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Thermophysical properties of lead and lead–bismuth eutectic

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Abstract

Among different heavy liquid metals, lead (Pb) and lead–bismuth eutectic (Pb–Bi) are considered at present as the potential candidates for the liquid spallation targets of neutron sources and accelerated driven systems and for the coolant of new generation fast reactors due to their very good neutron and thermal features. Up to now, the published data on the properties of the lead alloys of interest are still limited and significant discrepancies exist between the values given by different sources. This work is a critical review of old and new data reported in the open literature on the main thermo-physical properties of the molten Pb and Pb–Bi: characteristic temperatures, latent heats of melting and evaporation, surface tension, density, heat capacity, viscosity, electric and thermal conductivity. In general, the reliability of data is satisfactory, however, a large uncertainty still exists in the saturation vapour pressure, sound velocity, heat capacity and thermal conductivity. The critical parameters of Pb and Pb–Bi are not well defined yet, and this hinder the development of the equations of state for these coolants. The correlations developed on the basis of the fundamental physical models and the 'best fit' approach are proposed for engineering estimations and design calculations.

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1. Introduction

Many fields of experimental physics, material research and nuclear engineering need a very intense source of neutrons. Recently, the tests of the spallation neutron source (SNS), where neutrons are produced in the mercury target due to spallation process initiated by a high-energy proton beam, started in Oak Ridge National Laboratory of USA [1]. Another application of a liquid metal spallation source is the accelerator driven subcritical systems (ADS) under consideration in different countries as a possible way of nuclear waste burning [2].

Among different heavy liquid metals (HLM), lead (Pb) and lead–bismuth eutectic (Pb–Bi) composed of 44.5 wt% Pb and 55.5 wt% Bi are considered as the potential candidates for both the liquid metal spallation neutron source and the coolant of critical and subcritical (accelerated driven) reactors of a new generation [2,3]. Information about the main physical properties of the HLM such as the melting and boiling points, vapour pressure, density, specific heat, thermal conductivity, viscosity and surface tension is very essential for the optimum

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design and operation of the installations where they are used.

This article compiles data on the thermo-physical properties of the molten Pb and Pb–Bi reported in the open literature. In some cases, significant discrepancies exist among the values given in different sources. Therefore, recommendations based on the 'best fit' of data are usually used. The main sources are material handbooks published in the fifties to seventies [4–12], few compilations appeared (or were updated) after that [13–23] and articles non-included in the above cited compilations and handbooks. Unfortunately, few recent Russian compilation reports and publications could not be taken into account in this article because of difficulties with their accessibility.

2. Melting, boiling and critical points

The melting, boiling and critical temperature of HLM determine the margins of its operation region. The normal melting temperature of the pure lead was measured with a high precision. The value of 600.652 K (327.502 °C) was communicated by Gokcen [24] and 600.612 K (327.462 °C) was given in the last, 8th edition of the Smithells Handbook [21]. For technically pure lead the most probable value is $T_{\mathrm{M(Pb)}} = 600.6 \pm 0.1$ K [23]. The melting temperature of Pb increases by 0.0792 K per 1 MPa when pressure increases from about 15 up to 200 MPa. The increase continues at a lower rate, 0.0671 K per 1 MPa, in the range of 800–1200 MPa, and an increase of 5.4 K for a pressure increase from about 2 to 3 GPa was cited in [25].

The latent heat value for Pb melting at the normal pressure cited in [4,12–15,20,26] lies in the range of 4.76–5.11 kJ mol^{-1}. The mean value of $\Delta H_{\mathrm{M(Pb)}} = 4.9 \pm 0.2$ kJ mol^{-1} was recommended in [23].

The normal melting temperature of Pb–Bi was measured with a lower precision than for lead. Different sources give $T_{\mathrm{M(Pb\text{–}Bi)}}$ from 396.65 K (123.5 °C) up to 398.7 K (125.5 °C) [4,5,11,21,26] at the normal atmospheric pressure. The mean value of $T_{\mathrm{M(Pb\text{–}Bi)}} = 397 \pm 0.6$ K is recommended in the recent review [23]. Similar to the melting temperature of lead, it increases with pressure [24].

The most cited results for the latent heat of melting of Pb–Bi at the normal atmospheric pressure were published in the handbook edited by Friedland [10] $\Delta H_{\mathrm{M(Pb\text{–}Bi)}} = 8.08$ kJ mol^{-1}. In the recent compilation of IAEA $\Delta H_{\mathrm{M(Pb\text{–}Bi)}} = 8.07$ kJ mol^{-1} is recommended [20], which can be used as reference.

The variation in the Pb boiling temperature given by different literature sources [4,5,12,14,15,20,21] is larger than for its melting temperature. The most probable value is $T_{\mathrm{B(Pb)}} = 2016 \pm 10$ K [23]. The information on the latent heat of vaporization of Pb and Pb–Bi even at the normal boiling point (the enthalpy change on boiling) is not numerous. The following sources were included in the database: [4,13,14,20,21]. The minimum value of $\Delta H_{\mathrm{B(Pb)}} = 177.5$ kJ mol^{-1} was given for Pb in the old handbook of Lyon [4], and $\Delta H_{\mathrm{B(Pb)}} = 178$ kJ mol^{-1} was recently given in the recommendations of the IAEA Report [20]. The higher value of $\Delta H_{\mathrm{B(Pb)}} = 178.8$ kJ mol^{-1} was published in Smithells Metals Reference Book [21]. The difference between these values is less than 1%. The mean value of $\Delta H_{\mathrm{B(Pb)}} = 178.1 \pm 0.6$ kJ mol^{-1} can be recommended.

For Pb–Bi the boiling point of $T_{\mathrm{B(Pb\text{–}Bi)}} = 1943$ K (1670 °C) at the normal atmospheric pressure is given in the old handbooks [4,5,10,11]. This value, reproduced later in the IAEA compilation [20] $T_{\mathrm{B(Pb\text{–}Bi)}} = 1943 \pm 20$ K, can be recommended. For the latent heat of vaporization of Pb–Bi at the normal boiling point, $\Delta H_{\mathrm{B(Pb\text{–}Bi)}} = 178.352$ kJ mol^{-1} was published in the handbook of Friedland [10]. The IAEA compilation [20] gives a value of 178 kJ mol^{-1}. The mean values of $\Delta H_{\mathrm{B(Pb\text{–}Bi)}} = 178.2 \pm 0.4$ kJ mol^{-1} is recommended in [23].

A very large uncertainty exists in the critical temperatures of both Pb and Pb–Bi. The recent critical review of Morita et al. [27] showed that different sources give $T_{\mathrm{c(Pb)}} = 3370\text{–}5400$ K and $T_{\mathrm{c(Pb\text{–}Bi)}} = 2411\text{–}4890$ K.

3. Saturation vapour pressure

The saturated vapour pressure of a liquid metal is an important property which is directly related to the latent heat of evaporation. For the equilibrium between the liquid and vapour phases, the Clausius–Clapeyron equation states that

$$\left(\frac{\mathrm{d}p}{\mathrm{d}T}\right)_s = \frac{\Delta H_{\mathrm{B}}}{T_{\mathrm{s}} \cdot (V'' - V')}, \qquad (1)$$

where p is pressure, T – temperature, ΔH_{B} – enthalpy (heat) of evaporation, V'' and V' specific volumes of vapour and liquid phases.

Therefore, assuming that the vapour behaves as a perfect gas and neglecting the volume of liquid in comparison with that of the gas, it can be obtained for the saturated vapour pressure:

$$p_s = A \cdot \exp(-\Delta H_B/RT), \tag{2}$$

where A is a constant of integration and $R = 8.314\ \mathrm{J\ mol^{-1}\ K^{-1}}$ is the universal gas constant. For Pb and Pb–Bi at low pressure, the above equation can provide approximate values for equilibrium vapour pressures over a wide range of temperature due to a relatively small variation of ΔH_B with temperature.

The data for the saturation vapour pressure of lead at different temperatures were found in [4,10,12,14,15,21,25,26]. Fig. 1(a) shows the Pb vapour pressure from the above-cited literature and plotted as a function of temperature. At first view, the dispersion is small at temperatures higher than 1500 K. However it becomes about 15% when temperature decreases down to 1000 K. The following correlation was recommended in [23] for the saturated vapour pressure (p_s) of the molten lead from the normal melting temperature up to the normal boiling point:

$$p_{s(\mathrm{Pb})}\ [\mathrm{Pa}] = 6.572 \times 10^9 \cdot \exp\left(-\frac{184\,960}{RT}\right), \tag{3}$$

where temperature is in K.

The value of 184.96 kJ mol^{-1} in the exponential function is in a good agreement with $\Delta H_{B(\mathrm{Pb})} = 177.8\ \mathrm{kJ\ mol^{-1}}$ indicated above. First experimental

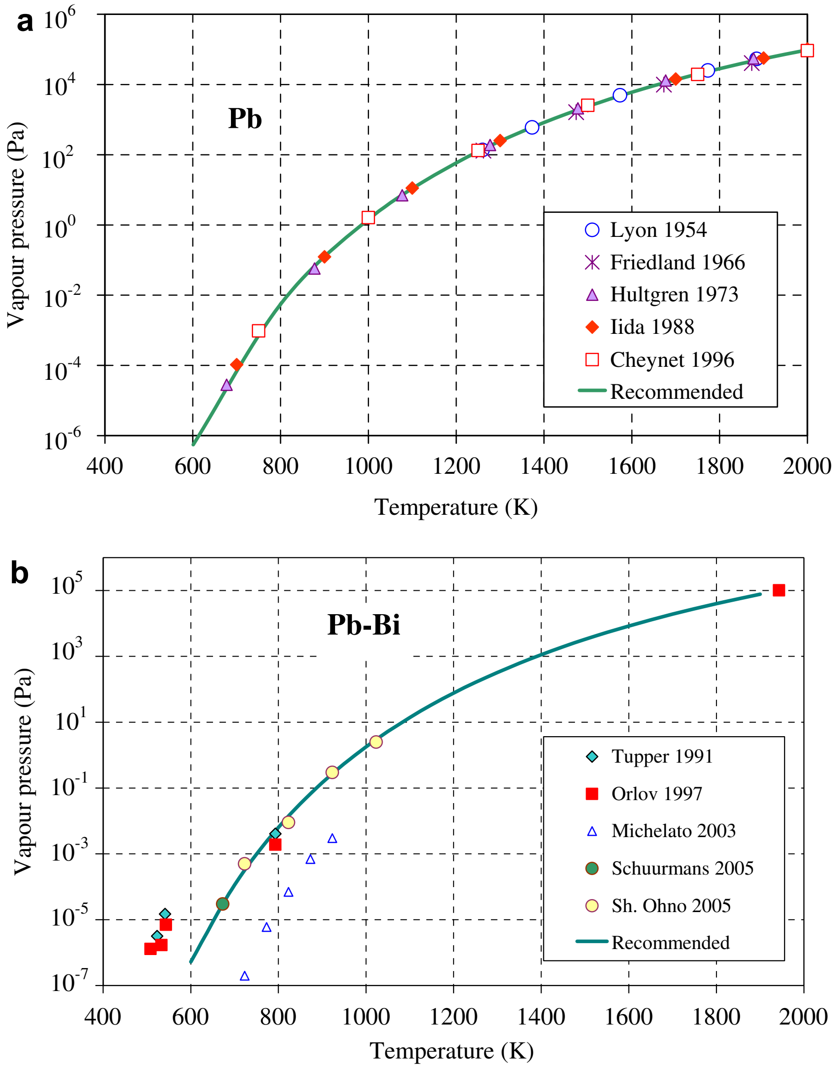

Fig. 1. Saturated vapour pressures of Pb (a) and Pb–Bi (b) at the normal pressure versus temperature.

results giving the Pb–Bi (saturation) vapour pressure at temperatures 508–541 K and 793–823 K were published by Tupper et al. [28]. Later, Orlov [29] presented the experimental values for five different temperatures. In both publications, it was mentioned, however, that a very large uncertainty exists at temperatures below 773 K. Some new results have recently been communicated by Schuurmans et al. [30] and published by Ohno et al. [31]. All these results are plotted in Fig. 1(b).

In the temperature range of 650–1940 K, the vapour pressure of Pb is very close to the experimental values for Pb–Bi available from the considered sources. At temperatures of 500–550 K, Tupper et al. [28], and Orlov [29], give Pb–Bi vapour pressure that is an order of magnitude higher above the liquid–vapour equilibrium lines of lead and bismuth extrapolated to the lower temperatures. However, it should be noticed that at the expected level of saturated pressure ($\sim 10^{-8}$ Pa) it is very difficult to measure the pressure correctly, and overestimation by several orders of magnitude can be obtained due to contamination of the liquid metal or of the experimental rig. Therefore it is proposed that these points are not considered at present. The remaining values can be described with

$$p_{\mathrm{s(Pb-Bi)}}\ [\mathrm{Pa}] = 1.11 \times 10^{10} \cdot \exp\left(-\frac{187500}{RT}\right). \tag{4}$$

The obtained value $\Delta H_{\mathrm{B(Pb-Bi)}} = 187.5\ \mathrm{kJ\ mol^{-1}}$ is in a good agreement with the latent heat of Pb–Bi evaporation recommended in the previous paragraph ($\Delta H_{\mathrm{B(Pb-Bi)}} = 177.8\ \mathrm{kJ\ mol^{-1}}$), taking into account the large uncertainty in the experimental results used for its calculation. The maximum dispersion of the experimental data from the correlation (4) is less than ±60% in the temperature range of 673–1943 K.

4. Surface tension

A surface tension of liquid surfaces (σ) is related to tendency to minimize their surface energy. It is a measure of the cohesive energy of atoms in a liquid metal, therefore it correlates with the latent heat of evaporation. The surface tension of normal liquids decreases with temperature linearly to zero at the critical temperature (according to the Eötvös' law).

The temperature dependence of the molten lead surface tension was found in [4,10,13,14,19–21,32–34]. In 1993, Keene [34] reviewed many previous works to obtain values of the coefficients in his linear empirical correlation. The coefficients obtained by him on the basis of all considered sources are very close to those recommended in 1986 by Jauch and Schulz [33]. More recent compilations [19,20] give only the tables of the recommended data. Fig. 2 shows the surface tension of lead in function of temperature from the selected sources. A difference of ±5% exists between the surface tension values given by different sources at $T < 1000$ K. At higher temperatures, the scatter increases. The correlation proposed by Jauch and Schultz [33] allows

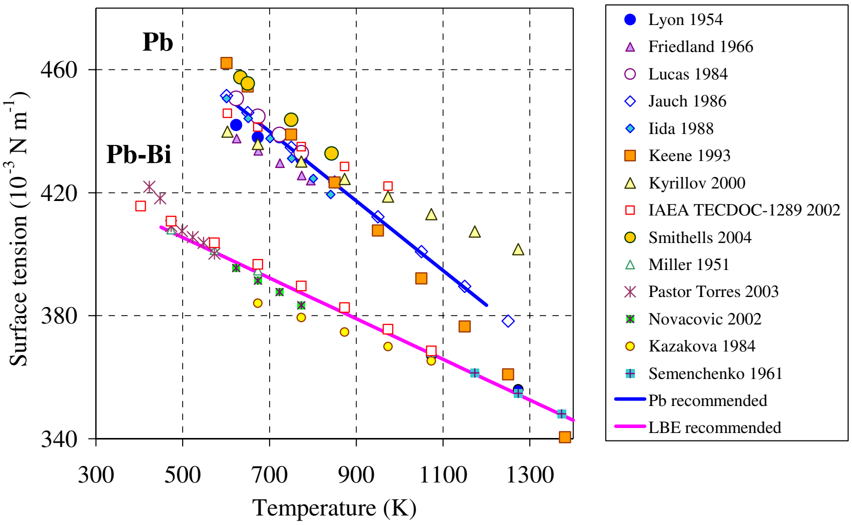

Fig. 2. Surface tension of the molten lead and Pb–Bi as a function of temperature.

a compromise represented by the following formula for the surface tension (σ) of lead:

$$\sigma_{(\mathrm{Pb})}\ [\mathrm{N\ m^{-1}}] = 0.519 - 1.13 \times 10^{-4} \cdot T. \tag{5}$$

The surface tension of the molten Pb–Bi was found in [4,35–40]. The data from the above cited sources are presented in Fig. 2. The analysed data and recommendations for the Pb–Bi surface tension can be described with an uncertainty of less than 3% by the following linear correlation:

$$\sigma_{(\mathrm{Pb–Bi})}\ [\mathrm{N\ m^{-1}}] = 0.437 - 6.6 \times 10^{-5} \cdot T. \tag{6}$$

The above correlation is recommended for estimation of the surface tension of molten Pb–Bi at normal pressure up to 1400 K.

5. Density and thermal expansion

The temperature dependence of density provides essential information for the development of an equation of state (EOS). The data for the density of the molten lead from [4,5,9,13,14,19–21,41] were analysed. The Pb–Bi density was taken from [4,5,20,42]. Fig. 3 shows the molten lead and Pb–Bi densities values from the selected sources versus temperature. The linear regression of the Pb and Pb–Bi density (ρ) values presented in Fig. 3 yields

$$\rho_{(\mathrm{Pb})}\ [\mathrm{kg\ m^{-3}}] = 11367 - 1.1944 \cdot T, \tag{7}$$

$$\rho_{(\mathrm{Pb–Bi})}\ [\mathrm{kg\ m^{-3}}] = 11096 - 1.3236 \cdot T. \tag{8}$$

The deviation of the selected values from correlations (7) and (8) does not exceed 0.7% for Pb and 0.8% for Pb–Bi.

Literature sources describing a direct measurement of the coefficient of thermal expansion (CTE) of molten Pb and Pb–Bi were not found. Therefore, the above information on the temperature dependence of their density was used for deduction of the isobaric volumetric coefficient of thermal expansion (β_p). In a general case, a link between the density and CTE is defined by the following formula:

$$\beta_\mathrm{p}(T) \equiv \frac{1}{V} \cdot \left(\frac{\partial V}{\partial T}\right)_\mathrm{p} = -\frac{1}{\rho} \cdot \left(\frac{\partial \rho}{\partial T}\right)_\mathrm{p}. \tag{9}$$

Substituting the correlations (7) and (8) into definition (9) yields the following formulae for the isobaric volumetric CTE of the molten lead and Pb–Bi, respectively:

$$\beta_{\mathrm{p(Pb)}}\ [\mathrm{K^{-1}}] = \frac{1}{9516.9 - T}, \tag{10}$$

$$\beta_{\mathrm{p(Pb–Bi)}}\ [\mathrm{K^{-1}}] = \frac{1}{8383.2 - T}. \tag{11}$$

CTE increases with temperature for both Pb and Pb–Bi due to decreasing the interatomic forces (configurational internal energy). Comparison of the last two correlations (slopes of two lines in Fig. 3) also shows that Pb–Bi has a higher CTE than Pb. In principle, the lower attractive forces between Pb and Bi atoms in Pb–Bi than in the pure lead can explain this.

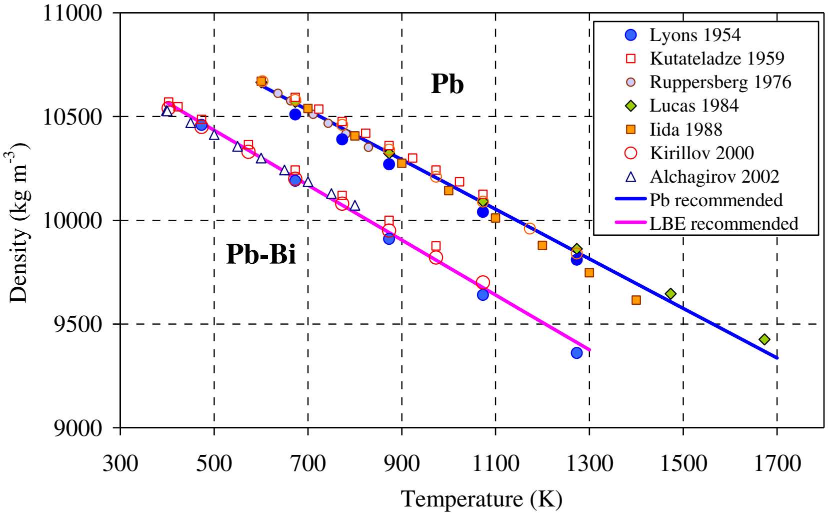

Fig. 3. Density of the molten lead and Pb–Bi as a function of temperature at the normal pressure.

6. Compressibility and sound velocity

There are no results for direct measurement of the compressibility for molten Pb, and Pb–Bi in the literature. Usually, the adiabatic compressibility K_S (or the adiabatic elastic modulus B_S) is found from the results of measurement of the (ultra)sound velocity $\mathbf{v}_{us}$ and density ρ using the following thermodynamic relationship [43]:

$$B_S \equiv -V\left(\frac{\partial p}{\partial V}\right)\bigg|_S \equiv \frac{1}{K_S} = \frac{\rho}{(\partial\rho/\partial p)|_S} = \rho \cdot \mathbf{v}_{us}^2. \tag{12}$$

The velocity of propagation of the longitudinal ultrasound waves in molten lead was measured by Kleppa [44] close to the melting point, by Gordon [45] over a temperature interval from the melting temperature to 643 K, by Mustafin and Shaikhiev [46] over the temperature range 601–2000 K and by Konyuchenko [47] over the temperature range 673–973 K. Rather a good agreement is observed among the first three sources in the region of the melting temperature. The sound velocity values presented by Konyuchenko [46] are 3% higher. No information was given on Pb purity and on the measurement method used. The most complete and reliable data seems to be that from Mustafin and Shaikhiev [46], therefore the correlation recommended for the estimation of the sound velocity in the molten lead is

$$\mathbf{v}_{us(Pb)}\ [\mathrm{m\,s^{-1}}] = 1951.75 - 0.3423 \cdot T + 7.635 \times 10^{-5} \cdot T^2. \tag{13}$$

The ultrasound velocity in the molten Pb–Bi has only been measured by Kažys et al. [48] over the temperature range 433–603 K. Azad [49] calculated the sound velocity in molten Pb–Bi using the experimental data on the sound velocity in the molten Pb [46] and in the molten Bi [50] using the Vegard's law. The values for the sound velocity in Pb–Bi are plotted in Fig. 4 together for lead and Pb–Bi with plots for the recommended correlations. The following correlation for the sound velocity in Pb–Bi is based on the experimental results of Kažys et al. [48]:

$$\mathbf{v}_{us(Pb\text{–}Bi)}\ [\mathrm{m\,s^{-1}}] = 1773 + 0.1049 \cdot T - 2.873 \times 10^{-4} \cdot T^2. \tag{14}$$

In the respective temperature range and at the normal atmospheric pressure, the temperature dependence of the elastic modulus can be described with the help of parabolic functions:

$$B_{S(Pb)}\ [\mathrm{GPa}] = 42.15 - 1.652 \times 10^{-2} \cdot T + 3.273 \times 10^{-6} \cdot T^2, \tag{15}$$

$$B_{S(Pb\text{–}Bi)}\ [\mathrm{GPa}] = 35.18 - 1.541 \times 10^{-3} \cdot T - 9.191 \times 10^{-6} \cdot T^2. \tag{16}$$

7. Heat capacity

Available experimental data on the heat capacity of HLM are even less numerous than those for density. The theoretical calculation of heat capacity of liquids is restricted by the extreme complexity of the motion of atoms. Often, the same types of empir-

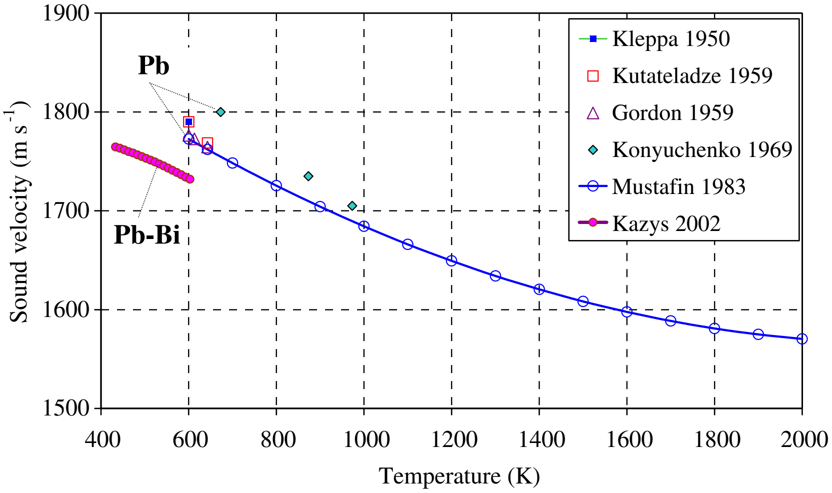

Fig. 4. Ultrasound longitudinal velocity in the molten lead and Pb–Bi as a function of temperature.

ical correlations as for solids are used to fit the experimental results. An extensive review of the existing experimental data on the heat capacity of lead and Pb–Bi was performed by Imbeni et al. [17,18]. The heat capacity data of lead from [4,5,10,12,14,51] have been analysed. Fig. 5(a) shows the Pb heat capacity from these sources as a function of temperature. Lyon [4] gives higher values for the heat capacity of lead than those reported by other sources. The values recommended by Kutateladze et al. [5] are independent of temperature. Hultgren et al. [12] presented the data selected from many previous publications and the heat capacity, which decreases linearly with temperature up to 1270 K and then remains constant. Kubaschewski et al. [26] presented an analytical equation based on the data from many previous sources. Gurvich et al. [51] gave tables with data obtained by benchmarked models. Smithells Material Reference Book [21] reported the values taken from the sources of the fifties and sixties. The recent compilation of IAEA repeats the data of Kutateladze et al. [5] in contrast to the conclusion of Chapman (based on a simplified statistical model) that heat capacity of liquid metals should decrease with temperature at least in the region of the melting point [52]. A simple parabolic correlation is proposed for estimation of the isobaric specific heat capacity (c_p) of the molten Pb in the temperature range of T_M to 1500 K:

$$c_{p(\mathrm{Pb})}\ [\mathrm{J\ kg^{-1}\ K^{-1}}] = 162.9 - 3.022 \times 10^{-2} \cdot T + 8.341 \times 10^{-6} \cdot T^2. \tag{17}$$

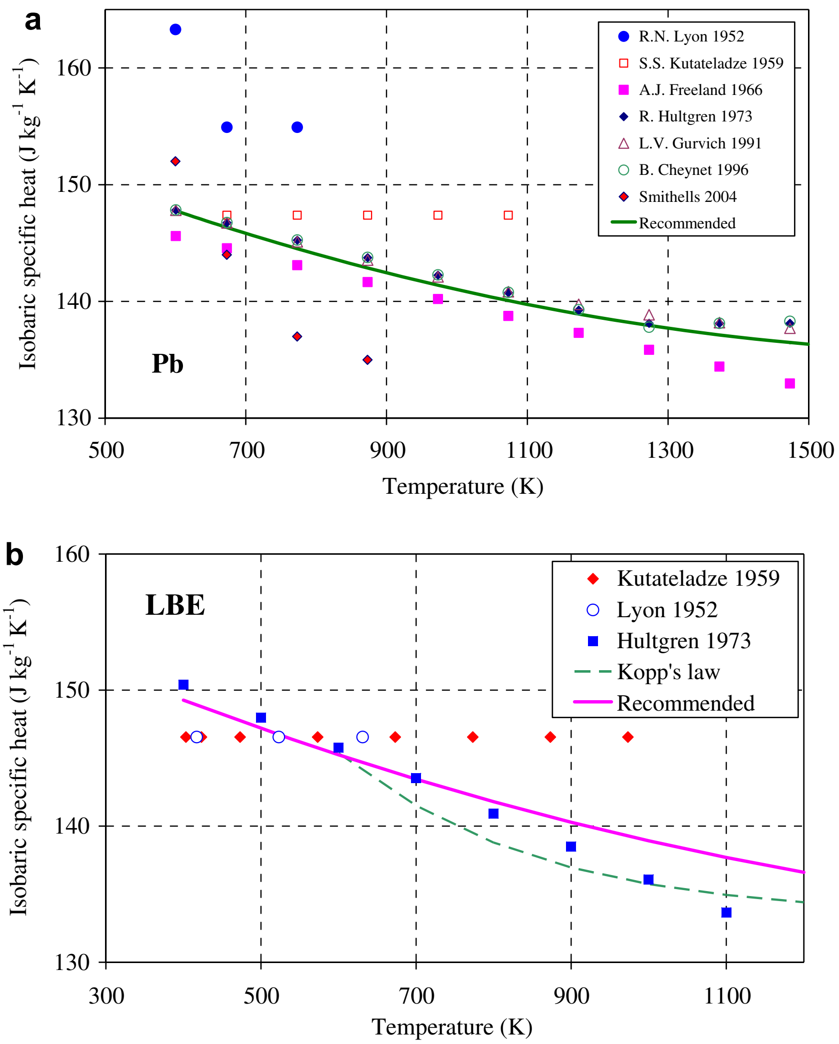

Fig. 5. Isobaric specific heat of the molten lead (a) and Pb–Bi (b) at the normal pressure as a function of temperature.

Only three independent sources, where heat capacity of the molten Pb–Bi was given at different temperatures, were found [4,5,11] The two older publications [4,5] recommend the same value of $c_{p(\mathrm{Pb–Bi})} = 146.5\ \mathrm{J\,kg^{-1}\,K^{-1}}$ ($0.035\ \mathrm{cal\,g^{-1}\,°C^{-1}}$) for a range of temperatures. In the second chapter of the original handbook of Lyon [4] it was only indicated that this value has been obtained in the temperature interval from 417 to 631 K. In [5] this value was recommended for temperatures from 403 to 973 K. This recommendation was reproduced in later handbooks [6–10] (some of them even extended it to higher temperatures without any explanation) and was recently repeated in [19,20]. The only source which reports that the Pb–Bi heat capacity depends (decreases) on temperature is Hultgren et al. [11], which refers to a report of Douglas and Dever [53] as the original source. The data from [11] were reprinted in later compilations [18,27]. In Fig. 5(b), the recommendations for the specific heat of Pb–Bi and also a line obtained with the additive Kopp's law (often used for calculation of the heat capacities of the binary systems) are also plotted. A reasonable agreement between the values of [11] and those given by the Kopp's law exists; however, the temperature dependence is different, leading to a larger difference at higher temperatures. Attempting, on the one hand, to take into account the similarity between Pb–Bi and its components (Pb and Bi) and, on other hand, to use the available experimental data (also the values obtained with the Kopp's law), a fitting with the following parabolic polynomial was performed in the temperature range of 400–1100 K:

$$c_{p(\mathrm{LBE})}\ [\mathrm{J\,kg^{-1}\,K^{-1}}] = 159 - 2.72 \times 10^{-2} \cdot T + 7.12 \times 10^{-6} \cdot T^2. \tag{18}$$

The deviation of the literature values from the recommended correlations (17) and (18) does not exceed 5% in the temperature range of 601–1500 K for Pb and 400–1100 K for Pb–Bi.

8. Viscosity

Accurate and reliable data on viscosity of LHM are not abundant. Some discrepancies between experimental data can be attributed to their high reactivity, to the difficulty of precise measurements at elevated temperatures, and to a lack of a rigorous theory for calculations. This physical parameter is never additive for mixtures or alloys. All HLM are believed to be Newtonian liquids and the temperature dependence of their viscosity are usually described by an Arrhenius type formula:

$$\eta = \eta_0 \cdot \exp\left(\frac{E_\eta}{RT}\right), \tag{19}$$

where E_η is the activation energy of motion of viscous flow and η_0 is a coefficient. The Pb and Pb–Bi viscosity data were taken for analysis from [4,5,7,13,14,19–21,25,54,55].

Fig. 6 shows the viscosity values for the molten Pb (a) and Pb–Bi (b) taken from the above cited literature. A good agreement exists among the different sets of experimental data and the values calculated by means of the empirical equations. It can be seen that viscosity of Pb–Bi is significantly

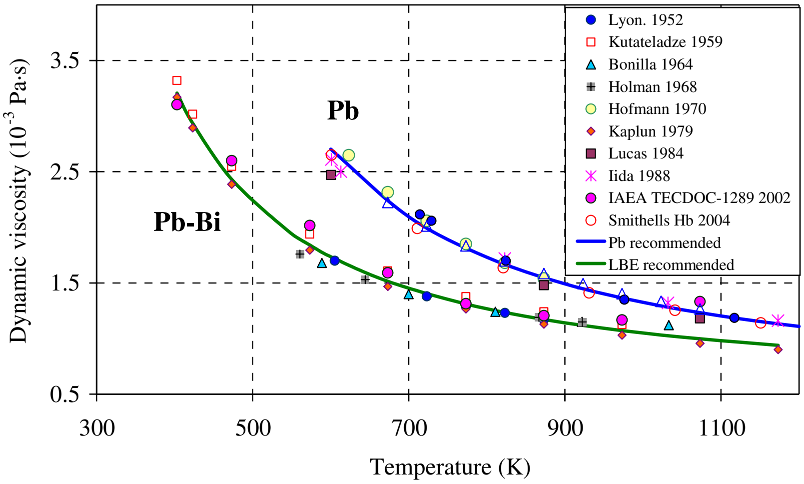

Fig. 6. Dynamic viscosity of the molten lead and Pb–Bi at the normal pressure as a function of temperature.

lower than that of lead, especially at lower temperatures. The more recent data for Pb–Bi from the IAEA compilation [20] are very close to the old data of Kutateladze et al. [5] up to about 900 K. At higher temperatures, they deviate to higher values, probably, due to the presence of oxides or other impurities. A reliable choice of an empirical equation to describe the temperature dependence of the dynamic viscosity of molten lead and Pb–Bi can be obtained by fitting the selected values into an Arrhenius type equation of the form

$$\eta_{(\mathrm{Pb})}\ [\mathrm{Pa\ s}] = 4.55 \times 10^{-4} \cdot \exp\left(\frac{8888}{RT}\right), \tag{20}$$

$$\eta_{(\mathrm{LBE})}\ [\mathrm{Pa\ s}] = 4.94 \times 10^{-4} \cdot \exp\left(\frac{6270}{RT}\right). \tag{21}$$

These correlations are valid in the temperature range from T_{M} to 1470 K for Pb and in the temperature range of 400–1100 K for Pb–Bi with an estimated uncertainty of about ±6%.

9. Electrical resistivity

The electrical resistivity of pure liquid metals increases linearly with temperature (with rare exceptions) and rises with impurities content. The data on the electrical resistivity of lead as a function of temperature were found in the different handbooks cited above [4,7,14,19–21,25] and in a review of Bretonnet [56] who treated different sources to obtain his correlation. The agreement between different sources is very good (Fig. 7). A semi-empirical liner correlation with the coefficients given by Iida and Guthrie [14] can be recommended for calculation of the specific electrical resistivity (χ) of liquid Pb as a function of temperature:

$$\chi_{(\mathrm{Pb})}\ [\Omega\ \mathrm{m}] = (66.6 + 0.0479 \cdot T) \times 10^{-8}. \tag{22}$$

This correlation is valid in the temperature range of 601–1273 K. The deviation of the selected data from this correlation is less than 1%.

Only two reliable data sources for the electric resistivity of molten Pb–Bi were found in the literature: Lyon et al. [4] (probably repeated in [5,7]) and the IAEA Report [20]. Friedland [10] reported only one value at 811 K. Fig. 7 shows the values of the electrical resistivity for molten lead and Pb–Bi taken from the literature cited above. The Pb–Bi resistivity data recommended in the IAEA report [20] fit very well with those calculated with the Vegard–Kopp's law using the resistivities of lead and bismuth. The recommendations of Lyon [5,7] yield higher values. The average linear interpolation produces the following linear correlation for the Pb–Bi electric resistivity:

$$\chi_{(\mathrm{Pb\text{–}Bi})}\ [\Omega\ \mathrm{m}] = (86.334 + 0.0511 \cdot T) \times 10^{-8}. \tag{23}$$

The maximum difference does not exceed 2.5% in the temperature range of 400–1100 K.

10. Thermal conductivity

Experimental determination of thermal conductivity of HLM is difficult because of the problems

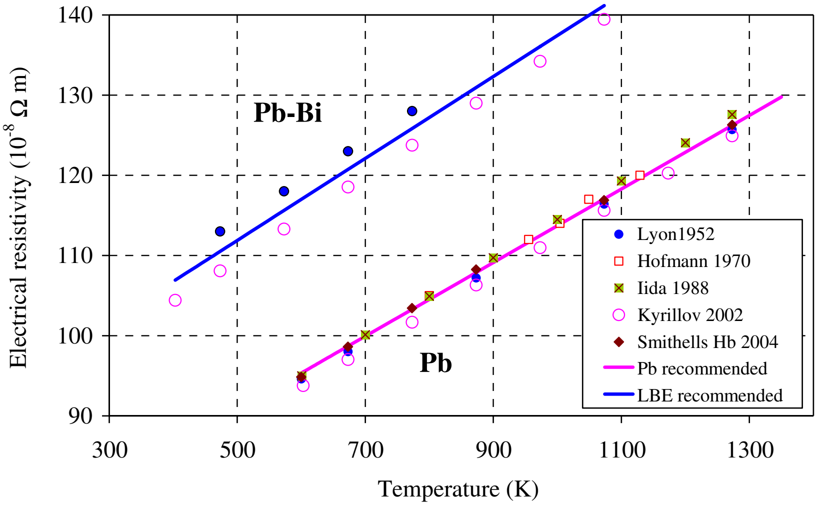

Fig. 7. Electrical resistivity of the molten lead and Pb–Bi at the normal pressure versus temperature.

related to convection and to wetting. At present, only few experimental data are available with large discrepancies between different sets of data. The high thermal conductivity of liquid metals is mainly due to free electrons. A simple theoretical relation exists for metals between electrical and thermal conductivities known as the Wiedemann–Franz–Lorenz law [57]:

$$\kappa_e = L_0 \cdot T/\chi, \tag{24}$$

where κ_e is the electronic thermal conductivity, χ is the electric resistivity and $L_o = 2.45 \times 10^{-8}$ W Ω K^{-2} is the Lorenz number. The contribution of atomic vibrations and movements into the thermal conductivity of the liquid metals of interest is small and can be neglected in the temperature range of 400–1200 K. Therefore, the prediction of thermal conductivity of liquid metals and alloys is possible by combining the Wiedemann–Franz–Lorenz law and the existing, reliable values of the electrical resistivity.

The following sources containing data on the lead thermal conductivity were analysed: [4,5,9,10,14,19–21,33,58,59]. A very large dispersion of data from different sources is present (Fig. 8(a)). The earlier results reported by Lyon et al. [4] and by Kutateladze et al. [5] show very different temperature dependencies – the thermal conductivity decreases with temperature in the first, while it increases in the second. A decrease of the lead thermal conductivity with temperature reported by Lyon et al. [4] and Yamasue et al. [58] and an anomaly reported by Kutateladze et al. [5] could be attributed to oxidation effects. Millis et al. [59] considered

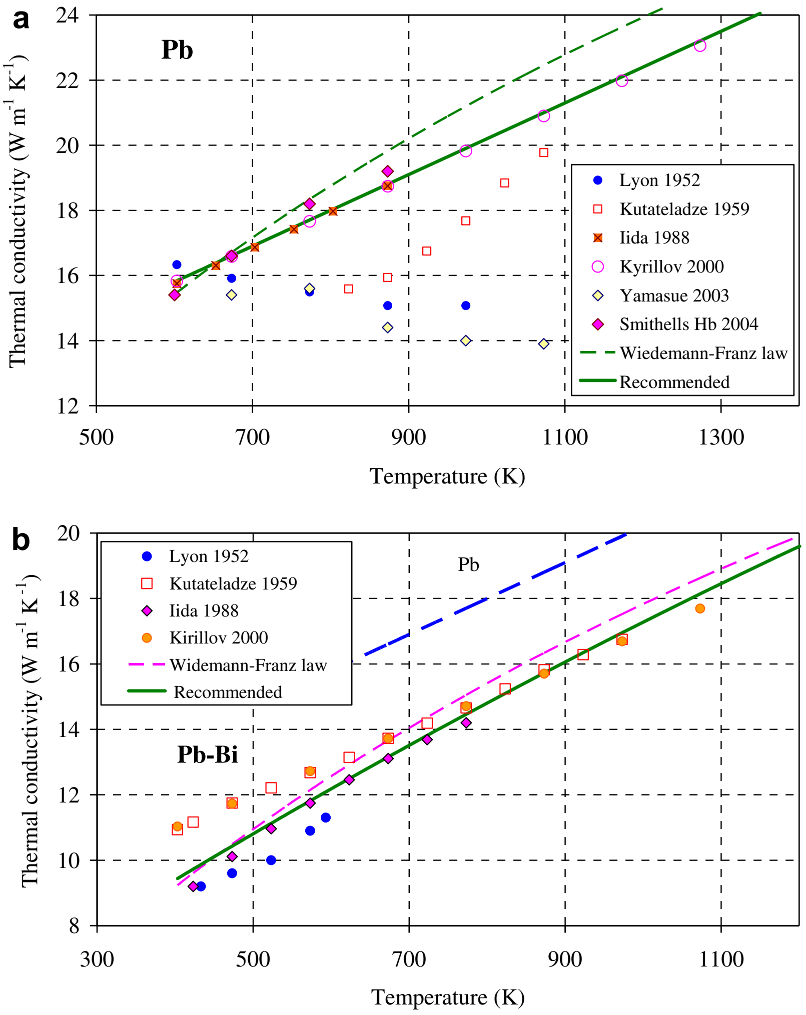

Fig. 8. Thermal conductivity of molten lead (a) and Pb–Bi (b) at the normal pressure versus temperature.

many sources and recommended a linear function for the temperature dependence of thermal conductivity of molten metals. Similar recommendation was given in [14,19,20].

In an effort to find a physically reasonable compromise between the available data and taking into account the data obtained with the Wiedemann–Franz–Lorenz law, a linear correlation was chosen as recommendation for the thermal conductivity of molten lead:

$$\kappa_{(\mathrm{Pb})}\ [\mathrm{W\ m^{-1}\ K^{-1}}] = 9.2 + 0.011 \cdot T. \tag{25}$$

This correlation is applicable in the temperature range of $T_{\mathrm{M}} - 1300$ K.

The information on thermal conductivity of the molten Pb–Bi mainly comes from the old handbooks [4,5]. Later handbooks or reviews reference these data. Only in the handbook of Iida and Guthrie [14], some values appear that differ from those cited above. In the Russian literature, new data were summarised in the handbook of Kyrillov and Bogoslovslaya [19] without direct references to the sources. Later, the last recommendations were practically reproduced in [20]. The values of the Pb–Bi thermal conductivity extracted from the selected sources are presented in Fig. 8(b) together with the recommended values for lead. A non-negligible difference exists between the old Western and Russian recommendations at lower temperatures, the latter giving 15–20% higher values. The recommendation of Iida and Guthrie [14] provides intermediate values.

Similar to viscosity and electric resistivity, the results on thermal conductivity are strongly affected by the purity of Pb–Bi. Thermal conductivity is not an additive parameter; therefore, the Vegard–Kopp's law cannot be used to estimate it. Contrarily, the Wiedemann–Franz law, which is valid for most metals, can be applied. Using the above recommended correlation for the Pb–Bi electrical resistivity, one can obtain the following correlation for the thermal conductivity:

$$\kappa_{\mathrm{W-F(Pb-Bi)}}\ [\mathrm{W\ m^{-1}\ K^{-1}}] \approx \frac{2.45 \cdot T}{86.33 + 0.0511 \cdot T}. \tag{26}$$

Eq. (26) gives values close to the recommendations of [14] at lower temperatures: 400–550 K, and close to the recommendations of [5,20] at higher temperatures: 600–1100 K. So, the Wiedemann–Franz law can be used for the estimation of the Pb–Bi thermal conductivity if its electrical resistivity is known.

Fitting the data of [4,5,14,20] with a parabolic function yields the following recommended correlation:

$$\kappa_{(\mathrm{Pb-Bi})}\ [\mathrm{W\ m^{-1}\ K^{-1}}] = 3.61 + 1.517 \times 10^{-2} \cdot T - 1.741 \times 10^{-6} \cdot T^2. \tag{27}$$

The maximum deviation of the analysed experimental data for the Pb–Bi thermal conductivity from the correlation (27) is 15% and observed at low temperatures.

11. Conclusions

The review of the available data on the thermophysical properties of the molten Pb and Pb–Bi shows that significant dispersion exists between different sources (except the melting and boiling points, and the density). A large uncertainty still exists for the heat capacity and the thermal conductivity – different sources give even different temperature dependence for these parameters. The critical parameters, the saturation vapour pressure and the temperature dependence of the latent heat of vaporisation of Pb and Pb–Bi are not yet well measured, and this hinders the development of the correct equation of state for these coolants. Nevertheless, the reliability of the existing data is satisfactory and the agreement among the certain sources is observed. This allows developing the empirical and semi-empirical correlations for main thermophysical properties of the molten Pb and Pb–Bi at normal pressure. The proposed correlations can serve as temporary recommendations in engineering estimations and design calculations.

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