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Temperature dependences of density and ultrasound velocity of the eutectic Bi-44.6 wt.% Pb melt

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Measurements of temperature dependences of density d and ultrasound velocity V_s of the eutectic Bi-44.6 wt.% Pb alloy were performed from eutectic temperature up to 550–900°C. Density was studied using gamma absorption method. Ultrasound velocity was determined by pulse phase method. The dependences $V_s(T)$ and $d(T)$ are linear in temperature interval of measurements and can be approximated by the equations:

$$V_s(T) = 1788 - 0.269\ (T - 398)\ \mathrm{m \cdot s^{-1}}.$$
$$d(T) = 10271 - 1.29\ (T - 398)\ \mathrm{kg \cdot m^{-3}}.$$

These results are found to be in good agreement with data of other works.

1. Introduction

The liquid Bi-Pb alloy of the eutectic composition (44.6 wt.% Pb) is widely used as a heat-carrier for nuclear reactors due to its low chemical activity with air, water and vapor, high boiling temperature excluding its boiling in hottest parts of the installation and permitting to keep low pressure inside and some more factors. The information about its physical properties is necessary for designing reactors and improvement of modes of their functioning.

However, the structural rearrangements of this alloy in liquid state is a very important and unsolved problem. For example, using internal friction and DSC measurements, Zu *et al*. discovered distinct signs of structural transformation at 598°C [1]. They explained these effects by dissolution of crystalline fragments existing in the melt at not too high overheating above eutectic point.

These explanations are in a good agreement with the hypothesis of metastable microheterogeneous states of liquid eutectic alloys proposed by P.Popel *et al*. in 1982 [2] and confirmed by the small angle neutron scattering (SANS) experiments [3]. According to our concept, a microheterogeneous melt of colloidal type is formed just after melting of a heterogeneous eutectic ingot. After rather long relaxation process (several hours at a temperature slightly above liquidus, according to our data [2]), a metastable equilibrium between disperse particles enriched with one of components and surrounding melt enriched with another one, establishes. This state can exist tens of hours at a temperature slightly above liquidus or can be destroyed irreversibly as a result of overheating of the melt up to a much higher temperature specific of each composition. By measuring property temperature dependence of a melt in heating and subsequent cooling mode, one can determine the temperature of its irreversible transition to a true solution corresponding to branching points of heating and cooling curves. Various anomalies in property temperature dependence obtained while heating precede usually this transformation.

Therefore, measurements of ultrasound velocity V_s and density d of the eutectic Bi-44.6 wt.% Pb melt have been performed in order to check existence of structural transformation related with the melt homogenization.

2. Experimental

The most original part of this work is measurement of ultrasound velocity in an interval of 400 to 1100°C using pulse phase method. The block-scheme of the device is shown in Fig. 1. The high frequency pulses ($f = 30.3$ MHz) pass through two identical acoustic cells containing separately investigated melt and a standard liquid (distilled water in present case). Both the transmitted signals interfere after amplification and the result of their interference is observed on the screen of the oscilloscope. The upper wave-guide in the measuring cell can be moved along vertical axis and a micrometer fixes its displacement. During the movement, a sequence of extremes of the interference signal can be observed. The distance between two nearest maximums or minimums is the ultrasound wavelength.

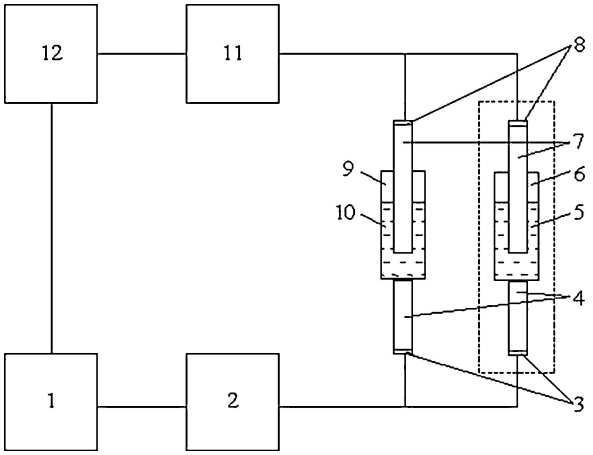

Figure 1 The block-scheme of the device for acoustic measurements: 1—high frequency generator; 2—pulse generator, 3—active piezo-ceramics, 4—down wave-guides, 5—investigated melt, 6—measuring cell, 7—upper wave-guides, 8—passive piezo ceramics, 9 -standard cell, 10—distilled water, 11—amplifier, 12—oscilloscope.

Precisely, the upper wave-guide displacement, Δh, corresponding to appearance of n subsequent maximums or minimums has been measured. The measurements has been followed by the calculation of the wavelength as $\lambda = \Delta h/n$ and, finally, the ultrasound velocity $V_s = (\Delta h/n)f$ in a very thin (not more then 0.5 mm) liquid layer with the thickness Δh near the lower end of the upper wave- guide. The accuracy of V_s values is estimated to be $\pm 0.2\%$.

The ability to measure practically local values of V_s at given distances, h, from the bottom of the crucible is the most attractive feature of the technique. By moving the upper wave-guide through the melt from the lower to the upper surface of the sample, one can fix V_s vs. h dependence and investigate the sedimentation of disperse particles in microheterogeneous melt.

Density d was measured using gamma absorption method. The scheme of the gamma densitometer has been outlined in our previous work [4]. The gamma beam (3 mm in diameter) penetrated horizontally through the beryllium oxide crucible having inner diameter of 22 mm filled with investigated melt, in horizontal direction. The intensity of beam was measured by gamma counter with NaJ(Tl) detector. In the course of the experiment stepwise heating and subsequent cooling of the sample and measuring of the intensity of gamma beam penetrating through the melt I continuously with exposition time of 400 s have been performed. As temperature stabilizes, the values of Ihave been measured for 1200 s. Later, the experiment with an empty crucible of the same set in the same time-temperature mode has been performed in order to get the initial intensity I_0. Corrections related with background count, dead time of the counter, curvature of the sample, inhomogeneity of the gamma beam and effects of superposition of pulses have been taken into account for the calculations of absolute density values. The counting rate (more than 600 pulses per second) permitted to fix more then 250,000 pulses at each exposition in the course of temperature changes and more then 750,000 pulses at isothermal expositions. It corresponds to accidental error of the order of 0.4% while temperature changes and 0.2% in isothermal conditions. The accuracy of absolute density values determination is estimated to be less then 0.5%.

3. Results

The results of measurement of ultrasound velocity V_s, and density d temperature dependences are shown in Figs 2 and 3.

The temperature dependence of ultrasound velocity is linear and can be approximated by the equation:

$$V_s = 1788 - 0.269(T - 398)\ \mathrm{m \cdot s^{-1}}. \qquad (1)$$

Unfortunately, we have not found any reference data on V_s that could be compared with the results of present investigation.

No dependence of ultrasound velocity on vertical coordinate were fixed both in the course of this

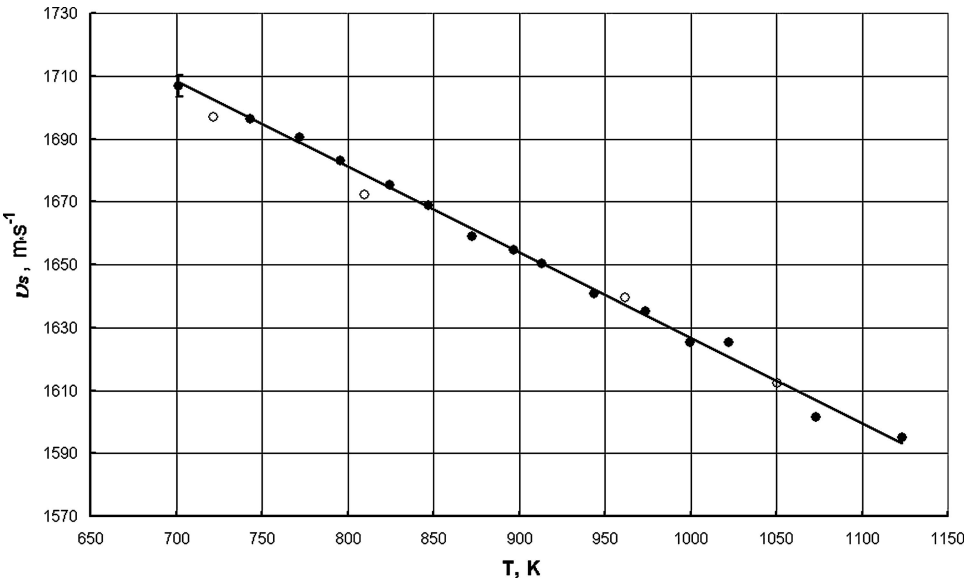

Figure 2 Temperature dependences of ultrasound velocity V_s while heating (●) and subsequent cooling (○) the sample.

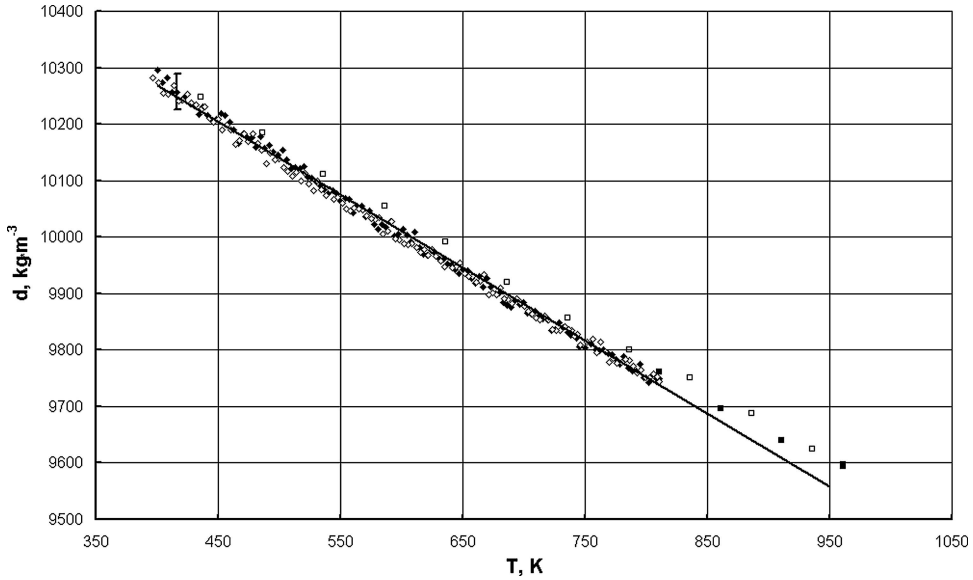

Figure 3 Temperature dependence of density d of the Bi-44.6 wt.% Pb alloy. (■) and (□)–heating and cooling results at isothermal expositions, (⧫) and (◇)—the same while continuous temperature changes.

measurements and in a special experiment where the melt has been kept at a temperature slightly above liquidus for several hours in order to discover the signs for the sedimentation of disperse particles enriched with one of the components. Obviously, at very close density values of bismuth and lead, this process is pronounced in the melt weakly. As in the present investigation the densities obtained in the case of variable temperature and at isothermal expositions are of different statistical error, we fit them in Fig. 3 separately. It was discovered that both the dependences are linear and can be approximated by Equations 2 and 3, correspondingly:

$$d(T) = 10270 - 1.30\,(T - 398); \qquad (2)$$

$$d(T) = 10287 - 1.24\,(T - 398); \qquad (3)$$

Combination of these dependences by taking the difference of their statistical weights into account gives:

$$d(T) = 10271 - 1.29\,(T - 398)\ \mathrm{kg \cdot m^{-3}}.$$

These results are in a good agreement with data of B.Alchagirov *et al*. [5] obtained using picnometer method. Unfortunately, these data cover temperature interval from the eutectic point up to 726 K only. The authors did not report the accuracy of their absolute density values. As the accuracy of gamma absorption method is relatively high, thus the data from present investigation can be used as a reference data.

Based on the results of density and ultrasound velocity measurements shown in Figs 2 and 3 we can not confirm the presence of any structural transformation in the liquid Bi-Pb eutectics similar to the results reported in Ref. [1]. Obviously, even if the transformation exists, its influence on density and ultrasound velocity is within accuracy of our measurements.

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