DIFFUSIVITY AND THERMAL CONDUCTIVITY IN SOLID AND LIQUID TIN

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A method for determination of diffusivity of liquid metals in a hemispherical volume is described and experimental results are presented.

Many studies have been dedicated to investigation of thermal and electrical properties of metals in the liquid state (cf., for example, [1]). Nevertheless, conflicts in available data and the limited temperature range of these studies makes development of new experimental methods desirable for study of conductivity and diffusivity at high temperatures. Impulse heating or temperature wave methods [1] employed for these purposes usually require the use of metallic containers with liquid metals, which creates difficulties with respect to corrosive metals at high temperatures.

In connection with this problem, we have developed a method of diffusivity measurement in solid and liquid metals in which the geometrical parameter entering into the formula was independent of the form of the vessel containing the melted metal. The basic idea of this method is the creation of a heat pulse in the center of a hemispherical crucible with the metal, in which is placed a temperature sensor (for example, a thermocouple) at a definite distance from the center. Measurement of the temperature pulse delay time gives information on the metal's diffusivity.

Since the theoretical solution of the thermal conductivity equation for a hemisphere in the general case is quite complex, in the first approximation we will consider the theory of the method for a thermally insulated hemisphere. In a hemisphere $0 < r \leq R$ at moment $\tau = 0$ at a point with coordinates (0, 0, 0) let there occur an instantaneous liberation of an amount of heat Q. Then the conductivity equation will have the form

$$\frac{\partial[rT(r, \tau)]}{\partial\tau} = a\frac{\partial^2[rT(r, \tau)]}{\partial r^2} \quad (1)$$

with initial conditions $\tau = 0$; $T(0, r) = 0$ and boundary conditions $\partial T/\partial r|_{r=0} = 0$; $\partial T/\partial r|_{r=R} = 0$ (the condition for thermal insulation of the lateral surface); $\partial T/\partial\theta|_{\theta=\pi/2} = 0$ (absence of thermal flux on plane surface). The solution of Eq. (1) in dimensionless coordinates will be

$$\vartheta = \frac{1}{\tilde{r}}\sum_{n=0}^{\infty}\frac{1+\mu_n^2}{\mu_n}\sin(\mu_n\tilde{r})\exp(-\mu_n^2\,\mathrm{Fo}) + \frac{3}{2}, \quad (2)$$

where $\vartheta = (T/Q)\rho c_p \pi R^3$ is the relative temperature; μ_n are the positive roots of the characteristic equation

$$\mu_n \operatorname{ctg}\mu_n - 1 = 0. \quad (3)$$

Figure 1 shows the temperature distribution as a function of τ for $\tilde{r}$ = 0.5, 0.55, and 0.75. By measuring the time interval over which the temperature signal at point $\tilde{r}$ reaches half of its maximum value, the curves of Fig. 1 can be used to determine the diffusivity coefficient of the material. The method is most easily realized not with a heat source (for which a laser or a well-focussed flash lamp is necessary),

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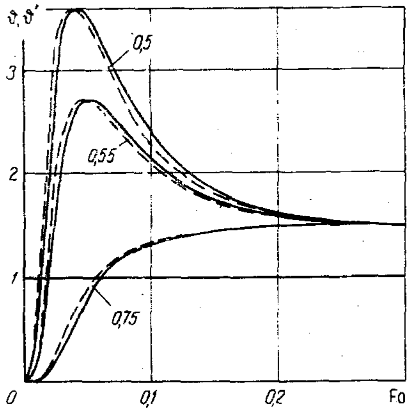

Fig. 1

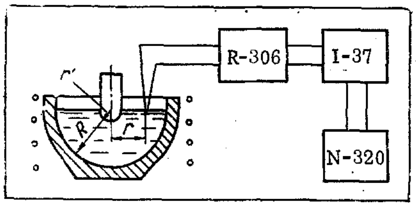

Fig. 2

Fig. 1. Relative temperature ϑ and ϑ' distribution as a function of Fo for instantaneous source (solid curve (ϑ), point source; dashed curve (ϑ'), hemispherical source); relative radius of hemispherical source $\tilde{r}'$ = 0.16. Numbers on curves are values of $\tilde{r}$.

Fig. 2. Apparatus used.

but with a heat discharge, which can easily be created by boiling or melting some substance in the center of the crucible. In this case the instantaneous heat discharge will have finite dimensions and it is convenient to confine it to the shape of a hemisphere with r < R. The solution of Eq. (1) in dimensionless coordinates in this case will be

$$\vartheta' = \frac{1}{\tilde{r}\tilde{r}'} \sum_{n=0}^{\infty} \frac{1+\mu_n^2}{\mu_n} \sin(\mu_n \tilde{r}) \sin(\mu_n \tilde{r}') \exp(-\mu_n^2 \mathrm{Fo}) + \frac{3}{2}. \tag{4}$$

The temperature ϑ' distribution as a function of coordinate and time is also presented in Fig. 1.

To verify the effectiveness of the method we conducted a study of the diffusivity a of pure tin (0.001% impurities). The specimen was melted in a porcelain crucible of radius R = 25 mm (see Fig. 2). The heat discharge was created by a drop of water falling into a thin walled steel hemisphere with radius 4 mm. Chromel—alumel thermocouples were located at various distances from the center. The constant temperature component was compensated with an R-306 potentiometer, with which the reference temperature was determined. The variable component was amplified by an I37 amplifier and recorded on a high speed recording milliammeter, type N320. Droplet boiling time in the experiments was approximately 0.1 sec; signal amplitude was 10°, and delay time to the half level about 0.9 sec. These conditions are not optimal, and the corresponding uncertainties can be evaluated as follows.

1. Under these conditions the pulse duration is not infinitely small. A more exact solution of the conductivity equation in this case can be achieved in the form of the difference of solutions for two continuous point sources of constant power, acting at moments $\tau = 0$ and $\tau = t'$ respectively, at the point r = 0 (t' is the pulse duration). The corresponding solution for a continuous source is contained in [2, 3]. A comparison of this refined solution with the solution for an instantaneous source which we used shows that the latter leads to a systemic error of 5%.

2. The value of the Bi criterion at our temperatures did not exceed 0.02. A coarse estimate of the influence of heat exchange on the surface [2, 3] showed that its contribution does not exceed 2% of the total thermal flux, which produces approximately the same contribution to a.

3. The error due to thermocouple inertia was evaluated in accordance with the data of [1], and did not exceed 1%.

4. Random measurement error connected with uncertainty of determining thermocouple position and dimensions, was approximately 1.5-2%.

5. Scattering of diffusivity values at one temperature, due primarily to inaccuracy in determining temperature pulse delay time were within the limits of 2%.

Thus, in these preliminary experiments the total uncertainty was about 10%.

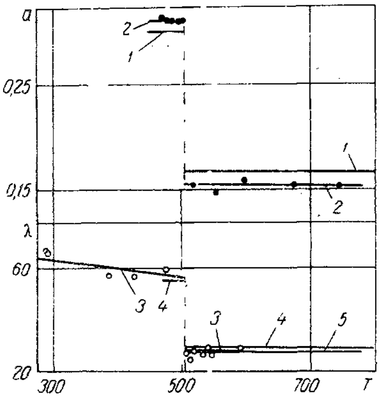

Fig. 3. Tin diffusivity: 1) data of [1]; 2) our data; thermal conductivity of tin; 3) our data; 4) [1]; 5) calculations with use of our diffusivity data, data of [1] on heat capacity, and data of [5] on tin density; $a \cdot 10^4$, m²/sec; λ, W/m·deg; T, °K.

The results of measuring tin diffusivity in the temperature range 450-750°K are presented in Fig. 3. The averaged data of [1] are also presented. It is evident that our data differs from the results of [1] by not more than 10%.

For complex studies of the thermal properties, the **conductivity** of the same tin was measured by the small connector method [1,4], which is a variation of the Kohlrausch method. The essence of the method is the determination of the change in resistance of a thin short connector when current passes through it. The connector geometry is chosen to minimize thermal losses. Our specimen was melted metal, located in interior (Ø = 10 mm) and exterior (Ø = 30 mm) crucibles. In the bottom of the interior crucible a hole was drilled with Ø = 0.1 mm and depth 0.3 mm. The resistance shift, of the order of 10^{-5}-10^{-6} Ω for currents up to 7 A, was measured by a bridge circuit with an F-116/1 meter. Measurements were made in a vacuum of 10^{-3} mm Hg. The formula used for calculation corresponded to [5]. The temperature coefficient appearing in the formula was determined experimentally, as described in [4]. The error in determination of λ, similarly related essentially to error in measurement of $\partial\rho/\partial T$ was about 10%. The results of a study of λ of tin in the solid and liquid states are presented in Fig. 3. It is evident that the data obtained by us for λ agrees satisfactorily with the data of the literature, and with the values calculated with use of the diffusivity measured by us together with the data of [1, 5] on heat capacity and density.

Thus a method has been developed for pulse measurement of hemispherical specimens with an instantaneous head discharge (source). The data on diffusivity of tin obtained by this method agree satisfactorily with the literature. By making corrections for heat transfer this method can be employed for studies at higher temperatures.

Measurement of heat conductivity of solid and liquid tin by the "small conductor" method also showed agreement with the results of other authors, and with calculated results based on the diffusivity measurements.

NOTATION

r	is the radius;
R	is the hemisphere radius;
r'	is the radius of hemispherical source;
$\tilde{r} = r/R$, $\tilde{r}' = r'/R$	are the dimensionless coordinates;
τ	is the time;
Q	is the quantity of heat;
T	is the temperature;
ϑ and ϑ'	are the dimensionless temperatures;
$Fo = a\tau/R^2$	is the Fourier number;
a	is the diffusivity coefficient;
λ	is the thermal conductivity coefficient.

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