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METHODS OF MEASURING THE THERMAL DIFFUSIVITY OF MOLTEN FERROUS AND NONFERROUS METALS

A. D. Ivliev,[1] V. V. Morilov,[2] A. A. Kurichenko,[2] V. V. Meshkov,[1] and S. A. Goi[1]

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Different methods of measuring the thermal diffusivity of molten ferrous and nonferrous metals are considered using examples of modern experimental equipment with a brief description of their constructional features. The possibility, in principle, of solving the problem of these measurements by experimental methods is pointed out.

Keywords: *thermal diffusivity, molten metals and alloys, pulse method, temperature-waves method.*

Molten ferrous and nonferrous metals are specific media which possess high chemical activity and sublimability, the ability to dissolve many metal and nonmetal materials, etc. Investigation of the properties of such metals is a complex experimental problem. Steady-state methods widely used at low temperatures are inapplicable for molten metals. The need to obtain information on the thermal properties of molten metals and alloys has existed for a long time and is still of interest today [1, 2].

Modern methods of investigating the thermal properties of molten metals and alloys are based on the use of unsteady (quasi-stationary) heat fluxes [3, 4]. A method of measuring the thermal characteristics during melting was proposed in [5], but it can only be used in temperature regions where the sample being investigated is a two-phase sample.

The same methods that are used to investigate the properties of solids are also used to investigate the properties of materials over a wide range around the melting point, namely, pulsed and temperature waves [6, 7]. These methods enable one to measure the thermal diffusivity of a material, defined as

$$a = \lambda/(C_{sp}\gamma), \tag{1}$$

where λ is the thermal conductivity, C_{sp} is the specific heat, and γ is the density of the material [8, 9].

We will consider the main features of the use of these methods to investigate the properties of molten metals and alloys.

The pulse method. The basis of this is the relationship between the propagation time of a heat pulse and the thermal diffusivity, established by Parker [6]. The main ideas, which enable this method to be used to investigate the properties of molten metals, were developed by Stankus [2, 10]. The measuring cell of the equipment is shown in Fig. 1. It consists of an inner cylindrical crucible *4* and an outer cylindrical crucible *2*, made of high melting-point metals, arranged coaxially. The sample to be investigated *3* is placed between the crucibles. The parameters of the system, which determine the fundamental geometrical dimensions of the cell, enable one to calculate the thermal diffusivity of a material from the results of measurements of the time taken for a heat signal to propagate.

[1] Russian State Professional-Pedagogic University, Ekaterinburg, Russia; e-mail: ad_i@r66.ru.
[2] Ural State Mining University, Ekaterinburg, Russia.

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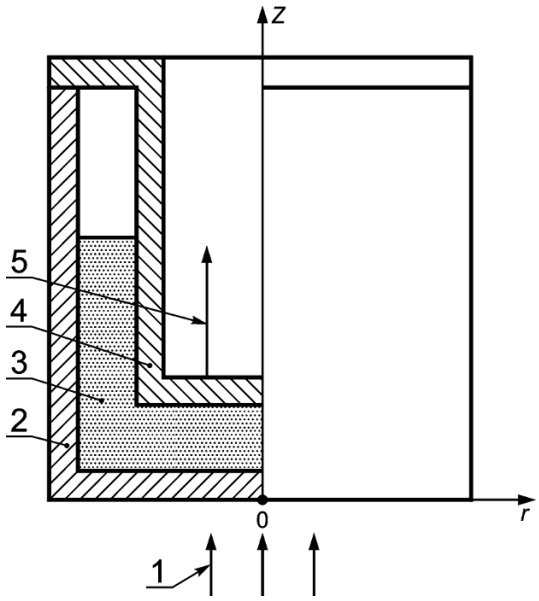

Fig. 1. Cross section of the measuring cell of the pulsed apparatus.

A pulsed heat flux *1* produced by a pulsed laser acts on the lower end surface (the bottom) of the outer crucible *2* and heats it up. The heat pulse then propagates through the components *2*, *3*, and *4*. Radiation *5* emerging from the side of the upper end surface of the crucible is converted into an electric signal by means of an infrared sensor. The measuring system only analyzes the time taken for the heat signal to propagate through the cell. This cell is monitored in the vacuum chamber using an LFA-427 instrument manufactured by the NETZSCH Company (Germany). The heater of the vacuum chamber ensures a change in the mean temperature of the sample from room temperature to 2300 K in an atmosphere of inert gases (helium or argon). The change in the temperature of the upper surface of the inner crucible due to the action of the laser pulse also has a pulsed form. Under adiabatic conditions, for an infinitely short heat pulse and for a uniform sample of thickness l, the relation between the thermal diffusivity and the time $t_{1/2}$ for the output pulsed signal to reach a value equal to half its amplitude, has the form [6]:

$$a = 1.38 l^2/(\pi^2 t_{1/2}).$$

The ideal conditions for which this relationship was derived cannot be produced in practice. Hence, a calculation using (1) will contain a considerable error, particularly, at high temperatures. Nevertheless, this relation is recommended by the manufacturer for calculating thermal diffusivity.

A procedure has been developed [2, 10] which enables the thermal diffusivity of molten metals to be estimated more correctly. A two-dimensional heat problem was considered, in which the heat signal propagates through a multilayer structure (see Fig. 1). It was assumed that the properties of the materials of the crucibles are known, the thickness of the bottom of each of them was 0.5 mm, and the thickness of the melted sample was 1–2.5 mm. Heat exchange between the cell and the environment was taken into account. It was assumed that the laser uniformly illuminates the surface of the bottom of the outer crucible, the transverse dimensions of the sample are bounded, and the actual heat pulse is not infinitesimally short. Convection occurs in the system which is largely suppressed by the inner crucible and by the absence of a free surface on the melted sample.

During the measurements, the thermal diffusivity was determined from the agreement between the theoretical and experimental curves of the pulse change in the temperature against the time [2, 10]. The error was estimated to be 5%.

The temperature-wave method can be used to investigate melts in two modifications [7]. First, it can be employed in the version used when solid-phase samples are irradiated, and secondly, in a specific version in which the temperature

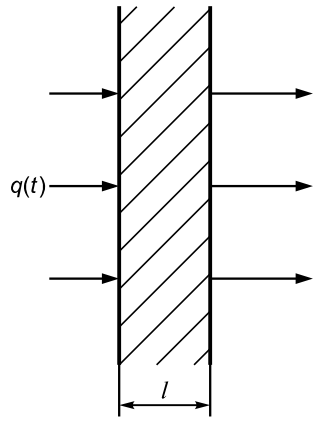

Fig. 2. The one-dimensional thermal model.

oscillations of the sample are estimated in a region of the surface on which a modulated heat flux acts, exciting a temperature wave. We will consider these versions in general outline.

1. In the first case, a one-dimensional model is the theoretical basis of the method [7, 11]. Consider the propagation of a plane temperature wave through an infinite uniform plate of thickness l = 0.5–2.5 mm (Fig. 2). A heat flux

$$q(t) = \bar{q} + \Delta q(t) \tag{2}$$

acts on the left surface of the plate, where $\bar{q}$ is the constant component of the heat flux, and $\Delta q(t)$ is the variable (periodic) component and t is the time.

The heat flux (2) (see Fig. 2) facilitates the propagation of a one-dimensional temperature wave in the sample. For small wave amplitudes, the heat conduction equation and the boundary conditions of the problem are linear. Using the ideas of the theory of harmonic waves, we note that on the right-hand surface of the plate temperature oscillations $\Delta\theta(t)$ are observed, which are shifted in phase by an amount φ with respect to the oscillations of the heat flux $\Delta q(t)$ and have an amplitude $\Delta\theta$. As a result of the analysis, we can conclude that, for sufficiently large phase delays φ and for small heat-exchange coefficients of the sample with the environment, the relation between the phase and the thermal diffusivity of the sample has the form [7, 11]:

$$a = \omega l^2 / k^2, \tag{3}$$

where ω is the angular frequency of the temperature wave and $k = \sqrt{2}[|\varphi| - (\pi/4)]$.

When the transverse dimensions and thickness of the sample are limited, one can also use this one-dimensional adiabatic method, but only for a long phase delay of the temperature wave. For the samples used in practice, the conditions for (3) to be applicable are as follows: $Bi < 0.012$ and $k > 2.5$, where Bi is the Biot number [3, 4, 7], which defines the intensity of the heat exchange between the sample and the environment. The error of this method, compared with the exact result for the two-dimensional model, does not exceed 1%, i.e., the temperature-wave method enables the effect of heat exchange and the dimensions of the sample to be taken into account.

Hence, for known l and ω, to calculate the thermal diffusivity of a sample it is necessary to measure φ. Consequently, the instrument for investigating thermal diffusivity is a phase meter. This phase meter must process fairly weak signals, since small amplitudes of the temperature-wave oscillations are an important condition, which enables the heat conduction equation to be linearized. This procedure can be employed in equipment with different types of heat-flux sources.

Consider *electron heating*. In this case, the heat flux (2) is produced by a modulated electron beam. Measurements are therefore only possible in a vacuum. The samples being investigated must be conducting, since the sample acts as an anode.

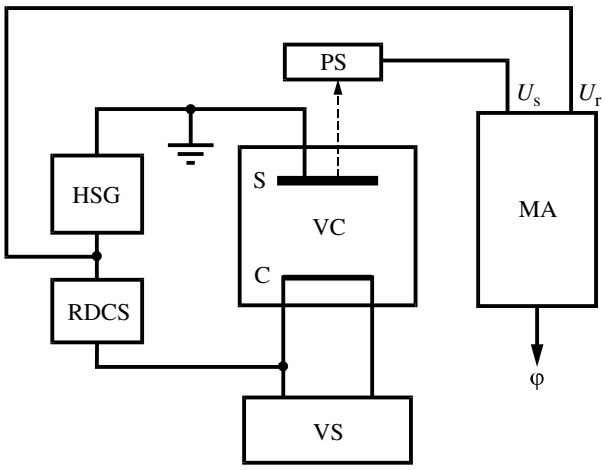

Fig. 3. Sketch of the equipment with electron heating.

A sketch of the equipment with electron heating is shown in Fig. 3; it is similar to those described in [12, 13]. The sample *S* has the form of a flat plate with transverse dimensions of 8–12 mm, placed horizontally in a vacuum chamber *VC* perpendicular to the plane of the diagram. It acts as the anode of a vacuum diode. The cathode *C* is a flat tungsten helix placed perpendicular to the plane of the diagram. The cathode is heated by an electric current from a voltage source *VS*. The electron flux acts on the lower plane of the surface of the sample. An harmonic signal generator *HSG* and a regulated dc voltage source *RDCS* are connected between the diode cathode and anode. As a result, the intensity of the electron flux is determined both by the constant value-varying voltage and by the variable harmonic voltage. The temperature oscillations of the upper plane of the sample surface are analyzed using a photosensor *PS*.

The heat flux consists of two components: a relatively slowly increasing component and the harmonic component, and hence the temperature waves are produced on a background of a monotonically varying (increasing) temperature of the sample. By selecting the rate of change of the mean temperature of the sample, one can measure the thermal diffusivity not only in the solid state but also during melting, provided the sample does not lose its shape. The voltages of the signal from the photosensor U_c and the reference voltage U_r, which carries information on the frequency of the temperature wave and the phase of the heat flux acting on the sample, are applied to the input of the measuring apparatus *MA*. The signal is averaged over one period, since, in the dynamic method, the signal is not repeated. The rate of the monotonic increase in temperature in the equipment can be varied in the range 0–100 K/sec, while the frequency of the temperature wave can be varied in the range 4–400 Hz. Measurements are carried out in the usual (quasistatic) and dynamic modes. The working temperature range is from 700 K to the melting point of the sample. The root mean square error of a thermal diffusivity measurement is approximately 2–3%. Data are given for solid-phase samples.

Another way of constructing the equipment with electron heating was proposed by Filippov in [14]. This equipment was designed to measure the thermal diffusivity of molten metals. Unlike Fig. 1, the substance being investigated is poured between the side cylindrical surfaces of the cell. The cell is the anode system, and the cathode is a tungsten helix placed inside the cell along the axis of the system. The isotherms in this problem are not plane. By an appropriate choice of the radii of the cylinders, between which the molten sample is situated, one can use the one-dimensional plane method considered above for the calculations. To suppress convection currents in the sample, horizontal plane partitions are placed in the sample, which hinder the motion of the molten metal. In the experiment, the phase of the oscillations of the temperature of the outer cylindrical surface of the cell is compared with the phase of the oscillations of the heat flux. The measurement error is 2%.

Radiation heating of the sample is carried out in the equipment using a laser [7]. A sketch of the equipment is shown in Fig. 4.

Radiation from a continuous laser propagates horizontally, passes through the amplitude modulator *M* and is converted into a train of pulses $q(t)$. This radiation acts on the left surface of the sample *S* (see Fig. 4) placed in a vacuum cham-

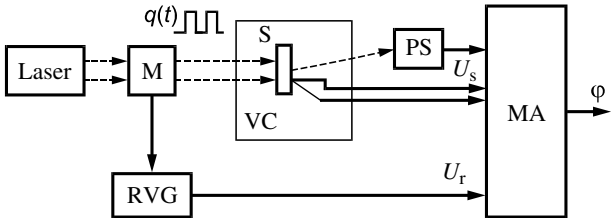

Fig. 4. Sketch of the equipment with radiation heating.

ber *VC*. The sample is a high melting point metal container, in which the material being investigated is placed, and where it is converted to the liquid phase at its melting point. The container has a plane-parallel shape. The thickness of the material of which the container is made is usually 0.2 mm, the distance between the walls of the container is 1.5–2 mm (the sample is placed in this region) and the transverse dimensions of the container–sample are 12–15 mm.

The container with the sample is situated in a vacuum chamber, supplied with a special heater, which varies the mean temperature of the sample, since the laser merely modulates the temperature, i.e., it produces a temperature wave. The mean operating temperature range can be varied from room temperature to 2300 K. Measurements can be carried out in a vacuum or in an atmosphere of helium. Free convection occurs in the molten sample. In this equipment, as in the equipment described in [2, 10, 14], the thermal signal passes through a multilayer system consisting of the two walls of the container and the material being investigated. The effect of the container properties in the temperature-wave method is also taken into account [15]. Temperature oscillations of the right-hand surface of the sample are converted, using a photosensor *PS* or a thermocouple, into an electrical signal U_s which enters the measuring apparatus. The reference voltage U_r, generated by the reference voltage generator *RVG*, is fed to its second input. The operation of the generator is connected with the operation of the modulator. The reference voltage contains information on the phase of the heat flux $q(t)$, acting on the left-hand surface of the sample, and the frequency of the temperature wave ω. Estimates show that the overall root mean square error of a measurement in this equipment is 5%.

2. The second version of the temperature-wave method was specially developed to investigate the properties of molten metals and alloys. The source of information, which enables the values of the thermal characteristics of a material to be established, can also be the left-hand surface of the sample (see Fig. 4) [16]. Thus, one can calculate the thermal diffusivity from the phase of the temperature oscillations of the left-hand surface. This version is of interest since it can be used for samples, taking into account semi-infinite media in the framework of the model considered [17]. There is therefore no need to monitor the linear dimensions (in particular, the thickness) of the molten samples, which are extremely unstable. This drawback is common to all the methods considered above.

Hence, in the method considered here, a time-varying heat flux (2) is applied to the surface of the sample. The diameter of the heat flux has finite dimensions of 1–3 mm. As a consequence of this, a temperature wave is generated in the sample, which propagates deep into the sample and over its surface. Calculation shows that the phase of the oscillations of the surface temperature of the sample at the central point of the incident heat flux is related to the thermal diffusivity of the material. The analytic expressions are fairly complex [17], but the problem is solvable in principle.

Using this procedure, equipment has been constructed which employs laser radiation to excite a temperature wave in the sample [18, 19]. Radiation heating enables one to investigate not only conducting materials, but also dielectrics that are nontransparent for laser radiation. The apparatus is similar to that considered above (see Fig. 4). The radiation of a continuously operating laser propagates in a horizontal direction, passes through a modulator and is converted into a periodic train of pulses. The pulse radiation obtained, on reflection from a mirror, enters a vacuum chamber on to the surface of the sample being investigated. The sample is molten and hence its surface is situated horizontally. The vacuum chamber is supplied with a heater, which enables the average temperature of the sample to be changed from room temperature to 2300 K. A photosensor, which converts the temperature oscillations into an electric signal, is aimed at the central region of the sample surface, which is subjected to the action of the laser radiation. The laser and the photosensor are chosen so that the latter

does not react to the laser radiation reflected from the surface of the sample, and only the natural radiation of the sample is recorded. The signal from the output of the photosensor is incident on the measuring apparatus (*MA*). By processing it, one can measure the phase delay and calculate the thermal diffusivity of the sample. The root mean square error of a measurement of the thermal diffusivity, achieved using this equipment, is estimated to be 6%. A considerable fraction of this error is due to the uncertainty of the effect of convective currents on the results of measurements of thermal diffusivity. Similar problems also arise in the measuring systems considered earlier.

It should be noted that none of the proposed systems measured the average temperature of the molten sample itself by contact methods. Either an estimate of the temperature of the solid-phase cell or the temperature of the gas in the neighborhood of the surface of the sample were used as the average temperature. Thermocouples were employed for these measurements. In practice, pyrometer measurements of the average temperature of the sample were not carried out in these systems.

Conclusion. Various types of equipment designed to investigate the thermal diffusivity of molten metals and alloys have been developed and used in Russia at the present time. However, the operating quality of these systems is far from perfect. Many components of the errors have not been investigated, as a result of which a rigorous estimate of the actual measurement errors has not yet been carried out on any of them.

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