Stretchable AgX (X = Se, Te) for Efficient Thermoelectrics and Photovoltaics

ABSTRACT

Transition metal chalcogenides (TMCs) have gained worldwide interest owing to their outstanding renewable energy conversion capability. However, the poor mechanical flexibility of most existing TMCs limits their practical commercial applications. Herein, triggered by the recent and imperative synthesis of highly ductile α-Ag_2S, an effective approach based on evolutionary algorithm and ab initio total-energy calculations for determining stable, ductile phases of bulk and two-dimensional Ag_xSe_{1-x} and Ag_xTe_{1-x} compounds was implemented. The calculations correctly reproduced the global minimum bulk stoichiometric $P2_12_12_1$-Ag_8Se_4 and P21/c-Ag_8Te_4 structures. Recently reported metastable $AgTe_3$ was also revealed but it lacks dynamical stability. Further single-layered screening unveiled two new monolayer P4/nmm-Ag_4Se_2 and C2–Ag_8Te_4 phases. Orthorhombic Ag_8Se_4 crystalline has a narrow, direct band gap of 0.26 eV that increases to 2.68 eV when transforms to tetragonal Ag4Se2 monolayer. Interestingly, metallic $P2_1$/c-Ag_8Te_4 changes to semiconductor when thinned down to monolayer, exhibiting a band gap of 1.60 eV. Present findings confirm their strong stability from mechanical and thermodynamic aspects, with reasonable Vickers hardness, bone-like Young's modulus (E) and high machinability observed in bulk phases. Detailed analysis of the dielectric functions ε(ω), absorption coefficient α(ω), power conversion efficiency (PCE) and refractive index n(ω) of monolayers are reported for the first time. Fine theoretical PCE (SLME method ∼11–28%), relatively high n(0) (1.59–1.93), and sizable α(ω) (10^4–10^5 cm^{-1}) that spans the infrared to visible regions indicate their prospects in optoelectronics and photoluminescence applications. Effective strategies to improve the temperature dependent power factor (PF) and figure of merit (ZT) are illustrated, including optimizing the carrier concentration. With decreasing thickness, ZT of p-doped Ag–Se was found to rise from approximately 0.15–0.90 at 300 K, leading to a record high theoretical conversion efficiency of ∼12.0%. The results presented foreshadow their potential application in a hybrid device that combines the photovoltaic and thermoelectric technologies.