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Supporting Information

Construction of 3D-rGO network-wrapping architecture in $Yb_yCo_4Sb_{12}$/rGO composite for enhancing thermoelectric performance

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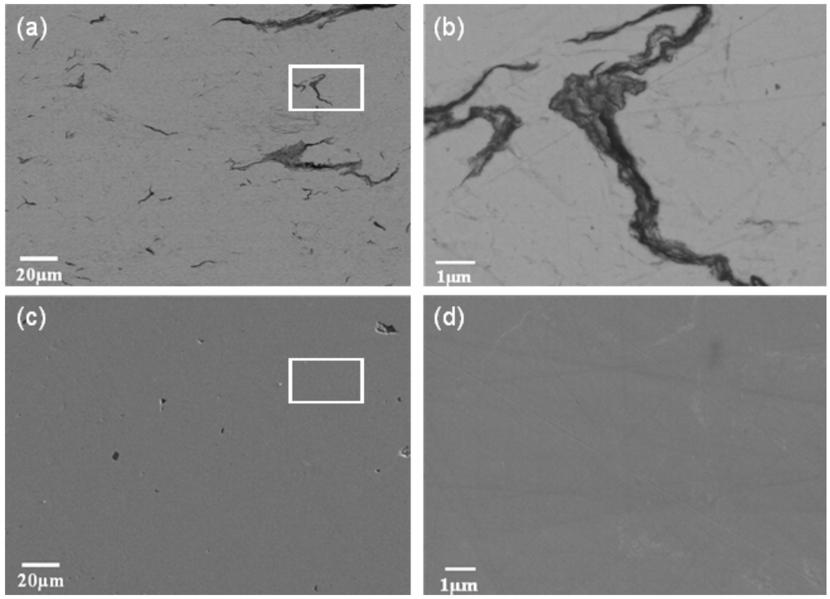

Figure S1 SEM images of bulk $Yb_{0.27}Co_4Sb_{12}$ /1.8 vol % rGO (a, b) particle dispersion sample; (c, d) 3D-rGO wrapping sample

For comparison, the microstructures of $Yb_{0.27}Co_4Sb_{12}$/y vol % rGO (y = 0.72, 1.8, 3.6) particle dispersion samples and 3D-rGO wrapping samples are characterized. The SEM photographs of the polished surface of the bulk y = 1.8 particle dispersion sample (Figure S1a,b) show that rGO was seriously agglomerated, even of a thickness of ~1 μm. On the contrast, no distinct second phase was found in the polished surface of y = 1.8 3D-rGO wrapping sample (Figure S1c,d), attesting to a homogeneous distribution of rGO realized by in-situ reduction method with GO as the precursors.

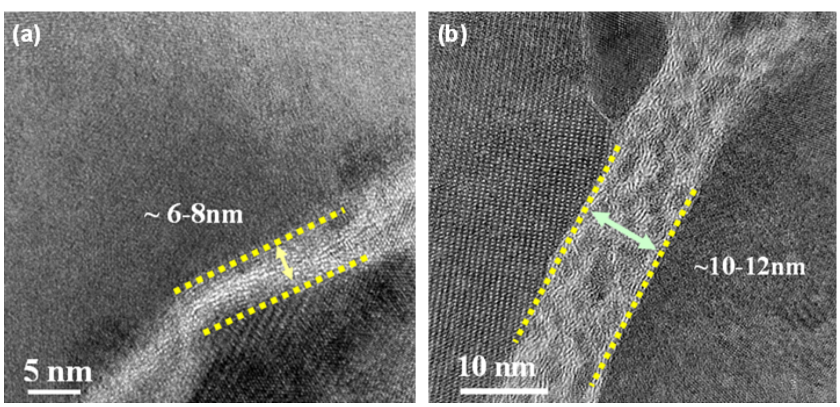

Figure S2 High-magnification TEM images showing rGO layers in 3D-rGO wrapping samples: (a) ~6-8 nm thick for y = 1.8 3D-rGO wrapping-sample; (b) ~10-12 nm thick for y = 3.6 3D-rGO wrapping-sample.

Figure S2 shows high-magnification TEM images for rGO-3D network-wrapping samples. It reveals that the rGO wrapping layer grows from ~3-5 nm thick (y = 0.72,

Figure 3d) to ~6-8 nm thick (y = 1.8, Figure S2a) and ~10-12 nm thick (y = 3.6, Figure 2b), which deteriorates the carrier mobility and leads to decrease of electric conductivity.

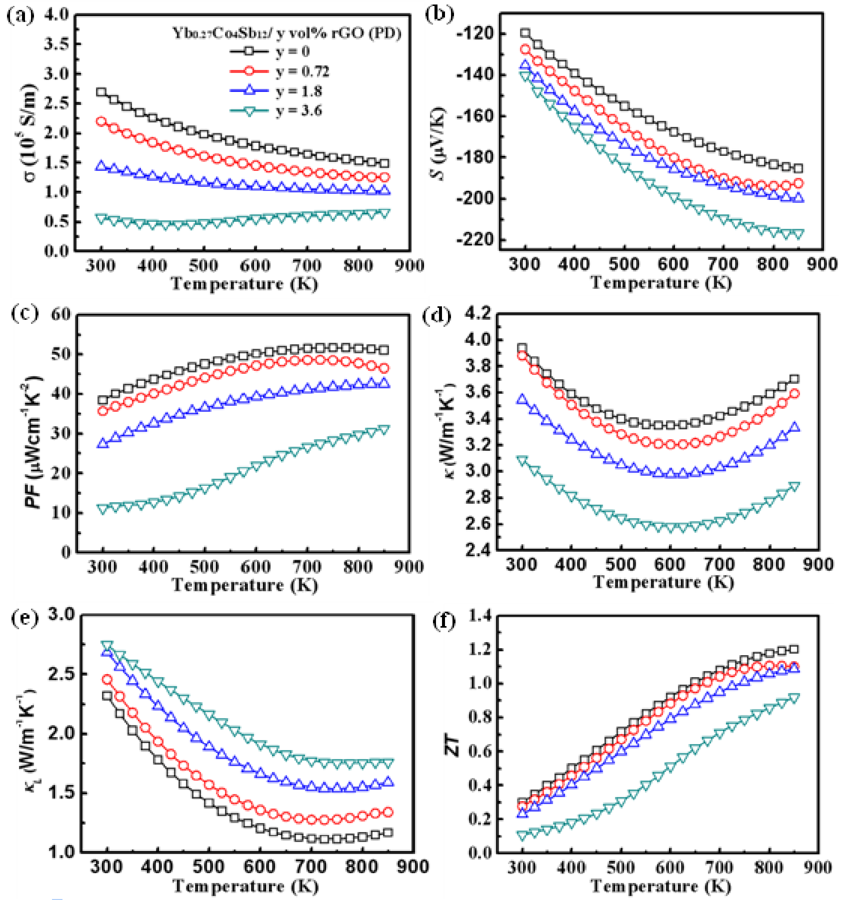

Figure S3 Thermoelectric properties as a function of temperature for the particle dispersion (PD) samples of $Yb_{0.27}Co_4Sb_{12}$/y vol % rGO (y = 0, 0.72, 1.8, 3.6): (a) electrical conductivity , (b) Seebeck coefficient, (c) power factor, (d) total thermal conductivity, (e) lattice thermal conductivity, (f) Figure of merit *ZT*.

Figure S3a shows the temperature-dependent electrical conductivity of particle dispersion samples. Due to the effect of agglomeration, the electrical conductivity deteriorates vastly comparing with 3D-rGO wrapping samples with the same y value. Figure S3b,c indicate that $|S|$ is increased with y, the power factor (σS^2) goes down with y addition, due to the fact that increase of $|S|$ can not compensate the deterioration of electrical conductivity. The total thermal conductivity (Figure S3d) is depressed mainly due to the decrease of electronic thermal conductivity. But the lattice thermal conductivity (Figure S3e) is increased with y, due to agglomeration of rGO by particle dispersion method. Figure S3f shows *ZT* as a function of temperature for the particle dispersion samples of $Yb_{0.27}Co_4Sb_{12}$/y vol% rGO (y = 0, 0.72, 1.8, 3.6) from 300-850K. The *ZT* values decrease with y, manifesting that particle dispersion method of SKD/rGO is unwise for ZT-plus due to the inferior dispersibility of rGO.