

**Vitrification of Actinide Solutions in SRS Separations
Facilities (U)**

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Vitrification of Actinide Solutions in SRS Separations Facilities

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ABSTRACT

The Savannah River Site (SRS) Separations Canyons will eventually undergo Decontamination and Decommissioning (D&D). In order to D&D these facilities, material (such as Americium and Curium) currently in tanks must be dispositioned. Actinide solutions in these tanks contain extremely valuable nuclear isotopes for which viable applications exist. The National Heavy Element and Advanced Neutron Source (ANS) program at Oak Ridge can use this material; however, a process to convert this solution to a solid form for safe transport does not currently exist. Actinide glass compositions were developed through Office of Technology Development (OTD) research. This research formed the cornerstone of a process to vitrify the Americium and Curium (Am/Cm). This process has been recommended by Westinghouse Savannah River Company as the most favorable option for Am/Cm disposition. Work continues towards developing a full scale prototype system to demonstrate the process and train operators.

The system would be installed in the Multi-Purpose Processing Facility (MPPF) of F-Canyon. This is an ideal location since it provides viewing windows, good shielding and access to the process with manipulators. The system will consist of a melter capable of at least 1-2 kg/hr glass output, feed systems, and an off-gas system. The glass will be poured into small canisters so the material can be safely stored or shipped to its destination. Two feed streams will be directed to the melter; glass marbles and a nitric acid solution which contains the material to be vitrified. The two streams are designed so that they will mix in the melter and form the appropriate composition to produce an acceptable glass. A significant amount of off-gas will be produced as a result of wet feeding which dictates the requirement to handle condensate in the off gas. The condensate will be collected, sampled for fissile material and discharged to the canyon waste tank.

The melter proposed for this program is a platinum / rhodium alloy bushing melter commonly used in the glass fiber industry; however, this is a new and unique application for this melter technology. The melter is an

alloy box with tabs on each end for an electrical connection. Current is passed through the bushing and heats it similar to a resistance heater. The unique features of the bushing melter for this application include the following: It will be enclosed so the off-gas can be contained and processed. The melter feed will be wet, that is the melter will be required to supply adequate power to vaporize the liquid portion of the feed and then melt the solids along with the glass beads entering the melter in a separate feed stream.

There are many specialized waste forms in the DOE complex which can not be easily processed in facilities like the Defense Waste Processing Facility (DWPF) at SRS or have a potential future use. The process being developed will provide the capability to convert specialized or unique forms of nuclear material into a stable, solid and safe form which can be stored indefinitely, shipped or reprocessed as the need arises. The successful demonstration of this technology to vitrify isolated actinide material could have future application in numerous locations across the DOE complex.

INTRODUCTION

The actinide vitrification system is being developed for several reasons. A long term goal for the Savannah River Site (SRS) is to Decontaminate and Decommission (D&D) facilities. At least one tank in the F-Area Separations Canyon contains valuable isotopes which the National Heavy Element and Advanced Neutron Source (ANS) program at Oak Ridge can utilize. A minimum of three options were investigated for dispositioning the Americium / Curium (Am/Cm) solution currently stored in an F-Area Separations facility tank. The evaluation determined that vitrification was the favored option since the end product is a solid glass log which can be safely stored or transported.

In order to D&D F-Canyon, stored and inprocess material must be processed or removed from tanks in the facility. This program was initiated to process a specific tank which contains Am/Cm in a nitric acid solution. Some tanks in the facility may be emptied to waste tanks. However, the solution in question could impose adverse effects if this were done. The replacement value for the Americium and Curium isotopes was estimated to be between one and two billion dollars. The quantity of material in this tank will supply all of the ANS program's needs for these isotopes for the foreseeable future. This program is serving two goals of dispositioning the tank of solution and supplying Oak Ridge needed isotopes. By stabilizing the isotopes in glass, they can be safely stored indefinitely for future use.

ACTINIDE VITRIFICATION SYSTEM DISCUSSION

The vitrification system must be operated remotely due to the radioactivity of the Am/Cm solution. The design of the equipment used in this system must ensure high reliability to minimize the amount of maintenance required. The vitrification system will be installed in the Multi-Purpose Processing Facility (MPPF) in F-Canyon. The MPPF is a unique facility in section eighteen of the canyon. It is the only section which has shielded oil filled windows for viewing the operation. Master Slave Manipulators are used for operating equipment in the shielded cells. There are eight window / manipulator stations in the facility, however, only one or two will be required for the vitrification system.

Each station has a removable rack that process equipment is mounted on. Two of the current racks which are no longer in service will be removed to accommodate the vitrification system. New racks will be fabricated to support the vitrification system. The new racks will be fully assembled and tested in a non-radioactive facility to ensure all the integrated systems function properly prior to installing them in the MPPF. Current estimates indicate that it will take approximately 300 hours of operation to process the Am/Cm solution. The expected process parameters are shown in table I. Table II and III show anticipated glass compositions for this process to vitrify Am/Cm. A schematic of the system is shown in Figure 1 and each major system is described in detail below.

Feed System

The feed consists of two components, a nitric acid solution which contains dissolved Am/Cm and a glass forming frit. The two feed streams must be fed in the correct proportion to make an acceptable glass product. Frit is a dry material similar to sand. Separate systems will be used to feed each stream into the melter lid. The two feed materials will be mixed in the melter and be poured out the bottom as a glass product.

The nitric acid solution will be pumped from a storage tank to a smaller feed tank in the MPPF. The solution is fed using a constant head arrangement as shown in Figure 1. The solution is airlifted from the bottom into a small vessel above the feed tank at a rate slightly higher than will be fed to the melter. An overflow from this small vessel is returned to the feed tank and a outlet below the liquid level feeds the melter. The fluid head pressure delivers the liquid feed to the melter through an orifice to regulate flow. The liquid feed system has a fixed flow

as determined by the equipment setup. Orifice changes can be made as required to alter the process parameters or feed a completely different solution. The liquid feed tube at the melter top will likely require cooling to prevent the solution from boiling in the feed line.

The dry frit feeder will have an adjustable feed rate which must be coordinated with the fixed solution feed rate so the ratio is in the target range. The frit will be fed using a weight loss feeder which determines the feed rate by monitoring the weight loss in the frit hopper over time. This stream will be delivered to a conveying system that will transport the material to the top of the melter where an air lock transfer device will ultimately discharge the dry feed into the melter. The air lock is necessary to maintain a seal on the melter and minimize the quantity of air infiltration so the melter off-gas system is not overloaded. It will also prevent the vapors in the melter head space from contaminating or possibly plugging the dry feed system.

Melter

The melter chosen for this vitrification system is called a bushing melter, which is basically a platinum / rhodium alloy box with tabs on each end. Electrical bus bars are connected to the tabs on each side of the alloy box which acts like a resistance heater when current is passed through it. Up to 15,000 amps at 2 volts are used to melt the glass forming materials. This melter technology is widely used in the glass fiber industry for drawing glass fibers. The vitrification bushing will have one drain hole as opposed to several hundred fiber drawing holes in a typical glass fiber bushing.

The novel attributes of the bushing melter that will be used for vitrification include wet feed and an enclosure above the melt. The feed for this melter includes dry frit and a solution of material to be stabilized as opposed to standard bushings which are fed glass beads or molten glass. The vitrification bushing must provide the heat of vaporization for the transport fluid as well as the heat necessary to melt the glass frit and oxides delivered in the feed solution. Using wet feed creates off-gas which must be contained and treated. A top will be installed on the melter which will seal the system so the off-gas can be drawn into the off-gas system. The top will also have two feed nozzles, one each for the wet and dry feed to enter the melter. In addition, a large port in the melter top directs the off-gas stream into the treatment system.

The glass product is poured into stainless steel canisters through a single nozzle in the bottom of the melter. The nozzle is sized to ensure the desired pour rate at the expected glass viscosity. Pouring is controlled by an air jet that freezes the glass in the pour nozzle and/or a platinum alloy flow stopper which acts as a heat sink.

Off-Gas System

This system will function to contain and treat the off-gas originating from the melter. It is desirable to maintain control over the stream coming from the melter since it may contain some small amount of radioactive particulate. The system will operate to cool and scrub this process byproduct and any air introduced into the system for pressure control purposes. The volume of off-gas is dictated by the vaporization of the nitric acid solution and the plant air introduced to control pressure in the bushing melter. A slight negative pressure is maintained inside the system to ensure that if a leak occurs the flow is into the system not out. This feature minimizes the possibility of further contamination of the shielded cells facility.

The off-gas system originates with the melter. A melter top is used to contain any volatiles emanating from the melter. This is a unique feature which has not been used on bushing melters before. The melter top has a port where the off-gas exits and enters the treatment system. There is a control air connection in the off-gas line immediately above the melter. An automatic valve that supplies control air is directed by the signal from a pressure transmitter connected to the melter vapor space. As dynamic changes effecting the melter pressure take place, a closed loop control system reacts to supply more or less air to the off-gas system which maintains a pressure of negative five inches of water vacuum inside the melter.

The stream then travels through the pipe to a quencher where the off-gas is quenched with a water spray. The quencher acts to cool, remove particulate and dilute the condensate. The two phase flow from the quencher flows into an off-gas condensate tank (OGCT) where particulate settles out and condensate is collected. The condensate level is monitored and pumped to a waste tank as needed. The vapor fraction of the off-gas stream exits the condensate tank through a port in the top and is directed into a high efficiency mist eliminator (HEME) where particulate is entrapped. The HEME condensate is drained back to the OGCT.

Once the off-gas exits the HEME it is directed to a heater which raises its temperature about ten degrees Celsius. This raises the temperature above the stream's dew point to prevent condensation in the High Efficiency Particulate Air (HEPA) filter. Condensation in the HEPA filter reduces its effectiveness and increases the pressure drop across the filter. The HEPA filter is the last component the off-gas flows through prior to entering the canyon process vessel vent system (PVVS).

Glass Storage Canister System

The glass product will be poured into stainless steel canisters which are approximately three inches in diameter and fifteen inches high. An automated system for handling the canisters remotely is required due to the radioactivity of the vitrified material and the fact that it will be performed in a shielded cells facility. A turntable capable of holding approximately eight canisters will be used to position empty canisters under the melter drain nozzle. Once a canister is filled, glass flow from the melter is terminated and the turntable rotates to position a new canister under the melter pour spout. Glass pouring can then be resumed so the next canister is filled.

Several of the turntable positions will hold full canisters and allow them to cool. Heat transfer from these canisters will be evaluated to determine the cooling rate. Insulated canister slots on the turntable may be required if the cooling rate needs to be adjusted to ensure the glass anneals properly. One station on the canister turntable will be setup to weld a cap on each canister. A mechanism to inventory and position canister caps so they can be welded is necessary in this remote application. The turntable location after cap installation is used to remove the full canister and insert an empty one.

A material handling system for the canisters must be developed. It will include a pick and place type device to automatically remove the full canisters and insert a new one. The full canisters must not only be removed from the turntable but transported to a location where they can be removed from the shielded cell facility and transported to their destination. Empty canisters will be delivered to the turntable to replace the full ones that are removed. The outer surface of the sealed canisters will be decontaminated to facilitate transportation of the sealed vitrified material.

CONCLUSION

The actinide vitrification system being developed at SRS provides the capability to convert specialized or unique forms of nuclear material into a stable solid glass product that can be safely shipped, stored or reprocessed according to the DOE complex mission. This project is an application of technology developed through funds from the Office of Technology Development (OTD). This technology is ideally suited for vitrifying relatively small quantities of fissile or special nuclear material since it is designed to be critically safe. Successful demonstration of this system to safely vitrify radioactive material could open up numerous opportunities for transferring this technology to applications throughout the DOE complex.

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TABLE I. EXPECTED PROCESS PARAMETERS

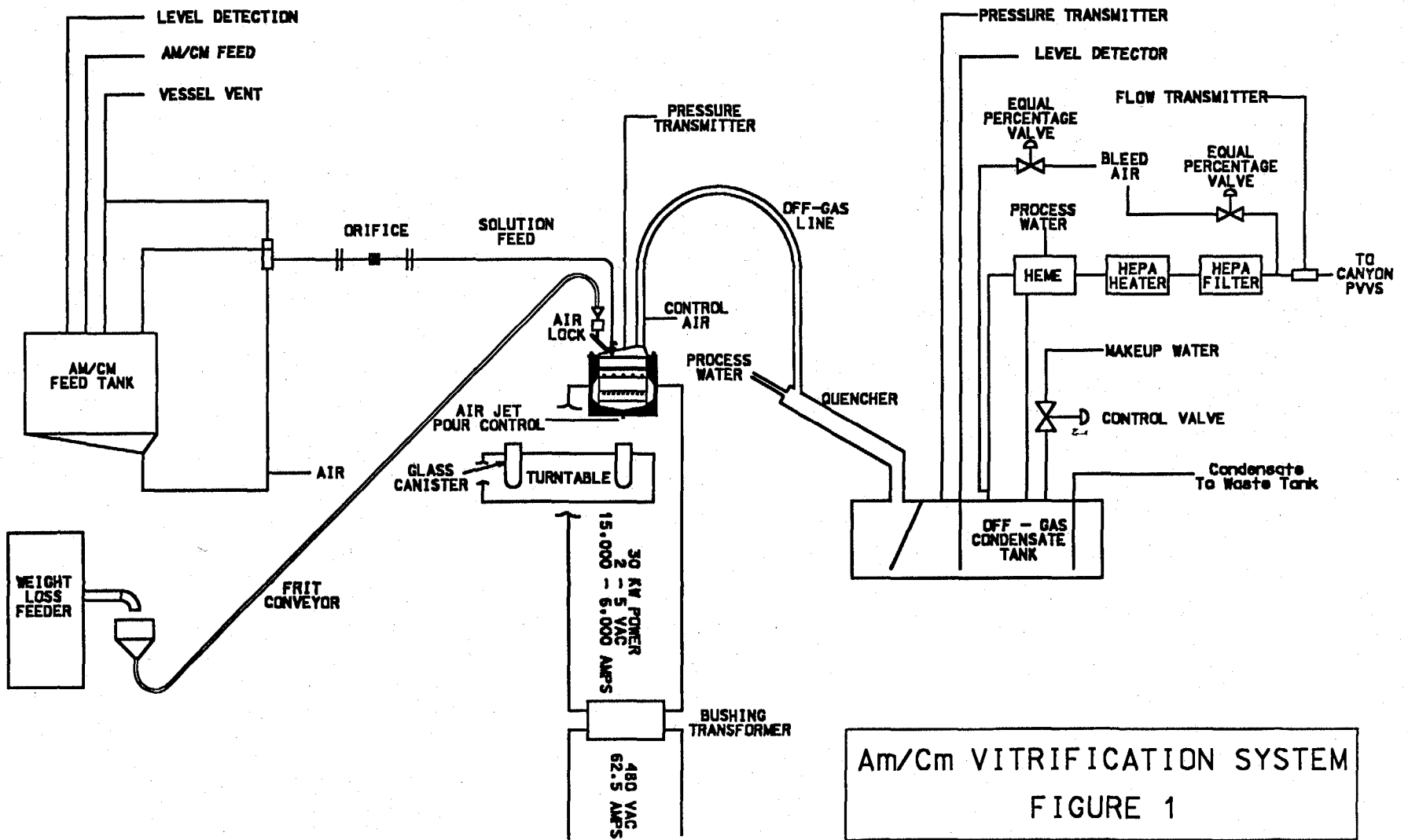
Process Parameter	Am/Cm Glass
HNO ₃ feed solution concentration	1 normal
Feed Solids Content (as oxides)	100 gm/l
Maximum Actinide Oxide in Glass	42 wt %
Nominal Glass Production	1 kg/hr
Maximum Glass Production	2 kg/hr
Maximum Off-Gas Flow (includes control air)	200 kg/hr
Melter Power Required	~30 KVA

TABLE II. TARGET GLASS COMPOSITION

Constituent	Weight %
SiO ₂	21 - 30
B ₂ O ₃	5 - 8
BaO	2
Al ₂ O ₃	3 - 13
PbO	8 - 14
Lanthanides & Actinides	Residual

TABLE III. TARGET SURROGATE GLASS COMPOSITION

Constituent	Weight %
SiO ₂	29.0
B ₂ O ₃	6.6
BaO	2.3
Al ₂ O ₃	12.4
La ₂ O ₃	24.7
Nd ₂ O ₃	6.8
Eu ₂ O ₃	1.6
CeO ₂	3.1
PbO	13.3



Am/Cm VITRIFICATION SYSTEM
FIGURE 1