METHOD OF MEASURING THE THERMAL DIFFUSIVITY OF TRANSLUCENT MATERIALS OF HIGH TEMPERATURES

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Vestnik Moskovskogo Universiteta. Fizika,
Vol. 34, No. 6, pp. 41-47, 1979

UDC 550.362

We propose a method of measuring the thermal diffusivity of partially transparent materials at high temperatures. The method is based on the application of the method of plane temperature waves for various thicknesses of the objects under study in the limit of an optically thin layer over a wide interval of heating frequencies. The method is tested by measuring the thermal diffusivity of high-quality fused quartz, which has an appreciable photon component of the heat transfer.

The study of the thermal properties of translucent materials is one of the important, though difficult, problems in high-temperature thermophysics. During measurements on objects of this type one encounters, along with the usual difficulties accompanying high-temperature experiments, additional difficulties due to radiative heat transfer, which is characteristic of some dielectrics and semiconductors. The fundamental difference between radiative (photon) transfer of thermal energy and the usual (conductive) transfer makes it impossible to design an unambiguous experiment. One can be convinced of this by examining Fig. 1, which shows the temperature dependence of the thermal conductivity λ of fused quartz in the high-temperature region as obtained by various authors. It is seen from the figure that the data of different authors are substantially different. The main reason for this disagreement is the partial transparency of fused quartz in the infrared region of the spectrum. The effect of the optical properties on the results of the thermal-conductivity measurement requires making specific allowance for the condition of the surface, for the characteristic dimensions of the sample, and, in the course of the experiment, for the magnitude of the temperature gradient created in the sample. If these conditions are not identical, there will be a substantial disparity of the data, exceeding the error of measurement on the different apparatus.

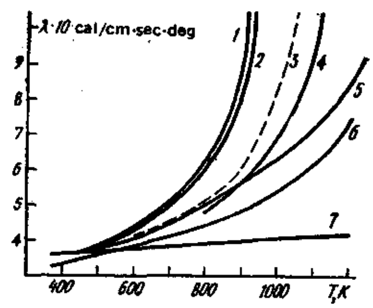

Fig. 1. Thermal conductivity of fuzed quartz. Curves 1-6 are constructed according to the data of [1-6] respectively, 7 is λ_{bkgd} from the data of [7].

We note that in most cases the experiments to measure the thermal conductivity of partially transparent materials, including fused quartz, reach temperatures of 1000 K. This is just the temperature at which radiative heat transfer can become important.

It is known that in radiative heat transfer in a material, the size of the radiative component of the thermal conductivity (diffusivity) can depend on the optical properties of the boundary surfaces and the characteristic dimensions of the sample. Such a dependence arises in the case when the mean free path of a photon is comparable to the characteristic size of the object (or rather, with the optical thickness, which is the product of the characteristic size of the sample and the absorption coefficient). The radiative thermal conductivity is small for optically thin samples and increases as the thickness increases. The effect of the thickness of the sample on the size of the radiative component of the thermal conductivity can be used to determine this component and to find the average (integrated) absorption coefficient of the material.

Thus, to determine the radiative heat transfer in measurements on optically thin objects, one should obtain data for various thicknesses. The extrapolation of these data to zero thickness in accordance with the ideas about radiative heat transfer should give the phonon component.

This approach to the separation of the phonon component of the thermal conductivity is not fundamentally new. We know of two papers in which the effect of an optically thin layer was used to study the role of radiative heat transfer in liquids [8,9]. However, as far as we know, this approach has not been used to investigate the analogous phenomenon in solids. The main reason, though apparently not the only one, is that most of the methods of measuring the thermal conductivity are not used to advantage in experiments on thin (fractions of millmeter thick) samples in the high-temperature region. (The temperature intervals in which the radiative effect is manifested are significantly different: in liquids, 400 K; in solids, 1000 K and higher. Furthermore, in liquids the absolute magnitudes of the thermal conductivity are lower than in solids).

Table 1

T,K	ε	D	ΔT (computer)	ΔT (gradient)
1000	0,112	0,066	93,6	92,1
1300	0,148	0,043	85,0	83,2
1500	0,192	0,035	75,2	75,9

In using the approximation of an optically thin layer, as in the case of an optically thick layer, it is assumed that the gradient representation for the heat flux is valid under conditions of combined conductive and radiative heat transfer [10,11]. For example, a calculation of the temperature gradient for a quartz plate 2.4 mm thick according to the gradient representation and as obtained by solving the integrodifferential heat-conduction equation using a computer has led to the results given in Table 1 [10] (ε and D are the degree of blackness of the surface and the optical thickness of the plate, respectively).

It is seen from the table that the temperature drops obtained by these two methods differ by not more than 2.5%. This example is typical for optically thin media.

The proposed method of measuring the thermal diffusivity of partially transparent materials involves doing an experiment over a wide range of heating frequencies for various sample thicknesses (within the limits of optically thin layers) and analyzing the dependence of the data on these factors. It will be helpful to illustrate the method with a specific example. The most suitable material for this is fused quartz, a typical translucent material with an appreciable radiative component. The samples chosen for study were cut in the shape of disks 12 mm diameter with thicknesses ranging from 0.2 to 2 mm. Prior to the measurements the samples were coated with a thin metallic layer. The coating provided a stable degree of blackness and electrical conductivity of the sample surfaces. The success of the measurements of the thermal properties of partially transparent materials depended on the coating used. Three different types of coatings were tried in the experiment: 1) plasma spray-coating with a high-melting metal, 2) coating with pyrolytic graphite; 3) coating with high-melting metals by a diffusion method. The experiments showed that the last was the most successful of these. A film of thickness ~5 μm almost completely precludes the direct penetration of radiation through the translucent object and gives a stable thermal contact (sample - film) all the way up to its melting temperature. The metallic conductivity of the sample surfaces was necessary because the apparatus employed periodic electron heating.

The apparatus used was essentially the same as that described in Ref. [12]. The thermal diffusivity was measured in the usual way for periodic-heating methods. The heating frequencies were varied from 0.5 to 50 Hz for the thicknesses of fused quartz indicated above. The lower limit was determined by the capabilities of the V5-2 device which recorded the phase of the thermal oscillations of the surface of the sample. The measured a_{ef} were functions of the frequency, a fact which is explained by a qualitatively new mechanism of energy transfer - the radiative mechanism. In reliably developed methods of measurements on standard materials (Mo or W), such a dependence is absent. Consequently, for such a case the mathematical relationships obtained under the assumption that there is only conductive heat transfer are not applicable.

As an example, we show in Fig. 2,a the dependence of the phase shift of the temperature oscillations on the heating frequency as obtained in high-quality quartz at a thickness of 0.69 mm for various temperatures. It follows from the

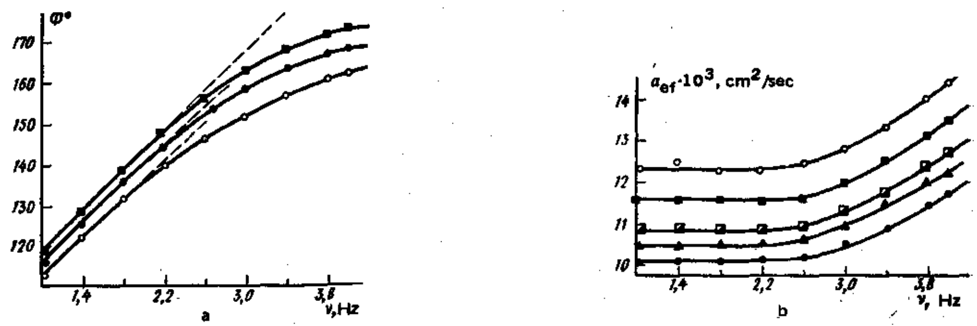

Fig. 2. Phase shift as function of frequency (a): L = 0.69 mm, T = = 1225 (■), 1315 (●), and 1550 (○) K; the straight parts of the lines and the dashed lines are the calculated functions for a purely conductive transfer. Frequency dependence of the effective thermal diffusivity (b): L = 0.69 mm, T = 1255 (●), 1275 (▲), 1315 (◪), 14525 (■), 1550 (○) K.

figure that the phase shift increases almost linearly with increasing frequency and then grows less rapidly as the dependence of Φ on ν begins to differ substantially from that calculated with a purely conductive heat transfer. Thus, a behavior like that shown in Fig. 2,a indicates the presence of radiative heat transfer. Analogous behavior was also found for thicknesses of 0.2, 0.3, 0.79, and 0.97 mm. It is appropriate here to point out a basic regularity in the behavior of $\Phi = f(\nu)$: as the thickness of the object increases, the linear part broadens and shifts to higher frequencies (for example, for a thickness of 0.3 mm the region of operating frequencies was 4-12 Hz). An increase in the frequency of the periodic heating corresponds to an increase in the optical thickness and, hence, in the role of radiative heat transfer.

The increase in the role of radiative transfer with increasing frequency as observed in the experiment thus agrees with the conclusions of Refs. [12-15].

The thermal diffusivity evaluated according to the usual relations of the method of plane temperature waves is shown as a function of the heating frequency in Fig. 2,b. As was the case for the curves in Fig. 2,a, the curves in Fig. 2,b also consist of two parts: a region in which the thermal diffusivity does not depend on the frequency, and a region in which it increases markedly with increasing frequency. Such a behavior indicates that the temperature distribution in the presence of combined conductive and radiative heat transfer differs from the distribution for purely conductive heat transfer. It turns out that it is in the low-frequency region that the mathematical relations obtained for the periodic temperature distribution during conductive heat transfer in optically thin objects are valid for the case of combined heat transfer as well.

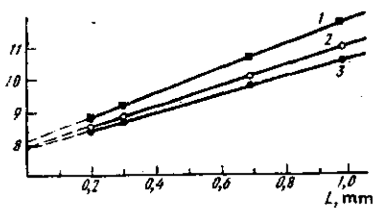

Fig. 3

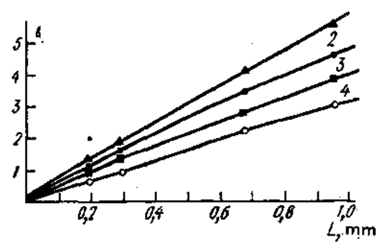

Fig. 4

Fig. 3. Extrapolation of a_{ef} to zero thickness. T = 1300 (1), 1225 (2), and 1150 (3) K. The scales on the vertical axes in Figs. 2, 3, and 4 coincide.

Fig. 4. Radiative component of the thermal diffusivity $a_{r.ef}$ of high-quality glass for various sample thicknesses. T = 1500 (1), 1400 (2), 1300 (3), and 1200 (4) K.

The measurements for fused quartz at various thicknesses show that the thermal diffusivity grows with increasing thickness and temperature. This is illustrated in Fig. 3, where the results of measurements of the thermal diffusivity of fused quartz are shown for thicknesses of 0.2, 0.3, 0.69, and 0.97 mm at three different temperatures.

The 1300 K isotherm in Fig. 3 was obtained for samples which were coated with molybdenum by the plasma method, while the 1225 and 1150 K isotherms were obtained for samples coated with tungsten by the diffusion method. The effective value of the thermal diffusivity for these temperatures decreases linearly as the sample thickness decreases. The extrapolation of the a_{ef} values to zero thickness gives the value of the phonon part of the thermal diffusivity, which amounts to $(7.9 \pm 0.2) \times 10^{-3}$ cm^2/sec. The extremely weak dependence of the phonon thermal diffusivity on the temperature (for $T \geq 1000$ K) for fused quartz can be explained on the basis of existing ideas about heat transfer in amorphous materials.

Using the value obtained for the phonon component of the thermal diffusivity, we separated out the radiative component for the various thicknesses and temperatures. The results are shown in Fig. 4. One can see that the radiative component is small for practically all the investigated thicknesses at lower temperatures and increases as the temperature is raised. The shape of these curves is reminiscent of the shape of the curves calculated from the formula for the radiative component of the thermal conductivity for an optically thin layer in the gray-body approximation [16]:

$$\lambda_{r.ef.} = 4\sigma n^2 T^3 \varphi(\alpha L, R), \qquad (1)$$

where σ is the Stefan-Boltzmann constant, n and α are the index of refraction and the absorption coefficient of the material, T is the absolute temperature, L is the sample thickness, and φ is a function of the optical thickness and the reflectivity of the walls of the sample R [16].

For R = 0 (black walls) this equation assumes the simple form:

$$\lambda_{r.ef.} = \frac{4\sigma T^3 n^2}{1+(3/4)\alpha L} L. \qquad (2)$$

Formula (1) contains two unknown quantities: α and R. To find them it is necessary to measure $a_{r.ef}$ for at least two thicknesses. Starting with the values of $a_{r.ef}$ for different thicknesses, we evaluated the average absorption coefficient of high-quality quartz for various temperatures.

The values of the refractive index was taken from [17]; the specific-heat data, from [18]. The thermal conductivity $\lambda_{r.ef}$ was evaluated from the values of $a_{r.ef}$ under the assumption that the phonon and radiative components of the thermal conductivity are additive.

Table 2

T,K	1000	1100	1200	1300	1400	1500	1600
α, cm^{-1}	(1,54)	(1,53)	1,53	1,53	1,53	1,54	1,55

The value of the absorption coefficient was chosen in such a way that the isotherm of the effective thermal diffusivity would agree with the values obtained from formula (1). The calculated results for several temperatures are given in Table 2.

It is seen in the table that the average absorption coefficient remains practically unchanged as the temperature increases. It should be stressed that in these calculations the refractive index was assumed to be constant, since its temperature dependence has not been studied experimentally. An insignificant increase in the refractive index (by, say, 0.2) can cause a to increase as the temperature is raised.

With the data on the average absorption coefficient of fused quartz, one can find the effective values of the radiative component using formula (1) and the "true" radiative thermal conductivity during the well known formula of Rosseland, which is valid for optically thick layers [13].

Comparison of the results of this paper with those in the literature is complicated by the fact that our measurements were taken at temperatures ~ 1200 K and higher. The experimental values of the average absorption coefficient at 1000 K agree with the results given in the monograph [10] for high-quality glass, which were obtained from spectral data on the absorption coefficient.

A similar comparison of the thermal-conductivity results shows that our data, when extrapolated to a temperature of 1000 K, lie above the data given in [10]. The reason is evidently that the data of [10] are effective values, which depend on the sample thickness and the particular optical properties of the surface.

In summary, it can be remarked that the method of plane temperature waves, when carried out over a wide spectrum of heating frequencies and for various sample thicknesses, provides information on the mechanism of heat transfer, on the values of the different components of the thermal conductivity (diffusivity), and on the average absorption coefficient in translucent materials and their temperature dependence as well, without requiring a theoretical examination of the corresponding heat equation, which does not have an exact analytical solution.

Analysis of the heat-conduction data on fused quartz at high temperatures indicates that quartz may prove useful in the development of methods for measuring the thermal properties of partially transparent materials, while at the same time it cannot be recommended as a reference material for studying the temperature dependence of the thermal conductivity of dielectrics.

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