Supporting Information

Maximizing phonon scattering efficiency by Cu_2Se alloying in AgCuTe thermoelectric materials

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Results and Discussion:

The Lorenz number

In our work, the Lorenz number used in the calculation of κ_e is obtained based on the single parabolic band:

$$L=\left(\frac{k_B}{e}\right)^2\left(\frac{(r+7/2)F_{r+5/2}(\eta)}{(r+3/2)F_{r+1/2}(\eta)}-\left[\frac{(r+5/2)F_{r+3/2}(\eta)}{(r+3/2)F_{r+1/2}(\eta)}\right]^2\right) \quad (1)$$

In the above equation, $F_n(\eta)$ is defined as

$$F_n(\eta)=\int_0^\infty \frac{\chi^n}{1+e^{\chi-\eta}}d\chi \quad (2)$$

$$\eta=\frac{E_f}{k_BT} \quad (3)$$

Where $F_n(\eta)$ is the n-order Fermi integral, and η is called the reduced Fermi energy. η can be obtained by the following formula:

$$S = \pm \frac{k_B}{e}\left(\frac{(r+5/2)F_{r+3/2}(\eta)}{(r+3/2)F_{r+1/2}(\eta)} - \eta \right) \quad (4)$$

In the above equation, η can be obtained by the measured Seebeck coefficients. The scattering factor r is $-1/2$ in acoustic phonon scattering.

The modified Cahill's model calculation

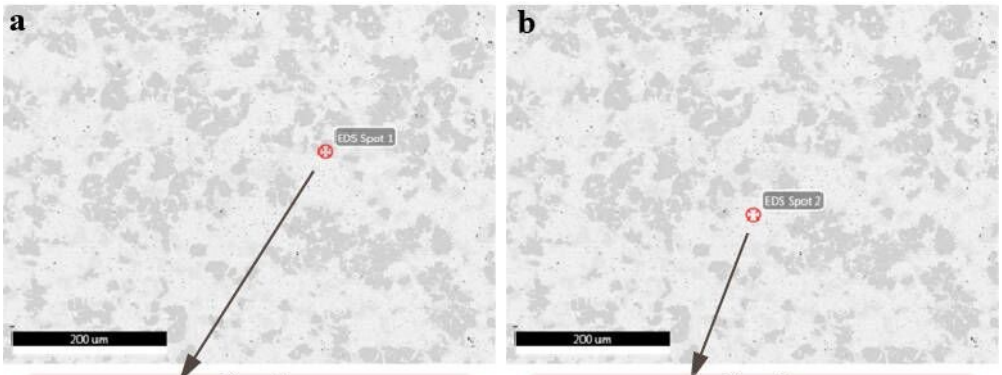
For AgCuTe, it is widely accepted that the transverse component of sound velocity is reduced or completely vanished at higher temperature due to its liquid-like behavior[1,2,3]. Because AgCuTe is a liquid-like material, the modified Cahill's model is used to calculate the minimum lattice thermal conductivity[4]:

$$\kappa_{\min} = \frac{1}{2}\left(\frac{\pi}{6}\right)^{1/3} \kappa_B V^{-2/3}(v_1) \quad (1)$$

where κ_B is the Boltzmann constant, V is the average volume per atom and v_1 is the longitudinal sound velocity. The v_1 can be calculated by[5]

$$v_1 = \sqrt{\frac{B + 4/3G}{\rho}} \quad (2)$$

Where B is the bulk modulus, G is the shear modulus and ρ is the density of a compound. Based on statistical-learning prediction of the bulk and shear modulus of cubic AgCuTe in the materials project[6], we get that B is 57.3 and G is 23.6. Therefore, the minimum lattice thermal conductivity was calculated to be ~0.21 $Wm^{-1}K^{-1}$ for AgCuTe.



Spot1

Element	Weight %	Atomic %	Error %
Ag	0.90	0.71	16.43
Te	49.48	32.95	1.98
Cu	49.62	66.34	3.92

Spot2

Element	Weight %	Atomic %	Error %
Ag	65.38	67.23	1.90
Te	31.56	27.44	3.65
Cu	3.05	5.33	20.03

Fig. S1 The percentage diagram of the EDS element content of the secondary phase in AgCuTe-1%Cu_2Se.

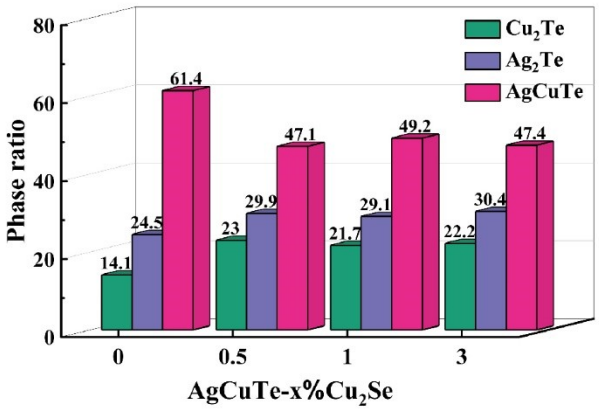


Fig. S2 The calculation results of the phases content in the AgCuTe - x%Cu_2Se (x = 0, 0.5, 1 and 3) sample.

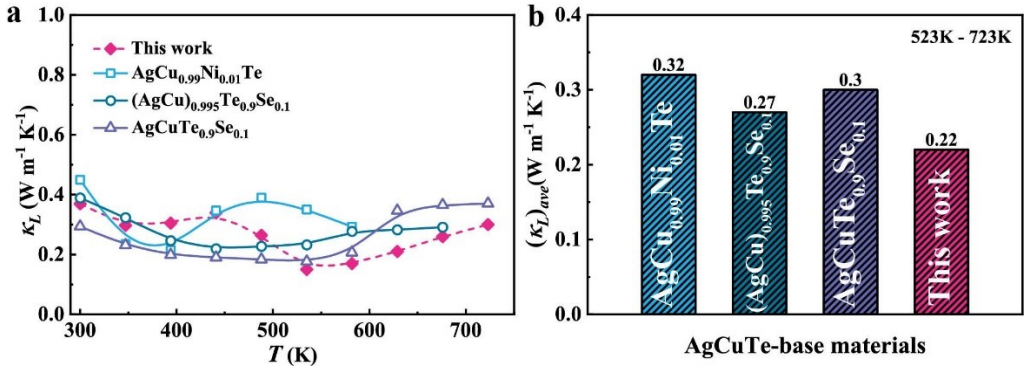


Fig. S3 a) Comparison of lattice thermal conductivity (κ_L) as a function of temperature in AgCuTe-based materials[7,8,9]; b) comparison of temperature-dependent average κ_L value in AgCuTe-based materials. Among them, due to the lower test temperature of $AgCu_{0.99}Ni_{0.01}Te$[7], it only calculated the average κ_L from 523 K to 582 K.

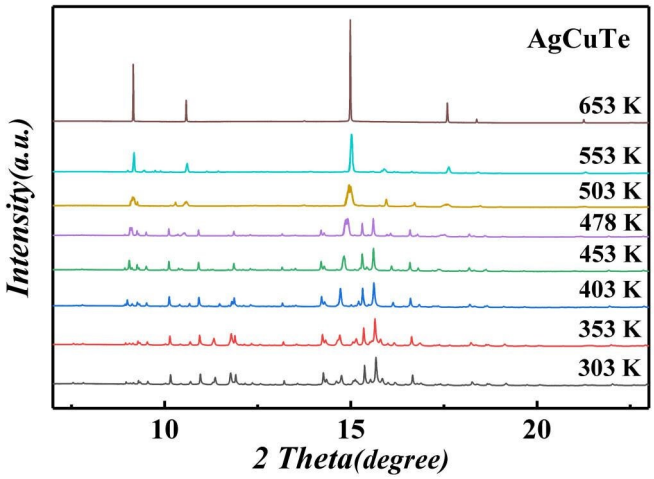


Fig. S4 Synchrotron powder diffraction (SPD) patterns of AgCuTe collected with the temperature increasing from 303 K to 653 K.

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