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Vacuum Plasma Spray Forming of Refractory Metals and Ceramics for Space Furnace Containment Cartridges—An Extended Abstract*

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1. Background

VACUUM plasma spray (VPS) is being evaluated as a method to fabricate high-temperature furnace containment cartridges (e.g., retorts) for the U.S. Microgravity Laboratory (USML) crystal growth furnace. These refractory metal cartridges are being developed to provide a sample containment vessel for growing single-crystal semiconductors and other materials in microgravity aboard the space shuttle. Currently, there are no feasible cartridges satisfying the requirements for space furnaces operating in the temperature range of 1260 °C (2300 °F) and 2000 °C (3632 °F). Inconel 718 has been satisfactory for previous experiments operating at 1150 °C (2100 °F). To provide containment for experiments up to 2000 °C, a variety of refractory metals (i.e., Re, W, Ta, Mo, Nb-Hf, W-25Re, and Mo-40Re) were considered. While these metals provide adequate strength at these high temperatures, they would be less able to withstand high-temperature oxidation or liquid metal corrosion should the sample containment (SiO_2) leak molten semiconductor material (i.e., GaAs). A variety of ceramic materials (BN, SiC, Si_3N_4, SiO_2, and Al_2O_3) are impervious to the aggressive attack of the molten semiconductors and provide a high service temperature. However, the ceramics are too brittle to be fabricated and handled in the thin 0.77 mm sections required for this application. The formation of a ceramic and refractory composite structure was of interest to utilize the desirable properties of both materials while compensating for their weak points. The high-temperature capabilities and corrosion resistance of the ceramics, combined with the ductility and toughness of the refractory metals, would lead to a very robust cartridge for USML.

The VPS process has been adapted to a wide range of applications at the NASA Marshall Space Flight Center VPS facility, from depositing thin thermal barrier coatings on turbine blades to net shape fabrication of complex-geometry parts, such as rocket engine main combustion chambers and these furnace cartridges. Novel tooling, leachable/removable mandrel technology, powder injection, and gun and part manipulation methods were applied in the fabrication of these cartridges.

Novel powder injection techniques were investigated to form a ceramic/refractory metal gradient structure. The composite tube was formed in situ during a single spray cycle in the VPS chamber. The gradient between the ceramic and metal is being utilized to lessen the difference between thermal expansions through a controlled gradient.

2. Experimental Procedure

2.1 *Powder*

Each of the powders received was evaluated for size distribution via Micro-Trak Systems, Inc., Mankato, MN and morphology via scanning electron microscopy. The powders are listed in Table 1. All of the powders were found to be very angular; none was gas atomized. The powders were all −325 mesh.

2.2 *Spray Mandrels*

Leachable or removable mandrel concepts were considered, depending on the necessity to spray the inside diameter to net shape. Mandrel materials included mild steel, zirconia, and graphite. Mandrels were evaluated for deposition efficiency and stability in the VPS environment, as well as ease of removal.

2.3 *VPS Parameter Development*

Evaluation of material microstructure via optical microscopy was used in developing the parameters and the gun manipulation program to spray these materials on the mandrels. The microstructures of the VPS-processed materials were qualitatively evaluated for density, grain size, and homogeneity. Spray pa-

Table 1 Powders being evaluated for fabrication of the space furnace cartridge tube

Refractory metal powders	Ceramic powders	Refractory/ceramic powder blends
W	Alumina	30%Mo-70%BN
Ta	Silica	50%Mo-50%BN
Mo	SiC	70%Mo-30%BN
Re	SiN	30%Alumina-70%BN
Nb-Hf (WC103)	BN	
W-25Re		
Mo-40Re		

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rameters and gun manipulation programming were then adjusted accordingly. Tungsten powder was used in the development stage of this program due to its low cost compared to that of the other refractory metal powders.

3. Results

3.1 *Spray Mandrels*

Graphite mandrels were found to best survive the harsh VPS environment. The ceramic (zirconia) mandrels could not withstand the thermal shock from preheating with a plasma gun and shattered and fell to the bottom of the chamber. The steel mandrels survived the high temperature but were more difficult to remove from the sprayed deposits. Therefore, development has concentrated on the graphite.

Initial work was carried out on a graphite mandrel that had sulfur outgassing due to the coal pitch tar used as a binder. Current emphasis has switched to graphite that receives a high-temperature bakeout to volatilize and drive out the organic binders.

3.2 *VPS Tungsten*

Tungsten powder was vacuum plasma sprayed on graphite mandrels to a thickness of 0.030 in. The mandrels were cross sectioned, mounted, and metallurgically examined. It was found that tungsten could be deposited to 97% density. The as-sprayed microstructure exhibited a strong dependence on the substrate temperature during powder deposition. Tungsten deposited on substrates at 816 °C (1500 °F) or below revealed a splat, laminar morphology.

Tungsten powder that was VPS processed on mandrels preheated above 1150 °C (2100 °F) showed the beginning of recrystallization and high density. A transition temperature range of 927 to 1093 °C (1700 to 2000 °F) was found where recrystallization began on some, but not all, specimens. Powder particle solidification at high temperatures, combined with recrystallization and grain growth, increased the tungsten density to 99%.

3.3 *VPS Alumina/Tungsten*

Samples of tungsten were sprayed over and with VPS-deposited alumina to develop techniques for applying a protective barrier coating over the refractory metal. The composite layering of alumina and tungsten specimens was accomplished in one continuous operation. Alumina was first deposited to 0.05 mm thickness. Then, without interruption, tungsten powder was phased into the deposition in continually increasing proportion. The alumina/tungsten transition layer was formed to 0.10 mm thickness, after which 100% tungsten was deposited to 0.58 mm (0.023 in.) thickness.

The microstructure consisted of a dense alumina layer with a noncontinuous gradient where isolated areas rich in alumina were encapsulated by a tungsten matrix. The VPS deposition of alumina and tungsten tubes did demonstrate the integral in situ formation of the ceramic-coated refractory material.

4. Discussion

Tungsten furnace containment cartridges have been formed via vacuum plasma spray. As-deposited tungsten was 97% dense in a splat structure and 99% dense in a recrystallized structure. The microstructure varied from a splat structure to a recrystallized structure, depending on substrate spray temperature. Tungsten powder deposited below 815 °C (1500 °F) showed no signs of recrystallization. In situ recrystallization of the VPS tungsten occurred at temperatures above 1150 °C (2100 °F), whereas specimens deposited between 815 to 1150 °C (1500 to 2100 °F) exhibited only occasional recrystallization.

The higher deposition temperatures induce recrystallization of the tungsten, producing a denser, more homogeneous product. Metallographic analysis shows that prior powder particle boundaries are consumed in the recrystallization process. Past experience has shown that the recrystallization and grain growth are necessary for good elevated-temperature strength and ductility.

Techniques to deposit a layer of alumina and transition to tungsten were successful. A gradient layer was produced between the 100% alumina and 100% tungsten.

5. Conclusions

- Tungsten can be VPS formed to 99% density in furnace retort shapes.
- The VPS tungsten microstructure is dependent on substrate temperature. Material deposited below 815 °C (1500 °F) displayed a splat structure, whereas material deposited above 1150 °C (2100 °F) was recrystallized.
- The VPS process can be used to make transitions from alumina to a gradient layer with a continuously increasing proportion of tungsten to a pure tungsten layer in a single spray operation.