Supplementary Information

Growth mechanisms and morphology engineering of atomic layer deposited WS_2

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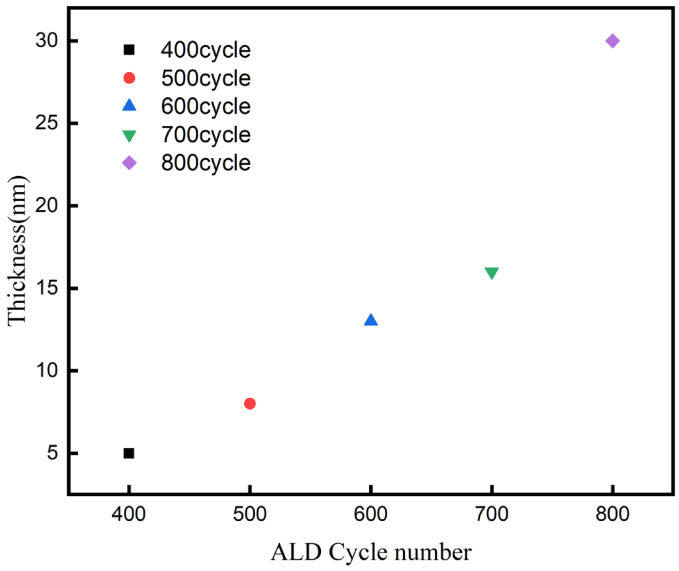

Figure S1. Growth rate of the ALD WS_2 process. The film thicknesses were measured by cross-sectional TEM and AFM. After 400 ALD cycles of continuous film formation, thickness of WS_2 film shows linear growth, indicating that the ALD process in this paper can well control the film thickness. After 700 cycles, the growth rate increases sharply, which is because the vertical growth of WS_2 starts to dominate.

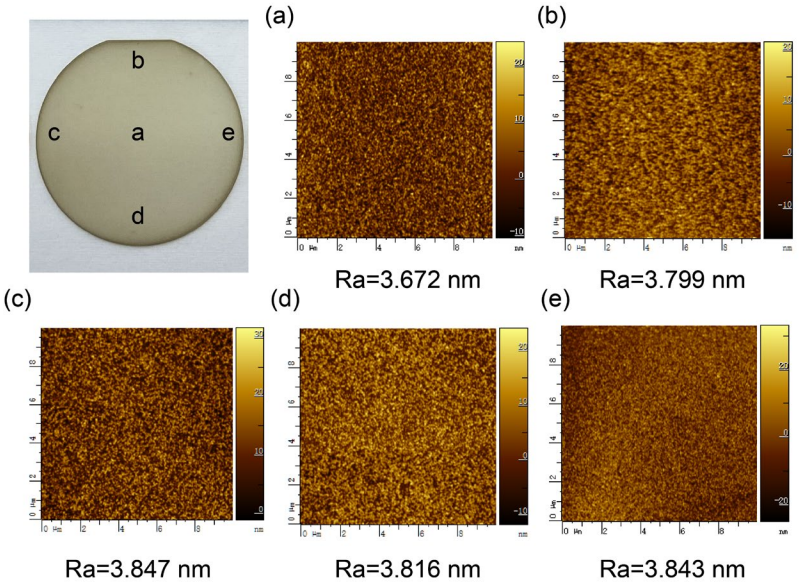

Figure S2. AFM images at five positions of 600-cycle annealed WS_2 film. (a-e) AFM measurements were carried on 600 cycles WS_2 films at the coexistence stage of lateral growth and vertical growth. The films showed good uniformity due to ALD growth mechanism. The appearance of vertical growth results in an increase in roughness.

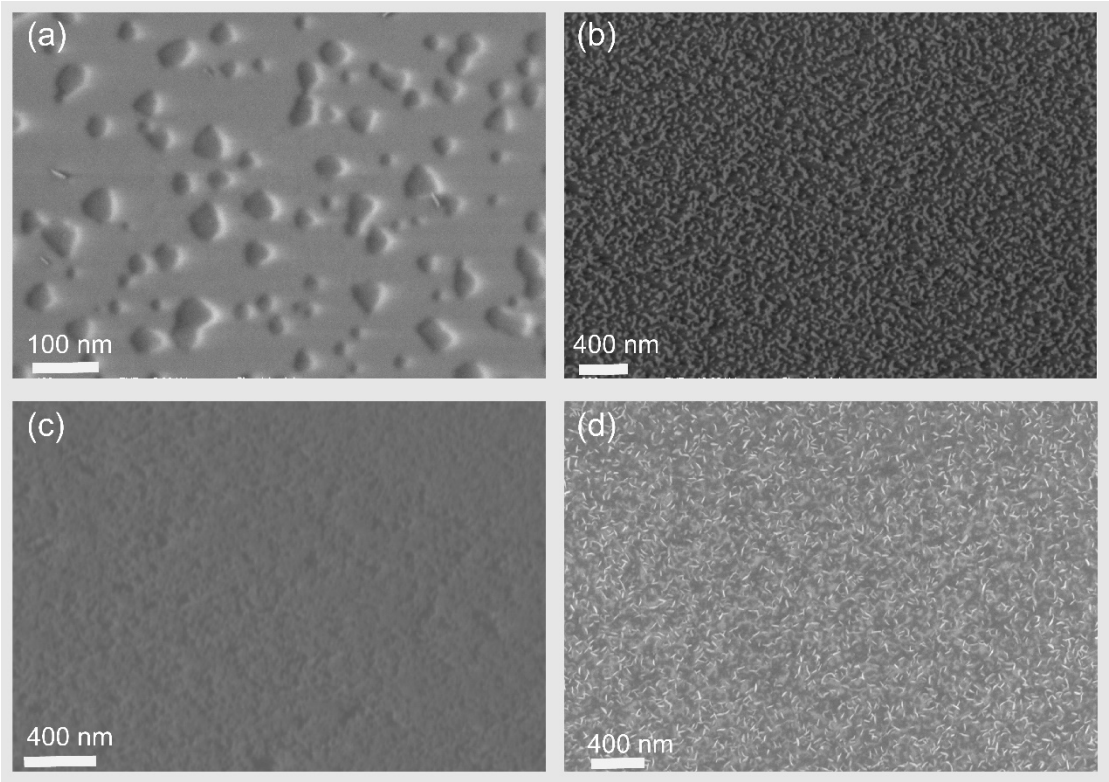

Figure S3. Plan-view SEM images of films at different growth stages. (a-b) Island lateral growth stage. The density of the island grains increased, that is, the film grew laterally until continuous films were formed. (c) Continuous film stage. (d) Vertical growth stage. A large number of nanowire structures WS_2 appeared, and the growth rate of the films increased rapidly.

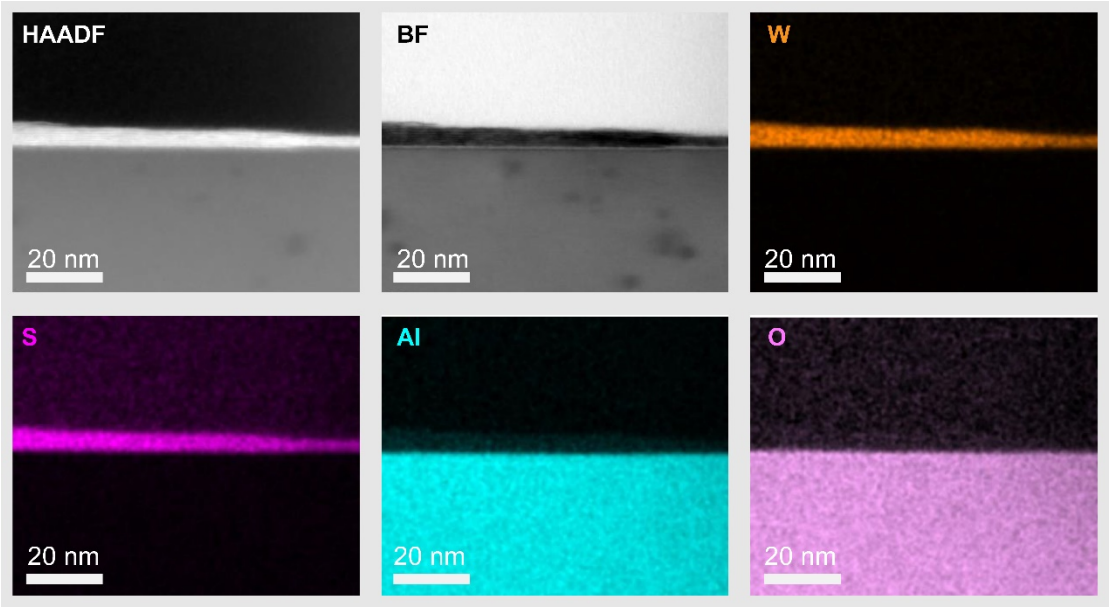

Figure S4. Energy dispersive X-ray spectroscopy (EDX) mapping of smooth planar film WS_2 films with 400 ALD cycles after annealing. The flat films were clearly observed in TEM images. Only W and S elements were found in the film, and Al and O elements were detected in the sapphire substrate region, indicating the high quality of WS_2 film after annealing.

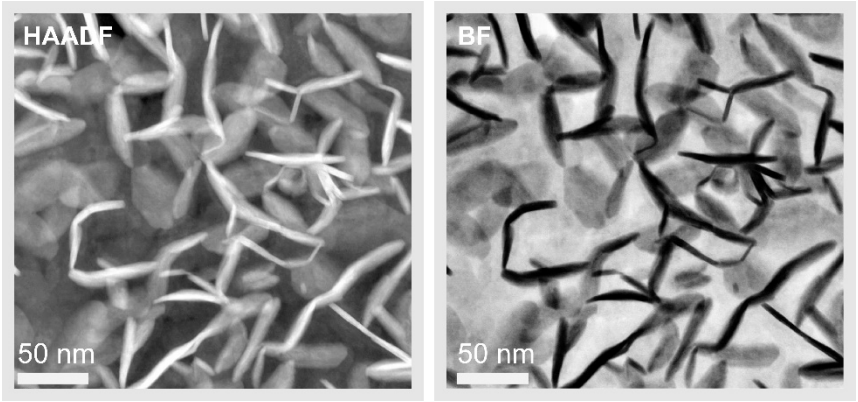

Figure S5. High angle annular dark field-STEM (HAADF-STEM) and bright field TEM (BF-TEM) of perpendicular WS_2 nanowires. A large number of WS_2 grains with nanowire structure were found, and the growth direction of these grains was perpendicular to the substrate. The nanowire grain length has reached about 50nm, which is due to the precursor being easy to attach to the dangling bond at both ends of the nanowire, resulting in a rapid increase in the growth rate of the film in this period.

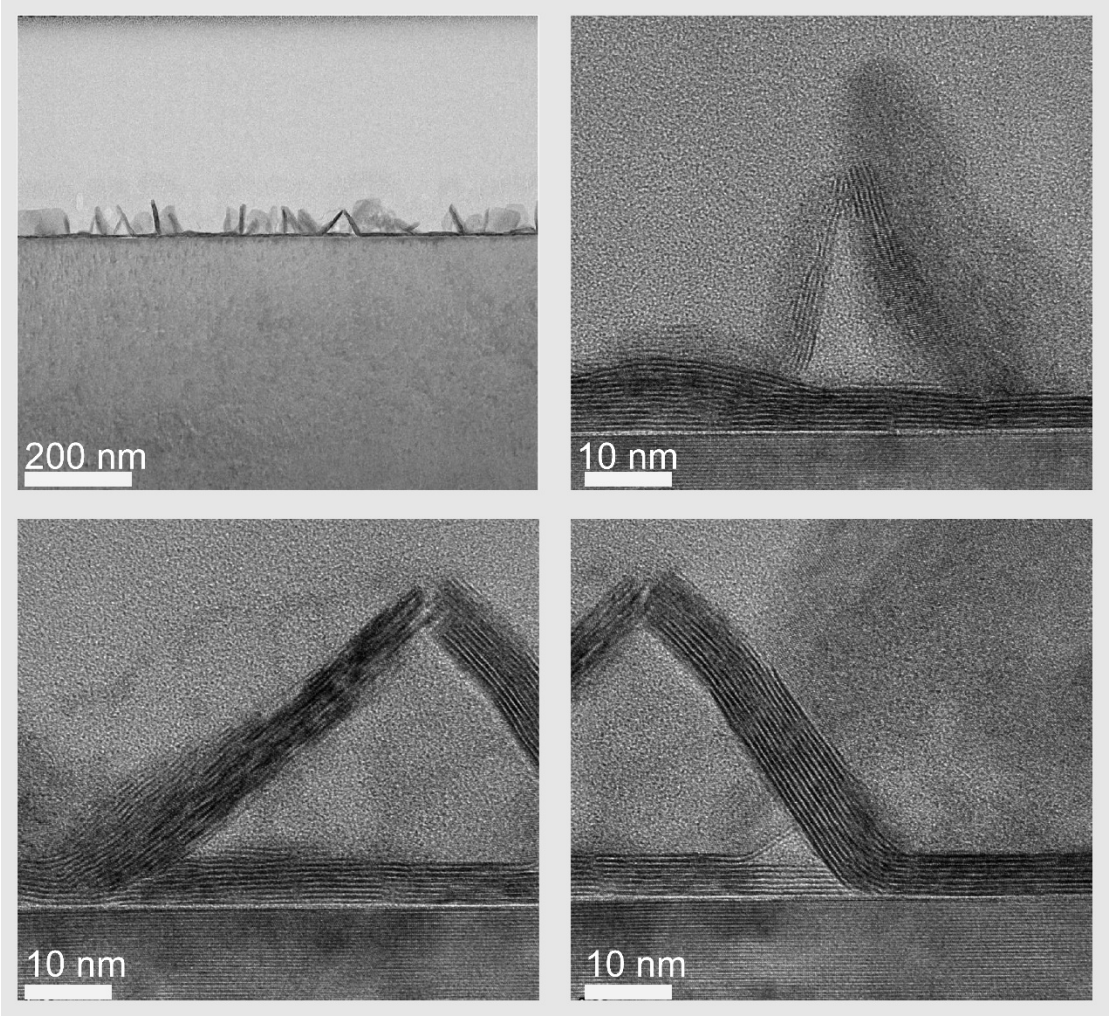

Figure S6. Cross-sectional TEM of perpendicular WS_2 nanowires. Perpendicular WS_2 nanowires were observed after vertical growth dominated. It can be found that the grains of WS_2 grow rapidly along the warping direction, and most of the grains growing vertically are due to the interaction between the two warped grains, which leads to the final growth direction vertically upward.

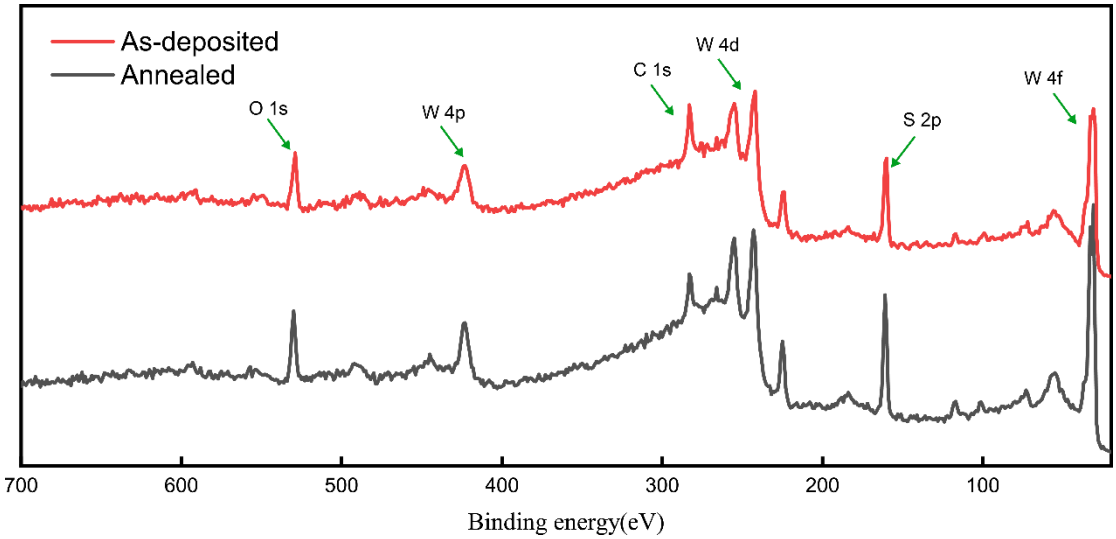

Figure S7. The full spectra of as-deposited and annealed WS_2 film. O 1s and C 1s peaks were observed due to the atmosphere of XPS equipment, and C 1s peaks at 282.5 eV was used for calibration. After annealing, the relative intensity of C and O decreased. The calculated W/S ratios of the deposited and annealed WS_2 are 1/2.3 and 1/2.1, respectively. The reduction of sulfur in WS_2 after annealing suggested the necessity of annealing process.

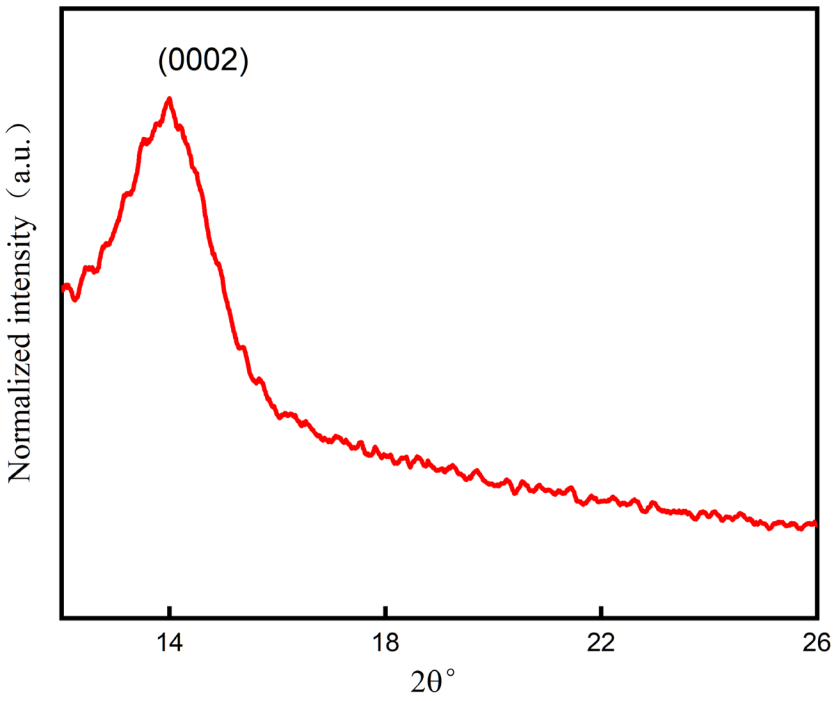

Figure S8. XRD pattern of WS_2 films with 400 ALD cycles. XRD pattern of the smooth planar film with 400 ALD cycles shows highly intense peaks at 2θ values 14.08º, which can be indexed to (0002) planes of 2H-WS_2 system. It confirmed WS_2 grains mainly grew in two-dimensional transverse direction in this stage.