Supporting Information

Precision grain Boundary Engineering in commercial $Bi_2Te_{2.7}Se_{0.3}$ thermoelectric materials towards high performance

Shuankui Li, ^a Zhongyuan Huang, ^a Rui Wang, ^a Chaoqi Wang, ^a Wenguang Zhao, ^a Ni Yang, ^a Fusheng Liu, ^b Jun Luo, ^c Yinguo Xiao, *^a and Feng Pan*^a

^{*a*} School of Advanced Materials, Peking University Shenzhen Graduate School, Shenzhen, 518055, China.

^bCollege of Materials Science and Engineering, Shenzhen University and Shenzhen Key Laboratory of Special Functional Materials, Shenzhen, 518060, China.

^c School of Materials Science and Engineering, Shanghai University, Shanghai, 200444, China.

*Corresponding author: y.xiao@pku.edu.cn (Prof. Y.G. Xiao)

panfeng@pkusz.edu.cn (Prof. F. Pan)



Fig. S1. The microstructure of the as-prepared BTZ sample. (a) low magnification TEM image. (b) the widespread nanopores and Bi_2O_x nanoprecipitates at the grain boundaries. (c) the TEM image (c) and corresponding EDS line-scan profiles. (e) the HRTEM image of the O-related dislocation cluster.



Fig. S2.The microstructure of the as-prepared M-BTZ sample. (a) low magnification TEM image. (b) TEM image and corresponding EDS elemental maps. TEM images of (c) the nanopores and (d) sawtooth grain boundaries.



Fig. S3. XPS spectra for the as-prepared ALD coated BTS sample. (c) Ti 2p peaks of BT, BTZ and M-BTZ sample.



Fig. S4.The thermoelectric transport properties of the BZ samples with different ALD cycles. (a) Electrical conductivity (σ), (b) Seebeck coefficient (S), (c) power factor (S² σ). (d) Total thermal conductivity (κ_{tot}).



Fig. S5. The thermoelectric transport properties of the BT samples with different ALD cycles. (a) Electrical conductivity (σ), (b) Seebeck coefficient (S), (c) power factor (S² σ). (d) Total thermal conductivity (κ_{tot}).



Fig. S6. The thermoelectric transport properties of the BTZ samples with different ALD cycles. (a) Electrical conductivity (σ), (b) Seebeck coefficient (S), (c) power factor (S² σ). (d) Total thermal conductivity (κ_{tot}).



Fig. S7.The thermoelectric transport properties of the M-BTZ samples. (a) Electrical conductivity (σ), (b) Seebeck coefficient (S), (c) power factor (S² σ). (d) Total thermal conductivity (κ_{tot}).



Fig. S8. Thermoelectric figure of merit ZT of the as-prepared sample. (a) BZ samples. (b) BT samples. (c) BTZ samples. (d) M-BTZ samples.