USE OF PLASMA SPRAYING IN THE MANUFACTURE OF CONTINUOUSLY GRADED AND LAYERED/GRADED MOLYBDENUM DISILICIDE/ALUMINA COMPOSITES

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Using platinum coatings on alumina (Al_2O_3) sheaths for thermocouples is a widely used practice in the glass industry. Protection of the thermocouple wires and alumina (Al_2O_3) sheathing is necessary to avoid corrosion and dissolution of the temperature-sensing unit. The cost associated with providing platinum coatings on the Al_2O_3 sheath material can be prohibitively high when taking into consideration the infrastructure needed at the glass plants to maintain and secure an inventory of available platinum. There are also issues associated with improving the performance of the platinum coated Al_2O_3. The failure rate of the thermocouples can be as high as 50%. The U.S. glass industry has been in search of alternative materials that can replace platinum and still provide the durability and performance needed to survive in an extremely corrosive glass environment.

Investigations by Y.S. Park et al [1] have shown that molybdenum disilicide ($MoSi_2$) has similar performance properties in molten glass as some refractory materials that are currently being used in glass processing applications. Molybdenum disilicide is a candidate high temperature material for such applications because of its high melting temperature (2030°C), relative low density (6.24g/cm^3), high thermal conductivity (52 W/mK), a brittle to ductile transition near 1000°C, and stability in a variety of corrosive and oxidative environments [2,3]. Additionally, the cost of $MoSi_2$ is significantly lower as compared to platinum coatings.

Plasma spraying has been shown to be a very effective method for producing coatings and spray formed components of $MoSi_2$ and $MoSi_2$ composites [4]. Investigations on plasma spray formed $MoSi_2$-Al_2O_3 composite gas injection tubes were shown to have enhanced high temperature thermal shock resistance when immersed in molten copper and aluminum [5]. The composite tubes outperformed high-grade graphite and SiC tubes when immersed in molten copper and had similar performance to high-density graphite and mullite when immersed in molten aluminum. Energy absorbing mechanisms such as debonding (between the $MoSi_2$ and Al_2O_3 layers) and microcracking in the Al_2O_3 layer contributed to the composites' ability to absorb thermal stresses and strain energy during the performance test (shown in Figure 1). Molybdenum disilicide and alumina are chemically compatible and have similar thermal expansion coefficients [6,7].

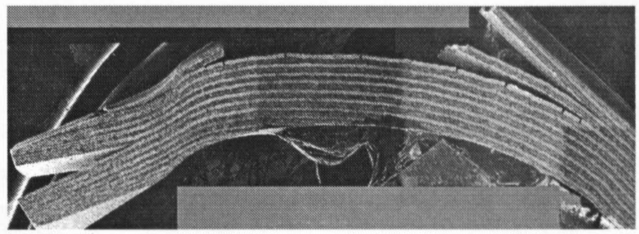

Figure 1. Four-point bend test beam after testing at 1400°C. Extensive debonding at the $MoSi_2/Al_2O_3$ interface and cracking within the Al_2O_3 was observed [5].

For thermocouple applications that require immersion of the thermocouple directly into molten glass, $MoSi_2$ coatings on Al_2O_3 protective sheaths will need to be optimized in order to perform in both a high-temperature (>1300°C) oxidizing environment (above the glass line) in addition to performing in the highly corrosive molten glass environment (below the glass line). We are currently evaluating the potential use of a graded coating of Al_2O_3 to $MoSi_2$ to enhance the performance of the $MoSi_2$ coating in molten glass. The graded microstructure of the coating will reduce the residual stresses that can develop during the spray deposition process, which can cause cracking and spallation of the coating limiting the coatings' lifetime. Preliminary results will be presented on the methodology used to produce the plasma sprayed $MoSi_2$-Al_2O_3 graded composites and on the microstructure and mechanical behavior.

Use of conventional plasma spraying equipment allows the flexibility of producing a variety of $MoSi_2$-Al_2O_3 microstructures, including laminate and graded structures. The plasma spraying equipment used for producing the graded structures included a Praxair Surface Technologies SG100 plasma torch and two Model 1264 powder feed hoppers. The plasma torch was mounted on a Fanuc S10 6-axis robot. A Technar DPV 2000 in-flight particle analyzer was used to measure the temperature, velocity and particle distribution of the $MoSi_2$ and Al_2O_3 particles as they exited the plasma torch. A computer control system was used to monitor and control the processing gases and the powder hoppers.

To determine the mechanical behavior of the graded $MoSi_2$-Al_2O_3 structures, C-rings were machined from the material deposited on the graphite rods. All of the samples were machined and tested in accordance with ASTM Standard C 1323-96. Twelve samples for each composite tube were tested. Four samples of monolithic plasma sprayed $MoSi_2$ and Al_2O_3 were tested and used for comparison. A Weibull statistical approach [8] was used to obtain the strength distributions in the coated and uncoated samples.

Macrographs of two types of $MoSi_2$- Al_2O_3 graded composites produced by plasma spraying are shown in Figure 2. The white phase is the Al_2O_3 and the dark phase is $MoSi_2$. Figure 2A, shows a layered and graded microstructure where the cross-section of the sample consists of discrete individual layers that have been graded from Al_2O_3 to $MoSi_2$. Figure 2B, shows the cross-section of a continuously graded structure where pure Al_2O_3 was first deposited on a graphite rod followed by increasing amounts of $MoSi_2$ until pure $MoSi_2$ is deposited on the outside diameter of the tube.

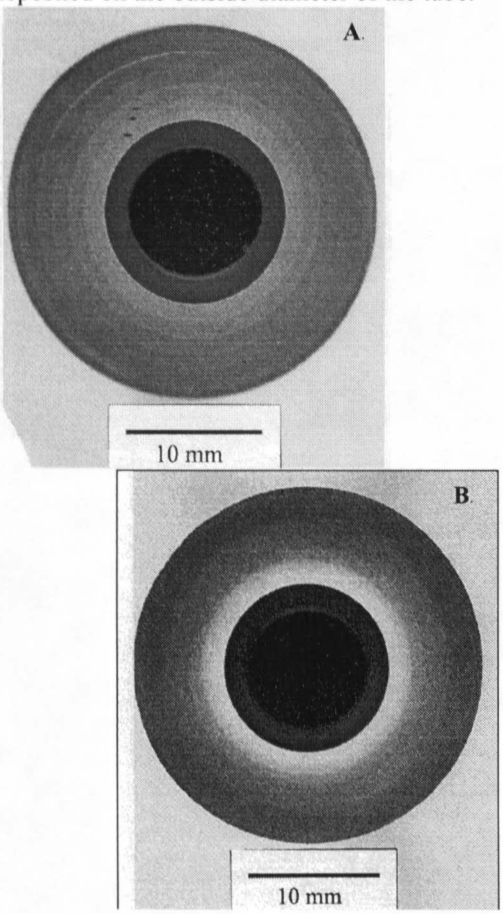

Figure 2. Macrographs of $MoSi_2$-Al_2O_3 graded tube cross-sections. a) Discrete individual layers that have been graded from Al_2O_3 to $MoSi_2$ and b) continuously grade structure from pure Al_2O_3 on the inside diameter of the tube to pure $MoSi_2$ on the outside diameter of the tube.

The mechanical behavior of the FGMs was evaluated using C-ring tests. Probability of failure at a given strength was obtained using Weibull analysis. Figure 3 shows the strength distribution plots for the layered and continuous FGMs. Both the continuous and layered FGM microstructures were found to exhibit similar mean Weibull strengths (~70 MPa). However, the spread of the data for the continuously graded material was smaller (hence a larger Weibull slope; 13.38 for the continuously graded samples, versus 7.635 for the layered graded samples). Interestingly, the fracture energy of the FGMs (qualitatively determined from the area under the load-displacement plot of the C-ring tests) was observed to be significantly higher (~3 times) than that of monolithic Al_2O_3 or $MoSi_2$ (monolithic Al_2O_3-285 J/m^2, monolithic $MoSi_2$-496 J/m^2, continuously graded $MoSi_2/Al_2O_3$ composite-766J/m^2, layered and graded $MoSi_2/Al_2O_3$ composite-955J/m^2. We are in the process of conducting independent fracture toughness tests to validate these preliminary observations and to determine if the graded microstructures exhibit R-curve behavior.

The fracture surface exhibited extensive microcracking and roughening in the center portion of the C-ring. We believe that the increased toughening of the composite is a direct result of microcracking. Preliminary analysis has indicated the strength and toughness of these FGM tubes to be more than acceptable for the proposed applications.

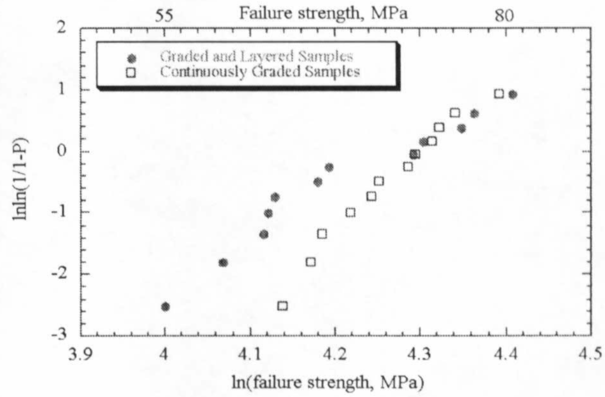

Figure 3. Results of C-ring tests performed on continuously graded and layered graded Al_2O_3-$MoSi_2$ coatings.

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