# DETERMINATION OF THE THERMAL CONDUCTIVITY OF AMORPHOUS METALLIC ALLOYS

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*Two techniques are suggested for identifying the temperature dependence of the thermal conductivity of a new class of materials. The results have been obtained for the metallic glass* $Co_{77}Fe_4Cr_7Si_8B_4$.

Amorphous metallic alloys or, as they are also called, metallic glasses (MG), represent a new class of materials whose systematic study began in the last 10-15 years. Investigations of the electric, magnetic and mechanical properties of MG have demonstrated the exceptional potential of these materials for manufacturing magnetic wires of transformers and magnetic heads, corrosion-resistant protective coatings, structural reinforcements, cutting tools, etc. There are virtually no losses for remagnetization of some MG, their corrosive stability is by several orders higher than that of special stainless steels; the high strength and hardness combine with a high rigidity and elasticity. Since MG specimens can be obtained only in the form of thin foils or film coatings, experimental investigation of their thermophysical characteristics (TPC) and thermodynamic properties was virtually not carried out, because the familiar methods of determining the above-indicated properties do not always allow one to obtain reliable results for such specimens. Therefore, it seems worthwhile to look for nontraditional ways of identifying TPC which would be appropriate for such specific bodies as MG specimens. One of these ways is the employment of the procedures used for solving inverse heat conduction problems (IHCP), in particular, the methods of solving internal IHCP's [1] which allow one to determine the TPC of materials from information obtained experimentally for the temperature field of the specimens made from these materials.

We tested the indicated way of identifying TPC when we determined the temperature dependence of the thermal conductivities for several types of amorphous metallic alloys. In doing so, we used nontraditional techniques not only in the computational portion of the study, but also at the stage of thermophysical experiment.

Investigation of the thermal conductivity $\lambda$ of MG was carried out on a universal low-temperature thermophysical apparatus (ULTA) which had been modified for the purpose and which was one of the most perfect adiabatic calorimeters with the operational range of temperatures from 4.2 to 373 K.

The thermal conductivity measurements by the steady-state method, allowed for on the ULTA, is extremely difficult to realize technically in view of the fact that one can ensure a steady thermal regime for a specimen only in the case when heat removal from the "cold" surface of the specimen is closely controlled. However, in the case of measurement errors greater than unity, it is more expedient to use the nonsteady-state method which is more easily realizable.

The studied material, in the form of a foil strip, is rolled up into a coil and fixed at the ends faces of two massive (as compared with the specimen) cylindrical body-heaters 1 and 2 (Fig. 1). The thermal contact between the heaters and specimen 3 is ensured by the careful treatment of contracting surfaces and by applying a thin layer of a heat conducting lubricant (graphite with vacuum lubricant or diffusion oil) on these surfaces before assembly. Upon assemblage, tight pressing is provided by tightening thin capron threads.

At the places of contact with the specimen, the junctions of a differential copper–constantan thermocouple were positioned in the body-heaters. The thermocouple measured the temperature difference on the specimen. The body-heaters are surrounded by internal thermostatic control screens 4 whose temperatures are kept equal to those of the corresponding body-

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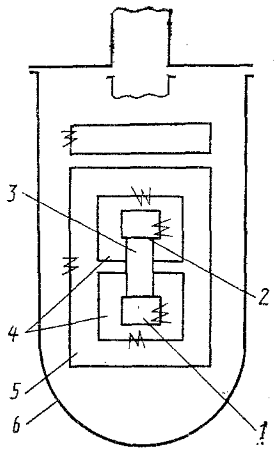

Fig. 1. Schematic of the thermal conductivity measuring cell.

heaters. The external thermostatic control screen 5 secures the minimum possible temperature difference between itself and the internal screens and takes over the major portion of heat exchange with the vacuum chamber cap 6. To reduce convective heat transfer, the measuring module is exhausted to a pressure of not higher than $5 \cdot 10^{-5}$ mm Hg.

The measured values in the experiment were the absolute temperature of the body 1 $T_1$ , the difference of temperatures $\Delta T$ between bodies 1 and 2, the powers $P_1$ and $P_2$ of body-heaters 1 and 2, the time between measurement $\Delta\tau$. The mean cross-sectional area of the specimen S was found by measuring its volume and actual length. Formally, the distance between bodies 1 and 2 is considered to be the specimen length $l$.

To calculate the thermal conductivity from the familiar formulae of the method of monotonous heating, the data on the heat flux in the specimen itself are required. This can be both the heat flux density at the place of specimen contact with body-heater 1 $q_1$ and with body-heater 2 $q_2$. When $\lambda(T)$ is calculated by the methods of inverse problems, it is desirable to have both of these values to assign the boundary conditions (BC). Since the measurement of these values is not allowed for in the above-described apparatus, we applied their indirect determination by carrying out a special test in the same apparatus but without a specimen.

In the experiment with a specimen, an electric heater for body 1 is a heat source which monotonically heats the system. Its electric energy $W_1$ is spent for heating the entire system and for heat exchange with the thermostatic screens, i.e.,

$$W_1 = Q_1' + Q' + Q_2' + Q_1'' + Q'' + Q_2'',$$

where $Q'$, $Q_1'$, and $Q_2'$ are the quantities of heat lost (obtained) by the specimen, body 1, and body 2, respectively, owing to incomplete adiabaticity of the system; $Q''$, $Q_1''$, and $Q_2''$ are the quantities of heat spent raising the temperature of the specimen, body 1, and body 2, respectively.

The heat supplied to the specimen through body 1 is determined by the expression

$$Q_1 = W_1 - Q_1' - Q_1'', \tag{1}$$

whereas that removed from it by heat conduction through body 2 – by the expression

$$Q_2 = Q_2' + Q_2''. \tag{2}$$

Having measured the heat capacities of the specimen and bodies 1 and 2 in separate tests, it is possible to calculate the quantity of heat $Q'' + Q_1'' + Q_2''$ spent during any length of time for system heating. To determine $Q_1(T_1)$ and $Q_2(T_1)$, a test is conducted in which, although the specimen is absent, all the geometric parameters of the main experiment and the depth of the vacuum are maintained. The rate of heating and the remaining thermal quantities are maintained the same as in the main experiment due to the switching-in of the heater for body 2. The powers of the heaters are controlled automatically in the course of the experiment with the aid of a computer.

In the experiment with an empty cell (without a specimen), the entire heat $W_1^n$ emitted by the heater of body 1 is spent for heating this body and for heat exchange with the surrounding screens and body 2. The heat $W_2^n$ emitted by the heater of body 2 is distributed similarly. We shall assume that the heat losses in experiments with a specimen and without it are equal, though there are differences in the heat transfer conditions owing to the screening of a portion of the surfaces of bodies 1 and 2 in radiative heat exchange between them. Taking into account the small temperature differences and insignificant screening area, these differences can be neglected. Then, the sought heat losses from the surfaces of bodies 1 and 2 are found from relations

$$Q_1' = W_1^{n} - Q_1'', \quad Q_2' = W_2^{n} - Q_2''.$$

Equations (1) and (2), with the above relations taken into consideration, acquire a form convenient for calculations:

$$Q_1 = W_1 - W_1^{n}; \quad Q_2 = W_2^{n}.$$

Proceeding to determination of the heat flux densities at the places of specimen butting with body 1 ($q_1$) and 2 ($q_2$), we shall consider two important circumstances: first, in the case at hand all of the measured and computed quantities can be regarded conveniently as functions of $T_1$ (of the temperature of body 1, "temperature of assignment"), which in turn is a discrete function of time; and, second, the discrepancy between the rates of heating body 2 in the experiments with a specimen and without it is possible. We obtain

$$q_1(T_1) = \frac{P_1(T_1) - P_2^{n}(T_1)\,k(T_1)}{S},$$

$$q_2(T_1) = \frac{P_2^{n}(T_1)\,k(T_1)}{S},$$

where $P_1$ is the power of the electric heater for body 1 in the experiment with a specimen; $P_1^n$, $P_2^n$ are the powers of the heaters for bodies 1 and 2 in the experiment without a specimen; k is the factor which takes into account the different rate of heating in the experiments with a specimen and without it, $k = (dT_2/d\tau)/(dT_2^n/d\tau)$ ($T_2$, $T_2^n$ are the temperatures of body 2 in the experiments with a specimen and without it).

Based on the heat flux densities calculated in this way and on the linear heat conduction model, we obtain two formulae for calculating the thermal conductivity:

$$\lambda^{\mathrm{I}}(T_1) = \frac{q_1(T_1)\,l}{\Delta T(T_1)}, \quad \lambda^{\mathrm{II}}(T_1) = \frac{q_2(T_1)\,l}{\Delta T(T_1)}.$$

The values of the thermal conductivities $\lambda^{\mathrm{I}}$ and $\lambda^{\mathrm{II}}$ are aggravated with systematic error components due to the fact taht the specimen heat capacity not taken into account in this case and to the heat losses from its surface into the ambient medium. These systematic error components are compensated for in great part if the thermal conductivity is found as the arithmetic mean of two values, i.e.,

$$\lambda(T_1) = (\lambda^{\mathrm{I}}(T_1) + \lambda^{\mathrm{II}}(T_1))/2. \tag{3}$$

In this case the errors are compensated because the neglected quantity of heat spent for heating the specimen and the losses from its surface will overshoot the result when calculating $\lambda^{\mathrm{I}}(T_1)$ and undershoot it when calculating $\lambda^{\mathrm{II}}(T_1)$, since the quantity of heat $Q_1(T_1)$ supplied to the specimen from body 1 is greater than the quantity $Q_2(T_1)$ removed to body 2 (provided the heat losses from the specimen surface are small as compared with the heat spent for its heating). The remaining systematic error components in the measurement of $\lambda(T)$ due to the nonideal contact between the specimen and the clamps, due to the errors in the measurements of the length and cross-sectional area of the specimen, etc., are to be related to the noneliminated remainder of the systematic error and are to be evaluated experimentally while qualifying the procedure of measurements. The experiments have shown that elimination of a portion of the systematic error in calculations by formula (3) is effective only for the functions $T_1(\tau)$ being close to linear ones. In the case of a more or less strong nonlinearity of the functions $\lambda(T)$ and $c(T)$, which is responsible for the nonlinearity of $T_1(\tau)$, the method described leads to errors as large as tens of percents. The sole way for these errors to be eliminated is the use of the nonlinear heat conduction equation with the subsequent solution of the internal IHCP.

To process the data of the experimental thermophysical investigation of MG, of all the methods used for solving internal inverse problems, we have selected that one which relates to the so-called extremal methods of solution. In contrast to nonextremal (mostly analytical) methods, in which the temperatures measured are identified with the mathematical model potentials and the inverse problem solution is reduced to the solution of a system of differential or algebraic equations in heat transfer pa-

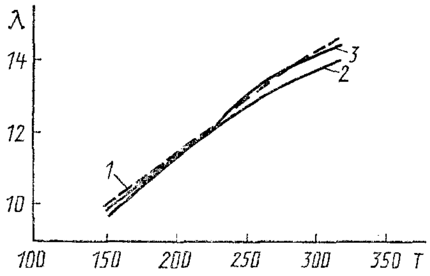

Fig. 2. Values of $\lambda$ (W/(m ·K)) for 12Kh18N10T steel as a function of T(K).

parameters, in extremal methods the difference (residual) between the measured and predicted temperatures is minimized by a purposive selection of the unknown parameters in order to numerically realize the mathematical model of the heat conduction process observed. On the one hand, this allows one to consider rather complex heat-conduction problems, because numerical methods of solving direct problems have been rather well developed (and this is the key procedure in extremal methods as to the amount of computations). On the other hand, a consecutive controlled decrease in the residual makes it possible to apply diversified methods for solution regularization. The main principle of obtaining a stable solution is: a minimized residual must be commensurable with the error of the initial data.

The original tabulated information on the experimentally obtained temperatures, fluxes, and other time-dependent quantities is transformed into analytical functions of time by approximation methods. This needs to be done because the moments of measuring different values may not coincide with each other or may not correspond to the temporal layers in the numerical solution of the problem.

Moreover, approximation of input functions leads to their smoothing-out, which is an extra factor for ensuring the stability of a computational process.

The functions $q_1(\tau)$ and $q_2(\tau)$ obtained in this way are used as boundary conditions of the second kind when forming a mathematical model of the thermal process proceeding in a specimen, whereas the functions $T_1(\tau)$ and $T_2(\tau)$ serve as the reference functions in solving IHCP's. With the experimental conditions taken into account, the model has the following form

$$\frac{\partial}{\partial x}\left(\lambda(T)\frac{\partial T}{\partial x}\right)-q_V(x,\ T)=c_V(T)\frac{\partial T}{\partial \tau},\quad 0<x<l,\ \tau>0;$$

$$T(x,\ 0)=T_{\text{in}};$$

$$-\lambda(T)\left.\frac{\partial T}{\partial x}\right|_{x=0}=q_1(\tau),\quad -\lambda(T)\left.\frac{\partial T}{\partial x}\right|_{x=l}=q_2(\tau);$$

$$T(0,\ \tau)=T_1(\tau),\ T(l,\ \tau)=T_2(\tau),$$

where x is the space coordinate along the specimen axis; $T_{\text{in}}$ is the initial temperature of the specimen; $q_v$ the inner heat sink. The introduction of the function $q_v(x, T)$ into the heat conduction equation is associated with heat removal from the specimen surface as a result of radiative exchange with the screens and reradiations between the points at the inner cylindrical surface of the specimen being taken into account.

In addition to the reference values, boundary conditions, and geometric information, the original data involve the functions c(T) and some values of the parameters which determine $\lambda$(T) and which are selected on the basis of the evidence concerning the thermal conductivity of the materials similar in chemical composition to that studied. The unknown function $\lambda$(T) is assigned in the form of the sum of the Chebyshev series whose coefficients are the unknown parameters.

To numerically simulate the phenomenon of heat conduction, we carried out the finite-difference approximation of the system of differential equations entering into the mathematical model of the phenomenon. The heat conductions between the nodes correspond to $\lambda[(T_i + T_{i+1})/2]$ (i is the number of the nodal point), whereas the accumulation of heat for the time step is determined by the heat capacity $c[(T_j + T_{j+1})/2]$ (j is the number of the temporal layer). In view of this, the simulation procedure represents a program for solving a system of nonlinear algebraic equations with a band matrix. In the given one-dimensional case, the band width is equal to 3. The nonlinear nature of the equations accounts for the necessity to conduct com-

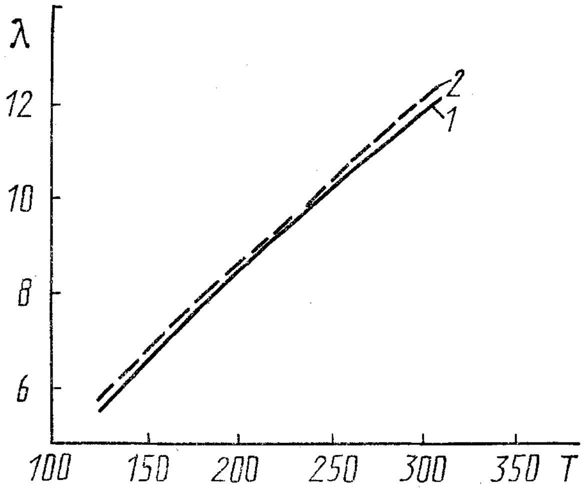

Fig. 3. Value of λ (W/(m·K)) for amorphous alloy $Co_{77}Fe_4Cr_7Si_8B_4$ as a function of T (K).

putations with iterational verification of the system coefficients. If the results of two successive solutions of the system of finite-difference equations differ from one another by less than $\delta \cdot 0.1$ ($\delta$ is the error of temperature measurement), the iterations are stopped. Usually, no more than 10 iterations are required. If there are more than 10 iterations, the residual is increased by adding a penalty function to it. The penalty is also assigned when $\lambda(T)$, if only at one point of the temperature range $[T_{min}, T_{max}]$, becomes smaller than or equal to zero. The minimum $T_{min}$ and maximum $T_{max}$ values of the temperature are determined by the initial data of the problem.

The minimization of the residual as a function of the parameters of analytical representation is an essential point underlying the solution of IHCP. A direct result of the solution are those values of the indicated parameters with which the residual is minimal and satisfies the regularization condition (i.e. the minimal residual is not smaller than the temperature measurement error).

To check the validity of the proposed approach, we carried out identification of the thermal conductivity of 2Kh18N10T stainless steel. In Fig. 2 the temperature dependence of the thermal conductivity of very familiar material is given plotted on the basis of reference data [2] (curve 1), those calculated from Eq. (3) (curve 2), and those obtained by solving IHCP (curve 3). Comparison of these results allows a conclusion about the possible use of the described experimental apparatus and procedures for identifying the thermal conductivities of solids. The error of identification by Eq. (3) and by solving the IHCP did not exceed 10 and 5%, respectively. Especially large deviations from the standard data were observed with the use of traditional equations within the temperature range of 220-270 K. This is attributed to the fact that the system of automatic regulation of specimen heating within this temperature range did not ensure linearity of the function $T_1(t)$, with curve 3 in Fig. 2 approaching the standard relation closer than curve 2, since the result of the solution of IHCP depends little on the heating rate.

The conclusions drawn when conducting a check experiment and processing its results were taken into account for identifying the temperature dependence for amorphous alloy $Co_{77}Fe_4Cr_7Si_8B_4$. The specific features of a sample of this material, which at present can be produced only in the form of a miniature thin tape preventing its investigation by traditional heat conduction measurement techniques, are rather fully taken into account in the apparatus suggested. Moreover, a substantial temperature dependence of the thermal conductivity of MG is taken into account most correctly and is determined when processing the test data by the IHCP methods based on the nonlinear model of heat conduction. In view of this, we regard that the most reliable result of the study of $\lambda(T)$ for $Co_{77}Fe_4Cr_7Si_8B_4$ is the dependence presented by curve 1 in Fig. 3. It has been obtained by solving the IHCP.

Curve 2 reflects the results of heat conduction calculations by Eq. (3). The greatest difference in the values of λ obtained by these two techniques is observed, as should be expected, at temperatures exceeding 220 K when the thermometry results varied nonlinearly. This led to a higher error of calculations by Eq. (3).

In conclusion, it should be noted that reliable information on the thermal conductivity of amorphous metallic alloys makes it possible not only to carry out thermal engineering calculations of products made from these unique materials, but also to judge their operating characteristics. The thermophysical properties of MG are structurally sensitive physical quantities. In some cases they can be adopted as a foundation for developing the methods of nondestructive strength testing, magnetic, and other properties. The methods of solving IHCP help in increasing the accuracy of identification, especially required in this case, and to reduce the limits imposed on the mode of heating a specimen when conducting a thermophysical experiment.

## NOTATION

$\lambda$ is the thermal conductivity; T, temperature; P, power; $\tau$, time; $l$, S, specimen length and its cross-sectional area; q, heat flux density; Q, quantity of heat; W, electric current energy; k, factor accounting for differences in heating rates; c, $c_V$, mass and volumetric heat capacities; x, space coordinate; $q_V$, specific volumetric power of internal heat source; $\delta$, error of temperature measurements; $T_{min}$, $T_{max}$, limits of operational temperature range.

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