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Defect formation enthalpy of Cu2ZnSnS4 and Cu2ZnSnSe4 revisited: ab initio insights into the limitations of CZTS technology JULIEN VIDAL, EDF R&D, Chatou, France, PAWEL ZAWADZKI, National Renewable Energy Laboratory, Golden, CO, United States, VLADAN STEVANOVIC, Department of Physics, Colorado School of Mines, Golden, CO, United States, STEPHAN LANY, National Renewable Energy Laboratory, Golden, CO, United States — The defect physics of earth abundant quaternary compound Cu2ZnSnS4 (CZTS) and Cu2ZnSnSe4 (CZTSe) is extremely complex not only because of the many competing phases but also because of the many possible cationic substitutions. Previous theoretical studies have indicated that CZT(S,Se) has a quite high hole concentration originating from intrinsic defects such as Cu vacancies and Cu-on-Zn antisites. In this study, we have carried out state-of-the-art defect calculations including thermochemical corrections to the phase diagram and specific correction to the formation enthalpy of shallow defects. The latter was found to be a critical point in the analysis of intrinsic defects in both CZTS and CZTSe. Indeed, and at variance with previous studies, our GW-corrected ab initio defect calculation reveals that both Cu-on-Zn and Zn-on-Cu antisites have comparable formation enthalpy, which results in the pinning of the Fermi level in the mid-band gap region. The latter has two important consequences: the relatively low carrier concentration in CZT(S,Se) and the limitation of the open circuit voltage. It is also found that higher carrier concentrations are achievable under growth conditions where CZT(S,Se) is only marginally stable and may decompose into binary or ternary competing phases.

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