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Method for Measuring High Temperature Spectral Emissivity of Nonconducting Materials

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A new method, based upon the integral blackbody technique, for measuring the normal spectral emissivity of nonconducting materials at high temperatures is described. A small cylindrical sample (5.21 mm diam × 9.53 mm length) is mounted within a long, thin-walled, metallic tube that is heated by the passage of direct current. Lateral holes in the tube wall permit an optical system to alternately detect the spectral radiance from the sample and from a cavity when both are at the same temperature. The major sources of error have been analyzed and the overall performance evaluated. Data are presented on a series of new, commercial ceramics, silicon carbide and silicon nitride, for the spectral region 1.5-15 μm and the temperature range 1300-1900 K. Temperature levels to 3000 K have been achieved with a specially configured graphite composite sample. These results indicate that the method provides a useful tool for studying the effects of fabrication processes and environmental influences on the radiative properties of materials at high temperatures.

Nomenclature

h = separation (radial) between sample and heating tube
H = detector signal proportional to blackbody radiance
k = thermal conductivity
L_λ = spectral radiance
r = radial coordinate
r_0 = cavity (lateral tube opening) radius
S = detector signal proportional to sample radiance
T = temperature, K
Z = detector signal proportional to cold reference blackbody radiance (background radiation)
ϵ = emissivity
λ = wavelength, μm

Subscript

λ = spectral condition

Introduction

RELIABLE data on the high temperature radiative properties of technical materials, particularly newer alloys, is lacking. The effects on the material properties due to thermal cycling, environmental interactions, and fabrication methods are known to be important. Without a body of experimental data generated under well-characterized conditions, rational prediction of material behavior is not possible. High technology applications such as turbine blades, combustion liners, laser weaponry, aerospace equipment, and electronic devices have stringent requirements for radiative properties of advanced materials, particularly nonconducting ones, at elevated temperatures. Unfortunately, few laboratories have the capability to provide these measurements.

The ceramics of silicon carbide and silicon nitride are typical of the new generation of materials. Improvements in their high temperature mechanical and thermal properties have resulted from modified hot-pressing fabrication methods using various dopants and temperature treatments. Graphite composites are another class of materials which, through new fabrication methods, have superior strength-to-weight characteristics at high temperatures. As will be shown in the results of the present study, the thermal radiative properties are strongly dependent upon the fabrication method. To effectively utilize these advanced materials there is a need for base-line data to study their behavior at high temperature.

Spectral emissivity is one of the most basic properties for describing material behavior at high temperatures. The shape of the spectra can provide information on oxidation effects, impurity migration, and structural changes. Furthermore, if the spectral region is selected properly, the total emissivity, most useful for heat exchange calculations, can be derived simply. Recall that spectral emissivity is defined as the ratio of the spectral radiance emitted from the sample to that from a blackbody when both are at the same temperature. There are two direct radiometric methods used to measure spectral emissivity. The first is the separate blackbody method wherein the sample and blackbody are physically separated, with their temperatures being carefully controlled.[1] The second is the integral blackbody technique wherein the sample and blackbody are in close proximity to one another in a nearly isothermal region. For temperatures below 1800 K in the infrared region, the requirement of matching the sample and blackbody temperatures can be met. Hence, the separate blackbody method is the preferred approach and can be realized, in general, by a simpler experimental apparatus. It should be recognized that nonmetallic materials pose special problems for both these methods, since their low thermal conductivities can cause temperature gradients during the heating process.[2] The rotating cylindrical sample technique of Clark and Moore[3] and the shallow-blackbody cavity technique of Moore[4,5] are studies wherein systematic errors due to temperature gradients have been considered carefully.

The emissometer used in the present investigation was initially developed for determining the normal spectral emissivity of conducting samples.[6] The method is based upon the integral blackbody technique wherein the blackbody cavity is formed by a lateral hole in a thin-walled, direct-electrically heated tube fabricated from the sample material.[7] To extend the method to nonmetallics, a short, solid-cylindrical sample is mounted within the tube, near the blackbody cavity opening, but behind a second lateral hole so that the spectral radiance from the nonconducting sample can be sensed.

The results of an investigation on several commercial formulations of silicon carbide and silicon nitride are presented. A detailed characterization of the samples including their composition and method of fabrication accompanies the data. Spectral emissivity values are given for

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the temperature range 1300-1900 K and wavelength region 1.5-15 μm. Results on an advanced graphite composite near 3000 K are also presented and described. The objective of this investigation was to determine if the spectral emissivity was markedly dependent upon temperature.

Measurement Method

Principle

Spectral emissivity is defined as the ratio of the spectral radiance emitted from the sample surface to that from a blackbody when both are at the same temperature. In this integral blackbody method, the sample-blackbody configuration is at the midsection of a long metallic tube as shown in Fig. 1. The upper and lower ends of the metallic tube are clamped to water-cooled electrodes. For measurement of the spectral emissivity of conducting materials, the sample is of thin-walled tubular form. The blackbody target is formed by a small lateral hole on the sample tube surface (*H*) and the cavity is the volume within the tube. The spectral emissivity is then determined from the ratio of radiometric observations made on three target areas

$$\epsilon_\lambda = (S-Z)/(H-Z)$$

where *S* is the signal proportional to the spectral radiance from the sample, *H* is the signal proportional to the blackbody spectral radiance, and *Z* the signal from the cold reference blackbody, serving as the radiometric zero for the optical system. This is used to account for background radiation. The values for *Z* are a function of wavelength and their magnitude typically ranges from 1 to 10% of the values for *H*. For conducting materials, this method has been well studied by DeVos,[8] Larrabee,[9] Riethof,[10] and Rodgers.[11]

For measuring the spectral emissivity of nonconducting materials, the sample-blackbody configuration is as shown in Fig 2. The sample, in solid-cylindrical plug form (5.21 mm diam × 9.53 mm length), is supported within the heating tube by a lateral pin. The sample is closely fit to the inner diameter of the tube so that isothermal conditions can be maintained. A lateral hole (1.57 mm diam) near the midsection of the heating tube allows the sample to be viewed by the optical system of the emissometer.

Emissometer

The emissometer represents an added capability to the multiproperty apparatus which has been described previously in detail.[12-14] The sample holder or heating tube is supported vertically between two water-cooled electrodes. The lower electrode is provided with a strain-relief device to accomodate the thermal expansion effect and maintain a constant load on the tube. The sample holder is contained within a large, water-cooled vacuum enclosure (5 kW heat removal capacity) operating in the mid 10^{-5} N/m^2 range. The enclosure features two windows, one which is used by the emissometer optical system for sighting the three targets and the other which is used by the radiation thermometer (Pyro Automatic Optical Pyrometer) for determining the temperature of the midsection of the heating tube. The enclosure rests on a feedthrough collar that contains mechanical positioning devices, instrumentation leads, water cooling lines, and electrical power leads. Sensor signals are patched into a terminal board that is connected to the mini-computer. Power to the heating tube is provided by regulated dc power supplies complete with calibrated shunt and remote-controlled, current-reversing circuitry.

The emissometer optical system is mounted on a table located in front of the multiproperty apparatus. The optical system, shown schematically in Fig. 3, views one of three targets – the sample (*S*), the blackbody cavity (*H*) on the heating tube, or the radiometric zero cavity (*Z*) – through the vacuum enclosure window.

Plane mirror M1 can be controlled to view one of the three targets. Spherical mirror M2 places an image of the target upon the aperture A of 0.8 mm diam. Spherical mirror M6 places an image of the aperture A, with a magnification of two, onto the entrance slit ES of the prism monochromator, a Perkin-Elmer Model 98 utilizing a sodium chloride prism. An Infrared Associates, liquid-nitrogen cooled, Hg-Cd-Te detector (1.5-15 μm) is used with a 1000 Hz chopper C located just behind the aperture plane A. The amplifier is an Ortec-Brookdeal Model 9503 lock-in amplifier interfaced to the impedance matching preamplifier of the detector.

The PDP-8/E minicomputer controls the positioning of M1 such that the proper target is viewed by the optical system and it also increments the wavedrum of the monochromator by a

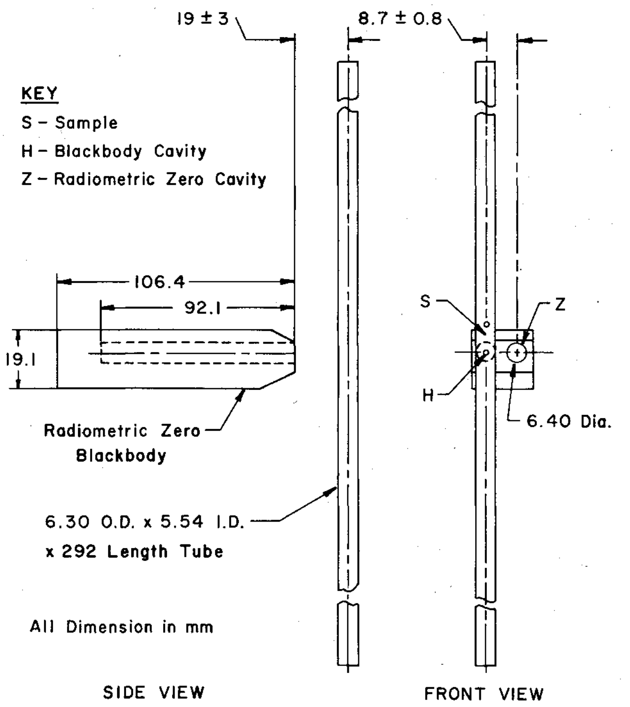


Fig. 1 Sample heating tube showing the location of the sample *S*, blackbody cavity *H*, and radiometric zero cavity *Z*.

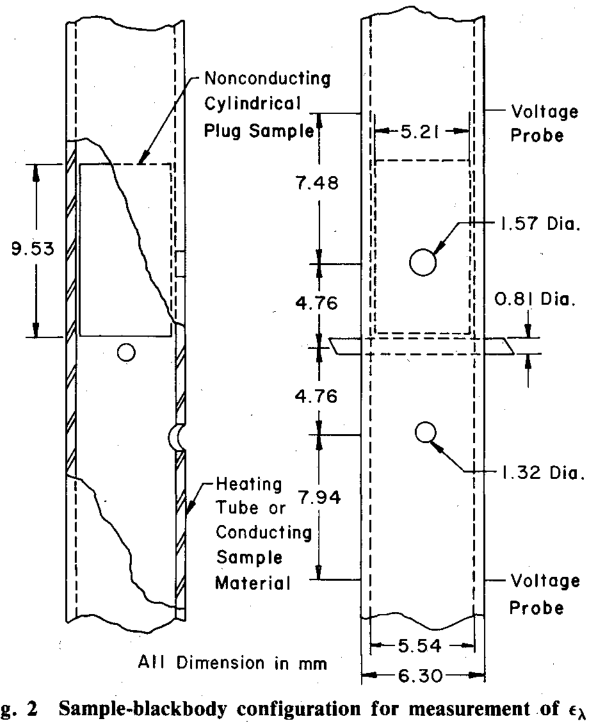


Fig. 2 Sample-blackbody configuration for measurement of ϵ_λ of nonconducting samples.

specified value. The positions of mirror M1 are set by solenoids controlled via programmable switches. The wavelength drum position is determined from a potentiometer signal that is related to the prism gauging curve. For each target the computer averages the detector signals for a specified number of data points (typically 25 at 167 ms intervals). It then calculates the emissivity according to Eq. (1), and determines the standard deviation for each set of signals. The computer also monitors the detector signal level and indicates by a message on the display unit, when the operator must take corrective action, usually a matter of increasing amplifier gain settings. The graphics display terminal provides a plot of the spectral emissivity vs wavelength, as the data are obtained, so that the operator can monitor the measurement process.

Conducting Sample Results

The initial trials to evaluate the performance of the emissometer were made on electrically conducting samples. Measurements on high-purity, well-polished tantalum are compared in Fig. 4 with the available literature data. Much of the difference between our data and that of Riethof[15] can be explained by surface conditions. Riethof[10] reported that his tantalum spectral emissivity values were high (by 0.01 emittance units) due to an unidentified surface film that had formed on the measured sample surface. In contrast, our tantalum surface was highly polished and oxide free, and therefore lower spectral emissivity values would be expected. The data of Kovalev and Muchnik[16] at 1800 and 2200 K are in good agreement with Riethof's[15] data. Available sample characterization indicates their tantalum was of 98.8% purity and the vacuum conditions were not stated. Lacking suitable emissivity reference standards and comparative literature data for this spectral region and temperature range, it is only possible to estimate that accuracies of 1 to 2% are possible for measuring the spectral emissivity of metallic samples, utilizing the emissometer.

Performance Evaluation

Due to the lack of comparative data and reference standards for nonmetallic materials, the performance and measurement accuracy of the emissometer had to be evaluated by considering likely sources of error.

Sample Temperature Distribution

It is necessary that the surface temperature distribution be nearly uniform for the nonconducting sample and that the temperature be the same as that of the heating tube (Fig. 2). To predict the spatial temperature distribution in the vicinity of the lateral hole, a one-dimensional heat transfer model was developed based upon the geometrical arrangement shown in the inset of Fig. 5. Note that the tube temperature is uniform at T_0, r_0 is the radius of the lateral hole opening, and h the distance between the nonmetallic sample and the tube. This model considered radiation exchange between the sample and the tube, radiation exchange between the sample and the enclosure (through the lateral opening), and diffusion within the sample. The thermophysical properties required in the model were the thermal conductivity k and the hemispherical total emissivity ϵ.

The results of the analysis are shown in Fig. 5. The difference between the tube temperature and the sample temperature as a function of radius, $T_0 - T(r)$, is shown as a function of tube temperature T_0 for typical values of k, ϵ, and r_0. The spacing between the sample and the tube (0.15 mm) was found to have little effect on the temperature distribution.

To estimate the error produced in measured spectral emissivity values due to the temperature depression of the nonconducting sample, it is only necessary to calculate the blackbody spectral radiance error, $\Delta L_{b\lambda}/L_{b\lambda}$ (%) produced by a temperature difference between the sample and blackbody, ΔT, $[T_0 - T(0)]$. This error in the blackbody spectral radiance value produces an equal error in the calculated spectral emissivity. At 1775 K, the maximum temperature depression (r = 0) is 1.8 K corresponding to $\Delta T/T_0 = 1.0\%$. For the worst spectral condition encountered, λ = 1.5 μm, the spectral radiance error is 0.5%. For a tube temperature at 2175 K this error is 0.9%, while at 2575 K the error is 1.4%. For the conditions encountered in the present investigation,

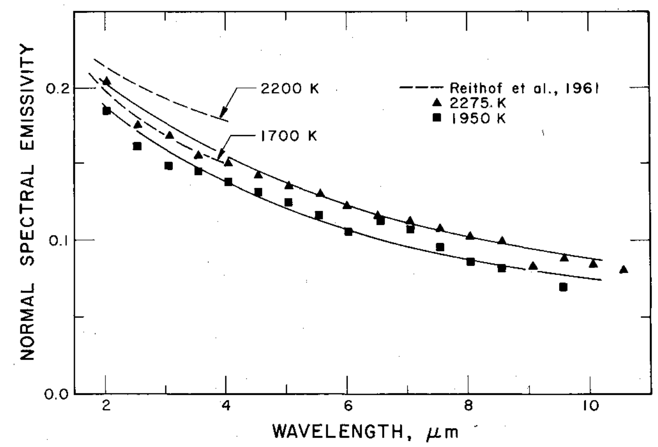
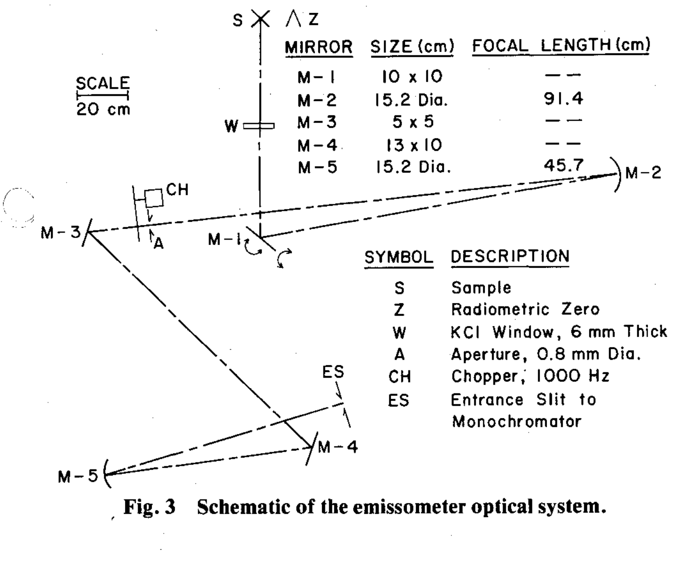


Fig. 4 Spectral emissivity of tantalum compared to literature values.



MIRROR	SIZE (cm)	FOCAL LENGTH (cm)
M-1	10 x 10	– –
M-2	15.2 Dia.	91.4
M-3	5 x 5	– –
M-4	13 x 10	– –
M-5	15.2 Dia.	45.7

SYMBOL	DESCRIPTION
S	Sample
Z	Radiometric Zero
W	KCl Window, 6 mm Thick
A	Aperture, 0.8 mm Dia.
CH	Chopper, 1000 Hz
ES	Entrance Slit to Monochromator

Fig. 3 Schematic of the emissometer optical system.

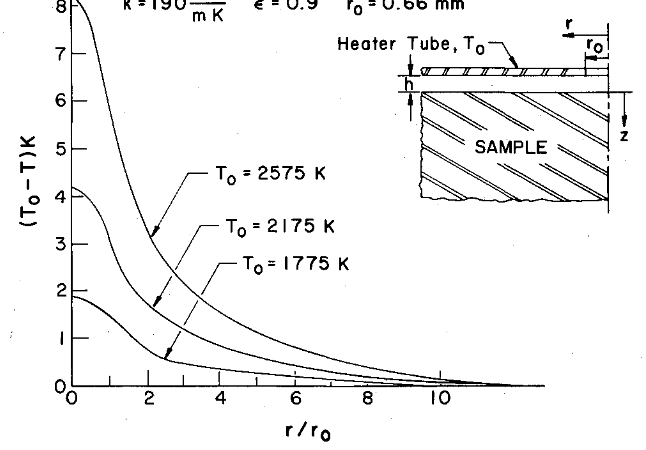


Fig. 5 Nonconducting sample surface temperature distribution for sample properties k = 190 W/m-K and ϵ = 0.9 and for r_0 = 0.66 mm.

the systematic error due to the sample temperature nonuniformity was estimated to be less than 1%.

Heating Tube Temperature Distribution with Sample Present

To ensure that the test section remained isothermal when a nonconducting sample was placed in the heating tube, an experiment was performed to measure the longtitudinal temperature distribution of the tube. The influence of the sample on the temperature distribution of the tantalum heating tube at 1800 K can be seen in Fig. 6. The spectral radiance temperature (0.65 μm) observations were taken under matched electrical resistivity conditons for "without" and "with" sample configurations; that is, the average temperature between the voltage probes was the same for each configuration. The general trend was that the sample did not really alter the temperature distribution. The temperature differences, particularly for the "with sample" case, are most likely due to emissivity variations resulting from grain growth and minor imperfections on the tantalum tube surface. This can be appreciated fully when noting that the radiation thermometer target is 0.5 mm in diameter while the total temperature range difference between the two configurations can be explained by a ± 0.010 emittance unit variation of the tantalum surface. That is, the spectral radiance temperature distribution is more likely explained by variations of emissivity than by a difference in temperature for the two configurations. Thus, the test section comprised of the sample and the blackbody cavity meets the isothermal requirements to determine emissivity without significant error.

Blackbody Cavity Quality with Nonconducting Sample Present

The quality of the lower cavity was expected to be dependent upon the emissivity of the nonconducting sample, since the cavity is bounded at the upper end by the sample. Because of the complicated geometry of this configuration, an analytical treatment would be difficult. To determine how the quality of the lower lateral hole was affected by the presence of nonconducting cylindrical sample, an experimental procedure was developed.

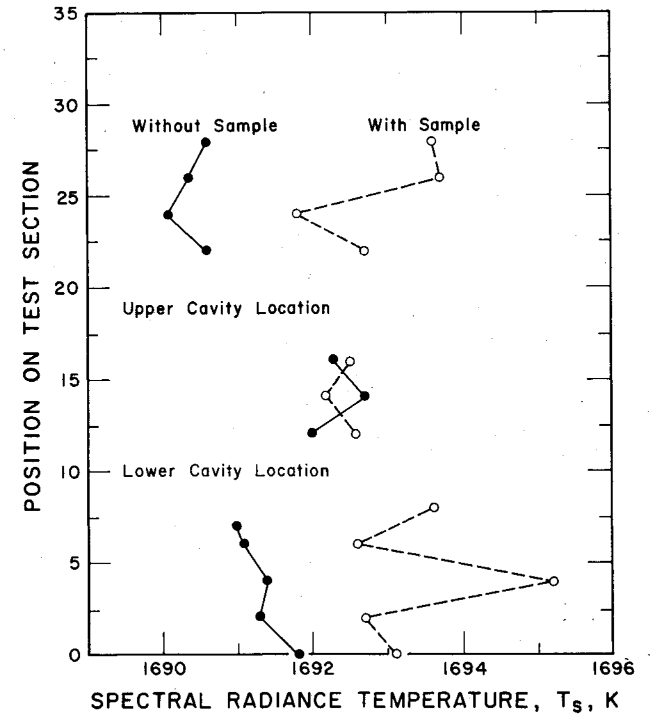


Fig. 6 Tantalum heating tube spectral radiance temperature distribution T_S(0.65 μm), with and without a silicon carbide sample in position.

This method assumed that the quality of the lower cavity was unity without the sample in position. The experimental approach relied on the use of the tantalum heating tube as a resistance thermometer. Voltage probe leads were spot welded to the tantalum tube and the distance between probes accurately determined.

The heating tube, without a nonconducting sample in position, was brought to an operating temperature as determined by sighting with the radiation thermometer into the lower cavity. The electrical resistivity was then determined from the voltage drop across the test section and the current through the tube. The solid line of Fig. 7 represents a least-squares fit to a series of resistivity-temperature measurements taken around 1800 K. Using the relationship[17]

$$r_e\,(\mu\Omega-\mathrm{cm}) = A + BT + CT^2 \tag{2}$$

where T is in kelvins and A = 4.43633, B = 4.1944 × 10^{-2}, and C = 2.3220 × 10^{6}, the tantalum heater tube could be used as a resistance thermometer.

A nonconducting cylindrical sample was then inserted into the heating tube and the current carefully adjusted to match the resistivity of the "tube only" configuration. The spectral radiance temperature, T_s, of the lower cavity was then measured by the radiation thermometer. The observed spectral radiance temperature of the lower cavity as a function of the electrical resistivity is represented by the dashed line of Fig. 7. The temperature offset of the two curves can be used to calculate the emissivity of the lower cavity with the silicon nitride sample in place by using Wien's approximation to the Planck spectral distribution curve. The results of these observations and calculations for two nonconducting materials, silicon carbide and silicon nitride, are summarized in Table. 1.

These results suggest that the presence of the sample has little effect on the quality of the lower cavity and that the effect of the sample emissivity on the quality, at least for the emissivity range studied, is negligible. It should be noted that the quality is not likely to be a function of wavelength, provided spectral emissivity values do not change markedly.

Blackbody Cavity Quality as a Function of Tube Lateral Hole Diameter

Any decrease in the blackbody quality produced by increasing the size of the viewing port would cause an equivalent decrease in the measured blackbody radiance, which would produce an equal increase (error) in the calculated spectral emissivity as obtained by Eq. (1). Thus, it needed to be shown that the blackbody quality was a weak function of the diameter of the viewing port.

Consider the situation where the upper cavity is 20% larger in diameter (44% larger in area) than the lower cavity. Without a sample in position, the DeVos relation[18] estimates the quality of the lower cavity to be 0.993 and the quality of the upper cavity to be only 0.003 emittance units less. A tantalum tube was heated to 1800 K and radiation thermometer observations were taken by sighting into the upper and lower cavities. The lower (smaller) cavity was assumed to have a quality of unity so that the spectral radiance temperature measured for this cavity would yield the true tem-

Table 1 Cavity quality with nonconducting sample present

Target cavity	Spectral emissivity (0.65 μm, 1800 K) Without sample	With sample	
		SiC	Si_3N_4
Upper	0.990	0.801	0.654
Lower	1.000	0.993	0.994

perature of the tantalum tube. The quality of the upper cavity was then determined from the well-known Wien's approximation of the Planck spectral distribution law. From these measurements the quality of the upper cavity was determined to be 0.010 units less than the lower cavity instead of 0.003 units as predicted by DeVos. This difference can be explained by the combined precision of the radiation thermometer and the variation of the observed radiance temperature when sighting into either of the cavities.

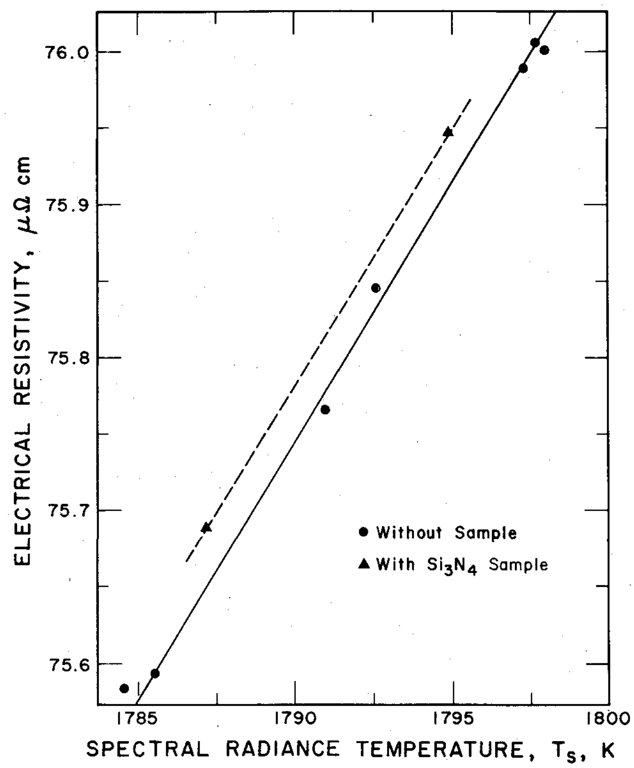


Fig. 7 Lower cavity quality determination with silicon nitride sample in position from spectral radiance temperature, T_s (0.65 μm), and electrical resistivity observations.

This study shows that increasing the hole surface area by 44% decreases the quality by less than 1%. It was therefore concluded that the size of the hole, at least within the diameter range considered, has very little effect on the quality of the blackbody.

Summary

To estimate the accuracy of the measurement method several features were considered. The systematic error due to the sample temperature nonuniformity in the vicinity of the lateral hole was found to be less than 2%. The effect of the sample on the longitudinal temperature distribution of the heating tube was found to be minimal. The presence of a nonconducting sample was shown to have very little effect on the quality of the blackbody cavity. Varying the size of the tube lateral hole was found to affect the blackbody cavity quality by less than 1%. Furthermore, errors associated with the scattered flux effect (veiling glare) and the wavelength calibration have also been found to be negligible. With all of these features taken into account, the overall performance limit of the emissometer is estimated to be ± 2 to ± 4%.

Spectral Emissivity of the Ceramics

Sample Descriptions

Samples of silicon carbide where obtained from Carborundum, Ceradyne, and General Electric. The commercial suppliers of silicon nitride were Airesearch, Kawecki-Berylco, Kyocera, and Norton. Sample descriptions are given in Table 2.

The Carborundum silicon carbide samples were formed by cold-pressing silicon carbide powder and then sintering the material at temperatures in excess of 2275 K. The final cylindrical shape was obtained by diamond grinding. The sample was reported as being 99% silicon carbide with the remainder being composed primarily of sintering aide, boron, and carbon.

Two types of silicon carbide were obtained from Ceradyne: Ceralloy 146A and Ceralloy 146I. Both types were prepared by hot-pressing and the final sample shape was formed by diamond grinding. Both samples were reported as having no free silicon or carbon. Ceralloy 146A was composed of about 98% silicon carbide and 2% Al_2O_3, with traces of tungsten

Table 2 Description of the ceramic samples

Manufacturer	Major constituent	Designation	Phase	Density, g/cm^3	Major impurities	Grain size, μm	Process
Carborundum	SiC		α	3.15	1% B+C	7	Cold-press and sinter
Ceradyne	SiC	Ceralloy 146A	α	3.30	2% Al_2O_3	5-10	Hot-press
Ceradyne	SiC	Ceralloy 146I	α	3.00	2% B_4C	coarse	Hot-press
General Electric	SiC		β	3.00	W trace 0.8% B+C	3-5	Cold-press and sinter
General Electric	SiC		α	3.00	W Trace	3-5	Cold-press[a] and sinter
Airesearch	Si_3N_4		α+β	2.75	1% Fe	5-10	Reaction-bonded
Kawecki-Berylco	Si_3N_4		70-80% α 30-70% β	2.38	3/4% Si	2-4	Reaction-bonded
Kyocera	Si_3N_4			2.70	1% Al_2O_3		Cold-press and sinter
Norton	Si_3N_4			3.07	...		Hot-press

[a] This is the β-phase sample that was heated (600 K/hr) to 2475 K in helium at 1.01 × 10^5 N/m^2 and soaked for 30 min. This caused a transformation from cubic β-SiC to hexagonal α-SiC.

present. Ceralloy 146I was 98% silicon carbide and 2% boron carbide.

The silicon carbide sample supplied by General Electric was prepared by cold-pressing submicron β-phase silicon carbide powder at 2.76×10^7 N/m^2 and sintering for 30 min at 2335 K in a vacuum. The resulting material was diamond ground to the appropriate diameter. After measuring the spectral emissivity of the β-phase material, the same sample was heated at 600 K/h, to 2475 K in helium at 1.01×10^5 N/m^2 and held at that temperature for 30 min by the Physical Chemistry Laboratory at General Electric. This caused a transformation from cubic β-SiC to hexagonal α-SiC.

The Airesearch silicon nitride samples were reaction bonded. Silicon powder was slip cast in a nitrogen atmosphere and small amounts of iron were added to aid in converting silicon to silicon nitride. The final iron content was estimated as being 1%. The sample was made of a mixture of α and β phase materials. The surface was measured as received and no diamond grinding was employed to obtain final dimensions.

A reaction-bonded silicon nitride sample was supplied by Kawecki-Berylco. This sample was prepared from silicon powder but no iron was added as a nitriding aid. The final sample shape was obtained by diamond grinding. The sample was reported to be 70-80% α phase (hexagonal), with the remainder being β phase. Some residual free silicon (½-1%) was also present.

The Kyocera silicon nitride sample (SN205) was prepared by cold-pressing silicon nitride powder and then sintering at temperature in excess of 1875 K. The sample shape was obtained by diamond grinding the rough form produced by cold-pressing the powder. The sample was reported as being 99% pure silicon nitride and 1% Al_2O_3.

The Norton silicon nitride sample was prepared by hot-pressing. The sample had been heat treated for 30 h; however, no further conditioning information was made available. Diamond grinding was used to obtain the sample shape from the rough form.

Spectral Emissivity Results - Silicon Carbide

The normal spectral emissivity data of SiC for each sample was measured (after a 10 min pretreatment at 1950 K in high vacuum) first at 1900 K, then at 1800 K, and in one case at 1700 K. Repeat measurements were also made on each of the four samples. The data for the first run on the Carborundum and the Ceralloy 146A samples and the data for the repeat run on the Ceralloy 146I and General Electric samples are plotted in the composite summary, Fig. 8. From this figure, it can be seen that the spectral emissivity of Ceralloy 146A has no structure, that the emissivity data of the Ceralloy 146I and Carborundum samples exhibit a small peak at about 11 μm and a large valley near 12.5-13 μm. The results were reproducible to generally better than 1% except for the Carborundum and Ceralloy 146I samples in the spectral regions where there is spectral character. The solid and dashed curves represent the (provisional) recommended curves given by the Center for Information and Numerical Data Analysis and Synthesis (CINDAS).[19] Since these curves are based upon very limited information, their uncertainties are nearly 30%. Especially interesting to note is the spectral character near the 10-12 μm region, which may be due in part to impurities (such as boron) as well as the fabrication process.

In Fig. 8, the General Electric silicon carbide is in the β phase. It is apparent that its spectral character is substantially different from the three other α phase samples. To determine if the difference was due to phase, a virgin GE β phase sample was measured and then this same sample was transformed by a heat treatment process to α phase. The results of spectral emissivity measurements on the β phase and transformed α phase samples at 1800 and 1900 K are shown in Fig. 9. Note that there is little difference between the α and β-phase materials.

Spectral Emissivity Results – Silicon Nitride

Because of the instability of silicon nitride at high temperatures under vacuum, emissivity data for the Airesearch, Kawecki-Berylco, and Kyocera samples were first taken at 1600 K, then 1700 K and in some cases at 1800 K. Emissivity values changed rapidly at 1800 K. The data all show an increase in emissivity from 3 to 8 μm, a decrease from 8 to 11 μm, and an increase in 11 to 14 μm range. The data obtained at 1600 K show the reproducibility to be generally better than 1% over the entire wavelength region. However, the data continued to change at 1800 K during the measurements and the results were not reproducible. The data for the Airesearch

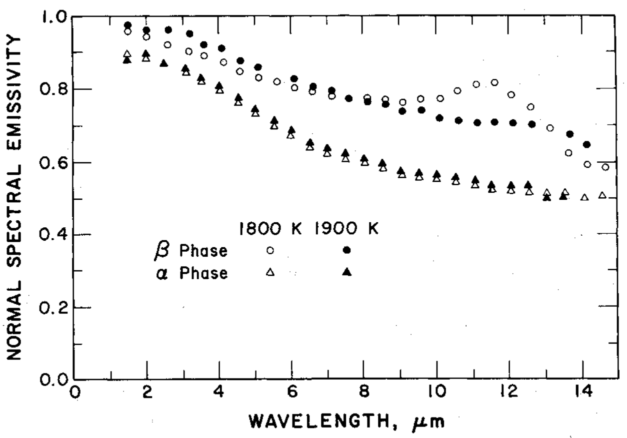


Fig. 9 Comparison of the spectral emissivity of General Electric silicon carbide, β-phase and General Electric silicon carbide, α-phase transformed from a β-phase sample.

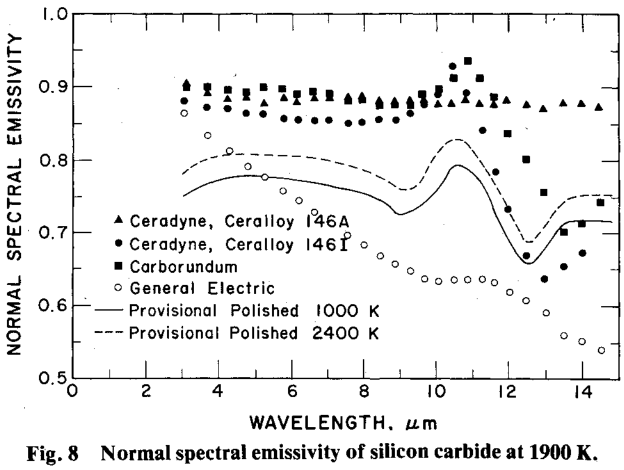


Fig. 8 Normal spectral emissivity of silicon carbide at 1900 K.

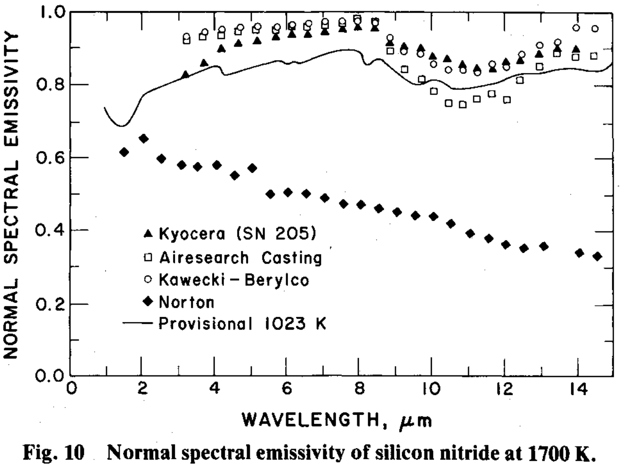


Fig. 10 Normal spectral emissivity of silicon nitride at 1700 K.

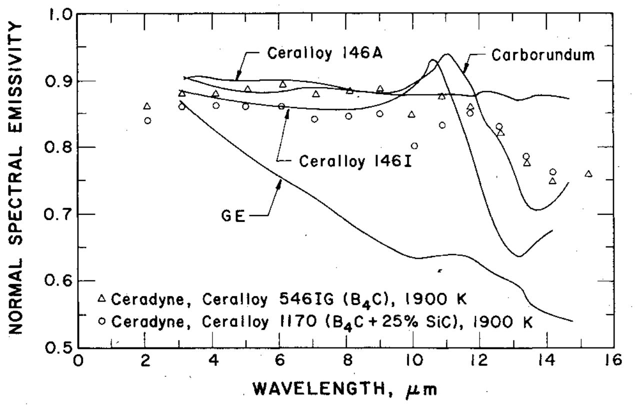


Fig. 11 Comparison of the spectral emissivity of silicon carbide and boron carbide at 1900 K.

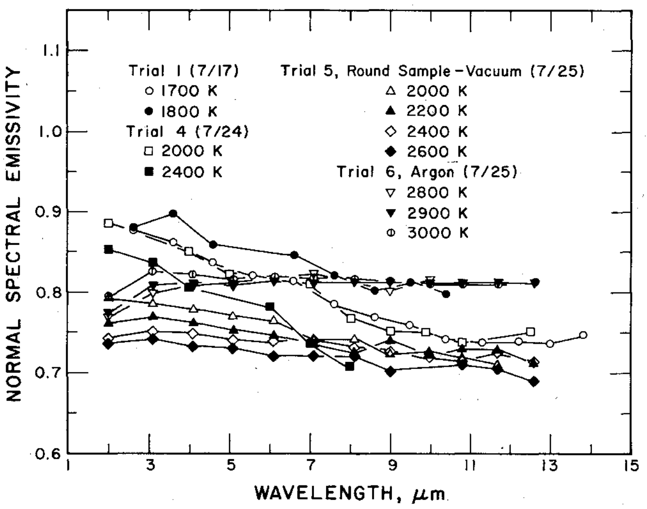


Fig. 12 Spectral emissivity of a graphite composite samples.

and Kyocera sample show that the emissivity values changed markedly for silicon nitride at 1800 K under high vacuum. Emissivity data for the Norton sample were first taken at 1400 K, then the temperature was incremented in 100 K steps to 1800 K. The sample was found to be quite unstable. The data from 1400 to 1500 K show an increase in emissivity from 3 to 8 μm, a decrease from 8 to 11 μm, and increase from 11 to 14 μm. The spectral emissivity values continued to drop in magnitude and to flatten out in shape from 1500 to 1600 K. The sample definitely deteriorated, as evidenced by the difference in the before and after weighings as well as dimensional changes. Unfortunately, the temperature at which this occurred is unknown. This might explain the sudden increase in emissivity at 1700 K. The results for the four silicon nitride samples at 1700 K are shown in Fig. 10.

Discussion of the Results

Silicon Carbide

It appeared that the presence of boron/carbon caused a peak in the spectral emissivity data at about 10.5 μm followed by a valley at about 13 μm. This peak/valley may not be stable at 1900 K and above in high vacuum. Although the material was sintered or hot-pressed at significantly higher temperatures, it probably was not under high-vacuum conditions. The emissivity of the β-phase material decreased markedly with increasing wavelength in contrast to α-SiC. Even in the β-phase material, it was conceivable that the presence of boron/carbon could be noted at about 11 μm by the presence of a plateau. To help verify this suspicion, two types of boron carbide were obtained from Ceradyne. The normal spectral emissivity for each sample was obtained at 1900 K and the data are compared with the silicon carbide data in Fig. 11. The spectral character of the boron carbide samples is quite unstable from about 8 μm on. This data seems to indicate that the cause of the plateau is not due to boron carbide. It is presently suspected that tungsten may be the cause of the peak at about 10.5 μm, but further verification is needed.

In any event, it is obvious that the spectral emissivity of silicon carbide can be controlled at the longer wavelengths for particular applications. Since the present silicon carbide samples were diamond ground, they exhibited a polished appearance. The recommended (provisional) values given by CINDAS[19] for average polished SiC at 1000 and 2400 K are also shown in Fig. 8. These values are about 15% below the present values for wavelengths less than 11 μm and include the peak/valley found in our results.

Silicon Nitride

The spectral emissivity of all of the silicon nitride samples, except the Norton sample, exhibited a maximum at 8μm followed by a sharp decline to 11 μm and an increase from 11 to 14 μm. No relationships to impurities were noted, so it was concluded that this behavior is typical of Si_3N_4. The provisional curve by CINDAS[19] is included in Fig. 10. The general features of the provisional curve agree with the present results, but this curve generally lies about 10% below the present results.

Spectral Emissivity of a Graphite Composite

The feasibility of using the multiproperty apparatus with the emissometer for determining the spectral emissivity of a graphite composite material has been demonstrated. The objective of this set of measurements was to determine whether the spectral emissivity, particularly at longer wavelengths, changed rapidly with increasing temperature and not necessarily to obtain highly accurate data.

In order to obtain a sample that would fit between the electrodes, three pieces of the graphite composite had to be separately machined and then fit together. This configuration had a severe limitation in that the sample continued to fail at the slip joints between the sample and the extender end pieces. These joints could not be made to fit as tightly as desired, resulting in high resistance at the interfaces. A series of specially configured samples were fabricated until finally a suitable geometry was obtained. Measurements were made in a vacuum up to 2800 K, at which point vaporization became noticeable. Measurements were then made in 4600 N/m^2 of argon from 2800 to 3100 K, where sample failure occurred at the blackbody hole. The results of these measurements are shown in Fig. 12. The objective of determining whether there was any anomalous behavior of the spectral emissivity from 2 to 13 μm between 1700 and 3100 K was met. It is apparent that no irregularities occurred over these regions.

Summary

An emissometer for generating high temperature spectral emissivity values of nonconducting materials has been described and its systematic sources of error identified. The performance evaluation indicates that accuracies of 2-4% can be expected. The emissometer has several advantages over other measurement techniques. The sample requirement, short cylindrical form, is especially convenient for materials research, since most new formulations of exotic materials can be easily fabricated.

The measurements on the new ceramics, silicon carbide and silicon nitride, will be of special interest to the high temperature technologist. The results for the range 1400-1900 K and 1.5-15 μm indicate that there is a wide variation (50% in one case) in property values of the same ceramic prepared by different manufacturers. For both the ceramics, the spectral emissivity results are in poor agreement with the recommended (provisional) curves given by the CINDAS data series.[19] The stated uncertainties for their recommended (provisional) curves are quite high and the comparison with our results further points out the need for reliable baseline data as the first step in understanding the behavior of materials at high temperatures. The results on the graphite composite sample indicate that the emissometer is capable of measuring the radiative properties of advanced materials at very high temperatures.

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