Supplementary Information

Optimization of the thermoelectric performances for SnTe alloys by In & Se co-doped

Hongchao Wang[1,3], Teng Wang[1], Junphil Hwang[2], Wenbin Su[1], Hoon Kim[2], Jinze Zhai[1], Xue Wang[1], Chunlei Wang[1*], Woochul Kim[2*]

1. School of Physics, State Key Laboratory of Crystal Materials, Shandong University, Jinan, China

2. School of Mechanical Engineering, Yonsei University, Seoul, Korea

3. State Key Laboratory of Metastable Materials Science and Technology, Yanshan University, Qinhuangdao, China

* Corresponding authors:

E-mail: woochul@yonsei.ac.kr (W. Kim)

E-mail: wangcl@sdu.edu.cn (C. Wang)

Thermal conductivity calculation. In Figure S4, theoretical thermal conductivities, k, were calculated by the Debye-Callaway model[1] as shown in Eq. * MERGEFORMAT (1) where I_1, I_2 and I_3 are shown in Eq. * MERGEFORMAT (2).

$$k = \frac{k_B}{2\pi v_g}\left(\frac{k_B T}{\mathrm{h}}\right)^3 \left(I_1 + \frac{I_2^2}{I_3}\right) \quad \text{* MERGEFORMAT (1)}$$

$$I_1 = \int_0^{\theta_D/T} \tau_c \frac{x^4 e^x}{\left(e^x - 1\right)^2} dx$$

$$I_2 = \int_0^{\theta_D/T} \frac{\tau_c}{\tau_N} \frac{x^4 e^x}{\left(e^x - 1\right)^2} dx \qquad \text{* MERGEFORMAT}$$

$$I_3 = \int_0^{\theta_D/T} \frac{1}{\tau_N} \left(1 - \frac{\tau_c}{\tau_N}\right) \frac{x^4 e^x}{\left(e^x - 1\right)^2} dx$$

(2)

Here, k_B, $\hbar$, T, and v_g are Boltzmann constant, reduced Planck constant, absolute temperature, and phonon group velocity, respectively. τ_N and τ_c are the phonon relaxation time for the normal scattering and the combined phonon relaxation time. τ_c is calculated using Matthiessen's rule as shown in Eq. * MERGEFORMAT (3).

$$\tau_c^{-1} = \tau_{N,L}^{-1} + \tau_{N,T}^{-1} + \tau_U^{-1} + \tau_A^{-1} + \tau_B^{-1} \text{* MERGEFORMAT}$$

(3)

$\tau_{N,L}$, $\tau_{N,T}$, τ_U, τ_A, and τ_B are the relaxation times of normal scattering for longitudinal mode and transverse mode[2], Umklapp scattering[3], alloy scattering[3], and boundary scattering. Each relaxation time was evaluated as in Eq. * MERGEFORMAT (4)-* MERGEFORMAT (8).

$$\tau_{N,L}^{-1} = \frac{k_B^3 \gamma^2 V_0}{M_{avg} \hbar^2 v_L^5} \omega^2 T^3 \qquad \text{* MERGEFORMAT (4)}$$

$$\tau_{N,T}^{-1} = \frac{k_B^4 \gamma^2 V_0}{M_{avg} \hbar^3 v_L^5} \omega T^4 \qquad \text{* MERGEFORMAT (5)}$$

$$\tau_U^{-1} = \frac{2\gamma^2 k_B}{\mu V_0 \omega_D} \omega^2 T \qquad \text{* MERGEFORMAT (6)}$$

$$\tau_A^{-1} = \frac{V_0 \Gamma}{4\pi v_g^3} \omega^4 \qquad \text{* MERGEFORMAT (7)}$$

$$\tau_B^{-1} = \frac{v_g}{d} \qquad \text{* MERGEFORMAT (8)}$$

Here, ω and ω_D are the phonon frequency and the Debye frequency. γ, M_{avg}, V_0, and μ are the Grüneisen parameter, the average atomic mass, and the volume per atom, and the shear modulus, respectively. d is the grain size and assumed to be 800 nm. The relation of the average group velocity, v_g, and the group velocities of each mode is shown in Eq. * MERGEFORMAT (9).

$$v_g^{-3} = \left(\frac{1}{3} v_L^{-3} + \frac{2}{3} v_T^{-3} \right) \qquad \text{* MERGEFORMAT (9)}$$

The mass-fluctuation phonon scattering parameter Γ is represented as in Eq. * MERGEFORMAT (10) where f_i is the fractional concentration of the atom and M_i is the atomic mass of each atom i.

$$\Gamma = \sum_i f_i \left(1 - \frac{M_i}{M_{avg}} \right)^2 \qquad \text{* MERGEFORMAT (10)}$$
$$M_{avg} = \sum_i f_i M_i$$

Required parameters are the followings. Phonon group velocities were chosen as 4000 m/s for the longitudinal mode and 1250 m/s for the transverse mode[4]. Debye temperature was set to be 132K[5] and shear modulus was 32.2 GPa as the Voigt average[6]. Grüneisen parameter was picked as a fitting parameter as chosen as 1.7, and the grain size for the boundary scattering was assumed to be 800 nm.

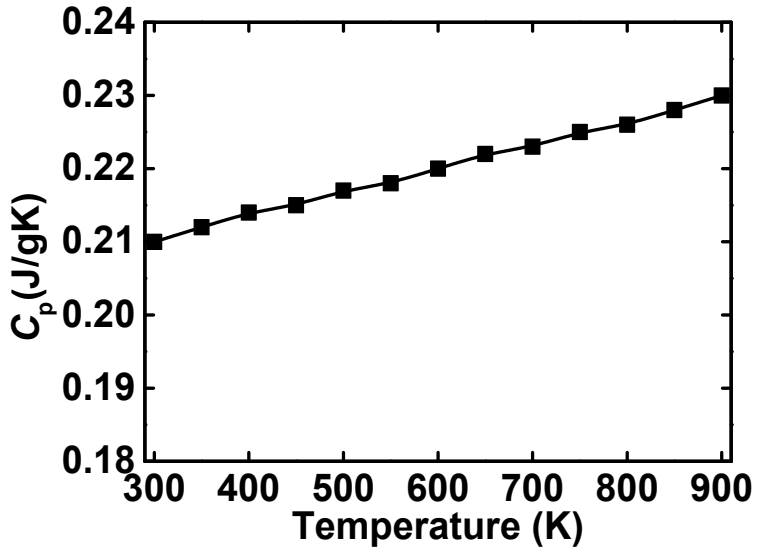

Figure S1 The used heat capacity, C_p, of each sample, which was estimated by the relationship C_p (K_B Per atom)= 3.07 + 4.7 $\times 10^{-4} \times$ (T/K − 300).

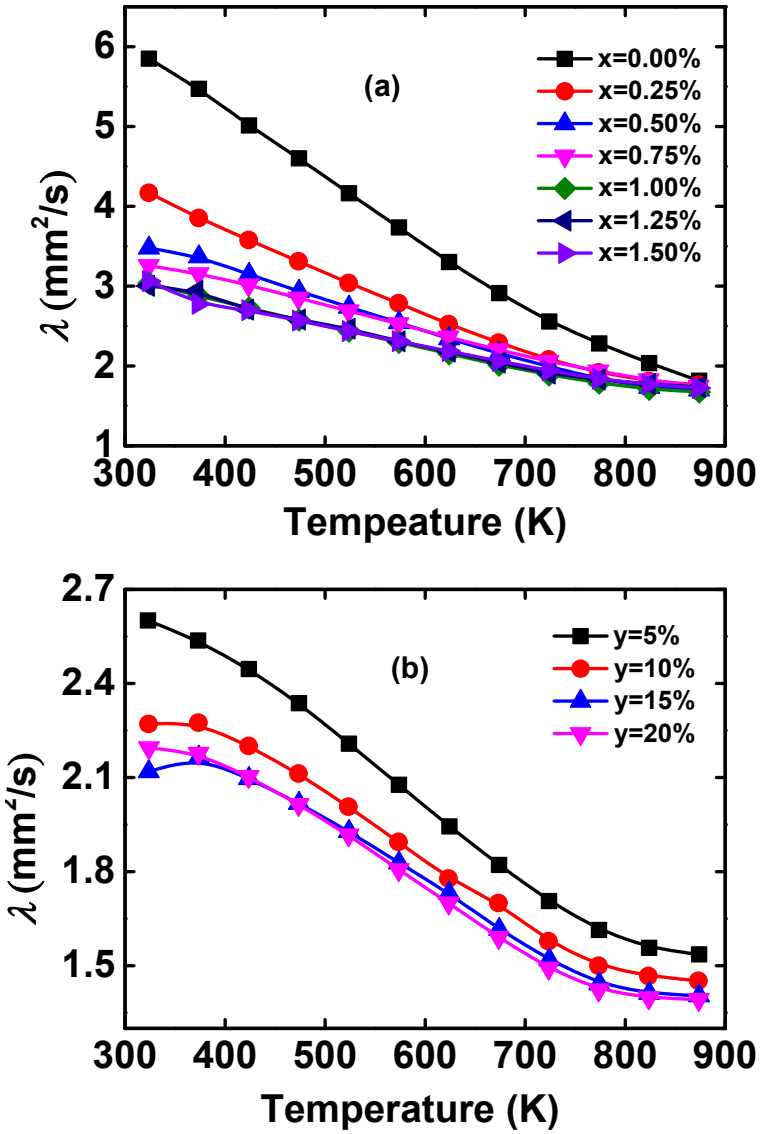

Figure S2 Thermal diffusivity, λ, for $Sn_{1-x}In_xTe$ **(a)**, and $Sn_{0.99}In_{0.01}Te_{1-y}Se_y$ alloys **(b)**.

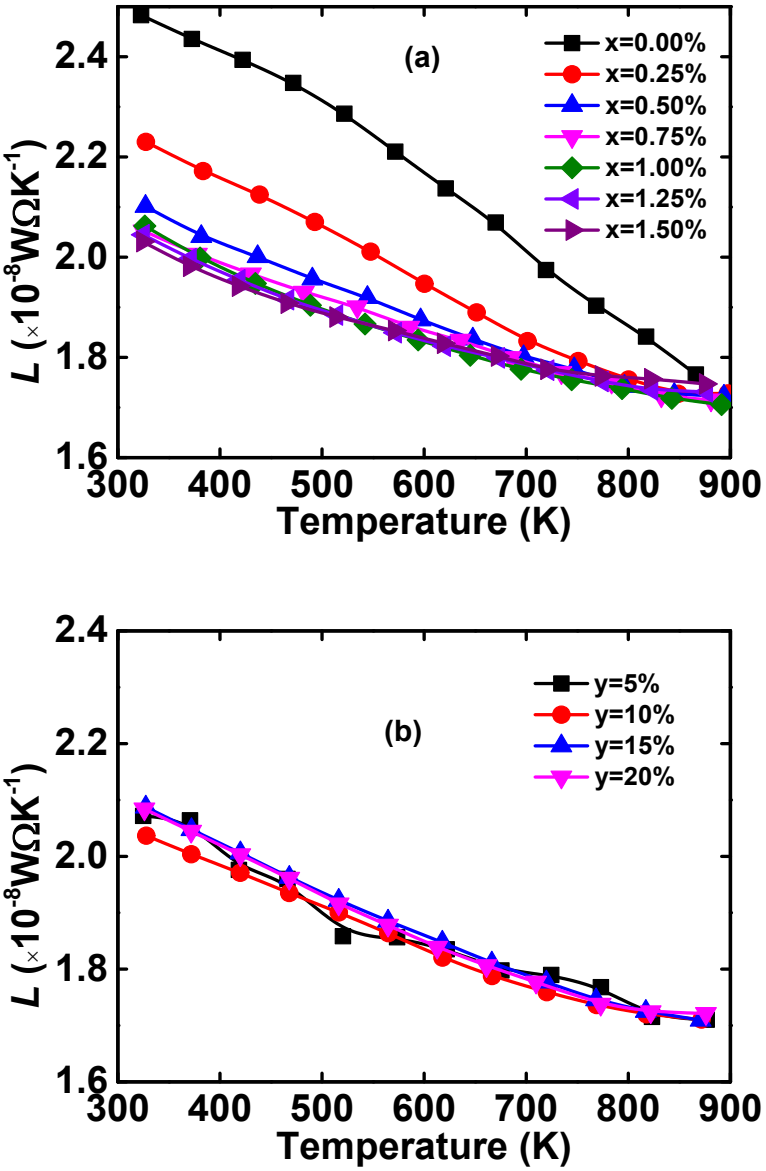

Figure S3 The Lorenz number, *L*, for $Sn_{1-x}In_xTe$ **(a)**, and $Sn_{0.99}In_{0.01}Te_{1-y}Se_y$ alloys **(b)**, which were derived from the equation of L=1.5+exp (-|S|/116).

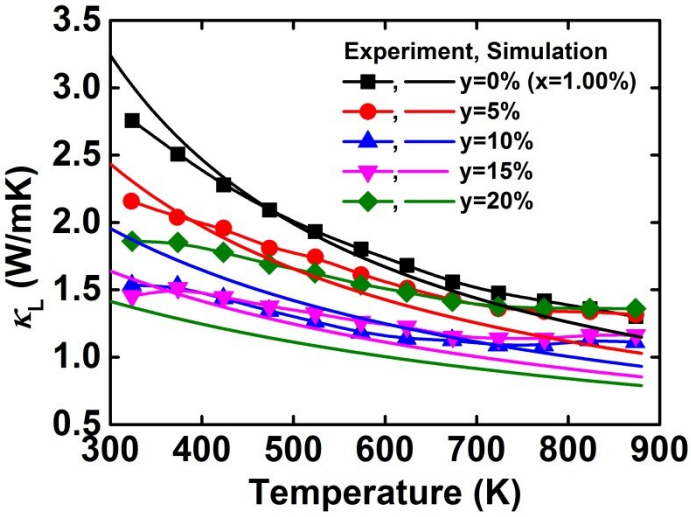

Figure S4 Lattice thermal conductivities of $Sn_{0.99}In_{0.01}Te_{1-y}Sey$ (Figure 8(b)) and theoretical thermal conductivities by the Debye-Callaway model.

Table S1. The lattice constants, a, V, theoretical densities, d_T, experimental densities d_E, and relative densities d_R, for $Sn_{1-x}In_xTe$ alloys.

x	a (Å)	V (Å^3)	d_T (g/cm^3)	d_E (g/cm^3)	d_R
0.00%	6.331	253.75	6.45	6.35	98%
0.25%	6.329	253.51	6.46	6.32	98%
0.50%	6.327	253.27	6.46	6.30	98%
0.75%	6.326	253.17	6.46	6.28	97%
1.00%	6.325	253.03	6.46	6.25	97%
1.25%	6.326	253.16	6.45	6.23	97%
1.50%	6.325	253.02	6.45	6.23	97%

Table S2. The lattice constants, a, V, theoretical densities, d_T, experimental densities d_E, and relative densities d_R, for $Sn_{0.99}In_{0.01}Te_{1-y}Se_y$ alloys.

y	a (Å)	V (Å^3)	d_T (g/cm^3)	d_E (g/cm^3)	d_R
5%	6.278	247.52	6.54	6.23	95%
10%	6.264	245.81	6.53	6.22	95%
15%	6.280	247.62	6.41	6.21	97%
20%	6.261	245.46	6.40	6.22	97%

References

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