

Numerous theoretical and experimental investigations on multinary Cu-based compound semiconductors have followed the successful materialization of chalcogenide-based energy devices. This work aimed to identify the potential energy applications of the  $\text{Cu}_2\text{HgSn}(\text{Se}_x\text{S}_{1-x})_4$  compound by investigating its composition with  $x = 0, 0.25, 0.50, \text{ and } 1$ . The bandgap tunability is observed to be between 0.81 to 1.33 eV for whole composition, according to the electronic structure calculations. Its excellent ductile strength and substantial absorption coefficient of around  $10^4 \text{ cm}^{-1}$  across all compositions reveal its promise as a flexible optoelectronic device working in the visible-to-infrared spectrum. Considering realistic material parameters such as defect concentration, doping concentration, and interface effect, device modeling for single/tandem junction and graded bandgap absorber-based photovoltaics suggests that the grading bandgap absorber-based device could be the best option for maximizing efficiency by harnessing the maximum solar spectra. Our research shows that a 2  $\mu\text{m}$  thick absorber performs well when the concentration of Se increases rapidly towards the back contact side, allowing for positive bandgap grading in the absorber. According to our findings, the device's efficiency is maintained up to a bulk defect density of  $10^{15} \text{ cm}^{-3}$ . However, it is extremely susceptible to interface defects, which diminish its performance even after a concentration of  $10^{12} \text{ cm}^{-2}$ . A substantial efficiency may be achieved with a positive conduction band offset of approximately 0.3 eV, which inherently provides a large built-in field at the junction and helps the device extract the photocarrier effectively with an efficiency of over 20%.

We have also investigated the material's thermoelectric potential in detail by studying the phonon dynamics of  $\text{Cu}_2\text{HgSnS}_4$  in the intermediate temperature range, with special emphasis on the role of the heavy element Hg substitution in the parent compound  $\text{Cu}_2\text{ZnSnS}_4$ . Our overall lattice thermal conductivity is  $0.53 \text{ W m}^{-1} \text{ K}^{-1}$  ( $0.33 \text{ W m}^{-1} \text{ K}^{-1}$ ) at 300 K (700 K), when we account for both the particle and wave-like phonon treatments. The heavy metal mercury introduces a small amount of anharmonicity, and its significant vibrational amplitude is comparable to the rattling mode in compounds with a cage-like structure. As a result of extremely low thermal conduction, the highest possible ZT value for a p-type (n-type) system is 2.28 (0.77), suggesting that it might be used for thermoelectric applications at intermediate temperatures.

This study could help experimental efforts to design graded bandgap absorber-based photovoltaics as well as serve as a guide for intermediate temperature thermoelectric devices using quaternary chalcogenides or Diamond Like Semiconductors.

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