**High-yield synthesis of nanometer-thick S-doped MoTe2 by a facile chemical vapour deposition method**

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**Introduction**

Transition metal dichalcogenides (TMDs), with unique strong in-plane covalent band and weak out-of-plane van der Waals interaction, have attracted significant research interest in the recent years. However, high-yield synthesis of single- or few-layer TMDs remains one of the main barriers for fundamental studies and device applications. In this study, we realize the high-yield growth of nanometer-thick S-doped MoTe2 by introducing zero-valent S8 intercalation. This study provides a new insight into the role of zero-valent intercalation in TMDs and a possible approach for high yield synthesis of ultrathin MoTe2 layers.

**Methods**

MoTe2 and S-doped MoTe2 plates were grown by chemical vapour deposition (CVD), using powders of MoO3, MoCl5, Te, and S as precursors and a mixture of Ar (100 sccm) and H2 (20 sccm) as carrier gas. Products were collected using SiO2 (285 nm)/Si (100) wafers. The horizontal tube furnace was heated to 760 and then hold for 2 hours before cooling down naturally. The TEM specimens were prepared by ultrasonic vibration in ethanol.

**Results and discussion**

Morphological characterizations show that the as-grown products vary from well-faceted hexagonal MeTe2 plates to high-density irregular plate-like S-doped MoTe2 after adding S powders in precursors. The later dimension increases from several micrometers for binary MoTe2 to several tens of micrometers for S-doped MoTe2, and the corresponding surface varies from smooth surface to a surface showing randomly-stacked nanometer-thick layer steps. It is interesting to note that after being ultrasonically vibrated, the exfoliated S-doped MoTe2 shows nanometer-thick feature.

Detailed structural and compositional analyses confirm the existence of elemental S8 phase in the as-grown S-doped MoTe2 plates, however, no element S8 phase was seen in TEM studies (noted that the TEM specimens were prepared by ultrasonic vibration, in which the S8 phase might be removed), including selected-area electron diffraction (SAED) and electron energy loss spectroscopy (EELS). The bonding state and role of S in the growth of S-doped MoTe2 have been investigated and illustrated as follows: (1) S2- ions are doped into the MoTe2 matrix and replace Te2- ion sites. (2) Individual S is intercalated into the van der Waals gap of S-doped MoTe2, leading to the lattice expansion along the *c* direction. (3) Elemental S8 crystals can suspend the out-of-plane growth, and play a role of facilely exfoliating S-doped MoTe2 layers.

**Conclusion**

We realize the high-yield growth of nanometer-thick 2H-phased S-doped MoTe2 by a facile CVD method. Elemental S8 crystals exist in the as-grown products and are responsible for the facile exfoliation of S-doped MoTe2 layers.

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