Supporting information of

Homogenizing Liquid-like Cu Sublattice for High-performance $Cu_2Te_{0.5}Se_{0.5}$ Thermoelectric Materials

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Contents

1. Details in the Theoretical Calculations

1.1 Lorenz number[1]

The total thermal conductivity κ_{tot} is divided into electrical (κ_{ele}) and lattice thermal conductivity (κ_{lat}), the subtraction of κ_{ele} from κ_{tot} is calculated by Wiedeman-Franz relation $\kappa_{lat} = \kappa_{tot} - \kappa_{ele}$ ($\kappa_{ele} = L \cdot \sigma \cdot T$), where σ and L is electrical conductivity and Lorenz number, respectively. L was calculated according to the equation (S1 – S3).

$$L = \left(\frac{k_B}{e}\right)^2 \left[\frac{3F_0(\eta)F_2(\eta) - 4F_1(\eta)^2}{F_0(\eta)^2}\right] \tag{S1}$$

$$S = \pm \frac{k_B}{e}\left(\frac{2F_1(\eta)}{F_0(\eta)} - \eta\right) \tag{S2}$$

$$F_n(\eta) = \int_0^\infty \frac{\chi^n}{1 + e^{\chi - \eta}} d\chi \tag{S3}$$

where S is the Seebeck coefficient, $F_n(\eta)$ is the nth order Fermi integral, η is the reduced Fermi energy, e is the charge of an electron, and k_B is the Boltzmann constant.

1.2 Weighted mobility[2]

To evaluate the manufacturing of engineering semiconductor devices, the understanding of the charge carrier mobility is required. At room temperature and above, and for mobilities as low as 10^{-3} $cm^2 \cdot V^{-1} \cdot s^{-1}$, the weighted mobility μ_w can be calculated from the Seebeck coefficient and electrical resistivity measurements according to the equation (S4).

$$\mu_w = 331 \frac{cm^2}{Vs}\left(\frac{m\Omega\, cm}{\rho}\right)\left(\frac{T}{300\,K}\right)^{-3/2}\left[\frac{exp\left[\frac{|S|}{\kappa_B/e} - 2\right]}{1 + exp\left[-5\left(\frac{|S|}{\kappa_B/e} - 1\right)\right]} + \frac{\frac{3}{\pi^2}\frac{|S|}{\kappa_B/e}}{1 + exp\left[5\left(\frac{|S|}{\kappa_B/e} - 1\right)\right]}\right]$$

(S4)

where ρ is the electrical resistivity measured in $m\Omega \cdot cm$, T is the absolute temperature in K, S is the Seebeck coefficient, and $\kappa_B/e = 86.3$ $\mu V \cdot K^{-1}$.

1.3 Theoretical lattice thermal conductivity [3]

The minimum limit of the theoretical lattice thermal conductivity (κ_{min}) was estimated to be based on Cahill's formulation

$$\kappa_{min} = \left(\frac{\pi}{6}\right)^{1/3} k_B n^{2/3} \sum_i v_i \left(\frac{T}{\Theta_i}\right)^2 \int_0^{\Theta_i/T} \frac{x^3 e^x}{(e^x - 1)^2} dx \tag{S5}$$

The sum is taken over the three sound modes (two transverse and one longitudinal) with speeds of sound v_i, Θ_i is the cutoff frequency for each polarization expressed in degress K, $\Theta_i = v_i(\hbar/k_B)(6\pi^2 n)^{1/3}$, and n is number density of atoms.

2. Supporting Table

Table S1. Density of the Quench at liquid, Quench at cubic, and Quench at trigonal samples after SPS process.

Samples	Density (g cm^{-3})	Relative density (%)
Quench at liquid	7.41	99.65
Quench at cubic	7.46	98.89
Quench at trigonal	7.48	98.72

3. Supporting Figures

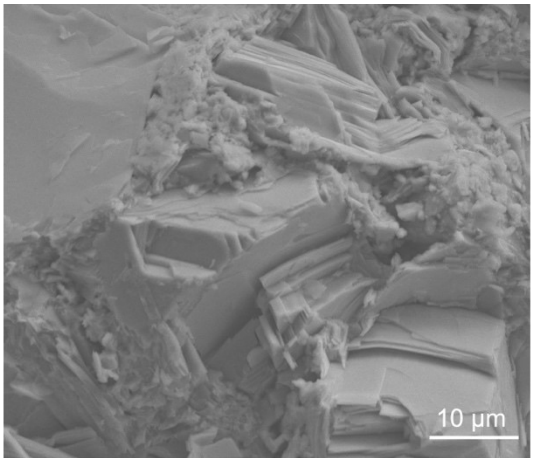

Figure S1. The scanning electron microscopy (SEM) image for $Cu_2Te_{0.5}Se_{0.5}$ with layered cleavage plates in nanoscale

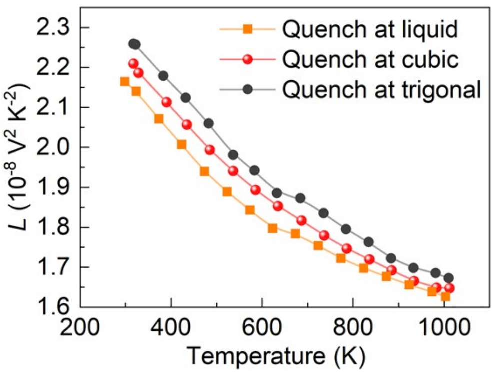

Figure S2 The Lorenz number (L) of the of Quench at liquid, Quench at cubic, and Quench at trigonal samples with respect to temperature.

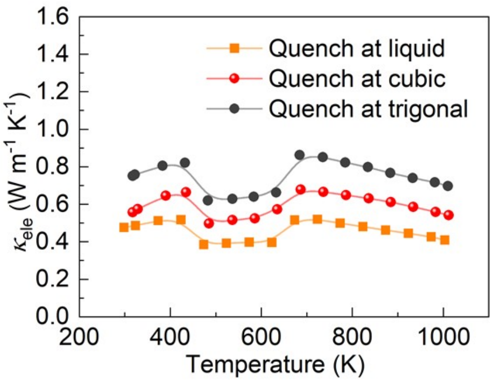

Figure S3 The temperature-dependent κ_{ele} for Quench at liquid, Quench at cubic, and Quench at trigonal samples

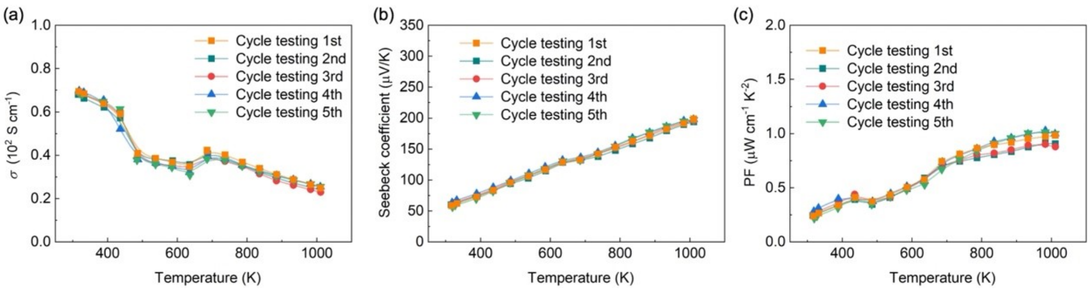

Figure S4. The stability of the $Cu_2Te_{0.5}Se_{0.5}$ samples of Quench at liquid was tested for 5 cycles for temperature-dependent (a) electrical conductivity (σ), (b) Seebeck coefficient (S), and (c) power factor (PF)

3. References

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2. G. J. Snyder, A. H. Snyder, M. Wood, R. Gurunathan, B. H. Snyder and C. Niu, *Adv. Mater.*, 2020, **32**, e2001537.
3. D. G. Cahill, S. K. Watson and R. O. Pohl, *Phys Rev B Condens Matter*, 1992, **46**, 6131-6140.