Supporting Information

Synthesis of Large-Area Uniform Si₂Te₃ Thin Film for p-type Electronic Devices

Xuefen Song^a, Yuxuan Ke^a, Xiaosong Chen^a, Jidong Liu^a, Qiaoyan Hao^a, Dacheng Wei^b, Wenjing Zhang^{a,*}

^{a.} International Collaborative Laboratory of 2D Materials for Optoelectronics Science and Technology,
Shenzhen University, Shenzhen 518060, P. R. China
^{b.} State Key Laboratory of Molecular Engineering of Polymers, Fudan University, Shanghai 200433, China

Email address : wjzhang@szu.edu.cn

Synthesis of silicon telluride (Si₂Te₃) crystals

Bulk crystals of Si_2Te_3 were prepared from the reaction of tellurium (Macklin, 99.99%) and silicon (Macklin, >99.99%). Mixtures with a total weight of about 250 mg and an initial stoichiometry of Si:Te = 2:3 were loaded into a quartz tube, which was then evacuated and sealed by an oxy-hydrogen flame. The sealed tubes were heated to 1000 °C at a rate of 80 °C/h and then kept at 1000 °C for 48 h. After that, they were cooled to 200 °C for 40,000 min followed by natural cooling. The product was a gray-red plate. Because of the extreme sensitivity of the product to moisture, all processes were completed under a dry argon atmosphere in a glovebox.



Figure S1. (a) Crystal structure of the Si_2Te_3 unit cell viewed from the top showing possible partial occupancies of the Si atoms. (b) Typical optical image of an as-prepared Si_2Te_3 film on a (300-nm) SiO_2/Si substrate grown in the temperature zone of 600-650 °C. (c) Temperature-controlled sequence typically used for growth of Si_2Te_3 thin films.



Figure S2. (a) Low-resolution TEM images of a CVD-grown p-type Si_2Te_3 film (600-650 °C) supported on a TEM grid. EDS mapping of (b) Si and (c) Te within part of the corresponding Si_2Te_3 film, demonstrating the uniform distributions of Si and Te.

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Figure S3. The Raman maps of (a) $A_{1g(143)}/A_{1g(127)}$ intensity ratio and (b) distance between $A_{1g(143)}$ and $A_{1g(127)}$ peaks.



Figure S4. (a) Root mean square of roughness (Rq) of Si₂Te₃ films grown in the temperature zones of 650-700, 600-650, 550-600, and 500-550 °C, and their corresponding substrates. (b) Statistical diagram of the number of different crystal domain sizes of corresponding Si₂Te₃ films. The statistical distribution of crystal domains size divides the domains into 0-250nm, 250-500nm, 500-750nm, 750-1000nm and > 1 μ m. Obviously, the roughness of Si₂Te₃ films prepared on 600-650 °C is minimum compared with the films grown in other temperature zones, but the crystal domain size of them are larger, and even larger than 1um, showing good crystal structure.



Figure S5. (a) Dependence of the bandgap of the Si_2Te_3 films measured by PL spectroscopy on film thickness. (b) Dependence of the distance (green) and intensity ratio (blue) of two A_{1g} on Si_2Te_3 film thickness.

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Figure S6. Representative optical microscope images of crystals grown in the different temperature zones of (a) 500-700, (b) 400-500, (c) 300-400, and (d) 200-300 $^{\circ}$ C.



Figure S7. (a) XRD patterns of CVD-grown materials, showing the crystal structures formed in the different temperature zones of 500-700, 400-500, 300-400, and 200-300 $^{\circ}$ C. Red, purple, and green asterisks indicate peaks from the crystal structures of tellurium (p3121), Si₂Te₃ (p-31c (163)), and their mixed phases, respectively. (b) Raman spectra of the corresponding samples.

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Figure S8. (a) XRD patterns of Si_2Te_3 film and bulk. (b) High-resolution XPS spectra of Si 2p and Te 3d core levels for Si_2Te_3 films (blue) and a bulk Si_2Te_3 crystal (red). (c) Low-resolution XPS profiles of Si_2Te_3 film and bulk.



Figure S9. (a) XRD patterns, (b) Raman spectra, and (c) PL spectra of Si₂Te₃ films grown on h-BN/SiO₂/Si and SiO₂/Si substrates 600-650 °C.

Table S1. Electrical transport characteristics of Si₂Te₃ FETs on the surface of different dielectrics. All the Si₂Te₃ films used in FETs were prepared in the temperature zone of 600-650 $^{\circ}$ C.

Growth	Thickness	Mobility	ON/OFF	dielectric	electrode
time	(nm)	(cm²V ⁻¹ s ⁻¹)			
(min)					
30	3.3	0.6	10 ³		
35	4.2	1.0	10 ³		10nm Ti
40	5.0	2.87	6x10 ²	300nm SiO ₂	&
45	5.8	3.19	5.5x10 ²		60nm Au
50	7.0	6.23	4x10 ²		
60	8.0	10.0	10 ²		
35	4	3.6	104	BN(2nm) &SiO ₂ (300nm)	10nm Pd
35	3.7	3.15	104		&
35	4	4.04	8x10 ³	BN(5nm) &	60nm Au
				SiO ₂ (300nm)	