

A review on development and application of self-healing thermal barrier composite coatings

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ABSTRACT

To improve the hot section metallic parts durability in advanced gas-turbine operating in power generation and aircraft, thermal barrier coating (TBCs) are extensively utilized to increase their lifetime. The reason for applying coatings on these components is the improvement of their physical properties, mechanical properties, and outer look. The self-repairing ability of materials is very promising due to expanding the service time of materials and it is also beneficial in terms of human safety and financial aspects. In this review article, structure, properties, limitations, and the modification approaches of TBCs were studied. In addition, self-healing agents for TBCs including SiC, MoSi₂, TiC were introduced, which release their oxide by reaction with air and O₂ that are able to heal the pores/cracks in the coatings. In this regard, their coating methods, mechanism, and applications in TBCs were reviewed.

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1. Introduction

Most industrial processes involve operating under harsh environments of high temperature, high pressure, increased temperature gradients, and foreign object damage (FOD), corroding and oxidizing atmosphere, and large stresses on individual components. Marine, aero, and industrial gas turbines are examples of such industrial processes [1].

Coatings on the surface of materials improve materials appearance, mechanical, and physical properties [2-4]. A thick barricade will be built on the surfaces of metals by the coating against atmospheric oxygen and water [5-7]. Thermal barrier coatings (TBCs) are extensively utilized for the durability enhancement of hot metallic components operating in advanced gas turbines or power generation and aircraft [8]. However, the initiation of cracks, their propagation, and coalescence result in the coating spallation, which exposes the hot-section metallic components

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to high temperature and oxidation [9, 10].

In the past 15 years, a considerable section of research activities has been allocated to self-healing materials [11-13]. The aim of using self-healing materials is to restore the mechanical and physical integrity and properties rapidly after being damaged. The self-repairing ability of materials is a treasured characteristic due to expanding the material service period offering benefits in terms of financial aspects and human safety requirements [5, 14].

This review article focuses on the TBCs and their structures, restrictions, and modifications techniques [15]. Subsequently, self-healing materials and self-healing composite TBCs are introduced, and finally, their coating methods, mechanism, and applications in TBCs are discussed [16].

2. Structure of TBCs

In order to protect metallic parts of vanes, blades, and combustion chambers in jet engines and gas turbines from oxidation and corrosion at high temperatures, TBCs are often applied on their surfaces [17-19]. The TBCs primary function is to enable the hot operating parts to bear higher service temperatures beyond the underlying metals limit temperature. When the hot sections are protected by TBCs with a thickness in the range of 150 to 800 μm , the inlet temperature can be increased by 100 to 300 $^{\circ}\text{C}$ [20].

There are three layers in the TBC structure applied on the surface of a metallic substrate: 1) the bond-coat (BC) applied on the surface of the substrate; 2) thermally grown oxide (TGO), which is generated at the BC/TC interface; 3) the ceramic top-coat (TC) [21]. BC and TC commonly consist of MCrAlY (M can be Co, Ni, Fe, or their combinations) layer, and zirconia that is partially stabilized with Yttria (YSZ), respectively. The TGO oxide layer mostly composed of Al_2O_3 , is generated at the BC/TC interface during high-temperature service in the air [22, 23]. YSZ has a porous structure and with the increase in service time, oxygen penetrates the coating or cracks and reacts with the bond coat materials. Due to this reaction, the TGO layer grows by the metallic BC selective oxidation and is mainly composed of alumina. One of the main reasons for cracking, delamination, and spallation of YSZ has been reported to be the TGO layer growth [24-27].

Zirconia-based coatings are favored in TBCs owing to their relatively high thermal expansion coefficient (TEC), good oxidation resistance, good thermal shock resistance low thermal conductivity, and good microstructure stability [28, 29].

3. TBC problems

Propagation of cracks in TBCs during the service period leads to the coating delamination and failure of TBC coatings on hot components [9, 30]. Spallation of the coatings occurs by the sequential steps of crack initiation, propagation, and coalescence leaving hot-section components unprotected against the high-temperature environment [1, 31]. TC is a high-temperature resistant oxide layer and BC is a connecting layer between TC and the substrate. The TGO layer formed by the diffusion of O_2 through TC and the BC layer oxidation has considerably different TEC with the top coat. Due to the thermal expansion coefficient mismatch, thermal cycling may cause stress in the coating resulting in the crack formation and the coating peeling leaving the substrate unprotected [32, 33]. Understanding the failure mechanisms in the TBC coatings during operation can help us improve the durability of these coatings. Several failure mechanisms have been reported in the literature [34]. Generally, three main mechanisms contribute to TBC failure:

1) Most TBC coatings fail at or near the BC/TGO interface result-

ing from elastic strain energy and stress concentration caused by the thickening of TGO and thermal expansion mismatch [35]. Upon cooling down from service temperature to room temperature, the mismatch of TEC between the substrate, BC, and TGO causes high compressive stresses in the ceramic layer [36]. Because of growth strain, high compressive stresses are applied to the TGO. Consequently, the strain energy is released by TGO distortion in the system, which leads to the creation of the tensile and compressive stress gradient. The key reason for nucleation and propagation of cracks in TGO and thereby, the ultimate spallation of the coating is tensile stress [34, 35, 37].

2) In air plasma-sprayed TBCs, spallation also occurs at the splat boundaries of the ceramic top coat. The spallation resistance of the coatings can be increased by altering the coatings architecture, sintering suppression within the top-coat, and using an innovative plasma-spray process [34, 38].

3) Besides thermal-driven damages, TBC damages might be due to mechanical causes such as mechanical fatigue caused by vibration and FOD [34, 39].

4. TBC modification

4.1. New TBCs

The refractory materials are preferably considered for developing new TBC materials. In this regard, the properties of four ceramic material groups are believed to be more promising for further investigations. These groups are mainly highly-defected zirconium oxide compounds, rare fluorite, and pyrochlore-type earth zirconates, hexa-aluminates, and perovskites. Fluorite and pyrochlore-type earth zirconates have been considered as the most promising choices for the development of new TBC coatings [26, 40].

Predicting the lifetime of TBCs is essential for the improvement of the safety of TBC components [41]. The turbine inlet temperature can be increased to 2000 K in the new generation of engines having high thrust ratios [42]. In these cases, YSZ-based traditional TBCs could not satisfy the requirements of thermal barriers [43]. In recent years, new TBC materials have been widely explored in order to build coatings that can bear higher temperatures than the traditional TBCs [25]. Some new TBCs include: 1) Lanthanum cerium oxide ($\text{La}_2\text{Ce}_2\text{O}_7$) that possess a lower thermal conductivity and a higher TEC than those of YSZ; Lanthanum cerium oxide ($\text{La}_2\text{Ce}_2\text{O}_7$) was proposed as a new material for TBC for its low thermal conductivity and large TEC, which is close to the BCs thermal expansion coefficient. Thermal cycling studies using a $\text{La}_2\text{Ce}_2\text{O}_7$ coating showed that the thermal stability of the $\text{La}_2\text{Ce}_2\text{O}_7$ coating is comparable to the stability of the more traditional YSZ coating. Thus, the lifetime of a single layer of $\text{La}_2\text{Ce}_2\text{O}_7$ is similar to the lifetime of a single layer of YSZ. However, for multi-layer coatings, the $\text{La}_2\text{Ce}_2\text{O}_7$ coatings showed a longer lifetime than the YSZ coatings [44-46].

2) Pyrochlore oxides ($\text{A}_2\text{B}_2\text{O}_7$) are structures comprised of 2 cations with a +3 oxidation state, such as La^{3+} , or Lu^{3+} (indicated by A), 2 cations with a +4 oxidation state, such as Zr^{4+} , Hf^{4+} or Ti^{4+} (indicated by B), and 7 oxygen anions. $\text{Gd}_2\text{Zr}_2\text{O}_7$ and $\text{La}_2\text{Zr}_2\text{O}_7$ are common examples of these pyrochlore oxides. These materials typically have a lower thermal conductivity, a higher melting point, and a relatively higher TEC compared to YSZ, which makes them suitable for TBC [24, 47-49].

3) Neodymium cerate ($\text{Nd}_2\text{Ce}_2\text{O}_7$) has a fluorite type crystal structure, which is stable at high temperatures up to 1600 $^{\circ}\text{C}$ and has a lower thermal conductivity than YSZ [29]. These thermal properties make this material very promising to be used in advanced TBCs [50-53], such as those used in turbine engines [54, 55].

4) Another material proposed as a new TBC is NiCoCrAlY, which is thermally stable at temperatures ranging from 1200 to 1400 $^{\circ}\text{C}$ [56].

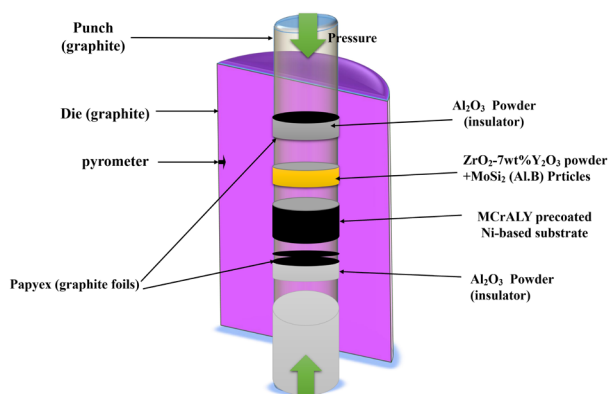


Fig. 1. Self-healing mechanism in TBCs with MoSi_2 particles.

This material is used on Ni-based superalloys to improve its resistance to oxidation. At high temperatures, NiCoCrAlY forms an oxide layer between the BC and the ceramic top coat. The underlying metal is protected from further oxidation by this oxide layer. This oxide layer is also the first place where this TBC fails and thereby, the spallation of TC is promoted [57-59].

4.2. Changing structure

Along with the new gas turbine development, there have been many investigations focusing on the introduction of advanced TBC coatings with higher temperature stability and lower thermal conductivity [60]. As mentioned above, the most popular material for TC in TBCs is 7-8 wt. % YSZ. It shows many advantages; however, it suffers from a few drawbacks and limitations. It is possible for the YSZ coating to undergo an adverse phase transformation upon thermal cycling; also, it may be sintered at elevated temperatures during the service period [34].

The coatings deposited by electron-beam physical vapor deposition (EB-PVD) exhibit improved erosion, strain, and thermal shock resistance due to their columnar microstructure [61]. Three types of EB-PVD thermal barrier coatings exist; two-layered, multilayered, and graded TBCs. TBCs with the two-layered structure is the conventional coatings involving the MCrAlY bond coat and YSZ top coat [56]. It has been recently proposed that graded composites of lanthanum hexa-aluminate and alumina present elevated fracture toughness and ductility [47, 62]. The feasibility of functionally graded thermal barrier coatings by spark plasma sintering was also reported [63, 64]. The stress between TC and BC that is the result of different TEC can be relaxed by graded TBCs. A transition layer with alumina between TC and BC is present in these coatings. It has been indicated that the oxidation and thermal shock resistance of graded TBCs are much higher than those of two-layered TBCs [61]. Multi-layered TBCs are commonly utilized for particular applications [56].

4.3. Nano Structure

New proposed structural, chemical, and technological approaches for developing new TBCs are not able to deal with all failure factors. Indeed, enhancements of some properties cause a reduction in other characteristics of the materials [65]. To obtain a combination of desired properties, the TBC architecture can be changed and multilayer coatings with engineered layers can be applied, according to their requirements [66]. Compared to conventional TBCs, nanostructure thermal barrier coatings have shown enhanced thermal insulation property and thermal cycling lifetime. Nanozones that consist of unmelted particles shows an utterly porous morphology. These nanopores are homogeneously dis-

tributed and besides microcracks, existing voids, and fine grain structure act as phonon scattering centers. Therefore, the thermal conductivity of the coatings is significantly reduced [34]. It has been reported that nanostructured ceramic coatings have excellent mechanical properties, and high TEC [67-74].

Despite the aforementioned properties, the applicability of nanostructured TBCs is skeptical. It is proposed that when nanostructured YSZ coatings are exposed to high-temperature environments, their densification or sintering rates are higher than that of conventional TBCs [73]. As a result of the high sintering rate, coating stiffening (elastic modulus values) and thermal conductivity/ diffusivity increase beyond critical levels; therefore, it might cause premature failure and limit the application of nanostructured TBCs [73].

5. Self-healing composite TBCs

In the 1970s, self-healing ceramic materials were first introduced. Self-healing materials are able to repair cracks in ceramics upon heat treatment. Many polymeric, metallic, and concrete-based materials have been reported as self-healing materials over the past few years [75]. Majority of studies in the field of self-healing ceramics are allocated to the use of SiC for healing, which occurs through the oxidation of SiC particles and the resulting volume expansion into the cracks [75].

Sloof et al. [76] took the first step to fabricate thermal barrier coatings with self-healing capability. They incorporated boron alloyed MoSi_2 (B- MoSi_2) into the YSZ coating and indicated that the addition of these particles did not create extra stresses during thermal cycles [77]. In the absence of oxidative atmospheres, B- MoSi_2 particles remain solid at 1100 °C, and their TEC is close to that of the YSZ matrix. Additionally, the fluidity of SiO_2 at the service temperature increases by alloying the MoSi_2 with B, which facilitates the filling of cracks [78]. When cracks are formed, B- MoSi_2 particles embedded into the thermal barrier coating are exposed to a high oxygen potential atmosphere followed by the formation of amorphous SiO_2 by the oxidation reaction Amorphous silica flows into generated cracks and comes into direct contact with their walls. This amorphous oxide then reacts with zirconia and form solid reaction products, mainly ZrSiO_4 , that adhere to the walls [79]. The mechanism of self-healing in TBCs with MoSi_2 particles is shown in Fig. 1.

6. Coating methods

As mentioned earlier, in an oxygen-containing atmosphere, the material is oxidized rapidly at 1000-1200 °C [80]. Besides, due to the very porous structure of YSZ, oxygen easily penetrates YSZ, and the ceramic layer is not able to accommodate the compressive stresses to a specific extent [76, 81]. The thickness of the protective SiO_2 coating on the bulk MoSi_2 formed above 800 °C reaches usually several μm ; therefore, a significant amount of the material in particles is consumed. Moreover, formed SiO_2 would react with ZrO_2 to form zircon. To prevent these reactions, applying a coating is necessary. Various materials have been studied, such as ZrSiO_4 , $\text{Al}_6\text{Si}_2\text{O}_{13}$, and Al_2O_3 . However, Al_2O_3 is commonly used for coating the self-healing ceramics in TBCs [82]. To enhance their corrosion and oxidation resistance, silicon-containing particles such as MoSi_2 and SiC have been coated by Al_2O_3 [83-85].

Atomic layer deposition (ALD) and sol-gel are the two common methods for alumina coating deposition. The sol-gel method is very versatile; however, the system should be in the form of a sol or at least not fully gelled [86]. Although Al_2O_3 is the most studied material prepared by sol-gel, the majority of investigations are associated with bulk Al_2O_3 , particularly for catalytic applications. For the production of particles from the environment, several studies have been carried out on particle

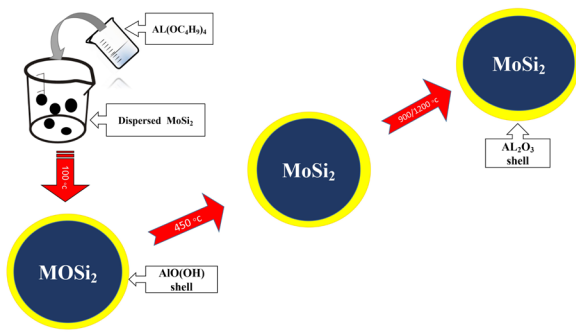


Fig. 2. Coating of MoSi₂ particles by the sol-gel method.

coating by sol-gel, for instance, on SiC [87, 88], magnetic particles [89], and phosphors [90]. As a result of very similar surfaces (a native SiO₂ layer), there have been several useful investigations of alumina coating on SiC by the sol-gel method; however, studies on MoSi₂ sol-gel coating with Al₂O₃ are rare [91]. The sol-gel production of the Al₂O₃ coating on MoSi₂ particles is shown schematically in Fig. 2.

ALD is a technique based on chemical vapor deposition, which is an important method to deposit thin films from gas-phase reactants. In this technique, two self-limiting half-reactions occur for the deposition of solid films on the surface. Controlling the thickness of the film is possible due to these self-limiting reactions [92]. ALD is mainly used for the deposition of thin coatings on relatively flat substrates and wafers. Although there are some challenges regarding deposition on particles, it is possible to use this technique for particles. Particles possess a significantly larger surface area compared to wafers requiring the supply of more reactants. The slow mass transfer occurring in a bed of particles is another problem associated with the particle coating, even in the case of high porosity. This leads to the formation of non-homogeneous coatings [93, 94].

Using a fluidized bed is a solution to these problems. In this process, the particles are suspended in a gas flow by blowing the gas with sufficient velocity through the bed of particles. As a result, both the gas and particles act as fluids enhancing contact and mixing gas and solid enormously [95].

7. Self-healing mechanism in thermal barrier coatings

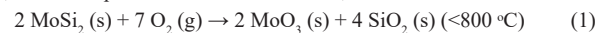
The addition of SiC particles to brittle materials such as mullite (3Al₂O₃·2SiO₂), Al₂O₃, and Si₃N₄ could increase the toughness of these materials and it also could give self-healing capability to these materials [96-98]. When SiC particles interact with cracks at high temperatures, SiC reacts with O₂ resulting in the formation of a silica-based reaction product with volume expansion, which flows into the crack [64]. It is desirable to use self-repair mechanisms at high temperatures to enhance the lifetime of TBC coatings. Using MoSi₂ particles in the YSZ matrix for high-temperature crack-healing has been reported [79, 99]. The healing ability of MoSi₂ particles is due to the release of viscous SiO₂ into the cracks and a subsequent reaction of the formed SiO₂ with the matrix through a solid-state reaction. As a result of the reaction, load-bearing crystalline ZrSiO₄ is formed, providing enhanced adhesion between the fractured surface and the healing agent [100].

The porous structure of YSZ facilitates the diffusion of oxygen, which might lead to the premature oxidation of the particles, even in the absence of cracks interfering in the healing process in TBCs [101]. Therefore, to prohibit the premature initiation of the healing mechanism in TBCs and maintain the particles in a dormant state, it is required to apply an inert shell that is impenetrable against oxygen on the surface of

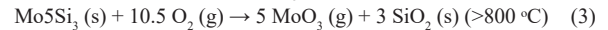
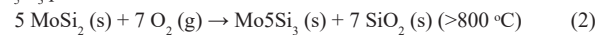
the actual intermetallic particles [102]. The ideal material for the protection of the particles is α-alumina, due to a low permeability for oxygen [103, 104] and good thermal stability [105]. When a crack penetrates the Al₂O₃ shell surrounding active MoSi₂, the healing mechanism is activated [17].

The healing agent incorporated in thermal barrier coatings should have some properties [106] including: 1) it should be in the solid phase at the operating temperature because liquids have a large thermal conductivity and large TEC; 2) it should be turned into a liquid to wet the crack surfaces and fill the crack; 3) the liquid medium should turn into a solid phase by the reaction between the TBC material and the liquid phase through a solid-state chemical reaction. As a result of this process, the crack is healed by a well-bonded crystalline material having low thermal conductivity [107].

The high-temperature oxidation of MoSi₂ has been studied extensively, due to its use as heating elements in many high-temperature furnaces. There are two oxidizable components in the system: molybdenum and silicon, which make the behavior of the system rather complex [108]. The oxidation temperature of MoSi₂ ranges between 400 and 500 °C, and at temperatures lower than 800 °C, reaction starts as follow:



Zhu et al. [76] found that above the melting point of MoO₃ (approximately 800 °C), the formation of Mo₅Si₃ and SiO₂ is more thermodynamically favored than that of MoO₃, according to reaction 3.2. Mo₅Si₃ can be further oxidized in the presence of adequate oxygen (according to reaction 3.3). At this temperature, however, the vapor pressure of MoO₃ becomes significant, causing most of MoO₃ to be removed. Therefore, the formation of the SiO₂ scale starts at 800 °C, protecting the MoSi₂ and Mo₅Si₃ phases.

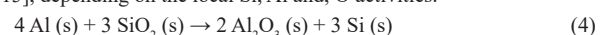


Porosity in the formed scale due to the significant formation of MoO₃ below 800 °C occurs as a phenomenon referred to the oxidation of MoSi₂, which prevents a protective coating formation. If no initial protective coating is formed, oxidation of MoSi₂ continues at high rates, which is a challenge in bulk MoSi₂ applications. However, a protective coating is needed for particles due to the required thickness of the SiO₂ coatings in several μm, which is already mentioned in the theory section [109]. Therefore, the consumption of a significant part of the healing particle to form this coating is necessary.

Attempts have been made to alloy MoSi₂ with boron and aluminum, due to its interesting high-temperature properties and intermetallic nature [110]. Mao found that boron stabilizes the amorphous phase of SiO₂. On the contrary, MoSi₂ tends to form separate phases with molybdenum. However, in the presence of molybdenum, no borides are formed from the reaction of boron and silicon [111].

The results indicate that only boron may form a separate phase with molybdenum, which mainly depends on the applied processing conditions for the production of MoSi₂. As most of the molybdenum borides have a significant high hardness, boron may increase MoSi₂ hardness [112]. In the structure of MoSi₂, Al can easily substitute B, although it stabilizes the metastable MoSi₂ hexagonal phase [113]. As a result, a single phase of MoSi_xAl_y or a two-phase system with both the hexagonal and tetragonal phase of MoSi_xAl_y coexists, depending on the molar ratios of the present elements.

However, the main effect of the presence of aluminum is on its oxidation behavior. In fact, aluminum is preferentially oxidized and reduces SiO₂, according to reaction 3.4. This is due to lower ΔG of Al₂O₃ per mole of oxygen than that of either Mo or Si [114]. As a result, pest oxidation is limited and the Al₂O₃ scale is formed instead of the SiO₂ scale [115], depending on the local Si, Al and, O activities.



The oxidation of components in MoSi₂ and subsequent formation of

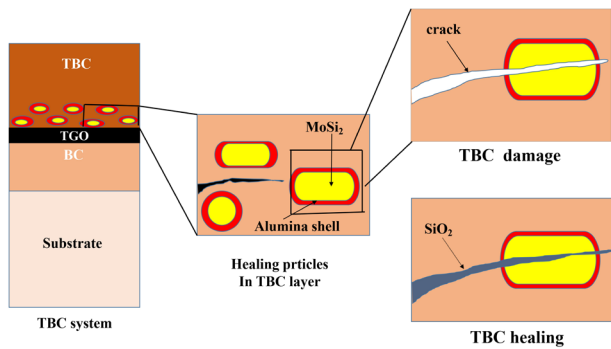


Fig. 3. Preparation of self-healing TBCs using SPS.

ternary oxides lead to significant changes in molar volume and help to fill and close cracks that are desired for the self-healing process. Premature oxidation also results in the accumulation of stress and subsequently coating fracture, as mentioned in the theory section [116].

8. Application of self-healing composites in TBCs

The material chosen as a healing agent for an autonomous self-healing TBC must fulfill criteria as follows: (i) the healing material should have a melting temperature higher than 1000 °C (maximum TBC operating temperature), and its TEC should match the TBC materials, (ii) it should be oxidized and transformed into a liquid to fill cracks and provide direct contact with the surfaces of cracks, (iii) it should be able to wet the crack faces and form a load-bearing material through a solid-state chemical reaction between the TBC material and the liquid [79].

Nozahic et al. [64] embedded encapsulated MoSi₂ (B) particles in the YSZ coating to prepare a self-healing TBC. The composite coating deposition on a Ni-based superalloy was carried out using spark plasma sintering (SPS). The coating process is shown in Fig. 3. According to the results, no cracks resulting from thermally induced stresses appeared in the SPS-sintered TBC. This is the result of the CTE difference of the YSZ matrix and MoSi₂-based particles.

Ouyang et al. [75] showed that the self-healing agent TiC in YSZ/Al₂O₃/TiC composite coatings was able to release TiO₂ to heal the pores and cracks in the coatings. Pre-oxidation treatment of the self-healing coating based on TiC in the furnace at 600°C may form a self-healing layer with low-porosity at the interface between TC and BC, and as a result, it could prevent the formation and growth of the detrimental oxide materials. Hence, the oxidation resistance of the coating was improved by the self-healing process. Nevertheless, according to the thermal cycle test, a thick layer of the TiC-self-healing coating induced the stress above the stress limit in ceramic coatings.

Ouyang et al. [117] used APS to fabricate self-healing TBCs based on YSZ/SiC-self-healing (SAZ) structure. They demonstrated that the self-healing occurred during SAZ coating after oxidation at the temperature of 720°C, which resulted in filling the remaining cracks/pores in the SAZ coating. Moreover, due to the comprised oxidation resisting materials, Al₂O₃ in the SAZ coating and its dense structure, the oxidation resistance of TBCs was improved after coating with SAZ. Consequently, the resistance against spallation in the YSZ/SAZ also increased, which was verified using an ideal mode assessing the TBC stress intensity. They also applied a cyclic high-temperature oxidation test on YSZ and YSZ/SAZ coatings. The results showed that the spallation resistance was increased in the YSZ/SAZ coatings by a toughening effect occurred after high-temperature oxidation.

Nguyen et al. [118] developed a self-healing material based on a composite of Ni and Y₂Ti₂O₇. In order to study the crack-healing behavior, Vickers indenter was employed to intentionally generate the cracks

on the surface of the composite, followed by an annealing process in an oxidizing atmosphere. They suggest that the depositing of NiO into the cracks is the main crack-healing mechanism. In fact, the oxidation of the Ni fillers led to the formation of NiO during annealing. According to XRD and SEM results, the complete healing of the cracks was obtained using 10 vol% Ni filler.

Kulczyk-Malecka et al. [119] established Ytria-partially stabilized zirconia (YPSZ)/MoSi₂ composites to render self-healing ability and enhance the thermal stability of the matrix. In this method, decomposition of MoSi₂ at the elevated temperatures results in the volumetrically expanding product which heals the cracks. Furthermore, they focused on the comparison between the conventional YPSZ and the composites containing MoSi₂ particles in terms of the TEC and the fracture toughness. It was found that the produced MoSi₂ composites have a similar TEC and fracture toughness to the conventional YPSZ, indicating small mismatch stresses induced by elevated temperature. The cracks introduced by indentation have shown such a composite system is able to independently perform the self-healing reaction.

Kurniadi et al. [120] encapsulate MoSi₂B particles in YSZ coatings using a novel approach for in situ shell formation. The healing particles were alloyed with Al. The results showed that the addition of 12 wt% of Al resulted in a homogeneous distribution of Mo(Si_{1-x}Al_x)₂ and the formation of exclusive alumina through selective oxidation of aluminum at 1100°C. In comparison with pure Argon, oxidation in lower O₂ pressure using CO/CO₂ led to a more exclusive alumina formation. But, composite oxidation in the CO/CO₂ exhibited the simultaneous formation of Al oxide, Si oxide, and mixed alumina-silica. This shows that partial reduction of YSZ plays a role in particle oxidizing. However, the shell provided protection for particles was observed in microcapsule stability test in laboratory air and at 1100°C for 100 h. The crack-gap filling experiment demonstrated no significant difference in the healing behavior of in situ encapsulated MoSi₂B and pre-encapsulated when the composites were exposed to the temperature of 1100 °C for 16 h.

Wang et al. [77] developed YSZ-La-Mo-Si coatings (YSZ-LMS) using the plasma spraying method for protecting carbon/carbon (C/C) composites. Raw materials for the production of the coating were YSZ, MoSi₂, and LaB₆. The results showed that the YSZ-LMS coating protected the composites at 1500 °C for 50 h. A denser oxide glass layer of Zr-Y-La-Si-O was formed due to the formation of La₂O₃, Y₂SiO₅, Zr-SiO₄, and SiO₂, which leads to the solid phase volume expansion and a decrease in the SiO₂ volatilization. As a result of the volume expansion at high temperatures, compressive stress is generated within the coating restraining the crack initiation and propagation, leading to the improvement of the oxidation resistance of the coatings. The activation energy of high-temperature oxidation for the coated C/C composites was obtained to be 74.466 kJ mol⁻¹ at 1300-1500 °C.

To prepare TBCs with the self-healing ability, Carnicer et al. [121] used an aqueous suspension of Al₂O₃ and SiCY-TZP. To prevent the SiC oxidation as a self-healing agent, suspension plasma spraying was utilized for the deposition of the coating. The results demonstrated that the SiC particles were oxidized during the PS process and the dispersion of the unmelted particles was observed in a partially melted Al₂O₃/Y-TZP matrix. This reveals the potential self-healing ability of the prepared coating.

Using embedded MoSi₂(B) healing particle oxidation, Derelioglu et al. [79] developed a new method for the fabrication of self-healing TBCs. The oxidative decomposition of the healing agents resulted in the amorphous SiO₂ formation which flowed into created cracks and wetted the crack faces. Thereafter, solid ZrSiO₄ was formed due to the reaction between SiO₂ and ZrO₂ based TBC coating establishing a strong bond between the matrix material and the healing agent causing a complete filling of cracks. Even when cracks are not present in the coating, the decomposition reaction can occur resulting from the oxygen transparen-

cy of the ZrO₂-based matrix. This led to premature decomposition and undesirable pore filling of the coatings.

In general, the self-healing TBC samples made by SPS indicate an enhanced resistance against thermal cycling in the air at the temperature of 1100 °C. The results show that the borosilicate or silicate phase partially fills some cracks and there is a zircon phase that connects both surfaces of the crack. The intended self-healing effect of partially yttria-stabilized zirconia-based TBCs reinforced with MoSi₂(B)-based particles was confirmed by the observations [64, 122, 123].

9. Conclusions and future insights

Thermal barrier coatings have been offered for several applications in the industry to improve the efficiency of advanced gas-turbine engines. SiC, MoSi₂, TiC, etc. have been shown to act as a self-healing agent and are able to heal the cracks/pores in the coatings. The good resistance of self-healing TBC samples against thermal cycling at high temperatures in the air indicates the feasibility and the good performance of these coatings in-service conditions. It has been deduced that the borosilicate or silicate phase partially fills some cracks and there is a zircon phase that connects the surfaces of cracks. In the near future, hopefully, other composite materials will be tested for self-healing applications in TBCs and a further improvement in the physical and thermal properties of the barrier coatings will be investigated.

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Conflict of Interest

All authors declare no conflicts of interest in this paper.

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