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Thermal Conductivity of Liquid Tin and Indium[1]

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The present paper reports new measurements of the thermal conductivity of liquid tin and indium. The measurements have been performed at atmospheric pressure in a range of temperatures from 450 to 750 K using a new experimental method based on the principle of the transient hot wire technique. The particular version of the technique employed for molten metals has been shown to have an accuracy in the measurement of the thermal conductivity of molten metals of ±2%. Ultimately, it is intended that the technique operate in a wide range of temperatures, from ambient up to 1200 K, and work is in progress to increase the working temperature and to extend the range of measurements. The results are compared with experimental data reported in the literature by other authors and with predictions of the Wiedemann and Franz law.

KEY WORDS: indium; liquids; metals; thermal conductivity; tin; transient hot wire.

1. INTRODUCTION

In previous papers [1–4] we have emphasized the importance of thermal conductivity data for molten materials in fields such as metallurgy, ceramic engineering, and glass manufacture. Molten metals and salts are also used as heat transfer fluids, for heat storage, and for high-temperature thermometers. Although all these industries and processes require accurate data, there has been a limited number of experimental studies on the thermal conductivity of molten metals [5] and the existing data show discrepancies of as much as 50% [5, 6]. In the present work we apply a new experimental technique to the measurement of the thermal conductivity of molten

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metals; this new method has been successfully applied to the measurement of the thermal conductivity of molten mercury and gallium [1–3] in the range of temperature between 300 and 541 K. Ultimately, the technique is intended to work up to 1200 K, and in this and in previous papers [1–4] the temperature has been slowly increased. The intention is to validate the technique at successively higher temperatures while preserving an accuracy of $\pm 2\%$; in the present work the temperature range is extended to 750 K.

2. EXPERIMENTAL

The present method is based on the transient hot wire technique, and it has been briefly described in previous papers [1–4] and is described in full elsewhere [5, 7]. Because in the present work we study electrically conducting materials, a 99.99% pure platinum wire of 25-μm diameter is symmetrically sandwiched between two thin sheets of 96% pure alumina substrate. The connections to the wire are printed on one sheet of alumina using platinum ink and screen printing technology before making the sandwich.

The assembly is baked at 1600°C, and the resulting rigid rectangular sensor is immersed in the molten metal of interest, which is itself contained in a crucible mounted in a high temperature furnace with a controlled atmosphere. A voltage is then applied to the wire to induce heat dissipation and a consequent temperature rise from the initial equilibrium temperature T_0. The temperature rise is determined, in part, by the thermal conductivity of the molten metal. The measurements of the temperature rise of the wire, which are accomplished by means of a dc Wheatstone bridge configuration, start 20 μs after the heat step initiation and are completed at a time of 1 s. The process is described by a set of partial differential equations and appropriate boundary conditions rather than an approximate analytical solution. Therefore, a two-dimensional finite element (2D-FEM) program has been specially developed for the solution of the working equations [7].

To obtain the thermal conductivity of the molten metal, calculations are made using the 2D-FEM program and the known values of the thermophysical properties of platinum [8] and alumina [9], and the density and heat capacity of tin [8, 10, 11] and indium [12–14], as well as the value of the heat flux, q, used during the experiment. The thermal conductivity of the molten metals is then obtained by matching the measured and calculated temperature rise data by trial and error. Because the different materials (platinum, alumina, melt) influence the temperature rise of the wire at different times, as shown in Fig. 1, the agreement between both temperatures can be achieved by adjusting the thermal conductivity of

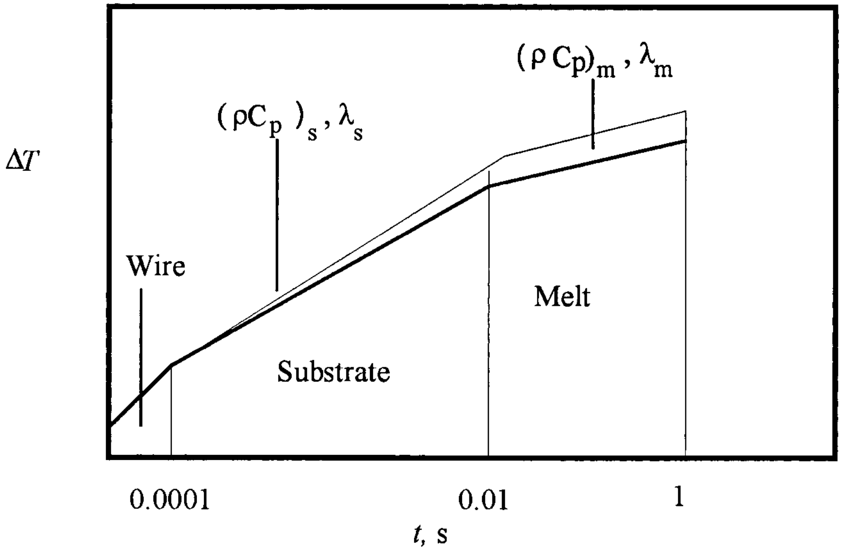

Fig. 1. Schematic representation of the measured and simulated temperature rise as a function of time. The thick line indicates the measured temperature rise of the wire; the thin line indicates the temperature rise calculated theoretically.

alumina for times $t < 10$ ms and the thermal conductivity of the melt for times $t > 10$ ms.

Experiments and simulations were carried out for tin and indium in a range of temperatures from 530 to 730 and from 450 to 743 K, respectively, at atmospheric pressure in a controlled atmosphere of argon (99.999%

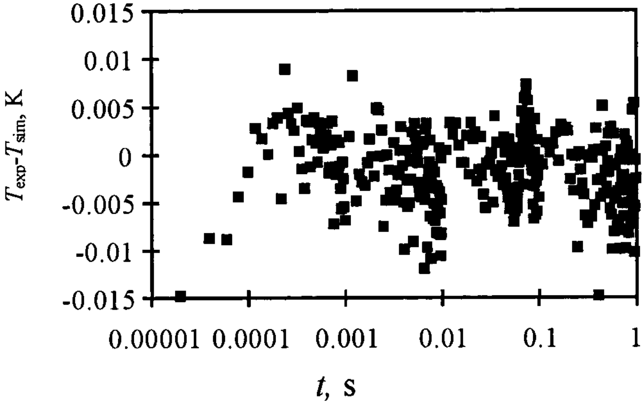

Fig. 2. Comparison between measured and simulated temperature rise data for liquid tin at 603.2 K.

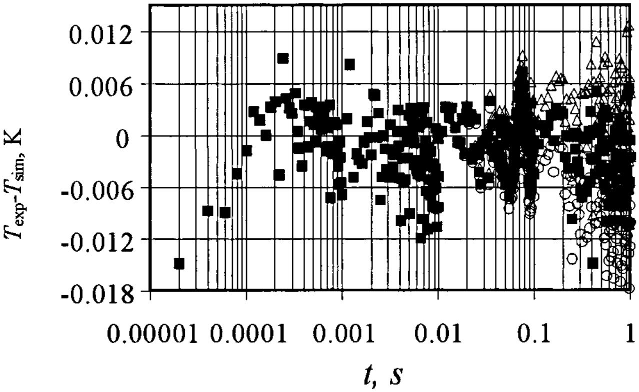

Fig. 3. Sensitivity to the thermal conductivity of liquid tin at 603.2 K: (■) $\lambda_{Sn} = 33.0\ W \cdot m^{-1} \cdot K^{-1}$; (△) $\lambda_{Sn} = 33.33$ (+1%) $W \cdot m^{-1} \cdot K^{-1}$; (○) $\lambda_{Sn} = 32.67$ (−1%) $W \cdot m^{-1} \cdot K^{-1}$.

purity) to avoid any problem with oxidation. To examine the extent to which the theory of the method describes the experiment, we plot the differences between measured and simulated temperature rise data, as shown in Fig. 2, for tin at 603.2 K. It can be seen in this example that the deviation of the experimental temperature rise from that calculated with the optimal physical properties is seldom more than 0.1%, given that the total temperature rise is about 5 K. To demonstrate the sensitivity of the technique, we change the optimal thermal conductivity of the melt by ±1%; the effect of this small change is clearly discernible, as an example for tin, given in Fig. 3, demonstrates.

3. RESULTS

Figure 4 illustrates the dependence of the thermal conductivity on temperature for liquid tin and indium. The error bars denote the estimated error of ±2%. Within this band the thermal conductivity has been represented by the following equations:

$$\lambda_{Sn} = -10.204 + 32.063(T/273.15) - 5.686(T/273.15)^2 \tag{1}$$

for $530 < T < 730$ K and

$$\lambda_{In} = -1.805 + 29.116(T/273.15) - 4.030(T/273.15)^2 \tag{2}$$

for $450 < T < 750$ K.

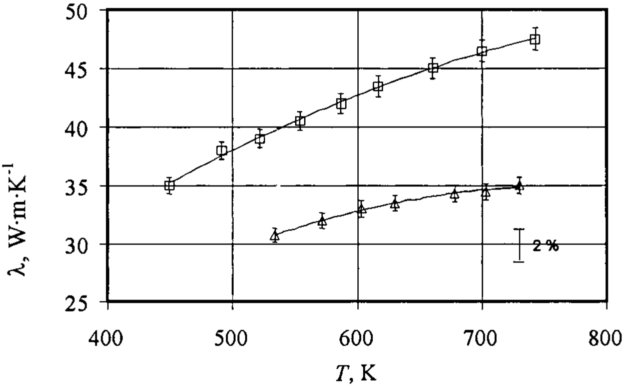

Fig. 4. Thermal conductivity of liquid tin (△) and indium (□) as a function of temperature.

4. DISCUSSION

Figure 5 compares the present correlation for tin with the data measured in earlier studies. Here the good agreement with the recommended values by Ho et al. [15] can be seen. The differences amount to no more than ±3%. The deviations from the results of other authors are often as much as ±10%.

Figure 6 shows a comparison of the correlation of the present results for indium with data reported in the literature. Here we can discern that

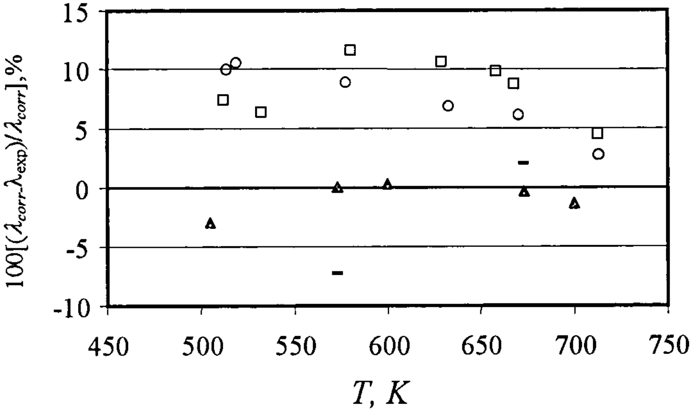

Fig. 5. Comparison of the correlation of the present thermal conductivity data for tin with the data from other authors: (▲) Ho et al. [15]; (○) Hemminger [16]; (□) Hemminger [16]; (——) Pashaev [17].

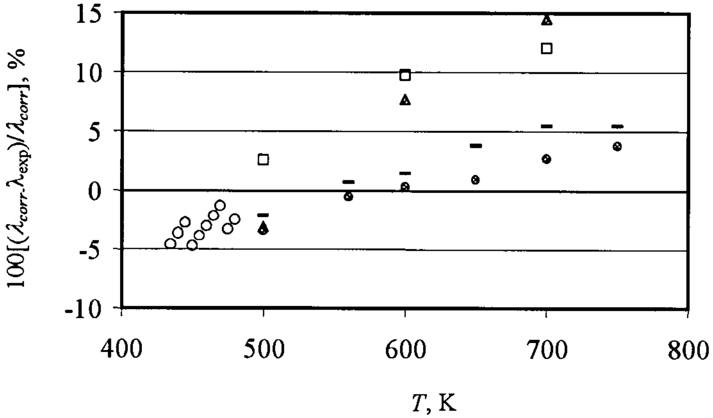

Fig. 6. Comparison of results of earlier measurements of liquid indium with the correlation of the present work: (●) Ho et al. [19]; (——) Yurchak and Smirnov [20]; (▲) Duggin [21]; (○) Goldratt and Greenfield [18]; (□) Touloukian et al. [22].

the results of the present work broadly support the data reported by Goldratt and Greenfield [18]; the deviations are within ±5%. Similar deviations are observed for the recommended values of Ho et al. [19] and Yurchak and Smirnov [20]. Larger deviations are observed for the data reported by Duggin [21] and the recommended values given by Touloukian et al. [22], where the deviations rise from about ±5% at 500 K to 15% at 700 K.

The present experimental data are thought to be of a higher accuracy than for data reported earlier, and, in this sense, Figs. 5 and 6 illustrate the lack of accurate data and the rather few experimental studies for the thermal conductivity of liquid tin and indium. Generally, the thermal conductivity of molten metal is estimated from the electrical resistivity, which is significantly easier to measure, by means of the Wiedemann and Franz relationship based on an electron gas theory of conduction in solids [23]:

$$L_0 = \frac{\lambda_e \rho_e}{T} \tag{3}$$

Here λ_e is the thermal conductivity, attributable to free electrons, ρ_e is the electrical resistivity, and L_0 is the Lorenz number, $L_0 = 2.445 \times 10^{-8}\ \mathrm{V}^2 \cdot \mathrm{K}^{-2}$.

It is important to note that this relationship was originally developed for solid-state metals and applied to an ideal gas of electrons within the metal. This relationship therefore applies only to the thermal conduction brought about by free electrons, and not to the total thermal conductivity, λ. It therefore neglects any other contribution to thermal conduction from phonons. We evaluate here a value of L_{T} for the total thermal conductivity, λ, from the relationship

$$L_{\mathrm{T}}=\frac{\lambda_{\mathrm{T}}\rho_{\mathrm{e}}}{T} \tag{4}$$

using values reported in the literature for the electrical resistivity of liquid tin [11, 24, 25] and indium [11, 25]. If the Wiedemann–Franz law were valid and there were no contributions to the total thermal conductivity except the electronic one, this calculation would yield the Lorenz number L_0. The results obtained are shown in Fig. 7, where the thick line represents the Lorenz number L_0. Figure 8 shows the deviations of L_{T} from L_0.

It can be seen in Figs. 7 and 8 that, for indium, the L_{T} is broadly constant, with the deviations from L_0 about $+5$ to 6%. For the case of tin, we can observe that the L_{T} tends to decrease as the temperature increases. The fact that the experimental values of L_{T} exceed L_0 probably indicates that the single-electron gas theory neglects any other contribution to thermal conduction. In the case of liquid metals, rather than the solid state for which the theory was originally derived, it might be argued that the proximity of the experimental and theoretical results may arise from a cancellation of errors. The accuracy with which it is now possible to measure the thermal conductivity of molten metals suggests that the theory of the transport properties of such systems might be revised since there have been almost no developments beyond the Wiedemann–Franz law.

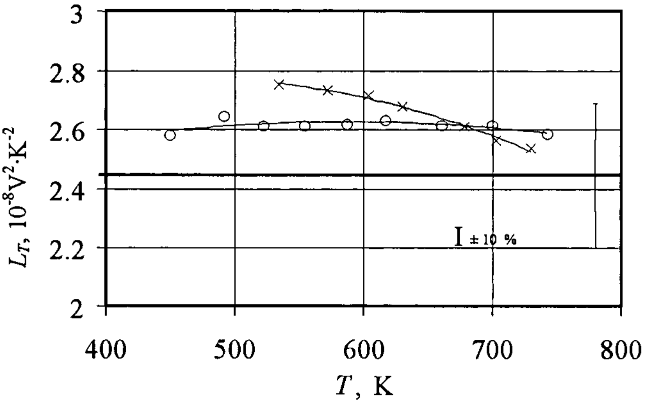

Fig. 7. Dependence of the measured L_{T} with temperature for (○) indium and (×) tin.

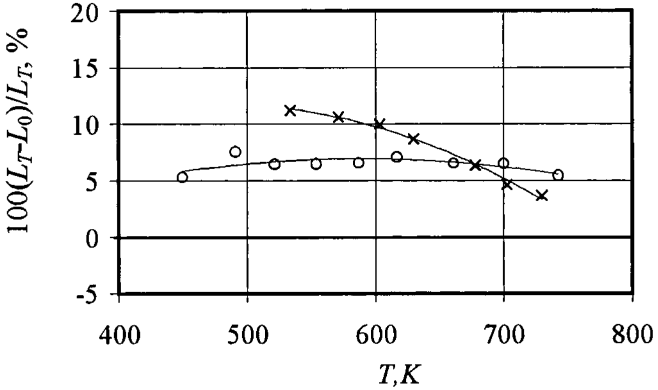

Fig. 8. Deviation of the measured L_T from L_0 for (○) indium and (×) tin.

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