# SUPPORTING INFORMATION

## Surfactant-Free Synthesis of $Bi_2Te_3$-Te Micro-Nano Heterostructure with Enhanced Thermoelectric Figure of Merit

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**The calculation of lattice contribution in thermal conductivity:**

According to Wiedemann-Franz law (Equation S1), the thermal conductivity can be split into two parts: the lattice part ($\kappa_L$) and electron part ($\kappa_e$). $\kappa_e$ is the product of the Lorentz number (L) with electrical conductivity and temperature. For heavily degenerated semiconductors (including our materials), $2.0 \times 10^{-8}$ $V^2$ $K^{-2}$ is accepted as the appropriate value of the Lorentz number L.

$$K = K_L + K_e = K_L + L\sigma T \quad \text{(S1)}$$

Due to the anisotropic properties of bismuth telluride, in this work we tested the in-plane charge transport properties and cross-plane thermal transport properties. Z. Ren and co-workers have done a very detailed study on the anisotropic TE properties of $Bi_2Te_{2.7}Se_{0.3}$.[1] In-plane conductivity (perpendicular to press direction) is reported to be higher for both electrical and thermal test than cross-plane (parallel to press direction) by about 10%-20%. Therefore, in this work, we applied a modified factor ~0.8 to estimate the cross-plane electrical conductivity from the experimental in-plane electrical conductivity, resulting in the modification of Equation S1 to Equation S2.

$$K_{L\perp} = K_{\perp} - K_{e\perp} \approx K_{\perp} - 0.8 \times L\sigma_{\parallel} T \quad \text{(S2)}$$

Here, $K_{\perp}$, $K_{L\perp}$ and $K_{e\perp}$ are thermal conductivities of the total, lattice part and electron part in the cross-plane orientation respectively; and $\sigma_{\parallel}$ is the electrical conductivity in the in-plane

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orientation. The estimated lattice contributions of the three samples (BiTe_bare, BiTe_101, and BiTe_51) in cross-plane direction are shown in the inset of Fig. 3 (D).

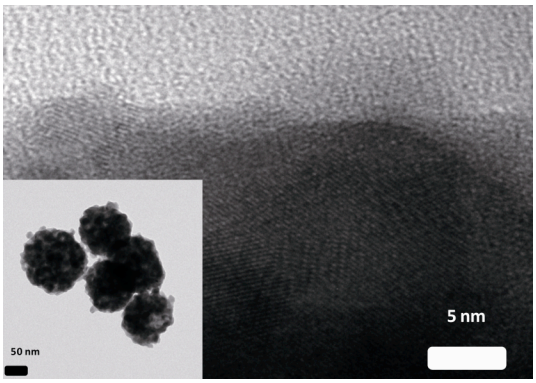


**Figure S1**. HR-TEM images of the solid bismuth telluride nanospheres; the inset is the overview image.

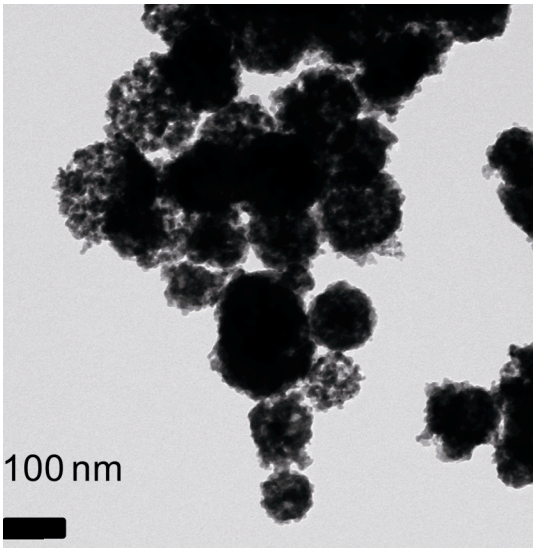


**Figure S2.** TEM images of some porous bismuth telluride nanospheres.

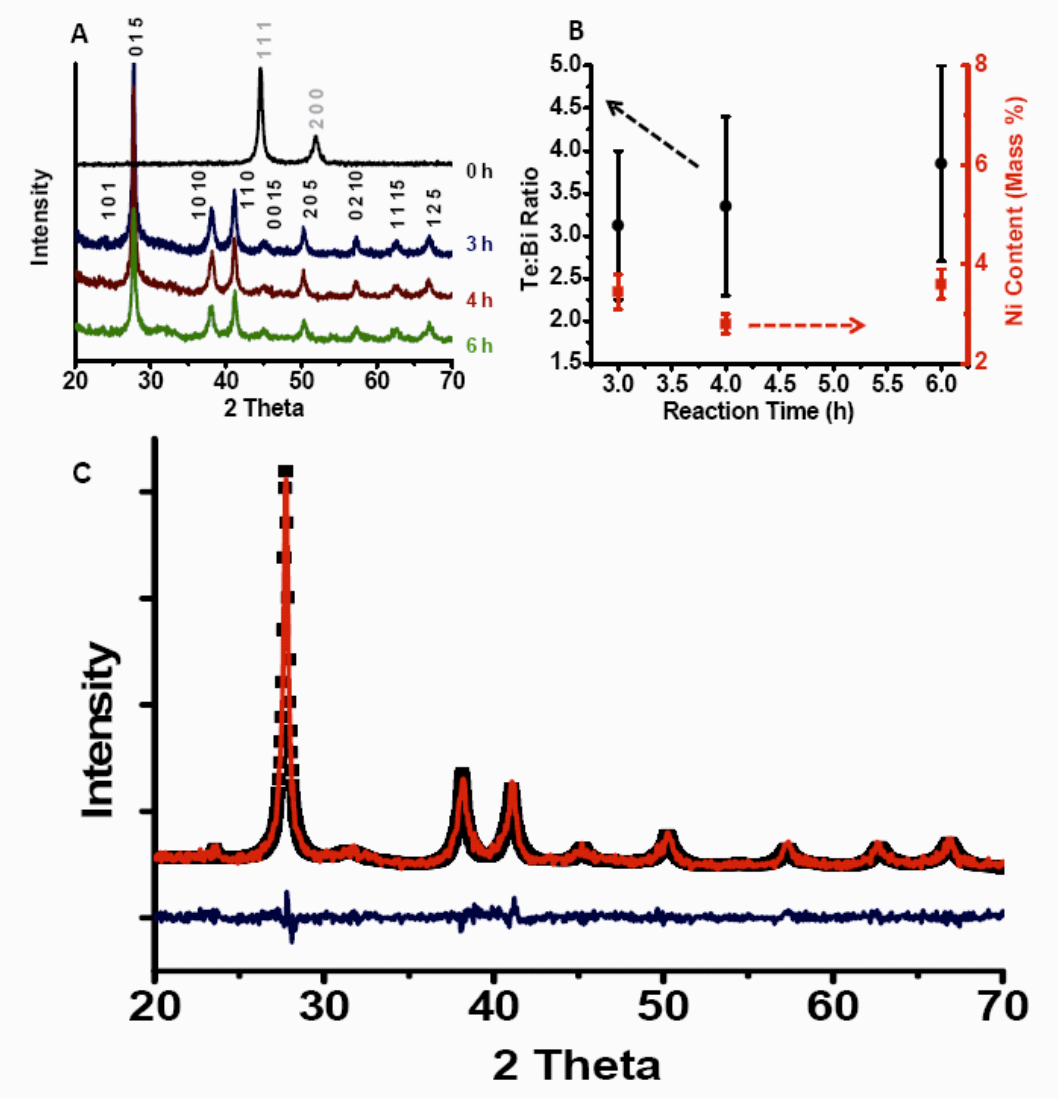


**Figure S3.** (A) XRD patterns of starting materials (0 h) and the products after reacting for 3 to 6 hours. Indexing according to space group $Fm\bar{3}m$ (0 h/nickel) and $R\bar{3}m$ (3-6 h/bismuth telluride) are shown. (B) ICP results of Te/Bi stoichiometry and Ni contents of as-prepared powders. (C) Rietveld refinement results of the product after reacting for 4 hour. (R value is 0.126)

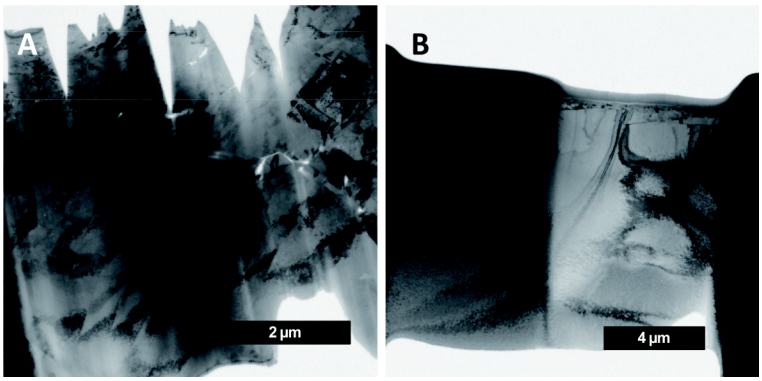


**Figure S4.** TEM images of prepared lamellas A and B. A shows polycrystalline structure and B is a single crystal.

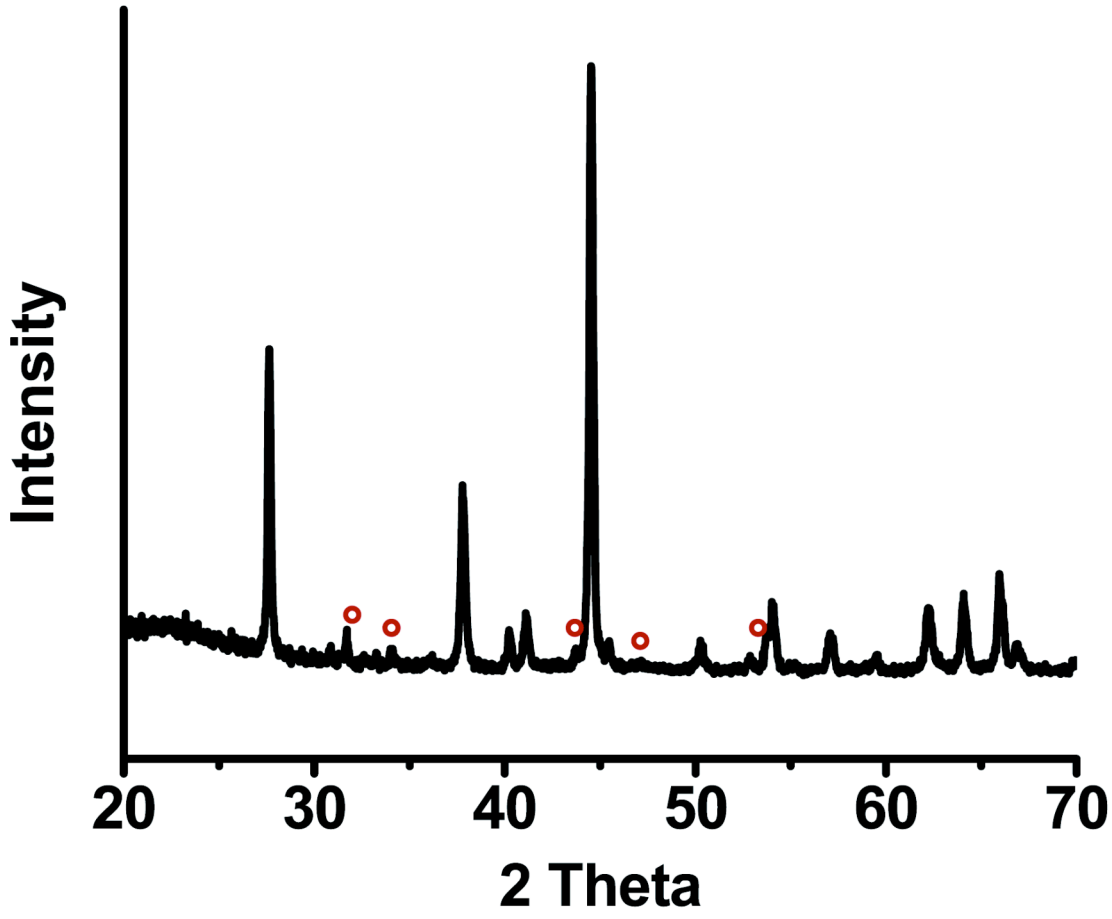


**Figure S5.** Enlarged XRD pattern of BiTe_51, the marked peaks are from crystal phase of $NiTe_2$ (PCPDF#89-2021)

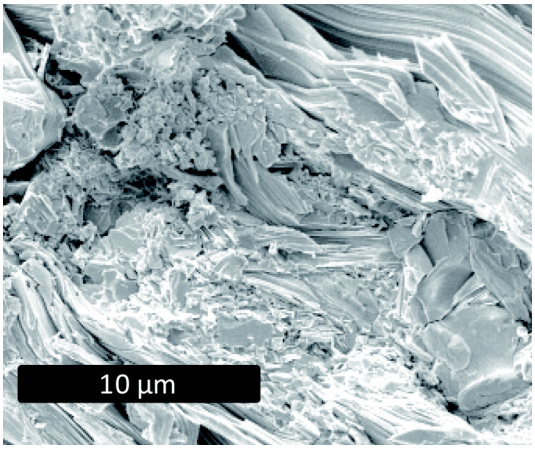


**Figure S6.** The selected area of EDX mapping.

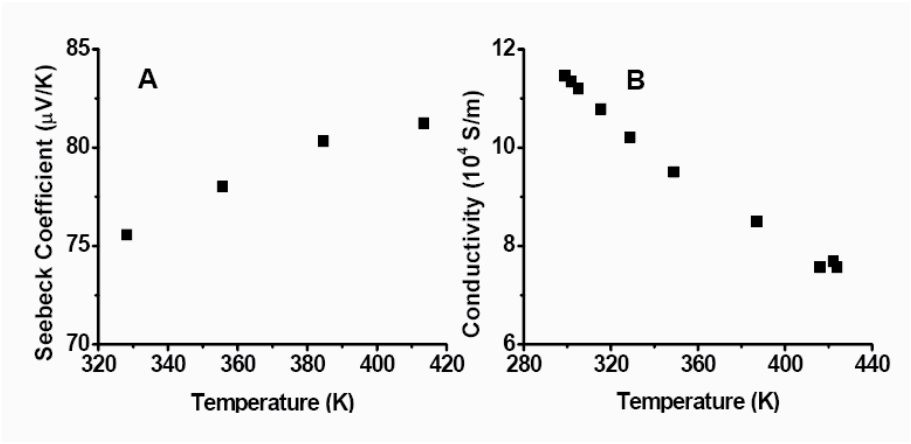


**Figure S7.** Temperature dependence of Seebeck coefficient (A) and conductivity (B) of BiTe_21

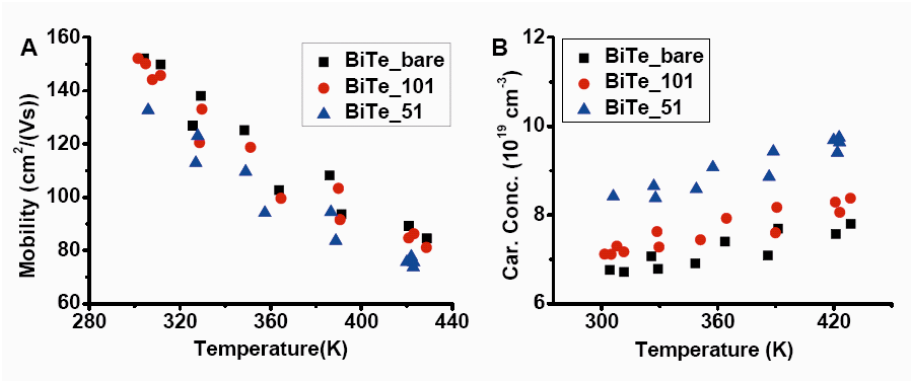


**Figure S8.** Temperature dependence of (A) mobility and (B) carrier concentrations of BiTe_bare, BiTe_101, and BiTe_51.

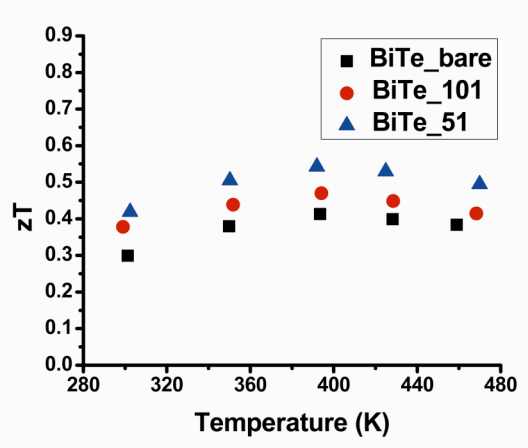


**Figure S9.** Temperature dependence of estimated figures of merit (zT) of BiTe_bare, BiTe_101 and BiTe_51.

Reference

1. Yan, X.; Poudel, B.; Ma, Y.; Liu, W. S.; Joshi, G.; Wang, H.; Lan, Y.; Wang, D.; Chen, G.; Ren, Z. F., Experimental Studies on Anisotropic Thermoelectric Properties and Structures of n-Type $Bi_2Te_{2.7}Se_{0.3}$. *Nano Lett.* **2010**, *10*, 3373-3378.