Supporting information

Investigating the Fundamental Conditions for Quantitative Growth to Obtain High-Quality WS2 Using a Process of Physical Vapor Deposition

Yassine. Madoune [1] ✉, Sid Ali. Madoune [2], Luzhi . Zhang [3], Reyadh A.M. Taha[4], Fuad A. Awwad [5], Emad A.A. Ismail [5]

[1] Hunan Key Laboratory of Super-microstructure and Ultrafast Process School of Physics and Electronics, Central South University No.932, South Lushan Road, Changsha, Hunan Province,410083, P. R. China

[2] School of Information and Software Engineering, University of Electronic Science and Technology of China, Chengdu, 610054, China.

[3] School of Physics & Astronomy, University of Glasgow, Glasgow G12 8SU, United Kingdom

4 School of Aerospace Engineering, Central South University, Changsha, Hunan Province 410000, P. R. China

[5] Department of Quantitative Analysis, College of Business Administration, King Saud University, P, O .Box 71115, Riyadh 11587, Saudi Arabia .

✉ Correspondence: madouneyassine001@csu.edu.cn

0000-0003-1739-1573

- **The effect of basic conditions on obtaining double growth and Size**

In this part, we discussed the initial conditions for obtaining good growth, monolayer ML or Bilayer BL. Figure 1 represents the methodological scheme we followed

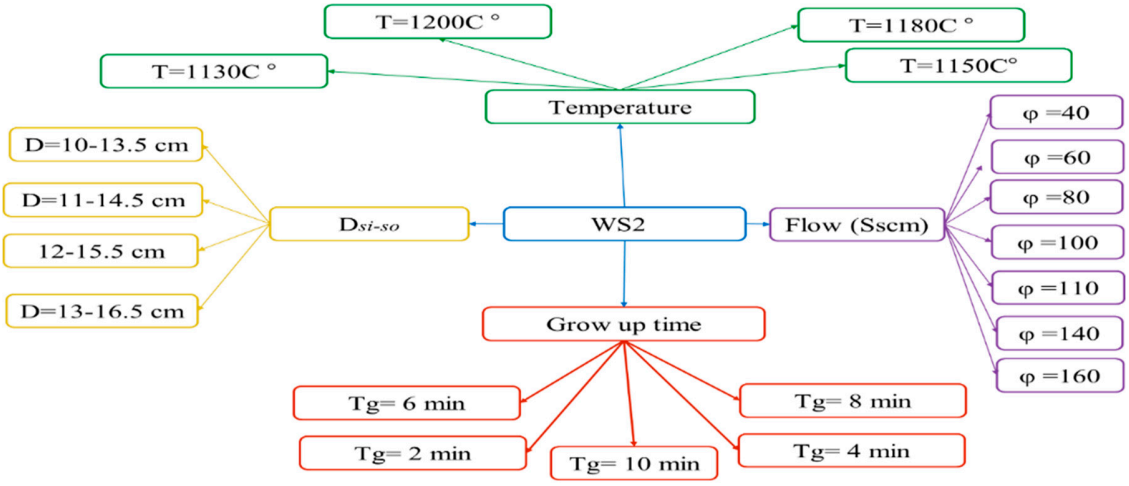


Figure S1 represents the initial conditions in an attempt to reach growth.

We fix all the requirements with only one change (e.g., T=1130, D=10 to13.5cm, t_g= 10 min and change the flow, for example, in each experiment from 40 to160 Sccm) to study analytically and to know the relationship between these conditions. In the end, we were able to find out the ideal conditions for obtaining good growth and good Size

We were able to find out the basic conditions for the growth of WS2 on the Si/Sio2

- Methods

Figure S2. Schematic diagram of the PVD synthesis device.

To prepare WS2 single-layer single crystals, a ceramic boat containing WS2 powder (0.5 g) is placed in a quartz tube (inner diameter 50 mm, The length is 100 cm). The silicon substrate (with 100 nm SiO2) is placed on the ceramic boat and pushed into a suitable position in the low-temperature zone of the tube furnace. Before heating, the N2 flow was introduced into the reaction chamber at a rate of 200 Sccm for 15 minutes to ensure a suitable environment for sample synthesis (anhydrous and oxygen-free environment).

A- ***Relationship between temperature and size*** $f(t_{tem}) = t_g$ *with* $(t_g\ C^{st})$.

Under precisely controlled conditions, traditional chemical vapor deposition has successfully prepared single-layer TMDs. Still, the synthesis conditions of two-dimensional materials are usually highly sensitive, making the current process of preparing large single crystals still very difficult.

Temp(°C)	1000	1100	1150	1180	1200	1220
Size(μm)	8-10	15-20	25-35	40-75	110-140	/

Table S1: Relationship between temperature and Size at constant growth time t_g=C^{st}=6 min,

Table S1 shows the growth from the traditional source supply method, with variable time (2-10 minutes). It can be seen that the crystal thickness changes when the temperature and growth time change.

We can notice that to get a better size, we must consider the inverse relationship between the temperature of growth and the temperature in Figure 2.

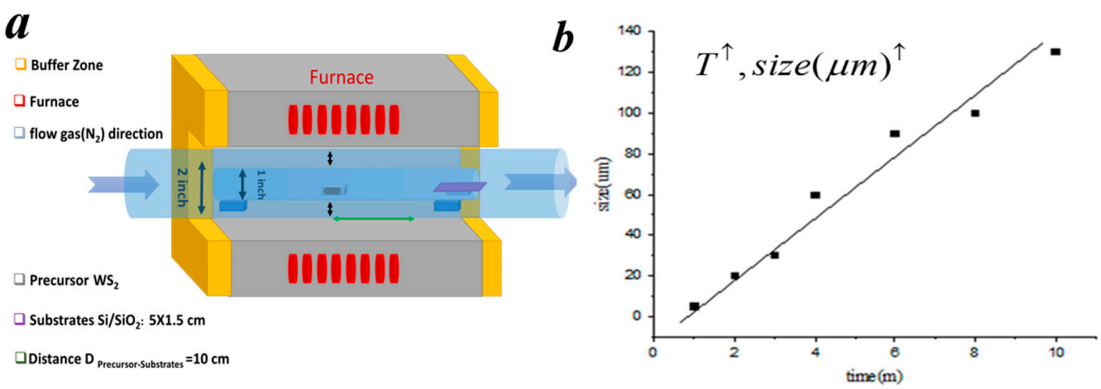


Figure S3 WS2 crystal synthesized by a PVD synthesis device. (a) Schematic diagram of the PVD system used for the growth of WS 2 (b) Relationship between size and disposition temperature

Figure S3 shows the uncontrollable growth the traditional source supply method brought about with a constant time (6 min). It can be seen that the uncontrollable crystal thickness will appear when the source evaporation temperature exceeds 1150℃, especially when the temperature is 1180℃, there will be uncontrollable crystal thickness. The two-dimensional growth structure induces a large number of non-two-dimensional planar growth at 1200°C, and the crystal thickness

and domain size will be uncontrollable. It can be seen that the ultra-high temperature evaporation of the evaporation source causes the transport of very high vapor pressure, which is beneficial to accelerate the growth rate. However, the problem is that the existing synthesis method cannot achieve precise controllability of the solid source vapor delivery. During the heating process, the turbulent evaporation source temperature disturbance and the violent fluctuation of the substrate temperature during the synthesis will cause uncertainty in the material synthesis.

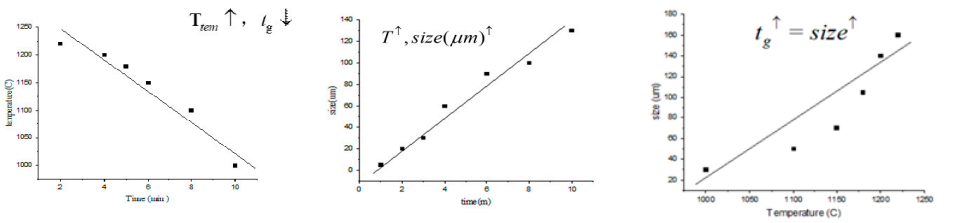
B- The *relationship between temperature, growth time, and size $f(t_{tem}, t_g) = Size$.*

Table S2 shows the growth from the traditional source supply method, with variable time (2-10 minutes). It can be seen that the crystal thickness changes when the temperature and growth time change.

We can notice that to get a better size, we must consider the inverse relationship between the growth temperature and the temperature.

Temp(°C)	1000	1100	1150	1180	1200	1220
Tg (min)	10	8	6	5	4	2
Size (μm)	35>x>40	40>x>60	60>x>80	80>x>100	110>x>160	160<

Table S2 :Relationship between temperature, growth time, and the obtained Size.



Relationship between time grow up and disposition temperature

Relationship between size and disposition temperature

Relationship between size and time grow up

Through the the table S2, we see an inverse relationship between temperature and growth time. The lower the time period and the higher the temperature, the larger the monolayer volume obtained. There is a direct relationship between temperature and Size (Figure S4).

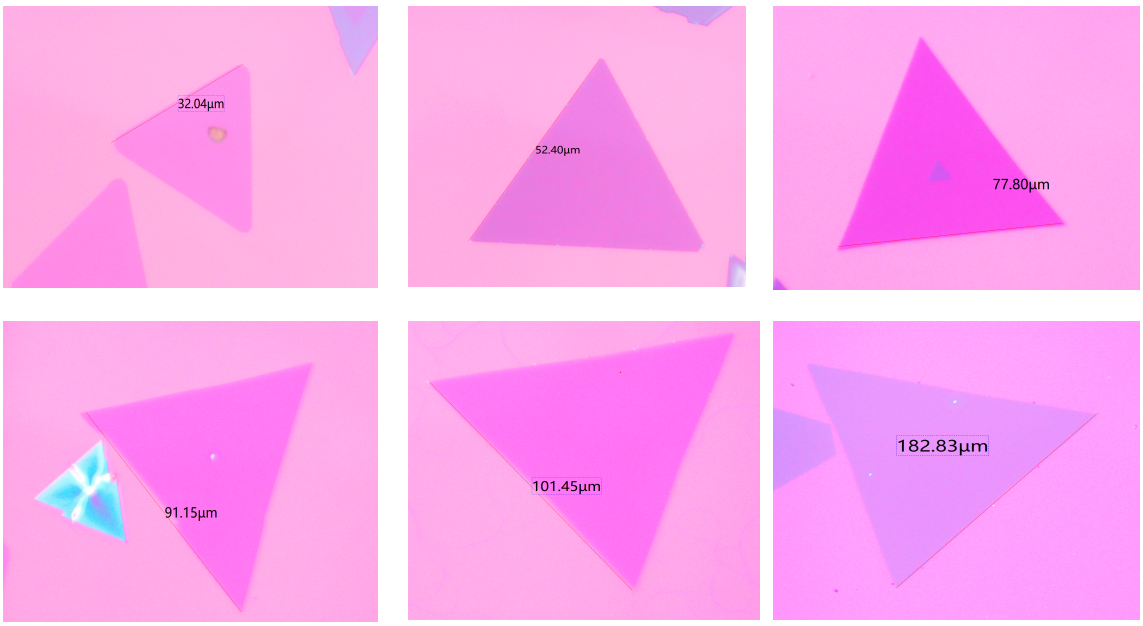


Figure S4: Ultra-fast growth regulation of single-layer WS2 single crystal. Modified PVD process with direct airflow at different condition.

Represents the sizes obtained and the agreement for the above information (Table S3).

Temp(℃)	1150	1150	1150	1150	1150	1150
φ(Sccm)	100	100	100	100	100	100
Tg (min)	10	8	6	5	4	2
Type Size (µm)	bulk	3L+bulk	3L	3L > x	3L<x<2L	No-R

Table S3:Relationship between growth time and the obtained Size at temperature and flow Constant.

we see an inverse relationship between time growing up and the type of growth. The lower (100 Sccm) the time period and the temperature (1150) constant, the more significant and monolayer volume obtained.

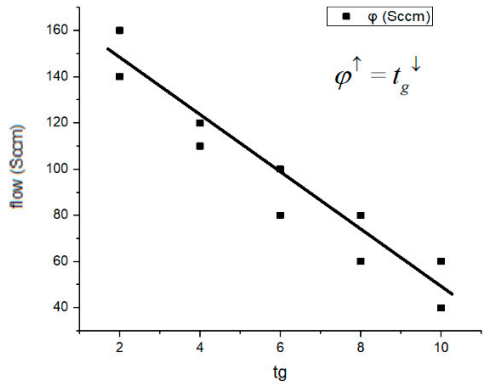
tg (min)	10	10	8	8	6	6	4	4	2	2
φ(Sccm)	40	60	60	80	80	110	110	120	140	160
Size(µm)	8-15	10-25	30-45	40-75	70-85	90-100	110-130	140-160	180	180<

Table S4 :Relationship between flow, growth time, and the obtained Size.

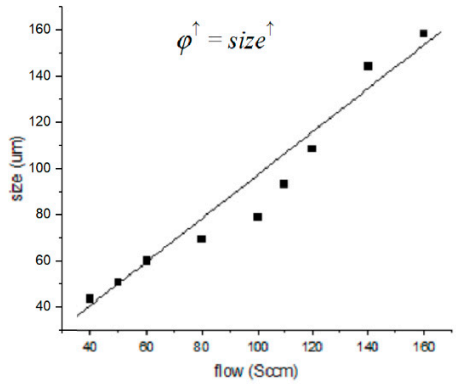
C Relationship between flow, growth time, and the obtained size f(φ Sccm)= size

Table S4 shows the growth from the traditional source supply method, with Constant temperature T=1180°C and variable time (2-10 minutes) and flow (40-160 Sccm). It can be seen that the crystal thickness changes when the flow and growth time change.

From the graph, we can note that it must consider the direct relationship between growth and flow velocity for a better size. An increase in flow means an increase in Size.



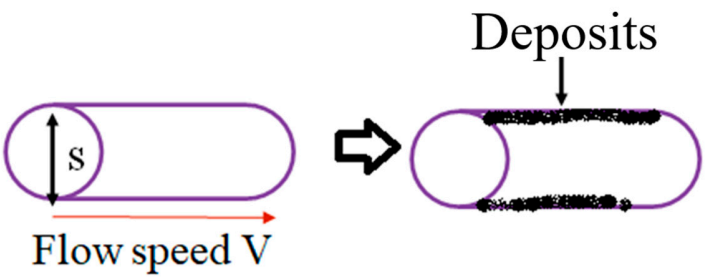
Relationship between time grow up and flow



Relationship between size and flow

Notice

- Here are neglected factors that affected the results of the experiments, such as the Gas leakage deposits in the tube ***φ=V/S***



- $\varphi,\ S^{\downarrow} \rightleftarrows V^{\uparrow}$ *: The Flow speed V increases because the tube section changes because of the sediment.*

- These results are not ideal because they are more than one layer.
- Chemical deposits are the result of previous experiments. It can cause contamination inside the tube, and the experiments may be inaccurate and contain impurities.
- Increased sedimentation can narrow the flow stream, which increases the speed, and this may affect the experiment.

Conclusion

In this chapter, we identified the primary conditions and the relationship between them, through which we can obtain good growth and Size.

We could define and adjust some initial conditions, such as temperature. We were able to notice that at a temperature of 1180°C, a flow of 110 Sccm, and a time growth up 2-4 min, we can get good growth and a size of 40 < Size< 150 µm with all this, but we cannot be certain that we were able to accurately determine these conditions because we found that there are factors that we neglected, such as the Size of the slice What can play a major role in growth is tube size.

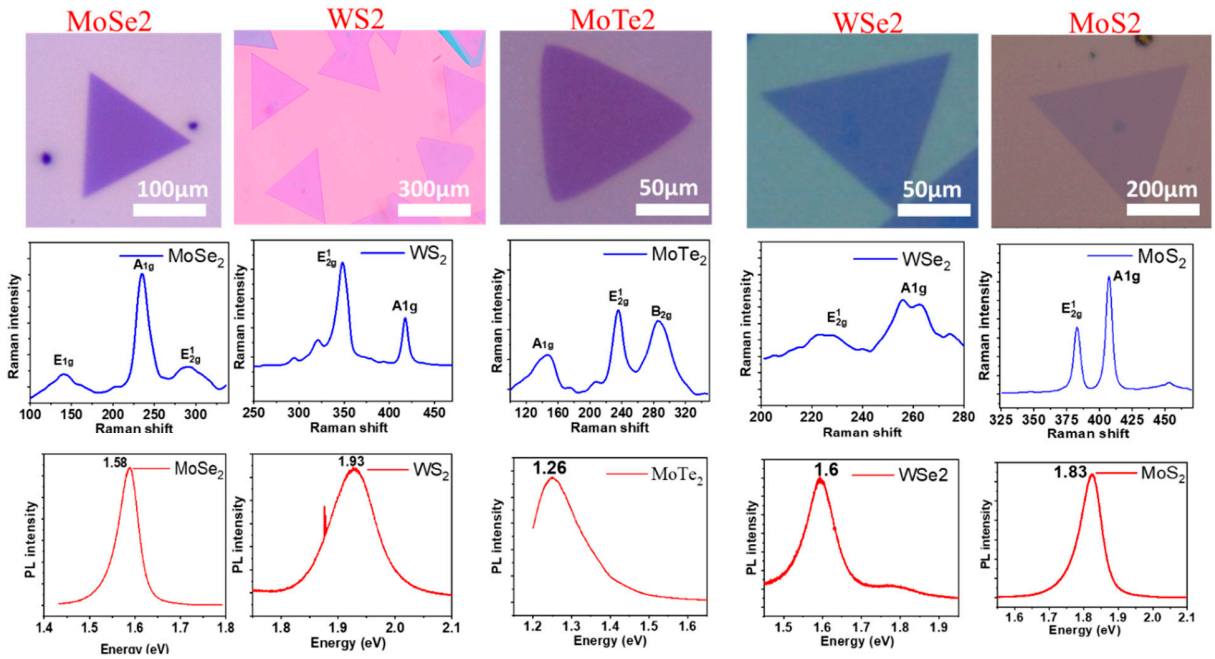


Figure S5: To synthesize and characterization of some 2D materials

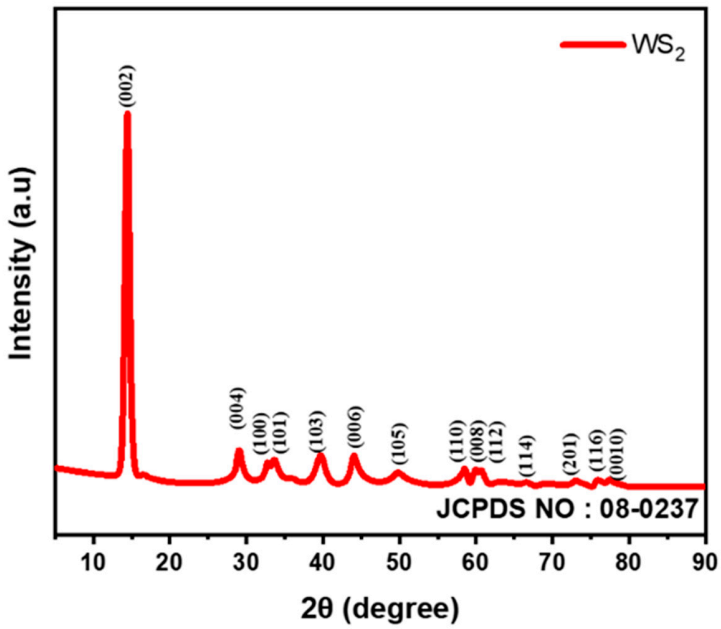


Figure S6: X-ray diffraction (XRD) pattern of the WS2 monolayer

The crystalline structure and phase composition of the synthesized samples were verified using X-ray diffraction. Figure S2a displays a representative X-ray diffraction (XRD) pattern of the WS2 nanosheets that were obtained. The reflection peaks observed in the experiment can be accurately matched to the hexagonal WS2 phase, with cell

constants of a = 3.154 Å and c = 12.362 Å. This finding is in agreement with the standard JCPDS card no. 08-0237. There are no discernible peaks corresponding to impurity phases in the observed pattern .

	Temperature (°C)	Time of growth (min)	Position (cm)	Flow φ (Sccm)	Gas directions	Source quantities (g)
MoSe2	1200	8	12	110	reverse flow	0.6
WS2	1180	10		120		
MoTe2	1100	6		50		
WSe2	1200	6		110		
MoS2	1180	10		120		

Table S5 :Represents the parameters of growth synthesis and characterization of some 2D materials.

- Note :

Reverse flow: During the temperature ramping and pre-annealing stage, the process entails the implementation of a reverse flow from the substrate to the source, while forward flow from the chemical vapor source is solely administered at the specific nucleation and growth temperature.