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Review article

Rare earth silicate environmental barrier coatings: Present status and prospective

Yue Xu[a,*], Xunxun Hu[b], Fangfang Xu[c], Kunwei Li[d]

[a] School of Chemistry and Environment, Beihang University, Beijing 100191, China
[b] School of Materials Science and Engineering, Beihang University, Beijing 100191, China
[c] Guobiao (Beijing) Testing & Certification Co., Ltd., 2 Xinjiekouwai Street, Beijing 100088, China
[d] China National Institute of Standardization, 4 Zhichun Road, Beijing 100191, China

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ABSTRACT

Due to drastic decreasing in mechanical properties at relative high temperature, traditional nickel based super alloys are replaced by Si-based non-oxide ceramics in the application of high temperature aero-engines. In order to reduce the spallation and deformation of aero-engine blades in the environment containing high temperature water vapor and oxygen, protection coatings on the surface of the ceramics are required. Owing to high temperature stability, superior oxidation resistance and corrosion resistance properties, rare earth (RE) silicates are promising as candidates and play an important role in improving the high-temperature mechanical/thermal properties of Si-based non-oxide ceramics. In this review, recent progress in the research and development of environmental barrier coatings (EBCs) are summarized. Development of EBCs is presented, and the multi-scale structures and properties of each part are introduced. In addition, the merits and demerits of each preparation technique are discussed. As a promising candidate for the application in high temperature aero-engines, Si/mullite/$Lu_2Si_2O_7$–Lu_2SiO_5 EBCs are highlighted.

1. Introduction

Developing heat engines with superior properties such as high flow ratio, high thrust weight ratio and high inlet temperature has received persistent attention for decades. Among these properties, high inlet temperature is a vital important factor in achieving the high thrust weight ratio and high thermal efficiency for heat engines [1]. For example, outlet gas temperature of 1650 °C in combustor corresponds a thrust weight ratio of 8, in order to achieve the thrust weight ratio of 10–12, an engine rear outlet temperature of 1850 °C or above is needed [2]. Ni-based super alloys, widely used as hot section material, can resist the temperature up to 1075 °C which is near their intrinsic limit. The super alloys has been unable to satisfy the rapid development and urgent requirement of the advanced aviation engines. Scientists have focused on the development of components made from high temperature ceramic materials [3,4]. The silicon-based non-oxide ceramic materials (such as silicon carbide, ceramic matrix composites), which exhibit superior high-temperature strength and durability, have promised to replace nickel based high-temperature alloy as hot section components of aero-engine [5,6]. Silicon carbide provides the high temperature mechanical properties needed for engine applications. However, it suffers from the rapid recession at fast combustion environment in which (oxygen, water vapor, etc.) cause degradation of its superior properties [7–13], as shown in Fig. 1.

These corrosion problems can be alleviated by EBCs, preventing Si-based non-oxide ceramic materials from reacting with water vapor [14–16]. Since EBC_S involves with the preparation techniques and application of new materials, investigating suitable preparation technology and screening out optimal materials, have become the hot-spot of investigation among scientists in the world.

This review tries to highlight the significant progress of EBCs and the preparation technology in the past decades. We first introduce the development and the necessity of EBCs, and then we highlight the current research progress, focusing on the most promising EBC candidates and the preparation techniques. Finally, we point out the existing problems and propose our research directions which could lead us to the next generation of EBC systems.

2. Development of EBCs

In the past few decades, EBC_S have been used as a protective layer for Si-based non-oxide ceramics substrate that inhibits the corrosion in the presence of water vapor and molten salt. Fig. 2 shows the change in the temperature capability of ceramic matrix composites with EBCs.

* Corresponding author.
E-mail address: xuyue@buaa.edu.cn (Y. Xu).

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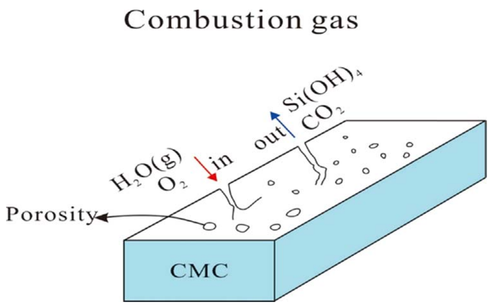

Fig. 1. Ceramic matrix composites reaction with oxygen and water vapor at high temperature.

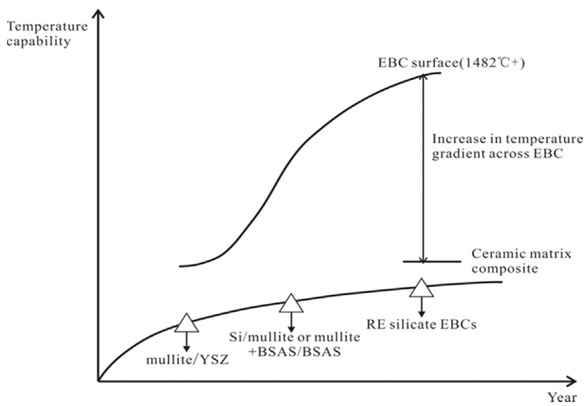

Fig. 2. Change in the temperature capability of ceramic matrix composites with EBCs.

The earlier EBCs consist of a mullite bond layer and an yttrium-stabilized zirconia (YSZ) top coating [17,18]. Mullite has low density, low thermal conductivity, high oxidation resistance and chemical compatibility, as well as similar thermal expansion coefficient to Si-based non-oxide ceramics [19–21], suitable for $EBC_{S.}$ However, mullite degrades seriously in combustion environment due to the selective corrosion of silica by water vapor [22–24]. Different approaches have been attempted to overcome these problems. It has been found that yttrium-stabilized zirconia (YSZ) shows a better stability in water vapor [25]. Therefore, a ZrO_2-8 wt% Y_2O_3 layer was coated over mullite by plasma spray. Unfortunately, thermal stress was generated in this EBC system due to mismatch on the coefficient of thermal expansion (CTE) [26], leading to delamination of the system eventually.

The second generation of EBCs is a system of Si/mullite or mullite +BSAS/BSAS (1-xBaO-xSrO-Al_2O_3-$2SiO_2$, 0≤x≤1) [27,28]. BSAS is used to replace YSZ as top coating material owing to its excellent CTE match with SiC and relatively low activity of silica when compared with mullite. Meanwhile, BSAS also shows excellent corrosion resistance, creep resistance and high strength at high temperature [29]. However, there are some intrinsic defects in the BSAS-based EBCs. BSAS suffers significant degradation at temperatures higher than 1400 °C. Fig. 3 shows the cross section Si/mullite/BSAS EBCs on MI CMC after 1000 h in 90% H_2O-balance O_2 at 1300 °C with 1 h cycles. The crack and pores are anticipated in Fig. 3(b), owing to the reaction of BSAS with SiO_2 on the surface of the Si layer, which forms a low melting point glass-state substances. The results demonstrate that the service temperature is limited to be lower than 1300 °C in Si/mullite/BSAS EBCs.

In order to overcome the limitations of the above two generations of EBCs and to increase the performances of Si-based non-oxide ceramics at high temperature, some advanced coating systems have been explored. RE silicates are considered as promising alternatives for the previous EBC due to their low thermal expansion coefficient and the ability to endure temperatures higher than 1482 °C for thousands of hours [30]. Based on the longer lifetime and superior comprehensive performance in aero-engine working environments of RE silicates, NASA developed a third generation of EBCs system, which consists of a Si bond coat, mullite intermediate layer, and rare earth silicates top layer, as shown in Fig. 4.

3. RE silicate EBCs

3.1. Optimizing RE silicate as EBC surface layer

Based on the report from NASA Glenn Research Center, Lee et al. have developed EBCs in which the surface coating can withstand a temperature higher than 1482 °C and the substrate can sustain a temperature of 1316 °C for over thousands of hours [31]. Since Si and mullite are the currently preferred bond coat and intermediate layer for SiC substrate, respectively. How to select a suitable RE silicate as top layer in optimizing the mechanical and thermal properties, as well as how to make thermo-chemical capability of substrates and intermediate layer match well, are the main task that has to be considered during the development of an EBC system.

Therefore, the thermo-physical properties of different RE silicates are of significantly importance for the design of EBCs. The relevant thermal expansion coefficients data for SiC and some promising rare earth silicates are listed in Table 1.

Alberto et al. [36] investigated the thermal expansion coefficients of RE pyro-silicate at 303–1873 K and found that the thermal expansion coefficients values differ significantly from one polymorph to the other. But, β-$RE_2Si_2O_7$ (RE=Sc, Lu and Yb) and γ-$Y_2Si_2O_7$ showed similar CTE values of $4.0\times10^{-6}\,K^{-1}$ with those of SiC and mullite, this is considered to be a suitable primary coating materials.

It is also known that the phase transformations occurs with most materials at elevated temperatures. However, this is not likely to be the situation for an EBC on ceramic matrix composites. Liddell et al. [37] reported that RE disilicates (RE=Y, Tm, Er and Ho) have several polymorphs with substantial disparity in density, which makes them unsuitable as the candidates of EBCs. RE monosilicates, (RE=Lu Yb, Y and Er) do not have polymorphs [38,39]. Al Nasiri et al. [33] concluded that RE_2SiO_5 (RE=Y, Yb, Er and Lu) had a great potential to be used as top layer for EBCs after studying the thermal properties such as thermal expansion, thermal diffusion, and thermal conductivity. Since β-$RE_2Si_2O_7$ (RE=Sc, Lu and Yb), γ-$Y_2Si_2O_7$ and RE_2SiO_5 (RE=Y, Yb, Er and Lu) show significant advantages of thermal-physical properties when compared with other RE silicates, they satisfy one of the primary design requirements for the EBC application. Their thermochemical compatibility with mullite, as well as durability in service conditions need to be studies systematically.

3.2. RE silicate EBC systems

The thermo-properties of numerous RE silicates have been studied extensively in terms of their applications in EBC, Based on the materials, the coating includes the RE monosilicate EBCs, the RE disilicate EBCs, and the combination of both. These are shown in Table 2.

3.3. Characteristics of RE silicate EBCs

Through the comprehensive understanding of the RE EBCs development, their composition has experienced a change from single to multiple layers. The tri-layer coatings usually have a Si bond layer, a mullite intermediate layer, and a RE silicate top layer.

The Si-bond layer usually acts as an oxygen absorbent to prevent oxidation of silicon based non-oxide substrate. The mullite intermediate layer, serves not only as a diffusion barrier against liquids and high temperature gases, but also a preventor for the reaction between thermally grown silica and top layer. The RE silicate top layer is characteristic with high melting point, low steam volatility and low