Materials and Design 143 (2018) 204–213

Contents lists available at ScienceDirect

Materials and Design

journal homepage: www.elsevier.com/locate/matdes

Self-healing thermal barrier coating systems fabricated by spark plasma sintering

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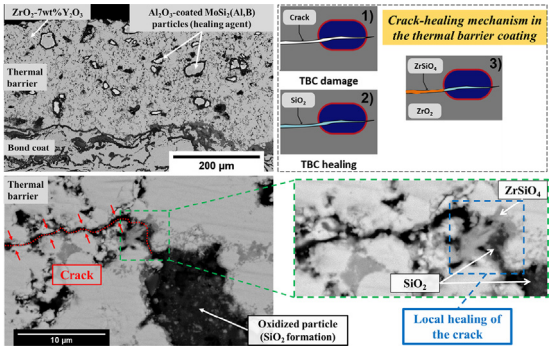
HIGHLIGHTS

- TBC made of YPSZ and encapsulated $MoSi_2$(Al,B) particles was spark plasma sintered on MCrAlY coated Ni-based superalloys.
- TBC self-healing ability was proved with cracks locally filled with silica and a zircon phase that links the crack surfaces
- Al_2O_3 shell protects the $MoSi_2$-based particles against premature oxidation under thermal cycling conditions at 1100 °C in air.
- Detrimental reaction between $MoSi_2$ particles and MCrAlY bond coating may occur but preventive solutions have been found.
- After high temperature exposure the initial protective Al_2O_3-shell around the particles is converted to more complex shell.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 7 November 2017
Received in revised form 31 January 2018
Accepted 1 February 2018
Available online 02 February 2018

Keywords:
Spark plasma sintering
Self-healing thermal barrier coatings
High-temperature cyclic oxidation
Intermetallic particles
Multi-layer materials
Ceramic matrix composites

ABSTRACT

The present paper focuses on the Spark Plasma Sintering (SPS) manufacturing of a new type of self-healing thermal barrier coating (TBC) and a study of its thermal cycling behaviour. The ceramic coating consists on an Yttria Partially Stabilized Zirconia (YPSZ) matrix into which healing agents made of $MoSi_2$-Al_2O_3 core-shell particles are dispersed prior to sintering. The protocol used to sinter self-healing TBCs on MCrAlY (M: Ni or NiCo) pre-coated Ni-based superalloys is described and the reaction between the particles and the MCrAlY bond coating as well as the preventive solutions are determined. Thermal cycling experiments are performed on this complete multi-layer system to study the crack healing behaviour. Post-mortem observations highlighted local healing of cracks due to the formation of silica and the subsequent conversion to zircon at the rims of the cracks.

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https://doi.org/10.1016/j.matdes.2018.02.001
0264-1275/

1. Introduction

Thermal Barrier Coatings (TBCs) made of Yttria Partially Stabilized Zirconia (YPSZ), deposited by Electron Beam Physical Vapour Deposition (EBPVD) or plasma-spraying techniques onto metallic superalloy turbine blades, are widely used to increase the durability of internally cooled hot-section metal components in advanced gas-turbines for aircrafts and power generation [1–4]. The role of such a ceramic coating is to act as a thermal insulator by decreasing the temperature of the underlying Ni-based superalloy blade material. The TBC is deposited on a bond coating that promotes and maintains the adhesion of the TBC and provides oxidation resistance to the system due to the formation of a Thermally Grown Oxide (TGO) consisting mainly of α-Al_2O_3. The failure of plasma sprayed TBCs under thermal cycling is a complex phenomenon that is mainly due to the stresses induced by the mismatch between thermal expansion coefficients of the different components of the TBC system and/or by the growth of the α-Al_2O_3 TGO as a result of the oxidation of the underlying bond coating. In case of plasma sprayed TBCs deposited on bond coatings with high surface roughness, failure was shown to be controlled by a sequence of initiation, propagation and coalescence of cracks, just above the TGO, that finally leads to the spallation of the TBC and expose the hot-section metal components to the high-temperature environment [5]. Hence, a TBC with higher toughness value or that exhibits self-repair ability at high temperature in an oxidizing environment is highly desirable to extend its lifetime and enhance its reliability.

Recently, embedment of SiC particles was shown to be a promising way to increase the toughness of brittle materials such as silicon nitride (Si_3N_4), alumina (Al_2O_3) or mullite ($3Al_2O_3 \cdot 2SiO_2$) but also to provide a self-healing ability to these materials [6–8]. When cracks interact with SiC particles, these latter react at high temperature with oxygen leading to the formation of an expanding silica based reaction product that will flow into the crack. Another example is given by the self-healing of nano Ni particles reinforced alumina composites [9]. To create an autonomously self-healing TBC, the material chosen as healing agent must satisfy some criteria: (i) it should have a high melting point, higher than the maximum operating temperature for TBCs (1000 °C or above) and a thermal expansion coefficient that matches reasonably with the one of the TBC material, (ii) it should be able to oxidize and form a liquid which fills the crack and establishes direct contact with the TBC crack surfaces, (iii) the wetting of the crack surfaces should be followed by a solid state chemical reaction between the liquid and the TBC material leading to the formation of a load bearing material [10]. Based on a literature study of potential healing agents, boron doped $MoSi_2$ particles covered with an alumina shell have been proposed as healing agents for TBC [10,11]. When intercepted by cracks, the healing agent will oxidize leading to the formation of a low viscosity and amorphous borosilicate phase that will flow into the cracks. Subsequently, this phase will react with the surrounding ZrO_2-based TBC to form a load bearing and crystalline $ZrSiO_4$ phase that closes the crack gap and exhibits a strong adhesion with the fracture surfaces. However, industrial ZrO_2-based TBCs are porous (≈20 vol%) and known as being an excellent oxygen ion conductor at elevated temperatures [12,13], thus encapsulation of the $MoSi_2$ particles by a high temperature stable and oxygen impenetrable shell is highly desirable to prevent the spontaneous oxidation of these particles. It has been proposed to deposit a shell of alumina around the $MoSi_2$ based particles via a precipitation process [14] or via a sol-gel process [15].

Recently, it was demonstrated the feasibility to deposit mixed layers of $MoSi_2$ based particles and YPSZ by Atmospheric Plasma Spraying (APS) technique leading for the optimal process conditions to a significant life time extension [16]. While APS is a suitable technique for coating complex shaped components, the Spark Plasma Sintering (SPS), has attracted great interest for the manufacturing of multi-layer samples for scientific studies on material degradation under relevant thermal cycles [17]. Already the feasibility of one-step manufacturing of YPSZ nanocrystalline TBC and Pt-rich γ-Ni/γ'-Ni_3Al bond coating sintered on AM1 substrates was demonstrated [18]. Thereafter, the same protocol was used to fabricate complete TBC systems with a NiCrAlY bond coating [19]. SPS was also used to simultaneously produce YPSZ/MCrAlY multi-layer coatings on Hastelloy X substrates with a rough YPSZ/MCrAlY interface [20]. Recently, the feasibility of functionally graded TBC by SPS was reported [21].

The aim of the present study is to evaluate the self-healing of model TBC systems. For this purpose, SPS technique was first used to sinter mixtures of YPSZ powder and $MoSi_2$ co-doped with B and Al based particles on a NiCoCrAlY coated Ni-based superalloy. Prior to sintering, these particles were coated with an α-Al_2O_3 protective shell deposited via a sol-gel route. The particles were alloyed with Al to allow the formation of Al_2O_3, by selective oxidation of Al, in case of shell breakage during the SPS sintering. The nature and composition of the protective shell, before and after SPS sintering and short annealing treatment, were characterized by means of X-ray Diffractometry (XRD), Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM). Thermal cycling tests were performed in order to study the behaviour and the self-healing response of the model self-healing TBC systems. The advantage of the SPS method over the industrially more relevant APS method is that the protective alumina coating on the $MoSi_2$ healing particles remains intact during the synthesis process. The integrity of the particle coating is crucial to ensure the absence of non-crack related decomposition of the particle [22,23].

To the best of authors' knowledge, this paper presents for the first time a complete study showing: i) the manufacturing and investigation of the thermal cycling behaviour and ii) the crack healing of self-healing TBC made by SPS.

2. Experimental procedure

2.1. Synthesis of the encapsulated $MoSi_2$(B) based healing particles

Coating of the $MoSi_2$ based intermetallic particles was performed using a modified Yoldas method [15], based on the polycondensation and formation of boehmite sol. The $MoSi_2$ based particles (with 99.5% purity) were alloyed with 2 wt% B and 6 wt% Al and delivered by ChemPur GmbH, Karlsruhe, Germany. The starting materials used for this sol-gel synthesis were: aluminium tri-sec-butoxide ($Al(OCH(CH_3)C_2H_5)_3$, 97% purity, Sigma Aldrich Co. LLC, St. Louis, MO) as a precursor, nitric acid (HNO_3 solution, Sigma-Aldrich Co. LLC, St. Louis, MO), isopropanol (99.8%, Sigma-Aldrich Co., LLC, St. Louis, MO), ethanol (99.8%, Sigma-Aldrich Co., LLC, St. Louis, MO), and deionized water (18.2 MΩ·cm at 25 °C). All these chemicals were used without any further purification.

Prior to the encapsulation process, the $MoSi_2$ based powder was wind sifted to remove the fine fraction. Wind sifting was performed using an Alpine 100 MRZ laboratory zig-zag classifier (Alpine Multi-Plex 100 MRZ, Hosokawa Micron Powder System, Summit, New Jersey, USA). Airflow was fixed at 15 m^3/h and the classifier rotational speed was kept at 5000 rpm. The particle size distribution was measured with laser diffraction using a Malvern Mastersizer X (Malvern Instruments Ltd., Worcestershire, UK). Prior to this measurement, the particles were dispersed in deionized water for 20 min by applying ultrasonic vibration. The wind sifting resulted in a powder having an average particle diameter of 33 μm and 16 and 60 μm at 10 and 90% cut-off, respectively.

The coating process of the $MoSi_2$ based powder was done by first heating a mixture of 600 mL of deionized water and 15 mL of 1.0 M HNO_3 to 80 °C to obtain a molar ratio of 2:150 of $Al(OC_4H_9)_3$ to H_2O. First, 10 g of $MoSi_2$ powder was added to the solution mixture of deionized water and HNO_3 and 200 sccm nitrogen (5 N purity) was purged through the suspension to improve the dispensability of the particles in the solution. When the temperature was stable at 80 °C again, 10 g of $Al(OC_4H_9)_3$ was added with approximately 25 mL of ethanol to