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THERMOPHYSICAL PROPERTIES OF ALLOY COMPOSITIONS **$(2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3)_{100-x} (2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_x$ ($x=0, 10, 50$)****S.I.Bananyarli, Sh.S.Ismayilov, R.N.Gasimova, L.A.Khalilova***M.Nagiyev Institute of Catalysis and Inorganic Chemistry, NAS of Azerbaijan**ishr_az.@yahoo.com*

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The thermophysical properties, namely, the temperature dependence of thermal conductivity, thermal resistance and heat capacity of the alloys compositions $(2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3)_{100-x} (2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_x$ in the (300–600) K temperature range have been investigated. An increase in thermal conductivity $\chi(T)$ above 500 K is probably associated with the softening of alloys and the presence of blurred phase transitions, which are accompanied by partial breaking of chemical bonds. It was revealed that the heat capacity in alloys of the compositions $(2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3)_{100-x} (2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_x$ increases with an increase in the GeO_2 concentration. In the studied samples, that showed their own disorder during solidification, the thermal conductivity is strongly reduced due to the enhancement of the anharmonicity of phonon – phonon interactions. In turn a small "disorder" introduced by defects due to the difference in masses is not noticeable against the background of the huge "disorder" inherent in oxide substances.

Keywords: thermal conductivity, thermal resistance, heat capacity, phonons, germanium oxide, alloys.

doi

Introduction

Amorphous and glassy materials based on bismuth borates are being intensively studied [1–6]. This is due to nonlinear optical properties were found in BiB_3O_6 [3, 7]. In addition, glasses based on Bi_2O_3 have high refractive indices, a wide transparency region in the visible and IR ranges, and are promising materials for practical application [8, 9]. It was found [7] that bismuth borates can be used as a frequency converter of laser radiation based on stimulated Raman scattering. Authors of [9–11] shown that the equilibrium system Bi_2O_3 – B_2O_3 is characterized by the existence of five compounds. There are a large number of works devoted to the preparation and study of the properties of these and other compounds based on Bi_2O_3 [12]. In [13], the heat capacity, thermal conductivity, and thermal expansion of the $\text{Bi}_{12}\text{SiO}_{20}$ single crystals were studied in a wide temperature range, while the heat capacity of the GeO_2 – PbO oxide systems was considered by authors of [14]. The heat capacity of glasses of bismuth borates containing 50, 51.5, 60, 62.5 and 65 mol% B_2O_3 in a wide temperature range is measured and thermodynamic properties of the $\text{Bi}_3\text{B}_6\text{O}_{12}$ и BiBO_3 compounds are determined [15].

Previously, we studied the electrical conductivity (σ), dielectric permeability (ϵ) and thermal conductivity (χ) of alloys of the $2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3$ – $2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2$ system [16–18]. It was found that in the system, depending on the ratio of the components, alloys with different electro-physical properties were obtained, and in some samples are detected semiconducting properties.

However, despite a large number of works devoted to the study of physical and electrophysical properties, works on the study of their thermophysical properties, namely, thermal conductivity and thermal resistance, is insufficient.

The present work aims to study the thermal conductivity, thermal resistance, and heat capacity of alloys of the $2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3$ – $2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2$ system in the 300–650 K temperature range.

Experimental part

Alloys of the compositions $(2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3)_{100-x} (2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_x$ ($x=0, 10, 50$) were synthesized from high-purity oxides GeO_2 (CAS No 1310-53-8, purchased from Alfa Aesar) and Bi_2O_3 (CAS No 1304-76-3) and boric acid H_3BO_3 (CAS No 10043-35-3) at 1273–1373 K.

The melts were poured into a titanium plate at room temperature. All samples were glassy from yellow to brown color. After synthesis, the glasses were annealed at 646 K for 200 hours.

The DTA of the alloys of the system was studied on a thermal analyzer brand NETZSCH STA 449F3 "Yupiter" [Range 20/10 (K/min)/650]. Bruker D2 PHASER diffractometer was used for XRD analysis. The density of the alloys was determined by the hydrostatic weighing method, distilled water was used as the working liquid [16, 17].

The measurements of the thermal conductivity and thermal resistance ($1/\chi \approx \omega$) were carried out on rectangular samples (thickness $d = 2\text{--}4$ mm; width $b = 7\text{--}8$ mm and length $l = 14$ mm) by a stationary method in the of 573–723 K temperature range.

Results and discussions

The heat capacity of bismuth borate was investigated by the authors of [6, 15–17, 19, 20]. Figure 1 shows the dependence of the standard heat capacity on the borate composition according to the literature data.

Figure 1 shows that the C_p^0 values are significantly higher than the standard heat capacity of bismuth borates. With the introduction of germanium oxide, the heat capacity increases and this agrees with the effect of atomic masses. The phonon frequencies should be higher for oxides with a high GeO_2 content, that leads to an increase in the Debye temperature and, accordingly, an increase in the heat capacity of oxides with a higher concentration of light atoms ($M_{\text{Ge}} \approx 72.59$ $M_{\text{Bi}} \approx 208.98$) [21]. The value of the heat capacity of the initial component $2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3$ obtained by us approximately coincides with the data of the authors [21]. Figure 1 also shows the results for samples №1 [10 mol% ($2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2$)] and №3 [50 mol% ($2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2$)] [16, 17]. As can be seen, the heat capacity of samples №1 and №3 is higher than the standard heat capacity of bismuth borates, that is apparently associated with an increase in the GeO_2 concentration in the alloys.

Figure 2 shows the temperature dependence of thermal conductivity of samples №1 ($2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3$)₉₀($2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2$)₁₀ (curve 1) and №3 ($2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3$)₅₀($2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2$)₅₀ (curve 3) and heat capacity of the alloys №1 and №3 (curves 4 and №5) of the compositions ($2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3$)_{100-x} $T(2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_x$. The electrophysical properties of above-mentioned samples were previously studied by us [16]. As can be seen, the total thermal conductivity decreases with increasing temperature, passing through a minimum in the 490–540 K temperature intervals. The change in the value of the thermal conductivity $\chi(T)$, depending on the GeO_2 concentration, occurs different. For example, in sample №1 (10 mol% $2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2$) in the 300–380 K temperature range the thermal conductivity $\chi(T)$ decreases slightly. Starting from 400 K, a relatively intensive decrease of $\chi(T)$ from 9.8 to $4.5 \cdot 10^{-3}$ $\text{W}/\text{sm}^{-1} \cdot \text{K}^{-1}$ is observed. With a further increase in temperature, a monotonic increase in thermal conductivity occurs (curve 1).

In sample №3 (50 mol% $2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2$), the value of the thermal conductivity $\chi(T)$ decreases relatively weakly with increasing temperature to 500 K and passing through a minimum ($\chi = 4.6 \cdot 10^{-3}$ $\text{W}/\text{sm}^{-1} \cdot \text{K}^{-1}$) at 520–540 K increases again (Figure 2, curve 3). For comparison, Figure 2, also present the thermal conductivity of the initial component $2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3$. As can be seen, the value of $\chi_0(T)$ changes very little with temperature in comparison with the results for samples №1 and 3 in the 300–500 K temperature range (curve 0). This is apparently due to the occurrence of anharmonicity of phonon processes in the samples due to the difference in the ionic sizes of boron and bismuth ($\text{B}_2^{+3} = 41\text{pm}$ and $\text{Bi}_2^{+3} = 117\text{pm}$). An increase in thermal conductivity $\chi(T)$ with a temperature above $T > 500$ K is associated with softening of alloys and the presence of blurred phase transitions, which are accompanied by partial breaking of chemical bonds.

Figure 2 also shows the results of the heat capacity measurements of samples № 1 and № 3 (Figure 2, curves 4 and 5).

The obtained data are higher than the literature data (Figure 1) [22, 23].

Fig. 1. Dependence of the standard heat capacity of bismuth borates on the composition: ● – [6], ○ – [15], □ – [19], ▲ – [20].

Fig. 2. Temperature dependences of thermal conductivity (1, 3) and heat capacity (4, 5) for the alloys of the compositions $(2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3)_{100-x} (2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_x$. 0 – $x=0$; $2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3$, 1 and 4 – $x=10$; $(2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3)_{90} (2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_{10}$, 3 and 5 – $x=50$; $(2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3)_{50} (2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_{50}$.

The measurements showed that with an increase in the GeO_2 in the samples (№1 – $C_p = 0.4$, №3 – $C_p = 0.7 \text{ J/g} \cdot \text{K}^{-1}$), the C_p values increase which probably leads to an increase of the Debye temperature. It is known that the thermal resistance and thermal conductivity of alloys (or solids) are very sensitive to a defect structure. In glassy substances, thermal conductivity varies from point to point.

The temperature dependence of the thermal resistance of alloys of the compositions $(2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3)_{100-x} (2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_x$ was also studied. Figure 3 presents the temperature dependence of the thermal resistance of the initial compound $2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3$ and samples № 1 $x=10$ mol% $2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2$ and № 3 $x=50$ mol% $2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2$. According to Figure 3, when the samples are heated to 500 K, the thermal resistance increases nonlinearly. With a further temperature increase, the thermal conductivity of the initial component is almost constant, and for sam-

ples № 1 and № 3 it decreases. This is because, apparently, besides phonon–phonon interaction, heat fluxes are scattered over large-scale point defects, i.e., over cluster centers [25]. With an increase in temperature ($T > 500 \text{ K}$), softening intensifies, smearing of phase transitions and partial desorption of cluster centers occurs. This is accompanied by the breaking of chemical bonds. As a result, a relative increase in thermal conductivity and a decrease in the thermal resistance of the samples are observed.

As is known, there is no long-range order in a glassy substance, including the alloys of the system $2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3 - 2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2$ studied by us. The studied samples are disordered substances in which some other mechanisms different from the processes occurring in crystals play an essential role in heat transfer. In glassy or non-crystalline substances, including the studied alloys (glasses), phonon processes (heat fluxes) change from point to point [24].

Fig. 3. Temperature dependence of thermal resistance (ω) of the samples of the compositions $(2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3)_{100-x} (2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_x$. 0 – $2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3$; 1 – $x=10$ mol%; $(2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3)_{90} (2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_{10}$; 3 – $x=50$ mol%; $(2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3)_{50} (2\text{Bi}_2\text{O}_3 \cdot 3\text{GeO}_2)_{50}$.

In the investigated alloys, heat propagation, i.e. the velocity (v_{ph}) of phonons is related to the size of structural units, and varies from point to point in different ways [26], the thermal resistance of elastic heat waves also changes.

For glassy and amorphous substances, thermal conductivity is determined by the relation [26]

$$\chi = \frac{1}{3} L \cdot C_v \cdot v_{\text{ph}}$$

where, L – is force run length (on phonon perturbations), i.e. free displacement of phonons, v_{ph} – is the speed of phonons, C_v – is the heat capacity of alloys. Taking into account the values of thermal conductivity (χ), heat capacity (C_v), and $v_{\text{ph}} \approx$ of the phonon velocity, L (for sample № 1 10 mol% $2\text{Bi}_2\text{O}_3 \cdot \text{B}_2\text{O}_3$) at 320 K is determined, $L \approx 10.2$ Å, that is comparable with interatomic distances.

The scattering of phonons by point defects occurred mainly due to the difference in the masses of the components and impurity atoms, as well as changes in the interatomic interaction forces near the defects.

Conclusions

1. A small "disorder" introduced by defects due to the difference in masses is not noticeable against the background of the huge "disorder" inherent in oxide substances.

2. In the studied samples, which showed their disorder during solidification, the thermal conductivity is strongly reduced due to the enhancement of the anharmonicity of phonon – phonon interactions.

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ТЕПЛОФИЗИЧЕСКИЕ СВОЙСТВА СПЛАВОВ СОСТАВОВ ($2\text{Bi}_2\text{O}_3\cdot\text{B}_2\text{O}_3$) $_{100-x}$ ($2\text{Bi}_2\text{O}_3\cdot 3\text{GeO}_2$) $_x$ ($x=0, 10, 50$)

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Исследованы физико-химические свойства: температурная зависимость теплопроводности, теплового сопротивления и теплоемкости сплавов составов $(2\text{Bi}_2\text{O}_3\cdot\text{B}_2\text{O}_3)_{100-x}(2\text{Bi}_2\text{O}_3\cdot 3\text{GeO}_2)_x$ в интервале температур 300 ÷ 650 K. Увеличение теплопроводности $\chi(T)$ выше 500 K связано с размягчением сплавов и наличием размытых фазовых переходов, которые сопровождаются частичным обрывом химических связей. Выявлено, что теплоемкость в сплавах составов $(2\text{Bi}_2\text{O}_3\cdot\text{B}_2\text{O}_3)_{100-x}(2\text{Bi}_2\text{O}_3\cdot 3\text{GeO}_2)_x$ возрастает по мере роста концентрации GeO_2 . В исследованных образцах, проявивших при затвердевании собственный беспорядок, сильно снижает теплопроводность за счет усиления ангармонизма фонон-фононных взаимодействий. В свою очередь небольшой "беспорядок", вносимый дефектами, возникающими из-за разности масс, не заметен на фоне огромного собственного "беспорядка", присущего оксидным веществам.

Ключевые слова: теплопроводность, теплосоппротивление, теплоемкость, фононы, оксид германия, сплав.

($2\text{Bi}_2\text{O}_3\cdot\text{B}_2\text{O}_3$) $_{100-x}$ ($2\text{Bi}_2\text{O}_3\cdot 3\text{GeO}_2$) $_x$ TƏRKİBLİ ƏRİNTİLƏRİN İSTİLİK FİZİKİ XASSƏLƏRİ ($x=0; 10; 50$)

S.İ.Bənənyarlı, Ş.S.İsmayilov, R.N.Qasimova, L.Ə.Xəlilova

Bu məqalədə $(2\text{Bi}_2\text{O}_3\cdot\text{B}_2\text{O}_3)_{100-x}(2\text{Bi}_2\text{O}_3\cdot 3\text{GeO}_2)_x$ tərkibli ərintilərinin fiziki-kimyəvi xassələri: istilikkeçiriciliyin, istilik müqavimətinin, istilik tutumunun 300÷650K intervalında temperatur asılılığı öyrənilmişdir. 500K temperaturdan yuxarıda istilikkeçiriciliyin $\chi(T)$ artması ərintilərin yumşalması və yaygın faza keçidlərinin mövcudluğu ilə bağlıdır, hansı ki, kimyəvi rabitələrin qismən qırılması ilə müşayiət olunur. Müəyyən edilmişdir ki, $(2\text{Bi}_2\text{O}_3\cdot\text{B}_2\text{O}_3)_{100-x}(2\text{Bi}_2\text{O}_3\cdot 3\text{GeO}_2)_x$ tərkibli ərintilərin istilik tutumu GeO_2 -nin qatılığının artması ilə mütənasib artır. Tədqiq olunan nümunələrdə, bərkimə zamanı ortaya çıxan özünəməxsus nizamsızlıq, öyrənilən tərkiblərdə fonon-fonon qarşılıqlı təsirinin anqarmonizminin artması hesabına istilikkeçiriciliyini güclü azaldır. Öz növbəsində defektlərin yaratdığı cüzi "nizamsızlıq" oksid maddələrə xas olan kütlə fərqləri nəticəsində əmələ gələn böyük "nizamsızlığın" fonunda nəzərə çarpır.

Açar sözlər: istilikkeçirmə, istilik müqaviməti, istilik tutumu, fononlar, germanium oksid, ərinti.