-, 10-, 15- and 20-minutes) of 1 nm thick Mo layers

correspond to a band gap of 1.27 and 1.80 eV, respectively. This is highly related to the metallic phase that converted into semiconductor phase with increasing dwell time. However, rising the dwell time from 15 minutes to 20 minutes destroyed the MoS_2 structure due to higher interaction of Mo film with more S atoms in high temperature [8, 34].

Figure 3 shows the AFM image (1µm x 1µm) of MoS_2 film obtained on the Si/SiO_2 substrate by sulfurization 1nm-thick Mo film in 15 minutes at 700 °C. The film exhibits similar surface morphology across the film, which is the confirmation of high coverage and homogeneity of the film.

We observed that the AFM images of the synthesized film is a few-layer, homogenous with < 0.5 nm surface (RMS) roughness and these results are in line with those obtained through the Raman measurements.

4 Conclusions

In the present study, 2D-MoS_2 films were synthesized using a two-step method. The first step included the synthesis of 1nm-thick Mo film using magnetron-sputtering system while the second step concerned the sulphurization

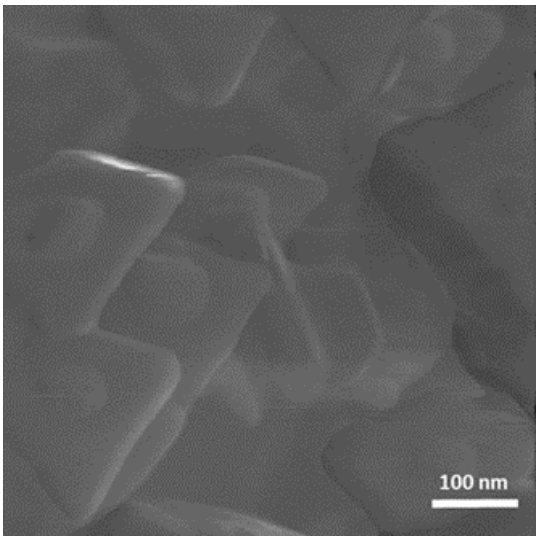

Figure 3. The AFM image (1µm x 1µm) of MoS2 film obtained on the Si/SiO2 substrate by sulfurization 1nm-thick Mo film in 15 minutes at 700 °C

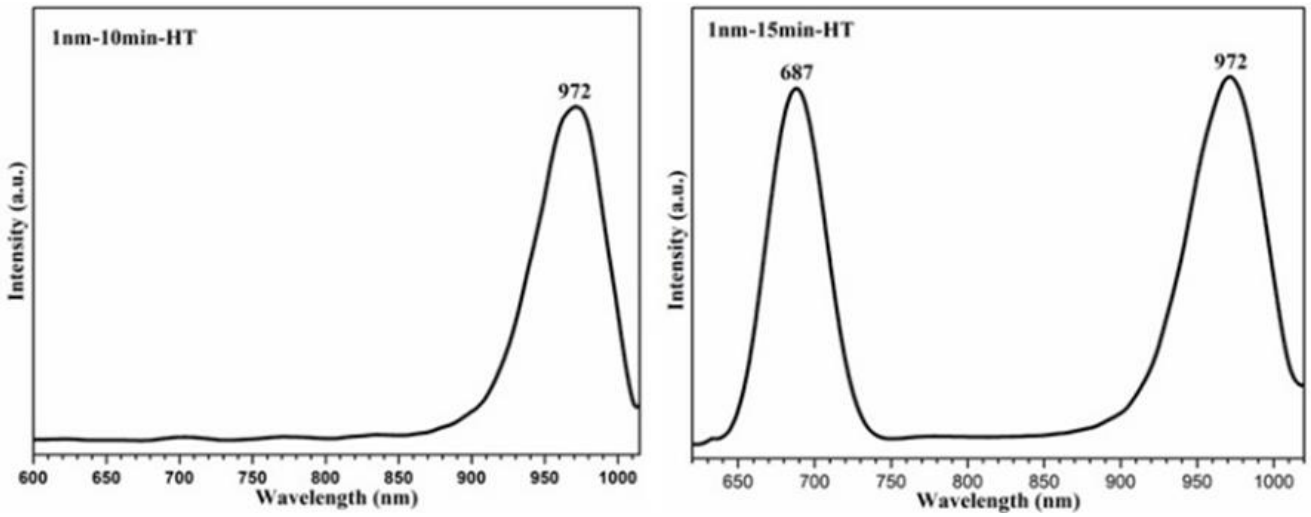

Figure 2. PL spectrum Raman spectrum of MoS2 film obtained with different sulphurization times (5-, 10-, 15- and 20-minutes) of 1 nm thick Mo layers

of the Mo films with different dwell times in the CVD system. According to the Raman and PL spectroscopy measurements of MoS_2 films, 10 minutes of dwell time was found to be insufficient for the formation of single phase MoS_2 as the obtained film showed metallic phase. With the increase of dwell time from 15 to 20 minutes, the metallic phase changed to the semiconductor phase and the indirect band transition evolved to a direct band transition. However, we observed that the MoS_2 structure could not be obtained when the dwell time was increased to 20 minutes and the optimum dwell time for the HT method was set to 15 minutes. Hence, we conclude that the two-step approach is highly effective to synthesize high quality MoS_2 films by the sulfurization of 1 nm-thick Mo film in 15 minutes at 700°C. These results enable the use of MoS_2 films in optoelectronic applications.

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Conflict of interest

The author declares that there is no conflict of interest.

Similarity rate (iThenticate): 7%

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