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Thermal Conductivity and Thermal Diffusivity of Liquid Indium –Tin Alloys

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Abstract Thermal-conductivity and thermal-diffusivity coefficients of indium–tin alloys have been determined using the laser flash method over the temperature range from the liquidus line to 1173 K. Measurements were performed using the setup LFA-427 of NETZSCH company in an argon protective atmosphere, and cells were produced from molybdenum. The equations for temperature dependences of the thermal conductivity and thermal diffusivity of In–Sn alloys have been obtained. The results of measurements were compared with data available in the literature.

Keywords Indium–tin alloys · Laser flash method · Melts · Thermal conductivity · Thermal diffusivity

1 Introduction

Liquid metals and their alloys are widely used in different areas of science and technology. The theory does not allow predicting properties with the accuracy required for the practice yet; therefore, experiment remains as the main source of necessary information. Despite a significant number of investigations already carried out, the thermal conductivity of liquid substances and materials is still one of the least studied properties. At present, there are no data on the thermal conductivity of melts that can be considered as standard reference values. Results of measurements overstep

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the limits of total errors and rather frequently have different signs of temperature coefficients. This is connected with the complexity of heat-flux measurements at high temperatures where the heat loss grows considerably, accounting for radiation and convective transfers. In this study, the thermal-conductivity and thermal-diffusivity coefficients of indium–tin alloys have been determined using the laser flash method over the temperature range from the liquidus line to 1173 K.

2 Experimental

Experiments were performed with the automated setup LFA-427 of NETZSCH company, designed for measurements of the thermal diffusivity of solid materials by the laser flash method in the range of temperatures from 20 °C to 2000 °C. A laser impulse with an energy up to 30 J and a duration of 0.3 ms to 1.0 ms was developed by a solid laser of yttrium–aluminum garnet doped by neodymium with a wavelength of 1.064 μm. The temperature change of the sample is registered by an IR-detector (InSb), cooled by liquid nitrogen.

In Fig. 1, a schematic diagram of the measuring cell design developed for the measurement of liquid metal thermal diffusivity and thermal conductivity is shown. Typical thicknesses of the crucible bottom and insert bottom are about 0.5 mm, of the melt layer 1 mm to 2.5 mm, thicknesses of the side walls of crucible and insert are about 0.3 mm, and their external diameters are 12.6 mm and 9 mm, respectively. For the determination of the thermal diffusivity of melts according to the experimental thermograms, it is necessary to develop a mathematical model that serves to calculate the time evolution of the temperature field in the cell.

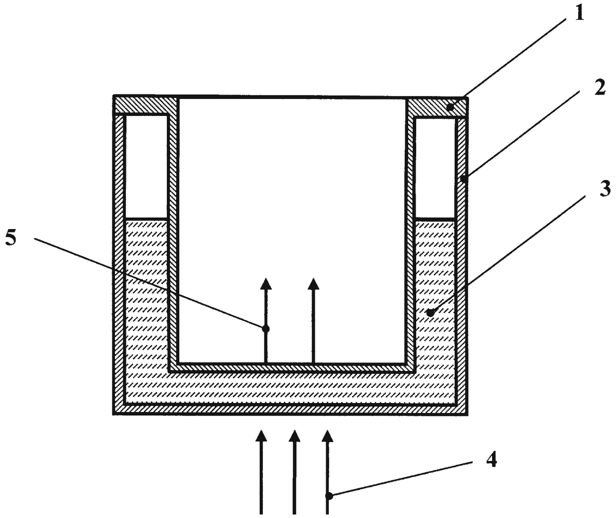

Fig. 1 Schematic diagram of cell for thermal-diffusivity and thermal-conductivity measurements of liquid metals [1]. *1* insert, *2* crucible, *3* melt, *4* laser radiation, *5* radiation on the infrared detector

Assume that the properties of the crucible, insert, and melt material do not depend on the temperature in the range of laser impulse heating; the measuring cell in the initial moment of time has a constant temperature, and the heat loss depends solely on radiation from all the surfaces. Equations of radiation transfer were linearized, since in the real experiment, the cell heating after the laser impulse impact does not exceed several degrees. In these approximations, the equation of the thermal conductivity describing propagation of the heat impulse in the cell takes the form

$$\frac{\partial \theta}{\partial t} = a\left(\frac{\partial^2 \theta}{\partial r^2} + \frac{1}{r}\frac{\partial \theta}{\partial r}\right) + a\frac{\partial^2 \theta}{\partial z^2} \tag{1}$$

where $\theta = T - T_0$, T_0 is the temperature of the sample in kelvin prior to radiation of the crucible by the laser impulse, r is the radius, z is the distance from the lower part of the cell, and a is the thermal-diffusivity coefficient. Equality of the temperatures and heat fluxes were assumed on boundaries of the melt with the crucible and insert. Equation 1 with the initial and boundary conditions was solved numerically. A correction for the resulting duration of the laser impulse and its real shape [2] was introduced. Details are described in [1,3,4].

Processing of experimental data included calculation of the time dependence of the upper surface temperature of the insert cavity normalized on the maximum temperature ($T_{\max}$) and its comparison with the experimental thermogram. For calculation of the specified thermogram, properties of the measuring cell material as well as the density and heat capacity of the studied liquid were considered preset. Adjustment parameters were the thermal diffusivity of the liquid and the total hemispherical emissivity of the boundaries of the cell. The criterion of adjustment completion was the minimum value of the mean square deviation of the calculated and experimental values. One observes that the value of the total hemispherical emissivity, per se, is the effective parameter responsible for all possible heat losses of the sample to the ambient medium (including convective heat transfer) and assumes the same for all boundaries. As an example, in Fig. 2, there is a comparison of the calculated and experimental curves obtained in the experiments with the $In_{80.53}Sn_{19.47}$ alloy. Calculations have demonstrated that concordance of the curves in the area of heating (the area of temperature rise up to its maximum) was provided mostly by the variation of the melt thermal diffusivity, and in the dip area (the section after the maximum) by the total hemispherical emissivity.

Initial metals, indium, tin (99.996 % and 99.999 % purity, respectively), and parts of the measurement cells were preliminarily degassed in a vacuum of 1 mPa at a temperature of 1200 K for (1 to 2) h. Alloy samples ($In_{34.05}Sn_{65.95}$, $In_{80.53}Sn_{19.47}$, and $In_{49.93}Sn_{50.07}$—eutectic) were synthesized by melting indium and tin masses to the required ratios. Thereafter, the melt was stirred thoroughly with a mechanical mixer. Argon with an initial purity of 99.998 vol% was used as a protective atmosphere. Different cells produced from molybdenum were used. The thermal diffusivity of the cell material was measured in additional experiments for the samples of the same material from which the cells were produced. Values of the density and heat capacity of molybdenum and eutectic were taken from reference data [6–8]. The density and heat capacity of $In_{34.05}Sn_{65.95}$ and $In_{80.53}Sn_{19.47}$ were calculated by the additivity rule for an ideal solution. Changes of the melt thickness caused by thermal

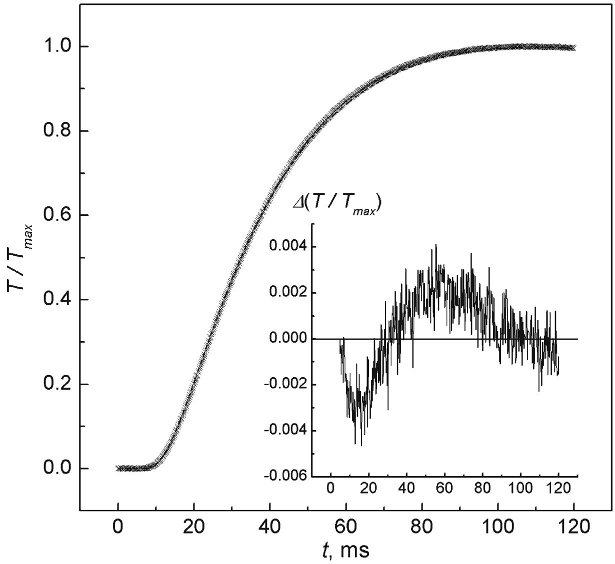

Fig. 2 Comparison of calculated (*line*) and experimental (*crosses*) thermograms for the measuring cell with liquid $In_{80.53}Sn_{19.47}$ alloy at 1173 K. $\Delta(T/T_{\max}) = (T/T_{\max})_{\exp} - (T/T_{\max})_{\mathrm{cal}}$

expansion of molybdenum were taken into account during the treatment. The instrumental error of the LFA-427 setup was determined in experiments with standard solid samples of Inconel and Pyroceram. They showed that the difference from the reference data lies within the limits of 0.5 % to 2.0 %. The error of the measured values of the melts' thermal diffusivity due to the errors of the cell material properties, as well as of the heat capacity, C_P, and the melt density, ρ, may be estimated according to the expression:

$$\delta a^2 = B_{a\mathrm{c}}^2 \delta a_c^2 + B_{\rho c}^2 \delta \rho_c^2 + B_{Cp\mathrm{c}}^2 \delta C_{P\mathrm{c}}^2 + B_\rho^2 \delta \rho^2 + B_{Cp}^2 \delta C_P^2 \tag{2}$$

Here

$$B_{a\mathrm{c}} = \frac{a_c}{a}\left(\frac{da}{da_c}\right), \quad B_{\rho c} = \frac{\rho_c}{a}\left(\frac{da}{d\rho_c}\right), \quad B_{Cp\mathrm{c}} = \frac{C_{P\mathrm{c}}}{a}\left(\frac{da}{dC_{P\mathrm{c}}}\right),$$
$$B_\rho = \frac{\rho}{a}\left(\frac{da}{d\rho}\right), \quad B_{Cp} = \frac{C_P}{a}\left(\frac{da}{dC_P}\right), \tag{3}$$

where a_c, ρ_c, and $C_{P\mathrm{c}}$ are the cell material thermal diffusivity, density, and heat capacity; δa, δC_P, $\delta \rho$, δa_c, $\delta C_{P\mathrm{c}}$, and $\delta \rho_\mathrm{c}$ are the relative errors of the thermal diffusivity, density, and heat capacity of the melt and cell material, respectively. The dominant functions (Eq. 3) were determined numerically. From these data and Eq. 2, the thermal diffusivity error due to the errors of the melt and molybdenum properties is less than 1.5 %, the main portion being contributed by the error of the determination of

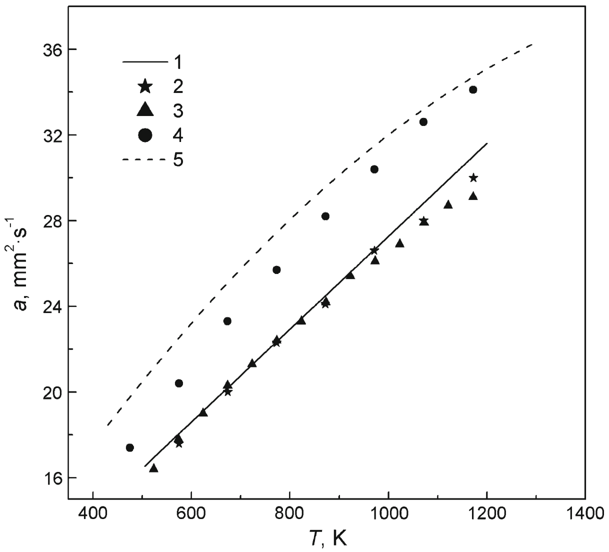

Fig. 3 Experimental thermal diffusivity of alloys: *1* Sn [4], *2* $In_{34.05}Sn_{65.95}$, *3* $In_{49.93}Sn_{50.07}$, *4* $In_{80.53}$ $Sn_{19.47}$, *5* In [3]

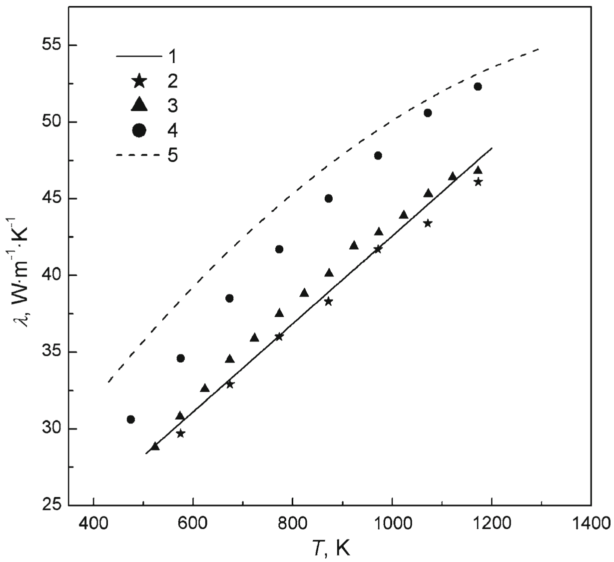

Fig. 4 Experimental thermal conductivity of alloys: *1* Sn [4], *2* $In_{34.05}Sn_{65.95}$, *3* $In_{49.93}Sn_{50.07}$, *4* $In_{80.53}$ $Sn_{19.47}$, *5* In [3]

the measuring cell material thermal diffusivity. It is easy to show that the error of the thermal-conductivity values, in fact, coincides with that of the thermal diffusivity [3,4].

Table 1 Experimental thermal conductivity and thermal diffusivity of liquid alloys

T (K)	a ($mm^2 \cdot s^{-1}$)	λ ($W \cdot m^{-1} \cdot K^{-1}$)
$In_{34.05}Sn_{65.95}$		
574.5	17.6	29.7
673.6	20.0	32.9
773.0	22.3	36.0
872.0	24.1	38.3
971.6	26.6	41.7
1071.6	28.0	43.4
1173.0	30.0	46.1
$In_{80.53}Sn_{19.47}$		
872.8	24.2	40.1
474.6	17.4	30.6
574.5	20.4	34.6
673.5	23.3	38.5
773.0	25.7	41.7
872.1	28.2	45.0
971.8	30.4	47.8
1071.8	32.6	50.6
1172.6	34.1	52.3
$In_{49.93}Sn_{50.07}$		
523.0	16.4	28.8
573.5	17.8	30.8
623.5	19.0	32.6
673.3	20.3	34.5
723.1	21.3	35.9
773.0	22.4	37.5
823.0	23.3	38.8
872.8	24.2	40.1
922.8	25.4	41.9
972.9	26.1	42.8
1022.8	26.9	43.9
1072.6	27.9	45.3
1121.5	28.7	46.4
1172.3	29.1	46.8

The general systematic error of the melts heat transfer coefficients, taking into account all sources, is estimated as 2 % to 3.5 %, depending on overheating above the melting point. Random error has a much smaller value.

The value of the Rayleigh number for our experiments does not exceed 50; this is much less than the critical Rayleigh number (of about 1700); therefore, we can assume that in our experiments free-convective flows do not arise. This is confirmed by the independence of measurement results of the liquid mercury thermal diffusivity from the melt layer thickness [1].

3 Results and Discussion

In Figs. 3, 4 and Table 1, results of the thermal-diffusivity and thermal-conductivity measurements of melts over a wide range of temperatures are presented. As is seen, the heat transfer coefficients of the $In_{34.05}Sn_{65.95}$ and $In_{49.93}Sn_{50.0}$ alloys practically coincide with a and λ of pure tin. The experimental data for the thermal diffusivity and thermal conductivity were approximated by:

$$a(T) = b_0 + b_1 T + b_2 T \tag{4}$$

$$\lambda(T) = c_0 + c_1 T + c_2 T \tag{5}$$

where T is the temperature in K, $a(T)$ is the thermal diffusivity in $\mathrm{mm^2 \cdot s^{-1}}$, and $\lambda(T)$ is the thermal conductivity in $\mathrm{W \cdot m^{-1} \cdot K^{-1}}$. The values of the coefficients b_j and c_j are given in Table 2. The standard deviation of the experimental points from Eqs. 4 and 5 vary from 0.35 % ($In_{80.53}Sn_{19.47}$) to 0.65 % ($In_{34.05}Sn_{65.95}$).

We have found only one publication [5] devoted to measurements of the thermal conductivity of liquid indium–tin alloys. Comparison of our data and [5] are presented in Fig. 5. Differences are less than 3.5 % at temperatures above 500 K. It is apparent that our measurements were carried out over a wider range of temperature.

Isotherms of the thermal conductivity of liquid indium–tin alloys are shown in Fig. 6. At a concentration of indium less than 50 at%, the thermal conductivity of alloys does not depend on the composition and coincides with λ of liquid tin in the limit of estimated errors. A further increase of the indium concentration leads to an increase of the thermal conductivity of the alloys, and the measured values are in good agreement with calculations from the additive rule.

4 Conclusion

A new laser flash technique for measurements of liquid metal heat transfer coefficients has been presented. Experimental data on the thermal conductivity and thermal diffusivity of liquid indium–tin alloys in the range of temperatures from liquidus to

Table 2 Coefficients of Eqs. 4 and 5

Material	b_0	b_1	$b_2 \times 10^5$	c_0	c_1	$c_2 \times 10^5$
In [3]	3.533	0.0394	−1.091	13.06	0.0537	−1.66
Sn [4]	5.592	0.0217	0	13.90	0.0287	0
$In_{34.05}Sn_{65.95}$	1.418	0.0319	−0.647	8.11	0.0427	−0.886
$In_{49.93}Sn_{50.07}$	0.315	0.0360	−0.965	6.30	0.0509	−1.370
$In_{80.53}Sn_{19.47}$	0.120	0.0413	−1.046	6.60	0.0582	−1.630

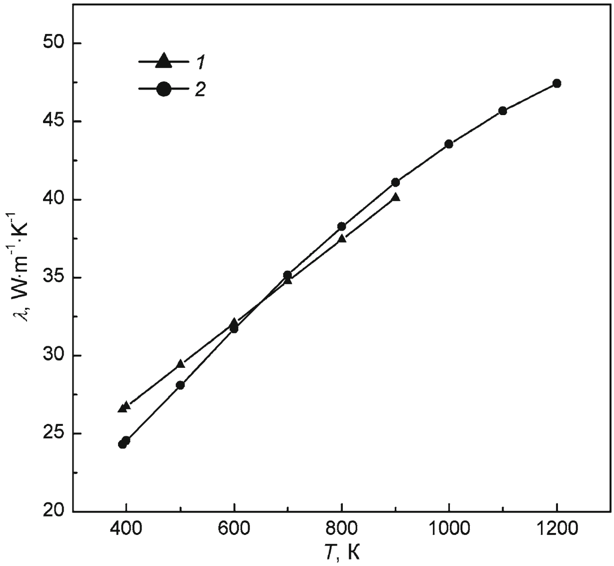

Fig. 5 Comparison of data for the thermal conductivity of a liquid eutectic alloy $In_{49.93}Sn_{50.07}$: *1* [5], *2* this study

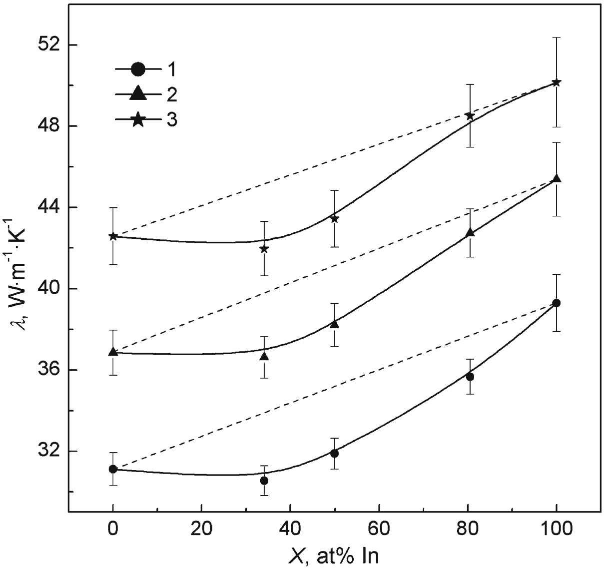

Fig. 6 Thermal conductivity of liquid indium–tin alloys as a function of composition. Points are experimental data, *solid lines* are spline approximation, and *dashed lines* are calculations from an additive rule. *1* 600 K, *2* 800 K, *3* 1000 K

1173 K have been obtained, and their errors have been evaluated. It has been shown that the thermal conductivity of the alloys does not depend on the composition at concentrations of indium less than 50 at%.

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