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Spectral and Total Emissivity Measurements of Highly Emitting Materials[1]

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Carbon, carbides, and nitrides are materials having a relatively high emissivity over the entire spectral range. They are important both as a material and as material components of composites for high-temperature applications, e.g., in space technology (reusable space transport systems) or energy systems (hot gas turbines). The normal total and spectral emissivities of these three materials have been investigated in the temperature range 1000 to 2000 K and at wavelengths between 0.6 and 6.8 μm. The results have been used to interpret emissivity results of fiber-matrix composites. They are also discussed with regard to potential application as reference materials for high-temperature emissivity measurements.

KEY WORDS: composites; emissivity; graphite; high temperatures; silicon carbide; solicon nitride.

1. INTRODUCTION

Great efforts are being directed worldwide to the development of high-temperature materials possessing good mechanical properties and high stability even at extreme conditions such as the hot gas stream in turbine engines or reentry of space vehicles from the orbit into the atmosphere. Favored materials are ceramic matrix composities (CMC) based on carbon, silicon carbide, and silicon nitride. For thermal analysis in design of components the thermal conductivity and emissivity has to be known as precisely as possible. The calculation of the maximum temperatures is especially important because these materials are often used up to their upper temperature limit. In the range above 1500°C, the emissivity becomes important in

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order to support heat dissipation by radiation. The high emissivity of this type of ceramic contributes additionally to an efficient surface cooling. Not only is the total emissivity important to calculate radiation heat transfer, but also the spectral emissivity is needed for radiation thermometry. The wavelength dependency is especially of interest because, to an increasing degree, ratio and multispectral pyrometers are used or proposed to be used, whereby special assumptions such as greybody or other emissivity/wavelength functions are made and have to be carefully checked before use in order to prevent uncontrollable errors [1].

As an example, Fig. 1 shows the results of one investigation [2] of the spectral emissivity of a carbon fiber/silicon carbide matrix composite. The figure illustrates specific behavior of this type of material:

- The spectral emissivity varies with wavelength over a wide range between 0.92 and 0.55.
- The wavelength dependency varies with temperature.
- The emissivity values undergo changes after thermal treatment.

At temperatures below 1600 K the behavior is similar to that of metals: The emissivity curves forme a so called x-point, defined as the wavelength at which the spectral emissivity does not change with temperature. Below this x-point, the spectral emissivity decreases with increasing temperature, and at wavelengths higher up the x-point the values increase. After annealing of the sample (in vacuum) for about 1 h between 1600 and 1700 K, this

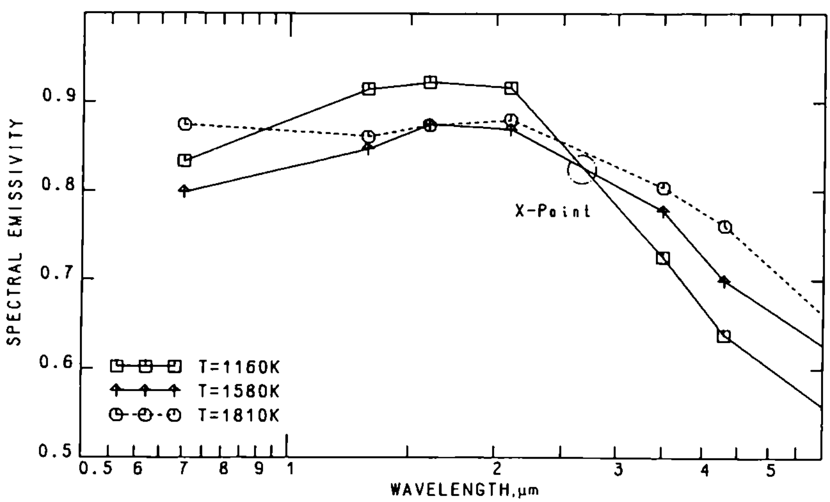

Fig. 1. Spectral emissivity of C/SiC composite at different temperatures.

behavior disappears, and at further increasing temperatures the emissivity increases slightly over the complete wavelength range. The total emissivity increases during this annealing procedure from 0.8 to 0.86 and remains high during cooling down. Detailed description of these investigations is given in Ref. 2.

In order to be able to compare the emissivity results of composites with those of pure substances, the emissivity of the three most important components of types of a CMC has been investigated. An additional aim of this study was to check whether one of these materials could be recommended as a reference material to compare emissivity measurements of different laboratories. There is an urgent demand for such high-temperature emissivity standards in order to improve the measurement accuracy.

2. MEASUREMENT TECHNIQUE

The measurement technique has been described earlier [3, 4] and is summarized briefly. The radiation comparison technique is used to measure the normal total and spectral emissivity of disk-shaped samples, heated in vacuum by means of an electron beam. The specimen temperature is measured inside a small radial hole positioned close to the surface. The total emissivity is measured directly by means of a thermoelectric sensor and the spectral values are measured by introducing interference filters. In order to calculate the temperature difference between the blackbody hole and the measurement surface, the thermal conductivity has to be known. This is the main source of error for materials having a low thermal conductivity, especially if the thermal conductivity is not uniform and reproducible.

As demonstrated in Ref. 5 the emissivity error caused by imperfect reproducibility of the thermal conductivity of composities can reach values up to 15%, depending on temperature and wavelength. The emissivity error caused by temperature error increases with decreasing wavelength. The thermal conductivities of the materials decribed in this paper are well defined and therefore the inaccuracy of the measurements is between 2 and 6%.

3. MATERIAL DESCRIPTION

3.1. Graphite

The samples were prepared by SGL, Carbon, Ringsdorff-Werke, Bonn, manufactured by isostatic molding and subsequent heat treatment up to

3000°C. Density, 1.9 $g \cdot cm^{-3}$; thermal conductivity,[3] 65 $W \cdot m^{-1} \cdot K^{-1}$ (at 1000 K), 45 $W \cdot m^{-1} \cdot K^{-1}$ (at 2000 K).

3.2. SiC

The sintered material was produced by Elektroschmelzwerk, Kempten, by axial hot pressing; 1.3 wt% aluminium was added to support sintering. Density, 3.2g $\cdot cm^{-3}$; thermal conductivity,[3] 45 $W \cdot m^{-1} \cdot K^{-1}$ (at 1000 K), 32 $W \cdot m^{-1} \cdot K^{-1}$ (at 1500 K).

3.3. HPSN

The silicon nitride was produced by hot pressing at the Institute für Nichtmetallische Werkstoffe, Technical University, Berlin. To support the sintering process 1.3 wt% Y_2O_3 and 2.0 wt% Al_2O_3 were added. Density, 3.15 $g \cdot cm^{-3}$; thermal conductivity [6], 21 $W \cdot m^{-1} \cdot K^{-1}$ (at 1000 K), 16.7 $W \cdot m^{-1} \cdot K^{-1}$ (at 1500 K).

4. DETERMINATION OF THE SURFACE ROUGHNESS

The emissivity of nontransparent materials depends on its surface profile. Unfortunately, the typical roughness parameters do not sufficiently describe a surface with respect to its influence on emissivity. The shape and distribution of cavities and, additionally, the surface structure of the cavity walls are much more important. The cavities yield an enlarged total surface S_R in comparison to the plane surface S_0. The ratio of both can be defined as roughness factor $R_F = S_0/S_R$. Published theoretical and experimental investigations [7, 8] have shown that the dependency of the emissivity can be approximated by means of the relation

$$\varepsilon_R = [1 + (\varepsilon_W^{-1} - 1) R_F]^{-1} \tag{1}$$

where ε_W is the emissivity of the cavity wall.

Among determination of profile lines the combination of electron scanning-microscope (ESM) pictures and measurements using the light microscope (LM) has proved to give representative results. The depth of the cavities can be determined with the LM by variation of the focus plane, while the ESM pictures show the distribution of the cavities. The surfaces of the present specimen can be described as follows:

[3] Own, unpublished measurement results.

4.1. Graphite

This had a homogeneous, diffuse surface with irregular but essentially funnel-shaped cavities. The angle of the cones varied between 30 and 70°, and the depth between 1 and 12 μm, with the following distribution.

Depth (μm):	0	2	5	8	11
Share of the total area (%):	12	20	30	28	10

The roughness factor R_F was approximately 0.8.

4.2. SiC

This had a plane surface with tub-shaped cavities of irregular geometry of the following distribution.

Mean area (μm^2):	10.000	6.000	2.000
Mean depth (μm):	14	10	8
Number/mm^2:	0	16	40

The estimated roughness factor R_F was 0.6.

4.3. HPSN

This sample was highly polished with diamond paste of 1-μm grain size. Therefore it can be classified as smooth, i.e., $R_F = 1.0$.

5. RESULTS OF EMISSIVITY MEASUREMENTS

Figures 2 to 4 show the spectral emissivity as a function of the wavelength at different temperatures. The curves were measured during the heating run. At high temperatures the specimen especially HPSN are not very stable in vacuum and start to evaporate leading to contamination of the windows. A first check for some kind of material change is to repeat emissivity measurements during cooling down. The contamination of windows can be excluded by transmittance measurement before and after the emissivity measurements.

The temperature dependence of the emissivity taken from the same results is presented for the HPN sample in Fig. 5. Both the spectral and the temperature behavior of HPSN is completely different from that of graphite and SiC. The difference of the wavelength dependency between graphite and SiC eventually can be explained by small dimensions of the

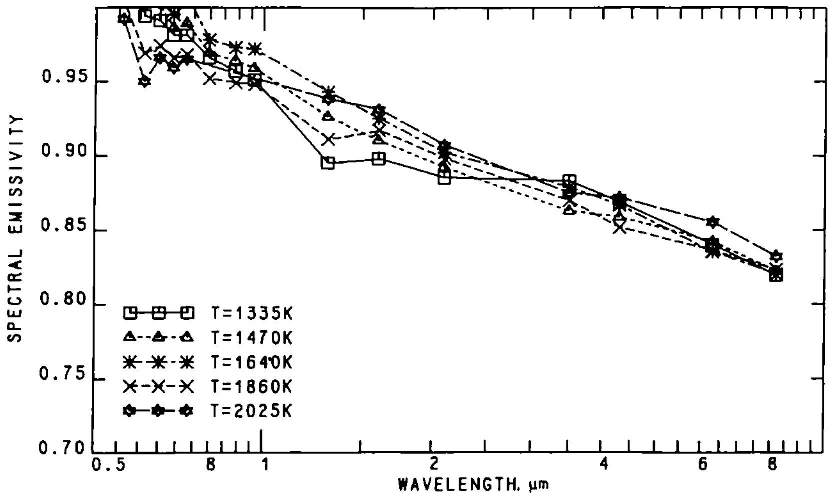

Fig. 2. Spectral emissivity versus wavelength of graphite at different temperatures.

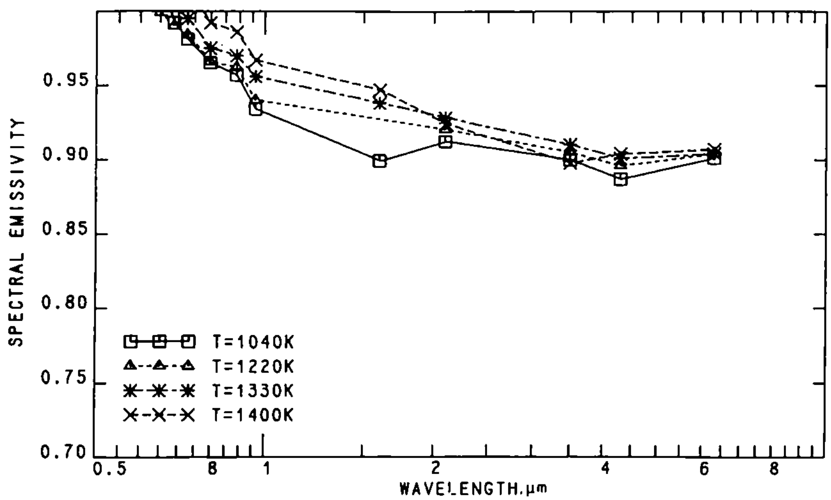

Fig. 3. Spectral emissivity versus wavelength of SiC at different temperatures.

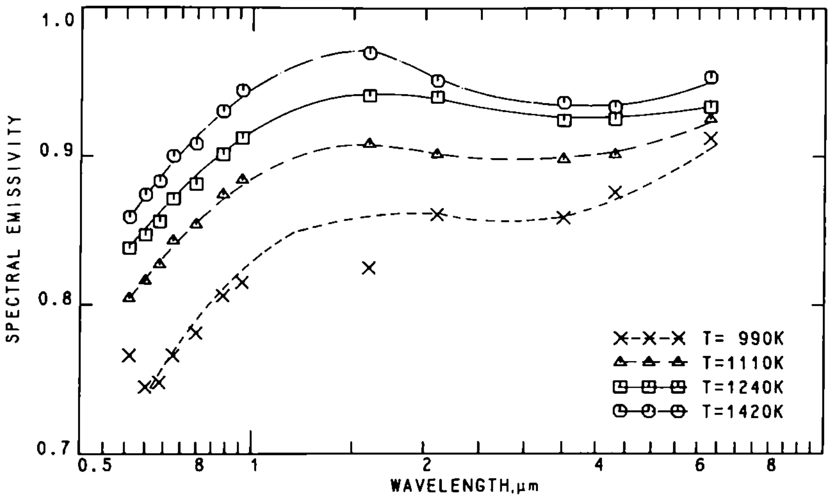

Fig. 4. Spectral emissivity versus wavelength of HPSN at different temperatures.

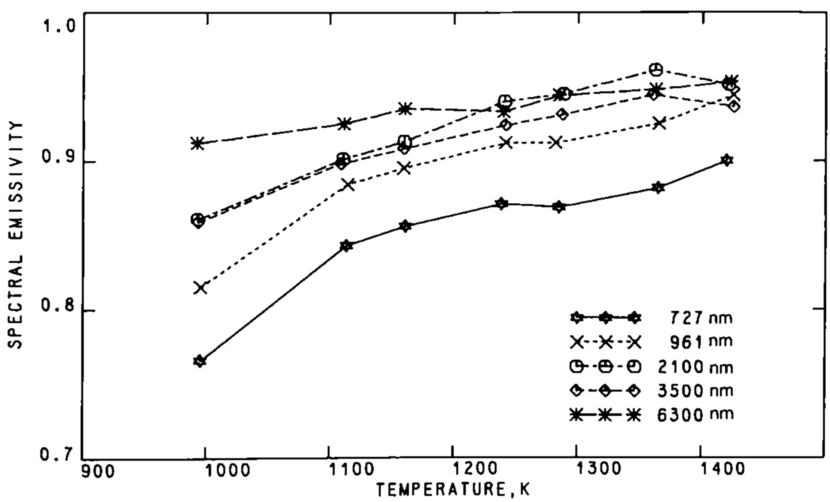

Fig. 5. Spectral emissivity versus temperature of HPSN at different wavelengths.

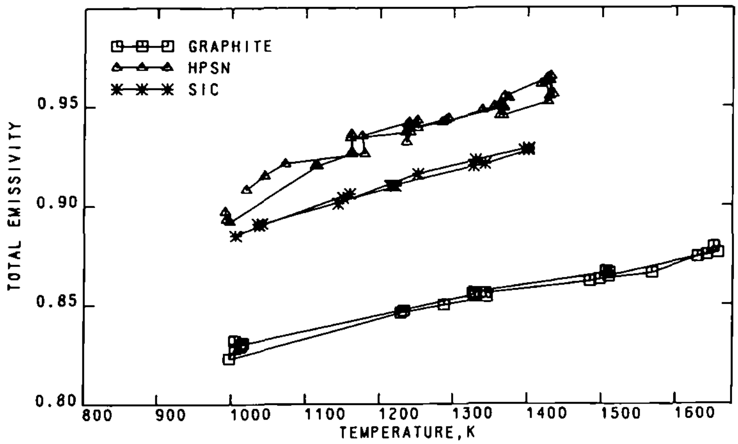

Fig. 6. Total normal emissivity of graphite, SiC, and HPSN measured during heating and cooling of the sample.

surface cavities of the graphite which affect the emissivity mainly at short wavelengths. The large cavity diameters, in the range above 50 μm, lead to an emissivity increase in SiC also at longer wavelengths. The spectral emissivity of both graphite and SiC is fairly independent on temperature. The small variations at short wavelengths are in the range of the uncertainty in measurement.

The total emissivity curves in Fig. 6 were measured during heating and cooling and their matching demonstrates that a material change arose during the measurement.

6. CONCLUSIONS

The comparison of the spectral emissivity of C/SiC (Fig. 1) with that of graphite (Fig. 2) and SiC (Fig. 3) shows that the emissivity of the composite is dominated by the influence of carbon fibers even though the fiber bundles are surrounded by SiC. The low emissivity of the fiber surface combined with the cavity effect caused by the groves between the filaments in the bundle lead to the pronounced wavelength dependency. An x-point cannot be observed either with graphite or with SiC. Fibers, however, are good electric conductors in the longitudinal direction. Spectral emissivity curves with an x-point have been measured only on metal surfaces until now, however, the x-point of metals was found at shorter wavelengths.

With respect to the application as reference material, silicon nitride is not suitable due to the high wavelength and temperature dependency of the spectral emissivity. Silicon carbide is the best of the three investigated materials because of its high emissivity and its insignificant variation with wavelength and temperature. Unfortunately it is extremely hard material and therefore difficult to machine. Although machining is much easier with graphite, samples have to be carefully handled in order to prevent damage of the surface structure.

In summary, it is recommended that SiC be investigated further with smooth surfaces and the reproducibility of graphite be evaluated.

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