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HEAT AND MASS TRANSFER AND PROPERTIES OF WORKING FLUIDS AND MATERIALS

Recommendations on Adopting the Values and Correlations for Calculating the Thermophysical and Kinetic Properties of Liquid Lead

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Abstract—Recent years have seen an essentially increased interest in studying the properties of liquid lead, which is primarily connected with the possibility of using it as coolant in nuclear power installations, first of all, in reactors based on fission of heavy nuclei by fast neutrons. The article presents an analysis of published data on the thermophysical and kinetic properties of lead in liquid state, the results of which served as a basis for selecting and recommending correlations to be used in carrying out scientific and engineering calculations. A general assessment of the state of experimental investigations into the thermophysical properties of liquid lead is presented. The presented value of lead solidification temperature is the maximally reliable one. The data on the boiling temperature, melting and vaporization enthalpies, and saturated vapor pressure have been determined with satisfactory accuracy. The published data on the liquid lead heat capacity differ considerably from each other; therefore, the recommended values should be experimentally checked and determined more exactly. The available experimental data on surface tension density, volumetric expansion coefficient, sound velocity, viscosity, and thermal conductivity do not cover the entire range of liquid phase existence temperatures. The temperature region above 1200 K and the crystal–liquid phase transition region are the least studied ones. Additional investigations of these properties in the above-mentioned temperature intervals are necessary. The question about the influence of impurities on the thermophysical properties of lead still remains to be answered and requires experimental investigations.

Keywords: lead, thermophysical properties, kinetic properties, experimental investigation, correlation

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In the period from the 1950s to 1980s, a great amount of experimental investigations into the thermophysical properties of lead was carried out. In the last decade, a lot of reviews and handbooks (e.g., [1–3]) on the thermophysical properties of lead have been published; however, they as a rule contain data borrowed from earlier works aimed at determining reference data [4–6].

At present, experimental investigations are carried out (although in a smaller amount) in order to refine or supplement the previously obtained results.

As more experimental data become available, they should be analyzed and put in systematic order. In this article, we briefly analyze the data available in the literature and make references to the works containing, in our opinion, the most reliable information.

Some notes should be made about the data selection criteria. As a rule, two methods for selecting the recommended values of thermophysical parameters are used in the literature. First, attempts to statistically analyze the data from a large number of works are encountered. In applying such an approach, data should be dealt with very carefully, and its use for heterogeneous data seems to be incorrect. The main difficulties arising in applying such an approach to analyzing the published data on thermophysical properties are described in detail in [7]. The other approach to analyzing experimental data is aimed at revealing the most reliable ones from them, which are then recommended for use. The authors of this article predominantly used the latter approach in the present work. The following conditions were adopted as validity criteria: consistency of the results with the data of other experimental works, qualitative agreement with the well-known theoretical regularities, the values of errors, and carefulness of the performed measurements. In case of lack of new reliable experimental information about the considered properties, it was proposed to use data from well-proven handbooks.

Phase transition temperatures. The lead solidification temperature at standard pressure is the secondary reference point of the ITS-90 temperature scale [8]; it has been determined with accuracy of 0.001 K and in all likelihood cannot be determined more exactly until

Recommended values of the lead thermophysical properties

Parameter	Value of formula for determining the parameter	Error	Temperature range
Solidification temperature, K	600.612	0.001	
Boiling temperature, K	2016	10	
Melting enthalpy, J/kg	23225.1	193	
Vaporization enthalpy, J/kg	858150	193	
Density, kg/m^3	$\rho = 11200.23 - 0.66285T - 0.55397 \times 10^{-3}T^2 + 0.17453 \times 10^{-6}T^3$	0.3%	600.6–1500
Volumetric thermal expansion coefficient, 1/K	$\beta = 7.324 \times 10^{-5} + 0.57384 \times 10^{-5}T$	3%	600.6–1500
Density jump in melting and crystallization, %	3.3	0.25	
Saturated vapor pressure, Pa	$\log(p_s) = 15.78397 - 10330.87/T + 0.0001545T - 1.81309\log(T)$	10%	600.6–2016
Surface tension, N/m	$\sigma = 0.50183237 - 0.08614 \times 10^{-3}T$	0.5%	600.6–1300
Sound velocity, m/s	$u_s = 1921.79 - 0.12464T - 0.07298 \times 10^{-3}T^2$	0.2%	600.6–1000
Heat capacity, J/(kg K)	$c_p = 175.1 - 0.04961T - 1.524 \times 10^5T^{-2} + 1.985 \times 10^{-5}T^2 - 2.099 \times 10^{-9}T^3$		600.6–1300
Viscosity, Pa s	$\eta = 4.55 \times 10^{-4}\exp(1069/T)$	5%	650–1400
Electric resistivity, Ω m	$R \times 10^8 = 65.73 + 4.65 \times 10^{-2}T$	1%	600.6–1200
Thermal conductivity, W/(m K)	$\lambda = 3.289 + 0.0274T - 7.87 \times 10^{-6}T^2$	4%	600.6–1300

a new temperature scale is adopted. Investigations of the effect of impurities on this parameter can be of certain interest for engineering applications.

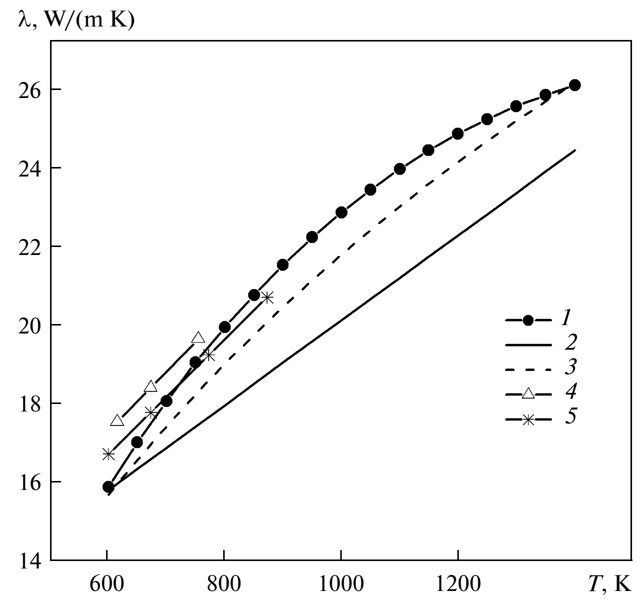
According to [3], the data on the lead boiling temperature at standard pressure presented in different sources differ from each other by no more than 25 K. Attempts to further refining this value are of no point unless matters relating to the kinetics of boiling processes and to the effect of impurities on this parameters are studied in detail.

Heat of phase transitions. Data on melting enthalpy are presented in a large number of sources. In the review [3] it is shown that the data from different sources may differ from one another by 7%. In our opinion, the most trustworthy value of melting enthalpy is given in [9], which was adopted based on the results from generalization of eight experimental works. It should be pointed out that the data on the lead vaporization enthalpy are limited in number and differ from one another by no more than 1%. We recommend to use the value presented in the review [3].

Density, volumetric thermal expansion coefficient, and density variation in melting and crystallization. The tables of standard reference data published in 2007 [10] contain recommendations on the lead density values obtained during experiments on determining density using the gamma-raying method. The main attention in these experiments was paid to carrying out the main measurements with high accuracy and to checking the purity of studied samples before and after the experiment. According to our estimates, the error of the results obtained in [11] for liquid lead is 0.2% at temperatures below 1000 K and 0.3% at higher temperatures. Since the thermal expansion coefficients are usually calculated taking the sample density into account, we also recommend to use the data presented in [10].

The value of density jump in melting is given in a few handbooks and reviews and is equal to 3.6–3.8%. A very careful review of experimental data presented in [11] has shown that the results from direct measurements of density jumps in lead melting "stratify" in two groups. The first group encompasses works with the average value $\langle \Delta\rho_f \rangle_1 = 3.3\%$ with the maximal deviation from it $\Delta_1 = 0.03\%$, and the second group encompasses works with the average value $\langle \Delta\rho_f \rangle_2 = 3.7\%$ with $\Delta_2 = 0.015\%$. We, like the author of the review [11], recommend to use the value $\Delta\rho_f = 3.3\%$. Since a rigorous substantiation in favor of selecting the presenting recommendations is lacking, the data on density jump in melting need to be determined more exactly by experiment.

Saturated vapor pressure. It is shown in the review [3] that the data on saturated vapor pressure given in different sources are in satisfactory agreement with each other and differ from the recommendations suggested in [3] by no more than 15%. In view of the fact that the pressure of saturated lead vapor in the interval from the



Data on the liquid lead thermal conductivity. (*1*) [15], (*2*) [3], (*3*) Wiedemann–Franz law, (*4*) [17], and (*5*) [18].

melting temperature to the boiling temperature varies by 13 orders of magnitude, mismatches by 20–30% seem to be insignificant. In our opinion, the most reliable recommendations are given in the handbook [12], in which the experimental works that had been carried out up to 1961 are scrutinized, the measurement procedures and their errors are analyzed, the most reliable experimental data are selected, and the table of recommended values is developed on their basis.

Surface tension. The results obtained from different experimental works aimed at measuring surface tension differ significantly from one another (by around 25%), especially at temperatures close to the lead melting temperature. However, there is a clearly distinguishing large group of works (approximately 20 sources) the data in which differ from one another by no more than 4%. For carrying out the liquid lead surface tension calculations in the temperature range from the melting point to 1300 K, we recommend to use extrapolation of the data presented in [13]. High-purity lead was used in [13] for carrying out measurements, and a set of measures was taken for additionally purifying the studied samples and to prevent them from becoming contaminated during the experiments; modern experimental equipment and computer-aided techniques for processing the results were used; according to the authors' assessments, the measurement error is 0.1%.

Sound velocity. The most reliable data on sound velocity in liquid lead are presented in the works [7, 14]. The review [7] presents, as recommended ones, the results from careful statistical processing of the data from 15 experimental works that had been carried out up to 1982. The experimental investigations described in [14] were carried out with the aim to obtain the most precision results for refining the existing data and to recommend them as standard reference data. The recommendations of works [7, 14] are in good agreement with each other within the total errors, and the temperature range of the presented data is limited to 1000 K.

Heat capacity. The data on liquid lead heat capacity presented in a large number of handbooks are based on an analysis of the same experimental results that had been obtained up to 1980. Nonetheless, their recommendations differ from each other in some cases by 10% or more. The data presented in the handbook [9] seem to be presently the most reliable ones. In our opinion, there is a need to experimentally refine the heat capacity values because modern calorimeters make it possible to measure this parameter in the entire temperature range in which the liquid phase exists with an error not exceeding a few fractions of percent.

Viscosity. The data on liquid lead viscosity presented in handbooks are in good agreement with one another. An analysis of experimental works has shown that the data in the majority of them also coincide within the limits of measurement errors. For viscosity calculations, we recommend to use the dependence borrowed from [3], which fairly well approximates the data of the most reliable experiments.

Electric resistivity. In the review [7], which contains an analysis of the works carried out up to 1982, the electric resistivity is determined with sufficiently high accuracy (the measurement error does not exceed 1%). The correlation recommended in [7], which was obtained by statistically processing the results from 12 experimental works, is sufficiently reliable one.

Thermal conductivity. The data of the majority of experimental works on measuring the liquid lead heat conductivity are in satisfactory agreement with one another near the melting point; however, the mismatch between the results grow significantly with increasing the temperature, reaching in some cases 100%. Selection of the most reliable data seems to be a difficult task. We recommend to use the measurement results presented in [15] for the following reasons: the measurements were carried out using the newest experimental equipment; the possible experimental errors have been carefully analyzed, and measures for eliminating them have been taken based on the results of that analysis; the measurement errors have also been analyzed; and the measurement procedure has been qualified at the RF State Service of Standard Reference Data [16] for being applied to liquid metals and was calibrated on low-melting metals in liquid state. It is also known that the Wiedemann–Franz law makes it possible to calculate the thermal conductivity coefficient of liquid metals from the data about their electrical conductivity with sufficient accuracy (with an error of no more than 10%). As is seen from the figure, the results of presented in [15] are in good agreement with the data of works [17, 18] and with the results of calculations according to the Wiedemann–Franz law, being somewhat higher than the latter. This systematic mismatch toward increasing is attributed to a small influence of the heat conductivity mechanism that differs from the "electronic" one. The data presented in the majority of other experimental works are significantly lower than the values obtained from calculations according to the Wiedemann–Franz law, which gives rise to doubts.

CONCLUSIONS

(1) Based on an analysis of the data on the thermophysical properties of lead available in the published sources, the most reliable of them have been revealed and recommended for use in carrying out engineering and scientific calculations (see the table). The presented value of solidification temperature is the most reliable one. The data on the boiling temperature, melting and vaporization enthalpies, and on saturated vapor pressure have been determined with satisfactory accuracy. The published data on the liquid lead heat capacity differ considerably from one another; therefore, the recommended values need experimental checking and refinement.

(2) The experimental data on surface tension, density, volumetric expansion coefficient, sound velocity, viscosity, and thermal conductivity do not cover the entire range of temperatures in which the liquid phase exists.

(3) The temperature region above 1200 K and the crystal–liquid phase transition region remain the least studied ones.

(4) It is necessary to experimentally investigate the above-listed properties in the indicated temperature ranges. The question about the influence of impurities on the thermophysical properties of lead still remains unanswered and requires experimental investigation.

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