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Interaction and infiltration behavior of Eyjafjallajökull, Sakurajima volcanic ashes and a synthetic CMAS containing FeO with/in EB-PVD ZrO_2-65 wt% Y_2O_3 coating at high temperature

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ABSTRACT

Yttria rich-zirconia thermal barrier coatings (TBCs) with a nominal composition of 65 wt % Y_2O_3 balanced ZrO_2 were deposited by electron-beam physical vapor deposition (EB-PVD) and tested for calcium-magnesium-aluminum-silicate (CMAS) infiltration resistance. The infiltration studies were performed with a set of one synthetized CMAS composition and two real volcanic ashes from the Eyjafjallajökull volcano located in Iceland and the Sakurajima volcano located in southern Japan. The coatings were tested at 1250 °C for short term (5 min) and long term (intervals from 1 to 20 h). The results indicate a significantly different reaction process for the synthesized CMAS compared with the natural volcanic ashes. The yttria-rich zirconia coatings demonstrate promising results against infiltration by vigorously reacting against the molten glass inducing its crystallization by forming apatite and garnet phases. The formed reaction products effectively sealed the columnar gaps of the TBC and generated a uniform reaction layer that prevented further infiltration.

1. Introduction

Thermal barrier coatings (TBCs), which are extensively used in gas turbines and aero-engine parts, protect the metallic surfaces and engine components from high temperature exposure for extended duration. State of the art TBCs used on gas turbine engines provide thermal insulation to the underlying super-alloy components in the hot gas path sections and, thus, allow engines to operate at temperatures as high as 1200 °C, thereby increasing their thermodynamic efficiency [1–3]. The commonly employed state of the art material for TBCs at present is 7–8 wt % Y_2O_3 stabilized ZrO_2 (7YSZ). The 7YSZ TBCs are commonly deposited by air plasma spray (APS) or electron-beam physical vapor deposition (EB-PVD). While 7YSZ TBCs served the industry for many years, currently, significant attention is paid towards the development of novel TBCs which can withstand higher temperatures ($\geq$1300 °C) [4]. Such TBCs with high temperature tolerance can minimize the fuel consumption by maximizing the gas turbine efficiency, which will subsequently help protect the environment through the reduced carbon emissions. Additionally, different fuel compositions ranging from natural gas to broad range of syngas with high hydrogen contents are being tried for next-generation gas turbine power plants [5,6]. Therefore, the TBCs for next-generation gas turbine systems must have the reliability and durability in diverse chemical, thermal and mechanical environments.

The increasing demand for higher operating temperatures in aero-derivative and land-based gas turbines has brought into attention the effects of infiltrated mineral deposits into hot engine's components [7–11]. The siliceous mineral deposits, commonly consisting of calcium-magnesium-aluminum-silicate i.e., CaO-MgO-Al_2O_3-SiO_2 (CMAS), are ingested as sand, runway debris, volcanic ash, air pollution, and fly ash into gas turbines. The CMAS particles deposit on the hot gas path components and melt generating infiltration into the porous TBC structure (regardless of the deposition technique used) resulting in premature failure due to microstructure degradation, sintering and spallation [12]. The molten CMAS infiltrates the columnar gaps, pores and cracks. While flowing inwards into the TBC it hardens as it flows down the thermal gradient. The hardening of the CMAS compound that also occurs during cooling of the whole component generates a stiffening of the TBC which leads to loss in strain tolerance of the

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coating, thereby generating delamination [13–15]. Subsequently, the interaction of the CMAS melt with the TBC at high temperatures generates a thermo-chemical damage system characterized by the dissolution of the initial metastable tetragonal (t') 7YSZ due to the depletion of Y_2O_3 phase and re-precipitation of Zr with a composition and structure that depends on the CMAS chemistry [14,16].

Due to the serious effects that CMAS attack has on reducing the service life of 7YSZ TBCs, scientific and engineering research community is challenged to better understand the attack mechanisms and design methods for CMAS infiltration resistance. Furthermore, due to the eruption of the Eyjafjallajökull volcano located in Iceland on 2010, the safety regulations for the aviation industry had to be reconsidered due to the large widespread of the volcanic ash cloud generating a substantial economic loss due to the extended air space closure [17,18]. This event motivated the aircraft engine manufacturers to effectively address this volcanic ash (VA) attack issue and reduce economic losses. The most common infiltration mitigation strategies focus on using a reactive material that upon interaction with molten CMAS glass induces its partial or full crystallization sealing the porous features of the coating stopping any further infiltration [7,19–22]. Only a few studies have tested their mitigation strategies with a natural CMAS source such as real volcanic ash or sand [23–25]. Some of the methods proposed include the utilization of overlying coatings on top of the 7YSZ in arresting the CMAS infiltration [26,27]. For instance, Eils et al. [28] have performed studies on a variety of yttria-zirconia coatings (ZrO_2, $Y_2O_3 \cdot ZrO_2$ and Y_2O_3) for CMAS mitigation, where the formation of oxyapatite and garnet phases exhibited promising results against CMAS infiltration. The objective of this work was to study the CMAS infiltration resistance of yttria rich-zirconia with a composition of 65 wt % Y_2O_3 (rest ZrO2) produced by EB-PVD process (65YZ). The ultimate goal is to employ the proposed coating system in multilayer TBC systems based on a standard 7YSZ coating for thermal insulation and a yttria rich-zirconia top coat for corrosion resistance. While high yttria based coatings show a potential for CMAS arrest in TBC systems [19,24,25,28–30], there is no available literature for infiltration tests using real volcanic ash on EB-PVD yttria rich coatings at this time. Thus, a better understanding of the interaction between CMAS/VA with –high yttria coatings at high temperature, as presented and discussed in this paper, can lead to significant benefits in the performance and lifetime extension of TBCs for aero-derivative and land based gas turbine engines.

2. Experimental details

2.1. CMAS and volcanic ash preparation

Two real volcanic ashes (VA) and one synthetized CMAS powder were used for the infiltration studies. The volcanic ashes were collected from site corresponding to the Eyjafjallajökull volcano located in Iceland and the Sakurajuma volcano located in southern Japan. The CMAS powder (CMAS 1) was synthetized matching the chemical composition of CMAS deposits in screened engine hardware, its preparation is described elsewhere [9,31]. The chemical composition and summary of the used powders are presented in Table 1. Differential scanning calorimetry (DSC) measurements were performed to determine their melting temperatures which are listed in Table 1. The powders were analyzed between room temperature and 1300 °C in a STA 449 F3 Jupiter equipment (Netzsch, Selb, Germany).

2.2. Sample preparation

Yttria-zirconia samples with higher yttria content were coated on alumina (Al_2O_3) substrates using EB-PVD technique. Evaporation was performed using a jumping beam system on dual evaporation sources namely 7YSZ (source 1) and 100 wt % Y_2O_3 (source 2) as seen in Fig. 1. The deposition set up used generated a variation in the yttria content with respect to the location to the evaporation source meaning the closer the sample to the 7YSZ source, the lower the yttria composition. The selected sample composition had a 65 wt % yttria (rest zirconia) and a thickness of 150–165 μm.

2.3. Infiltration experiments

Infiltration experiments were carried out by depositing CMAS/VA on top of the coated TBC samples in the amount of 20 mg/cm^2. Infiltration temperatures were chosen at 1250 °C for short term (5 min) and long term (1–20 h). Short term tests were performed in a cyclic furnace by heating up the samples to 1250 °C in 8 min, following isothermal heating for 5 min and air quenching to room temperature. Long term tests were performed in a chamber furnace with a heating range of 10 K/min, following isothermal heating for ranges of 1–20 h and furnace cooling to room temperature.

2.4. Characterization

CMAS/VA were mixed with yttria rich zirconia-powder obtained from the as coated samples to determine the formed phases upon CMAS/VA-high yttria reaction. The 65YZ powder was mechanically removed from the as coated samples by scratching it off the substrates. The final powder mixture used for XRD analysis had a composition of 40 wt % CMAS/VA and 60 wt % of yttria rich powder (65 wt % yttria). Subsequently, the powder mixture was heat treated on platinum sheets for 10 h at 1250 °C, followed by furnace cooling to room temperature. The post heat treated powder mixture was mechanically removed from the platinum sheets and crushed using mortar and pestle for30 min to obtain fine powder in order to facilitate the XRD. The measurements were carried out using Si single crystal holders in a Siemens D5000 diffractometer using CuKα radiation with a secondary graphite monochromator (EVA/Topas 4.2 software package, Bruker AXS, Karlsruhe, Germany). Standard metallographic techniques were used for sample preparation in the cross-section analysis. Scanning electron microscopy (SEM) (DSM ultra 55, Carl Zeiss NTS, Wetzlar, Germany) techniques were performed. The system was equipped with an energy-dispersive spectroscopy (EDS) (Inca, Oxford Instruments, Abingdon, UK) to characterize the infiltrated zones and depth of infiltration. The reaction products formed by the interaction of

Table 1
Chemical composition summary for used CMAS/VA.

Label	Chemical composition (Mol. %)								Crystalline phase	Melting point
	SiO_2	CaO	MgO	Al_2O_3	FeO	TiO_2	Na_2O	K_2O		
CMAS 1	41.7	24.7	12.4	11.1	8.7	1.6	0.0	0.0	Pryoxene-Anorthite	1250°c
Japan ash	66.3	11.4	0.3	15.7	1.4	0.0	4.3	0.7	Anorthite-Amorphous	1170°c
Iceland ash	49.7	12.5	6.1	7.4	17.6	4.3	2.0	0.4	Amorphous	1150°c