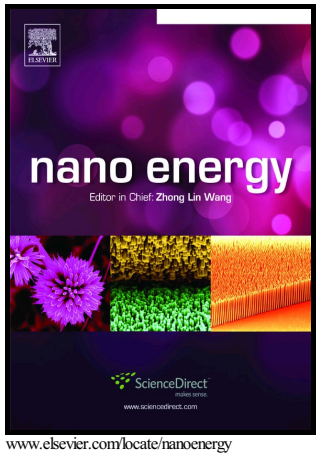

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Elemental distributions within multiphase quaternary Pb chalcogenide thermoelectric materials determined through three-dimensional atom probe tomography

Sima Aminorroaya Yamini[1]*, Tong Li[2], David R. G. Mitchell[1], Julie M. Cairney[2]*

[1] *Australian Institute for Innovative Materials (AIIM), Innovation Campus, University of Wollongong, Squire Way, North Wollongong , NSW 2500 , Australia*
[2] *Australian Centre for Microscopy and Microanalysis, The University of Sydney, Madsen Building F09, NSW 2006, Camperdown, Australia*

***Corresponding authors**: Sima Aminorroaya Yamini, Sima@uow.edu.au and Julie M. Cairney, Julie.Cairney@sydney.edu.au*

Abstract

Nanostructured multiphase p-type lead chalcogenides have shown the highest efficiencies amongst thermoelectric materials. However, their electronic transport properties have been described assuming homogenous distribution of dopants between phases. Here, we have analyzed elemental distributions in precipitates and matrices of nanostructured multiphase quaternary Pb chalcogenides doped to levels below and above the solubility limit of the matrix, using three-dimensional atom probe tomography. We demonstrate that partitioning of sodium and selenium occur between the matrix and secondary phase in both lightly- and heavily-doped compounds and that the concentrations of sodium and selenium in precipitates are higher than those in the matrices. This can contribute to the transport properties of such multiphase compounds The sodium concentration reached ~3 at.% in sulfur-rich (PbS) precipitates and no nano precipitates of Na-rich phases were observed within either phase, a result that is supported by high resolution TEM analysis, indicating that the solubility limit of sodium in PbS is much higher than previously thought. However, non-equilibrium segregation of sodium is identified at the precipitates/matrix interfaces. These findings can lead to further advances in designing and characterizing multiphase thermoelectric materials.

Keywords: Thermoelectric materials, sodium, dopant, atom probe tomography, segregation, and solubility

1. Introduction

Bulk lead-chalcogenides provide the highest thermoelectric efficiencies amongst mid-range temperature (600-900 K) thermoelectric materials [1-5]. Intrinsic semiconducting lead chalcogenides can be tuned to either *n*-type or *p*-type to charge carrier concentrations of 10^{18} to 10^{19} cm^{-3} due to excess lead or chalcogen atoms respectively, where point defects act as donor or