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Plasma spray deposition of yttrium oxide on graphite, coating characterization and interaction with molten uranium

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Abstract

Yttrium oxide coatings on graphite substrates were prepared by atmospheric plasma spray technique. Temperature and velocity of the yttrium oxide particles during the plasma spray process were measured by 'spraywatch' diagnostics system. Coatings were characterized for phase composition, microstructure and thermal stability. Dense adherent coatings could be deposited at 24 kW power. Corrosion behaviour of the coatings in molten uranium was studied by using differential thermal analysis (DTA) up to 1473 K. The results showed that the coatings offered sufficient protection to graphite against corrosion by molten uranium.

Keywords: Plasma spray deposition; Yttrium oxide coating; Corrosion resistant coatings

1. Introduction

Plasma sprayed ceramic coatings are extensively used for thermal barrier, corrosion barrier and wear resistant applications.[1–5] High density graphite is used for processing uranium and its alloys. Since molten uranium is not chemically compatible with graphite, a protective ceramic coating should be provided on the crucible to prevent the reaction between uranium and carbon. Although many refractory oxides, carbides and nitrides have been suggested for this purpose,[6–8] yttrium oxide (Y_2O_3) is the most preferred coating material. Yttrium oxide has high melting point (about 2700 K) and is chemically stable in many reactive environments.[9,10] Besides, it does not react with uranium and many reactive metals. By virtue of these reasons, yttrium oxide is used as a protective coating on graphite crucibles and metallic moulds used for processing uranium and its alloys.[11–14]

Tournier et al.[15] studied the reaction of sintered yttrium oxide with molten uranium metal. These authors found that yttria reacted with molten uranium above 1675 K to form uranium oxide and sub-stoichiometric Y_2O_{3-x}. The dissolved oxygen in uranium, after the experiment, was reported to be 1.2×10^{-3} mole fraction. Yasushi et al.[10] melted uranium in sintered yttria crucible and reported a 100 μm thick layer of UO_2 after 96 h of operation at 1675 K. Padmanabhan et al.[16] reported that yttrium oxide was chemically stable against attack by uranium up to 3000 K from thermodynamic calculations. The authors also studied the reaction of yttrium oxide with molten uranium and found that there was no reaction between the molten metal and the oxide. Alangi et al.[17] reported that yttrium oxide coating offered sufficient protection to tantalum substrate against corrosion by molten uranium.

The present study focuses on plasma spray deposition of yttrium oxide on graphite substrates and characterization of the coatings. Yttrium oxide coating was applied directly over graphite without any bond coat. The coatings were characterized for phase composition and microstructure. Temperature and velocity of yttrium oxide particles in the plasma jet were measured by 'spraywatch' diagnostics system. Differential thermal

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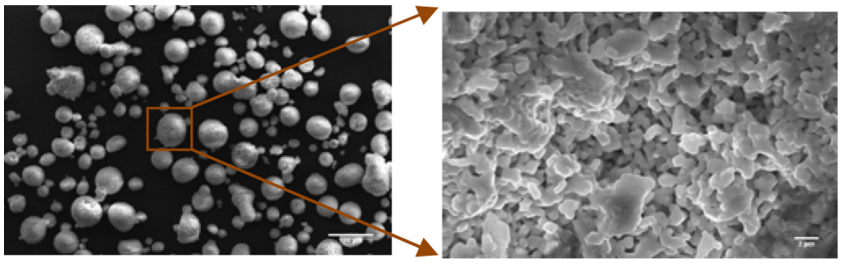

Fig. 1. SEM image of Y_2O_3 raw powder.

analysis (DTA) from room temperature to 1475 K was used to investigate the reaction between yttrium oxide and uranium.

2. Experimental methods

2.1. Plasma spray deposition

Chemically pure yttrium oxide powder (Grade: MEC2100P1 by M/s MEC Pvt. Ltd., Jodhpur, India) with particle size in the range of 15–40 μm was used for plasma spray deposition. The scanning electron microscope (SEM) photograph of the raw powder is shown in Fig. 1. The SEM image of a single particle is also shown alongside. It is evident from the figures that the powder consists of agglomerates of spherical particles.

A 40 kW DC arc plasma spray system, developed in our laboratory, was used for plasma spray deposition. The torch consists of a tungsten cathode (10 mm in diameter) with a conical tip and a copper nozzle, which acts as the anode. The anode is 40 mm in length and the nozzle diameter is 7 mm. The electrodes are cooled by water and a Teflon insulator separates the electrodes. A mixture of argon and nitrogen was used as the plasma gas, which was injected into the torch through an opening in the insulator segment.

Input power to the plasma torch was varied from 16 kW to 30 kW by controlling the gas flow and arc current. The flow rate of argon gas was varied from 25 to 30 standard litres per minute (SLPM). Trial experiments had shown that erosion rate of the nozzle was very high for gas flow rate of 20 SLPM and below. Therefore, experiments were carried out only at 25 and 30 SLPM of primary gas flow rate. Based on a large number of trial experiments and operational experience, the standoff distance between the torch and substrate was kept constant at 100 mm. Yttrium oxide powder was stored in a powder feeder and injected into the plasma jet by means of a carrier gas (Ar) through a side port located 2 mm before the exit of the plasma torch nozzle.

Table 1
Typical operating parameters for plasma spray coating of Y_2O_3.

Operating parameter	Experimental value
Input power	16–24 kW
Arc voltage	40 V
Arc current	400–600 A
Plasma gas	30 LPM
Plasma gas (N_2)	3 LPM
Powder carrier gas	10–12 LPM
Powder feed rate	~15 g/min
Particle size	15–40 μm
Torch-base distance	100 mm

The range of operating parameters for coating process is given in Table 1.

Spray deposition was carried out on graphite substrates, which were thermally etched by the plasma flame for about 1 min before starting the coating process. This was done to remove any organic binder and to increase the surface roughness of the substrate. The SEM image of the etched graphite surface is shown in Fig. 2. It is evident from the figure that the etched surface has better interlocking sites. The weight loss observed during thermal etching was small and found to be about 8.5 mg/cm^2. Plasma spray coating was carried out on cylindrical graphite specimens of 20 mm diameter and 10 mm thickness and rectangular specimens of 25 mm × 20 mm × 10 mm. Cubic graphite specimens (4 × 4 × 4 mm^3), coated on all sides with yttrium oxide, were used for differential thermal analysis (DTA) experiments. The coated cubic sample and a button of uranium metal were loaded in the DTA apparatus and the experiment was carried out from room temperature (RT) to 1475 K at a heating rate of about 25 °C/min. The specimen was held at 1475 K for about 1 h and then cooled to room temperature. Microstructure and energy dispersive X-ray (EDX) analysis of the coated sample after the DTA experiment was also carried out.

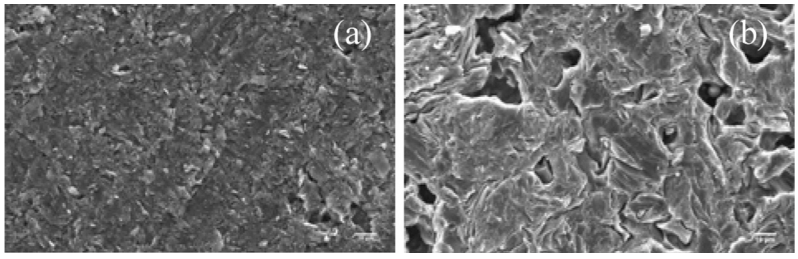

Fig. 2. SEM microstructure of graphite surface (a) before thermal etching and (b) after etching.