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Some physical data of the near eutectic liquid lead–bismuth

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Abstract

Lead–bismuth alloys are under intense consideration as target material of spallation sources. The thermohydraulic design of such a target or related coolant systems requires a reliable data basis regarding the temperature dependent physical properties of such alloys. We present measurements of the electrical conductivity and thermoelectric power up to about one hundred degree above the melting point for various alloy compositions. For the eutectic alloy, the measurements were performed up to much higher temperatures including, in addition, viscosity, thermal conductivity and surface tension. A comparison with data and scaling relations available in literature is given.

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1. Introduction

Lead–bismuth alloys are of recent interest for spallation neutron targets [1–3]. Various target designs are under consideration including the set-up and instrumentation of related liquid metal loops [2,4,5]. Obviously, for the design of such targets a reliable knowledge of relevant alloy properties is mandatory. Investigations on different alloy characterizations are, therefore, a continuous and recent subject of research [6–9].

The physical properties, such as density, viscosity, thermal and electrical conductivity or specific heat, are of direct relevance for the thermohydraulic design of a Pb–Bi target and the related liquid metal loop. Several reviews of those data exist. Former measurements performed in Russia have been summarized in the book of Kutateladze et al. [10]. Cevolani and Tinti [11] performed in 1998 a literature review resulting in suggested temperature scalings, which are often used meanwhile. The recent paper of Morita et al. [12] presented data mainly on density, the vapor pressure curve and the vapor equation of state, but included also some literature data on thermal conductivity, viscosity and surface tension. Due to the recent interest in Pb–Bi targets, further literature reviews of the interesting material data exist, see for instance the report [13] or very recently the review of Sobolev [14]. However, the discrepancy between the reported results, different investigated temperature ranges, and sometimes a very limited number of measuring points require new precise measurements in order to obtain reliable data on the temperature dependence of the above mentioned thermophysical properties over a wide temperature range.

In the present paper, new measurements are reported on the electrical conductivity, the thermopower, the viscosity, the thermal conductivity, and the surface tension of the eutectic Pb–Bi alloy in the temperature range between 400 and 1000 K. Related temperature correlation fits are derived and compared with those available in literature. In addition, the electrical conductivity and the thermoelectric power are given for Pb–Bi alloys of varying compositions. Of particular interest, thereby is the melting–solidification range of temperatures, where some hysteresis is observed depending on the heating–cooling cycles.

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2. Measuring methods

2.1. Electrical conductivity and thermoelectric power

The electrical conductivity, $\sigma(T)$, and the thermoelectric power, $S(T)$, were measured by a contact method with a 4-point scheme. According to this scheme the potential drop U is measured at two electrodes located along a straight line between the electrodes carrying the electric current I. Then, the electrical conductivity is determined by $\sigma = Il/US$, where l is the length and S is the cross-section of the sample. When it was difficult to determine precisely the geometric dimensions, σ was found from $\sigma = IG/U$, where G is the geometrical factor, which has been conveniently determined by calibration with mercury.

The experiments were performed in an argon atmosphere. Graphite electrodes for current and potential measurements were placed in the wall of the vertical cylindrical BN-ceramic measuring cell along its vertical axis. The potential electrodes were provided with thermocouples for temperature measurements. Single thermoelectrodes of these thermocouples were used for electrical conductivity and thermoelectric power determination. The melt temperature was determined by WRe-5/20 thermocouples in close contact with the liquid. Temperature gradients of 3–4 K/cm along the cell were additionally controlled to be within 0.1 K by a preliminary calibrated 5-point differential thermocouple. The cell construction permits to carry out the electrical conductivity and thermoelectric power measurements simultaneously in one run. For further details of this method and its experimental realization we refer to Ref. [15].

Pure Pb and Bi were melted and evacuated in sealed quartz ampoules at 10–15 Pa. Then each sample was inserted into the cell directly inside a high-pressure vessel. Thus, the sample composition was accurate within 0.02 wt%. The resultant error of the electrical conductivity measurements is about 2%, and about 5% for the thermoelectric power determination.

2.2. Viscosity

The measurements of the viscosity were carried out using a computer-controlled oscillating-cup viscosimeter [16]. Using the Roscoe equation [17], the dynamic viscosity, $\eta(T)$, has been calculated from the corresponding logarithmic decrement and the period of oscillations. The experiments were performed in helium atmosphere under a negligible excess pressure of about 0.02–0.03 MPa. The sample compositions of about 30 g were accurate to 0.02 wt%. Each sample has been weighed before and after the measurements, and no loss of mass has been observed. Cylindrical graphite crucibles with internal diameter of 14 mm were used. A homogeneous temperature field up to 0.3 K in the range of absolute values up to 800 K has been created inside the furnace. The temperature has been measured with a WRe-5/20 thermocouple arranged just below the crucible. The viscosity was measured with an accuracy of about 3%.

2.3. Thermal conductivity

An experimental arrangement based on the steady-state concentric cylinder method was used for thermal conductivity measurements [18]. The apparatus comprises two coaxial cylinders (stainless steel, BN or graphite) separated by a gap, into which the melt is poured. A central hole is drilled in the inner cylinder for an internal heater made of a molybdenum wire, wound on an alumina form. The inner heater is used for producing the necessary temperature gradient in the investigated melt layer. The cell is closed by a BN cover, which is sealed with a special compound based on a finely dispersed BN powder. The outer three-section furnace is made of molybdenum wire wound on a BN form. The outer heater produces an over-all temperature level, and its upper and lower sections permit regulation of the temperature field over the height of the apparatus. Tungsten–rhenium WR5/20 thermocouples were used in the experiments. Two thermocouples placed in the body of the inner cylinder allow the examination of the temperature distribution over the radius of the apparatus. The coefficient of thermal conductivity, $\lambda(T)$, can be calculated from the formula for the heat transfer in a cylindrical layer. The design of the apparatus assures a maximum reduction of the heat leakage and of convection. The resultant error of thermal conductivity measurements is about 7%.

2.4. Surface tension

The surface tension was measured using a 'large drop' method in the temperature range between T_m and 1000 K. The method is a modification of the sessile drop method and allows overcoming problems connected with a large sessile drop asymmetry [19]. This modification of the sessile drop technique has two advantages, namely it produces a large axisymmetric meniscus and can be used with both wetting and non-wetting systems. Besides, a drop enlargement allows to reduce the experimental uncertainty by almost one order of magnitude. A circular crucible, which has its upper circumferential edge chamfered to an acute angle, is overfilled with fluid, so that an axisymmetric meniscus, with a diameter exceeding the diameter of the crucible is produced and stands proud of the rim. The experiments were performed in an atmosphere of 90%Ar + 10%H_2 after initially evacuating the working volume of the chamber in order to avoid sample oxidation.

The temperature has been measured with the WRe5/20 thermocouple placed near the specimen and was kept constant within 1 K. A CCD camera and a computer-controlled equipment were used for determination of the drop parameters. Based on the Laplace–Young equation, the surface tension, $\gamma(T)$, was calculated by the Kozakevitch