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DEVELOPMENT OF A REMOTE BUSHING SYSTEM FOR AMERICIUM-CURIUM VITRIFICATION

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ABSTRACT

The Savannah River Site (SRS) and the Savannah River Technology Center (SRTC) are combining their existing experience in handling highly radioactive, special nuclear materials with commercial glass fiberization technology in order to assemble a small vitrification system for radioactive actinide solutions. The vitrification system or "bushing", is fabricated from platinum-rhodium alloy and is based on early marble remelt fiberization technology. Advantages of this unique system include its relatively small size, reliable operation, geometrical safety (nuclear criticality), and high temperature capability. The bushing design should be capable of vitrifying a number of the actinide nuclear materials, including solutions of americium/curium, neptunium, and possibly plutonium. State of the art, mathematical and oil model studies are being combined with basic engineering evaluations to verify and improve the thermal and mechanical design concepts.

INTRODUCTION

A solution containing the isotopes of americium-243 and curium-244 (Am/Cm) is stored in the F-Canyon, Separation Area at the Savannah River Site. This material was accumulated from processing plutonium targets during the 1970's. These materials are the only concentrated kilogram sources of these isotopes in the United States and have an extremely high intrinsic value for the nation. This is particularly true if the material can be further enhanced by transformation at the Oak Ridge National Heavy Element and Advanced Neutron Facility in Oak Ridge, Tennessee.

Alternatively, the disposal of this material to the SRS Tank Farm as a waste would create a number of problems and concerns due to the very high level of radioactivity involved. Transportation of the radioactive Am/Cm in liquid form to Oak Ridge is not considered acceptable under present standards. An acceptable solution is to vitrify this material in available space in the Separation Canyon prior to transporting to Oak Ridge in sealed containers within shipping casks. The vitrified form would be relatively safe to transport and the material would then be redissolved in acid at Oak Ridge. The Am/Cm would then undergo nuclear transformation to californium-252 (Cf) and other useful transplutonium isotopes.

After reviewing a number of available vitrification technologies which could be employed in the canyon environment, a technology was selected which has been employed in the glass fiber industry to remelt glass marbles and draw glass fibers. This technology involves the direct resistance heating of the platinum bushing as described in a later section of this report and in greater detail in a number of published works. [1-2]

Am/Cm PROCESS DESCRIPTION

At the present time, the Am/Cm is dissolved along with other lanthanides in 12,000 liters of 4 normal nitric acid and stored in a stainless steel tank in the Canyon. There are about 10 kg of Americium-243 emitting about 1,800 Curies and 3 kg of Curium-244 emitting about 210,000 Curies. This material is maintained inside a shielded cell behind heavy walls of concrete and glass. The thermal energy generated from the nuclear activity amounts to about 7 kw/hr total. During processing, oxalic acid will be added to the nitric acid solution and the actinides and lanthanides will be precipitated as oxalates. The remaining aqueous fraction will be removed and the lanthanide-actinide oxalates will be redissolved in nitric acid. The Am/Cm solution will be evaporated and steam stripped down to approximately 100 grams/liter solids in 1 to 3 molar nitric acid solution. The solution will be stored in a stainless steel tank until it can be fed along with a special glass frit to the proposed vitrification system. The glass frit and nitric acid solution will be fed in two separate streams and melted together to form a lanthanide borosilicate glass. A compositional description of an initial frit and the glass surrogate are presented as Table 1. Glass research is being undertaken to try to replace the lead and barium oxide constituents.

Complete recovery of the Am and Cm surrogates (La and Nd) from the glass has been demonstrated by a nitric acid extraction technique. While the glass can be attacked by concentrated acid it is several orders of magnitude more durable in water than the required specification for high level waste glass. [3-5]

Table 1
Surrogate Frit and Glass Formulation - Am/Cm

Oxide Composition	Frit (Wt%)	Am/CmGlass (Wt%)
SiO_2	44.8	26.3
B_2O_3	5.6	3.3
BaO	6.1	3.6
Al_2O_3	23.7	14.1
La_2O_3	1.5	16.5
Nd_2O_3	- - -	17.6
CeO_2	- - -	7.3
PbO	18.4	11.0

REMELT FIBERIZATION TECHNOLOGY

The technology selected for the vitrification was derived from the glass marble-remelt fiberization process, which was the primary method of continuous fiberization until the mid 1960's. Specialty glass fibers with diameters less than 6 microns are still manufactured by this type of process. The small platinum alloy melters or "bushings" are heated by passing large electrical currents (thousands of amps) through the end terminals and into the platinum sides and heater screens in the bushing.

The design of a typical marble remelt bushing is complicated by the number of functions the bushing must complete. These functions are to receive and melt glass marbles, condition the glass melt for fiberization, and distribute the glass melt evenly across the fiberization section while maintaining an even temperature. Two factors are employed to adjust and maximize this process. First the position of the water cooled connectors on the end terminals can significantly affect the temperature of the bushing ends. Also the location and thickness of the platinum alloys between the end terminals can influence the thermal energy distribution. The amount of heat generated in a bushing is a function of the current path and the electrical resistance (thickness) of the metal sheets. If the bushing is constructed of continuous sheets between the terminals it can be generalized that the amount of heat produced in a particular area of the bushing is directly proportional to the thickness of the alloy in that region. In order to provide an abundance of heat for the melting of the glass marbles an additional heating screen is included in the top section of the typical marble bushing design as shown in Figure 1.

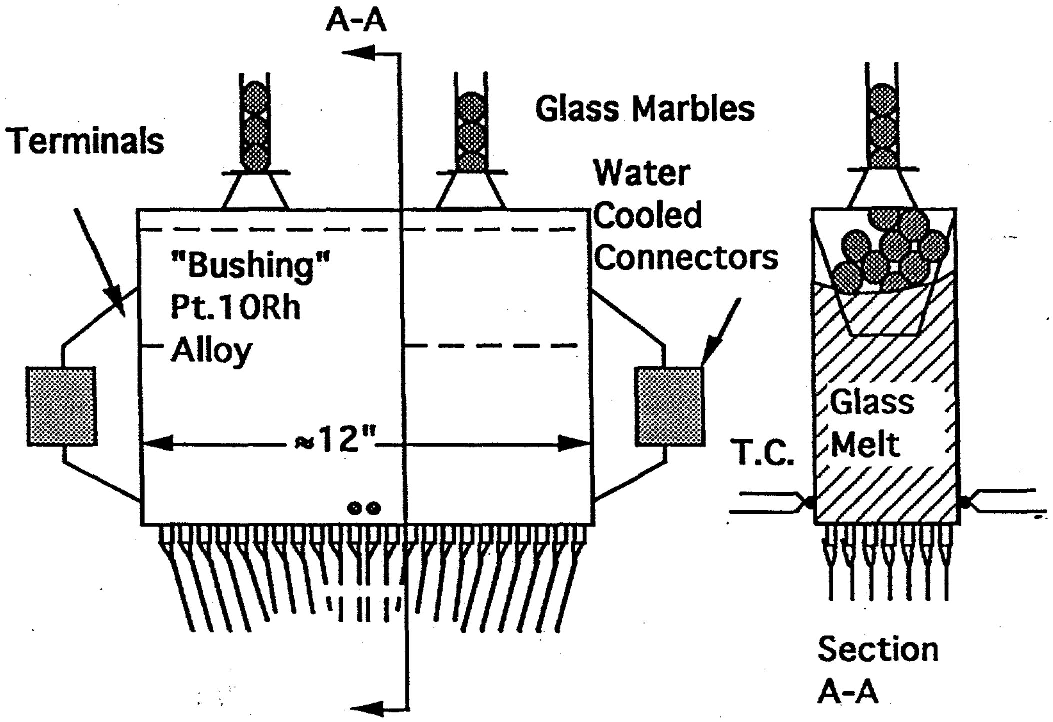

Figure 1. Typical Marble Remelt, Fiberization Bushing.

The typical operating parameters for a 408 fiber marble remelt bushing are presented in Table 2. [2] These parameters would be increased or decreased for other types of bushings.

Table 2
Operating Parameters for Typical Marble Remelt Bushing

Number of Fibers/Bushing	408
Available Power	25 to 35 KVA
Nominal Power	18 KVA
Nominal Voltage Drop	2-6 Volts
Throughput	15 kg/hr
Melt Energy	5 KVA
Platinum Alloy	4 kg. Pt10Rh

Since the bushing alloy is an excellent conductor of electricity it requires a great deal of current to generate thermal energy. This high current density requires heavy air or water cooled bus bar systems to carry the high currents from the step-down transformer to the bushing.

A very simple schematic of a typical remelt bushing process is shown as Figure 2. The temperature and power control systems can include over temperature control, ramping, and loss of power restrictions. Because the glass fiber industry requires a large number of these systems (hundreds/plant), the cost of a single control system is relatively low.

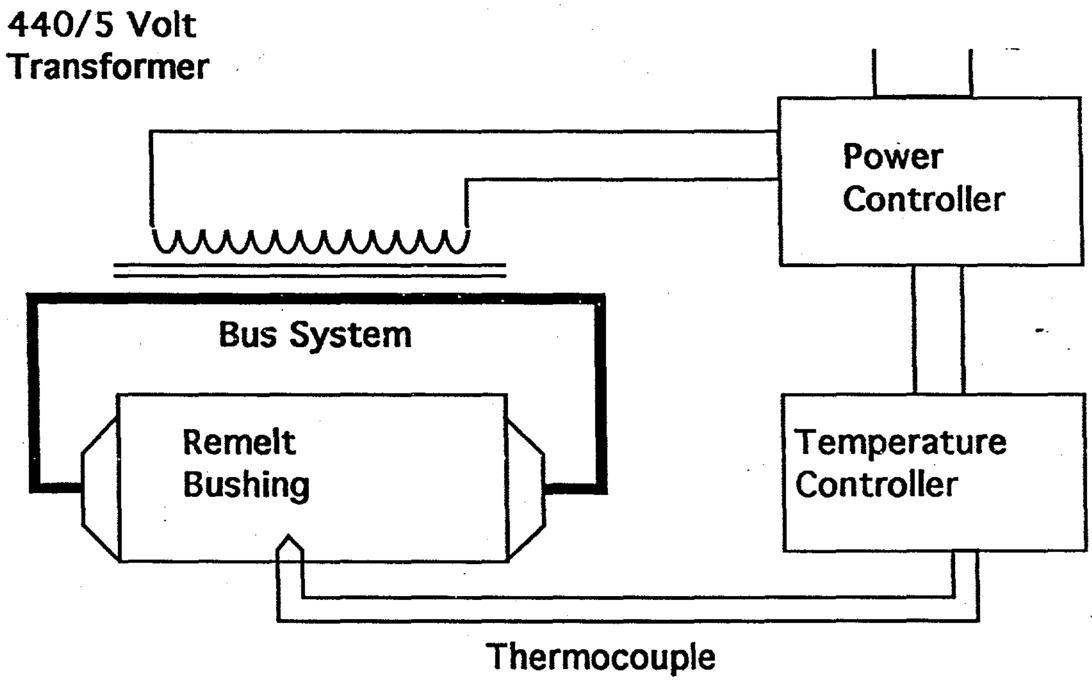

Figure 2. Simple Schematic of a Typical Marble Remelt Bushing.

Am/Cm BUSHING DEVELOPMENT

The melter for the vitrification of the Am/Cm solution was initially based on the marble remelt bushing technology. These types of melters offer several distinct advantages for the vitrification of actinides:

- Bushings can be cooled to room temperature and restarted as required,
- The bushings are chemically inert due to the use of platinum-rhodium alloys,
- The typical commercial bushing has a geometrically favorable design, similar to the slab tanks used in the nuclear materials industry, and
- The bushing melter is relatively small and simple to operate.

A number of design requirements were established for the Am/Cm project. The bushing melter is fed with two streams, one containing the ground frit and the other the nitric acid Am/Cm solution. The nitric acid feed contacting the hot glass was expected to be highly corrosive and generate a highly corrosive, acidic off-gas. It was required that the melter would produce about 300 kg of Am/Cm containing glass over a reasonable period of time. The average melt rate should be between 1 to 2 kg/hr. and a melt temperature approaching 1450°C. should be possible. The exiting glass flow would be controllable by remote means. The melter would be capable of being drained to an empty condition. It was expected that the complete melter portion of the system would fit on a single "process rack" which is normally used to support equipment in the canyons. The melter and in cell support systems had to survive the radiation field from the Am/Cm glass.

Some of the requirements for processing the Am/Cm were quite different from the conventional marble-remelt fiberization bushing technology. The liquid feed was a major departure and has caused considerable concern throughout the development program. Partially compensating the liquid feed issue is the fact that the glass quality does not have to be as high as the glass quality for fiberization. The first bushing designed for the Am/Cm program was similar to the marble remelt bushings. A second heating screen was installed near the bottom to further heat the glass prior to exiting. The tip section for fiberization was replaced with a solid sloping bottom and a single drain tube at the central axis. The inside diameter of the drain tube was 0.125 inch and its length about 0.75 inch. An air jet was installed at the bottom of the bushing, aimed at the drain tube, to stop the glass flow. A platinum wire wound resistance heater was placed around the drain tube to help start the glass flow. The bushing was fabricated and, set into a high alumina castable refractory.

A fairly large Inconel-690 off-gas plenum was developed which matched the bushing. A ceramic separator was placed between the two parts to maintain the electrical isolation of the plenum. A small viewing port was also included in the plenum, but is not shown in Figure 3 for the sake of simplicity.

The first bushing was operated and demonstrated the proof of concept. However, the operation over an extended period of time indicated several problem areas and the need for a number of design modifications:

- The air/resistance heat, flow control was successful in starting and stopping glass flow, however a fairly large number of droplets preceded and followed the stream flow and produced an unacceptable number of glass threads. These threads would tend to increase the spread of contamination.

- In order to minimize the number of glass threads formed during draining it was decided to drain at a continuous flow stream of about 7 to 8 kg/hr. This would require that the bushing be operated in a batch mode i.e., feeding to build level over a number of hours then draining glass for a number of minutes. This cycle would then be repeated.

- The large open area at the top of the bushing allowed excessive heat to exit the bushing to the plenum and limited the feed melting rate.

- The lower screen was an impediment to the melting operation. During evaporation and melting of the liquid/frit feed, when more power was required to enhance the melting rate, the bottom melt temperature exceeded the temperature limit setting for the system.

- The expansion of the alloy against the castable ceramic created warpage of the metal which eventually introduced a small tear in the sidewall.

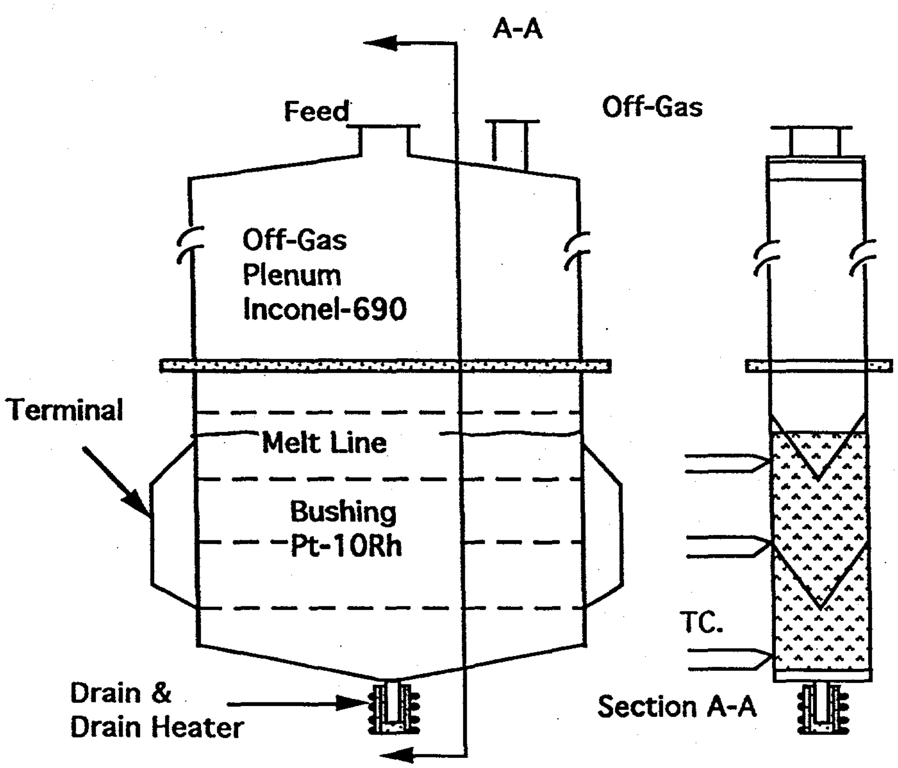

Figure 3. Simplified Schematic, First Am/Cm Bushing.

During this period of time additional commercial glass fiber and specialty glass producers were contacted and extensive discussions were held. It was decided that using geometry and the terminal clamps to control the heat distribution in the bushing would require numerous trial and error designs. A more expeditious route would be to install a two zone power supply and provide two pair of terminal connectors.

Oil modeling of the bushing and drain was established to investigate convection within the bushing and glass flow through the drain tube. In order to obtain more control in starting and stopping the flow a drain tube about six inches long with an inside diameter of about 0.200 inch was selected. Oil studies confirmed the ouput rate calculated using Poiseuille's equation:

$$F = K\, r^4\, h / l\, \mu,$$

where F is the rate of flow of glass from the drain tube, r is the radius of the tube, h is the height of the glass above the drain tube, l is the length of the tube, and μ is the viscosity of the glass in the tube. [2]

Additional mathematical computer modeling of heat transfer and fluid flow were initiated. An ABAQUS ™ Finite Element Computer program was utilized to map the current density in the two zone bushing. Results from this code indicated that the two zones should apportion power independently. In other words, during melting the power could be concentrated in the upper zone, while during pouring some power in the lower zone reduced the temperature gradient and helped initiate pouring. A CFX-4™ heat transfer and fluid flow program was used to study flow patterns in the bushing and flow control in the orifice tube. These mathematical computer models provided a great deal of support and guidance to what had previously been an "intuitive" design effort. A number of potential replacements for the "V" trough heater screens were considered and eventually the V screens were replaced with vertical heater strips which would permit the liquid feed to fall onto the glass surface at the expected melt levels. The vertical strips also reduced the stresses on the side-walls. The over-melt space was redesigned to reduce heat loss and improve evaporation of the liquid. A simple schematic of the second bushing is shown in Figure 3.

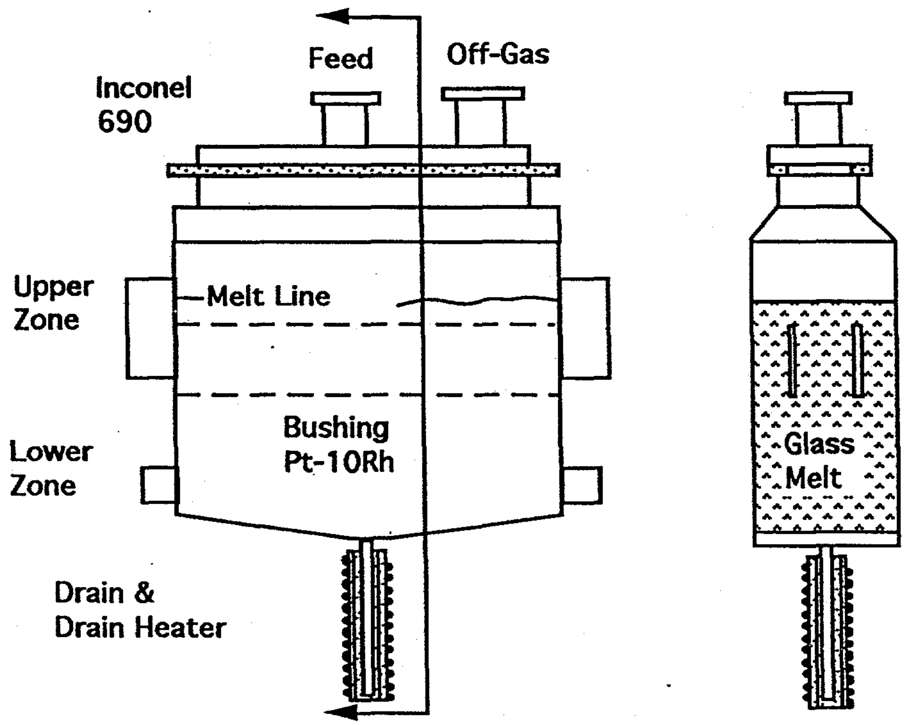

Figure 3. The Second Am/Cm Bushing Design.

The second bushing has now been evaluated and all of the systems appear to have operated as expected. Some problems still remain in obtaining the desired melt rate. The two zone control appears to operate as planned although the upper power level is presently somewhat limited. Efforts are under way to establish a separate bushing dedicated to flow control and to investigate the potential of inductively heating the drain.

CONCLUSIONS

Commercial marble remelt-fiberization technology was combined with existing experience in remote handling of special nuclear materials to provide equipment suitable for the vitrification of residual highly radioactive nuclear materials. Development of this process is continuing, with emphasis in the areas of melt rate enhancement, two zone power control, and remote glass flow control.

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