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Measurements of the Thermal Conductivity of Molten Lead Using a New Transient Hot-Wire Sensor

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The paper reports new measurements of the thermal conductivity of molten lead at temperatures from 600 to 750 K. The measurements have been carried out with an updated version of a modified transient hot-wire (THW) method, where the hot-wire sensor is embedded within an insulating substrate with a planar geometry. However, unlike previous sensors of the same type, the updated sensor works with the hot-wire divided into three thermally isolated parts. The operation of this sensor has been modeled theoretically using a finite-element (FE) analysis and has subsequently been confirmed by direct observation. The new sensor is demonstrated to have a higher sensitivity and a better signal-to-noise ratio than earlier sensors. Molten lead is used as the test fluid. It has the lowest thermal conductivity of any material we have yet studied. This allows us to probe the limits of our sensor system for the thermal conductivity of high-temperature melts. It is estimated that the uncertainty of the measurements is 3% over the temperature range studied. The results are used to examine the application of the Wiedemann–Franz (W-F) relationship.

KEY WORDS: molten lead; sensor design; thermal conductivity; transient hot-wire.

1. MODIFIED TRANSIENT HOT-WIRE TECHNIQUE

The modified transient hot-wire technique (THW) used in this work was first introduced in 1997 [1]. The method has been further refined [2] and used for measurements of the thermal conductivity of liquid mercury,

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gallium, indium, and tin [3,4]. In principle, the method consists of two main steps. First the temperature rise of the hot-wire, which is encapsulated in the ceramic substrate and immersed in the measured liquid, is observed over a period of 1 s after the initiation of a precisely defined heat flux within it. In the second step, the measured transient temperature response is compared to the temperature rise of the hot-wire obtained from a two-dimensional finite-element (FE) model of the measurement. The nature of the temperature rise of the hot-wire allows accurate measurements of the thermal conductivity of the materials surrounding the hot-wire, and a FE implementation of a solution of the heat transfer equations for the sensor/fluid complex is employed to deduce the thermal conductivity of the fluid and the sensor *in situ*. A detailed description of the method, its background theory and an analysis of the temperature response of the hot-wire have been published elsewhere [2–4]. These publications also discuss in detail the importance of radiation and natural convection for this kind of THW technique. In general terms, radiative heat transfer is unimportant relative to conduction because a very small temperature difference is applied in circumstances where there is a very large temperature gradient; natural convection is rendered unimportant by virtue of the timescale of the experiment.

We have significantly upgraded both aspects of the technique. A new enhanced FE model has been introduced and represents the true geometry of the THW sensor, unlike the old model which employed a rectangular cross section of the hot-wire. Moreover, the new model is capable of providing a more accurate solution because it uses more sophisticated meshing techniques, which results in a finer mesh in areas where a greater temperature gradient occurs and therefore the solution is more accurate. Another significant improvement is introduction of a new time-stepping methodology in the transient analysis. The sudden changes in time-stepping employed earlier have been removed and the FE model becomes even more accurate. The detailed features of the upgraded FE model, the fundamental mathematical equation as well as the validation of the FE model, have been presented elsewhere [5–9].

The experimental part of the modified THW technique has also been considerably improved by the introduction of a new design of the THW instrument. Additionally, the measurement of the temperature rise of the hot-wire has undergone minor but important improvements [9], which have helped to suppress the phenomenon of natural convection within the measured liquid. This paper further describes and discusses the most significant improvements in the design of the THW instrument.

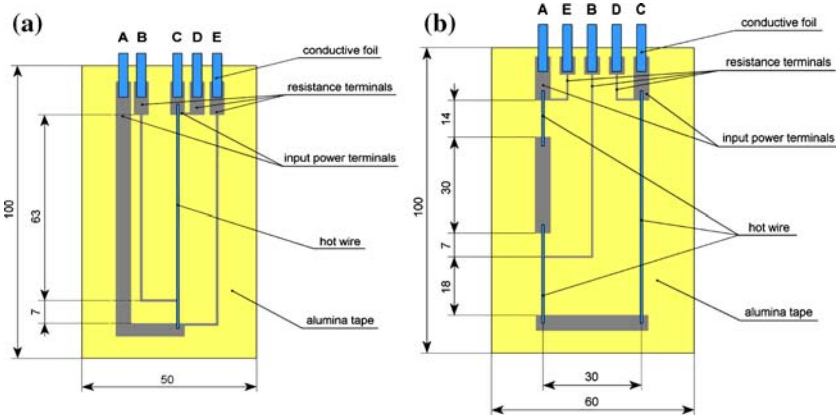

Fig. 1. Original and new design of the THW sensor (dimensions in mm).

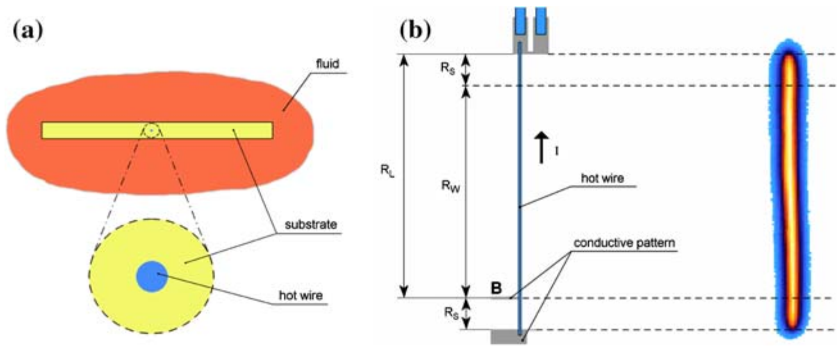

Fig. 2. (a) Cross section of the sensor of original design and (b) principle of the end-effect correction.

2. NEW SENSOR DESIGN

The original design of the sensor, which was first introduced about ten years ago [1], is illustrated in Fig. 1a. The resistance and input power terminals are printed using thick film technology onto a flexible alumina tape and a 25 μm bare platinum wire (hot-wire) and platinum conductive foils (thickness 25 μm) are positioned as shown. During the fabrication process, another alumina sheet is placed on the top and the whole 'sandwich' is hot-pressed and sintered in order to produce a rigid and robust sensor, which withstands a highly chemically aggressive environment over a wide range of temperatures. A cross section of the sintered sensor is shown in Fig. 2a.

During the transient measurements with the sensor of the original design, a step voltage is applied between terminals A and C and a single platinum wire originates a heat wave, which penetrates through the alumina substrate and later also reaches the measured liquid. The hot-wire is electrically divided into two parts, 63 and 7 mm long, which allows the automatic end-effect correction. The principle of the correction is shown in Fig. 2b together with an image acquired by an IR camera after 1 s from application of the heat flux to the hot-wire. The arrangement provides a finite part of an infinitely long hot-wire that is 56 mm long, and its resistance change can be then evaluated as

$$\Delta R_{\mathrm{w}} = \Delta R_{\mathrm{L}} - \Delta R_{\mathrm{S}} \tag{1}$$

We have introduced a new design, which uses the same materials and the same number of terminals and therefore does not require any further adjustments of the measuring devices. The new design, which is illustrated in Fig. 1b, also results in a sensor of similar dimensions and uses less platinum thick film ink for the terminals. However, the most significant advantage of the new design is an increased length of the finite part of the infinitely long hot-wire.

The hot-wire is divided into three thermally isolated parts as is illustrated in Fig. 3. The end-effect correction is again automatically applied, and the finite part of an infinitely long hot-wire is extended to 66 mm, i.e., an increase of 18% compared to the original design. The resistance change of the finite part is calculated as

$$\begin{aligned} \Delta R_{\mathrm{w}} &= \Delta R_{\mathrm{w1}} + \Delta R_{\mathrm{w2}} = 3 \cdot \Delta R_{\mathrm{s}} + \Delta R_{\mathrm{w1}} + \Delta R_{\mathrm{w2}} + \Delta R_{\mathrm{p}} - 3 \cdot \Delta R_{\mathrm{s}} - \Delta R_{\mathrm{p}} \\ &= \Delta R_{\mathrm{long}} - \Delta R_{\mathrm{short}} \end{aligned} \tag{2}$$

The longer hot-wire allows more accurate absolute measurements of the resistance changes, because the signal-to-noise ratio is improved by about 18%. From another point of view, the longer hot-wire allows values of the applied heat flux to be 18% smaller; hence, the temperature increase of the measured fluid near the hot-wire is smaller and the natural convection of the melt, which is driven by a buoyancy force, is minimized.

This new design has been tested in a number of experiments with molten indium, tin, and various solder alloys and the results compared to the values obtained from measurements of the thermal conductivity using the original design [7–9]. Both the new and old sensor designs provided similar results with deviations of no more than 2%, which is less than the mutual uncertainty.

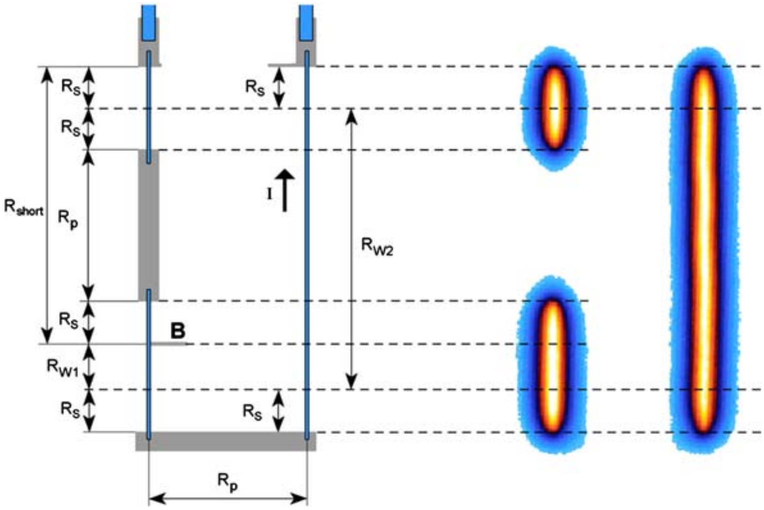

Fig. 3. Cancellation of the end effects in the new design.

3. THERMAL CONDUCTIVITY OF MOLTEN LEAD

Two newly designed sensors have been used to measure the thermal conductivity of pure molten lead kept in a cylindrical alumina crucible with an inner diameter of 60 mm and a height of 100 mm. In order to suppress natural convection, liquid lead at the top of the crucible has been kept at a higher temperature than at the bottom and the difference between these two temperatures has been typically between 3 and 5 K. The measurements have been carried out from the melting point ($T_{m,Pb} = 600$ K) to the maximum application temperature of the current instrument, which was found to be about 750 K. If it is required to use the instrument at higher temperatures, some of the materials used in the fabrication process must be replaced by alternatives with improved high-temperature performance.

As mentioned above, the extraction of the thermal conductivity of the metal from the raw measurements is based on comparisons between the measured data and the results from the FE model. The experimentally measured temperature responses have been acquired using various energy pulses, typically between 45 and 80 $W \cdot m^{-1}$. The range of energy input serves to verify further the FE model, because it has not been found necessary to change the parameters of the model over the whole range of input energy and the thermal conductivity of the lead extracted from the results remained constant. Figure 4 shows a typical absolute comparison

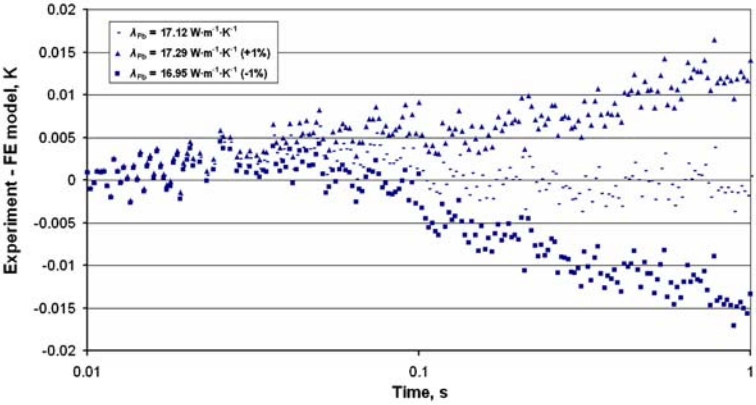

Fig. 4. Sensitivity of the measurements of the thermal conductivity of the molten lead (new sensor at 682 K, applied heat flux of 79 $W \cdot m^{-1}$).

Table I. Thermal Conductivity of Molten Lead as a Function of Temperature

T (K)	T (°C)	λ_{Pb} ($W \cdot m^{-1} \cdot K^{-1}$)
620.3	347.1	16.2
678.1	404.9	17.0
729.5	456.3	17.6

between the experimentally acquired temperature rise of the hot-wire (heat input of 79 $W \cdot m^{-1}$) and results obtained from the FE model. It can be seen that the absolute differences are well within ±0.01 K while the temperature rise itself is about 6.5 K. The ability to distinguish such small temperature differences between the experiment and the FE model is a key feature of our method. It can be seen in Fig. 4 that an assumed change of 1% in the thermal conductivity of molten lead in the simulation of the experiment gives rise to changes in the deviation plot that can be readily discerned.

Table I lists the average values of the thermal conductivity obtained from the two newly designed sensors at specific temperatures, and Fig. 5 graphically illustrates the data from the table.

A linear trend line illustrated in Fig. 5 is used to characterize the rise of the measured thermal conductivity of molten lead with temperature and its equation, valid for T from 600 to 750 K, can be written as

$$\lambda_{Pb} = 0.0128(T - T_{m,Pb}) + 15.95 \quad W \cdot m^{-1} \cdot K^{-1} \tag{3}$$

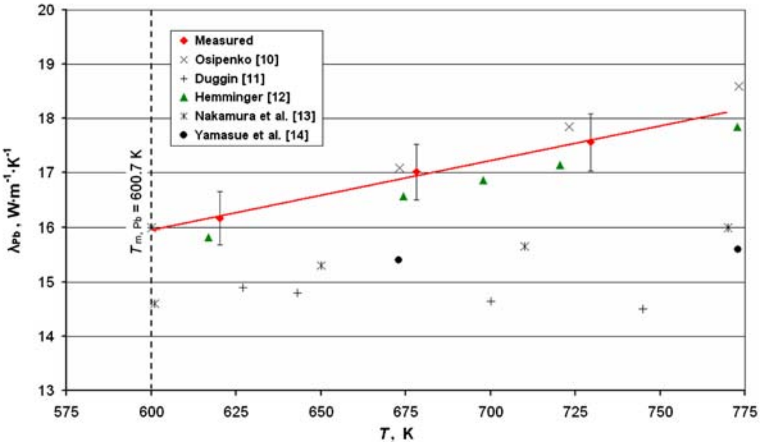

Fig. 5. Thermal conductivity of molten lead.

The measured values of the thermal conductivity are shown with error bars having a range of ±3%. Whereas we have shown that the sensitivity of the method can be less than ±1%, there are other uncertainties that arise. Indeed, the repeatability of the measurements is nearer 2%. The influence of other material properties, which need to be entered into the FE model, has been also investigated [9] and evaluated to be a maximum of 1.5%. Therefore, the overall uncertainty of the method has been estimated to be 3%.

Figure 5 also illustrates the thermal conductivity of molten lead measured by Osipenko [10], Duggin [11], Hemminger [12], Nakamura et al. [13], and Yamasue et al. [14]. The newly measured values are in very good agreement with the thermal conductivity data published by Hemminger and Osipenko who both used steady-state methods and stated uncertainties of 3 and 5%, respectively. Values published by Yamasue et al., who employed a THW instrument with a coated platinum wire, are much lower, but the standard deviation of their experimental values was 1.2 W·m^{-1}·K^{-1}, i.e., about 8%. Their method suffered many potential uncertainties such as non-symmetrical coating of the hot-wire as well as the fact that the materials properties of the coating were unknown and not measured *in situ*. Nakamura et al. also used an instrument based on the THW method, but they applied simplified analytical equations to an instrument with a complex geometry, which must greatly compromise the claimed accuracy. And finally the data published by Duggin differ significantly from all other sources, because his results suggest that the thermal conductivity of molten lead should be decreasing with increasing temperature rather than increasing. The values of Duggin also show a very large scatter (almost ±8%) of the thermal conductivity when examined in detail (Fig. 5 presents only the averaged values.), so that they cannot be considered of high accuracy.

It is the conclusion of this analysis that the results of the present measurements of the thermal conductivity of lead enjoy a high level of confidence and support the earlier work of Hemminger and Osipenko rather than the more recent measurements.

The measured thermal conductivity has been compared to the thermal conductivity calculated from the Wiedemann–Franz (W-F) relationship:

$$\lambda = \frac{T L_0}{\rho_e} \tag{4}$$

where the constant $L_0 = 2.445 \times 10^{-8} \cdot \mathrm{V}^{-2} \cdot \mathrm{K}^{-2}$ is the Lorentz number and ρ_e is the electrical resistivity of the molten metal at temperature T (in K). The data of the electrical resistivity of pure molten lead published by Monaghan [15] have been used, and the thermal conductivity calculated from the W-F law is shown in Fig. 6 together with recommended values published by Touloukian et al. [16], Ho et al. [17], and Mills et al. [18]. It can be seen that the values calculated from the W-F law and those published by Ho et al. and Touloukian et al. are in very good agreement (maximum deviation of ±3%) with values of the measured thermal conductivity reported here using a newly designed THW instrument. The values recommended by Mills et al. more or less follow the values measured by Nakamura et al., because Mills et al. believed that the thermal conductivity measurements carried out by Hemminger and Osipenko (steady-state techniques) were significantly affected by convection within the measured sample and chose to give them little weight.

4. CONCLUSION

The new design of the THW sensor has been presented and compared to the original design. The availability of the two different sensor designs, which use the same methodology to derive the thermal conductivity of molten metals, has significantly strengthened our belief in the validity of the method itself. Moreover, the newly designed sensor allows more precise measurements of the thermal conductivity, because it employs a longer hot-wire.

The new sensors have been used for measurements of the thermal conductivity of pure molten lead up to 750 K, and the linear relationship between the thermal conductivity and the temperature has been defined.

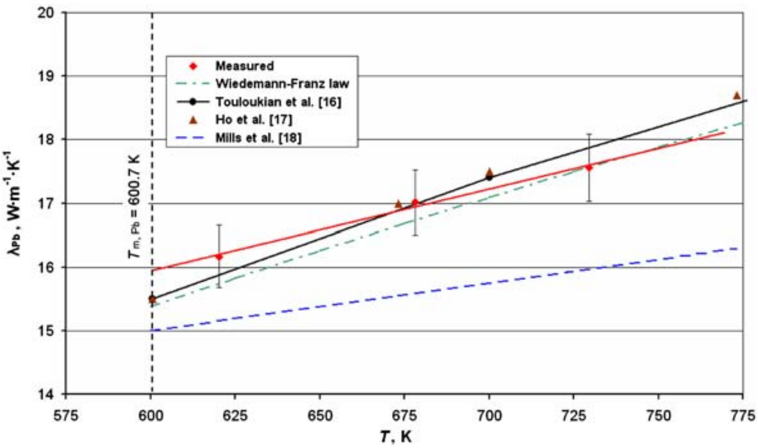

Fig. 6. Measured thermal conductivity of molten lead in comparison with recommended values.

The results are in a very good agreement with the W-F law as well as with data published by several other researchers. This suggests that the W-F law can be used for prediction of the thermal properties of molten lead up to 750 K with an uncertainty of about 3%.

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