

Mechanical and Thermoelectric Properties of $\text{PbSe}_{1-x}\text{Te}_x$ Solid Solutions

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Abstract—The microhardness, Seebeck coefficient and electrical conductivity of $\text{PbSe}_{1-x}\text{Te}_x$ ($x = 0 - 0.045$) solid solutions were investigated at room temperature. In the range of small concentrations of PbTe (in the vicinity of $x = 0.01$ and $x = 0.02$) an anomalous decrease in microhardness, increase in Seebeck coefficient and electrical conductivity were registered. The concentration anomalies were interpreted as the transition from diluted to concentrated solid solutions when the interaction between impurity atoms starts to make a significant contribution to the change of the properties of materials.

Index Terms— $\text{PbSe}_{1-x}\text{Te}_x$ solid solution, impurity, hot pressing, microhardness, Seebeck coefficient, electrical conductivity, percolation threshold

I. INTRODUCTION

PbSe-PbTe solid solutions are promising thermoelectric materials (TE) used in thermogenerators working in medium-temperature region (600 – 900 K) [1]. Pressing of materials is widely used in the manufacture of TE devices to increase strength and homogeneity degree of samples as compared to cast ones. In [2-4] we reported on the anomalous character of properties' dependences (microhardness, Seebeck coefficient, Hall coefficient, electrical conductivity, Hall mobility of charge carriers, thermal conductivity) of $\text{PbTe}_{1-x}\text{Se}_x$ solid solutions in the range of $x = 0.05$ for both cast and pressed samples. The observed effects stimulate more detailed investigations of the concentration dependences of TE properties, namely, under small amounts of the second component in the solid solutions based on IV-VI semiconductor compounds.

The purpose of this work is a study of the composition dependences of the mechanical and TE properties in the polycrystalline solid solutions $\text{PbSe}_{1-x}\text{Te}_x$ ($x = 0 - 0.045$) at room temperature.

II. EXPERIMENTAL

Pressed samples of $\text{PbSe}_{1-x}\text{Te}_x$ ($x = 0 - 0.045$) solid solutions were prepared at temperature of 670 K and pressure of 400 MPa with subsequent annealing at temperature of 720 K for 260 hours.

Microhardness was measured on PMT-3 microhardness tester, using a diamond pyramid, under constant load on the indenter $P = 0.49$ N. Measuring H of each sample was performed at least at 30 points of a sample with subsequent statistical processing of results. Relative mean-square fluctuation for all samples did not exceed 3 %.

The Seebeck coefficient S was measured by compensation method in relation to copper electrodes at no fewer than 20 sample points with subsequent statistical processing of measured data. The root mean square relative error for all samples did not exceed 3 %. All the samples were of p-type conductivity.

The electrical conductivity σ measurements were carried out by a four-probe method. Relative mean-square fluctuation for all samples did not exceed 7 %.

III. RESULTS

Fig. 1a shows the dependence of H on the composition in the $\text{PbSe}_{1-x}\text{Te}_x$ solid solutions. As it is seen that dependence of H has nonmonotonous character. Introduction of the first portions of PbTe (up to $x = 0.005$) leads to the crystal strengthening. The effect of solid-solution hardening is well known and related to blocking of dislocations by impurity atoms [5]. However, with further increase in PbTe concentration, two ranges of anomalous drop in H ($x = 0.0075 \div 0.01$ and $x = 0.0175 \div 0.025$) are retained. At $x \geq 0.025$ microhardness remains practically constant.

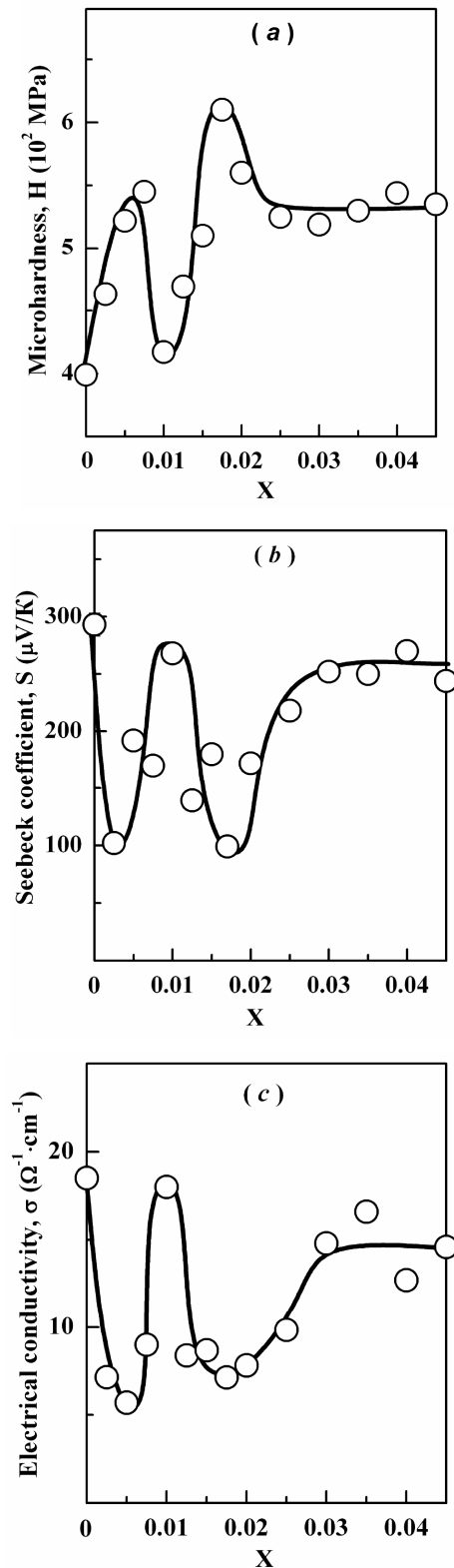


Figure 1. The microhardness H (a), Seebeck coefficient S (b) and electrical conductivity σ (c) versus the composition of $\text{PbSe}_{1-x}\text{Te}_x$ ($x = 0 - 0.045$) solid solutions for pressed samples at room temperature.

Complicated composition dependencies are observed for Seebeck coefficient (Fig. 1b) and electrical conductivity (Fig. 1c): there are anomalies on the property-composition curves for the above-mentioned concentration intervals. For isoivalent isostructural solid solutions PbSe-PbTe , under the condition of ideal anionic substitution scheme $\text{Se} \rightarrow \text{Te}$, the concentration of charge carriers (and, hence, the Seebeck coefficient) shouldn't change significantly. Meanwhile, the extreme character of the $S(x)$ dependence is clearly visible. Decrease in the electrical conductivity with a growth in PbTe concentration is connected with the increased imperfection of crystal lattice and the emergence of additional centers of charge carriers' scattering. However, the growth in electrical conductivity is unusual.

From our point of view [6] the appearance of concentration anomalies of properties in the range of small impurity content represents the universal physical phenomenon associated with the formation of percolation channels in impurity subsystem of crystal upon attaining percolation threshold [7].

The existence of anomalies of properties in the range of small impurity concentration should be taken into account while developing and using TE materials.

ACKNOWLEDGMENTS

The authors thank Anna Nikolaenko for preparing of pressed samples.

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