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Original Article

Aqueous suspension processing of multicomponent submicronic Y-TZP/Al_2O_3/SiC particles for suspension plasma spraying

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ABSTRACT

In order to obtain thermal barrier coatings by Suspension Plasma Spraying (SPS) process with potential new self-healing ability multicomponent submicronic Y-TZP/Al_2O_3/SiC suspensions were prepared. For this purpose, concentrated aqueous suspensions of individual components, as well as the multicomponent mixture were studied and characterised, in terms of colloidal stability and rheological behaviour to determine the best conditions for processing and preparation of the coatings. In the study, different dispersant contents and sonication times were tested. Subsequently, low concentrated suspensions were prepared to obtain preliminary thermal barrier coatings with the optimised feedstock. Thus, ceramic coatings were deposited by SPS and then characterised in order to assess the microstructure and phase distribution, in particular, the degree of preservation of the sealing agent, SiC, in the final coating as a previous indicator of its self-healing ability.

1. Introduction

In the last decades, different researchers have developed and studied ceramic coatings with the double purpose of improving the protection and thermal insulation and reducing the surface temperature of the metal components of gas turbine engines, such as rotators, blades... to operate at inlet higher temperatures, increasing the efficiency of the process and the lifetime of these components [1–4]. These materials are called thermal barrier coatings (referred to as TBCs). TBCs are composed of refractory ceramic oxides, including alumina, titania, magnesia and their mixtures; however, yttria-doped tetragonal polycrystalline zirconia (Y-TZP) represents the state-of-the-art material as a consequence of its excellent properties at high temperatures compared to metals, such as low thermal conductivity, low probability of phase transformations at operating temperature, chemical inertness and high melting point, superior to metals or superalloys, among others [5]. Although there are different coating techniques to produce TBCs, atmospheric plasma spraying (APS) is widely used due to its economic and technical feasibility at an industrial scale.

Nowadays, turbines need to operate at higher gas temperatures to improve the engine efficiency; nevertheless, these new conditions produce an additional thermal stress and increase the probability of mechanical failure, which causes the appearance and growth of cracks and laminations [2,6]. In the literature, several researchers have proposed to develop a new generation of TBCs with self-healing ability, which will withstand the new working conditions of the turbines and prolong their lifetime [6–11]. SiC is reported as the most efficient ceramic self-healing material, although other metals and alloys are being investigated [6,9,12]. However, self-healing functionality in a TBC has barely started to be developed and, one of the specific challenges is that silicon carbide cannot be easily deposited by plasma spray because it oxidises in the torch before melting [12,13]. The sealing ability (such as in silicon carbide) consists in full filling the cracks originated in the coating, which are produced by the fatigue work of extreme temperatures and aggressive hot gases, during the service time. The self-healing process develops by growing the volume of self-healing particles when the oxygen in the air along with the high environmental temperature goes through the crack, giving rise to the oxidation of self-healing particles located on the edge of the crack [6,8]. Nevertheless, the effectivity of sealing ability is based on the amount and size of non-oxidised particles of the self-healing material found in the coating.

Literature reports different strategies aiming to preserve the nature of the sealing agent in the final coating obtained by a thermal spray process. Thus, one strategy employs eutectic mixtures whose melting point is lower than the oxidation temperature of SiC [14,15]. Another sealing mechanism resides in encapsulating the sealing agent (core-shell) by metal compounds, to produce a selective oxidation of the metal elements which prevents the sealing agent from further oxidation [10,16]. Other strategies focus on the development of reducing atmospheres surrounding the sealing particle during the thermal spray process [17].

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Table 1
Characteristics of commercial powders employed in this research.

Characteristics	Y-TZP	Al_2O_3	SiC
Particle size / D_{10} (μm)	0.1	0.3	0.2
Particle size / D_{50} (μm)	0.4	0.5	0.6
Particle size / D_{90} (μm)	1.4	2.0	1.4
Specific surface area (m^2)	6.8	8.0	15.0
Density (g/cm^3)	6.05	3.97	3.21

Table 2
The composition of bond coat "Amdry 997".

Element	Ni	Al	Co	Cr	Ta	Y
Composition (in wt%)	43.9	8.5	23	20	4	0.6

Table 3
Plasma spray conditions employed for bond and top coats deposition.

Parameters	Bond coat (APS)	SAZ coat (SPS)
Ar flow rate (slpm[a])	65	37
H_2 flow rate (slpm[a])	8	8
Intensity (A)	650	700
Stand-off distance (m)·10^3	145	40
Surface speed ($m\,s^{-1}$)	1	1.25
Number of passages	1	5
Pre-heating (K)	423	573
Injector diameter (m)·10^4	15	1.50
Powder feed rate ($kg\,s^{-1}$)	0.04	–
Specimen holder velocity ($m\,s^{-1}$)	–	0.72
SAZ solid content (wt%)	–	35
Suspension feed rate ($m^3\,s^{-1}$). 10^7	–	4.86

[a] Slpm: standard litre per minute.

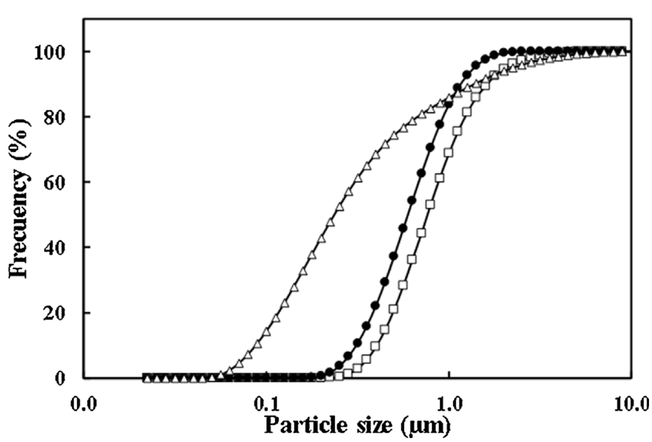

Fig. 1. Particle size distribution of the different powders. □: Al_2O_3, ●: SiC, △: Y-TZP.

More recently, some papers have reported the use of the Suspension Plasma Spray (SPS) technique to obtain coatings containing SiC particles [18]. SPS is an emerging thermal spray process in which the powder feedstock is replaced by a suspension feedstock. An important benefit of SPS process is the possibility of spraying very fine, poor flowability powders [3,19–23]. Moreover, the SPS technique would be very favourable to avoid undesirable oxidation of the sealing material because a lot of the plasma energy is destined to solvent (water) evaporation. However, limiting energy during plasma deposition can compromise coating microstructure and adherence since the particles may not melt as much as required [3,19,21]. For this reason, the optimisation of solid content and stability of SPS feedstock containing SiC particles is necessary to obtain coatings in which the amount of SiC particles can be maximised.

This paper reports the first part of an ambitious research based on a mixed strategy which has been chosen to avoid the oxidation of SiC (self-healing agent). On the one hand, SPS technique will be used for the thermal deposition avoiding the contact of SiC particles with an excessively energetic plasma plume. On the other hand, a third component (Al_2O_3) will be added to the Y-TZP matrix in order to enable the formation of a eutectic phase which can favour the protection of SiC particles against oxidation during plasma deposition. Therefore, the great challenge of this first part of the research addresses the preparation and stabilisation of multicomponent aqueous suspensions of submicronic particles of Y-TZP/Al_2O_3/SiC (referred as SAZ). The literature on the preparation and stabilisation of each of these oxides or their combinations is abundant [22,24–27]. There is also previous research on the preparation of highly concentrated, aqueous suspensions of SiC submicronic particles [28,29]. But the combination of the three ingredients as feedstock for SPS deposition has not been reported to the best of our knowledge. The main purpose is to demonstrate the feasibility of using this formulation to prepare concentrated suspensions to produce homogeneous, self-healing coatings by SPS. For so doing, the stability and rheological behaviour of this 3-component SPS feedstock is firstly studied. Later, a stable, selected feedstock is deposited by SPS and the corresponding coating is characterised in terms of microstructure and crystalline phase distribution. In conclusion, this first research can be useful to establish the basis for obtaining coatings from aqueous and very concentrated suspensions which can result in an enhancement of industrial scalability (lower energy consumption and higher deposition rate) of this new class of coatings.

2. Experimental

2.1. Suspension preparation and characterisation

As starting raw materials, the following commercially available powders were used: α-alumina (CT3000SG, Almatis, Germany), with an average particle size of 0.5 μm and a specific surface area of ~8 m^2/g; tetragonal zirconia polycrystals doped with 3 mol% Y_2O_3 (TZ-3YS, Tosoh, Japan), with an average particle size of 0.4 μm and a surface area of 6.8 m^2/g; and α-SiC (UF-15, Hermann C. Starck, Germany), with an average particle size of 0.6 μm and surface area of 15 m^2/g. The three powders are labelled as A, Z, and S in the following sections. The two oxides were mixed together in such concentrations as to produce after sintering the eutectic composition, i.e. with a relative volume concentration of zirconia/alumina of 50.8/49.2 (that means a weight ratio of 59.6/40.4). The main characteristics of these three powders are shown in Table 1.

Particle size distributions were measured using the laser diffraction technique (LD; Mastersizer S, Malvern, UK) and the morphology of the as-received powders was observed by field-emission gun environmental scanning electron microscopy (FEG-ESEM; Quanta 200 FEG, FEI Company, USA). The crystalline phases were identified by X-ray diffraction (XRD; Advance diffractometer, Bruker Theta-Theta, Germany). In the case of zirconia, the Garvie's approach was used to calculate the relative ratio of tetragonal phase (density = 6.07 g/cm^3, ASTM 83-113) and monoclinic phase (density = 5.82 g/cm^3, ASTM 37-1484), which was found to be 68/32.

Zeta potential measurements were performed using the laser Doppler principle combined with non-invasive back-scattering (Zetasizer Nano-ZS, Malvern, UK). Zeta potentials were firstly measured as a function of pH and secondly as a function of polyelectrolytes. According to previous results concerning the stabilisation of aqueous suspensions of the three types of powders used herein an ammonium salt of polyacrylic acid (PAA; Duramax TM D-3005, Rohm & Haas, USA, with 35 wt% active matter) was selected as a deflocculant for both oxides [24,30,31] and a synthetic polyelectrolyte (PKV, Produkt KV5088, Zschimmer-Schwarz, Germany) with unknown composition (but that it is thought to be of polycarboxylic nature), which has