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The effect of parametric variation on the mullite content of plasma sprayed zircon-alumina powder mixture

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ABSTRACT

Mullite was obtained by plasma spraying a mechanical mixture of alumina and zircon sand mixed in 3:2 M ratio. The following parameters were varied during coating deposition: arc current, secondary gas, i.e., hydrogen flow rate, stand-off distance (SOD) and angle of powder injection. These parameters were chosen since they significantly affect powder heating and consequently, mullite formation. Laser surface treatment was undertaken on a couple of selected coatings. The amount of mullite formed in the coating was estimated using Rietveld analysis of the X-ray diffraction patterns of the as-spayed and laser treated coatings. The effects of these parameters on coating hardness and porosity were also taken into account. Finally the mullite yield was correlated with coating porosity and hardness.

1. Introduction

The importance of mullite as a highly attractive engineering material for numerous industrial applications is well documented. Schneider et al. [1] reported that mullite possesses a number of technologically important properties like high thermal stability, low thermal expansion coefficient and thermal conductivity, high creep resistance and corrosion stability coupled with suitable strength and fracture toughness. Some of the applications of mullite are in panels for re-entry space vehicle, fused mullite refractory bricks, sintered mullite-based conveyor belt, substrate for catalytic converters, electronic devices and other advanced applications like optically translucent ceramics for high temperature furnace windows.

It is possible to produce mullite by various processing techniques. Mazdiyasni and Brown [2] prepared mullite by thermal decomposition of kaolinite at ~1200 °C. Ismail et al. [3] used sol-gel method to produce mullite by mixing boehmite and silica as ingredients. Khor and Li [4] obtained mullite by reaction sintering of zircon and alumina powder mixture.

Li and Khor [5] carried out an extensive study to produce mullite by plasma spraying a zircon and alumina powder mixture. At high temperature (above 1600 °C) zircon releases zirconia and silica. Alumina present in the feedstock reacts with the released silica to form mullite. Mullite has been detected in the X-ray diffraction of the coating produced. A similar observation has been made by Hazra et al. [6].

Li and Khor [5] also studied the effect of plasma arc current and secondary (helium) gas pressure on spheroidization of zircon and alumina powder mixture. It was also observed from the X-ray diffraction (XRD) of the coatings that the relative peak intensity ratio of zircon decreased with an increase in current, indicating a higher degree of zircon dissociation. This was accompanied by a corresponding increase in the peak intensity of mullite in the coating. This was expected since, at a higher arc current, i.e., at higher temperature, more zircon dissociated and hence, more silica was released. This silica combined with alumina to produce more mullite. An increase in secondary gas pressure from 0 to 275 kPa retarded the dissociation reaction resulting in a decrease in the mullite concentration in the yield. An increase in the secondary gas pressure in the plasma in this case was said to have produced a higher gas-cum-particle velocity, thus resulting in a reduced time of flight of the particles. Too short a residence time of the particles in the arc restricted particle heating and this in turn limited the dissociation reaction.

In a sequel work, Li and Khor [7] studied the microstructure of the coating prepared under different energy level and SODs. They found that at a very high arc power (100 kW) the coating showed a sponge like morphology. This was attributed to the evaporation of a fraction of particles in the high energy plasma before reaching the substrate. The other observation was that with an increase in the SOD (from 60 mm) melting condition became favorable, while the arc power was kept constant. But beyond a critical SOD (120 mm) the particles froze during flight and coating became porous.

Hazra et al. [6] discussed the microstructural characteristics of as-sprayed and laser treated zircon alumina powder mixture in a previously published report. The as-sprayed coating had a lamellar structure

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common to the thermally sprayed coatings. Some pores were also present. A focused ion beam cross section of the as sprayed splats showed good intersplat wetting in the as-sprayed coating. Laser melting was performed to remelt the upper part of the top coat. The remelted portion of the coating had a homogenous microstructure along with very few pores. The hardness and porosity of the as-sprayed and laser remelted layers were 1091 ± 45 HV & 5.8 ± 0.49% and 1363 ± 47 & 1.5 ± 0.54% , respectively.

Das et al. [8] produced a composite coating by plasma spraying a mixture of natural zircon and commercially available alumina powder. The mixing was carried out in a planetary ball mill in dry condition for 25 min. The purpose of this operation is mixing and hence, no grinding media was used. The X-ray diffraction of this coating had shown mullite peaks indicating presence of mullite in the coating. This mullite formed owing to partial conversation of the starting materials. The authors argued that a shorter flight time was not sufficient for the reaction to complete and suggested that variation in parametric condition might improve the mullite content in the coating. However, the authors did not report the corresponding parameters or the yield of mullite content in the coating.

Salimijazi et al. [9] plasma sprayed a commercially available mullite powder (−35 + 5 μm, Saint-Gobain, MA, USA) and mixture of alumina & silica (in 3:2 M ratio) on a grit blasted stainless steel substrate kept at three different substrate temperatures - room temperature, 300 and 600 °C. The porosity of the mullite coating was lower than that of the coating produced using the mixture of alumina and silica powder. Also, the porosity level decreased with preheating temperature for both coatings. It was found from the visual observation of the X-ray diffractograms, that the coatings produced from the alumina-silica powder mixture was possibly richer in crystalline phase, while the substrate preheating temperature was limited to 300 °C. In general, substrates are preheated to about 200 °C immediately preceding thermal spraying. Hence, the authors recommended an alumina-silica mixture instead of mullite for fabricating plasma sprayed mullite coatings. However, in this investigation no quantitative analysis of phases was performed.

It may be noted that, while sintered mullite is used a number of applications [1], the potential of mullite coatings are yet to be utilized fully. Chemical vapour deposition (CVD) processed mullite has a good potential for protection against jet burner exhaust or corrosive media like Na_2SO_4 [10]. A mullite based coating obtained using pack cementation process had been used to protect carbon composites from high temperature oxidation at 1600 °C [11]. Plasma sprayed thick mullite coating described in this paper, is likely to be suitable for diesel engine cylinder liners for its low tendency to crack with temperature variation [12].

Young [13] demonstrated that Rietveld analysis is a method of extracting quantitative information on the phase content of a solid consisting of multiple phases from its X-ray diffraction (XRD) pattern. It is a profile fitting method based on the least square approach. A profile fitting function is used to generate a calculated profile for a sample and then this profile is matched with the measured full range XRD pattern of the same, by refining the various parameters of the function. Thermally sprayed coatings often contain multiple phases and hence, this technique is very useful for quantitative phase analysis of such coatings. However, very little published data is available on this subject. In one such rare publication, Sabiruddin et al. [14] discussed quantitative phase analysis of plasma sprayed alumina coating using Rietveld refinement. The effects of some parameters like primary & secondary gas flow rate, nozzle diameter, SOD on retention of α-alumina in the coating were investigated. The phase fraction of γ alumina was found to increase with an improvement in melting condition. Wang et al. [15] sprayed anatase titania using various Ar-He-H_2 and Ar-He-N_2 plasma gas flow rate. The fractions of phases present in the coatings were calculated using Rietveld analysis and correlated with the variation of primary and secondary gas flow rates. Gualtieri et al. [16] used the Rietveld refinement technique to quantify the α, γ and amorphous phases in plasma sprayed alumina obtained using a single set of plasma spray parameters. Suffner et al. [17] performed Rietveld analysis of plasma sprayed alumina-zirconia composite coating using a single set of parameters.

Laser remelting can be used to consolidate thermally sprayed coating in terms of a low porosity and improved microhardness. Fu et al. [18] reported that after laser surface remelting of the plasma sprayed ZrO_2 coatings, the porosity and roughness of the as-sprayed coatings reduced significantly, while the adhesion strength between the coating and substrate increased. However, an extensive network of crack, as well as a few large bubbles, formed in the laser treated coatings. In another investigation, Yuanzheng et al. [19] had shown that plasma sprayed Al_2O_3 and Al_2O_3-13 wt% TiO_2 coatings become much harder and denser upon laser surface melting. The hardness of the as-sprayed Al_2O_3 coating increased from 993 to 1760 HV after a laser treatment. In the case of Al_2O_3-13 wt% TiO_2 coating the hardness value was found to increase to 1427 HV upon laser treatment from the original hardness value of 691 HV of the as-sprayed coating. The hardness was found to increase further with an increase in laser power. However, the authors did not report the porosity of either as sprayed or laser treated coatings.

Bandyopadhyay et al. [20] emphasized that it is important to understand the effect of parametric variation on the coating microstructure for any plasma sprayed coating. Additionally, formation of mullite coatings involves a reaction between two ingredients and a major goal in such an investigation is to improve the yield of the reaction product, i.e., mullite. In this case, it is imperative to find the effect of parameters on mullite yield. Laser treatment of such coating is expected to enhance mullite yield in addition to coating consolidation. This aspect has not yet been reported in any literature. In fact although it has been established by a number of researchers that it is possible to synthesize mullite by plasma spraying a mixture of zircon and alumina [4–8], no report is yet available showing the quantity of mullite produced in wt%.

Based on the above findings, the objectives of the present investigation have been formulated as follows:

i. To create a plasma sprayed zirconia-mullite coating from a mixture of zircon sand and indigenously available alumina powder using various process parameters.
ii. To study the phases, hardness and porosity of the coatings and their variation with process parameters.
iii. To monitor the amount of mullite in the above coating using Rietveld analysis and correlate the findings with the process parameters: SOD, arc power, primary and secondary gas flow rate and angle of powder injection.
iv. To observe the effect of laser remelting of the coated layer on mullite yield and coating microstructure.

2. Material and methods

Materials used in this experiment are alumina (Al_2O_3) and zircon ($ZrSiO_4$). The particulars of the powders are listed in Table 1. Alumina and zircon were mixed in 3:2 M, i.e., 45:55 weight ratio. The mixture was put in a planetary ball mill (Fritsch Pulverisette 6, Idar-Oberstein, Germany) and mixing was carried out for two hours. Ni-5 wt% Al was used as bond coat. The substrate material is C 20 low carbon steel having a dimension 60 × 40 × 5 (thickness) mm. The substrate was ground in both sides using a surface grinder. The ground substrates were grit blasted with alumina grit (grit size 60, air pressure 689 kPa and SOD 100 mm) inside a suction type grit blasting cabinet. The surface roughness of the grit blasted surface was measured using a Surtronic 3 P (Taylor Hobson, UK) surface roughness meter. The roughness value was 6.3 ± 0.5 μm Ra. An ultrasonic bath was administered to clean the grit blasted substrates. The coatings were deposited immediately after cleaning.