International Journal of Thermophysics, Vol. 18, No. 2, 1997

Toward Standard Reference Values for the Thermal Conductivity of High-Temperature Melts[1]

M. J. Assael,[2] M. Dix,[3] I. Drummond,[3] L. Karagiannidis,[2] M. J. Lourenço,[4] C. Nieto de Castro,[4] M. Papadaki,[3] M. L. Ramires,[4] H. van den Berg,[5] and W. A. Wakeham[3,6]

The paper describes the progress made in the development of an instrument for the measurement of the thermal conductivity of molten materials at high temperatures. The instrument is designed to provide experimental data of unique accuracy at temperatures up to 1500 K on a wide range of materials, some of which will be suitable as standard reference substances. In particular, the paper concentrates upon the method of analysis of the experimental data and upon those critical aspects of the experimental technique which will enable a high accuracy to be achieved. Demonstrations of the validity of the method of treating one correction and of its behavior under typical conditions are included.

KEY WORDS: high temperature; molten metals; thermal conductivity; transient hot-strip method.

1. INTRODUCTION

Measurement of the thermal conductivity of molten materials at elevated temperatures has proved inordinately difficult to perform with a high

---

[1] Paper presented at the Fourth Asian Thermophysical Properties Conference, September 5–8, 1995, Tokyo, Japan.

[2] Department of Chemical Engineering, Aristotle University of Thessaloniki, 54006 Thessaloniki, Greece.

[3] Department of Chemical Engineering and Chemical Technology, Imperial College, Prince Consort Road, London SW7 2BY, United Kingdom.

[4] Instituto de Ciência Applicada e Tecnologia da Faculdade de Ciências da Universidade de Lisboa, Campo Grande, 1700 Lisboa, Portugal.

[5] Van der Waals-Zeeman Institute, University of Amsterdam, 1018-XE Amsterdam, The Netherlands.

[6] To whom correspondence should be addressed.

0195-928X/97/0300-0439$12.50/0

accuracy, as is revealed by the discrepancies among results discussed in several reviews [1–3]. Given the importance of such data in the modeling of a number of production processes, improved accuracy is obviously essential. Two coupled steps are required to secure the requisite accuracy routinely. First, an experimental technique must be developed to secure a high accuracy for a carefully selected set of reference materials, and second, the values of the thermal conductivity obtained must be promulgated as standard reference data for use as calibrants for routine measurements. This paper describes progress on the first of these objectives under a collaborative program with the support of the European Union.

In view of the limitations of space here, we describe briefly the essential principles of the proposed method and the analysis of the experimental results. The principles of the method derive from those first employed by Nakamura and co-workers [4–6]. We describe here one special aspect of our instrumental design.

2. PRINCIPLE OF THE EXPERIMENTAL METHOD

In the ideal model of the experimental method, sketched in Fig. 1, a planar, electrically conducting element of a material with a density ρ_w, isobaric heat capacity C_{p_w} and thermal conductivity λ_w is contained within an electrically insulating substrate of density ρ_s, isobaric heat capacity C_{p_s}, and thermal conductivity λ_s. The substrate and the electrically conducting element are supposed to be infinitely long in the z-direction. In the x-direction the conducting strip has a width of $2b$, whereas the substrate has a width of w. The conducting strip is located symmetrically within the distance w. In the y-direction, the conducting strip has a thickness $2a$ and is

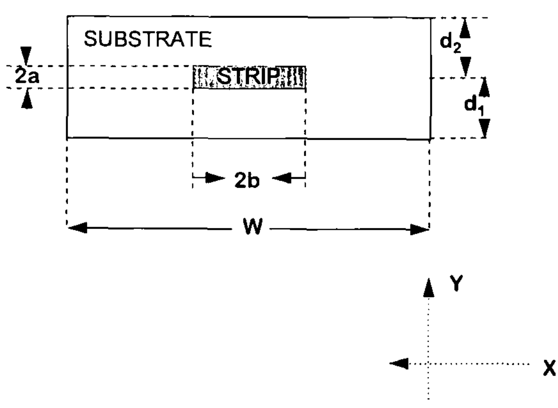

Fig. 1. A schematic diagram of the metallic hot-strip sensor.

located such that the central plane is a distance d_1 from one face and d_2 from the other face. The substrate is surrounded in the x- and y-directions by an infinite extent of a fluid with a thermal conductivity λ_m, density ρ_m, and an isobaric heat capacity C_{p_m}. At time $t = 0$, commencing from an initial equilibrium state at a temperature T_o, heat is generated in the electrical element at a rate q per unit length. In principle, the temperature rise of the metallic element, of the substrate, and of the melt are all determined by the thermal conductivity of the melt among other parameters. Its resistance change may be used to monitor the evolution of the temperature of the strip and related to the thermal conductivity of the melt.

2.1. Working Equations

The working equations for the experimental method are expressed by the following differential equations, for the temperature of the strip T_w, the temperature of the substrate T_s, and the temperature of the fluid T_m, together with the appropriate boundary and initial conditions,

$$\rho_w C_{p_w} \frac{\partial T_w}{\partial t} = \lambda_w \left[\frac{\partial^2 T_w}{\partial x^2} + \frac{\partial^2 T_w}{\partial y^2} \right] + \frac{q}{ab} \tag{1}$$

$$\rho_s C_{p_s} \frac{\partial T_s}{\partial t} = \lambda_s \left[\frac{\partial^2 T_s}{\partial x^2} + \frac{\partial^2 T_s}{\partial y^2} \right] \tag{2}$$

$$\rho_m C_{p_m} \frac{\partial T_m}{\partial t} = \lambda_m \left[\frac{\partial^2 T_m}{\partial x^2} + \frac{\partial^2 T_m}{\partial y^2} \right] \tag{3}$$

where, for the rectangular surface, $y = \pm a$, $x = 0$ to $\pm b$; $x = \pm b$, $y = 0$ to $\pm a$.

$$T_w = T_s, \qquad \lambda_w \frac{\partial T_w}{\partial x} = \lambda_s \frac{\partial T_s}{\partial x}, \qquad \text{and} \qquad \lambda_w \frac{\lambda T_w}{\partial y} = \lambda_s \frac{\partial T_s}{\partial y}, \qquad t > 0 \tag{4}$$

for $y = -d_1$, $x = 0$ to $\pm w/2$, and for $y = -d_2$, $x = 0$ to $\pm w/2$,

$$T_m = T_s, \; \lambda_m \frac{\partial T_m}{\partial x} = \lambda_s \frac{\partial T_s}{\partial x}, \qquad \text{and} \qquad \lambda_m \frac{\lambda T_m}{\partial y} = \lambda_s \frac{\partial T_s}{\partial y}, \qquad t > 0 \tag{5}$$

while for $x \to \infty$ or $y \to \infty$

$$T_m = T_o, \qquad t > 0 \tag{6}$$

and we have the initial condition, for $t = 0$,

$$T_w = T_s = T_m = T_o \qquad \text{for all} \quad x, y \tag{7}$$

There is no analytical solution known to these equations so that it is appropriate to use a numerical solution of them. This represents a valid basis for the analysis of experimental data. Equations (1) to (7) then remain the working equations of the method so that, in principle, from measurements of T_w as a function of time, it is possible to determine λ_m, provided that all other physical quantities are known given the availability of an appropriate numerical procedure. We have adopted a specially developed finite-element procedure for this purpose, which will be described in detail elsewhere [7]. The methodology has been validated in practice with simulated experimental data for $T_w(t)$ wherein all of the parameters other than λ_m are in fact prescribed. In practice, for a real sensor when the various parameters are not known, then recourse must be had to experimental determination of them in independent experiments but this does not change the principle.

3. THERMAL-CONDUCTIVITY SENSOR

The practical thermal-conductivity sensor is shown in Fig. 2. It comprises an alumina substrate which is 0.4 mm thick, 100 mm long, and 58 mm wide. Symmetrically in the middle of it is mounted a platinum strip 60 mm long, 5 μm thick, and 100 μm wide. This strip is formed in such a way that it has two current leads and one potential lead. This creates, notionally, three zones on the wire as is explained in the next section. The

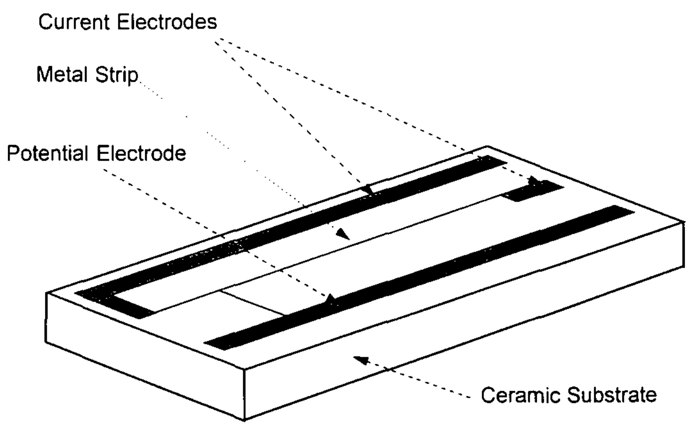

Fig. 2. A projection of the hot-strip sensor.

exact details of construction of the sensor are omitted here in the interests of brevity but it suffices to say that the entire sensor design has been founded on a rigorous study based upon the full working equations.

4. MEASUREMENT EQUIPMENT

The essential task of the measurement system is the determination of the resistance change of the central section of the platinum strip contained within the alumina substrate during transient heating. The heating current is typically 500 mA, the resistance of the strip about 50 Ω, and the resistance change typically 1 Ω, for a temperature change of a few degrees. The measurement must be completed in a period of the order of 1 s with a resolution and precision of the order of 0.1 %.

For this purpose we have constructed an automatic bridge shown in Fig. 3. In this diagram R_A represents a fixed-resistance element as does R_l. R_w represents the resistance of a central section of the platinum strip and R_s the resistance of an end portion of the strip between the potential lead and the current lead (see Fig. 2). Naturally, the temperature rise in this end portion is less than that in the majority of the strip owing to axial conduction. The same heat loss creates another segment of resistance R_s at the other end of the strip.

Physically, then, the upper R_s in Fig. 3 arises from a short segment of strip connected at one end to a current lead and at the other to a potential lead. The combination of R_w and the lower R_s arises from an exactly equivalent configuration which differs only in length. Thus, the subtraction of the two yields R_w, the resistance of an element of the strip with no ends, i.e. a finite part of an infinite strip.

If an analysis is applied to the circuit in Fig. 3, then it is readily shown that the transient changes in the voltages V_l and V_2 that accompany

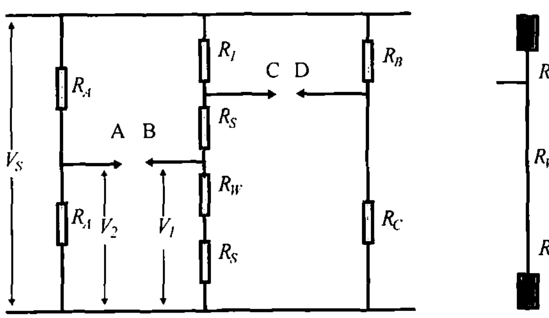

Fig. 3. The automatic-bridge configuration.

resistive heating in R_w and R_s can readily be used to determine the changes in R_w and R_s, which are denoted by r_w and r_s according to the equations

$$r_w \left\{ \frac{dV_1}{V_s} X - X + R_w + R_s \right\} = r_s \left\{ X - 2\frac{dV_1}{V_s} X - 2(R_w + R_s) \right\} - \frac{dV_1}{V_s} X^2 \tag{8}$$

wherein

$$X = R_1 + 2R_s + R_w \tag{9}$$

and

$$r_w + 2r_s = \frac{dV_2 X^2}{R_1 V_s - dV_2 X} \tag{10}$$

It follows that measurement of dV_1 and dV_2 during the transient heating of the strip is sufficient to evaluate r_w and r_s independently at any time by solution of the simultaneous Eqs. (8) and (10) at each time. These measurements are readily performed by modern methods and details of the procedures will be described elsewhere.

Once r_w has been determined, it is a simple matter to evaluate the temperature rise of this central section of the strip as a function of time to which data the working equations described earlier are to be applied.

5. RESULTS

There is no opportunity in a paper of this length to present detailed results so that we content ourselves with just two demonstrations of the performance of the instrument.

5.1. End-Effect Compensation

Figure 4 shows the fractional increase in the resistance of $R_w + R_s$, R_s, and R_w deduced from a measurement conducted in mercury at around ambient temperature. The figure concentrates only on the period between 800 and 1000 ms to reveal the effects of interest.

The fractional increase in the resistance of the strips is, in fact, proportional to the average temperature rise of the strip. We see that, as expected, the average temperature rise of the element that constitutes R_s is significantly less than that for the element that constitutes $R_w + R_s$. Since the element R_s has an approximate length of 5 mm, this is to be expected because the end effects must be substantial. Even for such a short wire,

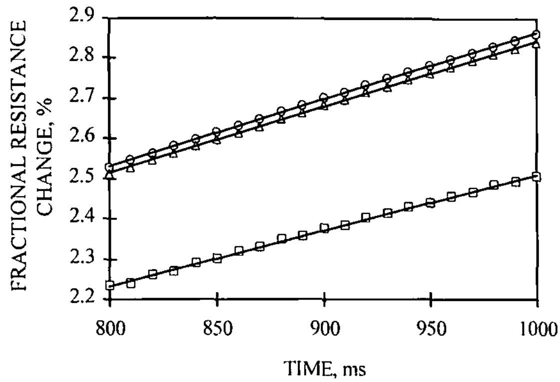

Fig. 4. Fractional resistance change of elements of the metallic strip under transient heating (○) R_w, the central segment of the strip; (△) $R_w + R_s$, the long element of the strip; (□) R_s the short element of the strip.

though, we note that the ends reduce the temperature rise by only 10% compared with the infinitely long wire. For the element $R_w + R_s$, which has a length of about 55 mm, the end effect has less than a 1% effect on the temperature rise. Confirmation of these effects has been revealed by infrared photographs.

It is readily deduced from this figure that the method outlined is entirely adequate to determine r_s with sufficient precision to allow r_w to be deduced with negligible error so that end effects are effectively eliminated.

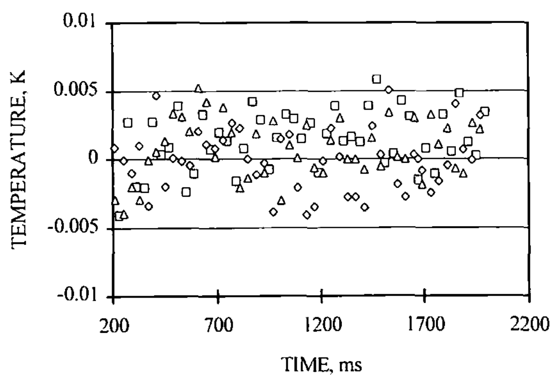

Fig. 5. Deviations of repeated measurements of the temperature rise of the metallic-strip sensor in mercury from a representative fit. (△) Run 1; (◇) Run 2; (□) Run 3.

5.2. Repeatability

A further essential element of the instrumentation must be the repeatability of the temperature-rise measurements for the platinum strip during the transient run. Accordingly, Fig. 5 shows the results of three repeated measurements of the temperature rise of the sensor under representative working conditions. The results are plotted in the form of deviations of the measured values of the temperature rise (~ 5 K) from all three runs to an arbitrary function fitted to one of them. It is clear that the deviations are typically $\pm 0.05\%$ of the total temperature rise, which is a figure that represents both the precision and repeatability of these direct measurements. At this level of precision an error of a few percent in the thermal conductivity of melts should be possible.

6. CONCLUSIONS

It has been shown how, for a transient-strip thermal conductivity assembly, it is possible to formulate exact working equations for an ideal model of the sensor. Further, the means to eliminate the effects of unwanted conduction in a third unwanted dimension has been described and demonstrated to work. The precision of the entire measurement system, demonstrated in conjunction with these measures to eliminate systematic errors, will lead to an accuracy in the measurement of the thermal conductivity of high temperature melts of the order of a few percent.

ACKNOWLEDGMENT

The authors gratefully acknowledge financial support from BCR of the European Union.

REFERENCES

1. R. Tufeu, J. P. Petitet, L. Denielou, and B. Le Neindre, *Int. J. Thermophys.* **4**:315 (1985).
2. Y. Nagasaka and A. Nagashima, *Int. J. Thermophys.* **12**:769 (1991).
3. N. Nagazawa, Y. Nagasaka, and A. Nagashima, *Int. J. Thermophys.* **13**:753 (1992).
4. S. Nakamura, T. Hibiya, and F. Yamamoto, *Rev. Sci. Instrum.* **59**:997 (1988).
5. S. Nakamura, T. Hibiya, and F. Yamamoto, *Rev. Sci. Instrum.* **59**:2600 (1988).
6. S. Nakamura, T. Hibiya, F. Yamamoto, and T. Yokota, *Int. J. Thermophys.* **12**:783 (1991).
7. M. J. Assael, M. Dix, I. W. Drummond, L. Karagiannidis, M. J. Lourenço, C. A. Nieto de Castro, M. Papadaki, M. L. Ramires, H. R. van den Berg, and W. A. Wakeham (to be published).