Supporting Information

Atomic-scale tuning oxygen-related defects in Bi₂Te_{2.7}Se_{0.3} to simultaneously enhance Seebeck coefficient and electrical conductivity

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Fig. S1 (a) Resulting TiO₂ film thickness as a function of the ALD cycle count. (b) AFM image of 50 cycles ALD deposited TiO₂ film. (c) HRTEM image of the as-prepared BTS. HRTEM image of the TiO₂@BTS with (g) 30 ALD cycles, (e) 50 ALD cycles and (f) 70 ALD cycles.



Fig. S2 XRD pattern of the as-prepared sample with different ALD cycles after HP process.



Fig. S3 (a) XPS spectra for the as-prepared bulk sample and after Ar etch. (b) Typical lowmagnification STEM -HAADF image and (c) the corresponding EDS spectra. (d) The corresponding EDS elemental maps.



Fig. S4 Cross-sectional scanning TEM (STEM) images and elemental mapping of the grain boundary. (a) Low-magnification STEM -HAADF image. (b) HAADF-STEM image of the selected grain boundary. (c) high-magnification HAADF-STEM image of the grain boundary and the corresponding EDS elemental maps.



Fig. S5 TEM images of the as-prepared BTS sample. (a) TEM image (b) HAADF-STEM image of the BTS grain (c) high-magnification HAADF-STEM image.



Fig. S6 Lattice and bipolar thermal conductivity, $\kappa_{latt} + \kappa_{bi} = \kappa_{tot} - \kappa_e$.



Fig. S7 The low magnified TEM image of the bulk sample of S70 and the corresponding EDS elemental maps.



Fig. S8 Temperature dependences of the specific heat for the as-prepared BTS sample.

 Table S1 Density of the as-prepared samples.

Sample ID	ALD cycles	Density (g cm ⁻³)
S0	0	7.60
S10	10cycles	7.42
S30	30cycles	7.47
S50	50cycles	7.32
S70	70cycles	7.50
S90	90cycles	7.45