UCRL-16152

University of California

Ernest O. Lawrence Radiation Laboratory

THERMAL SHOCK RESISTANCE OF TRANSPARENT CERAMICS, I.

Berkeley, California

Submitted to J. American Ceramics Society

UCRL-16152

UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
Berkeley, California

AEC Contract No. W-7405-eng-48

THERMAL SHOCK RESISTANCE OF TRANSPARENT CERAMICS, I.

D. P. H. Hasselman

July 8, 1965

THERMAL SHOCK RESISTANCE OF TRANSPARENT CERAMICS, I.

D. P. H. Hasselman

Inorganic Materials Research Division, Lawrence Radiation Laboratory, and Department of Mineral Technology, College of Engineering, University of California, Berkeley, California

July 8, 1965

ABSTRACT

The hypothesis is advanced that semitransparent ceramics under conditions of radiation heating have greater resistance to fracture by thermal shock than opaque ceramics. In semitransparent ceramics radiant heat can be transferred within the specimen being heated, rather than being absorbed at the surface. Temperature differences within the specimen are therefore reduced, resulting in a lower thermal stress level.

An approximate derivation is presented for the maximum temperature of a surrounding black-body source to which spherical shapes can be submitted without failure composed of materials the infrared absorption properties of which can be described to a good approximation to be completely transparent below a given wave length and entirely opaque above this wave length. Thermal stress resistance parameters are obtained. A calculation comparing the thermal shock resistance of transparent and opaque alumina spheres shows the transparent spheres to be far superior.

At the time this work was done the writer was graduate research assistant, Inorganic Materials Research Division, Lawrence Radiation Laboratory, and graduate student, Department of Mineral Technology, University of California, Berkeley, California.

I. INTRODUCTION

The thermal shock resistance of most refractory ceramics is quite low, generally due to low strength, low thermal conductivity, high coefficients of thermal expansion, and high values of Young's modulus of elasticity. Many theoretical studies[1-5] have shown that even in the best ceramic materials the maximum temperature difference that can be withstood within the ceramic body is quite low.

Recently a number of refractory oxides have been developed, such as alumina and magnesia, which are transparent over a considerable range of that part of the electromagnetic spectrum found in heat transfer by radiation at temperatures usually encountered in ceramic technology. At these temperatures (i. e., > 1000° C), as shown elsewhere, the contribution to the heat flux by natural convection is negligible, the transfer of heat occurring primarily by radiation. It is suggested here that under these conditions "transparent" ceramics can be employed advantageously in many applications when fracture by the thermal shock constitutes a major problem. For transparent or semitransparent ceramics, the thermal radiation may traverse the material over a considerable distance. The transfer of heat can take place throughout the interior of the body rather than at the surface only. As a consequence, the body is heated more uniformly, thereby avoiding large temperature differences.

The present paper is the first paper in what is intended to be a series discussing the various theoretical and experiments aspects of the thermal shock resistance of transparent ceramics. In general, the calculation of temperature distribution in semitransparent materials heated by radiation is quite complex, due to variations in reflectivity with angle of incidence, "focusing effects" due to refraction effects, multiple reflections within the body, and variations with wave length of most of the optical properties.

This paper presents an approximate treatment of the thermal shock resistance of ceramics whose absorption properties to a good approximation can be described as being completely transparent below a certain wave length and completely opaque above this wave length.

II. THEORETICAL

For the computation of the thermal stresses, discussion will be confined to specimens of spherical shape at low initial temperature. In addition, the specimens are considered to be submitted instantaneously into a radiation source completely surrounding the specimen. Also, the specimen size is considered to be small compared to the surrounding radiation source. Under these conditions the emissivity of the source does not have to be taken into account and allows a "black-body" approximation to the radiant heat emitted.

Figure 1 shows the transmission characteristics as a function of wave length of three ceramic oxides.[6] At short wave lengths, transmission takes place virtually unimpeded, the loss through the specimens primarily due to reflection at the surfaces. At longer wave lengths, however, due to absorption, transmission decreases very rapidly and becomes virtually nonexistent even for the relative thin specimens used. These absorption characteristics can also be shown for alumina by plotting absorption coefficient at 500°C against wave length,[6] as shown in Fig. 2. The absorption coefficient can be seen to increase by three orders of magnitude over a relatively narrow range of wave length.

The change in intensity of electromagnetic radiation in an absorbing medium can be expressed by[7]

$$I = I_o e^{-\alpha x} \tag{1}$$

where I = the intensity of the wave after propagation over a distance x,

I_o = the original intensity of the incident wave as it enters the surface,

and α = absorption coefficient.

Equation (1) reveals that for sizes of specimens employed in a laboratory or practice, say greater than two inches, that for high values of absorption coefficient ($\alpha \leq 0.1$ mm^{-1}) most of the radiation penetrates the body over a distance small compared to the body size. The material then can be regarded as practically opaque. For low values of α, however, the radiation propagates through the body practically undiminished and for practical purposes can be considered transparent. Due to the narrow wave length interval, Fig. 1 suggests that for silica, alumina, and magnesia the wave length separating the transparent region from the opaque region occurs at 4μ, 5μ, and 7μ, respectively. The high absorption at very short wave length, for instance for alumina at $\lambda < 0.16\mu$, can be neglected since the contribution to the total radiant heat flux at wave lengths less than this value can be neglected at the temperatures of radiation sources usually encountered in practice. Similarly, transparency at very long wave lengths can also be neglected for the spectral distribution of the radiation for almost all sources, but those at very low temperatures, which are of no consequence in the present discussion. The absorption properties of alumina and magnesia only increase slightly with temperatures, and a "cut-off" wave length of 5 and 7μ, respectively, will still hold. Silica, however, tends to become opaque; and unless the specimen is at low temperatures, the above discussion will require modifications.

From the above, in the opinion of the author, it can be concluded that under the conditions as described (i.e., specimen size, temperature of radiation source) that to a good approximation the absorption characteristics of the materials shown in Fig. 1 can be described to be transparent below a certain wave length and opaque above this wave length. A calculation of the actual heat flux to which a specimen is subjected, only that fraction of the total energy has to be considered which occurs at wave lengths above the "cut-off" wave length (λ_o).

The calculation of the fraction of the energy above a certain wave length (λ_o) to the total energy emitted by a radiation source can be calculated as follows: the radiant energy as a function of wave length can be expressed by the Planck equation:[8]

$$W_\lambda = \frac{C_1 \lambda^{-5}}{e^{C_2/\lambda T} - 1} \qquad (2)$$

where C_1 and C_2 are constants,

λ is the wave length,

and T is the absolute temperature.

The total energy emitted by a black body at temperature T can be obtained by integration of Eq. (2) from $\lambda = o$ to $\lambda = \infty$ which results in

$$W_{\lambda = o \rightarrow \infty} = \rho T^4 \quad (3)$$

where ρ is the Stefan-Boltzmann's constant.

The fraction F_{λ_o} of energy below the "cut-off" wave length (λ_o) can be defined:[9]

$$F_{\lambda_o} = \frac{W_{\lambda = o \rightarrow \lambda_o}}{W_{\lambda = o \rightarrow \infty}} \quad (4)$$

which upon substitution of Eq. (3) results in

$$W_{\lambda = o \rightarrow \lambda_o} = F_{\lambda_o} \rho T^4 . \quad (5)$$

The total heat flux at wave lengths $\lambda \geq \lambda_o$ then becomes

$$W_{\lambda = \lambda_o \rightarrow \infty} = (1 - F_{\lambda_o}) \rho T^4 \quad (6)$$

Table II lists values* of F_{λ_o} for a series of values of $\lambda_o T$. For instance, at 2000°K, 91.4% of the radiant energy is emitted at wave lengths shorter than 5μ.

With the knowledge of the heat flux, the thermal stresses can now be derived. Assuming the specimen to have a low initial temperature, the heat emitted by the specimen during the time until maximum stress can be neglected and allows the "constant-heat-flux" approach derived elsewhere.[5] The rate of heat absorption (q) by the specimen surface equals

$$q = (1 - F_{\lambda_o}) \epsilon \rho T^4 \quad (7)$$

where ϵ is the total hemispherical absorbtivity in the range of wave length where the material is opaque.

The maximum heat flux (q_{max}) to which a sphere can be subjected as derived previously[5] can be written

$$q_{max} = \frac{5 S_{ts} (1 - \upsilon) k}{b \alpha E} \qquad (8)$$

where S_{ts} = tensile strength

υ = Poisson's ratio

k = thermal conductivity

b = sphere radius

α = coefficient of thermal expansion

E = Young's modulus of elasticity.

The maximum radiation temperature (T_{max}) to which a sphere can be subjected can be expressed in terms of the maximum heat flux by:

$$q_{max} = (1 - F_{\lambda_o}) \epsilon \rho T_{max}^4 \qquad (9)$$

Substitution of Eq. (9) with Eq. (8) and rearranging yields:

$$T_{max} = \left[\frac{5}{\rho b}\right]^{\frac{1}{4}} \left[\frac{S_{ts} (1 - \upsilon) k}{\alpha E \epsilon (1 - F_{\lambda_o})}\right]^{\frac{1}{4}} \qquad (10)$$

Expressions similar to Eq. (10) can be derived for the infinite cylinder or flat plate, the constant being replaced by 4 or 3, respectively.

A thermal stress resistance parameter for transparent ceramics with the optical characteristics, as shown in Fig. 1, can be defined by:

$$R_{trans} = \left[\frac{S_{ts}\,(1 - \upsilon)\,k}{\alpha\, E\, \epsilon\,(1 - F_{\lambda_o})}\right]^{\frac{1}{4}} \qquad (11)$$

which, but for the factor $(1 - F_{\lambda_o})$, is identical to the thermal stress parameter R_{rad} for completely opaque ceramics.[5]

III. NUMERICAL EXAMPLE

At this time, as far as the author is aware, no experimental data exist to test the theory developed above. Instead, a sample calculation is presented for the maximum temperature to which transparent alumina spheres can be subjected and compared with the thermal shock behavior of opaque alumina spheres. The same external conditions will be assumed as for the theoretical derivations, namely, spheres at initial low temperature suddenly introduced in a completely enclosed black-body environment at high temperature. Table II lists the physical property values assumed for the calculation and are typical for dense polycrystalline alumina. For a given sphere size, T_{max} can be calculated by means of Eq. (10). This calculation, however, due to the strong temperature dependence of the factor $(1 - F_{\lambda_o})$, requires the laborious technique of successive approximations and also necessitates a table of values of $\lambda_o T$, considerably more extensive than Table I. A graphical technique, similar to the one employed elsewhere,[4] was used to overcome this difficulty. The heat absorbed per unit area at the surface of the specimen is plotted against the temperature of the black-body enclosure. With the known value of the maximum heat flux (q_{max}) calculated by means of Eq. (8), the maximum temperature is readily obtained from the temperature axis. Figure 4 illustrates the graphical technique for both transparent and opaque spheres. The superior thermal stress resistance of the transparent ceramics compared to the opaque ceramics is quite apparent. It appears to be quite difficult to fracture 2 in. and 4 in. diam. spheres made of transparent alumina, whereas the same spheres of opaque alumina can be fractured quite readily using temperatures commonly encountered in the laboratory. Even a 6 in. diam sphere of the transparent alumina would require a temperature in excess of its melting point for fracture to occur.

In Fig. 4 it is of interest to note that for the transparent spheres the heat absorbed rises approximately linearly with temperature rather than as the fourth power of temperature. This is due to the fact that the contribution of heat flux at successively higher temperatures more and more occurs at wave lengths shorter than the "cut-off" wave length of 5 μ.

Similar calculations can be carried out for other materials. Due to its relative high "cut-off" frequency, magnesia should be superior in thermal stress resistance to both alumina and silica under the conditions assumed.

IV. DISCUSSION AND CONCLUSIONS

The results of the numerical example appear to support the hypothesis of the superior thermal stress resistance of transparent ceramics as compared to opaque ceramics. The assumption of a sharp "cut-off" wave length conceivably could introduce some error. However, the small quantity of radiation which penetrates the specimen to some degree in the region assumed opaque is compensated for the partial absorption of the radiation in the region considered completely transparent. For materials in which the absorption coefficient rises slowly with wave length, the theory presented above would require extensive modifications, if applicable at all.

Many refinements are possible to make a more precise prediction of the thermal stress resistance. Inclusion of the temperature dependence of the physical properties of the ceramic material in the calculation could lead to a considerable improvement. The dependence of emissivity on wave length, such as shown in Fig. 3, might be taken into account. In the numerical example presented, emissivity was assumed constant and equal to 0.80. The decrease in emissivity at longer wave lengths suggests that (T_{max}) is underestimated. More refined calculation of the heat flux absorbed at the surface could take place by summing the contribution to the heat flux over each region where the emissivity can be considered constant. The spectral dependence in the equation can also be included in the expression for (T_{max}). However, examination will reveal that the calculation of (T_{max}) then becomes rather tedious and involved.

The theory presented can also be applied to materials with different absorption characteristics. For instance, for a material with high absorption coefficient between wave lengths λ_a and λ_b $(\lambda_b > \lambda_a)$ and practically transparent at all other wave lengths, the heat absorbed at the surface (q) becomes:

$$q = (F_{\lambda_a} - F_{\lambda_b}) \epsilon \rho T^4 \qquad (12)$$

F_{λ_a} and F_{λ_b} being defined by Eq. (4) by replacing λ by λ_a, λ_b, respectively.

The expression for T_{max} then becomes:

$$T_{max} = \left[\frac{5}{\rho b}\right]^{\frac{1}{4}} \left[\frac{S_{ts}\,(1 - \upsilon)\,k}{\alpha\, E\, \epsilon (F_{\lambda_a} - F_{\lambda_b})}\right]^{\frac{1}{4}} \qquad (13)$$

The actual experimental verification for the present theory may be difficult to perform. As shown by the results in Fig. 4, relatively large specimens of polycrystalline alumina are required if fracture by the thermal shock will occur at all. The preparation of sufficient specimens of sufficient size and of various shapes at the present state of the art might be rather difficult. From these results, however, it does appear that shapes and objects of a size usually encountered in the laboratory can be used without failure by thermal shock.

ACKNOWLEDGMENTS

The author is grateful to Messrs. Fulrath and Pask for helpful discussions and comments. The author should also like to acknowledge the Research and Development Laboratories of The Carborundum Company, Niagara Falls, where the basic idea of using transparent ceramics for thermal shock applications was initially conceived.

This work was done under the auspices of the U. S. Atomic Energy Commission.

REFERENCES AND FOOTNOTES

1. B. Schwartz, "Thermal Stress Failure of Pure Refractory Oxides," J. Am. Ceram. Soc., 35 [12] 325-33 (1952).

2. W. D. Kingery, "Factors Affecting Thermal Stress Resistance of Ceramic Materials," J. Am. Ceram. Soc., 38 [1] 3-15 (1955).

3. W. B. Crandall and J. Ging, "Thermal Shock Analysis of Spherical Shapes," J. Am. Ceram. Soc., 38 [1] 44-54 (1955).

4. D. P. H. Hasselman and W. B. Crandall, "Thermal Shock Analysis of Spherical Shapes," J. Am. Ceram. Soc., 46 [9] 434-37 (1963).

5. D. P. H. Hasselman, "Thermal Shock by Radiation Heating," J. Am. Ceram. Soc., 46 [5] 229-34 (1963).

6. "Optical Grade Sapphire," Linde Crystal Products Bulletin No. F-917-D. Union Carbide Corporation, Linde Division, East Chicago, Indiana.

7. M. Born and E. Wolf, Principles of Optics, 2nd ed., p. 614. The MacMillan Company, 1964. 808 pp.

8. F. K. Richtmyer, E. H. Kennard, and T. Lauritsen, Introduction to Modern Physics, 5th ed., p. 130. McGraw-Hill, 1955. 666 pp.

9. T. R. Harrison, Radiation Pyrometry and Its Underlying Principles of Radiant Heat Transfer, pp. 24-25. John Wiley, 1960. 234 pp.

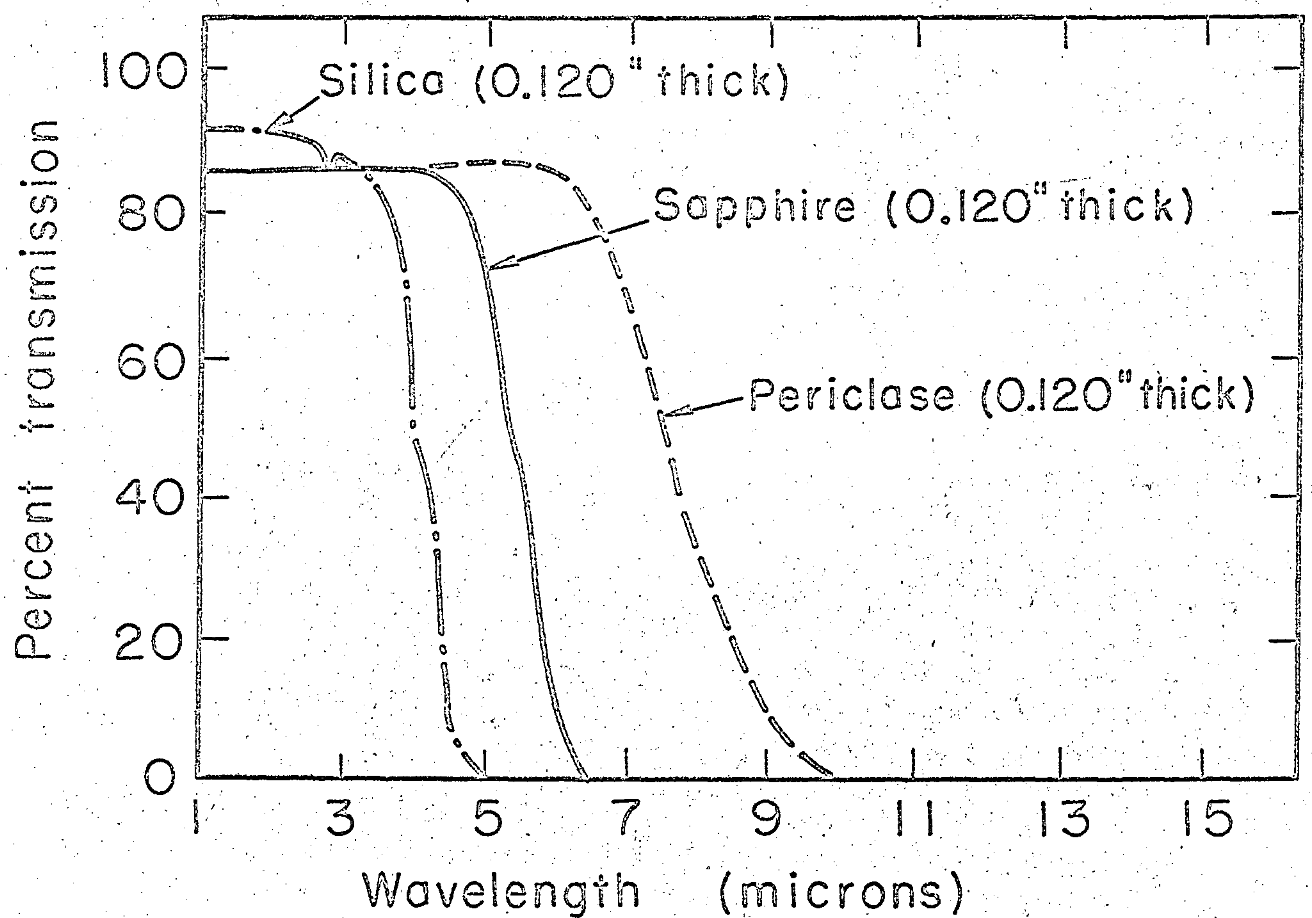
* Values listed in Table II were obtained from Reference 9.

Table I. Fraction (F_{λ_o}) of radiant energy from a black-body source at temperature (T) for a range of wave length from 0 to λ_o microns

$\lambda_o T$	F_{λ_o}	$\lambda_o T$	F_{λ_o}
500	0.129×10^{-8}	11,000	0.9318
1,000	3.207×10^{-4}	12,000	0.9451
2,000	6.673×10^{-2}	13,000	0.9551
3,000	0.2732	14,000	0.9629
4,000	0.4809	15,000	0.9689
5,000	0.6337	16,000	0.9738
6,000	0.7378	17,000	0.9777
7,000	0.8081	18,000	0.9808
8,000	0.8563	19,000	0.9834
9,000	0.8899	20,000	0.9856
10,000	0.9142	∞	1.0000

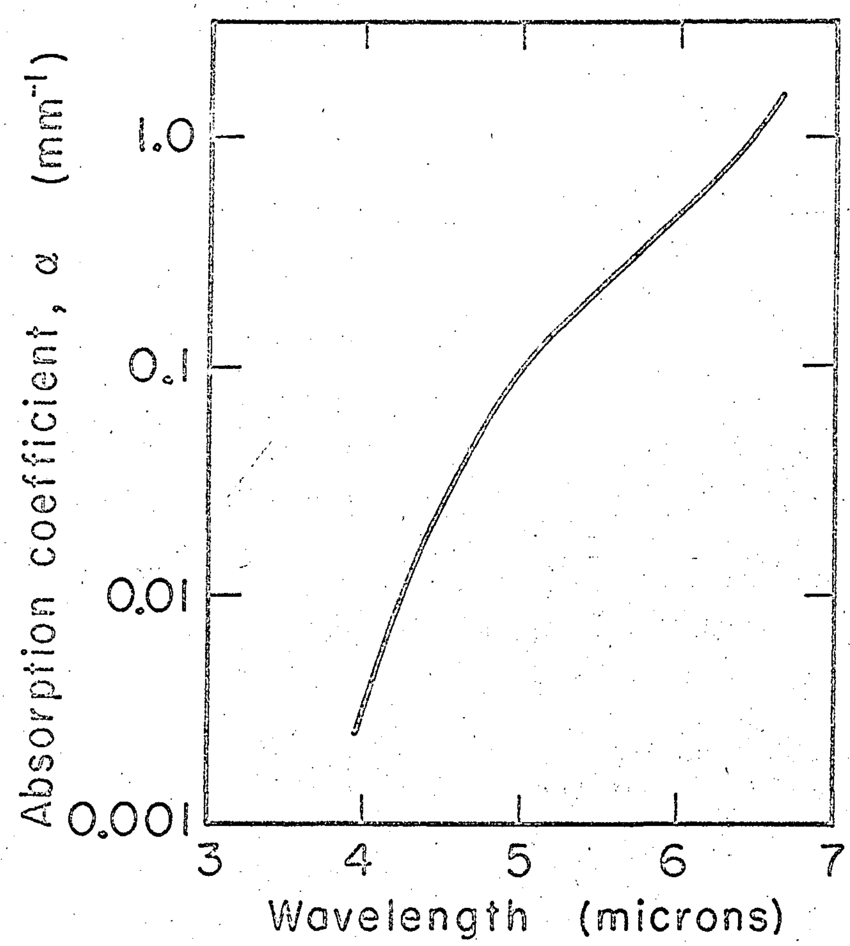
Table II. Physical properties of typical polycrystalline alumina ceramic

Tensile strength (S_{ts})	=	2.07×10^9 dynes/cm^2 (30,000 psi)
Thermal conductivity (k)	=	0.05 cal cm^{-1} sec^{-1} °K^{-1}
Poisson's ratio (υ)	=	0.27
Coefficient of thermal expansion (α)	=	7.0×10^{-6} °K^{-1}
Young's modulus (E)	=	4.14×10^{12} dynes/cm^2 (60×10^6 psi)
Emissivity	=	0.80



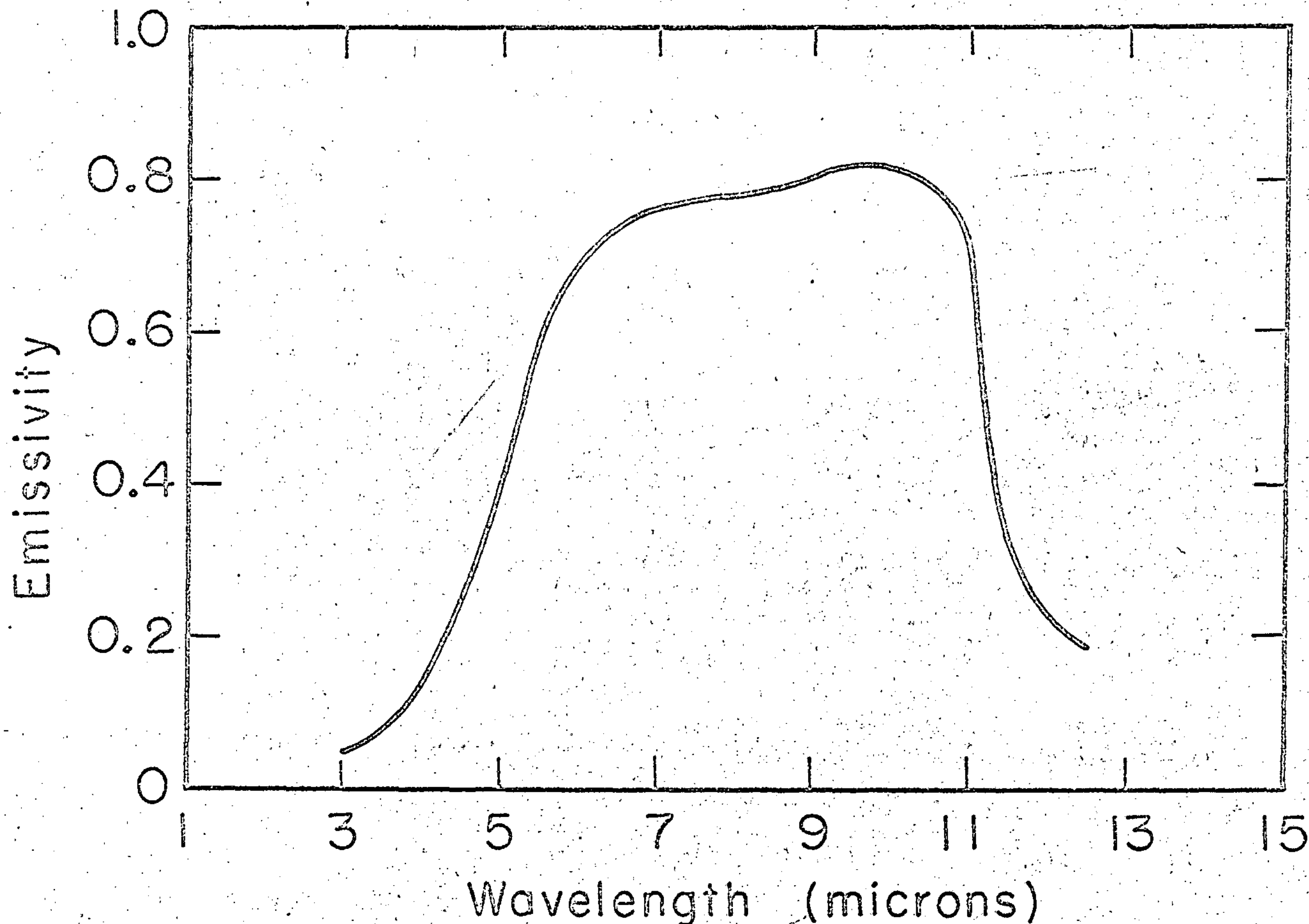
MUB-6398

Fig. 1. Spectral absorption characteristics of silica, sapphire, and periclase at room temperature (from footnote 6).



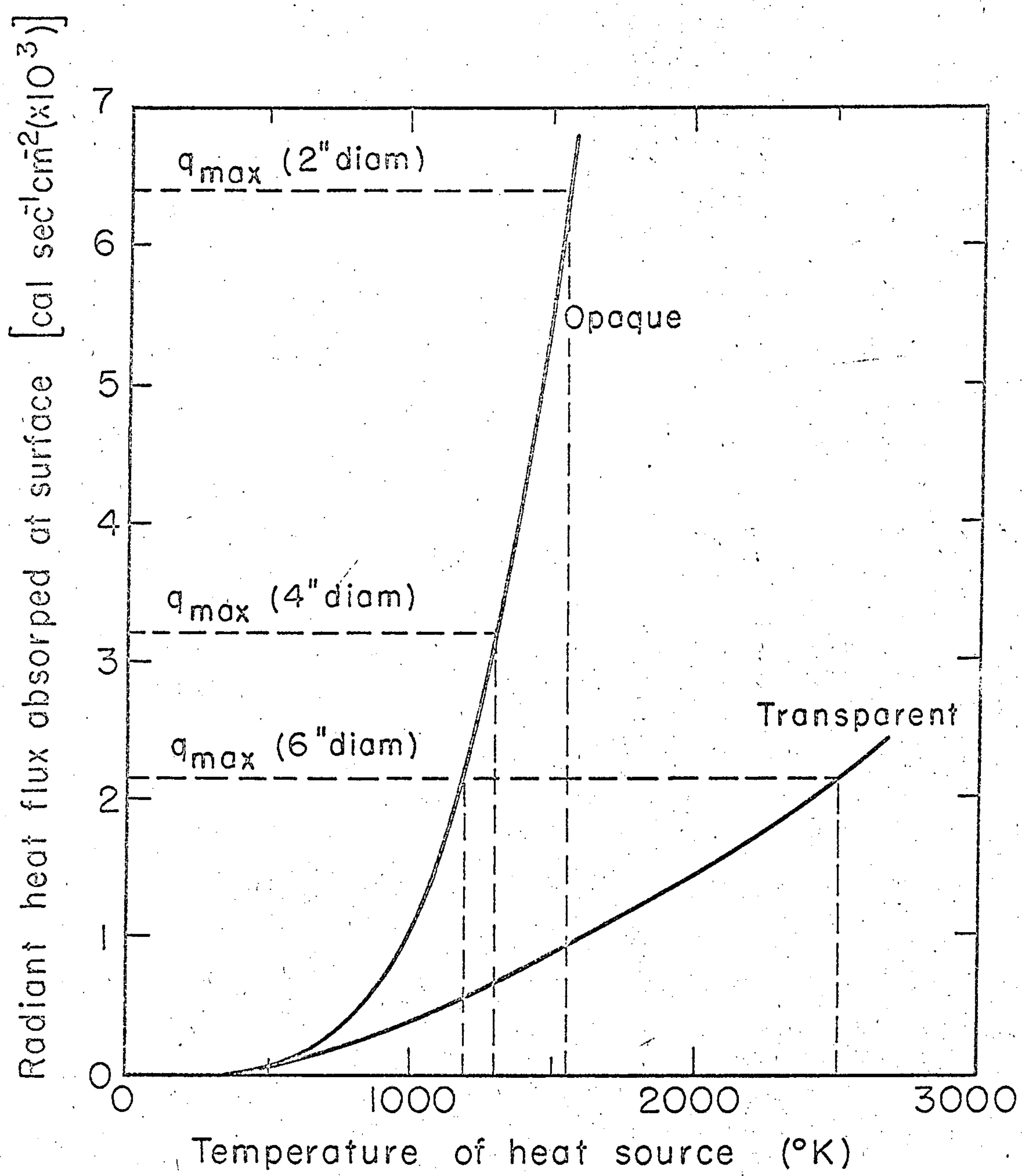
MUB-6397

Fig. 2. Absorption coefficient of sapphire at 500°C (from footnote 6).



MUB-6399

Fig. 3. Spectral emissivity of sapphire at 500°C (from footnote 6).



MUB-6400

Fig. 4. Graphical technique for determination of (T_{max}) for opaque and transparent alumina spheres.