



# Formation and growth of silica layer beneath environmental barrier coatings under water-vapor environment

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## ARTICLE INFO

### Article history:

Received 16 October 2017  
Received in revised form  
30 November 2017  
Accepted 25 December 2017  
Available online 29 December 2017

### Keywords:

Environmental barrier coatings  
Thermally grown silica oxide  
Oxidants  
Diffusion controlled processes

## ABSTRACT

Environmental barrier coatings (EBCs) have widely been employed as protective coatings for SiC-based ceramic matrix composites (CMC-SiCs) against combustion corrosion in advanced gas turbines. During service, a layer of oxides forms beneath EBCs, which plays key role in lifetime of coated CMC-SiC parts. In this study, the kinetics of thermally grown silica oxide (silica-TGO) were examined through rare earth disilicates coated CMC-SiC composites subjected to corrosion under 50% $H_2O$ -50% $O_2$  gas flow environment. Results revealed that growth of silica-TGO followed parabolic behavior, indicating diffusion controlled process. Calculated growth rates and activation energies of silica-TGO suggested that oxygen ion was the main active oxidant involved at relatively low partial pressure of water vapor. Corrosion environments containing different  $H_2O/O_2$  ratios were also investigated and the data indicated that water vapor was also involved in growth of silica-TGO. The role of EBCs in corrosion process was also analyzed and discussed.

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## 1. Introduction

One of the significant trends in gas turbine involved in aerospace industry is to increase the operation temperature [1–3]. Higher operation temperatures may lead to increased specific core power, better efficiency, and reduction in environmentally harmful byproducts [4,5]. To meet the requirements of advanced gas turbines, SiC-based ceramic matrix composites (CMC-SiCs) have partially been utilized as hot-section components to replace current superalloys due to their low densities and high strengths at elevated temperatures [3,6,7]. CMC-SiCs materials develop intrinsic oxidation resistances thanks to formation of a protective silica layer during operation under dry air [8]. However, combustion environments in gas turbine contain 5–10% water vapor, which could react with silica layer to yield volatile products [9–12]. This induces loss of CMC-SiCs protection and final failure of composite components, which usually are solved using protective coatings on CMC-SiCs like environmental barrier coatings (EBCs) [13,14]. EBCs are meant to insulate the composites from direct contact with combustion environment, as well as preventing the composites from reacting with water vapor [13,14].

The durability of EBC coated CMC-SiCs is a potential issue, which determines the successful application of these materials in turbine engines. One approach for improving the lifetime of composites is to enhance EBCs water-vapor resistances. To this end, several EBC systems were developed, including mullite/yttria-stabilized zirconium (YSZ) [13,15], barium-strontium aluminosilicate (BSAS) [13,16–20], and rare earth silicates [21–28]. However, a recent study by Lu et al. [29] indicated that thermally grown silica scale (silica-TGO) beneath EBCs played a key role in durability of coated composites, which could fail if silica-TGO is grown to reach certain thicknesses. Under such circumstances, EBC materials were well kept without erosion by water vapor flow. It is believed that stress induced by growth of silica-TGO forms cracks at silica-TGO/SiC bond coat interface and EBCs spallation, resulting in composites failure. Similar phenomena are widely observed in YSZ-based thermal barrier coating (TBC) systems, where alumina-TGO is found to potentially contribute to service lifetime of TBC [30–34].

Despite the importance of silica-TGO in durability of EBC systems, reports dealing with silica-TGO are still limited. For instance, Haynes et al. [35] confirmed the formation of a thin silica layer beneath dense mullite coated SiC/SiC composites after 500-hours exposure to air containing 15 vol% water vapor at 1204 °C and 10 atm pressure. During engine testing with art-three-layer EBCs like Si/mullite (or mullite+BSAS)/BSAS, silica-TGO forms at the interface between Si-bond coating and mullite (or mullite+BSAS)

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layer [19]. Liu et al. observed silica-TGO scale beneath dense BSAS [17] and  $\text{Sc}_2\text{Si}_2\text{O}_7$  coatings [26] after corrosion at 1250 °C under 50%  $\text{H}_2\text{O}$ -50% $\text{O}_2$ . In environments containing high water-vapor contents (90% $\text{H}_2\text{O}$ -10% $\text{O}_2$ ), silica-TGO could form on SiC/SiC composites beneath BSAS coatings at 1300 °C [16]. Although numerous studies based on silica-TGO have already been reported, only few of them investigated the relationship between silica-TGO and EBCs-coated CMC-SiCs performances. Richards et al. [36] examined the thermal cycle properties of  $\text{Yb}_2\text{SiO}_5$ /mullite/Si-coated SiC at 1316 °C with 1-hour cycle in 90% $\text{H}_2\text{O}$ -10% $\text{O}_2$ . They found that cristobalite TGO formed on the material contributes to poor steam cycling durability of tri-layer EBC system [36]. With regard to reasons leading to formed silica-TGO, most studies generally considered  $\text{O}_2$  and/or  $\text{H}_2\text{O}$  oxidants as the species diffusing through dense EBC layer or transported through cracks in EBCs layer to reach Si or SiC bond layer and form silica-TGO scale [16,17,26,36].

Despite the handful of reports published on silica-TGO, studies dealing with the effect of silica-TGO on CMC-SiCs performances are available [29,36]. This constitutes a critical parameter for evaluating the durability of EBC coated CMC-SiCs, and predicts their service time in combustion environments. However, a little is still known about the mechanism and kinetics of silica-TGO growth under water-vapor containing environments. Lu reported a parabolic growth behavior of silica-TGO in BSAS and  $\text{Y}_2\text{Si}_2\text{O}_7$ -BSAS coated C/SiC composites [29]. However, the growth kinetics were not fully elucidated. Therefore, a number of issues should be addressed to gain a comprehensive view of silica-TGO growth mechanisms. The main oxidants involved in formation of silica-TGO are often oxygen or water vapor. For oxygen, it is not clear what active species (molecular, atomic, or ionic) are involved in the oxidation process. Also, the function of EBCs that is insulating water vapor and/or retarding the diffusion of the oxidants are fully elucidated.

To clarify these unclear issues, the growth kinetics of silica-TGO beneath rare earth disilicates ( $\text{RE}_2\text{Si}_2\text{O}_7$ , RE = Yb, Y) coating under various corrosion environments were evaluated through estimating the growth rate constants of silica-TGO and activation energies of TGO growth. By comparing the obtained data with published values related corrosion of pure SiC, the aforementioned questions were answered.

## 2. Experimental

Two-dimensional (2D) C/SiC composites fabricated by chemical vapor infiltration (CVI) process were used in this study. The SiC matrix was deposited under the following conditions: 1000 °C, 5 kPa pressure, Ar and  $\text{H}_2$  flow rate of 350 ml  $\text{min}^{-1}$ ,  $\text{H}_2$  to methyltrichlorosilane molar ratio of 10 [37], and total deposition time of about 480 h. Specimens with sizes of 40 mm × 5 mm × 3.5 mm were cut from the as-prepared composites, and edges were beveled to 45° by 0.2 mm. A SiC bond coat of ~40 μm in thickness was then deposited on surfaces of the specimens under same deposition conditions as SiC matrix. The total deposition time was controlled to 80 h during the fabrication of SiC bond coat. The resulting C/SiC bars were then polished and cleaned with alcohol.

$\text{Yb}_2\text{Si}_2\text{O}_7$  and  $\text{Y}_2\text{Si}_2\text{O}_7$  were selected as models of coating materials due to their high water-vapor resistances, high service temperatures, and low steam volatilities [21,28]. The powders based on  $\text{Yb}_2\text{Si}_2\text{O}_7$ ,  $\text{Y}_2\text{Si}_2\text{O}_7$  and  $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Al}_2\text{Si}_2\text{O}_8$  (BSAS) were synthesized using the sol-gel process. The detailed preparation process was described in the literature [17,38]. The obtained powders were ball milled in a shock-type high-energy ball-milling machine (QM-3A High-Speed Vibrating Ball Mill, Nanjing T-Bota Sciotech Instruments & Equipment Co. Ltd., Nanjing, China) at 120 rpm for 30 min.

The  $\text{RE}_2\text{Si}_2\text{O}_7$  (RE = Yb, Y) coatings were deposited on clean C/

SiC composites using the slurry method. To prepare the slurries,  $\text{RE}_2\text{Si}_2\text{O}_7$ -BSAS powders were mixed with liquid-phased polysilazane (Institute of Chemistry, Chinese Academy of Science, Beijing, China) at a weight ratio of 85 to 15. For  $\text{Yb}_2\text{Si}_2\text{O}_7$  and  $\text{Y}_2\text{Si}_2\text{O}_7$  slurry, 10 wt% and 30 wt% BSAS were added to disilicates in order to lower the processing temperatures, respectively. All slurries were ball milled in a QM-3A machine at 120 rpm for 1 h, which then uniformly applied to surfaces of C/SiC composites using dip-coating process, followed by heat-treatment in an oven at 70 °C for 5 h. The resulting samples were heat-treated at 400 °C for 1 h under argon to allow the cross-linking of polysilazane. This was followed by pyrolysis at 900 °C for 2 h and heat-treatment at 1400–1450 °C for 3 h under protected argon atmosphere to yield  $\text{RE}_2\text{Si}_2\text{O}_7$  coatings. The polysilazane in the coating would derive to SiCN ceramic during the heat treatment. Its ceramic yield at 1000 °C was about 82% [39]. The pyrolysis of polysilazane during the heat treatment process would result in the formation of pores in the coatings. The small amounts of SiCN ceramic and the pores had little influence on water-vapor resistance according to the previous work [26,29].

The water-vapor corrosion behaviors of  $\text{RE}_2\text{Si}_2\text{O}_7$  coated C/SiC samples were evaluated in alumina tube furnace at elevated temperatures under simulated combustion corrosion atmospheres containing water vapor. The samples were placed in alumina boat crucible, and water vapor was introduced into the tube using oxygen carrier gas bubbling through distilled water using the Belton and Richardson method [40]. Heating tape was used to keep the tube at 110 °C on the furnace side, where water-vapor entrance was located. The condensed water vapor accumulated at the tube's exit side was collected to verify the experimental conditions. To obtain water-vapor partial pressures, distilled water was heated at 46 °C, 81.7 °C and 97 °C to maintain the corrosion environments at 10% $\text{H}_2\text{O}$ -90% $\text{O}_2$ , 50% $\text{H}_2\text{O}$ -50% $\text{O}_2$  and 90% $\text{H}_2\text{O}$ -10% $\text{O}_2$ , respectively.

The microstructural identification was carried out using a scanning electron microscope (SEM), frequently in back-scattered electron (BSE) mode (JEOL-6700F, Tokyo, Japan). Elemental analysis was performed using energy-dispersive spectroscopy (EDS, EDAX, USA). The average thickness of silica-TGO layer were determined from at least 30 different areas according to the coating cross-section microscopes. Most of the silica-TGO thickness were collected beneath the EBC layer which had the analogous thicknesses, pore-free and flat. A Rigaku D/max-2400 diffractometer (Tokyo, Japan) with Cu  $\alpha$  radiation was utilized to determine the X-ray diffraction (XRD) profiles. The data were digitally recorded at (2θ) ranged from 10° to 70° at scanning rate of 0.02°/s.

## 3. Results

### 3.1. Growth kinetics of silica-TGO in $\text{Yb}_2\text{Si}_2\text{O}_7$ coated C/SiC

The as-prepared  $\text{Yb}_2\text{Si}_2\text{O}_7$  coatings were first examined by XRD and SEM. The XRD profiles indicated that main phase of the as-prepared coatings consisted of monoclinic  $\beta$ - $\text{Yb}_2\text{Si}_2\text{O}_7$  (Fig. 1a). The typical surface topography shown in Fig. 1b revealed relatively smooth and uniform morphologies of the as-prepared  $\text{Yb}_2\text{Si}_2\text{O}_7$  coatings. Also, little flaws like cracks and pores could be observed on the surface. The inner coating depicted in Fig. 1c was rather dense and tightly adhered to SiC bond coating. The average thicknesses of the resulting coatings were estimated to 40–60 μm. According to EDS analysis shown in Fig. 1d, the particulates depicted in Fig. 1c consisted of  $\text{Yb}_2\text{Si}_2\text{O}_7$ . These particulates were dispersed in a continuous phase, confirmed by EDS to be BSAS (Fig. 1e).

$\text{Yb}_2\text{Si}_2\text{O}_7$  coated C/SiC samples were subjected to corrosion in 50% $\text{H}_2\text{O}$ -50% $\text{O}_2$  at 1150 °C, 1200 °C, 1250 °C and 1300 °C, respectively. The XRD patterns of the samples after 200 h corrosion time at various temperatures are shown in Fig. 2a. Compared to XRD

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