

# Controlled Preparation and Crystal Phase Control of MoTe<sub>2</sub> Thin Films by CVD Method

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**Abstract:** Transition metal chalcogenide molybdenum telluride (MoTe<sub>2</sub>) has three crystal phase structures of 2H, Td and 1T', among which 2H phase structure (2H-MoTe<sub>2</sub>) has attracted much attention due to its small band-gap width (1eV) and high carrier mobility (2500cm<sup>2</sup>·V<sup>-1</sup>·s<sup>-1</sup>). Hexagonal (2H-MoTe<sub>2</sub>) polycrystals are an indirect gap semiconductor, but refining them into several layers leads to the emergence of a direct band gap of 1.1eV. By comparing the different preparation schemes, as well as the growth preparation methods of other two-dimensional materials, we found that the reactivity of tellurium can be improved by introducing hydrogen, which has the same role in the preparation of other transition metal chalcogenides. In this paper, we successfully prepared two MoTe<sub>2</sub> films with different morphologies by chemical vapor deposition. In addition to introducing hydrogen as carrier gas, NaCl was added as catalyst in order to increase the concentration of Mo atoms during the reaction.

**Keywords:** Chemical Vapor Deposition; Molybdenum Telluride Film

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## 1. Introduction

Molybdenum disulfide (MoS<sub>2</sub>) is the most studied transition metal chalcogenide, but theoretical studies show that the carrier mobility of MoS<sub>2</sub> is not high enough compared with traditional materials at room temperature. Transition metal chalcogenide molybdenum telluride (MoTe<sub>2</sub>) has three crystal phase structures of 2H, Td and 1T', among which 2H phase structure (2H-MoTe<sub>2</sub>) has attracted much attention due to its small band-gap width (1eV) and high carrier mobility (2500cm<sup>2</sup>·V<sup>-1</sup>·s<sup>-1</sup>). Hexagonal (2H-MoTe<sub>2</sub>) polycrystals are an indirect gap semiconductor, but thinning them down to several layers leads to the emergence of a direct band gap of 1.1eV<sup>[1]</sup>, which is accompanied by an intense luminescence signal in the near infrared region (similar to Si), making 2H-MoTe<sub>2</sub> a viable candidate for optoelectronic devices. In addition, the mobility of 1T' phase molybdenum telluride (1T'-MoTe<sub>2</sub>) at room temperature is as high as 4000 cm<sup>2</sup>·V<sup>-1</sup>·s<sup>-1</sup>, the bandgap width is 60meV<sup>[2]</sup>, and the magnetoresistance of 1T'-MoTe<sub>2</sub> can reach about 16000% at a strong magnetic field of 14T. Compared with other transition metal chalcogenides, such excellent physical properties make MoTe<sub>2</sub> have a very wide application prospect<sup>[3]</sup>.

## 2. Experimental

In this experiment, MoTe<sub>2</sub> film was prepared by chemical vapor deposition under atmospheric pressure. Te powder was used as Te source, MoO<sub>3</sub> powder as Mo source, and SiO<sub>2</sub>/Si as substrate. The size of SiO<sub>2</sub>/Si substrate is about 0.6cm\*0.6cm. Te powder and MoO<sub>3</sub> powder were evenly spread in two quartz boats respectively, and the substrate and MoO<sub>3</sub> were placed in the same quartz boat. Te powder is placed in the first temperature zone and MoO<sub>3</sub> is placed in the second temperature zone. The distance between the two quartz boats is 18cm, and the distance between the MoO<sub>3</sub> powder and the substrate is 2cm. The carrier gas is hydrogen and argon. MoO<sub>3</sub> vapor reacts with Te vapor at high temperatures to form MoTe<sub>2</sub>.

## 3. Results and Conclusion

### 3.1 Growth of 2H-MoTe<sub>2</sub> film

In the experiment, it is found that the growth of the film can be controlled by adjusting the size of the carrier gas flow,

heating rate, maximum temperature, growth time, cooling rate and other conditions, and the most obvious is the change of its morphology. The 2H-MoTe<sub>2</sub> films catalyzed by no NaCl have no obvious boundary and mostly have irregular morphology. However, when NaCl is added for catalysis, hexagonal 2H-MoTe<sub>2</sub> films with clear boundary can be seen to grow, as shown in Fig.1 (a) and (b) below. After adjusting the growth time, the temperature zone of Mo source and substrate was controlled at 670°C for 45min, and monolayer 2H-MoTe<sub>2</sub> could be grown, as shown in Fig.1 (c) below.

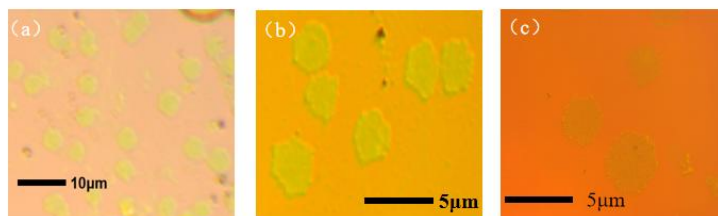
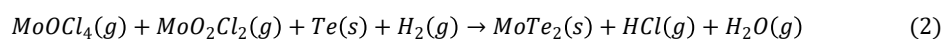
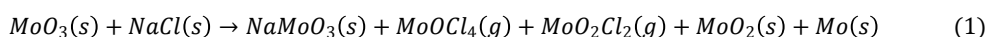


Fig.1 (a)2H-MoTe<sub>2</sub> with no NaCl catalyzed growth (b)2H-MoTe<sub>2</sub> with NaCl catalyzed growth (c)Single layer 2H-MoTe<sub>2</sub> optical microscope image

In general, it is extremely difficult to control the reaction between elements with low chemical activity, such as Te and Mo. In the preparation of transition metal tellurides (WTe<sub>2</sub>, MoTe<sub>2</sub>, etc.), due to the low chemical reactivity of Te, it is difficult to carry out telluride reaction if only argon is used as carrier gas<sup>[4]</sup>. Although the mechanism of H<sub>2</sub> in the preparation of transition metal tellurides is not well understood, the introduction of H<sub>2</sub> does improve the activity of telluride reactions<sup>[5]</sup>. It is difficult for Te and MoO<sub>3</sub> to directly react to get MoTe<sub>2</sub> mainly because it is difficult to balance the concentration of Te and MoO<sub>3</sub> in the reaction process, and the following two situations are usually prone to occur: First, in the case of insufficient Te precursor concentration, MoO<sub>3</sub> is reduced to non-volatile MoO<sub>x</sub> under the action of hydrogen (x<3, such as MoO<sub>2</sub>), so the reaction is not enough to proceed further and MoTe<sub>2</sub> cannot be obtained. Second, in the case of sufficient Te precursor or insufficient Mo precursor, the surface of MoO<sub>3</sub> powder will be tellurized first, hindering the evaporation of internal MoO<sub>3</sub> and directly blocking the reaction. Therefore, NaCl needs to be introduced as a catalyst, first to reduce the reaction temperature, and second to allow MoO<sub>3</sub> to react with it first to generate Mo-containing precursor, and then fully react with Te to generate MoTe<sub>2</sub><sup>[6]</sup>.

Due to the addition of NaCl as catalyst, the evaporation temperature of MoO<sub>3</sub> is greatly reduced. MoO<sub>3</sub> can first react with NaCl at a lower temperature to form gaseous Mo precursor, and then react with Te. The by-product produced will leave the furnace tube with the airflow at high temperature. The reaction steps are as follows<sup>[7]</sup>:



When only MoO<sub>3</sub> is used as the reaction raw material, it generally needs to be heated to 750°C, and the addition of NaCl significantly reduces the evaporation temperature of MoO<sub>3</sub>. At 450°C, the evaporation capacity within 5 minutes is an order of magnitude higher than that at 420°C, indicating that the mixture of MoO<sub>3</sub> and NaCl is easy to evaporate. The temperature at which a large amount of evaporation begins is between 420 °C and 450 °C.

The maximum temperature of Te powder is fixed at 650°C, and the temperature range of MoO<sub>3</sub> and substrate is between 650°C and 670°C for 45min. By controlling the size of air flow and cooling rate, 2H-MoTe<sub>2</sub> with hexagonal morphology with few layers and clear boundaries can be grown, and the area size is about 3~5µm, as shown in Fig.2 (a). The formation of 2H-MoTe<sub>2</sub> polycrystals is confirmed by the presence of Bg peaks at 233cm<sup>-1</sup> and Ag peaks at 289cm<sup>-1</sup> in the Raman spectra of the thin films<sup>[8]</sup>. Fig.2 (c) is the XRD pattern of MoTe<sub>2</sub>, which shows obvious characteristic peaks.

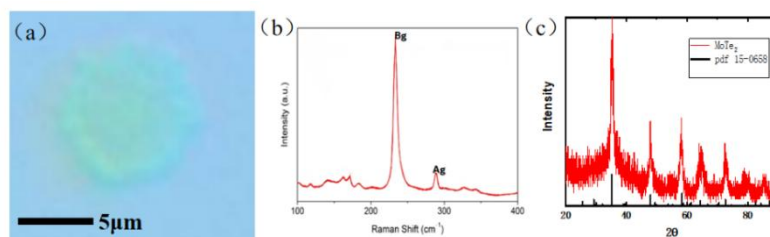


Fig.2 (a)2H-MoTe<sub>2</sub> optical microscope image;(b)2H-MoTe<sub>2</sub> Raman spectrum; (c)XRD of 2H- MoTe<sub>2</sub>;

The grown 2H-MoTe<sub>2</sub> film was prepared and the height information was collected by scanning under an atomic force microscope, as shown in Fig.3. It can be obtained that the surface of the film is relatively smooth and flat, and the height is about 3.9nm and 6.2nm, respectively, which is close to the multiple of 0.7nm, which corresponds to the thickness of single-layer MoTe<sub>2</sub>. Because the force between the growing MoTe<sub>2</sub> film and the substrate is much greater than the van der Waals force between the layers of the MoTe<sub>2</sub> film itself, the MoTe<sub>2</sub> film with fewer layers will be closely attached to the substrate. With the increase of the film thickness, the force between the atoms on the upper layer of the film and the substrate becomes weaker, and the film tends to be more smooth and flat.

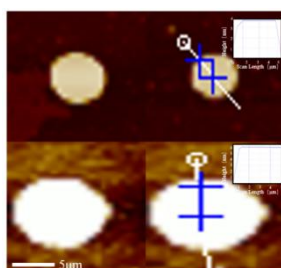


Fig.3 AFM of 2H-MoTe<sub>2</sub>

### 3.2 1T'-MoTe<sub>2</sub> film growth

It was found that accelerating the rate of heating and cooling, increasing the growth temperature to 710°C, and adjusting the tilt Angle of the quartz boat where the Mo source and substrate are located to be greater than 30° for 30min of growth usually induced more 1T'-MoTe<sub>2</sub> film growth. As shown in Fig.4 (a) below, the product morphology is mostly long lamellar morphology, with a width of about 0.3μm and a length of about 7~15μm. When the amount of carrier gas H<sub>2</sub> is slightly increased, Te powder can be reduced to TeH<sub>2</sub> more quickly and participate in the subsequent reaction more fully, while the Mo precursor evaporates out at a faster rate, increasing the reaction intensity and generating a large number of layers of 1T'-MoTe<sub>2</sub> films, as shown in Fig.4 (b), (c) Through SEM, it can be seen that the film thickness of the product is relatively uniform and the surface is relatively smooth. In Fig.4 (d) SEM can see the film growth structure with continuous large-area lamellar growth. Because the tilt Angle of the quartz boat where the Mo source and substrate are located is increased. However, due to the instability of the air flow, some cracks will appear in the structure of the product, and further improvement of the experimental conditions may be able to obtain a smooth single-layer film growing in a larger area.

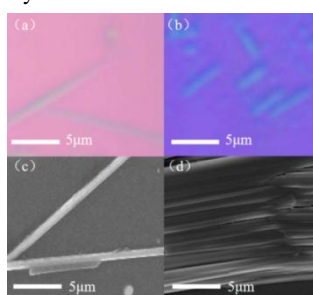


Fig.4 (a)optical microscope image of strip 1T'-MoTe<sub>2</sub>; (b)lamellar 1T'-MoTe<sub>2</sub>; (c)SEM of strip 1T'-MoTe<sub>2</sub>;(d)SEM of lamellar 1T'-MoTe<sub>2</sub>

As shown in Fig.5 (a), (b). The characteristic morphologies of 2H-MoTe<sub>2</sub> and 1T'-MoTe<sub>2</sub> can be clearly observed under the optical microscope, and MoTe<sub>2</sub> homojunction can be obtained in some areas, which can be divided into two cases. One is a 1T'-MoTe<sub>2</sub> film with strip layers extending from the edge of the flake 2H-MoTe<sub>2</sub> film, and the other is a strip 1T'-MoTe<sub>2</sub> film through the flake 2H-MoTe<sub>2</sub> film. The specific growth process of the film is still uncertain, and it is speculated that 2H-MoTe<sub>2</sub> will nucleate first, and 1T'-MoTe<sub>2</sub> will grow simultaneously with 2H-MoTe<sub>2</sub> and grow again after 2H-MoTe<sub>2</sub> is formed. Raman scattering spectrum was used to analyze the structure of samples with two morphologies. Fig.5 (c) shows the lamellar 1T'-MoTe<sub>2</sub> Raman scattering spectrum, and the characteristic peak of 1T'-MoTe<sub>2</sub> can be clearly seen: B<sub>g</sub> peak at 163cm<sup>-1</sup> and the A<sub>g</sub> peak at 125,269cm<sup>-1</sup>, Fig.5 (d) are Raman scattering spectra in the homojunction region, with characteristic peaks of two crystal phases.

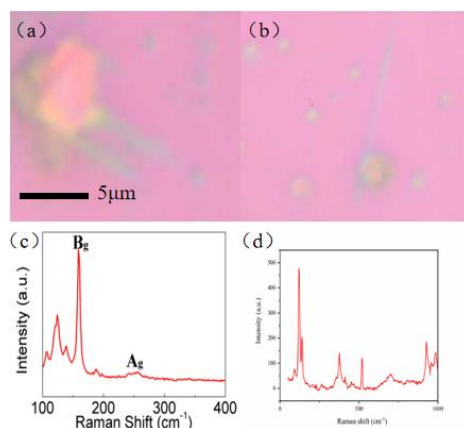


Fig.5 (a)The edge of the sheet 2H-MoTe<sub>2</sub> film extends the strip of 1T'-MoTe<sub>2</sub> film; (b)Strip lamellar 1T'-MoTe<sub>2</sub> film grown through 2H-MoTe<sub>2</sub> film; (c)Raman of 1T'-MoTe<sub>2</sub>; (d)Raman of MoTe<sub>2</sub> homojunction

## 4. Discussion

In summary, this chapter introduces the experimental steps of preparing MoTe<sub>2</sub> thin films by chemical vapor deposition. Under the premise of using MoO<sub>3</sub> as the precursor of Mo, MoTe<sub>2</sub> thin films of two crystal phases were prepared on SiO<sub>2</sub>(300nm)/Si substrate by CVD method. 2H-MoTe<sub>2</sub> and 1T'-MoTe<sub>2</sub> films were controllably prepared on SiO<sub>2</sub>/Si substrate by adjusting and changing different growth environmental conditions, and MoTe<sub>2</sub> crystal phases were obtained in the same batch of CVD growth, and MoTe<sub>2</sub> homojunction was successfully prepared.

## References

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