Electronic Supplementary Information

Surface-diffusion-limited growth of atomically thin WS_2 crystals from core-shell nuclei

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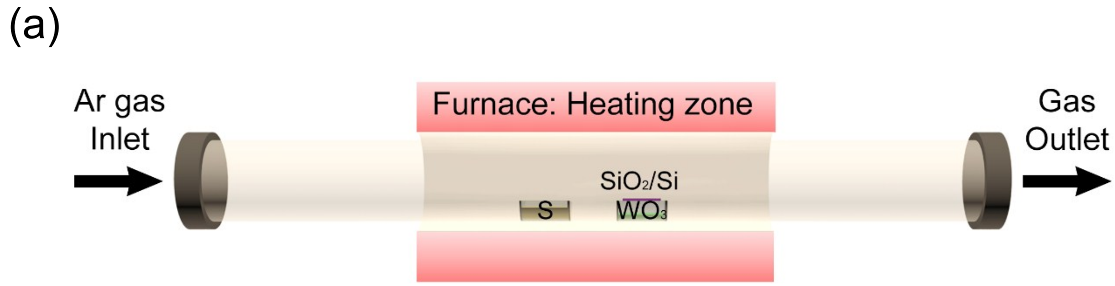
Experimental setup of CVD system to grow monolayer WS_2



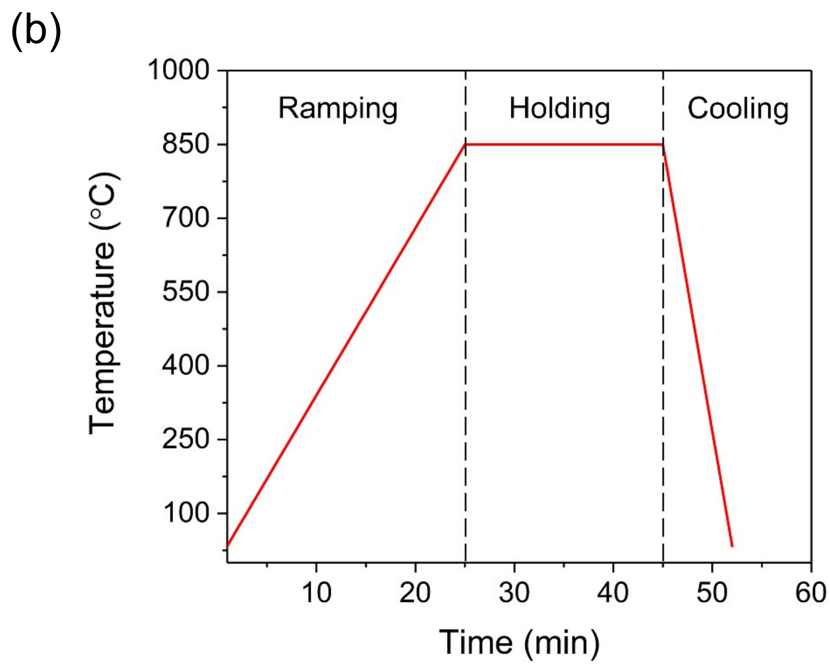


Fig. S1 (a) Schematic of the thermal CVD system used to synthesize monolayer WS_2. During the growth process of monolayer WS_2, growth conditions, such as the pressure, the temperature and the time, were precisely controlled through the CVD system. The CVD system used for this study consists of a rotary pump, an automatic pressure controller, a mass flow controller, a quartz tube chamber, and a tube furnace. For the reproducibility of the growth, the CVD chamber is sufficiently evacuated to the base pressure of about 5×10^{-4} Torr using the rotary pump. The automatic pressure controller and the mass flow controller were utilized to precisely control the growth pressure and the gas flow, respectively. This CVD system allows us to accurately maintain the pressure in the range from 0.1 to 700 Torr, and the flow rate of Ar gas in the range from 10 to 200 sccm. (b) The temperature profiles during the growth procedure of monolayer WS_2 in the heating zone of the furnace. The furnace was ramped from room temperature to a growth temperature of 850 °C at a rate of 33 °C/min, and the furnace was held at a constant temperature for 20 min and finally cooled down to room temperature.

Formation of nuclei in the initial stage: Core-shell nanoparticles

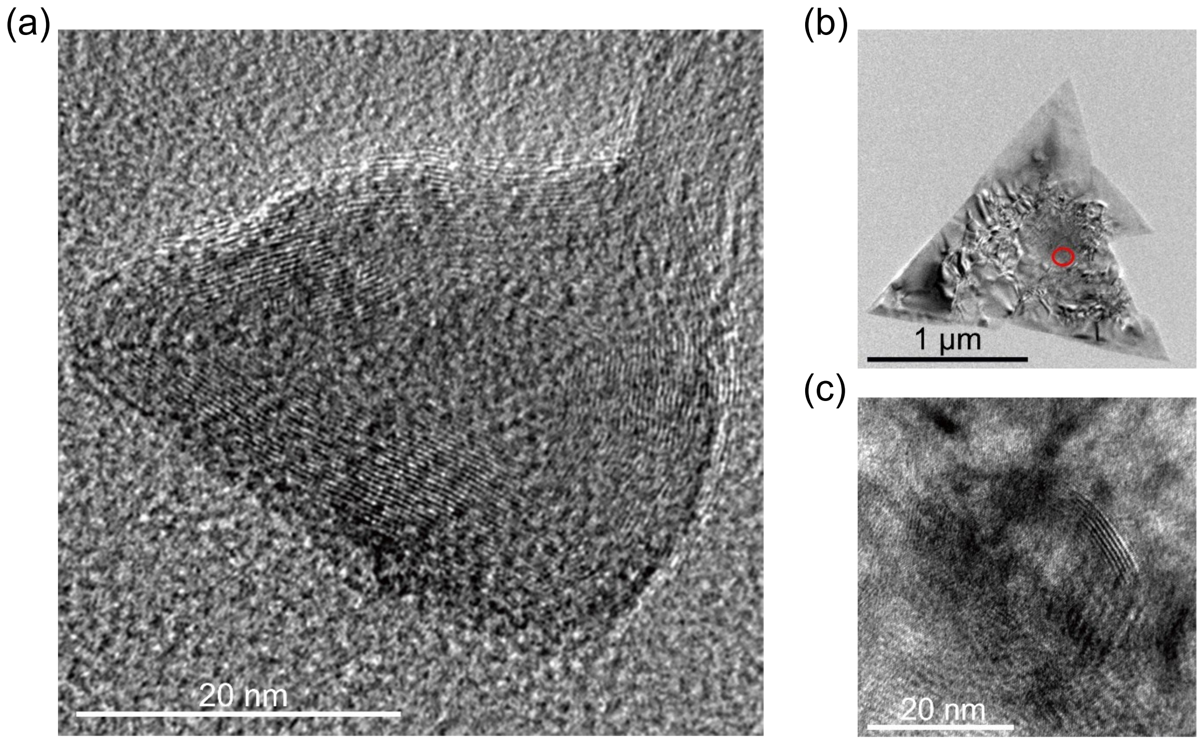


Fig. S2 (a) HR-TEM image of the core-shell nanoparticle measured from the sample grown at the temperature of 630 °C. (b) BF TEM image of WS_2 crystal grown at the temperature of 750 °C, showing the embedded core-shell nanoparticle into the 3D island region (red circle). (c) HR-TEM image of the core-shell nanoparticle embedded in the 3D island region (red circle) in (b).

Cross-sectional TEM and EDS mapping of WS_2 crystal

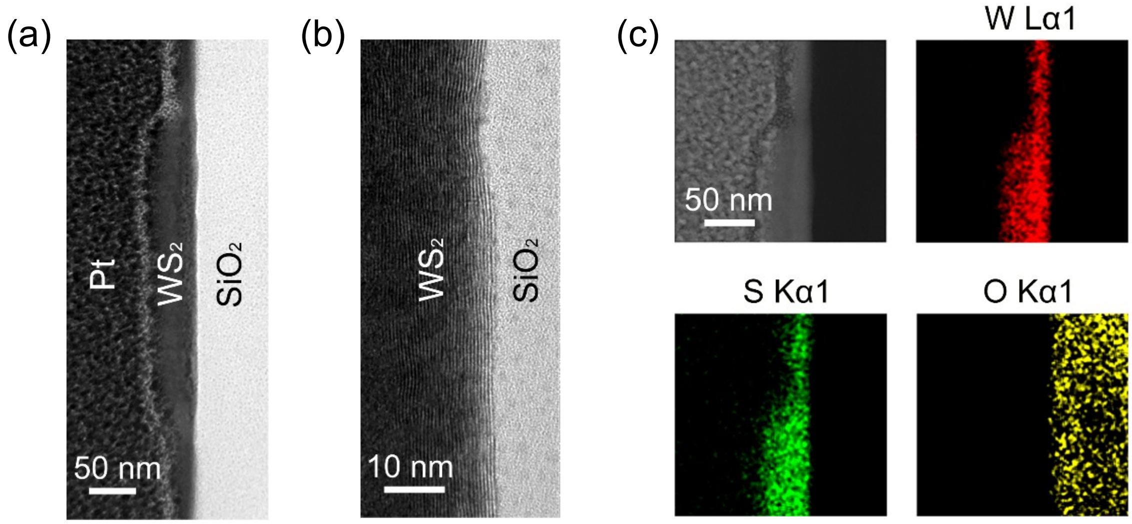


Fig. S3 (a) BF TEM image showing the cross-sectional 3D island and 2D crystal regions. (b) HR-TEM image of (a). (c) EDS map images for the cross-sectional 3D island and 2D crystal regions showing the spatial elemental distribution for tungsten (red), sulfur (green) and oxygen (yellow).

Exhaustion of sulfur powder during the ramping period

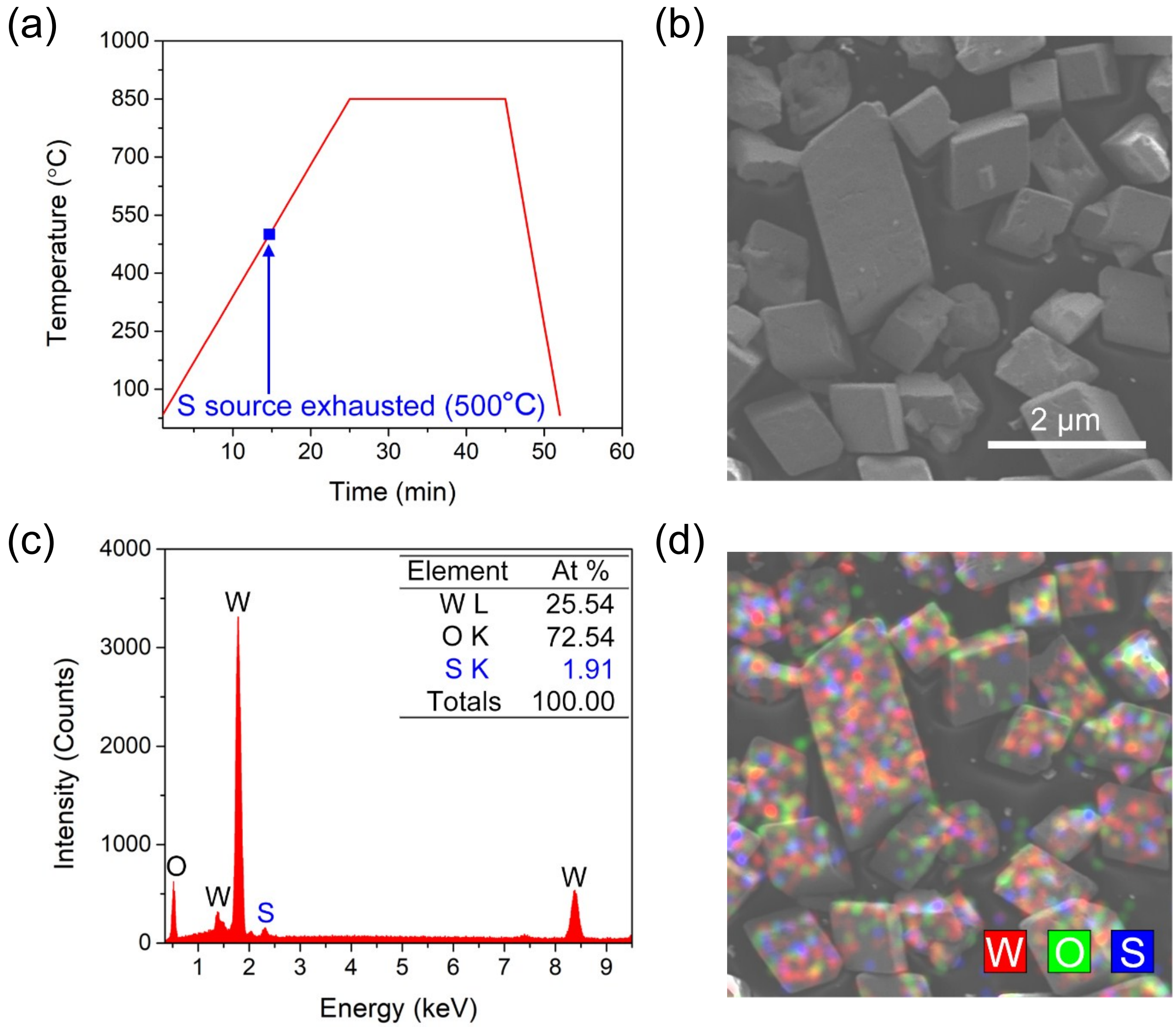


Fig. S4 (a) Temperature profiles for the growth of monolayer WS_2 in the heating zone of the furnace. However, S powder was fully evaporated and exhausted at the temperature of 500 °C before the growth of monolayer WS_2. The blue square point indicates the point at which the S powder is fully exhausted from the growth chamber. Note that full evaporation of S powder at the temperature of 500 °C was confirmed by stopping the growth process under the same growth conditions. Additionally, SEM-EDS elemental mapping was performed to check the remaining S element in the WO_3 powder, which was heated in the growth chamber, as shown in the following figures. (b) SEM image of the WO_3 powder heated to the blue square point in (a). (c, d) EDS spectrum (c) and mapping image (d) of W, O and S elements taken for the full area of the SEM image in (b).