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Repeatability and Refinement of a Transient Hot-Wire Instrument for Measuring the Thermal Conductivity of High-Temperature Melts[1]

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The paper reports an assessment of the repeatability of a method for the measurement of the thermal conductivity of high temperature melts. The main goal is to demonstrate that a novel approach to the transient hot-wire technique can yield highly accurate results that are consistent with previous, independent measurements. The paper summarizes the modified transient hot-wire method, presents improvements in the finite-element analysis of its operation, and briefly discusses deviations from available analytical equations. The transient hot-wire instrument and experimental configuration are also described. Results from measurements on molten metals, in particular, tin and indium, in the temperature range from their melting points up to 750 K are presented. A comparison with previously measured values is given, and the accuracy and repeatability of the method are discussed.

KEY WORDS: finite-element method (FEM); indium; molten metal; thermal conductivity; tin; transient hot wire.

1. INTRODUCTION

Measurements of the thermal conductivity of molten metals or other aggressive substances at high temperature present one of the most challenging tasks for thermophysics. The acquisition of accurate data from methods of measurement that have a complete theory and that have been

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shown to conform to it remains the objective of the highest quality work. At the end of the last century, a modified transient hot-wire method was introduced where the thermal conductivity was derived from measurement of the transient temperature increase in the wire using a finite-element analysis of the experiment. The hot-wire instrument described here was first introduced in that period as a result of a European Union collaborative project [1] and, subsequently, the thermal conductivity of several molten metals was measured [2]. The current work continues to improve the transient hot-wire instrument and the method itself and aims to provide highly repeatable and accurate results in order to prove the robustness of the method.

When the classic transient hot-wire method was introduced, considerable efforts were made to ensure that the experiment was designed so that an analytical solution to the energy equation for its circumstances, combined with a number of small corrections could be used to determine the thermal conductivity of a test material from the measured data [3]. The energy equation appropriate to all transient experimental methods for the measurement of the thermal conductivity of an isotropic fluid which has a temperature independent thermal conductivity, density, and heat capacity over small temperature ranges, can be written as [3,4]

$$\rho C_P \frac{\partial T}{\partial t} = \lambda \nabla^2 T + Q \tag{1}$$

where ρ is the density (kg·m^{-3}), C_P is the specific heat capacity (J·kg^{-1}·K^{-1}), T is the temperature (K), t is the time (s), λ is the thermal conductivity (W·m^{-1}·K^{-1}), and Q is the heat generation (W·m^{-3}).

For the simplest form of the transient hot-wire technique, a wire is suspended in direct contact with the material whose thermal conductivity is to be measured. In this case the problem of transient heating of the wire has radial symmetry, and it is possible to design instruments to make use of an analytical solution of Eq. (1) for the extraction of the thermal conductivity of the material from measurements of the temperature rise of the wire as a function of time [3]. For materials that are electrically conductive and/or aggressive, the hot wire must be encased in an insulating material and, in general, this makes the analytical solution of Eq. (1) impossible. As a consequence, numerical means of solving Eq. (1) have been adopted, particularly for applications to molten metals [5].

In this paper, we first describe the means by which we have improved the numerical description of the transient hot-wire method for application to molten metals. Subsequently, we apply this new methodology to measurements of the thermal conductivity of tin and indium. The recent measurements have been conducted with a variety of new sensors, which differ from those employed in earlier work since our intention is to examine the repeatability of our experimental technique.

2. IMPROVING THE NUMERICAL APPROACH

The finite-element method (FEM) was originally selected as the numerical means to solve Eq. (1) for applications of the transient hot-wire method to molten metals [5]. However, the numerical method was developed and tested for the case of a circular section wire immersed in an infinite material because, for that case, we have the analytical solution discussed above. In practical applications the circular wire was originally approximated by a rectangular section wire, and a relatively coarse spatial grid was employed for integration of the equations. Furthermore, the numerical analysis of the response to the imposition of a stepwise heat flux in the wire employed a linear time-stepping algorithm. At the time the methodology proved sufficiently accurate that it was possible to use it for measurements of the thermal conductivity for molten metals [5] with an uncertainty of less than 5%.

Subsequently, however, it has become possible to improve the spatial integration of the energy equation using a circular cross-section to model the wire and a finer spatial grid over the entire space. This new system has been tested on the radially symmetric hot-wire problem in a time range where the corrections to the analytical solution are small. Even under these conditions it is still possible to discern [6] small periodic discrepancies between the analytical solution of the transient hot-wire problem and the solution obtained numerically as is illustrated in Fig. 1.

The periodic discrepancies that are shown in Fig. 1 arise when linearly spaced time steps are employed in the temporal integration of Eq. (1). The discrepancies are not large because the overall temperature rise of the wire is a few degrees Kelvin. However, the effect is particularly important in the problem of interest because the period of the transient hot-wire experiment extends over five orders of magnitude of time. We have therefore now adopted the so-called 'log-spaced' time stepping for integration of the energy equation. Deviations of the numerical solution of the energy equation for the hot wire using this new algorithm from the same analytical solution are included in Fig. 1, and it can be seen that this new choice eliminates the 'periodic' effect, and gives a more accurate solution within every decade. We have used the new algorithm to analyse all of the experimental measurements reported in this paper. The new method gives a more precise representation of the real behaviour of the transient hot wire in the system employed to measure the thermal conductivity of molten metals than that employed earlier, but we have verified that the

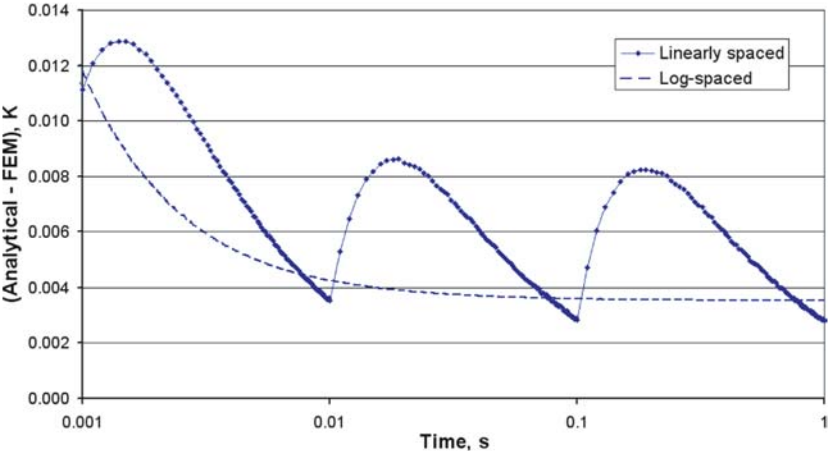

Fig. 1. Comparison between solutions with different time steps spacing (one hundred substeps per decade), average temperature of the wire is shown.

periodic nature of the earlier discrepancies was such as to not disturb the reported thermal-conductivity values in our earlier work by a significant amount.

3. EXPERIMENTAL SETUP

Figure 2 shows the design of the transient hot-wire instrument that we have employed in the present work, and Fig. 3 shows a cross-sectional view of the sensor after the pressing and curing stages. The ‘hot’ platinum wire is encapsulated in the substrate, which protects the wire from chemically aggressive molten compounds and also electrically isolates the whole sensor from the melt. The sensor design and fabrication process have been described in detail in previous studies [5], but it has undergone several modifications to yield altered characteristics of the substrate. The main difference is the peak temperature of the curing profile for the alumina substrate, where, instead of a temperature of 1850 K, a temperature of 1700 K was applied. This change of process resulted in a solid substrate with slightly modified thermal properties. This change has allowed us to confirm that our description of the experimental technique is correct because the technique allows us to derive the material properties of the substrate as well as the melt from a single experiment. We shall show that we are able to recover the same value of the thermal conductivity of the molten material from measurements with two sensors which employ substrates with different thermal conductivities.

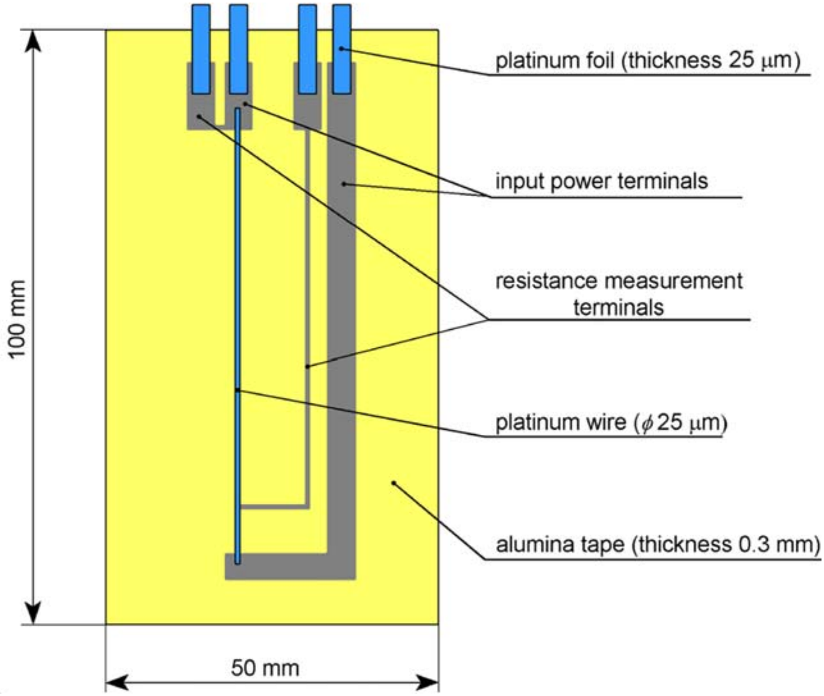

Fig. 2. Sensor design and configuration of terminals before hot-pressing.

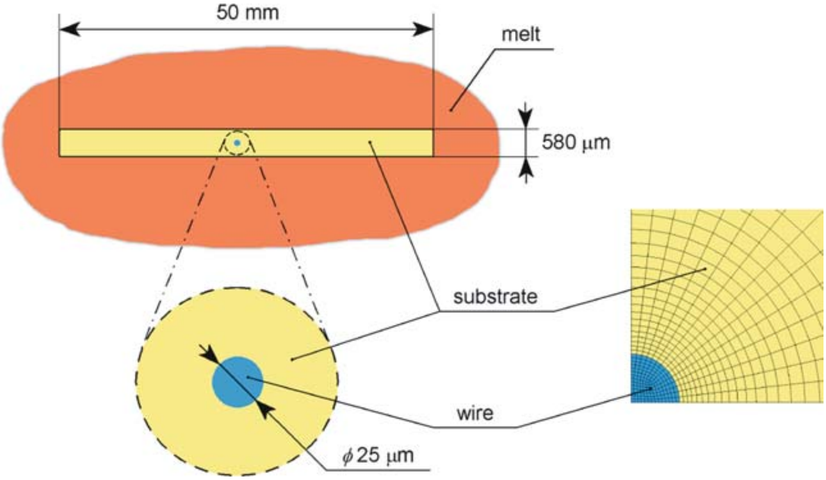

Fig. 3. Cross-section of the sensor and its FE-meshed 2D model.

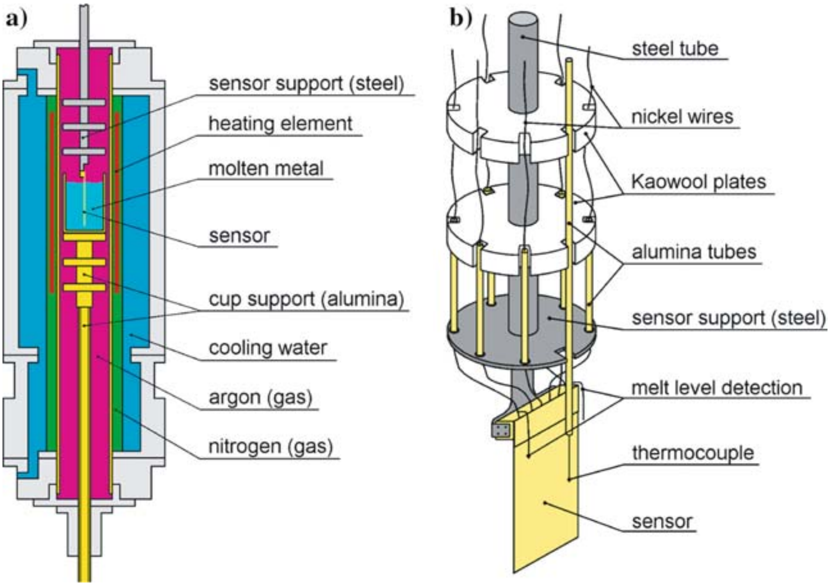

Fig. 4. (a) Position of the sensor inside the furnace and (b) a detailed view of a sensor mounted to a steel support.

The experimental configuration is shown in Fig. 4. The sensor is mounted on a tubular, steel support, and the platinum contacts are spot-welded to nickel wires that connect to the measurement bridge. The temperature of the melt is measured by a pair of thermocouples positioned at the bottom and top ends of the hot wire. These same thermocouples are used to ensure the temperature in the melt is slightly increasing so as to suppress convective flow. An appropriate temperature distribution is ensured by the use of an additional cartridge heater which is placed inside the steel tube of the sensor support. The sensor is connected into a bridge circuit described in detail elsewhere [5] that enables a known heat input to be applied at time zero and the resistance change of a section of the hot-wire to be determined over a period of 1 s as the wire warms by about 5 K. The resistance change is then converted to a temperature rise of the wire by means of a suitable calibration [5, 7].

4. DERIVATION OF THE THERMAL CONDUCTIVITY

The derivation of the thermal conductivity from the transient temperature response is carried out in an iterative fashion. A numerical (FEM) simulation of the transient temperature rise of the wire for the same heat input as applied in the experiment is conducted for values of the thermal conductivity and the product (ρC_P) of platinum, the substrate, and the melt for the defined geometry of the sensor. The simulated temperature rise is compared with that of the experiment and adjustments made to the thermal properties of the substrate and melt so as to secure agreement between the simulated and experimental temperature rises over the entire time period of the measurement from 100μ s to 1 s. The design of the sensor is such that the properties of the substrate $(\rho C_P, \lambda)$ and those for the melt $(\rho C_P, \lambda)$ affect different parts of the temporal history of the temperature rise so that they can be independently determined.

The typical experimental transient response with illustrated parts of the temperature rise is shown in Fig. 5. As has been explained elsewhere [2,5,8], it is necessary at each interface in the heat transfer system to allow for a temperature discontinuity. This is done within the simulation by allowing for the existence of a thin layer of air at both interfaces. Typically their thicknesses are about 5 nm for the wire-substrate interface and 0.5μ m for the substrate-melt interface. The thickness of the interfaces varies slightly for different sensors and also depends on the properties of the measured fluid. However, once the interface thickness is found to fit the model, it does not change by more than ±3% for all measured temperatures with the same sensor and for all applied heat fluxes. Figure 6 shows a typical comparison between experiment and modelled response after all material parameters in the model were set properly over four orders of magnitude in time. The difference is typically within ±5 mK which equates to about ±0.07% of the total temperature rise. This is very strong evidence that our theoretical and numerical model of the sensor is a very good description of the practical instrument.

We have also been able, in our current work, to investigate the effect of using different heat fluxes upon the thermophysical properties derived from the transient measurement. Obviously, if the model of our experiment is correct, the values derived for the thermal conductivity for a molten metal should not depend upon the heat flux used to measure it. We have performed measurements in which the applied heat inputs vary from 70 to $140\,\mathrm{W{\cdot}m^{-1}}$ and confirmed that the derived thermal conductivity for the fluid (and substrate) remains unchanged. After analysis of possible sources of uncertainty and experiments with several sensors, we have estimated the overall uncertainty of the current technique to be of ±3%.

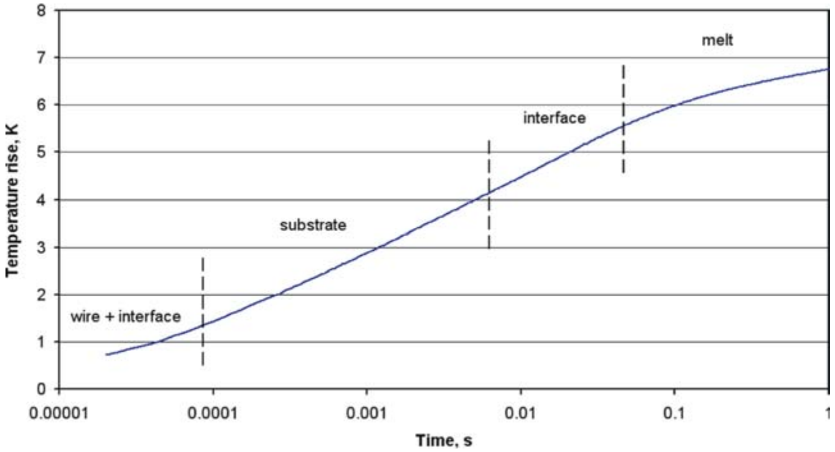

Fig. 5. Typical measured transient response from the sensor (increase of the hot-wire temperature) and the main time regions.

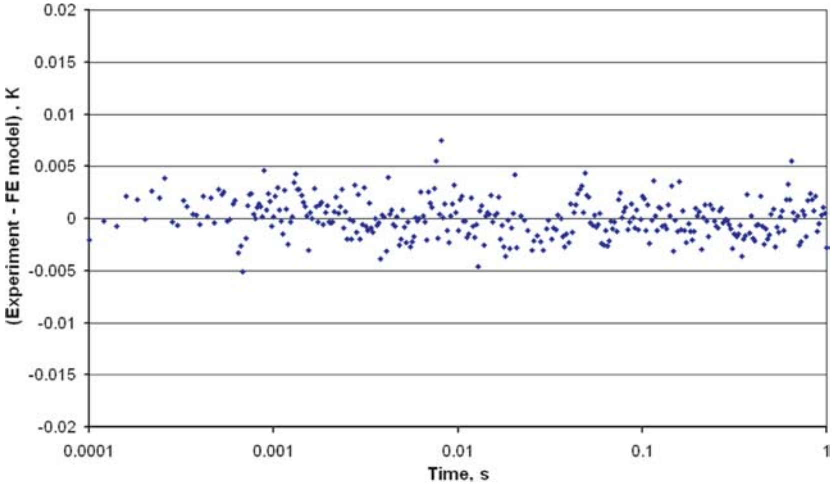

Fig. 6. Example of the comparison between experimental and modelled response for molten indium at 469 K (total temperature rise of the wire is 5.5 K).

5. RESULTS

Two metals with relatively low melting points, indium and tin, were chosen for a comparison with values derived using the same methodology but different sensors and a different numerical procedure. The measured

Table I. Measured Thermal Conductivity of Molten Indium

T (K)	$\lambda_{\mathrm{In}}(\mathrm{W \cdot m^{-1} \cdot K^{-1}})$	Linear Equation
467.0	36.3	
495.4	37.0	
522.2	38.2	$\lambda_{\mathrm{In}} = 0.0345(T - T_{\mathrm{m,In}}) + 35.0\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$
575.9	40.0	
608.8	41.4	equation valid if $T_{\mathrm{m,In}} < T < 750\,\mathrm{K}$
630.2	42.2	
684.5	44.0	$T_{\mathrm{m,In}} = 430\,\mathrm{K}$ (melting point of indium)
711.0	44.4	
734.1	45.3	

Table II. Measured Thermal Conductivity of Molten Tin

T (K)	$\lambda_{\mathrm{Sn}}(\mathrm{W \cdot m^{-1} \cdot K^{-1}})$	Linear Equation
523.1	27.3	
549.2	28.0	
580.0	28.6	$\lambda_{\mathrm{Sn}} = 0.025(T - T_{\mathrm{m,Sn}}) + 26.8\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$
603.7	29.1	
634.9	30.0	equation valid if $T_{\mathrm{m,Sn}} < T < 750\mathrm{K}$
657.0	30.6	
683.8	31.4	$T_{\mathrm{m,Sn}} = 505\,\mathrm{K}$ (melting point of tin)
707.6	31.9	
733.2	32.5	

thermal conductivity for molten indium (purity 99.99%) over a wide range of temperatures is shown in Table I, and the comparison with previous studies and other sources [9–14] is illustrated in Fig. 7. All of the previously measured data [12] fall into the ±5% uncertainty region of the current experiment, and the data are comfortably within ±3% at temperatures close to the melting point. The new method of analysis is felt to be superior, and thus the results reported here are thought to be more accurate than our earlier results because both the measuring technique and equipment have been improved. The comparison in Fig. 7 illustrates that these new data are in excellent agreement with data published by Goldratt and Greenfield [10].

The measured values for the thermal conductivity of molten tin are presented in Table II, and Fig. 8 shows the comparison with previous work using the same method [12] and with other sources (i.e., steady-state

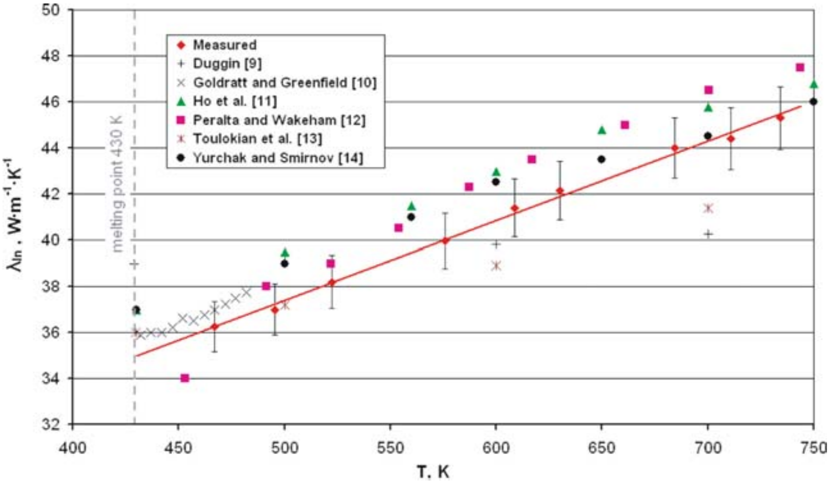

Fig. 7. Thermal conductivity of indium (melting point 430 K) and comparison with some previously measured values (error bars indicate ±3%).

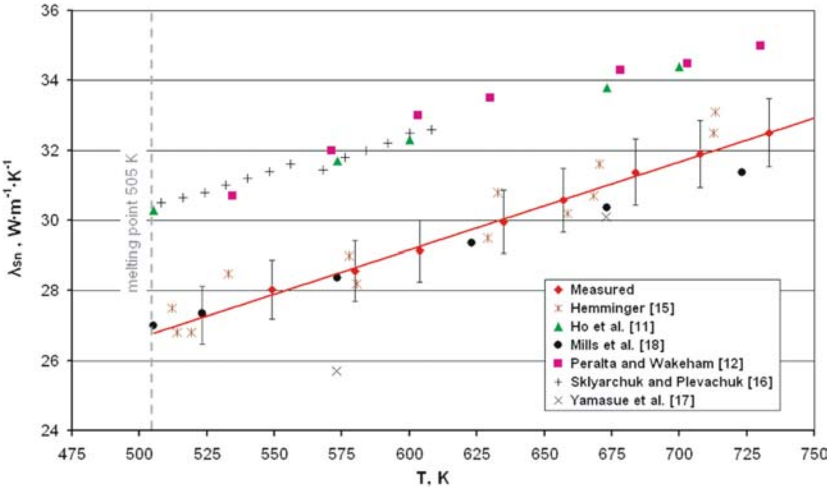

Fig. 8. Thermal conductivity of tin (melting point 505 K) and comparison with some previously measured values (error bars indicate ±3%).

methods [15,16], another transient hot-wire instrument [17], or recommended and correlated values [11,18]). The new experimental results are found in very good agreement with Hemminger [15] and Mills et al. [18]. However the differences between the results reported here obtained by the hot-wire technique and those reported earlier by the same technique amount up to 10%, which is very disturbing, because the previous studies [5] claimed an uncertainty of ±2%. The differences have been investigated and measurements of different samples of 99.99% pure tin with several different sensors with different properties of the substrate have been carried out. These additional measurements confirmed the newly measured results. The results of our previous studies lie up to 10% above those we reported earlier using an earlier version of our technique. We attribute most of the discrepancy to undetected reversals of the temperature distribution in the metal sample in the earlier work, which led to steady-state convective motion of the fluid around the wire which has now been suppressed by the use of larger temperature gradients. We have verified that larger apparent values of the thermal conductivity can be generated if the temperature distribution is not monotonically increasing. In addition, of course, improvements in the FEM modelling have had small effects.

6. CONCLUSIONS

We have employed new transient hot-wire sensors with different substrate properties to demonstrate that the theoretical description of an experimental technique for measuring the thermal conductivity of molten metals is robust and accurate. Furthermore, a new algorithm for the temporal integration of the energy equation for the transient experiment has also been applied to improve the precision.

Results from previous work [5] for molten indium fall comfortably into the ±5% uncertainty for indium. The previously measured thermal conductivity of molten tin [5] differs by up to 10% from the currently measured values, but the new, more accurate thermal conductivity has been justified by the analysis of two different samples of molten tin and usage of several transient hot-wire sensors. Other studies show a very good match for both molten metals, because the measured values are within the accuracy regions stated by other researchers. Based on experience and accuracy of the measurements of physical values, such as sensor geometry, the uncertainty of the measurements is estimated to have a maximum of ±3%. The main source of uncertainty for the above described experiments is found to arise from convective flow within the molten metal. However, provided a suitable temperature gradient along the hot wire is maintained, the sensor provides repeatable transient temperature responses and the measured thermal conductivity of the sample fluid is also stable. The comprehensive analysis of uncertainty of the measurement of the thermal conductivity of molten metals as well as a detailed description of the experimental setup and sensors can be found in recently published work [19].

Based on the facts mentioned above, it can be concluded that the method itself is robust and capable of accommodating various materials and temperature ranges.

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