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Influence of Radiation Losses on Thermal Conductivity Determination at Low Temperatures[1]

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The thermal conductivity of electrolytic iron has been measured in the temperature range from 100 to 390 K. Electrolytic iron is a standard material for the measurement of thermal conductivity. The thermal conductivity was measured on a commercial device Thermal Transport Option (TTO) of a Physical Properties Measurement System (PPMS) produced by the Quantum Design company. The temperature gradient on the sample was determined using small highly accurate Cernox chip thermometers. The thermal conductivity of the standard material showed higher values than those cited by NIST for the temperature range from 100 to 390 K (NIST's "Report of Investigation" for SRM 8420). The maximum deviation reached 30% at 390 K. Detailed analyses of the measured data and of the commercial software of the measuring device revealed that the large differences resulted from radiative losses of the interior parts of the device. The determination of the radiative losses takes into account the sample geometry, contacts, and cooling part of the device, and these differences in the thermal conductivity values were substantially reduced after accounting for these losses.

KEY WORDS: electrolytic iron; four-contact method; radiative losses; thermal conductivity.

1. INTRODUCTION

In longitudinal heat flow methods, the experimental arrangement is designed such that the flow of heat is only in the axial direction of

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a rod sample. The radial losses from the sample surface are prevented or minimized and evaluated. Under steady-state conditions and assuming no radial losses, the thermal conductivity is determined by the following expression from the one-dimensional Fourier-Biot heat-conduction equation,

$$\lambda = -\frac{q\Delta x}{S\Delta T} \tag{1}$$

where λ is the average thermal conductivity corresponding to the temperature $T_{\text{sample}} = \frac{1}{2}(T_1 + T_2)$, $\Delta T = T_1 - T_2$, q is the rate of heat flow, S is the cross-sectional area of the sample, and Δx is the distance between points of temperature measurements for T_1 and $T_2 (T_1 > T_2)$. The accuracy of the thermal conductivity obtained is determined not only by the accuracy of the measurement of the parameters in Eq. (1), but also by the overall design of the apparatus. There is a set of recomendations and requirements [1] to obtain thermal conductivity values with minimum errors.

The Physical Properties Measurement System (PPMS) (Quantum Design, USA) is an instrument where the thermal conductivity of a solid material is measured on small samples by the linear flow method at temperatures from 2 to 400 K. The sample, heater, cooler, and thermometers are placed in an evacuated chamber so that the heat losses from the periphery of the sample are purely radiant. Below 100 K the radiative heat losses are usually negligible and Eq. (1) holds. However, from 100 to 400 K the radiative losses cannot be neglected. In the instrument, the radiation losses are reduced by a suitable selection of the L/S ratio for the sample (where L is the length of the sample) and by a correction on the radiation losses from the sample. However, both conditions are not adequately satisfied, and the thermal conductivity values at 400 K for the metal samples were found to be about 20 to 30% higher than reference values. To obtain minimum errors during determination of the thermal conductivity further corrections on the radiative losses should be made. The analysis of this problem and the development of a new correction are the aims of this work.

2. EXPERIMENTAL

The thermal conductivity was measured on the Physical Property Measuring System, Thermal Transport Option (TTO) from Quantum Design (USA). The TTO system includes two measurement modes, a continuous measurement mode and a single-step measurement mode. Our results were obtained by measurements in a single-step mode, which uses a stationary, linear flow method. The thermal conductivity is determined by

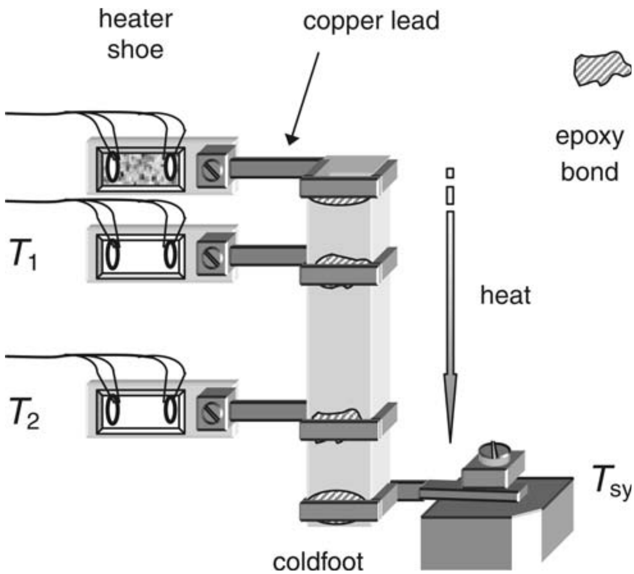

Fig. 1. Schematic diagram of the arrangement of the sample, heater and cooler, and both thermometers.

applying heat from the heater shoe in order to create a user-specified temperature difference between the two thermometers. A schematic diagram of the measurement parts is shown in Fig. 1.

The heater shoe assembly contains a resistive heater chip, and the temperature shoe assemblies contain a Cernox 1050 thermometer. The shoes are made of copper and coated by gold film, and the heater chip and/or thermometers are protected by an Al_2O_3 cover. Each shoe-type heater or thermometer is individually serialized. The copper isothermal radiation shield screws into the base of the puck and is designed to minimize radiation between the sample and environment. A copper shield plate is also placed between the sample stage and the PC board sockets to minimize radiation effects.

The thermal conductivity of three samples of electrolytic iron was measured in temperature range from 100 to 390 K. The samples were prepared from material obtained from the National Institute of Standard and Technology. The dimensions of the sample was 12 mm × 1 mm × 1 mm; Δx was about 7 mm. The connection between shoes and the sample was made from Cu contacts with gold coating, which were connected to the sample with silver-filled epoxy. The measurement was performed in high vacuum (10^{-4} Pa) to make thermal conduction by the residual gas negligible. The initial value of the power was obtained from the continuous measurement mode where the optimum heating power is determined. The uncertainty of the thermal conductivity measurements, as specified by the supplier, is about 5%.

3. RESULTS AND DISCUSSION

The thermal conductivity using the PPMS device is determined from a thermal conductance K and geometrical parameters as $\lambda = K(\Delta x/S)$. The thermal conductance is obtained as $K = P/\Delta T$ where P is the heat flow through the sample. Since the heat flux cannot be measured directly, the heat conducted throughout the sample is estimated as the power (I^2R) dissipated in the heater resistor minus losses due to radiation. Thus, the conductance is determined as follows:

$$K = \frac{I^2R - P_{\text{rad}}}{\Delta T} - K_{\text{shoes}} \tag{2}$$

K_{shoes} is the thermal conductance of the shoe assemblies, and P_{rad} is the radiant loss of heat. In the PPMS device only radiative losses from the sample are considered. They are determined using the following relation:

$$P_{\text{sample}} = \sigma_{\text{T}} \frac{A}{2} \varepsilon \left(T_1^4 - T_{\text{sys}}^4\right) \tag{3}$$

where A is the total sample surface area, ε is the emissivity of the radiating surface, T_1 is the temperature of the hot thermometer, T_{sys} is the lowest temperature of the system, and σ_{T} is the Stefan-Boltzmann constant. The factor of $1/2$ in the equation is due to the approximation that only one-half of the sample surface is radiating at the hot temperature, while the second half is at the cold temperature. It is difficult to estimate accurately the radiative heat losses, and the expected error in the measured thermal conductance since radiative losses above 300 K may be on the order of $\pm 1\,\text{mW}\,\text{K}^{-1}$.

Figure 2 shows the temperature dependence of the thermal conductivity for electrolytic iron (NIST standard material). It can be seen that the measured values are higher than the recommended values [1] by as much as about 30% at 390 K. A detailed analysis of this difference demonstrated the major problem in this measurement: neglect of the radiative losses from the hot parts of the device during measurement at the highest temperatures. It can be seen from Fig. 1 that the heater has maximum values of temperature for the whole system and, as result, the maximum heat

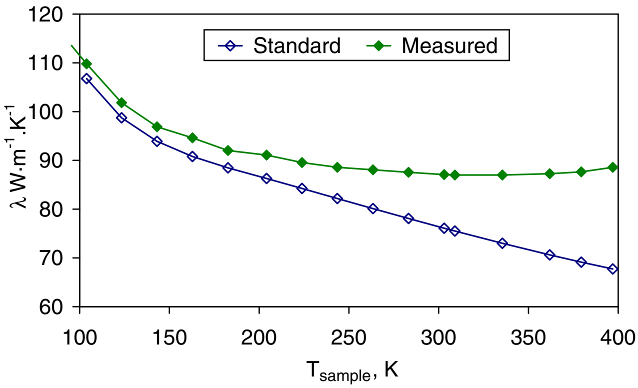

Fig. 2. Temperature dependence of the measured and recommended thermal conductivity for the electrolytic iron.

loss due to radiation. However, these heat losses as well as further thermal losses from all parts, where the temperature is higher than T_{sys}, are not included in the calculation of the thermal conductivity as provided by the instrument supplier. As a consequence of this fact, a higher heating power is used for the conductance calculation than in the actual experiments, and therefore, the measured thermal conductivity is higher (see Eq. (2)). Without an additional correction for radiation heat losses, the PPMS device is not reliable for measurements of the thermal conductivity at higher temperatures.

For an evaluation of the correction, a numerical model of the measuring process was developed to determine the heat losses due to radiation of the sample and system components, especially the heating and thermometer shoes. The numerical model was realized using the computational system Cosmos/M from the Structural Research & Analysis Corp. (SRAC). The Cosmos/M system solves the general heat transfer equation,

$$c_p \rho \frac{\partial T}{\partial t} = \nabla(\lambda \nabla T) \tag{4}$$

by a finite element method (c_p is the specific heat, ρ is the density, and t is the time). Tools for geometry and mesh generation, definition of boundary conditions (radiation, convection, heat generation, etc.), material properties, and/or other analysis parameters are included in the software.

The model is based on the arrangement of the measuring part described in Fig. 1. Steady-state thermal analyses were performed using temperature dependent material thermal properties. The values of the thermal properties (thermal conductivity, density, specific heat) were obtained from the literature: electrolytic iron sample [2–4], copper for the leads and heater/thermometer shoes [3–5], and Al_2O_3 for the heater/thermometer cover on the shoes [6–8]. The heat generation boundary condition on the heating shoe was the same as the heater power indicated by the measurement system. The heat conduction boundary condition (the heat transfer coefficient is determined to be about $20\,\mathrm{kW\,m^{-2}\cdot K^{-1}}$) is based on the thermal resistance between the cold-foot and the system (T_{sys}). It was found that this parameter can influence the maximum temperature rise, but it has only a small influence on the temperature difference ΔT.

Two cases were analyzed. The first one was solved for the radiation boundary condition set on the sample surface only. Such an approximation is used in the PPMS device for determining the thermal conductivity. The second one was solved for the radiation boundary condition on each surface of the sample and measuring system components (shoes and leads). The ambient temperature is assumed to be the temperature of the system T_{sys}. The emissivity was 0.3 for the iron sample, 0.03 for the copper leads and shoes covered by a thin Au-film, and 0.75 for the Al_2O_3 cover of the heater and thermometers.

The results of the thermal analysis using a numerical model Cosmos/M [m.s.1] are expressed as the temperature dependence of T_1 and T_2. [m.s.2] Comparisons of the calculated values of ΔT and the PPMS values when only the radiation of the sample, shoes and leads are considered can be seen in Fig. 3. The agreement between the calculated and PPMS values is very good. Figure 4 shows the temperature dependence of only the calculated values of ΔT for cases when radiation of the sample and the interior parts of the instruments is considered. These values of ΔT

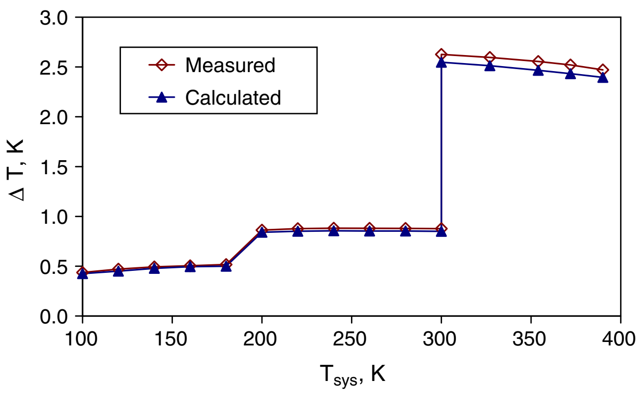

Fig. 3. Temperature dependence of the measured and calculated ΔT (during the calculation, the radiation of the sample and measuring system components is considered).

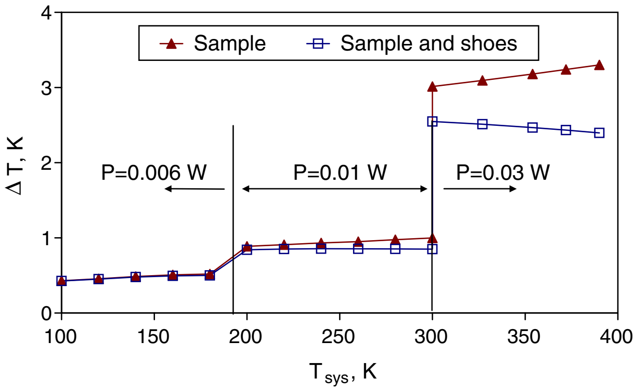

Fig. 4. Temperature dependence of the calculated ΔT.

were used for the calculation of the radiative losses using the following relation:

$$P_{\text{rad}} = I^2 R - \lambda S \frac{\Delta T}{\Delta x} - K_{\text{shoe}} \Delta T \tag{5}$$

The results are shown in Fig. 5. Three heating powers were used in obtaining these results (the same values of heating power were used during the measurements). The radiative losses are nearly nine times higher at 390 K for the case when radiation of all the parts of the measuring device is included in [m.s.3] the calculation than for the case when only the sample radiation is considered. This result is not surprising because the surface area of the shoes and leads (180 mm^2) is appreciably larger than the surface area of the sample (43 mm^2). Moreover, the highest temperature of this system is not at the hot thermometer (see Eq. (3)) but at the heater shoe. As expected, the radiative losses are strongly dependent on the heating power.

Using Eq. (2) we can determine the thermal conductivity from the PPMS values and P_{rad} from the sample and interior parts of the device. The corrected values of the thermal conductivity are presented in Fig. 6. These values are about 5% lower than the recommended values. We assume that there are additional effects influencing the thermal conductivity determination such as, for example, the heat losses in leads, radiation of the interior parts among themselves, etc. The influence of these effects on the thermal conductivity values may be removed using a correction factor. This factor $f = 1.03$ was obtained from the difference between the recommended and measured thermal conductivity values.

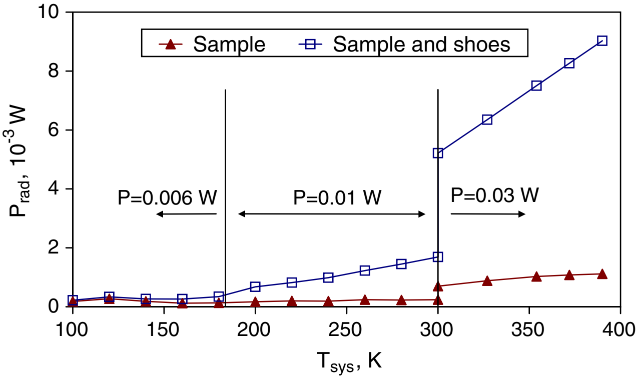

Fig. 5. Temperature dependence of the calculated radiative losses.

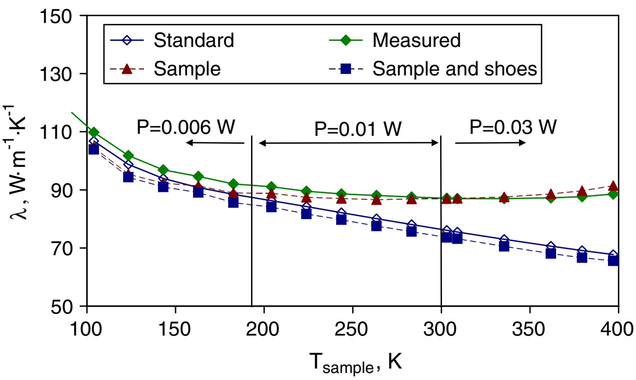

Fig. 6. Temperature dependence of the thermal conductivity of the electrolytic iron; measured, recommended, and corrected values are shown.

The use of both corrections is so far limited to materials with a thermal conductivity near the thermal conductivity of the used sample-electrolytic iron. The study of this problem for materials with lower and higher thermal conductivities will be the subject of future research. The dependence of both corrections on the heating power will also be studied.

4. CONCLUSIONS

The thermal conductivity of the electrolytic iron was measured at temperatures from 4 to 390 K. The measured values were higher than those recommended by NIST. The analysis showed that this difference is due to the radiation losses from the interior of the measuring part.

Without further correction, the PPMS device cannot be used for measurements of the thermal conductivity at higher temperatures because the error reaches values as large as 30% at 390 K. A numerical modeling approach was used to calculate the radiation losses; the Cosmos/M computational system uses a finite element method. The thermophysical parameters of all materials were used during this calculation. The radiation losses are nine times higher at 390 K than the losses considered by the measuring device supplier. The agreement between measured and recommended values was about 5% by the application of these radiation losses during the calculation of the thermal conductivity from measured temperatures.

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