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Original Article

Total hemispherical emissivity of sintered SiC up to 1850 K in high vacuum and in air at different pressures

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ABSTRACT

The total (0.6–40 μm) hemispherical emissivity of sintered silicon carbide samples was measured in different conditions of pressure and temperature, from high vacuum to atmospheric pressure in air and up to 1850 K using a direct method.

Emissivity data are correlated to the presence of different thicknesses of the silica layer and material characterization using scanning electron microscopy, ellipsometry and 3D profilometry was carried out to complete the interpretation of the emissivity evolution with temperature and pressure according also to surface roughness.

1. Introduction

SiC is a widely used ceramic due to its excellent properties and refractoriness and is mainly involved in space mirrors, heat exchangers, chemistry reactors, and more generally in the semiconductor industry and metallurgy sector.

Even though a lot of works were dedicated to SiC, it is roughly difficult to find reliable emissivity data at high temperature with a clear description of the conditions of measurement, surface state and composition of the material.

Emissivity is a property that is very dependent on temperature, wavelength, direction, surface roughness, porosity, environment, oxidation level related to surface composition for the main parameters. Moreover, it exists several polymorphs of SiC and among them the more frequently encountered are, for massive materials, α-SiC (6 H) and β-SiC, and also SiC as coating.

Different emissivity data can be found in the literature but few are correlated to the surface state of the material.

Touloukian and De Witt have reported normal total (1–15 μm) emissivity values from 500 to 1800 K in air close to 0.85-0.90 for a porous α-SiC material from Carborundum, the density being of 1.49 g cm^{-3} instead of 3.21 g cm^{-3} for the theoretical one, and also they have collected some normal spectral (0.6–15 μm) data for the same material with a main value of 0.90 from 1 to 10 μm, then a sharp decrease up to 12 μm and an increase again around 14 μm, due to the presence of silica on the surface as the measurement have been performed in air [1].

De Witt et al. have completed these measurements on a SiC from Carborundum with a purity of 99% (density of 3.15 g cm^{-3}, and with 1% B + C, 7 μm grain size) and also on a SiC from Ceradyne (Ceralloy 146 A) and they have given a nearly constant value for the normal spectral emissivity of 0.90 in high vacuum from 3 to 12 μm at around 1800–1900 K [2]. These SiC materials presented a different behavior from the one from General Electric for which the normal spectral emissivity reported is strongly decreasing with increasing wavelength from 3 μm (around 0.86) to 14.5 μm (around 0.55) after a plateau in between 10 and 12 μm (around 0.63) [2].

Neuer has measured the spectral and total emissivity of different sintered SiC samples from 1040 to 1400 K, and in the wavelength range 0.6–10.6 μm [3]. The normal spectral emissivity goes from 0.98 down to 0.90, and the total emissivity is increasing from 0.87 at 1000 K to 0.92 at 1400 K. Neuer and Jaroma-Weiland have also measured the total (1.3–9 μm) emissivity of SiC coating and gave values around 0.92 at 1150 K up to 0.96 at 1700 K [4].

Biasetto et al. have measured on sintered α-SiC from Saint-Gobain the normal emissivity but only around 1 μm (0.95–1.05 μm) in high vacuum (10^{-4} Pa) using a dual-frequency pyrometer and the normal spectral emissivity goes from 0.80 at 1370 K up to 0.85 at 2270 K [5].

Zolotarev has reported spectral emissivity data for SiC in air at 1700 K for the 1–15 μm wavelength range and they are around 0.83-0.85 with two minima at around 9.2 and 12.6 μm explained by the presence of Si-O and Si-C bonds respectively [6]. This minimum at 12.6 μm is also mentioned by Manara et al. for a SiC sample with 4% porosity at ambient temperature [7].

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Table 1
Chemical composition of Boostec® SiC in ppm except when % is added (from the product sheet of the manufacturer).

SiC	B	free C	SiO_2	free Si	Fe	Al	Ca	K, Mg, Na
98.5%	< 1%	< 0.2%	< 500	< 500	< 500	< 400	< 30	< 1 for each

Hatzl et al. measured the normal spectral emissivity in the wavelength range 0.6–15 μm from 800 to 1200 K on sintered SiC (from FCT Germany) using a direct method [8], and they have obtained values around 0.85 up to 9 μm followed by the Christiansen point around 10.5 μm and a decrease at around 12.6 μm [6,7]. The calculated total emissivity finally goes from 0.82 at 770 K up to 0.84 at 1270 K. It is noticed by the authors that according to Planck law, about 2.4% of the radiance is missing at 1270 K and about 8.5% at 770 K due to the wavelength range.

Finally, only Cagran et al. have measured the normal emissivity on oxidized β-SiC samples in the wavelength range 2–20 μm and from 570 to 1120 K in air using a direct method [9]. Emissivity is stable around 0.81–0.85 up to 8 μm to nearly reach 1 at 10 μm (Christiansen point) then there is a sharp decrease around 12 μm and again an increase to reach only 0.75 around 20 μm.

Wang et al. have obtained normal spectral (6–16 μm) emissivity data for SiC coating deposited by CVD (20 μm of β-SiC on C/SiC composite) from 1270 to 1870 K. As observed before, the emissivity of the SiC coating is stable – around 0.60 – from 6 to 9 μm, then increases up to only 0.80 around 10 μm and then a sharp decrease occurs down to 0.30 around 12.6 μm followed by an increase up to 0.60 at 16 μm [10]. The total emissivity of the SiC coating was calculated to be equal to 0.52 around 1270 K then increases up to 0.63 at 1870 K and then increases a little bit to 0.66 at 1970 K. These results are comparable to those we have previously obtained on a β-SiC coating on C/SiC composite at lower air pressures (4 and 200 Pa) with values for the total (0-6-40 μm) hemispherical emissivity of around 0.60 at 1070 K up to a stable value of around 0.72 from 1270 to 1900 K [11].

To conclude, even if some data are in agreement, few total directional and hemispherical emissivity values can be found at very high temperatures for SiC and the differences between vacuum and air conditions are not clearly shown with precise material surface characterization. SiO_2 being semi-transparent, it remains necessary to measure emissivity in several conditions in order to have different thicknesses of the oxide layer: with the semi-transparency comes the dependence of the thermal radiative properties to extrinsic parameters such as the thickness [12] as well as the porosity and texture [13].

This is why we have proposed the following study to show the effect of temperature and pressure, linked to the composition and roughness of the surface, on the total hemispherical emissivity of sintered α-SiC.

2. Material, methods and set-up

In this section, the SiC material, the methods used for emissivity measurement and for material characterization, and the experimental device are described.

2.1. Material

Sintered α-SiC cylindrical samples from Mersen Boostec (Bazet, France) called Boostec® SiC were delivered in the size 40 mm diameter and 2 mm thickness. This material is a polycrystalline ceramic obtained by pressureless sintering at T > 2300 K in a protective atmosphere. This process yields a pure silicon carbide with less than 1% B, few traces of free carbon (< 0.2%) and free silicon (< 500 ppm) as shown in Table 1. Its low residual porosity is fine and completely closed. The very strong covalent Si-C bond gives Boostec® SiC exceptional physical properties that are particularly stable over time: high stiffness and hardness, low thermal expansion, high chemical and thermal stabilities. Its density is equal to 3.15 g cm^{-3} (theoretical 3.21 g cm^{-3}).

2.2. Methods used

The direct method developed at PROMES-CNRS for the evaluation of the emissivity is based on the measurement of the directional radiance of the material in a given wavelength range and of the true temperature [14–16]. For the evaluation of the total hemispherical emissivity, the radiance measurement is made in the range of 0.6–40 μm for different incidence angles by 10° step (from 0° to 80°, plus 45° and 75°), 0° being normal incidence. Directional values are then extrapolated to zero for the angle of incidence of 90° by cubic spline functions which are integrated to obtain the hemispherical emissivity. The uncertainty is around 3% between 1500 and 2300 K for the spectral range of 0.6–40 μm.

The measurements have been performed in high vacuum (up to 10^{-4} Pa) or in controlled atmosphere in air.

The modification of the optical properties - refractive index and extinction coefficient - of the samples after different treatments were investigated with phase modulated spectroscopic ellipsometry measurements with a Jobin-Yvon UVISEL (0.75–4.75 eV and fixed 70° incident angle) apparatus. First, a pristine SiC sample is measured and considered as reference. Since this latter is few-centimeter thick and absorbent in the investigated wavelength range, this measure is used to simulate the other samples, where an oxidized layer was formed. This new oxide layer was simulated by a Tauc-Lorenz model [17,18] commonly used to describe the dielectric function of insulator or semiconductor thin films.

Scanning Electron Microscopy (SEM) was realized using a Hitachi S-4500 microscope and 3D profilometry was performed with the confocal microscope Leica DCM 3D to measure the surface topography via several parameters such as Sa the arithmetic surface roughness, Sq the root mean square surface roughness and Sz the maximum height.

2.3. Experimental set-up MEDIASE

The experimental set-up MEDIASE (Fig. 1) installed at the focus of the 1 MW solar furnace of Odeillo consists of a stainless steel vessel with a capacity of about 60 l, equipped with a turbo-molecular pumping system. A hemispherical silica window, 35 cm in diameter, is placed in front of this vessel and allows concentrated solar radiation to heat the sample (up to 10 MW/m^2). The sample (40 mm in diameter, 2 mm thickness) is maintained in the water-cooled sample-holder by three thin alumina needles placed at 120° with a spring, and is exposed to concentrated solar radiation by the gradual opening of the doors of the solar furnace. The sample temperature is measured on the rear face using the two-color pyro-reflectometer developed in our laboratory [19]. This pyro-reflectometer (accuracy 1%) is calibrated on a blackbody before and after each series of measurements. The temperature of the blackbody that can reach 3000 K is measured with a calibrated pyrometer Impac. The radiance is measured using a bolometer with an