Noncontaminating Plasma Arc Sprayed Crucible Coatings for Containing Molten Ceramic Oxides

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A study was performed to assess the suitability of several plasma arc sprayed coatings applied to graphite crucibles for melt processing Al2O3-ZrO2, Al2O3-Y2O3, and Al2O3 ceramics. Coatings of W, Ta over W, and Re over W were evaluated. Pressed compacts of Al2O3-ZrO2, Al2O3-Y2O3, and Al2O3 were each placed in refractory metal-coated graphite crucibles and heated to 2040, 2150, and 2200 °C, respectively. Compatibility of the coating/ceramic oxide systems was evaluated by optical and scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), and combustion chromatography. The Ta over W coating system was chemically nonreactive with all three molten oxides studied.

1. Introduction

AMES Laboratory recently established the Ames Laboratory Plasma Spray (ALPS) Facility. The operation currently is focused on atmospheric and low-pressure plasma arc spraying of ceramic and advanced metal alloy systems for numerous applications. One major activity involves developing coating systems for containment crucibles used in high-temperature processing techniques such as high pressure gas atomization (HPGA), self-heating synthesis (SHS), and solidification studies. As new refractory materials are processed through molten or reaction routes, crucibles that are nonreactive to the materials being processed are required; furthermore, process temperatures of new materials under evaluation frequently exceed 2000 °C. For example, the Ames Laboratory HPGA system has been used to prepare powders of copper/refractory metal alloys.[1] However, during this study, attempts to contain a molten Cu-21Nb-2Mo alloy in a molybdenum alloy crucible at 2000 °C resulted in substantial selective dissolution of Mo from the crucible inner wall. Figure 1 highlights the crucible and atomization region of the HPGA system. The stringent demands placed on the containment crucible are quite clear. The crucible must first be noncontaminating to the molten material, which typically is heated to 200 to 400 °C above its liquidus temperature. Second, it must withstand the mechanical load of the molten charge. Third, it must survive the thermal stresses induced at the melt line. The inherent static thermal gradient developed during melting, together with the dynamic thermal gradient produced as the molten charge leaves the vessel, increases the crucible susceptibility to catastrophic thermal shock damage.

Refractory materials that provide the required high-temperature chemical stability described above, e.g., ceramic oxides and intermetallic compounds, typically are characterized by brittleness and very poor thermal shock resistance. Graphite is often used as a containment vessel material because of its high-temperature mechanical stability and compatibility with many materials. However, systems that tend to form carbides cannot be used, nor can materials that suffer from interstitial carbon impu-

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Fig. 1 General configurations of containment crucible and atomization regions of Ames Laboratory HPGA system.
rities. Therefore, an approach has been devised that combines the chemical stability of various refractory materials with the physical stability of graphite. Figure 2 shows the composite crucible design that has been evaluated. The plasma sprayed coating system is chosen based on the thermodynamic stability of the interface between it and the process material. Because the coating is thin, thermal gradients are less steep, decreasing the potential of the coating for cracking or spalling by thermal shock. In addition, the authors' experience has shown that it is beneficial to precede the desired noncontaminating layer with a refractory metal layer, e.g., tungsten, directly onto the graphite surface. The refractory metal layer is intended to block carbon migration from the graphite to the subsequently sprayed protective layer and the process material and also increase adherence of the composite coating system to the graphite crucible. Recent programs have demonstrated the effectiveness of this composite crucible approach for containing molten Cu-15Cr (vol%) alloys. A coating system of ZrO2-8wt%Y2O3 over W was far more stable in the presence of the molten alloy at 1800 °C than a series of Ta-base coatings applied over W. A subsequent study found that the ZrO2-8wt%Y2O3 over W coating was quite effective in containing Cu-Cr alloys with Cr contents up to 90 vol%. In the current study, refractory metal coating systems were evaluated for containing molten Al2O3-base ceramics. This work was motivated by the desire to study the solidification behavior of ceramic oxides in an attempt to understand the structure/property relationships of ceramic materials having tailored microstructures.

2. Materials

2.1 Ceramic Oxides

The Al2O3-ZrO2, Al2O3-Y2O3, and Al2O3 used in this study were purchased from CERAC, Inc., Milwaukee, WI. All three oxides were reported to be 99.99% pure. The Al2O3-ZrO2 is a eutectic mixture of 60 at% Al2O3. The stoichiometric Al2O3-Y2O3 is 62.5 at% Al2O3, the yttrium aluminum garnet compound. The powders were dry mixed and isostatically pressed to form compacts for melting.

2.2 Graphite Crucibles

High-purity graphite crucibles were purchased from Beaucor, Epsom, NH. Total ash content for this material is reported to be <15 ppm by weight.

2.3 Plasma Arc Sprayed Powders

The coating systems evaluated were 0.025 in. (0.635 mm) W, 0.015 in. (0.381 mm) Ta over 0.010 in. (0.254 mm) W, and 0.015 in. (0.381 mm) Re over 0.010 (0.254 mm) W. Figure 3 shows SEM photomicrographs of the plasma spray feedstock powders.