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Structural stabilization of EBC with thermal energy reflection at high temperatures

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ABSTRACT

Structural stability of an environmental barrier coating (EBC) with thermal energy reflection function that has a periodic layered structure consisting of $Y_2Ti_2O_7$ (YT) and Al_2O_3 is essential to maintenance of the EBC performance at high temperatures. The effect of adding Al to YT layer on the structural stability was investigated using model samples in which Al_2O_3 layer was formed on both the YT and Al-doped YT (AYT) substrate by aerosol deposition (AD). Exposure to heat selectively dissipated the Al_2O_3 layer on the YT substrate near the interface between the layer and the grain boundaries of the substrate. In contrast, the Al_2O_3 layer on the AYT substrate remained intact upon heating when Al was added at the solid solution limit. The Al_2O_3 layer was found to exhibit a pronounced degree of (0001) basal plane texture. An increase in the impact velocity of particles during AD effectively developed the basal fiber texture.

1. Introduction

SiC fiber reinforced SiC matrix composites (SiC/SiC) are predicted to be the material of choice for the next generation of gas turbine engines, since they can tolerate both high pressure and temperature [1–3]. However, SiC/SiC materials suffer from oxidation and volatilization in high temperature combustion gas environments [1,3,4], and so environmental barrier coatings (EBCs) are essential to overcome these limitations. EBCs capable of effectively reflecting the thermal energy radiated from a high temperature heat source are anticipated to provide oxidation protection while also decreasing the SiC/SiC surface temperature so as to drastically reduce the oxidation of the SiC/SiC. Additionally, the requirement for air cooling of SiC/SiC components would be significantly reduced, since the temperature of the material would be lower.

Thermal reflectivity can result from the formation of a periodic layered structure consisting of two different oxide materials having a significant difference in refractive index (n), as a consequence of the interference of electromagnetic waves [5,6]. Based on this phenomenon, we have previously proposed EBCs exhibiting thermal energy reflection for use with SiC/SiC [7]. These EBCs have a periodic layered structure composed of $Y_2Ti_2O_7$ (YT) and Al_2O_3, with Si and mullite bond coats on the SiC/SiC substrate. The refractive index of YT is $n = 2.3$, which is higher than that of Al_2O_3 ($n = 1.7$). Because YT has excellent water vapor corrosion resistance, it is applied to the upper surface to prevent the reduction of underlying layers and of the substrate. This configuration also theoretically increases the reflection of thermal energy compared with an arrangement in which the Al_2O_3 (having a low n) is on the top surface [8]. Because YT is highly permeable to oxygen, the Al_2O_3 layers and the mullite bond coat layer act as oxygen shields.

Fig. 1 shows a scanning electron microscopy (SEM) micrograph of a cross-section of a multi-layer sample fabricated in a previous study [7]. In that study, the YT layers were applied using a sol-gel method, while the Al_2O_3 layers were applied by aerosol deposition (AD) (Fig. 1(a)). As a result, the YT layers were approximately 30% porous and the Al_2O_3 layers were dense. The optical reflectivity of the model sample produced in this prior study was found to be lower than the value estimated based on the matrix theory for the analysis of multi-layer systems [7]. However, the reflectance

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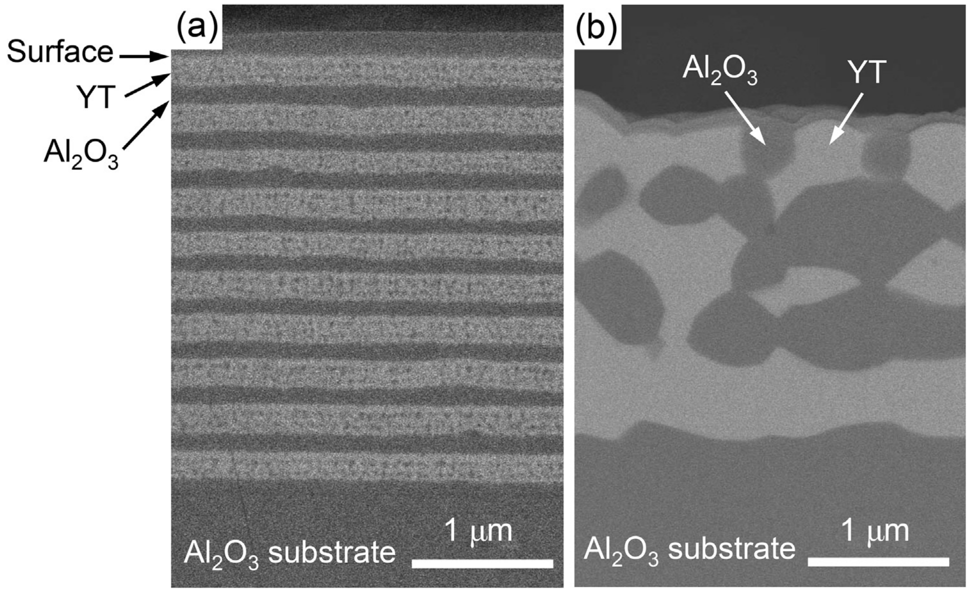

Fig. 1. SEM micrographs showing a cross-section of a multi-layered model sample from a prior study. (a) In the as-coated condition and (b) after heat exposure.

spectrum estimated by considering the decrease in refractive index resulting from pores in the layers was in good agreement with the experimentally acquired spectrum. Therefore, the lower than anticipated reflectivity was attributed to the many voids in the YT layers, which lowered their refractive indices and decreased the refractive index difference between the YT and Al_2O_3 layers. This result demonstrates that it is essential to generate highly dense layers to obtain a system that exhibits thermal energy reflection and acts as an environmental barrier. The exposure of this YT/Al_2O_3 multilayer sample to 900 °C during the sol-gel process did not damage the periodic layered structure, although exposing the same sample to 1400 °C for 1 h caused the periodic structure to collapse as the layers underwent shrinkage during sintering (Fig. 1(b)). Thus, it was determined that the YT and Al_2O_3 layers have a tendency to react with one another at high temperatures. Furthermore, the results of X-ray diffraction (XRD) analysis suggested that the Al in the Al_2O_3 layers might have diffused into the YT layers and dissolved in the YT crystals. Therefore, the diffusion and dissolution of Al into the YT crystals should be considered.

The AD method is a type of gas deposition based on shock loading solidification resulting from the impact of ceramics particles in a vacuum at room temperature, and is able to generate fully dense ceramic coating layers [9–11]. It is generally accepted that an Al_2O_3 coating layer formed by AD will have a random crystal orientation. However, it was recently determined that the layers resulting from this process have a fiber texture composed of (0001) basal planes [12] with a low surface energy [13,14]. The orientation distribution of these layers has been evaluated based on the analysis of coating cross-sections by transmission electron microscopy (TEM) electron diffraction [9,15–17], and Hasegawa has also assessed the texture on a coating plane using an XRD technique known as the Schulz reflection method in conjunction with the pole figures resulting from orientation distribution function (ODF) calculations [12]. The increased degree of texture of the Al_2O_3 coating on a mullite substrate following heat treatment has been demonstrated using electron back scatter diffraction, while that of the Al_2O_3 coating on a YT substrate was demonstrated by a quantitative evaluation of the volume fraction of the (0001) orientation from ODF analysis [12]. In published reports, nitrogen is typically used as the carrier gas during AD [12], although the impact velocity of particles during AD can be increased by employing helium as the carrier gas [9]. However, the effects of varying the impact velocity on the resulting layer texture are unknown.

The goal of the present study was to obtain a better understanding of the structural properties of Al_2O_3 deposited on a YT substrate. The solid solubility limit of Al in YT was initially investigated, and the structural stabilities of Al_2O_3 applied to both a YT substrate and an Al-doped YT (AYT) substrate by AD were evaluated at high temperature. Additionally, the crystal orientation of an Al_2O_3 coating layer formed by AD using a helium carrier gas was evaluated, and the effect of the particle impact velocity during AD on the formation of the texture was examined.

2. Experimental procedure

2.1. Investigation of the solid solubility limit of Al in $Y_2Ti_2O_7$

To ascertain the solid solubility limit of Al in YT, sintered bodies consisting of AYT were fabricated. The solid solubility limit of Al in YT was assessed at 1300 °C because this is the assumed operational temperature of the proposed EBCs with thermal energy reflection. As a first step, the raw AYT powder was synthesized by spray pyrolysis, using a precursor solution composed of $Y(NO_3)_3$ and a 0.05 M aqueous $TiCl_4$ solution to which 0.6–15 cation% $Al(NO_3)_3{\cdot}9H_2O$ had been added. Following the spray pyrolysis step, the product was calcined at 800 °C for 1 h, then molded using a uniaxial press at 20 MPa and finally subjected to cold isostatic pressing at 245 MPa. The green compacts were sintered at atmospheric pressure in air at 1500 °C for 5 h, annealed at 1300 °C for 50 h, and finally cooled by quenching in water. The resulting material was crushed and the lattice constant was measured by powder XRD, using Si (640c, NIST) as the internal standard.

2.2. Fabrication of coated specimens

The raw material for the AD process was commercially available α-Al_2O_3 (TM-DAR, Taimei Chem. Corp.). The YT and AYT substrates were fabricated in the same manner as the sintered bodies described in Section 2.1, except that a cooling rate of 5 °C/min was applied. Specimens with a width of Φ23.5 and a thickness of 0.25 mm were cut from the sintered bodies and their surfaces were polished to a mirror-like finish. The amount of Al added to each AYT