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Original Article

Protecting the $MoSi_2$ healing particles for thermal barrier coatings using a sol-gel produced Al_2O_3 coating

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ABSTRACT

A successful sol-gel process to encapsulate molybdenum di-silicide $MoSi_2$ particles with a closed and thermally stable Al_2O_3 layer using aluminium tri-sec-butoxide as a precursor is presented. The processing conditions such as precursor selection and temperature were optimized through analysing the interaction of the $MoSi_2$ particles with the sol. The application of the sol-gel based coating was followed by calcining the coated particles at temperatures between 900 and 1200 °C in Ar. The shell composition and the mechanical stability of the microcapsule were analysed by means of X-ray diffraction, scanning electron microscopy and thermogravimetric analysis. Upon calcining at 1200 °C in Ar, the $MoSi_2$ core remains intact as it is, covered by an α - alumina shell with a thickness of about 0.6 μm. The stability tests proved that the encapsulate particles are about five times more oxidation resistant than the uncoated $MoSi_2$ particles.

1. Introduction

Molybdenum di-silicide not only is an attractive monolithic high temperature material because of its excellent refractory and mechanical properties [1–6], but is also as attractive candidate for repairing crack damage in brittle ceramic coatings such as in thermal barrier coatings (TBCs) [7,8]. TBCs are often applied to metallic surfaces of combustion chambers, blades and vanes in gas turbines and jet engines to protect them from corrosion and oxidation at elevated temperatures [9,10]. Yttria stabilized zirconia (YSZ, ZrO_2 containing 4–5 mol% Y_2O_3) is the most employed TBC material due to its excellent chemical and thermomechanical properties, such as low thermal conductivity at elevated temperatures ($2.3\ W\ m^{-1}\ K^{-1}$), low density ($6.4\ g/cm^3$), high melting point (2700 °C), high toughness ($7.7\ MPa\ m^{0.5}$) and good corrosion resistance [9–11]. However, the durability of the TBCs is negatively affected by thermal cycling due to the starts and the stops of the engine resulting in temperatures varying over more than 1000 °C. High compressive stresses develop in the coating during cooling as a result of the mismatch between the coefficients of thermal expansion of the metal substrate ($14{\cdot}10^{-6}\ {}^\circ C^{-1}$) and that of the YSZ TBC ($10–11{\cdot}10^{-6}\ {}^\circ C^{-1}$) [11,12]. As a consequence, microcracks form within the ceramic coating. They propagate and coalesce during subsequent thermal cycling leading to coating debonding and ultimately engine failure [11,13–15].

As YSZ TBCs are thermodynamically stable and hence do not poses autonomous crack-healing capabilities, a self-repair mechanism active at the high temperature encountered in a turbine engine is highly desirable to prolong the lifetime of these coatings. The concept of autonomous high temperature crack-healing based on the presence of $MoSi_2$ sacrificial particles embedded in a YSZ matrix has been presented elsewhere [7,16]. It is based on the release of viscous silica (SiO_2), that fills the crack running through the matrix and re-establish the adherence in the ceramic coating. Subsequently, the formed SiO_2 reacts with the matrix via a solid-state reaction, resulting in a load bearing crystalline zircon phase ($ZrSiO_4$), which gives a better adhesion between the healing agent and the fractured surface. However, in porous yttria partially stabilized zirconia based TBC matrix the transport of oxygen ions is likely to prevail [17]. This will lead to the premature oxidation of $MoSi_2$ particles and, consequently, offsetting the healing reaction even in the absence of a crack. Thus, the presence of an inert and oxygen impenetrable shell around the actual intermetallic particles is critical to prevent the premature triggering of the healing mechanism and to keep the particles in a dormant state. α-Alumina seems an ideal candidate to protect the $MoSi_2$ particles from premature oxidation since

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it has a good thermal stability [18] and a low permeability for oxygen [19,20]. Hence, the healing mechanism will become active only when a crack penetrates the alumina shell protecting the active $MoSi_2$.

Several soft-chemical routes have been proposed to deposit alumina shell such as: precipitation [21,22], hybridization [23] and sol-gel [24]. Among them, the sol-gel method is the most economical way to develop coatings of sizeable thickness and to generate solids of high purity [25]. So far, alumina coatings have been applied successfully on silicon containing particles such as silicon carbide (SiC) grit and $MoSi_2$ to improve their oxidation and corrosion resistance [21,26,27]. However, in the case of $MoSi_2$-Al_2O_3 core-shell system in which the protective α alumina layer was produced by a precipitation reaction, a rather thin layer of alumina – silica mixed oxides of only 0.15 μm was obtained after annealing at 1000 °C [21].

The current paper aims to create a closed and continuous thicker shell of α alumina around $MoSi_2$ intermetallic particles through a simple sol-gel approach. Experimental results proved that by tuning the processing conditions (i.e. type of precursor and calcining temperature), a robust and stable shell of α alumina with a typical thickness of 0.6 μm could be obtained.

2. Experimental details

2.1. Materials

The chemicals for the sol-gel coating of $MoSi_2$ particles were aluminium oxalate tetrahydrate ($Al_2(C_2O_4)_3 \cdot 4H_2O$, 99% purity, Alfa Aesar, Karlsruhe, Germany), aluminium tri-isopropoxide ($Al(OC_3H_7)_3$, 98% purity, Sigma-Aldrich Co., St. Louis, Missouri, US), aluminium-tri-sec-butoxide ($Al(OC_4H_9)_3$, 97% purity, Sigma Aldrich Co., St. Louis, Missouri, US), nitric acid (HNO_3 99% purity, 65 wt% solution as a source, Sigma-Aldrich Co. St. Louis, Missouri, US), isopropanol (C_3H_8O, 99.8%, Sigma-Aldrich Co., St. Louis, Missouri, US), ethanol (C_2H_6O, 99.8%, Sigma-Aldrich Co., St. Louis, Missouri, US), molybdenum disilicide ($MoSi_2$, 99.5% purity, particle size of 29 μm; ChemPur GmbH, Karlsruhe, Germany) and deionized water (18.2 MΩ cm at 25 °C). All chemicals were used without any further purification.

2.2. $MoSi_2$-Al_2O_3 core-shell synthesis

To produce sol-gel encapsulated $MoSi_2$ particles, a modified Yoldas method [28,29] was employed. This method is based on a polycondensation reaction leading to the formation of a boehmite sol, which upon further annealing transforms into a dense and defect free α-alumina layer.

Prior to the encapsulation process, the $MoSi_2$ powder was wind sifted to remove the fine fraction and the impurities. 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 wind sifting resulted in a powder having an average particle diameter of 30 μm with a D_{10} and D_{90} value of 10 and 60 μm, respectively, as will be described further on.

Three precursors were selected and tested to obtain more detailed information on the reaction of $MoSi_2$ particles with the sol: aluminium oxalate ($Al_2(C_2O_4)_3$), aluminium tri-isopropoxide ($Al(OC_3H_7)_3$) and aluminium tri-sec-butoxide ($Al(OC_4H_9)_3$), respectively. The experiments were performed in a reactor consisting of a glass beaker (1000 mL) positioned on a magnetic stirring hot plate (IKA RET control-visc, IKA – Werke GmbH & Co, Staufen, Germany) with thermocouple to control the temperature of the sol and a magnetic stir bar with controlled speed. Nitrogen gas (5 N purity) was supplied to the reactor by a custom-made ring-shaped bubbler with a flow rate of 200 sccm, to ensure a better dispersion of the healing particles in the sol suspension by reducing the rate of oxidation.

First, aluminium oxalate tetrahydrated was dissolved in 20 mL of 1.0 M HNO_3 solution and 20 mL deionized water (Mili-Q ultrapure water type 1; 18.2 MΩ cm at 25 °C), to obtain a molar ratio of 1:110 of $Al_2(C_2O_4)_3 \cdot 4H_2O$ to H_2O. Then, the suspension was heated to 60 °C and stirred for 30 min at 800 rpm to dissolve the aluminium oxalate. Next, $MoSi_2$ particles (10 g) with an average size of 30 μm were added to the solution and the temperature was increased to 80 °C to initiate the gelation of the aluminium oxalate around the particles. The as-formed mixture was kept at this temperature for 90 min while continuing stirring.

In the case of aluminium tri-isopropoxide, the precursor was dissolved in 200 mL isopropanol and 50 mL ethanol with a molar ratio of 1–70 of $Al(OC_3H_7)_3$ to C_3H_8O. 10g of $MoSi_2$ powder was dispersed in 72 mL deionized water and 72 mL of isopropanol by ultrasonication, prior to the sol-gel process. After, the dispersed powder was added to the dissolved $Al(OC_3H_7)_3$ solution, together with 3mL of 1.0 M HNO_3 solution. The mixture was heated to 90 °C and left to gelate for 30 min.

The sample based on aluminium tri-sec-butoxide was prepared by 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 1 : 150 of $Al(OC_4H_9)_3$ to H_2O. First, 10 g of $MoSi_2$ powder was added in the solution mixture of deionized water and HNO_3 and 200 sccm nitrogen (5N 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 approx. 25 mL of ethanol to transfer the viscous liquid. After, the system was left to gelate for 60 min. Next, the nitrogen flow and stirring were stopped and the solution was left at 80 °C until the liquid fraction was evaporated.

All the three resulting gels were filtered using a glass - vacuum filtration system (Sartorius Stedium, Biotech, Goettingen, Germany) with a hydrophilic propylene membrane with a pore size of 0.45 μm. To remove the unreacted precursors, the samples were washed with warm deionized water (about 60 °C). Finally, the coated particles were dried in an autoclave (WTC binder, TAMSON, Tuttlingen, Germany) at 110 °C overnight and gently ground to break up the agglomerates.

To improve the structural integrity of the coating and obtain a thicker shell, the molar ratio of the selected precursor (aluminium tri-sec-butoxide) was increased from 1:150 to 2:150 of $Al(OC_4H_9)_3$ to H_2O, following the same coating procedure, as described above; see Fig. 1. Next, the coated particles were subjected to a two-step annealing treatment. First, a calcination treatment was applied at 450 °C for 30 min in laboratory air and 15 h in Ar, to slowly release the molecular water, followed by a post-treatment between 900 and 1200 °C for 1 h in Ar, to consolidate and to promote the formation of α-Al_2O_3 shell.

2.3. Characterisation

The particles size distribution of the wind sifted $MoSi_2$ particles was determined with a laser diffraction instrument (Malvern Mastersizer X, Malvern Instruments Ltd, Worcestershire, UK). Prior to the measurement, the particles were dispersed in water for 20 min by applying ultrasonic vibration.

The morphology of the sol-gel $MoSi_2$ coated particles was investigated by scanning electron microscopy (SEM) using a JSM 6500F (JEOL, Tokyo, Japan) operated with an electron beam energy of 15 keV and a beam current of approx. 600 pA. Prior to the SEM observation, the particles were coated with a thin carbon film to prevent charging.

The shell and core composition was analysed with X-ray diffractometry (XRD). A Bruker D500 diffractometer equipped with a Huber incident-beam monochromator and a Braun PSD detector operated with Cu Kα1 (1.54060 pm wavelength) was used to record the diffractograms in the 2θ range of 15–70° with a step size of 0.0387° 2θ.

The shell integrity was investigated by exposure of the coated particles to a quasi-stagnant oxidizing environment at high temperatures using a thermogravimetric analysis (TGA). For this purpose, a symmetrical thermogravimetric analyser (Setaram TAG 16/18, France) was

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