**Supplementary Materials**

**InSb nanoparticles dispersion in Yb-filled Co4Sb12 improves the thermoelectric performance**

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1. **Rietveld refinement analysis of XRD patterns**



**Figure S1** (a) Rietveld refinement of powder XRD pattern of Yb0.2Co4Sb12.



**Figure S1** (b) Rietveld refinement of powder XRD pattern of (InSb)0.2+Yb0.2Co4Sb12.



**Figure S1** (c) Rietveld refinement of powder XRD pattern of (InSb)0.3+Yb0.2Co4Sb12.



**Figure S1** (d) Rietveld refinement of powder XRD pattern of (InSb)0.4+Yb0.2Co4Sb12.



**Figure S1** (e) Rietveld refinement of powder XRD pattern of ball milled InSb.

1. **EPMA-WDS elemental mapping**



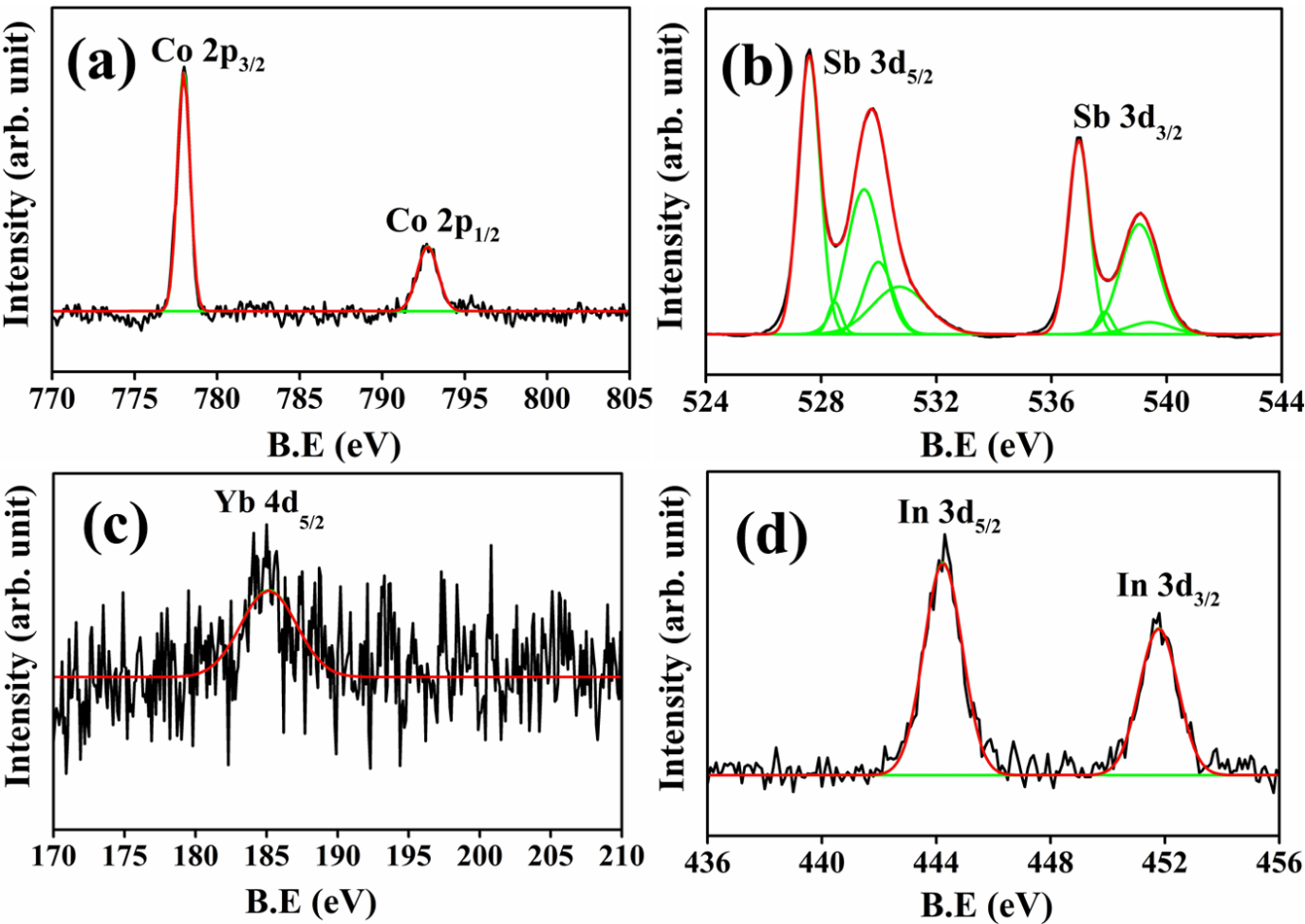
**Figure S2** (a) SE micrograph, (b) BSE micrograph of (InSb)0.4+Yb0.2Co4Sb12, EPMA-WDS elemental mapping of (c) Co, (d) Sb, (e) Yb, (f) In and (g) O.

1. **Inverse pole figure map**



**Figure S3** Inverse pole figure (IPF) map of (a) Co4Sb12, and (b) InSb phases showing grain size and their distribution, normal direction (ND) IPF of (c) Co4Sb12, and (d) InSb phase of (InSb)0.4+Yb0.2Co4Sb12.

1. **X-ray photoelectron spectroscopy**



**Figure S4** XPS spectra of the (a) Co 2p, (b) Sb 3d, (c) Yb 4d and (d) In 3d energy levels of the (InSb)0.2+Yb0.2Co4Sb12 composite.

Figs. S4(a-d) present the X-ray photoelectron spectra of Co, Sb, Yb, and In, which helped to identify all the elements' oxidation states and their bonding type. The XPS spectrum of Carbon 1s (at 284.4 eV) was also collected to calibrate the XPS spectra of all the elements, which showed a small shift towards higher binding energies. The XPS spectrum of Co (evaluated using a Gaussian fitting function [Fig. S4(a)]), exhibited peaks at 777.97 eV and 792.75 eV associated with the 2p3/2 and 2p1/2 energy levels, respectively. No significant shift in the XPS spectrum with respect to elemental Co [NIST XPS database] demonstrated covalent bonding between Co and Sb. A +3 oxidation state of Co was detected in Yb0.2Co4Sb12, verified from the absence of satellite features in the spectrum. The XPS spectrum of Sb 3d energy levels, shown in Fig. S4(b), reveals that each peak assigned to 3d5/2 and 3d3/2 energy levels is split into two peaks, mainly due to the two different Sb-Sb bond lengths. This particular characteristic of the XPS spectrum of Sb was observed by several other groups [1-3]. The different bonding states of Sb were demonstrated by deconvoluting the XPS peak associated with Sb 3d5/2 and Sb 3d3/2 energy levels into sub-peaks, and their percentages were evaluated from the area under the individual peak [3]. The first sub-peak at a binding energy of 527.52 eV with a content of 24.06 % is assigned to the short Sb-Sb bond [3]. A higher electron density at the short Sb-Sb bond generated the peak at a lower binding energy value. The next smallest sub-peak at 528.47 eV of 1.70 % is allotted to the Co-Sb covalent bond [3]. The consecutive two peaks are due to the large Sb-Sb bond, one at 529.49 eV with 19.86 % and another at 529.98 eV with a content of 8.21 %. The total content assigned to the large Sb-Sb bond is 28.07 %. The electron density is smaller at this bond than that of the short Sb-Sb bond. The peak at 530.72 eV with a content of 10.17 % is attributed to the bond perpendicular to the [Sb4]4- ring with the lowest electron density. Yb showed a faint signal associated with the Yb 4d5/2 energy level with a peak at 185.15 eV, indicating a +3 oxidation state of Yb [Fig. S4(c)]. Contrary to our observation, Nolas et al. [4] reported a fluctuation in the valence state of Yb between +2 and +3 from magnetic susceptibility measurement. Anno *et al*. [5] also observed an XPS peak at ~186 eV associated with the +3 oxidation state and a tiny peak at ~182 eV associated with the +2 oxidation state in the XPS spectrum for different content of Yb in Co4Sb12 voids, which also indicated an intermediate valence state between +2 and +3. However, in our experiment, the peak associated with the +2 oxidation state of Yb at ~182 eV could not be detected, probably due to the weak contribution from Yb. The peaks associated with In 3d5/2 and In 3d3/2 energy levels were obtained at 444.24 eV and 451.78 eV binding energies, respectively [Fig. S4(d)]. The similar binding energies of In 3d energy levels were reported (444.2 eV and 451.8 eV) for bulk InSb [6]. In conclusion, XPS analysis showed Co-Sb covalent bonding and a +3 oxidation state of Yb, indicating probable charge transfer from Yb to Co4Sb12 and consequently, a low electrical resistivity of Yb0.2Co4Sb12.

1. **Lorenz number**



**Figure S5** Temperature dependence of Lorenz number of (InSb)x + Yb0.2Co4Sb12 (where x=0, 0.1, 0.2, 0.3, 0.4).

1. **Reproducibility of thermoelectric properties**



**Figure S6** Temperature dependence of the electrical resistivity of (InSb)x+Yb0.2Co4Sb12 (x=0.1 and 0.3).



**Figure S7** Temperature dependence of the Seebeck coefficient of (InSb)x+Yb0.2Co4Sb12 (x=0.1 and 0.3).



**Figure S8** Temperature dependence of the power factor of (InSb)x+Yb0.2Co4Sb12 (x=0.1 and 0.3).

**Supplementary References**

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