

Infrared radiative properties of plasma-sprayed BaZrO₃ coatings

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Received 18 October 2013; received in revised form 30 April 2014; accepted 6 May 2014

Available online 28 May 2014

Abstract

The room temperature directional-hemispherical reflectance and transmittance spectra of free-standing atmospheric plasma-sprayed BaZrO₃ coatings with different thicknesses were measured in the wavelength range of 0.8–15.0 μm, and the absorption coefficient and scattering coefficient as a function of wavelength were extracted using the modified four-flux model. Results showed that BaZrO₃ is a high scattering, low absorption material at the wavelength <6 μm, where turbine engine thermal radiation is most concentrated. The absorption coefficient of BaZrO₃ starts to increase rapidly at wavelength of 6 μm, indicating that BaZrO₃ is high absorbing and opaque in the long wavelength range. A pronounced absorption peak occurs at a wavelength of 7 μm, and is associated with a BaCO₃ coating impurity. The scattering coefficient of BaZrO₃ decreases with the increase of wavelength in the whole measured wavelength range, caused by the decrease of the relative size of the scattering center compared with the wavelength.

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Keywords: BaZrO₃; TBCs; Thermal radiation; Plasma spray; Scattering

1. Introduction

The perovskite-type barium zirconate (BaZrO₃) is of interest for various applications. As a functional ceramic, it is reported as a potential material for humidity sensor, electro-optic device, and high capacity memory cells.^{1–3} As a well-known refractory material, BaZrO₃ has very high melting temperature (2600 °C), high thermal and chemical stability, good mechanical properties, and low thermal conductivity, which makes it a good candidate for high-temperature applications such as crucible material for growing crystals of high-temperature superconductors,^{4–6} substrate for thin and thick film deposition,^{7–10} and interfacial coating for alumina fiber-alumina matrix composites.^{11,12} It is also of interest in nuclear safety studies as it can be formed in the fuel of nuclear reactors at high temperature.¹³ BaZrO₃ additionally offers the possibility to create defects in its perovskite structure by proper doping, which enables the properties to be

selectively influenced and thus it can be used as solid electrolyte for fuel cells due to its proton conduction.^{14,15} These refractory characteristics and tailorable properties make BaZrO₃ very attractive for material development in thermal barrier coatings (TBCs) application.^{16–20}

TBCs are used to protect the superalloy components from the hot gas stream in gas-turbine engines. They are consisted of four layers: the substrate, the bond coat, the thermally grown oxide, and the ceramic top coat.²¹ Under the protection of so-called external film cooling, the ceramic top coat is exposed to the gas turbine hot stream, the temperature of which can reach up to 1700 °C. At such a high temperature, the thermal radiation property of top coat material is very important as the thermal radiation contributes to both the heat transfer from the hot gas to the coating surface and within the coating. What is more, with the continuous increase of turbine inlet temperature, driven by the requirement for an even higher efficiency, the radiation heat transport through TBCs becomes a larger contributor to the overall heat transfer due to the fourth power dependence of thermal radiation on the temperature. According to Wien's displacement law as a body increases in temperature, the emission spectrum

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Table 1
Plasma spray parameters.

Coating	Arc current (A)	Coating distance (mm)	Plasma gas Ar/H ₂ (L/min)	Carrier gas Ar (L/min)	Powder feed rate (g/min)
BaZrO ₃	660	80	30/15	4.0	40

becomes more pronounced and the peak of emission intensity shifts toward shorter wavelengths. For example, the peak radiant intensity of a blackbody occurs at 2.28 μm at 1000 °C and at 1.47 μm at 1700 °C. Therefore, for the typical range of temperatures in the gas turbine hot section, a considerable fraction of the radiant energy is in the infrared wavelength range. It was reported that 95% of the radiation in a combustor was in the wavelength range of 0.8–9.5 μm .²²

Because of the diverse applications of BaZrO₃, extensive research has been devoted to it, mostly focusing on the synthesis of high purity BaZrO₃ powders,^{23–31} development of high density bulk BaZrO₃ ceramics using different sintering profiles,^{26,29,32,33} and characterization of the thermophysical, thermodynamic, electronic, and photoluminescence properties of BaZrO₃.^{13,16,34–43} There are only a few reports related to the optical properties, which were the focus of the theoretical calculation of the refractive index of BaZrO₃ and ultraviolet-visible absorption.^{3,44–46}

Therefore, in this paper, free-standing BaZrO₃ coatings with different thicknesses were prepared by atmospheric plasma spray (APS) and the room temperature directional-hemispherical reflectance and transmittance spectra of these coatings were measured in the wavelength range of 0.8–15.0 μm . To determine the absorption and scattering coefficients, which are independent of the coating thickness, the modified four-flux model was employed. It should be noted that the scattering coefficients determined in this investigation are specific to the APS coating microstructure and should not be generalized to BaZrO₃ produced by other processing methods. While this limits generalization of the scattering coefficient results, the APS microstructure is of practical interest due to its widespread industrial use. By matching the predicted and measured reflectance and transmittance of free-standing coatings, the absorption and scattering coefficients of BaZrO₃ prepared by APS were obtained.

2. Experimental details

2.1. Preparation and characterization of BaZrO₃ coatings

BaZrO₃ powders of 99.9% purity with particle size less than 10 μm were obtained from Sigma–Aldrich. These powders were compressed into green disks under 400 MPa and sintered at 1600 °C for 4 h. The sintered disks were then crashed and sieved to make the plasma spray powders with particle size of 44–210 μm . Using Sulzer Metco 9MB plasma gun, five ceramic coatings with different thicknesses were atmospheric plasma-sprayed on sacrificial carbon disks with 25.4 mm in diameter and 2.0 mm in thickness. The spray parameters were listed in Table 1.

Free-standing specimens were obtained for reflectance and transmittance measurements by burning off the carbon disk substrate at 800 °C for 4 h in a furnace. Then the X-ray diffraction (MiniFlex XRD, Rigaku Corporation, Japan) with Cu K α radiation $\lambda = 1.54178 \text{ \AA}$ at a scan speed of 1°/min was used to characterize the composition of the plasma-sprayed free-standing coatings. The densities of BaZrO₃ specimens were measured by Archimedes method following the measurement process in reference^{47,48} and the porosities were then calculated by comparing with the theoretical density of BaZrO₃ which is 6.229 g/cm³ according to PDF#06-0399 and references.^{16,23} By comparing with both mercury intrusion method and image analysis method, Archimedes method was reported to be an easy and reliable method to measure the porosity of TBCs.⁴⁹ The coating thicknesses were obtained by dividing the measured coating volume by the coating area (5.065 cm²). This indirect thickness measurement is believed to have better average over the localized thickness variations inherent in plasma-sprayed coatings than determining thickness using a microscope to measure the distance between poorly defined coating boundary locations. To observe the cross-section microstructure, the specimen was sectioned, mounted, ground and polished by standard metallographic preparation techniques. The polished specimen was then platinum coated prior to observe using an FE-SEM (Quanta 3D FEG, FEI Company, USA). Fourier transform infrared (FTIR) spectrometer (Bruker Tensor 27, Billerica, MA) was also used to identify the impurity of the coating.

2.2. Reflectance and transmittance measurements

The room temperature directional-hemispherical reflectance and transmittance spectra of free-standing BaZrO₃ coatings were measured in the wavelength range of 0.8–15.0 μm using a Nicolet 760 FTIR spectrometer (Thermo Nicolet, Madison, WI) equipped with an integrating sphere accessory (Labsphere, North Sutton, NH). The specimens were illuminated at normal incidence. Spectra in the wavelength range of 0.8–2.5 μm were collected using a quartz halogen lamp source and a CaF₂ beam splitter. Spectra in the wavelength range of 2.0–15.0 μm were collected using an Ever-Glo Mid-IR source and a KBr beam splitter. All spectra were acquired using an uncooled deuterated triglycidyl sulfate (DTGS) detector. See reference⁵⁰ for more detailed test information. According to Kirchhoff's law, the absorptance (A) is equivalent to the emittance (E), and because the reflectance (R) and transmittance (T) were measured at room temperature, there was no emission of the coating in the measured wavelength range. So the absorptance can be calculated from closure using the following Eq. (1):

$$A = E = 1 - R - T \quad (1)$$

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