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Synthesis of mullite-based coatings from alumina and zircon powder mixtures by plasma spraying and laser remelting

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HIGHLIGHTS

- Mullite has been produced by plasma spraying of alumina–zircon powder mixture.
- As sprayed coating shows good integrity.
- Laser remelting reduced porosity and increased coating hardness.

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ABSTRACT

A mechanical mixture of alumina and pulverized zircon sand in 3:2 M ratio has been plasma sprayed to obtain mullite coating. Thereafter, the top layer of the coating has been remelted using laser. The presence of a mullite phase in the as-sprayed and laser remelted coatings has been confirmed qualitatively using X-ray diffraction. Both as-sprayed and laser remelted coatings have been characterized for their microstructure, hardness and porosity. The ultrafine grain structure of the coating produced by rapid quenching has been analyzed using transmission electron microscope. Presence of a mullite phase in the coatings has also been confirmed using small angle electron diffraction. Laser remelting has resulted in an appreciable reduction in porosity and increase in hardness in the coatings.

1. Introduction

Mullite is a highly attractive coating material for many industrial applications. Some examples of application of mullite are the following: panel for re-entry space vehicle, refractory bricks for glass melting tanks, sintered mullite based conveyor belt for continuous charging of annealing furnaces, etc [1]. While it has also been proved that it is possible to produce mullite coating by plasma spraying, very few reports are available on this subject.

Mullite can be produced either by thermal decomposition of kaolinite at a firing temperature of around 1800 °C [2,3] or by processing a mixture of Al_2O_3 and SiO_2. Some of the known processing techniques for mullite production are sol–gel technique [4–6], synthesis of hydroxyhydrogel [7,8] or reaction sintering of zircon and alumina [9]. The major limitation of mullite is its brittleness. To increase the utility of mullite, some toughness should be imparted to it by adding a relatively tough ceramic component like zirconia [10].

Li and Khor [11] have produced a mullite coating while plasma spraying a mixture of zircon sand and alumina powder. At a temperature above 1600 °C, the zircon sand dissociates into zirconia and silica. Molten alumina comes in contact with silica while particles hit the substrate and mullite forms. The equations of the reaction are as follows:

$$ZrSiO_4 \rightarrow ZrO_2 + SiO_2\text{, at around } 1600\ ^\circ C \quad (1)$$

$$3Al_2O_3 + 2SiO_2 \rightarrow 3Al_2O_3 \cdot 2SiO_2\ \text{(Mullite)} \quad (2)$$

The complete reaction is:

$$3Al_2O_3 + 2ZrSiO_4 = 3Al_2O_3 \cdot 2SiO_2 + 2ZrO_2 \quad (3)$$

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Li and Khor [12] have also analyzed the microstructure and composition of plasma sprayed coatings produced by $Al_2O_3/ZrSiO_4$ mixture. Five different types of coatings have been prepared using either spray dried or plasma spheroidized powders with a 40 kW or 100 kW plasma spraying equipment under varying arc energy. The spraying distances used in this study are 6, 8, 10, 12 and 15 cm. At a low power level, the plasma spheroidized particles have produced a denser coating as compared to the coatings produced from the spray dried powders. At a higher power level both powders have produced well molten splats. A further increase in arc energy while using a power of 100 kW has produced a coating with a sponge-like morphology. It suggests that some particles may have evaporated before they have reached the substrate owing to excessive plasma energy input. An increase in spraying distance at constant arc power has first improved melting conditions, followed by possible particle freezing beyond a critical distance of 12 cm. Li and Khor have also made notable contribution on crystallization behavior of zircon–alumina mixture upon plasma spraying [13], effect of plasma arc current and helium (secondary) gas flow rates on mullite formation [11] and mechanical properties of these coatings [14].

Plasma spraying of zircon and characterization of the dissociated products has been discussed elaborately by Das et al. [15,16]. In addition, Das et al. [17] have investigated on zirconia–alumina–mullite composite coating produced by plasma spraying a mixture of naturally available zircon sand and commercial alumina. During spraying, partial conversion of the ingredients to mullite has occurred and the coating has shown good microstructural integrity. The short time available for reaction has not been sufficient for the reaction to complete. It has been stated that it is possible to increase the yield of mullite to some extent by varying the plasma spraying parameters. However, neither the corresponding parameter values nor the mullite content has been included in this paper.

An et al. [18] have used andalusite (Al_2SiO_5) powder to prepare mullite coating on stainless steel substrate bond coated with NiCrAlY using atmospheric plasma spraying. At a lower power level (22.5 kW) strong peaks of mullite have been observed whereas at a higher power level the amount of amorphous phase has been found to increase. With higher arc power the powders have gained more enthalpy from the plasma plume resulting in a higher average particle temperature. Particles with a higher average temperature undergo faster quenching on impact with the substrate and hence yield a larger amount of amorphous phase.

One major limitation of thermally sprayed coatings is porosity. Laser remelting of the top coat is a technique to eliminate the pores from the coating. A remelted layer also offers a homogenized coating microstructure and a strong metallurgical bond at the coating-substrate interface. Laser processing also offers certain other advantages like short processing time, low thermal distortion and limited microstructural changes of the substrate.

Fu et al. [19] have investigated the effect of laser remelting on the wear behavior of plasma sprayed ZrO_2 ceramic coatings. The porosity and roughness of the coatings have reduced significantly after laser treatment, and the bonding strength has increased. However, there have been extensive network cracks, as well as a few large bubbles, in the laser treated coatings.

Yuanzheng et al. [20] have used laser treatment to modify the surface of the plasma sprayed Al_2O_3 and Al_2O_3-13wt%TiO_2 ceramic coatings. The laser treated layer has become much denser and the hardness has been greatly improved despite the presence of cracks. The hardness of the laser treated layers has been found to increase with an increase in laser power. Laser melting has also improved the wear resistance of ceramic coatings. The lamellar defect structure of the as-sprayed coating has been erased, and the compactness of the coating has been improved significantly after laser remelting [21]. A γ-Al_2O_3 to α-Al_2O_3 transformation has been found to occur upon laser remelting. To the knowledge of the authors no report is yet available in the literature on laser remelted mullite coating.

The objectives of the present investigation are:

1. To create plasma sprayed zircon–mullite coating from zircon sand and commercially available alumina powder.
2. To study the phases, microstructure, hardness and porosity of the coatings.
3. To observe the effect of laser remelting on the coating microstructure.

2. Materials and method

In this study, alumina (Al_2O_3) and zircon ($ZrSiO_4$) have been mixed in 3:2 M ratio, i.e., 45:55 weight ratio in a planetary ball mill (Make: Fritsch; Model: Pulverisette-6) for 2 h and the mixture has been plasma sprayed as top coat. Ni-5wt%Al has been used as bond coat material. The particulars of the powders are listed in Table 1. The powders have been characterized using a JEOL JSM 5800 Scanning Electron Microscope (SEM) and a Phillips X-ray diffractometer (PW 1729 generator and PW 1710 goniometer).

Rectangular sample of C 20 low carbon steel (60 × 40 × 5 mm) has been used as substrate material for this study. Both surfaces of the substrate have been ground first using a surface grinder (ALEX, NH 500, Mumbai, India). The ground substrates have been grit blasted inside a suction type grit blasting cabinet (Sandstorm, Bangalore, India) with alumina grits of grit size 60, 100 psi air pressure and 100 mm stand-off distance. The grit blasted samples have been ultrasonically cleaned in an ultrasonic bath and coating

Table 1
Powders used for coating.

Name	Size	Composition	Type	Supplier
Alumina	− 90 + 45 μm Individual platelet size: 0.5 μm–5 μm	Al_2O_3 99.443 wt%, TiO_2 0.007 wt%, SiO_2 0.045 wt%, Na_2O 0.280 wt%, CaO 0.025 wt%, Fe_2O_3 0.200 wt%,	Agglomerated	M/S Hindalco, India (powder type HT)
Zircon	−90 + 45 μm	ZrO_2 (+traces of HfO_2) 64–65 % SiO_2 32–33 % Fe_2O_3 0.08–0.13 % TiO_2 0.30–0.40 %	Crushed	Indian Rare Earths Ltd., Chavara, India. (powder type zirflor)
Nickel Aluminum	−90 + 45 μm	Ni 95 wt%, Al 5 wt%	Clad	M/S Sulzer Metco, Westbury, NY,USA, (powder type 450 NS)