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Thermodynamic Effects of Na on the Morphology of PbTe-PbS Nanostructured Thermoelectrics JEFF DOAK, JIAQING HE, IVAN BLUM, STEVEN GIRARD, LI-DONG ZHAO, DAVID SEIDMAN, MERCOURI KANATZIDIS, VINAYAK DRAVID, CHRIS WOLVERTON, Northwestern University, HUI-QIONG WANG, JIN-CHENG ZHENG, Xiamen University, GILBERTO CASILLAS, MIGUEL JOSE-YACAMAN, University of Texas at San Antonio —
The creation of nanostructures via phase separation provides a mechanism for decreasing the lattice thermal conductivity and increasing the figure of merit of bulk thermoelectric materials like PbTe-PbS. The addition of Na to PbTe-PbS drastically alters the morphology of PbS precipitates in the system. To see if this change in morphology can be attributed to equilibrium thermodynamics, we use first-principles density functional theory (DFT) calculations to study the energetics of Na partitioning between PbTe and PbS and Na segregation at PbTe/PbS interfaces. We calculate a variety of Na defects in PbTe and PbS and find that the lowest energy defect in both PbTe and PbS is Na substituted for Pb. From the Na defect formation energies, we find the solubility limit of Na in PbTe and PbS, as well as the partitioning coefficient between PbTe and PbS. We find that Na partitions to PbS over PbTe, in agreement with experiment. We calculate Na segregation energies by substituting Na for Pb at the PbTe/PbS interface and find that Na segregates at the PbTe-side of the interface, in qualitative agreement with atom-probe tomography analysis. Applying the Gibbs adsorption isotherm to Na segregation, we find a corresponding decrease in interfacial energy leading to a change in morphology.

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