THE PECULIARITIES OF ELECTRICAL CONDUCTIVITY IN $AgSbSe_2$ AND $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$

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Temperature dependence of electrical conductivity in $AgSbSe_2$ and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ solid solution was investigated in the temperature range 80-350K. It is determined that the electrical conductivity of $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ solid solution to have a hopping mechanism in the temperature range 127-233K. In both compositions, the activation energy of the charge carriers was calculated in different temperature ranges.

Keywords: hopping conductivity, activation energy, $AgSbSe_2$
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INTRODUCTION

Ternary chalcogenide $AgSbSe_2$ is great interest as a good thermoelectric material due to its low thermal conductivity. $AgSbSe_2$ crystallizes in a cubic rock salt structure with a space group of Fm3m. In this structure the Ag and Sb atoms randomly occupy the sites of their sublattice, forming a disordered substructure [1]. The investigations of $AgSbSe_2$ was carried out basically above room temperature for the purpose to improve thermoelectric properties by adding various elements [2, 3, 4]. In addition, the determination of the mechanism of transport of electric charge in $AgSbSe_2$ and its solid solutions are very interesting.

As is known, both $AgSbSe_2$ and PbTe are good thermoelectric materials that used at medium temperatures (400-800K) [5]. Both of compositions have the same crystal structure, which allows obtaining a number of solid solutions based on them [6, 7].

In this work, $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ solid solution was investigated for the purpose to study the influence of PbTe addition on the mechanism of transport of charge carriers in $AgSbSe_2$ and to determine the activation energy of charge carriers.

EXPERIMENTAL RESULTS AND DISCUSSION

The investigated samples were synthesized by direct fusion of high purity elements (99,99% purity) taken in stoichiometric ratios in quartz tube. The tubes were flame sealed under vacuum (10^{-4} Torr) and slowly heated in the furnace up to 1000 K over 12 h. The furnace temperature was gradually increased at a rate of 1 K/min. Then tubes held for 10 h in this temperature and were gradually cooled to room temperature at the same rate.

The X-ray diffraction analysis for the obtained samples were performed on a Bruker D8 Advance diffractometer. The lattice constant was determined from the analysis of XRD peak positions with the EVA and TOPAS programs. The results of X-ray analysis (shown in Figure 1) revealed that both of samples were single phase with the cubic NaCl-type structure (Fm3m).

The lattice constans of $AgSbSe_2$ and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ are a=5,762Å, a=5,845Å, respectavely. Ionic radius of Pb (Te) is larger than of Sb. Therefore, as bigger Pb (Te) is introduced in the place of smaller Sb, the unit cell undergoes a systematic expansion, leading to increase in the lattice parameter [8].

The electrical conductivity of $AgSbSe_2$ and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ was measured by four-point probe method on direct current.

Figure 2 presents the temperature dependence of electrical conductivity of $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ solid solution in comparison with $AgSbSe_2$. The experimental results show that, at temperatures below T=300K, the electrical conductivity of the samples is relatively small and does not change much. The value of electrical conductivity of both samples with the temperature rise begins to increase.

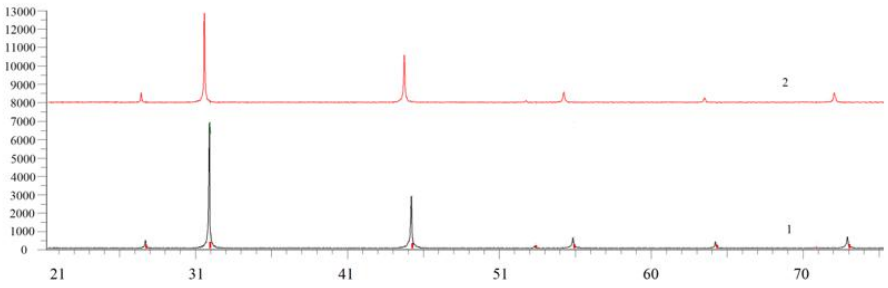

Fig. 1. X-ray diffraction patterns of $AgSbSe_2$ (1) and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ (2).

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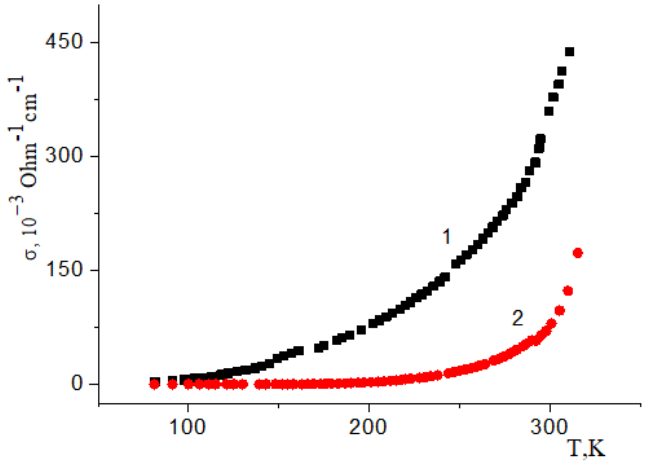

Fig. 2. Temperature dependences of electrical conductivity of $AgSbSe_2$ (1) and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ (2).

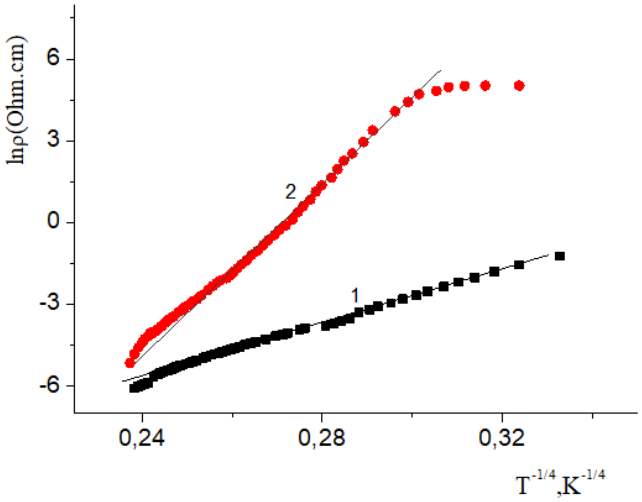

Fig. 3. Temperature dependences of specific resistance of $AgSbSe_2$ (1) and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ (2) in Mott coordinates.

The temperature dependences of the resistivity of $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ solid solution in Mott coordinates in comparison with $AgSbSe_2$ in the 80-350K temperature range are presented in Figure 3.

As seen in figure, the experimental points at certain temperatures correspond to the linear dependence in Mott coordinates. This fact to affirm that both $AgSbSe_2$ and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ solid solution the charge transfer occurs by the hopping conductivity of charge carriers over localized states lying in a narrow energy region near the Fermi level. As is known, in this case electrical conductivity is described by Mott's relation [9]:

$$\rho=\rho_0\exp(T_0/T)^{1/4}, \quad T_0=\beta/k_B g(\mu)a^3 \qquad (1)$$

where k_B is Boltzmann's constant, $g(\mu)$ is density of localized states near the Fermi level, a is radius of localized states near the Fermi level, β is number depending on dimension of the problem (for the three dimensial case $\beta=21$).

As can be seen in Figure 3, the Mott dependence (1) for the $AgSbSe_2$ and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ solid solution are satisfied in the temperature ranges 105K<T<230K and 127K<T<233K, respectively. The fact that the electrical conductivity of $AgSbSe_2$ has a hopping mechanism is reported in detail in [10].

As a result of calculations based on relation (1), for the density of localized states near the Fermi level in $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ the value of $g(\mu)=3{,}7\cdot10^{14}eV^{-1}cm^{-3}$ was obtained.

Thus, it was determined that the electrical conductivity of $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ solid solution, as in $AgSbSe_2$, has a hopping mechanism in the temperature range 127-233K. At higher temperatures only the band conductivity prevails.

One of the main problems in explaining the mechanism of conductivity is the calculation of the activation energy of charge carriers. For this purpose, in Figure 4 shows the dependence of the logarithm of the specific conductivity of $AgSbSe_2$ and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ versus 1/T. The values of the activation energy of charge carriers of $AgSbSe_2$ and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ solid solution were calculated based on the values of linear region at the coordinates $\ln\sigma\sim(1/T)$ in the temperature range 200-550K.

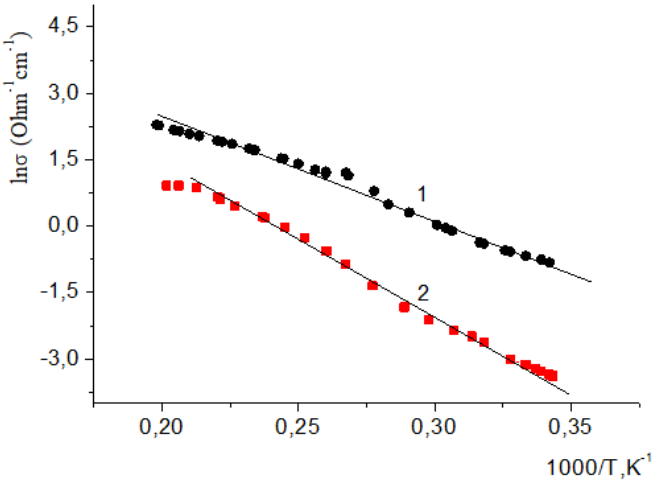

Fig. 4. Dependence of logarithm of electrical conductivity of $AgSbSe_2$ (1) and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ versus 1000/T.

As a result of the calculation, for the activation energy of charge carriers of $AgSbSe_2$ and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ in different temperature ranges the values of ΔE=78 meV (200-300K), ΔE=211 meV (300-400K), ΔE=130 meV (400-500K) and ΔE=173meV (200-300K), ΔE=312 meV (300-400K), ΔE=158 meV (400-500K) were obtained, respectively. It is seen from the obtained results, the activation energy of the charge carriers in both composition takes different values in different temperature range. The different values of activation energy of charge carriers is caused by the existence of different impurity levels in the band gap. Also, the value of activation energy of charge carriers of $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ increases compared to the value obtained for $AgSbSe_2$. The formation of impurity conductivity as a result of doped with PbTe increases the value of activation energy.

CONCLUSION

The electrical conductivity of $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ solid solution has hopping mechanism in the temperature range 127-233K. The activation energy of $AgSbSe_2$ and $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ are incresing with temperature rise. As a result of doped with PbTe the formation of impurity conductivity increases the value of the activation energy of charge carriers of $(AgSbSe_2)_{0,85}(PbTe)_{0,15}$ compared to $AgSbSe_2$.

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[1] *S. Geller and J.H. Wernick*. Ternary semiconducting compounds with sodium chloride-like structure: $AgSbSe_2$, $AgSbTe_2$, $AgBiS_2$ and $AgBiTe_2$. Acta Crystallographica, 1959, 12 (1), p. 46-54

[2] *S. Cai, Z. Liu, J. Sun et al*. Enhancement of thermoelectric properties by Na doping in Te-free p-type $AgSbSe_2$. Dalton Transactions, 2015. 44 (3), p. 1046-1051

[3] *Z. Liu, J. Shuai, H. Geng et al*. Contrasting the role of Mg and Ba doping on the microstructure and thermoelectric properties of p-type $AgSbSe_2$. ACS Applied Materials and Interfaces, 2015, 7 (41), p. 23047-23055

[4] *D. Li, X.Y. Qin, T.H. Zou et al*. High thermoelectric properties for Sn-doped $AgSbSe_2$, Journal of Alloys and Compounds, 2015, 635, p. 87-91

[5] *J.R. Sootsman, H. Kong, C. Uher et al*. Large enhancements in the thermoelectric power factor of bulk PbTe at high temperature by synergistic nanostructuring. Angew. Chem. Int. Ed., 2008, 47, p.8618-8622

[6] *Y. Xiao, L.-D. Zhao*. Charge and phonon transport in PbTe-based thermoelectric materials. npj Quantum Materials, 2018, 3(55), p. 1-12

[7] *H.Z. Wang, Q.Y. Zhang, B. Yu et al*. Transmission electron microscopy study of Pb-depleted disks in PbTe-based alloys. Journal of Materials Research, 2011, 26(7), p. 912-916

[8] *S.N. Guin, C. Arindom, S.N. Devendra et.al*. High thermoelectric performance in tellurium free p-type $AgSbSe_2$. Energy Environ. Sci., 2013, 6(9), 2603-2608

[9] *N.F. Mott, E.A. Davis*. Electronic processes in non-crystalline materials. New York: Oxford University Press, 1979, 590 p.

[10] *S.S. Ragimov, A.A. Saddinova, A.I. Aliyeva*. Mechanism of electrical conductivity and thermal conductivity in $AgSbSe_2$. Russian Physics Journal, 2019, 62 (6), p.1077-1081.

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