THERMOPHYSICAL PROPERTIES OF QUARTZ GLASS

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UDC 536.21:666.192

Available experimental data on the thermal conductivity, specific heat, and thermal diffusivity of quartz glass over the temperature range 60-1100°K are generalized. Empirical equations describing the temperature dependence of these thermophysical parameters are presented.

Quartz glass (fused quartz) is widely used in thermophysical measurements for calibration and verification of thermal devices [1, 2]. Therefore, its thermophysical properties are constantly studied in order to refine their accepted values.

Thermal Conductivity. A large number of original studies [3-27] have been dedicated to measurement of the thermal conductivity of quartz glass, with results also published in several reviews [28-31]. To estimate the reliability of such data, the authors of [2, 28] applied the following criteria: 1) The data must be obtained by absolute methods; 2) the study must contain an analysis of systematic and random measurement error; 3) information on the origin of the specimens used must be presented; 4) the study must present the experimental data.

Table 1 enumerates the original studies and presents information needed to apply the first of the above criteria. As is evident from the table, we may remove from further consideration the results of [5, 6, 8, 9, 11, 12] and a portion of those of [15, 25], since these were obtained by relative measurements. These data were not included in the calculations of the present study. Moreover, our calculations do not include data from [16], since those were replaced by refined values in [17]. Criteria 2 and 3 were applied to the remaining data.

Knapp's study [13] does not satisfy the second criterion. It provides practically no analysis of error sources, one of which is temperature differences, which reached 1800°K over a thickness of 2.5 mm. Kingerey's study [25] fails by the second and third criteria. In this case the computation formula, derived for an Archimedean cylinder, is invalidly applied to specimens of elliptical form. There is no analysis of systemic error. Due to their complex form the specimens were prepared by pulverization of blocks of transparent quartz glass, i.e., they differed in their structure from the continuous specimens considered in the other cases.

The data of [10] do not satisfy the third criterion.

The data of the above studies were not processed further. Moreover, the thermal conductivity value obtained by Euken [7] at 100°C was also excluded, since the author himself doubted its validity. Detailed analysis of the remaining studies revealed that many of them did not satisfy the third criterion. Data on the purity of the material studied is presented only in [4, 15], and also [16-20, 22], where the specimens used satisfied GOST 15130-79 "Quartz optical glass. Technical specifications," which closely regulates the purity of the material. Despite this fact, we did not assign special significance to this criterion for the following reasons: 1) In the temperature range 90-1100°K, for a substance such as quartz glass, where the natural structure is chaotic, the thermal conductivity is sensitive to changes in structure and composition (impurities) to only an extremely small degree; 2) the greatest contribution to inaccuracy of the results obtained is at present produced by unconsidered systemic measurement errors.

Our calculations also excluded the results of [19], since they differ so sharply from other determinations of quartz-glass thermal conductivity at high temperatures.

All-Union Scientific-Research Institute of the Metrological Surface, Moscow; Applied Physics Institute, Academy of Sciences of the Belorussian SSR, Minsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 43, No. 6, pp. 960-970, December, 1982. Original article submitted December 9, 1981.

0022-0841/82/4306-1375$07.50

TABLE 1. Information on Studies of Thermal Conductivity of Quartz Glass

Author	Temperature range, °K				Temperature range, °K
	absolute	relative	stationary	nonstationary	
Euken, 1911 [7]	+		+		80—400
Barrat, 1914 [8]		+			300—400
Kaye and Higgins, 1927 [9]		+			350—500
Seeman, 1928 [10]	+	+	+		230—1230
Barrat, 1940 [4]	+	+	+		300—800
Knapp, 1943 [5]		+			400—1100
Berman, 1951 [3]	+		+		20—90
Kingery, 1955 [25]	+	+	+		300—1000
Gafner, 1957 [11]		+		+	303
Wray and Connoly, 1959 [13]	+		+		303—2100
Ratcliffe, 1959 [12]		+	+		130—350
Pustovalov, 1960 [26]	+		+		500—1300
Devyatkova et al., 1960 [14]	+		+		80—1100
Kuperin and Platunov, 1961 [23]	+			+	
Aleksenko, 1962 [6]		+	+		293
Vasil'ev, 1964 [24]	+			+	80—380
Fritz and Bode, 1965 [27]	+		+		293
Sugawara, 1969 [15]	+	+	+		273—773
Sergeev et al., 1971—1977 [16—18]	+		+		90—1100
Zhdanovich and Chashkin, 1976 [22]	+		+		60—90
1976, 1978 [20]	+		+		400—1200
Bityukov, 1981 [19]	+		+		600—1100

TABLE 2. Thermal Conductivity of Quartz Glass vs Temperature

T, °K	Thermal conductivity, W/(m·°K)				
	data of present study	data of [17]	data of [22]	generalizations	
				Powell et al. [30]	Curwile et al. [29]
60	0,414	—	0,454	—	—
70	0,526	—	0,518	—	—
80	0,519	—	0,582	0,55	0,548
90	0,642	0,651	0,646	—	—
100	0,691	0,714	—	0,69	0,674
110	0,737	0,768	—	—	—
120	0,783	0,818	—	—	0,787
140	0,873	0,906	—	—	0,888
160	0,957	0,986	—	—	0,976
180	1,03	1,056	—	—	1,055
200	1,10	1,119	—	1,14	1,126
250	1,25	1,247	—	1,28	1,315
273,1	1,30	1,295	—	1,33	—
300	1,36	1,344	—	1,38	1,373
350	1,45	1,423	—	1,45	1,451
400	1,52	1,496	—	1,51	1,512
450	1,58	1,574	—	—	—
500	1,63	1,663	—	1,62	1,625
600	1,70	—	—	1,75	1,738
700	1,76	—	—	1,92	1,84
800	1,81	—	—	—	1,88

In accordance with the above, the data obtained in [4, 14, 17, 18, 20, 22, 23, 27] and a portion of the data from [7, 15, 24] were processed. In mathematical processing of data sets, varying weight factors are often assigned to various data depending on an estimate of the data's reliability. However, such an approach is only seemingly objective, because the selection of weight factors itself is always subjective. Therefore, we have considered all the available reliable data with equal weight, without regard to the accuracy specified by the individual authors.

A total of 239 experimental points were used to obtain a generalized quartz-glass thermal conductivity—temperature curve. The data were processed by a computer using the method of least squares. The approximating equation used was the best of the equations of form $\lambda = \Sigma a_i T^i$, in which the exponent was varied over the range —3 to +4, permitting description of λ over the entire temperature range by a single equation, rather than the two used in [28]. The best results were obtained with the equation

$$\lambda = a_0 + a_{-1}T^{-1} + a_{-2}T^{-2} + a_{-3}T^{-3}, \tag{1}$$

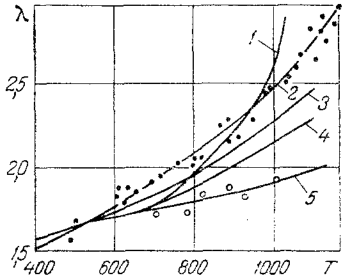

Fig. 1. Effective and true thermal conductivities of quartz glass versus temperature: 1) λ* result obtained in [14]; 2, 3) λ* of KV brand specimens 15 and 7 mm thick, respectively; dark circles, scattering of experimental data about smoothed curve 2; 4) λ* of KI brand specimen (processing of curve 4); open circles, some values of KV glass λ obtained by recalculation of curves 2 and 3. T, °K; λ, W/(m·°K).

with coefficents of the following values: $a_0 = 2.14749$ $a_{-1} = -298.76$ $a_{-2} = 20.72 \cdot 10^3$, $a_{-3} = -0.54 \cdot 10^6$. Table 2 (column 2) shows λ values calculated with Eq. (1). For comparison, the table also shows generalizations as carried out in [29, 30] and latter data obtained with the use of government references in [17, 22] for quartz glass of a specific brand (KV). The data of [17] are recommended for use as attested values for the corresponding standard specimens.

Analysis of deviations of experimental λ values from the average curve shows that the main mass of experimental points deviates by not more than 3%, while beyond the 5% boundary there are only 16 points, of which six are in the low temperature range below 180°K.

Comparison shows that the results of all the generalizations are close to each other. The differences that do exist are due primarily to the fact that these studies used data of different investigations in their data bases. Moreover, as was noted above, in the present study much data was used from studies of specimens in which no information on the structure and purity of the material was provided. As for the divergence between our results and the data of [17, 22], it comprises about 10% at 60°K, and does not exceed 5% over the range 70-180°K and 2% over the range 180-500°K. The large (9.7%) deviation of the experimental point in [22] from the average curve at the edge of the temperature interval (65°K) is remarkable. Despite the fact that in the range 65-80°K the results of [22] are the only existing ones obtained at a metrological accuracy level, the average curve cannot be fit to this point, since such fitting leads to abrupt physically unjustifiable inflections in the temperature coefficient of the thermal conductivity, i.e., in the quantity $\frac{1}{\lambda}\frac{d\lambda}{\lambda}$.

The results obtained indicate the necessity of performing additional studies of quartz-glass thermal conductivity, especially in the temperature range below 180-200°K.

In recent years quartz-glass thermal conductivity studies have been performed in the temperature range where the contribution of the radiant component becomes significant.

Figure 1 shows results obtained in [14] (curve 1) and [18] (curves 2-5). To recalculate the effective thermal conductivity to the true value (without radiant component), the reflection coefficient from the specimen boundaries and the function k(ν) for KI and KV brand quartz glass from [18] were used. It is evident from Fig. 1 that for the conditions studied, which, generally speaking, are typical of high-temperature thermal conductivity studies of quartz glasses, the radiant component begins to have a marked effect at a temperature of 550-560°K, so that the divergence of the quartz glass thermal conductivity values presented in the literature can be explained to a significant degree by this reason. The maximum value of

TABLE 3. True Thermal Conductivity Values of Quartz Glass at High Temperature

T, °K	Results of λ determination, W/(m·°K)				
	generalization of [29]	data of present study	calculation with λ^* [18]	calculation with λ^* [19]	calculation with a [18]
600	1,76	1,70	1,69	1,60	1,70
700	1,84	1,76	1,76	1,76	1,74
800	1,88	1,81	1,82	1,91	1,77
900	1,92	—	1,86	2,07	1,80
1000	1,96	—	1,92	2,23	1,84
1100	2,0	—	1,98	2,39	1,88

effective thermal conductivity occurs in the 15-mm-thick KV quartz glass specimen (curve 2), because of the increase in the radiant heat transfer component with increase in material thickness. The absence in the KI quartz glass spectrum of absorption bands occurring in the spectrum of KV glass in the range 3000-4000 cm^{-1} leads to a lower effective thermal conductivity for the 9.5-mm-thick KI specimen than that of the 7-mm-thick KV specimen.

Table 3 presents a comparison of true thermal conductivity values for quartz glass, obtained by various authors, with the data of the present study within the temperature range where radiant heat transfer is of significance. The data presented in column 6 of Table 3 are not calculated from the experimental values of effective thermal diffusivity, but by computing the true thermal diffusivity a, with subsequent determination of λ by using the known values of specific heat and density of the glasses.

As is evident from Fig. 1, the true thermal conductivity values of [18], obtained from data on the effective thermal conductivity λ^* of the specimens and their absorption spectra, agree within 4%. The true thermal conductivity values determined by the two different methods with proper elimination of the radiant component agree within 5% (columns 4 and 6 of Table 3). Considering the uncertainty of the boundary reflection coefficient values and the accuracy with which the effective thermophysical properties were measured, such an agreement of true thermal conductivity values obtained under wide variations in external conditions must be considered remarkable. In particular, the agreement indicates the validity of the calculation methods used and the internal consistency of all physical parameter values employed in [18].

The results obtained confirm the validity of the calculations of [29]. The fundamental equation used therein was

$$\lambda = c_p v l, \tag{2}$$

which relates the thermal conductivity λ of a dielectric, the specific heat at constant pressure c_p, the mean photon velocity v and the mean free path length l. The authors of [29] proposed that the product vl is a constant quantity. For quartz glass, with density practically constant up to 1100°K and equal to (2200 ± 10) $\mathrm{kg/m}^3$, this means that the true thermal diffusivity, produced solely by phonon transfer, is practically constant. On this basis, using values from the generalized temperature dependence of thermal conductivity and c_p values from Sosman's table, for the temperature range 473-623°K, the authors of [29] obtained an approximate relationship

$$\lambda/c_p = 16.6\cdot 10^{-5}\ \mathrm{kg/(m\cdot sec)}., \tag{3}$$

with the ratio λ/c_p increasing at both ends of the temperature interval. Such an approach, which the authors of [29] considered especially coarse and approximate, leads to results (column 2 of Table 3) which agree satisfactorily with the experimental data shown in columns 4 and 6.

The experimental results shown in column 5 appear elevated in comparison to the others considered. The divergence increases with increase in temperature, reaching 20% at 1100°K. This indicates the necessity of further studies of quartz glass at high temperatures in the range where they are partially transparent.

At the present time, in the temperature range above 600°K one can employ the results presented in columns 3 and 4 of Table 3 in practice.

Specific Heat. The specific heat of quartz glass has also been studied by many authors. The first estimates of reliability of specific heat data were carried out in 1927 by Sosman

TABLE 4. Values of Coefficients in Eq. (4)

Temperature range, °K	b_0	b_1	b_2	$b_{-2}\cdot 10^{-6}$
70—300	—84,32	3,871	—3,782	0
300—2000	931,3	0,256	0	—24,0

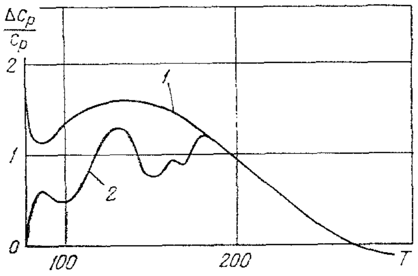

Fig. 2. Relative deviations of results of [36] (curve 1) and [34] (curve 2) from tabular values of Sosman (abscissa), $\Delta c_p/c_p$, %.

[32], who offered recommended values of quartz-glass specific heat. In 1934 a similar study was performed by Kelley [33]. As new experimental material appeared, he reconsidered his data in 1949 and 1960. In the latest ediction of [33] all results available as of September 1958 are considered. The specific heat values in this latest study show practically no difference from previous editions. The temperature range begins at 298.15°K (25°C).

According to Kelley, the specific heat of quartz glass as a function of temperature over the range 300-2000°K can be represented to an accuracy of 0.4% by the expression

$$c_p = b_0 + b_1 T + b_2 T^2 + b_{-2} T^{-2}. \tag{4}$$

Values of the coefficients in Eq. (4) are given in Table 4.

Unfortunately, Kelley presents no conclusions which would permit evaluating the uncertainty of the specific heat determinations. The uncertainty of 0.4% would appear to be too low. He also gives no specific heat values below 300°K. Such data can be found in Sosman's review article [32]. Processing of these data shows that they can be represented over the temperature range 70-3000°K by the same Eq. (4) with the coefficient values of Table IV. The approximation error in determining c_p with Eq. (4) is equal to 0.4%.

In recent years, the specific heat of quartz glass was studied in [34-36]. The results obtained in those studies are apparently more accurate than those offered in Kelley's and Sosman's tables. However, the data of [34-36] are insufficient to describe the temperature dependence of specific heat over a wide temperature range, while Eq. (4) does provide a convenient description, with the low temperature data of Sosman coinciding with Kelley's data at 300°K to an accuracy of 0.4%.

A comparison of Eq. (4) with the more exact results of [34-36] shows, e.g., that in the low temperature range (Fig. 2) the deviation reaches 1.6%, and in the high temperature range it reaches 2%. Thus, it seems appropriate to characterize the error of Eq. (4) by these values. Such a comparison also shows that further refinement of quartz-glass specific heat data is required. Thus, in this present time, in the temperature interval 70-1100°K, values obtained from Eq. (4) can be recommended. For higher and lower temperatures these data can be used as estimates.

Thermal Diffusivity. A description of a technique and apparatus for determining effective thermal diffusivity of quartz glass were presented together with measurement results in [18]. The data obtained in [18] can be approximated by the expression

$$a^* = 53.87 - 0.3882\ T + 7.10 \cdot 10^{-4}\ T^2. \tag{5}$$

Figure 3, whose abscissa consists of a^* values calculated with Eq. (5), compares the data obtained with other results available in the literature. The dashed lines are deviation levels

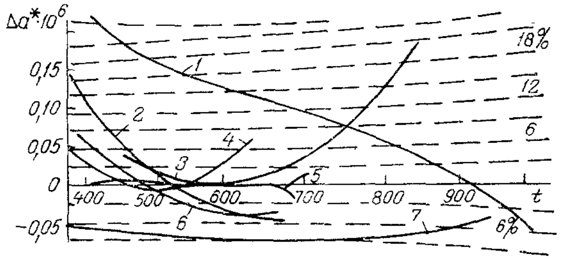

Fig. 3. Comparison of data presented in [18] for effective thermal diffusivity of KZ brand quartz glass (abscissa) with other data from literature: 1) [37]; 2) [37]; 3) [38]; 4) [39]; 5) [28]; 6) [23]; 7) [40]. $\Delta a \cdot 10^6$, m²/sec; t, °C.

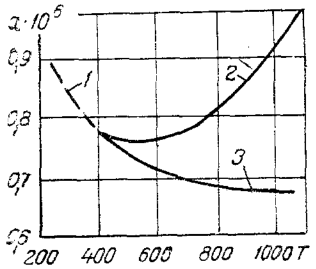

Fig. 4

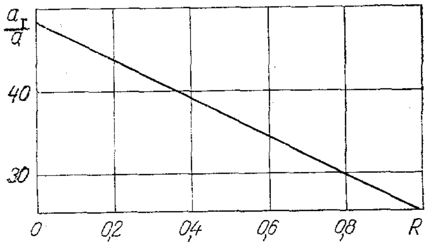

Fig. 5

Fig. 4. Effective and true thermal conductivities of quartz glass vs temperature: 1) generalization of data from the literature on a^* at relatively low temperatures [28]; 2) a^* values calculated with Eq. (5); 3) a calculations from data of curve 2 [18].

Fig. 5. Effect of specimen boundary reflection coefficient on radiant component of thermal diffusivity (plate thickness 20 mm, heating rate 80°K/hr, temperature 1100°K) [18]. a_r/a, %.

(in %) from the values used as the abscissa. Curves 1 and 2 are for quartz glasses used in two studies performed in the USA. Curve 3 is the result of measurements performed with an uncertainty of 10%. In the opinion of Plummer et al. [38], beginning at 400°C the results shown by curve 3 contain a systemic error due to radiant heat transfer. Curves 4 and 6 were obtained with the LITMO $\alpha\lambda$-calorimeter for KV brand quartz glass with an uncertainty of 8%. Curve 5 was obtained in [28] by generalization of then-available data on quartz-glass thermophysical properties. Curve 7 was constructed from the results of measurements in which the quartz glass was used as a reference material to monitor equipment constructed by the authors of [40] in measurements of thermal diffusivity by the Ångstrom method with an uncertainty of 5%.

Analysis of the data of Fig. 3 leads to the conclusion that over the temperature range 400-700°K the measurement results agree with each other satisfactorily. An exception is the data of [37], which apparently contain large systemic errors. In the high temperature range the sharp divergence between curve 3 and the results of [18] (the abscissa) can be explained by different effects produced by the radiant component in the relevant experiments.

In [18] it was demonstrated by calculations that expansion of the spectral interval over which the absorption coefficient is studied in the direction of higher ($\nu > 4000$ cm^{-1}) and lower ($\nu < 2000$ cm^{-1}) frequencies leads to lower or higher values of the absorption coefficient, which have practically no effect on calculation of the true thermal diffusivity. It was also shown that the transparency range of quartz glass (from 4000 to 40,000 cm^{-1}) does not affect the temperature field or a^*, in view of the symmetry of the boundary conditions.

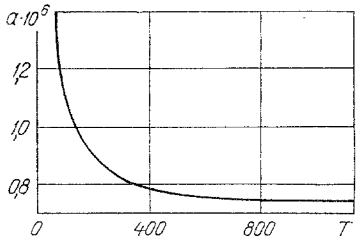

Fig. 6. Thermal diffusivity of quartz glass vs temperature, $a\cdot10^6$, m²/sec.

Effective and true thermal conductivity values are compared in Fig. 4. As is evident, the difference between a^* and a is significant, reaching 46% at T = 1100°K. The difference a_r between a^* and a is the radiant component of thermal diffusivity. Its temperature dependence is as follows (H = 20 mm; b = 800°K/h, R = 0.1):

T, °K	500	600	700	800	900	1000	1100
$a_r\cdot10^7$, m²/sec	0.18	0.41	0.79	1.33	1.85	2.32	3.01

Figure 5 shows the ratio of the radiant component of thermal diffusivity to the true thermal diffusivity for various values of the boundary reflection coefficient R. As is evident, uncertainty in R determination within the limits 0.08–0.15 [18] leads to insignificant error in determining the true thermal diffusivity — the maximum value of this error at T = 1100°K does not exceed 2%.

It is evident from Fig. 3 that the literary data do diverge from each other significantly. Therefore, to obtain recommendable values of thermal diffusivity it is best to define the value from the expression

$$a = \lambda/\rho c_p, \tag{6}$$

where the density ρ is taken equal to (2200 ± 10) kg/m³ for quartz glass.

The density of quartz glass changes insignificantly with temperature, since the specimen volume $V = V_0(1 + \beta t)$ changes little, being dependent on the volume expansion temperature coefficient β, which is approximately equal to $2\cdot10^{-6}$ °K^{-1} over the entire temperature range considered. In the worst case (at t = 800°C), the correction for thermal expansion of the specimen volume does not exceed 0.2%, i.e., is relatively small compared to the uncertainty (0.5%) of the density value taken. In estimating uncertainties this correction may be treated as an uneliminated systemic error residue.

Thus, the expression for calculating thermal diffusivity of quartz glass has the form

$$a = 454.5\cdot10^{-6}\,\frac{a_0 + a_{-1}T^{-1} + a_{-2}T^{-2} + a_{-3}T^{-3}}{b_0 + b_1T + b_2T^2 + b_{-2}T^{-2}}, \tag{7}$$

where the coefficients a_0, a_{-1}, etc., are the coefficients of Eq. (1), and b_0, b_1, etc., are the coefficients of Eq. (4). A graph of thermal diffusivity vs temperature is shown in Fig. 6. As is evident from the graph, beginning at 500°K the thermal diffusivity becomes practically constant and equal to $(0.75\text{–}0.77)\cdot10^{-6}$ m²/sec, so that the ratio λ/c_p which is in fact expressed by Eq. (3) is practically constant.

Over the temperature range 180–500°K the uncertainty in determination of a by Eq. (7) does not exceed 3%, and it does not exceed 6% over the entire temperature range.

NOTATION

T is the temperature, °K; λ and λ^*, true and effective thermal conductivity, W/(m·°K); a, a^*, a_r, true, effective, and radiant thermal diffusivity, m²/sec; c_p, specific heat, J/(kg·°K); ρ, density, kg/m³; β, volume expansion coefficient, °K^{-1}; R, specimen boundary reflection coefficient; V, V_0, specimen volume at temperatures t and 0°C; v, mean phonon velocity, m/sec; l, mean phonon free path length, m; ν, wave number, cm^{-1}.

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