THERMAL CONDUCTIVITY OF VITREOUS SILICA: SELECTED VALUES

by

Lois C. K. Carwile
and
Harold J. Hoge

July 1966

UNITED STATES ARMY
NATICK LABORATORIES
Natick, Massachusetts 01760

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FOREWORD

This report is the fourth of a series on the thermal conductivities of materials of scientific and engineering interest. The values selected are based on thorough study and critical evaluation of published investigations. In a critical survey such as this one, much depends on the judgment of the surveyors. The care that the authors of the present survey have exercised may be judged from the comments they have made on the individual papers examined. Their comments on the more important papers are in the text of the report. In addition, they have made many brief comments on less important papers; these comments are given as annotations, immediately following the listing of the paper in the references.

S. DAVID BAILEY
Director
Pioneering Research Division

APPROVED:

DALE H. SIELING
Scientific Director
U. S. Army Natick Laboratories

WILLIAM M. MANTZ
Colonel, QMC
Commanding
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The published literature on the thermal conductivity of vitreous silica has been assembled and the results critically evaluated. Best values of thermal conductivity as a function of temperature have been selected. These are presented in both graphical and tabular form. They cover the range 80 to 660° K and published data extending from 3° to 2100° K are shown in the graphs. An attempt was made to consult all work that could significantly affect the choice of best values. Published papers were located with the aid of Chemical Abstracts, Physics Abstracts, the Thermophysical Properties Retrieval Guide, and some other general sources. In addition, relevant references in the papers themselves were followed up until a substantially "closed system" had been generated, as shown by the fact that no new references were being turned up.
THERMAL CONDUCTIVITY OF VITREOUS SILICA: SELECTED VALUES

1. Introduction

Vitreous silica is an important material in research, and is used to some extent in industry. It is easily obtained in a state of high purity. It is chemically inert to most substances, including many strong acids; hence it is valued as a storage container, and as a reaction vessel for chemicals of the highest purity. It is much used in optical work because of its wide range of transmission. It has excellent elastic properties; it transmits ultrasonic waves with very little absorption of energy (except at low temperatures); and fibers of it, being relatively immune to permanent set, make excellent electrometer suspensions and delicate spring balances. Vitreous silica has a very small coefficient of thermal expansion, hence is highly resistant to thermal shock. As a refractory, it can withstand temperatures of $1300^\circ\text{K} (1027^\circ\text{C})$ for long periods, and higher temperatures for short periods. It is an excellent electrical insulator.

Vitreous silica is a noncrystalline material (glass) of the composition $\text{SiO}_2$. It is usually made either from large selected quartz crystals or from very pure quartz sand. It is available in two basic types: clear (transparent) and translucent (satiny, silky). Clear vitreous silica is made by melting large crystals of quartz, and for this reason it is often called fused quartz or quartz glass; however, we prefer to reserve the word quartz for the commonest of the crystalline forms of silica, and to refer to the glass as vitreous silica, fused silica, or silica glass. Sosman (75), in his book on silica published in 1927, accepted the name "vitreous silica", and Laufer (76) in a more recent paper recommends it.

When vitreous silica is made from sand, the translucent or satiny type is obtained. The satiny appearance is caused by tiny air bubbles that are trapped when the material is fused, and then are elongated into striations when the material is drawn or worked into useful shapes. The present study is almost entirely concerned with clear, vitreous silica, which contains no trapped bubbles of air or other gas.

Vitreous silica is marketed under the names of fused silica, quartz glass, vitreous silica, and also under a number of trade-mark names. Among these are Amersil, Homosil, Infrasil, and Vitreosil; some of these names refer to special varieties having, for example, high optical transmission in a certain region of the spectrum.
Because vitreous silica is a highly reproducible material with suitable mechanical, chemical, and refractory properties, it has been suggested as a standard material for thermal-conductivity work. However, we believe the large radiative transfer of heat through vitreous silica makes it less desirable than an opaque substance as a thermal-conductivity standard, at least at high temperatures. We will return to this point later.

**Characterization.** Vitreous silica consists of a network of tetrahedrons of SiO\(_2\), loosely packed. Each silicon atom is tetrahedrally surrounded by 4 oxygen atoms, and each oxygen is shared by 2 silicons; but long-range order is lacking. In a crystal, all corresponding Si-O-Si bond angles are equal, but in the glass this angle varies somewhat; also, according to Warren and Biscoe (77), the orientation of adjacent tetrahedra about these bonds is random in the glass. The X-ray analysis of vitreous silica shows the short-range, near-neighbor order that is characteristic of liquids and glasses, but no order extending over a distance as large as 3 or 4 times a unit-cell dimension.

The density of vitreous silica has a slight dependence on the rate at which the material is cooled from the molten state. Sosman (75) selected 2.203 g cm\(^{-3}\) as the best value for the density of clear vitreous silica, and stated that different samples were likely to differ in density by as much as 1 per mille. Subsequent work gives little reason for differing from Sosman's conclusions. Crawford and Cohen (13) observed both the density and the thermal conductivity of vitreous silica before and after neutron bombardment. Using a hydrostatic method, they found the density at room temperature to be 2.2002 g cm\(^{-3}\) before bombardment. This value was increased by 2.7 percent during bombardment, but subsequent annealing substantially restored the original value. The lack of long-range order in vitreous silica is associated with an open molecular structure with numerous vacant sites. These vacant sites reduce the density of vitreous silica to a value about 17 percent less than that of crystalline quartz.

Vitreous silica does not have a sharp melting point. The crystalline form of silica that is stable at the highest temperature is cristobalite (75); it melts at about 1710°C (1983 K), and becomes a liquid of high viscosity.

**Devitrification.** The high-temperature limit of usefulness of
vitreous silica is generally the temperature at which devitrification (crystallization) occurs, rather than that at which the material begins to soften or melt. Below about 1000°C (1273 K) it shows no tendency to crystallize, but at 1100°C and above, minute crystals gradually form. The crystals become visible when the material is cooled, as they undergo a transformation that gives them a white, chalky appearance. Devitrification is accompanied by a decrease in mechanical strength and an increase in tendency to shatter. It proceeds more rapidly as the temperature is raised; it may be complete in 4 to 8 hours, or it may be a matter of days. Devitrification begins at a surface; it is accelerated by water vapor, by alkalis, and by many other substances. It is retarded when the amount of oxygen in the vitreous silica is slightly less than stoichiometric (78). High-quality transparent vitreous silica, in the absence of catalysts, has been held at 1300°C (1573 K) for several hours with only a slight cloudiness resulting. The product of devitrification is usually cristobalite, even at temperatures where cristobalite is not the most stable form of silica.

Effects of OH. Although clear vitreous silica made by melting crystals is ordinarily very pure, there are usually traces of metals in it. For some optical work, these traces of metals must be avoided; for this purpose, silica is made by reacting SiCl₄ with water and depositing the SiO₂ in the glassy state. Vitreous silica made from SiCl₄ is metal-free, but it contains (79) a relatively large amount of OH, which comes from the water formed in the reaction. This OH can amount to more than 1 part per mille by weight, whereas vitreous silica made by melting natural crystals contains only about 3 parts of OH per million. This difference in OH-content causes small changes in density, thermal expansion, index of refraction, and other properties, but perhaps its greatest importance is in spectroscopic experiments that require transmission of the OH frequencies, the presence of OH in the glass causes these frequencies to be absorbed. The effect of OH-content on the thermal conductivity of vitreous silica has not been specifically studied, but presumably it is small in comparison with the scattering of even the better data now available.

Satiny vitreous silica, k-values. As discussed earlier, satiny vitreous silica contains elongated air bubbles that form striations in the material. Since the selected values in the present paper refer to clear, rather than satiny vitreous silica, we will present here the small amount of information that is available on the differ-
ence in thermal conductivity of the clear and satiny varieties. One would expect the striations to reduce the conductivity, especially when the direction of heat flow is perpendicular to the long axes of the striations. However, the effect appears to be rather small. Koenig and co-workers (34) reported measurements on clear vitreous silica, and on satiny material with the heat flow parallel to the striations. These measurements show the k-values of the satiny type to be about 1 percent below those of the clear material in the range 40 to 120°C. The authors did not consider this difference significant, but we note that it is in the direction to be expected. Later the same group (33) made similar measurements on two new satiny samples, one having the striations parallel, the other perpendicular, to the direction of heat flow. Again, in approximately the same temperature range, one sample had a conductivity about 3 percent lower than the other, but unfortunately the report does not tell us which sample was which. Both of the sets of measurements just referred to were made rather early in an extensive series of investigations. More accurate results were obtained later in the work, but the comparison between clear and satiny vitreous silica does not appear to have been repeated.

2. Selection of the Values

The methods used in evaluating the experimental data, drawing a master curve, and preparing a table of selected values of the thermal conductivity, k, will now be described. As indicated earlier, the variety of material to which the curve and tables refer is clear (transparent) vitreous silica. In some cases the same set of experimental data has been published in several different papers. In such cases we have selected one of the papers as the primary reference and have listed the others in the group of references "for which another reference is preferred." The governing factors considered in the selection of the primary reference will be found in the annotations to the various references.

Because of the large amount of data on the thermal conductivity of vitreous silica and the wide range of temperature covered, three separate graphs have been used to present the data. All three graphs start at 0°K; one extends upward to 100°K, another to 1000°K, and the third to 2100°K. The results of a particular research are given in one graph but are not duplicated in the others; thus overcrowding of the graphs is avoided.
The values of $k$ selected and recommended for use are given in Tables 1 and 2, and are represented in Figs. 1, 2, and 3 by heavy solid lines (the master curve). The values were selected by plotting the available data on large-scale graphs similar to Figs. 1, 2, and 3. A master curve was drawn, studied, revised, and tentatively accepted. Values were read from this master curve; they were differenced, smoothed, and checked for consistency with the master curve. When table and curve were smooth and mutually consistent, they were accepted. Table 1 resulted from this process. Table 2 was derived from Table 1 and is consistent with it.

The master curve and the tables extend only from about 80° to about 660° K (-193° to 387°C). At higher temperatures there is a dashed line joining with the master curve and extending it. This extension was derived by using heat-capacity data, and it is believed to represent roughly the thermal conductivity of vitreous silica in the absence of radiative heat transfer. The method of deriving the dashed curve is described later in this report.

Data Shown in Fig. 1. Figure 1 extends from 0° to 1000° K. It contains the data from references (1) to (11); these references include much but not all of the best data for intermediate temperatures.

Although the master curve is based on the data in all three figures, the data of Birch and Clark (1), shown in Fig. 1, are perhaps the most valuable single set of data. These authors used a guarded hot-plate apparatus and a steady-state absolute method. The temperature difference across the sample was usually about 5° K. Each test was run in a nitrogen atmosphere and again in helium, and between the two results the thermal-contact resistances were eliminated.

Considerable weight has been given to the data obtained by Ratcliffe (2), which cover the range 123° to 322° K. Steady-state measurements were made in 3 different apparatuses; each apparatus had the hot plate sandwiched between 8 specimens. Care was taken to reduce or eliminate contact resistance, and corrections were made for lateral heat losses. At least 12 different samples were measured.

Valuable data were obtained by Kamilov (3) from about 89° to about 500° K. The apparatus was contained in a Dewar flask within another flask, with provision for regulating the pressure between the walls of the inner flask. One unusual feature of the apparatus was the type of "thermocouple" used to register temperature equality between the top shield and the hot plate. This "thermocouple" consisted of a copper sheet coated with cuprous oxide; it actuated an
Thermal conductivity of vitreous silica

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<th>T (°K)</th>
<th>k (millical cm sec °C)</th>
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Thermal-conductivity data of references (1) to (11).
Kurepin and Platunov (4) described an apparatus for measuring either thermal conductivity or thermal diffusivity over a wide range of temperature. In measuring $k$, heat was supplied so as to get a steady rate of temperature rise in the entire sample, for example, 0.10 deg/sec, and the temperature drop across the sample was measured as a function of time. The amount of heat flowing was determined from the known heat capacity and temperature rise of a heat sink, which was a core of Armco iron. Their data appear to be of good accuracy.

Eacken (5) used a steady-state hot-plate method, and obtained surprisingly good results considering that his apparatus appears to have been unguarded.

Vasil'ev (6) used a quasistationary (steady-rise) method for 39 of his plotted points, and a transient method for 2 points. He used radial heat flow in a warded cylindrical apparatus that did not require a heat sink. Automatic temperature control and recording were used, and the data appear to be good. We note that the data in his Fig. 3 taken from Ratcliffe appear to be incomplete; and those taken from Knapp, to be misplotted.

Kaye and Higgins (7) reported careful measurements by a comparison method, using aluminum bars as the reference material. Data were obtained for 3 samples of vitreous silica; they are 6 to 8 percent lower than our accepted curve.

Kingery and Norton (8) used 2 different methods. The first method employed a prolate spheroidal envelope; it was an absolute, steady-state method. The second method was a comparative one. Their data are about 4 percent higher than our accepted curve; however, the data are valuable because they extend to relatively high temperatures.

Bil' and Avtokratova (9) used vitreous silica as a standard material for testing their apparatus. The values found are about 8 to 15 percent lower than our selected values. Their samples were stated to be of unsatisfactory purity and to contain air pockets.

The results obtained by Scholes (10) and by Koenig (11) were
Fig. 2 Thermal-conductivity data of references (12) and (13). The master curve begins at 80° K.
obtained by a comparison method. These values are higher and rise more rapidly with temperature than those we have selected.

Data Shown in Fig. 2. Figure 2 extends from 0° to 100°K. In this range, the work of Berman (12) appears to be the most reliable. He measured 3 samples by a steady-state method, with a radiation-shielded apparatus surrounded by a vacuum jacket. The entire apparatus could be cooled by liquid helium or other coolant.

The k-values of Crawford and Cohen (13) increase very rapidly with temperature, and do not extrapolate toward the accepted values at higher temperatures as Berman's do. Crawford and Cohen attributed the difference between their values and those of Berman to a difference in the materials tested, but the difference seems rather large for such an explanation.

Data Shown in Fig. 3. Figure 3 extends from 0° to 2100°K. Very good data were reported by Devyatkova, Petrov, Smirnov, and Moizhes (14), over a wide range of temperature. Their work was undertaken to determine the potential usefulness of vitreous silica as a standard of reference for measurements of thermal conductivity. They used 3 different apparatuses. Temperature gradients were measured by means of thermocouples fastened to metal pins inserted into holes drilled in the samples. Corrections were made for lateral heat losses. Their results have significantly influenced our selection of best values throughout the range for which a master curve and tables have been given. As will be seen below, we believe that radiant energy transfer has caused the results at higher temperatures to represent an apparent rather than true thermal conductivity.

Seemann (15) reported careful work by a radial-flow method with 2 different apparatuses and a single test sample. During measurements, both surfaces of the specimen were in contact with liquid metal: mercury at ordinary temperatures, lead-tin eutectic at higher temperatures. Seemann's results appear to be about 10 percent low in the range of temperatures for which we have drawn a master curve. However, at higher temperatures Seemann's results appear to be among the most successful in avoiding radiant-heat transfer.

Ito (16) also used the radial-flow method. His results are lower than those of most other workers; at the lower end of his range, he is about 20 percent below our master curve. Ito's results as reported by Birch and Clark (1) and by Rotcliffe (2) in the form
Fig. 3 Thermal-conductivity data of references
With Radiation

Without Radiation (Estimated)

T, °K

Refer to conductivity data of references (14) to (25).
of straight lines do not agree with the plotted points in Ito’s paper; the discrepancy can be explained if we assume that these authors used Ito’s equation with temperatures in °K, whereas it is actually for temperatures in °C. Whatever the cause, the discrepancy is substantial.

Vishnevskii and Dzyubenko (17) built a guarded apparatus for measuring k by a transient method. They report results obtained with this apparatus and also by a steady-state method; the two sets of results show good agreement and are represented by a single curve. However, these values are about 8 percent lower than those of our accepted curve.

An unusual method, involving steep temperature gradients, was used by Wray and Connolly (18) to measure k of vitreous silica at high temperatures “without radiative effects”. Each sample was in the form of a cylindrical rod with a tungsten wire along the axis, so prepared as to have continuous bonding between wire and sample as shown by microscopic examination. The wires were of very small diameter (0.008 cm or less), and served as both heater and thermometer. From the graphs of observed power vs. wire temperature, the slopes were determined; and these slopes were used to find k by a formula which Wray and Connolly derived theoretically. Their results were presented as smooth curves, and give values of k at higher temperatures than any other papers that we have located. The curves of Wray and Connolly remain substantially horizontal at higher temperatures, where those of others rise steeply.

Observations at high temperatures were also reported by Lucks, Matolich, and Van Velzor (19); Kingery and Norton (20); and Pustovalov (21). In all these data the steepening rise shows the presence of radiative heat transfer. Most observers made some effort either to reduce or to correct for radiative heat transfer. For example, Lucks, Matolich, and Van Velzor made some experiments with thin low-emissivity foil “adjacent to the surface of the transparent solid specimens” (they used a comparison method, with Armco iron as the accepted standard). They found that the k-values obtained without foil showed a more rapid increase than those obtained with foil, above about 450°C (723°K).

Kingery and Norton used 3 different apparatuses; all employed the comparison method. The paper of Lee and Kingery (64) reports the same work with a somewhat better description of experimental methods.
Pustovalov used an apparatus of cylindrical symmetry, with an axial heater; the positions of the thermocouples in the samples were determined by using X-rays (80). The $k$-values reported are about 25 percent lower than our master curve. It is possible that the porosity of the sample is responsible; Pustovalov gives (69) the apparent porosity of his sample as 1.0 percent; but from his measured density ($2.06 \text{ g cm}^{-3}$) and our accepted density ($2.204$), we calculate a porosity of 6.5 percent.

Also shown in Fig. 3 are the results of Colosky (22), and those of Knapp (23); these appear to be too high and to rise too steeply.

At the higher temperatures, the data reported by almost all the experimenters lie above the master curve and the dashed curve that extends it upward. As mentioned earlier, this dashed curve represents our estimate of the thermal conductivity of vitreous silica in the absence of radiative heat transfer. The method of obtaining this curve will be described in the next section.

3. Nonradiative Heat Transfer at High Temperatures

Thermal conductivity as defined does not include the transfer of heat by either convection or radiation. However, in many investigations of thermal conductivity some heat transfer by convection or radiation takes place in the sample. When the amount is small, this transfer is often included with the transfer by conduction, and an apparent thermal conductivity is calculated. In the case of vitreous silica such a procedure is permissible up to about $600^\circ{\text{K}}$, but as the temperature increases the radiative heat transfer becomes very large, and it is not desirable to lump it with thermal conductivity.

The problem of radiative heat transfer is of special importance in vitreous silica because the substance is highly transparent. Vitreous silica transmits a wider range of wave lengths than most glasses; the range extends from 0.2 to 4.0 microns, approximately, and in addition there are other bands of transmission or partial transmission at longer wave lengths.

In estimating the ordinary (nonradiative) thermal conductivity of vitreous silica at high temperatures we can follow either of two paths. We can estimate the effect of radiation and subtract it from the total observed heat transfer, or, we can assume a
relation between thermal conductivity and some other property of the material such as the specific heat $c_p$, and use this to give us $k$.

It is easy to make rough calculations of radiative heat transfer in vitreous silica, but much harder to make accurate ones using the absorption coefficient appropriate for each wave length. We have avoided such difficult and perhaps uncertain calculations by assuming that $k/c_p$ is a constant. Accepting $c_p$, we have calculated the non-radiative value of $k$, and have checked the results so obtained by some rough calculations of radiative heat transfer that are described in the next section.

It is well known that heat capacity and thermal conductivity are closely related. According to a very simple theory $k$ is proportional to the product $c_p v l$, where $v$ is the mean velocity of phonons and $l$ their mean free path. Our assumption that $k/c_p$ is constant is equivalent to assuming that the product $v l$ is constant. This assumption is used simply because no better one is at hand, and no great accuracy is claimed for it. If we were to go beyond the assumption $k/c_p = a$ constant, we would assume that this quantity decreases somewhat with temperature, because increasing lattice vibrations tend to reduce the mean free path $l$.

Sosman (reference 75, p. 314) has published a table giving $c_p$ of vitreous silica. Although there are more recent data in some temperature regions than those that Sosman used, his table is convenient for our present purpose because it covers a wide range, extending from 180 to 1975 K. With $c_p$-values from this table and $k$-values from our master curve, the quantity $k/c_p$ was calculated and plotted vs. T. Throughout the interval 473 to 623 K, this quantity had the value 16.6 mg cm$^{-1}$ sec$^{-1}$. At both ends of the interval the curve turned upward. The value 16.6 was accepted.

Multiplying Sosman's $c_p$-values by 16.6 gave the values of $k$ that are represented by the dashed extension of the master curve in Fig. 3. This dashed curve we believe to be a rough but reasonable estimate of the thermal conductivity of vitreous silica in the absence of radiative heat transfer. It will be seen that the results of Wray and Connolly (18), whose experiments were designed to minimize radiative heat transfer, are in much better agreement with the dashed curve than those of any other investigator.
Sosman's $c_p$ values have a change in slope between 1400° and 1500°K, which shows up in our dashed curve. Whether this change in slope is real or is due to experimental error we do not know. The uncertainty in assuming $k/c_p$ to be constant is so large that the irregularity in the dashed curve is probably comparatively unimportant.

4. Radiative Heat Transfer

As a check on the values of nonradiative heat transfer obtained as described above, a few calculations of the radiative heat transfer to be expected in vacuum and in our idealized model of vitreous silica have been made. First the "thermal conductivity" of a "slab of vacuum" 1 cm thick, with black-body walls, was calculated. Such a model would account for the radiative heat transfer in a perfectly transparent material and might be useful for a material that was a uniformly gray absorber at all wave lengths. It did not account satisfactorily for the sharp increase in slope shown by the experimental data for vitreous silica, principally because it caused the rise to occur at temperatures that were too low.

Next we tried a slightly more sophisticated model, one in which the sample of 1 cm thickness was considered to be a perfect transmitter for all radiation of wave lengths lying between 0.2 and 4.0 microns, and to be perfectly opaque to all other wave lengths. (Actually, the energy at wave lengths shorter than 0.2 μ was negligible, so a "window" extending from 0 to 4.0 μ would have given the same result.) This model was quite satisfactory. When the radiative "conductivity" of the model was added to the values of the master curve and the dashed curve, the dot-dash curve shown in Fig. 3 was obtained. This curve is a reasonably good representation of the experimental data in which radiation was important. Note that it comes close to the data of Kingery and Norton, and to those of Lucks, et al. "without foil".

Since the more sophisticated model of radiative transfer and the dashed $k$-curve based on $c_p$ are mutually consistent they tend to confirm each other, but even so the breakdown of $k$ into radiative and nonradiative parts can only be considered a rough approximation.

A few remarks pointing out the weakness of the model used above in the calculation of radiative transmission are in order. Perfect transmission of radiation gives a conductance that is independent of the sample thickness and a conductivity that is
proportional to sample thickness. For many wave lengths, vitreous silica is neither a perfect transmitter nor a perfect absorber, but is intermediate, with a mean free path for the radiation that may be of the order of a few millimeters. An individual quantum will, on the average, travel this distance before it is absorbed, but new quanta to replace those absorbed will be generated throughout the glass.

Gardon (81) has summarized the theory of heat transfer within glass by absorption and re-emission of radiation. A radiative thermal conductivity proportional to \( T^3 \) and inversely proportional to the absorption coefficient of the glass is found. However, the concept of a radiative thermal conductivity is not very useful because it is applicable only in the interior of large samples and not near the heat source and heat sink (hot plate and cold plate). The boundary conditions when radiative heat transfer is present are normally such that the temperature gradient within the sample is nonuniform, being steeper near the hot and cold plates than elsewhere. We see from this that when the absorption coefficient has any small non-zero value (as well as when it is actually zero) the "radiation conductivity" depends on sample thickness.

Some workers have reduced the effect of radiative heat transfer by using highly reflective metal foil such as aluminum or platinum, placed on either side of the sample, between it and the hot and cold plates. Although this arrangement substantially reduces the amount of radiative heat transfer, it does not eliminate it. Even if the foil had zero emissivity so that heat entered and left the sample only by conduction, a portion of the heat would be converted to radiation within the sample and would travel through the sample in the form of radiation.

5. Vitreous Silica as a Standard of Thermal Conductivity

Clear vitreous silica exhibits many of the characteristics desirable in a standard material for thermal-conductivity measurements. Among these characteristics are its high purity, uniformity, low thermal expansion, and chemical inertness. Vitreous silica has been used as a standard by a number of investigators; among them Benfield (27), Ballard and Kblett (82), Namilov (3), and R. W. Powell (67). Noting the wide use of vitreous silica as a calibrating standard, Devyatkova, Petrov, Smirnov, and Moizhes (14) made experiments to examine its suitability for this purpose.
Using three different apparatuses in overlapping ranges, they made measurements from $82^\circ$ to $1166^\circ$K. Comparing their data with those of Berman (12), Ratcliffe (2), and Kingery (60), they found good agreement, and concluded that vitreous silica is suitable for use as a standard substance in thermal-conductivity measurements.

Although we agree in part with this conclusion, we believe that the usefulness of clear vitreous silica as a standard is limited to low and moderate temperatures, where radiation is not important. As was brought out in the two preceding sections of this paper, radiative heat transfer in vitreous silica undoubtedly exceeds nonradiative transfer at the higher temperatures. The agreement of the high-temperature data of Devyatkoa, et al., with those of Kingery is better than we would expect. The sample used by the first group had a thickness of 1 cm. The thickness of Kingery's sample was probably 2.54 cm. The relative importance of radiation in the two samples would be different and should lead to different apparent values of $k$. It is possible that some compensating effect was present that kept the two sets of results in agreement.

Methods of making vitreous silica opaque have been devised, but so far as we are aware there are no measurements of the thermal conductivity of opaque samples. If vitreous silica could be rendered opaque without changing its thermal conductivity, it would make an excellent standard material for use at any temperature up to the point where significant devitrification may occur.

6. **Reliability of the Tables**

The tabulated values of $k$ between $150^\circ$ and $450^\circ$K are believed to be accurate to ±7 percent; near room temperature they may be somewhat better. Below $150^\circ$K the uncertainty increases and may reach ±12 percent. Likewise above $450^\circ$K the uncertainty increases and may be as large as ±15 percent at the upper limit of the table.

7. **Data for Conversion of Units**

\[
T \,(^\circ R) = T \,(^\circ K) \times 1.8.
\]

\[
T \,(^\circ K) = t \,(^\circ C) + 273.15.
\]

\[
T \,(^\circ R) = t \,(^\circ F) + 459.67.
\]

Watt $\text{cm}^{-1} \text{cm}^{-1} \text{K}^{-1}$ cal $\text{cm}^{-1} \text{cm}^{-1} \text{K}^{-1} \text{sec}^{-1} \times 4.1840.$

17
Btu in. °R⁻¹ft⁻²hr⁻¹ = cal cm °K⁻¹cm⁻²sec⁻¹ x 2902.9.

Btu ft. °R⁻¹ft⁻²hr⁻¹ = cal cm °K⁻¹cm⁻²sec⁻¹ x 241.91.

8. Acknowledgment

We are indebted to Mr. Ronald A. Segars of this laboratory for help on various phases of this survey.
9. ANNOTATED REFERENCES

Containing Data Plotted in Fig. 1

1. Francis Birch and Harry Clark, "The thermal conductivity of rocks and its dependence upon temperature and composition, Part I," Am. J. Sci. 238, 529-58 (1940). Values were read from their Fig. 6 and also from their Fig. 7, and agreed within the error of reading. Corresponding values from the two graphs were averaged before being plotted in our Fig. 1.

2. E. H. Ratcliffe, "Thermal conductivities of fused and crystalline quartz," Brit. J. Appl. Phys. 10, 22-5 (1959). In addition to the plotted points of Ratcliffe's "present measurements" in his Fig. 3, we have also accepted the point taken by Ratcliffe from D. W. Butler, "Unpublished data (National Physical Laboratory, 1950)."

3. I. K. Kamilov, "Investigation of the thermal conductivity of solids in the interval from 80 to 500°K," Instr. Exptl. Tech. No. 3, 583-7 (1962). This is a translation of reference (58). Figure 3 of this paper contains vitreous-silica data in the form of plotted points. We found that the x's represent Kamilov's data; the circles were taken by him from reference (14).

4. V. V. Kurepin and E. S. Platunov, "Apparatus for rapid wide-temperature thermophysical tests of thermally insulating and semi-conducting materials (dynamic α λ-calorimeter)," Izv. Vysshikh Uchebn. Zavendenii, Priboorost. No. 5, 119-26 (1961). Their Fig. 4 contains values of thermal conductivity (λ) and thermal diffusivity (α); we have used only the former.

5. A. Eucken, "The temperature dependence of thermal conductivity of solid nonmetals," Ann. Physik (4) 34, 185-221 (1911). The value for 100°C (373°K) is followed by a question mark in the paper; also it does not agree with the value given by Eucken for the same temperature in a later paper (reference 56). Hence we have omitted this value from our Fig. 1.
6. L. L. Vasil'ev, "Method and apparatus for the determination of the thermophysical properties of heat-insulating materials in the temperature range 80 - 500°K," Inzh.-Fiz. Zhur. 7, No. 5, 76-84 (1964). Thermal-conductivity values are given in his Fig. 3; thermal-diffusivity and specific-heat values are given in his Fig. 4. We have used only the thermal-conductivity data as given in his Fig. 3.


8. W. D. Kingery and F. H. Norton, "The measurement of thermal conductivity of refractory materials," U. S. At. Energy Comm. Rept. NYO-6444 (June 30, 1954), 16 p.; AD-40873. We have taken the experimental data from Fig. 1 of this report. This figure has been published in several different papers; we have selected this one as our primary reference because the figure in it was the largest. Reference (60), which contains a smaller version of the same figure, contains a fuller description of the work than the present reference.

9. V. S. Bil' and N. D. Avtokratova, "Temperature dependence of the thermal and temperature conductivities of some unfilled polymers," High Temp. 2, 169-75 (1964). This is a translation of reference (49). The data given in their Fig. 1 have been read off and are plotted in our Fig. 1.

10. William A. Scholes, "Thermal conductivity of bodies of high BeO content," J. Am. Ceram. Soc. 33, 111-7 (1950). The points for vitreous silica given in his Fig. 3 have been read off and are plotted in our Fig. 1.

11. John H. Koenig, "Progress Report No. 4 from September 1 to December 1, 1953," N. J. Ceram. Research Sta., Rutgers Univ., 159 p.; AD-29335. We have selected the data given in engineering units in Table II, p. 6 of this report, for conversion and plotting in our Fig. 1.
12. R. Berman, "The thermal conductivities of some dielectric solids at low temperatures," Proc. Roy. Soc. (London) A 208, 90-108 (1951). We have read off and used the plotted points in the upper section of Fig. 2 of Berman's paper.

13. J. H. Crawford, Jr. and A. F. Cohen, "The effect of fast neutron bombardment on the thermal conductivity of silica glass at low temperature," Bull. Inst. Intern. Froid, Annexe 1958-1 (1958), p. 165-72. These authors give data obtained before irradiation, after irradiation, and after the effects of irradiation have been removed by annealing. The lowest curve in their Fig. 1 represents the data obtained in the unirradiated state and the data obtained after annealing; we have used both of these sets of data.

14. E. D. Devyatkova, A. V. Petrov, I. A. Smirnov, and B. Ya. Molzhis, "Fused quartz as a standard material in heat conductivity measurements," Fiz. Tverdogo Tela 2, 738-46 (1960). Figure 3 of this paper contains 3 sets of data obtained by the authors and 4 sets of data taken from other workers. The data of the authors have been read from this figure. An English translation of the paper exists (reference 55), but it was considered preferable to take the data from the Russian original.

15. Herman E. Seemann, "The thermal and electrical conductivity of fused quartz as a function of temperature," Phys. Rev. 31, 119-29 (1928). The data were read from Fig. 3 of this paper.

16. Shuyo Ito, "Thermal conductivity of a transparent vitreous silica tube," J. Mazda Co. (Japan) Vol. 4, No. 2, 185-8 (1929). A copy of this paper was obtained for us from the Diet Library, Tokyo, by a Japanese colleague. The experimental points plotted in Fig. 3 were read from the graph in this paper.

18. Kurt L. Wray and Thomas J. Connolly, "Thermal conductivity of clear fused silica at high temperatures," J. Appl. Phys. 30, 1702-5 (1959). These authors give k-values only as the smooth curves plotted in their Fig. 3; values have been read from these curves. Fig. 5 of reference (74) could have been used instead, but there are slight differences in the two figures, and the present paper is presumably the later one. In our Fig. 3, the data for Wray and Connolly have been shown as short sections of smooth curves at 100°-intervals. There is no correspondence between these short sections and any plotted points of Wray and Connolly, since these authors gave their k-data only as smooth curves.

19. C. F. Lucks, J. Matolich, and J. A. Van Velzor, "The experimental measurement of thermal conductivities, specific heats, and densities of metallic, transparent, and protective materials, Part III" Wright Air Development Center AF Tech. Rept. 6145, Part III, March 1954. 71 p.; AD-95406. We have used the data for vitreous silica from Tables 7 and 8.

20. W. D. Kingery and F. H. Norton, "The measurement of thermal conductivity of refractory materials. Quarterly Progress Report for the Period Ending January 1, 1955." U. S. At. Energy Comm. Rept. NYO-6147 (1955), 16 p.; AD-55595. Data are given in Fig. 4 of this paper. They extend from 95 to 1214°C (368 to 1487°K), but not all have been shown in our Fig. 3. Those below 700°C are uniformly spaced, and we have omitted them on the assumption that they represent the same data that are presented in reference (8). The data at 700°C and above are less regular and we have accepted them as new data. The 3 highest-temperature points in the present reference are omitted because they have k-values beyond the range of Fig. 3.

21. V. V. Pustovalov, "Change of thermal conductivity of quartz glass in the process of crystallization," Steklo i Keram. 17, No. 5, 28-30 (1969). The plotted points from the top curve in Fig. 3 of this paper have been read off and used in our report. The paper has been translated into English (reference 68).
22. Benjamin P. Colosky, "Thermal conductivity measurements on silica," Bull. Am. Ceram. Soc. 31, 465-6 (1952). Figure 2 contains the experimental data. The two upper curves refer to crystalline quartz. The bottom curve represents data obtained with 3 different samples of vitreous silica. Two of these were clear and one was smoky, but there is no significant difference between the k-values of the 3 samples. The points for all three have been read from the graph and plotted in our Fig. 3.

23. W. J. Knapp, "Thermal conductivity of nonmetallic single crystals," J. Am. Ceram. Soc. 26, 48-55 (1943). The vitreous-silica data from Table II of this paper are plotted in our Fig. 3, except for the point of highest T and k, which comes beyond the range of the figure.

Containing Data Not Plotted in the Figures

These references contain the data judged to be less important than those in references (1) to (23). The arrangement is alphabetical. Each of the references in this group is followed by a brief annotation, in which the k-values reported in the paper are included. Numerical values given below are in cal cm⁻¹ sec⁻¹°C⁻¹.

24. Stanley S. Ballard, Kathryn A. McCarthy, and William C. Davis, "A method for measuring the thermal conductivity of small samples of poorly conducting materials such as optical crystals," Rev. Sci. Instr. 21, 905-7 (1950). A relative method was used, but the specific reference material used in each experiment is not stated. For vitreous silica they found k = 0.00282 at 41°C, using a "Homosil" sample from the W. C. Heraeus Co.

25. Thomas Barratt, "Thermal conductivity. Part II: Thermal conductivity of badly-conducting solids," Proc. Phys. Soc. (London) 27, 81-93 (1914). For vitreous silica of density 2.17 g cm⁻³, the thermal conductivity was found to be 0.00237 at 20°C, and 0.00255 at 100°C.

26. A. Deck, "A steady state method for the rapid measurement of the thermal conductivity of rocks," J. Sci. Instr. 34, 186-9 (1957). A relative method was used, with brass (calibrated versus crystalline quartz) as the reference material. For vitreous silica at 28°C, the thermal conductivity obtained was 0.00323.
27. A. E. Benfield, "Terrestrial heat flow in Great Britain," Proc. Roy. Soc. (London) A 173, 428-50 (1939). A comparison method was used, with brass bars as the reference material. The brass was calibrated versus crystalline quartz, and the effect of resistance at contact surfaces was eliminated by using 4 samples of different thickness. The value of k found for vitreous silica (Vitreosil from the Thermal Syndicate) was 0.00517 at 21.1°C.

28. E. C. Bullard, "Heat flow in South Africa," Proc. Roy. Soc. (London) A 173, 474-502 (1939). The thermal conductivity of vitreous silica was measured as a check on the accuracy of the apparatus, which had been calibrated against a crystalline quartz sample. For vitreous silica at 25°C, Bullard found k = 0.00307.

29. J. H. Crawford, Jr. and M. C. Wittels, "Radiation stability of nonmetals and ceramics", Proc. 2d U. N. Int. Conf. Peaceful Uses Atomic Energy, Vol. 5, p. 300-10, 1958. They report k = 0.0035 at 30°C; there is also a graph (Fig. 7) of low-temperature values. This graph appears to be identical with one in reference (53); reference (13) is preferred over both.

30. A. Eucken, "The heat conductivity of ceramic refractory materials. Its calculation from the heat conductivity of the constituents," Forschungsh. 353 (Suppl. Forsch. Gebiete Ingenieurw. B 3, Mar. - Apr. 1932), 16 p. The data in this paper appear to be a selection of values based on previously published work of Eucken (5), Kaye and Higgins (14), and Seemann (15). However, if this is the case, the values ought to be about 20 percent higher. We cannot be sure that these values are not based in part on new unpublished data obtained by Eucken.

31. G. Gafner, "The application of a transient method to the measurement of the thermal conductivity of rocks and building materials," Brit. J. Appl. Phys. 6, 393-7 (1957). A method of "steady rise" was employed; three samples of vitreous silica were measured in one apparatus and three others in another. The mean of the 6 measurements was k = 0.00323 with a total spread of 6 percent; the temperature of all the measurements was 30°C.
32. M. Jakob, "The material properties most important in heat transfer," Der Chemie - Ingenieur, A. Eucken and M. Jakob, editors, Akad. Verlag, Leipzig, 1933. Vol. 1, Pt. 1, p. 308-79. The information on vitreous silica, p. 340-1, includes a smooth curve of thermal conductivity vs. temperature, but no plotted points. It seems likely that this curve is based on previously-published work of other authors, but we are not sure that this is the case.


34. John H. Koenig, "Progress Report No. VI from March 1 to June 1, 1952," N. J. Ceram. Research Sta., Rutgers Univ., 98 p.; AD-163519. This paper is of interest because it contains a comparison of thermal conductivities of clear and silky vitreous silica. For quantitative data on clear vitreous silica we prefer reference (11), in which the measurements were made with improved equipment.

35. John H. Koenig, "Progress Report No. 2 from March 1 to June 1, 1953," N. J. Ceramic Research Sta., Rutgers Univ., 117 p.; AD-13154. This paper contains a comparison of thermal conductivities of silky vitreous silica measured parallel and perpendicular to the striations. Although it is not clear which set of data refers to each form, the data show that there was not much difference between the two.
36. A. V. Luikov, L. L. Vasiliev, and A. G. Shashkov, "A method for the simultaneous determination of all thermal properties of poor heat conductors over the temperature range 80 to 500°C," in Advances in thermophysical properties at extreme temperatures and pressures, Third Symposium on Thermophysical Properties, ASME, Purdue Univ., Mar. 22-25, 1965, Serge Gratch, editor (The American Society of Mechanical Engineers, New York, 1965), p. 314-9. This paper contains some ambiguities or errors that have caused us not to include the data in any of our graphs. The density given for the vitreous silica is 2.650 g cm\(^{-3}\), which is 20 percent higher than accepted values (it is roughly the accepted density for crystalline quartz). The data of other observers, plotted in Fig. 6 of the present reference, appear to be incorrectly given, at least in part. Also the data for vitreous silica in the present reference are from 8 to 17 percent lower than those given by Vasiliev (6) one year earlier.

37. M. V. Milnes, "Low-temperature thermal conductivity apparatus," North American Aviation, Inc., Rept. SDL 408, 9 Sept. 1963. 14 p. A sample of vitreous silica was measured, presumably to check out the apparatus. The k-value found was .0033, and the sample mean temperature, which we computed from the given thermocouple data, was 23.1°C.

38. Shiro Nukiyma, "The thermal conductivity of glass, chilled glass, quartz, fused quartz (transparent), Bakelite, India-rubber, coal, Isolite, porcelain, slate, granite and marble," Trans. Soc. Mech. Engrs. (Japan) 2, 344-5 (1936). English abstract, ibid., p. S-95, S-97. Three plotted k-values for vitreous silica are given, covering the range 15 to 33°C. An equation is given that appears to represent the points satisfactorily. The equation gives, at 300°C (26.85°C):

\[ k = 0.00294 \]
39. L. S. Phillips, "The measurement of thermoelectric properties at high temperatures" J. Sci. Instr. 42, 209-11 (1965). The apparatus was so designed that the Seebeck coefficient, the electrical resistivity, and the thermal conductivity of a specimen could each be measured. The thermal conductivity of vitreous silica was measured in the range 160 to 595°C "to assess the behavior of the apparatus." The results are given in the form of a smooth curve. In view of this, and in view of the fact that the apparatus emphasized rapid operation rather than high accuracy, we have not plotted the results in any of our figures. At the lowest temperature, Phillips's curve is 5 percent below our master curve; at the highest temperature it is 18 percent above it.

40. R. W. Powell, "Thermal conductivity as a non-destructive testing technique," in Progress in Non-destructive Testing, E. G. Stanford and J. H. Fearon, editors (Heywood and Company, London, 1958), Vol. 1, p. 199-226. The k-value of vitreous silica at "about 28°C" is given as 0.00330 in Table 4 and again in Table 5. This same value of k was given earlier by Powell in reference (67). However, the earlier paper does not give the temperature of the measurement.


42. James L. Weeks and Ralph L. Seifert, "Note on the thermal conductivity of synthetic sapphire," J. Am. Ceram. Soc. 35, 15 (1952). Measurements made on a 6-mm rod of vitreous silica from Hanovia Chemical Company yielded 0.0041 at 73°C and 0.0057 at 135°C.

44. R. Berman, "Thermal conductivity of glasses at low temperatures," Phys. Rev. 76, 315-6 (1949). This appears to be the place of first publication of Berman's data on vitreous silica. However, we prefer to use the fuller account, reference (12).


46. R. Berman, "Thermal conductivity of some polycrystalline solids at low temperatures," Proc. Phys. Soc. (London) A 65, 1029-40 (1952). This paper contains 7 tabulated values of k of vitreous silica. Six are at low temperatures, and agree with those in reference (12), which is preferred. The remaining value is for room temperature, and we have not found it elsewhere in Berman's published papers; however, it is given to only one significant figure, and we have not used it.

47. R. Berman, "The thermal conductivity of dielectric solids at low temperatures," Advances Phys. (London) 2, 103-40 (1953). This paper contains a smooth curve for vitreous silica. Reference (12) is preferred.


49. V. S. Bil' and N. D. Avtokratova, "Temperature dependence of thermal conductivity and thermal diffusivity of some unfilled polymers," Teplofiz. Vysokih Temperatur, Akad. Nauk S. S. R. 2, 192-8 (1964). We have not yet obtained a copy of this paper; it is the Russian original of (9).

51. A. Foner Cohen, "Low-temperature thermal conductivity in neutron-irradiated vitreous silica," U. S. At. Energy Comm. Oak Ridge Natl. Lab. Rept. ORNL-2413, 70-1 (Aug. 31, 1957). This appears to be the same work as that reported in reference (13), which we prefer.


53. Anna Foner Cohen, "Low-temperature thermal conductivity in neutron irradiated vitreous silica," J. Appl. Phys. 29, 591-3 (1958). This paper contains less than half as many plotted points as (13), although it covers a greater temperature range. We prefer reference (13), which appears to be a later and better report of the work.


56. A. Eucken, "The heat conductivity of solid materials at low temperatures," Z. Tech. Physik 6, 689-94 (1925). Reference (5) is preferred. The present paper contains a value at 21°K that is not given in reference (5) and another at 373°K that differs from the value given in reference (5). We have omitted the 21° value of the present report on the assumption that it is not an original determination but an extrapolation, and we have omitted both of the 373° values given by Eucken because they do not agree. The remaining values in the present reference agree with those in reference (5).


59. W. D. Kingery, "Factors affecting thermal stress resistance of ceramic materials," J. Am. Ceram. Soc. 38, 3-15 (1955). This paper contains two numerical k-values for vitreous silica; however, they appear to be only incidental to the subject of the paper. We prefer reference (8).

60. W. D. Kingery, "Thermal conductivity: XII, Temperature dependence of conductivity for single-phase ceramics," J. Am. Ceram. Soc. 38, 251-5 (1955). This paper is similar to reference (8); it has a better description of experimental techniques than reference (8).


62. P. G. Klemens, "The thermal conductivity of dielectric solids at low temperatures (Theoretical)", Proc. Roy. Soc. (London) A 208, 108-33 (1951). The k-data on vitreous silica in Fig. 2 of this paper are from Berman (12). However, the data in the present reference are less numerous than those in Berman's paper, which we prefer. In addition, the data in the present reference have been "adjusted."

63. John H. Koenig, "Progress Report No. 1 from December 1, 1952 to March 1, 1953," N. J. Ceram. Research Sta., Rutgers Univ., 100 p; AD-5552. This paper contains k-data for vitreous silica. However, we consider that they are superseded by the data reported in reference (11).

64. D. W. Lee and W. D. Kingery, "Radiation energy transfer and thermal conductivity of ceramic oxides," J. Am. Ceram. Soc. 43, 594-607 (1960). Reference (20) is preferred. However, the present reference contains a somewhat fuller description of the experiments than reference (20).
65. C. F. Lucks, H. W. Deem, and W. D. Wood, "Thermal properties of six glasses and two graphites," Bull. Am. Ceram. Soc. 39, 313-9 (1960). After studying this paper and plotting the k-values given for vitreous silica, we have concluded that the data are smoothed values taken from the original data of reference (19).

66. F. H. Norton, W. D. Kingery, "et al.", "The measurement of thermal conductivity of refractory materials," U.S. At. Energy Comm. Rept. NYO-3646, 1-8 (July 1, 1953). Reference (8) is preferred. Although the present reference appears to be the earliest report of this work, its graph of the data is smaller than the graph in reference (8).

67. R. W. Powell, "Experiments using a simple thermal comparator for measurement of thermal conductivity, surface roughness and thickness of foils or of surface deposits," J. Sci. Instr. 34, 485-92 (1957). Vitreous silica was one of the materials used in standardizing the apparatus. The k-value given here is also given in reference (40). Reference (40) is preferred because it contains the temperature of the measurement, whereas no temperature is given in the present reference.

68. V. V. Pustovalov, "Change in thermal conductivity of quartz glass in the process of crystallization," Translation by A. J. Peat, General Electric Research Lab. Rept. 60-RL-(2525M), September 1960. 8 p. English translation of reference (21). Graph showing data is smaller than the one in reference (21).

69. V. V. Pustovalov, "Thermal conductivity of some refractory materials," Ogneupory 26, 302-5 (1961). The unsmoothed values of reference (21) are preferred. However, the present paper gives the density of the vitreous-silica sample, from which we computed a greater porosity than that given by Pustovalov.

70. E. H. Ratcliffe, "Preliminary measurements to determine the effect of composition on the thermal conductivity of glass," Phys. Chem. Glasses 1, 103-4 (1960). Reference (2) is preferred. The smooth curve in the present reference agrees with the data in reference (2). It goes to a higher temperature, but we have assumed that this is an extrapolation of the previous results.
71. E. H. Ratcliffe, "A survey of most probable values for the thermal conductivities of glasses between -150 and 100°C, including new data on twenty-two glasses and a working formula for the calculation of conductivity from composition," Glass Technol. 4, 113-28 (1963). Reference (2) is preferred. The present reference contains a short table of "assessed best" values for vitreous silica. These values average 1.0 percent above our master curve.


Containing No Original Thermal Conductivity Data on Fused Silica


Thermal Conductivity of Vitreous Silica: Selected Values

July 1966

Lois C. K. Carwile and Harold J. Hoge

The published literature on the thermal conductivity of vitreous silica has been assembled and the results critically evaluated. Best values of thermal conductivity as a function of temperature have been selected. These are presented in both graphical and tabular form. They cover the range 80 to 660° K and published data extending from 3° to 2100° K are shown in the graphs. An attempt was made to consult all work that could significantly affect the choice of best values. Published papers were located with the aid of Chemical Abstracts, Physics Abstracts, the Thermophysical Properties Retrieval Guide, and some other general sources. In addition, relevant references in the papers themselves were followed up until a substantially "closed system" had been generated, as shown by the fact that no new references were being turned up.
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