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MEASUREMENTS OF THERMAL CONDUCTIVITY AND SPECIFIC HEAT OF DILUTE METALLIC ALLOYS IN THE FORM OF SMALL SAMPLES BY ELECTRICAL METHOD

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The method of determining the thermal conductivity depends upon a relation between the maximum temperature (θ_m) attained for a given current and potential difference (V) in a current carrying specimen. Heat is assumed to enter and leave specimen only through the surfaces through which electric current enters and leaves, other surfaces being insulated against flow of both heat and electricity. The plane ends of the rod were taken to be isothermal and equipotential surfaces held at a constant temperature.

For the measurement of the specific heat, a homogeneous constant Joule heating is imposed. The initial slope of (θ_m) as a function of time is inversely proportional of heat capacity regardless of heat losses. Measurements on dilute Ni base Cr alloy sample up to 360 °C.

Direct measurement of thermal conductivity "K" is difficult and in most cases inaccurate [1]. The reasons for this are heat losses and imperfect thermal contact. In an attempt to avoid such difficulties, measurements on ferromagnetic metals and alloys were made by an indirect method to investigate the variation of thermal conductivity with temperature in the region around the Curie temperature [2–4]. The method depends upon a relation between the temperature and the potential distributions in a current-carrying conductor kept under vacuum.

In the present work, measurements were performed on dilute Ni base Cr alloy sample up to 640 K. The electrical resistivity "ϱ" was accurately measured [5], and it has been found that the temperature coefficient of resistivity $\left(\frac{1}{\varrho}\frac{d\varrho}{dT}\right)$ diverges logarithmically at the Curie temperature T_c. The quantity "K" can be obtained by plotting the potential difference (V) across the sample against "θ_m", where "θ_m" is the maximum temperature attained for a given current in the conductor the ends of

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which are held at a constant temperature. Specific heat "C_p" is determined from the rate of temperature rise while the sample is ohmically heated [6].

Theory

Consider an isotropic homogeneous electrically heated conductor in the form of a thin rod mounted between main leads in vacuum with blackbody surroundings. For a circular conductor of diameter ($2r_0$) and cross-sectional area (A), the following steady state differrential equation holds:

$$-\frac{\mathrm{d}}{\mathrm{d}x}\left(K\frac{\mathrm{d}T}{\mathrm{d}x}\right)=\frac{I^2\varrho}{A^2}-\frac{2\pi r_0\sigma}{A}(\varepsilon T^4-\varepsilon_b T_b^4)-$$
$$-\frac{I\tau}{A}\frac{\mathrm{d}T}{\mathrm{d}x}-\frac{2\pi r_0}{A}K_g\left(\frac{\mathrm{d}T}{\mathrm{d}r}\right)_{r0} \tag{1}$$

where K = thermal conductivity of the sample
x = axial coordinate
I = current in the sample
ϱ = resistivity at temperature T
σ = Stefan–Boltzmann's constant
ε = emissivity of the sample surface at temperature T
ε_b = sample absorptivity at temperature T to blackbody radiation from container walls at temperature T_b
τ = Thomson coefficient
K_g = thermal conductivity of the surrounding gas.

Neglecting radiation and conduction through the surrounding gas, equation (1) reduces to:

$$\frac{I^2\varrho}{A^2}-\frac{I\tau}{A}\frac{\mathrm{d}T}{\mathrm{d}x}+\frac{\mathrm{d}}{\mathrm{d}x}\left(K\frac{\mathrm{d}T}{\mathrm{d}x}\right)=0 \tag{2}$$

Following Llewellyn Jones [7] and Flynn and O'Hagan [8], the equation can be written in the form:

$$K\varrho\frac{\mathrm{d}T}{\mathrm{d}x}+\left[\psi-\int_T^{T_m}\tau\,\mathrm{d}T\right]\frac{\mathrm{d}\psi}{\mathrm{d}x}=0 \tag{3}$$

where ψ is a pseudo-potential defined by:

$$I=-\frac{A}{\varrho}\frac{\mathrm{d}\psi}{\mathrm{d}x} \tag{4}$$

and T_m is the maximum temperature at some point on the rod (to be discussed later). If we choose the pseudo-potential ψ to be zero at the point where the temperature gradient vanishes (i.e. at $T = T_m$), then by integration from an isothermal and equipotential surface (T, ψ) to the surface of maximum temperature $(T_m, 0)$, the result is:

$$\psi^2 = 2\int_T^{T_m} K\varrho \, dT + 2\int_0^{\psi} d\psi \int_T^{T_m} \tau \, dT \tag{5}$$

where the second term on the right-hand side represents the contribution to ψ^2 due to Thomson effect. Since the Thomson effect is small, it can be shown that:

$$\psi = \frac{V}{2}, \tag{6}$$

where V is the potential difference between two surfaces of same temperature T_0 which in the present case are the two ends of the rod. Then

$$V^2 = 8\int_{T_0}^{T_m} K\varrho \, dT \tag{7}$$

From this equation, we can see that in an electrically heated conductor the maximum temperature rise $(T_m - T_0)$, in the region between two surfaces which are held at the same temperature is a function only of the voltage drop between the surfaces, and of K and "ϱ" of the material. It is independent of the geometry of the conductor provided the lateral losses and the Thomson heat are negligible.

The form in which the (V, θ) relation (where $\theta \equiv T$) actually used, is obtained by differentiating the above equation (7) partially with respect to θ_m. This gives

$$\frac{V}{4}\left(\frac{\partial V}{\partial \theta_m}\right)_{\theta_0} = K(\theta_m)\,\varrho(\theta_m), \tag{8}$$

where $\varrho(\theta_m)$ of the same sample is measured using a separate experiment.

Temperature distribution along the rod

If we include in equation (2) the temperature dependence of the electrical resistivity as $\varrho = \varrho_0[1 + \alpha_0(T - T_0^*)]$ where T_0^* is the temperature to which the constants ϱ_0 and α_0 correspond, and if we neglect the Thomson effects then we get:

$$\frac{d^2T}{dx^2} - PT = -Q, \tag{9}$$

where

$$P = -\frac{I^2\varrho_0\alpha_0}{A^2K}$$

and

$$Q=\frac{I^2\varrho_0}{A^2K}-\frac{I^2\varrho_0\alpha_0T_0^*}{A^2K}$$

Solution of equation (9) is:

$$T=\frac{Q}{P}+C_1e^{+\sqrt{P}x}+C_2e^{-\sqrt{P}x}, \tag{10}$$

where C_1 and C_2 are constants to be determined from the boundary conditions:

$$T=T_0 \qquad \text{at} \qquad x=0,\ L,$$

where L is the length of the conductor. Solving for C_1 and C_2, we found that:

$$T=\frac{Q}{P}+\frac{(T_0-Q/P)}{e^{\sqrt{P}L}-e^{-\sqrt{P}L}}[(1-e^{-\sqrt{P}L})e^{\sqrt{P}x}+ \\ +(e^{\sqrt{P}L}-1)e^{-\sqrt{P}x}] \tag{11}$$

This expression gives the temperature distribution along the rod. The condition for the maximum temperature is:

$$e^{2\sqrt{P}x}=\frac{e^{\sqrt{P}L}-1}{1-e^{-\sqrt{P}L}} \qquad \text{at} \qquad T=T_m, \tag{12}$$

If $e^{\sqrt{P}L}\gg 1$, then $x=\frac{L}{2}$ at $T=T_m$.

Specific heat

For the measurement of the specific heat, homogeneous constant Joule heating is imposed. The form of the heat conduction equation for a sample with one-dimensional temperature dependence and for small temperature differences is:

$$-\frac{\mathrm{d}}{\mathrm{d}x}\left(K\frac{\mathrm{d}\theta}{\mathrm{d}x}\right)=\frac{I^2\varrho}{A^2}-\frac{H}{A}\theta-C_p\frac{\mathrm{d}\theta}{\mathrm{d}t}, \tag{13}$$

where H is the rate of heat loss from the surface area per unit length of the sample per degree of temperature difference between the sample and its surroundings, C_p is the specific heat of the sample, t is the time, and x is measured from the mid-point of the sample. In deriving equation (13) the validity of the heat diffusion and a linear cooling law are assumed. Lateral gradients are ignored.

Let:

$$\frac{K}{C_p} = a = \text{thermal diffusivity,}$$

$$\frac{H}{AC_p} = h = \text{radial heat loss parameter,}$$

$$\frac{I^2 \varrho}{A^2 C_p} = s = \text{Joule heat parameter,}$$

Equation (13) takes the form:

$$\frac{\partial \theta}{\partial t} = a \frac{\partial^2 \theta}{\partial x^2} - h\theta + s \tag{14}$$

The subsidiary conditions of this equation are:

$$s = \text{constant}; \qquad \theta(x, 0) = 0 \qquad \text{and}$$

$$\frac{\partial \theta(x, t)}{\partial t} = 0$$

The solution of equation (14) is:

$$\theta(t) = \frac{s}{h}(1 - e^{-ht}). \tag{15}$$

For small values of "t" (typically one minute), we obtain first order in "t":

$$\theta(t) = st \tag{16}$$

Following the definition of "s", the initial slope of "θ" as a function of "t" is inversely proportional to "C_p" regardless of heat losses.

Experimental

Apparatus

The apparatus consists of two copper water-cooled heat sinks held to a steel base plate. To insure good thermal contact between the specimen and the heat sinks, the specimen is clamped symmetrically between two pairs of semi-cylinders made of brass fitted accurately into cylindrical channels in the heat sinks. The base of the copper heat sink is flat and well polished; (Fig. 1).

The sample under examination is machined into the form of a wire of 2.3 mm diameter. To reduce heat losses, the surface of the specimen and the sinks are

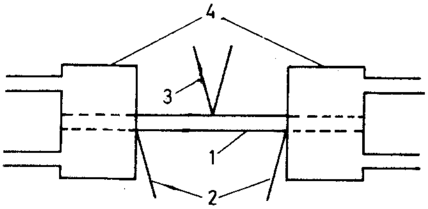

Fig. 1 Scheme of the apparatus. 1 – Specimen, 2 – Needle probes to measure V, 3 – Thermocouple to measure θ_m, 4 – Water cooled heat sinks

contained in a vacuum glass bell-jar which can be pumped easily to a pressure of 0.1 mtorr; this being found adequate to eliminate convection losses. The cooling water is held in a large temperature controlled bath, and is pumped through the two sinks simultaneously. The temperature of this cooling water is monitored with a Cu-constantan thermocouple and kept constant within ±0.2 deg.

The current is passed through the specimen using two thick leads via the cylindrical channels of the heat sinks. A d.c. source capable of delivering up to 80 A was used. The use of d.c. avoids difficulties associated with enhanced skin effect in ferromagnetic materials. The contacts for the measurement of the voltage across the specimen are made with two fine needle probes. The points of contact are precisely on the junctions of the flat ends of the specimen which are clamped to the heat sinks. The leads from the probes are connected to a vernier potentiometer measuring to the nearest 1 μV. The maximum temperature "θ_m" of the sample is measured by Cu-constantan thermocouple silverly-welded to it. The correct position of this thermocouple junction is the mid-potential points as proved in the previous section. The e.m.f of the thermocouple is read after thermal equilibrium is attained.

Measurements

Measurements were performed on Ni–0.12 at. % Cr sample. For each constant voltage across the sample, the readings of "V", "T_0", and "θ_m" were taken. Thermal equilibrium was attained in less than twenty minutes for low current values and in about one hour at high temperatures. From the $(V-\theta_m)$ plot, the value $\frac{1}{V}\left(\frac{\mathrm{d}V}{\mathrm{d}\theta}\right)_{T_0}$ for each θ_m was calculated. "K" as a function of temperature was determined using Eq. (8). The absolute accuracy of "K" is limited largely by the thermocouple calibration and in calculating the slope of the $(V-\theta_m)$ curve. It was found to be about 1%. Adding uncertainties in measuring "ϱ" the best accuracy claimed for "K" is 3%.

For the specific heat measurement, a current of (about 20 A) was transmitted through the sample and the temperature rise at the centre of the sample was simultaneously recorded. After an initial thermocouple delay (about 0.5 sec), the temperature rises linearly for about fifteen seconds. The slope of this line determines the specific heat, see (2c).

Results and discussion

Figure 2 gives a plot of the measured ($V-\theta_m$) relation in which one can notice a change of slope starting from 620 K where the sample begins to transform into the paramagnetic phase. In Fig. 3, the experimental thermal conductivity $K(T)$ is plotted. As can be seen K falls with the increase of temperature up to about the Curie point T_c where a minimum in K is observed (T_c = 628 K). We may suggest that the critical fluctuations cause a depression on the thermal conductivity below its general trend in the neighbourhood of T_c. In the paramagnetic phase, the thermal conductivity increases with a large positive slope. This is in contrast with the monotonic decrease of $K(T)$ in the case of ordinary metals, towards an approximately constant value at high temperatures. This behaviour of $K(T)$ has been previously observed in pure Ni by Powell et al. [9], in pure Ni and Ni–Cu alloys by Jackson and Saunders [10], and in Ni–Mn dilute alloys by Ammar et al. [4]. On measuring transport properties of monocrystalline sample of TbZn by Ausloos [11], no change of sign in $\frac{dK}{dT}$ for both sides of its T_c (~200 K) was observed.

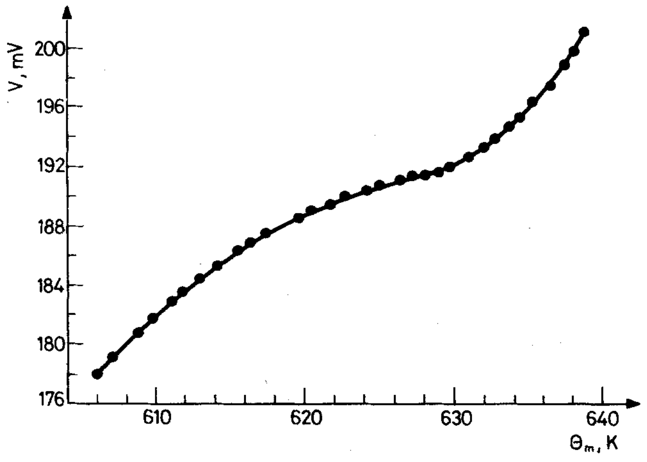

Fig. 2 Potential difference across the wire *vs.* mid-point temperature

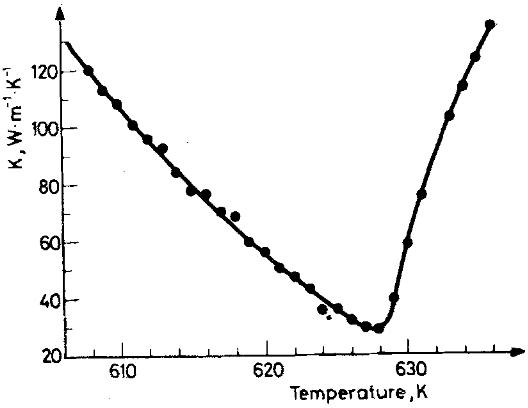

Fig. 3 Thermal conductivity *vs.* temperature

The electrical resistivity (ϱ) has been measured for the same sample [5] and it was found that the divergence of the temperature coefficient of resistivity $\alpha(T)$ is logarithmic in the near vicinity of T_c which is, as well nearly identical to that of C_p for the same sample [12]. The divergence behaviour of $\frac{dK}{dT}$ is expected to be more complex than that of $\alpha(T)$ or C_p.

Figure 4 shows a plot of the heating current (I) versus the voltage (V) across the sample under test. A scale giving the corresponding temperature, θ_m, of the middle of the specimen has been superimposed upon the voltage axis. It is clear that θ_m (or V) increases linearly as "I" increases until a certain temperature after which a small increase of "I" produces a large increase of θ_m. In order to explain this curve, the

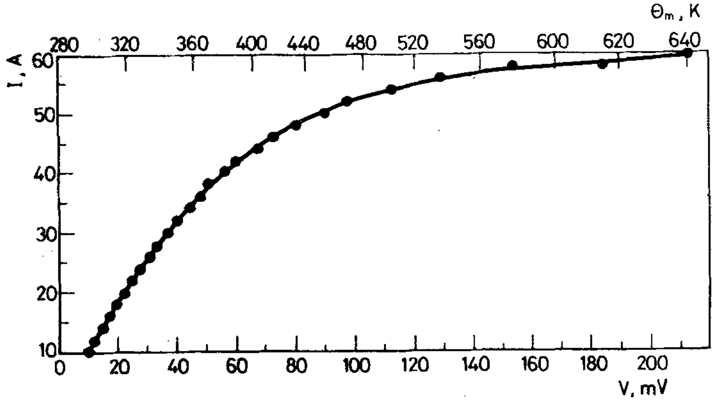

Fig. 4 Current-voltage curve (with specimen ends at 27 °C)

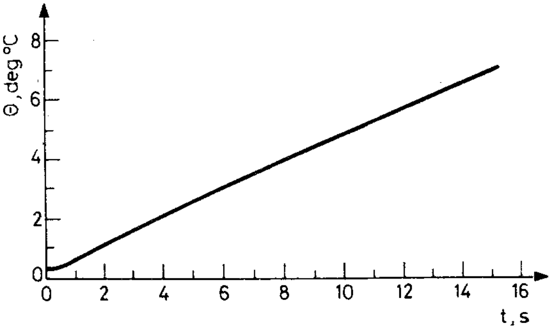

Fig. 5 Temperature rise due to joule heating of the wire *vs.* time at 27 °C

general equation of the heat-transfer problem, Eq. (1), must be solved to obtain the temperature profile along the sample. For the early stage of this curve, it seems reasonable to assume that the resistivity is an ohmic one and $\alpha(T)$ is linear. This particular feature of the $(I-V)$ variation has been pointed out by Greenwood and Williamson [13].

The specific heat result is determined from Fig. 5 and amounts to be 498 J kg^{-1} K^{-1} at 300 K, which is nearly close to the predetermined value [12]. In order to obtain other values of C_p at different temperatures, microfurnaces should be included at the ends of the sample.

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Zusammenfassung — Die Methode zur Bestimmung der Wärmeleitfähigkeit hängt vom Verhältnis zwischen der bei einer bestimmten Stromstärke erreichten Maximumtemperatur (θ_m) und dem Spannungsabfall an einem stromdurchflossenen Prüfling ab. Es wird davon ausgegangen, daß die Wärme nur an den Flächen in die Probe gelangt bzw. diese verlässt, an denen auch der elektrische Strom ein- bzw. austritt, da die übrigen Oberflächen gegen Strom- und Wärmefluss isoliert wurden. Die planen Enden des Messstabes wurden bei konstanter Temperatur gehalten und als isotherme bzw. äquipotentiale Flächen angesehen.

Резюме — Метод определения термической проводимости зависит от соотношения между максимальной температурой (θ_m), достигнутой при данном токе и разностью потенциалов (V) в токонесущем образце. Принято, что теплота проникает и покидает образец только через те поверхности, через которые подводится и выводится также и ток, а другие поверхности являются изоляторами теплоты и электричества. Принято также, что плоскостные концы электрода должны быть изотермическими и эквипотенциальными поверхностями, выдержанными при постоянной температуре. При измерении удельной теплоемкости накладывается однородная постоянная джоулево тепла. Начальный наклон θ_m в зависимости от времени, не считаясь с тепловыми потерями, обратно пропорциональный теплоемкости. Измерения были проведены на образце Ni–Cr сплава при температурах до 360°.