PERFORMANCE OF A LARGE-SCALE MELTER AND OFF-GAS SYSTEM UTILIZING SIMULATED SRP DWPF WASTE

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ABSTRACT

The Department of Energy and the DuPont Company have begun construction of a Defense Waste Processing Facility to immobilize radioactive waste now stored as liquids at the Department of Energy's Savannah River Plant. The immobilization process solidifies waste sludge by vitrification into a leach resistant borosilicate glass. Development of the process has been the responsibility of the Savannah River Laboratory. As part of the development, two large-scale glass melter systems have been designed and operated with simulated waste. Experimental data from these operations show that process requirements will be met.

BACKGROUND AND INTRODUCTION

The Savannah River Plant (SRP) has been operating for nearly thirty years producing nuclear materials for defense, space, and medical applications. The site spans approximately 300 square miles and is located along the Savannah River near Aiken, South Carolina. Included on the plant site are three production reactors, two chemical separations facilities, and a fuel fabrications facility.

Wastes produced from chemical separation processes have accummulated and are currently being stored in large specially designed underground tanks as a neutralized liquid containing sludge and soluble salts. The sludge consists primarily of hydroxides and hydrous oxides of iron, aluminum, and manganese. The primary radioactive species in the waste are strontium-90 and cesium-137.

Since 1974 the Savannah River Laboratory (SRL) has been developing a process to immobilize the waste sludge by vitrification.[1,2,3] In the reference process the waste sludge is pretreated to remove excess aluminum, soluble salts, and mercury. Glass-forming chemicals are then added as a premelted borosilicate glass frit, and the resulting slurry is fed to a continuous Joule-heated ceramic melter operating at 1150°C. Evolving effluents (mainly steam) are treated by an extensive off-gas treatment system which recycles contaminated solids back to the process. The solid portion of the slurry melts into a molten pool and is poured into 0.6-meter diameter by 3-meter tall stainless steel canisters. Before interim storage the canisters are decontaminated and then sealed shut by welding a 13-centimeter cap to the nozzle.

Early development work involved small-scale equipment. A large portion of recent development efforts have focused around a large-scale vitrification pilot plant that has been in operation since August of 1980 using simulated radioactive waste. Two separate, but similar, large-scale melter systems have been operated. Pilot plant campaigns have lasted up to 63 days and have demonstrated the slurry-fed vitrification process for the future Defense Waste Processing Facility (DWPF). Design differences between the two melter systems have followed a progressive path toward attaining the final design of the DWPF melter system. A final large-scale pilot system, which is a 40% scaled version of the DWPF design, is scheduled to begin operation in late 1984.

With the majority of the development effort complete, initial construction of the DWPF began in 1983. Operation of the facility with actual SRP waste will begin in 1989.

LARGE SCALE MELTER PERFORMANCE

Two large-scale melter systems have been tested since August of 1980 at SRL's Equipment Test Facility (ETF). Approximately 300 tons of simulated waste glass (Table I) have been produced. Simulated waste glass performs similarly to actual DWPF glass and consists of similiar chemical composition with non-radioactive isotopes substituted for key radioactive species. The two melters are identified as the Project 1941 and the Large Slurry Fed Melter (LSFM). Fourteen slurry-fed campaigns lasting up to 63 days have been completed between the two melter systems. The 1941 melter was initially dry-fed by calcining the waste prior to melting. However, substantial cost savings were realized by elimininating the calciner and feeding a waste/frit slurry directly to the melter. Melt flux rates on both melters have exceeded the DWPF reference of 39 kg/hr·m^2.

TABLE I

SRL Large Scale Melter Experience
Kessler and Randall

Melter Identified	Duration	Slurry-Fed Campaigns	Total Glass Production* metric tons
1941	8/80-9/81	3	73
LSFM	1/82-2/84	11	> 180

* Majority produced by slurry-feeding

A majority of SRL's slurry-feeding experience has been with the LSFM system. The LSFM design is shown in Fig. 1. This melter is octagonal with a 1.1 square meter melt surface area (43% of full scale). Main containment refractory material is Monofrax® K-3 (trademark of the Carborundum Company). Other refractory type materials surround the K-3 and provide good

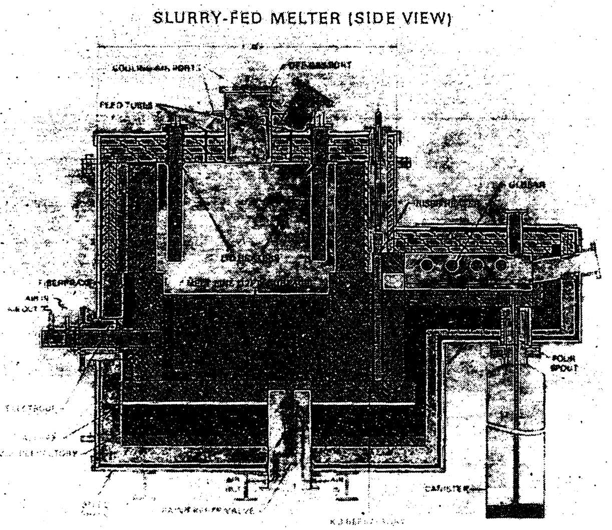

Fig. 1. Large Slurry-Fed Melter

thermal insulation, especially below the melt pool where sufficient insulation is critical for maintaining constant melt pool temperature. Joule heat is supplied to the pool by passing current between two pair of flat plate electrodes made of Inconel® 690 (trademark of Huntington Alloys, Inc.) One electrode pair contains "feet" attached at the bottom to divert energy toward the floor of the melter. This feature provides some ability to vary vertical power distribution within the melt pool. Energy can also be applied above the melt pool by silicon carbide resistant heaters (lid heaters) which are encased in protective Inconel® 690. Lid heaters are used primarily to help vaporize slurry and increase melt flux. They are also necessary for melter start-up to bring the melt body to sufficient temperature to initiate Joule heating with the electrodes. Under normal slurry-fed operation the lid heaters supply sufficient heat to maintain the melter plenum temperature at 650-800°C. Within this range the desired melt flux of 39 kg/hr.m^2 can be obtained.

Other heating systems are also used on the LSFM. While the actual melting of solid material occurs only in the main melt chamber, the molten glass must remain hot until after being poured into stainless steel canisters. To accomplish this task heating systems exist in the riser, trough, and pour spout sections of the melter. Joule heating between 2-1/2-inch Inconel® 690 bars is used in the riser section while resistance heaters are used in both the trough and pour spout areas. A final heating system exists beneath the melter on the drain valve. A glass plug is frozen in the drain valve by air cooling during normal operation, and heat is applied only to drain the melt cavity contents in an emergency situation or for shutdown.

Design differences between the earlier 1941 melter and the LSFM are small but still significant. First, rod electrodes entering the lid of the 1941 melter were used instead of side entering flat plates. Second, Monofrax® K-3 refractory material was used singly rather than a composite refractory construction. Third, the melter shell was water cooled while the LSFM is not. Finally, the 1941 melter design included a slanted Inconel® 690 resistance heated riser.

SRL's slurry-fed operating experience has affected melter design in several ways. During operation of the 1941 melter a 7-inch slag or spinel layer formed on the melter floor because the melter bottom temperature was below the liquidus temperature of the glass. Slag is undesirable because it reduces melter volume (and thus glass residence time) and may also affect glass quality by entraining spinel crystals in the final glass product. Spinel formation was virtually eliminated with the LSFM by incorporating a composite refractory design that has a relatively high thermal resistance to maintain uniform melt temperatures. The DWPF melter also contains composite refractory construction.

Melter electrode configuration has changed also. Optimal electrode configuration incorporates: 1) side entering buss bars (versus top entering), 2) large electrode surface area to reduce current density and thus corrosion, and 3) the ability to vary vertical power disposition within the melt pool. To satisfy all of the criteria above the DWPF melter design consists of 2 pair of side entering flat plate electrodes located with one pair directly above the other. Large flat plates provide ample surface area, and the over-under electrode pair placement allows flexibility for supplying power where it is needed within the melt pool. SRL's melter electrode designs have progressed to the current design of the DWPF melter.

Glass contact materials have performed well on both the 1941 and the LSFM. Measured corrosion rates for the 1941 melter were 0.5 centimeters per year for Inconel® 690 and 2.5 centimeters per year for Monofrax® K-3. Consultants from the glass and refractory industries confirmed that the refractories would easily last the required two years. Only the throat area has exhibited significant corrosion. The throat area was constructed of Monofrax® K-3. The DWPF design includes an Inconel® 690 lined throat and riser to reduce corrosion significantly in that area. Resistance heated Inconel® 690 has also been chosen for the lid heaters. Silicon carbide resistance heaters, used for both the 1941 and LSFM, have proven to be too fragile for DWPF operation.

Currently, SRL is involved in the construction of a third large-scale melter system, designated the Scale Melter (Fig. 2). Start-up is scheduled for late 1984. The Scale Melter is a 40% scaled version of the DWPF melter (1.0 square meter surface area versus 2.6 square meters). The Scale Melter design includes:

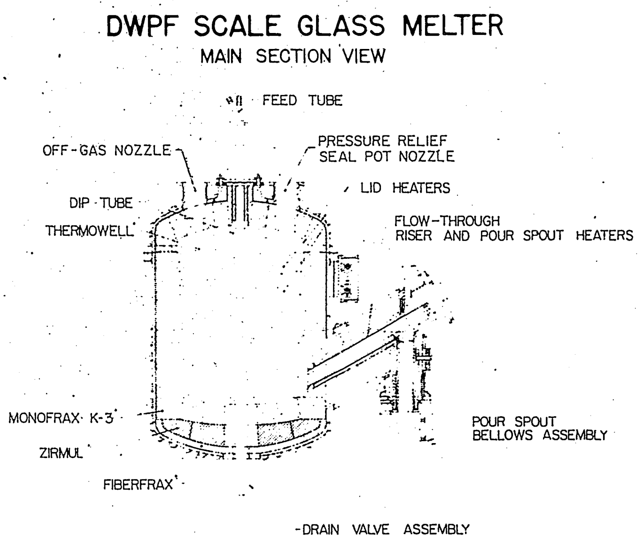

Fig. 2. Scale Melter

1) over-under side entering Inconel® 690 electrodes, 2) composite refractory construction, 3) Inconel® 690 lined, and resistance heated, riser and pour spout, and 4) Inconel® 690 resistance heated lid heaters. Operation of the Scale Melter will further demonstrate the slurry-fed melting process for the DWPF melter.

OFF-GAS SYSTEM PERFORMANCE

Process Requirements

The principal requirement of the DWPF off-gas system is removal of particulate contaminants from melter off-gases before they are exhausted to the atmosphere. In tests using non-radioactive isotopes, cesium has been found to be the radioactive contaminant of greatest concern. On a curie basis, it is forecast to be the most abundant element in the off-gas (Table II). In addition, it is present primarily as a submicron particulate (Fig. 3) making it one of the most difficult contaminants to remove. For these reasons, required overall off-gas system decontamination performance is being specified in terms of an allowable cesium release to the atmosphere. A yearly limit of 3 mCi Cs-137 has been established as a design basis. This translates to a required cesium DF for the DWPF melter off-gas system of 8 x 10^8.

TABLE II

Major Radionuclides in Untreated Melter Off-Gas
Kessler and Randall

	Ci/kg glass produced
Cs-137	2.75
SR-90	0.269
Ru-106	0.202
Pm-147	0.148
Ce-144	0.0602
Cs-134	0.0285
Pu-241	0.0070

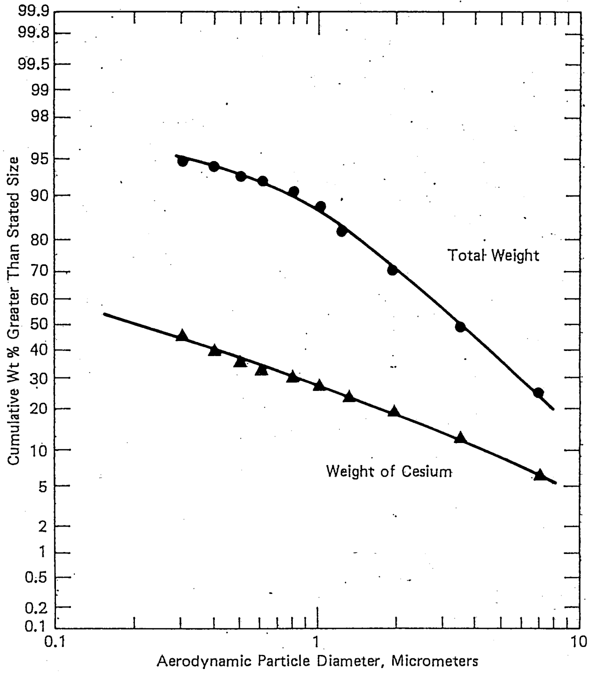

Fig. 3. Size Distribution of Untreated Melter Off-Gas Particulates

Additional requirements of the off-gas system are removal of elemental (non-radioactive) mercury, maintenance of a controlled negative pressure in the melter in the presence of surges in off-gas generation rate, and dilution of hydrogen and carbon monoxide generated in the melting process to below the lower explosive limit. Principle design bases for the off-gas system are summarized in Table III.

TABLE III

Principle Off-Gas System Design Bases
Kessler and Randall

Maximum Atmospheric Releases*	
Cesium-137	3-mCi/yr
Mercury	45.4 kg/yr
Melter Pressure Control	Maintain melter vacuum during 7X off-gas surge
H_2 and CO Concentration	70% of composite LEL during 3X H2-CO surge

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* Total DWPF release limit. Melter off-gas is primary contributor.

A particular problem associated with the off-gas system has been pluggage of the off-gas line exiting the melter with entrained particulates and droplets of molten glass and slurry as large as 2 millimeters in diameter. This problem has led to specification of a maximum temperature of 400°C in the pipeline exiting the melter to prevent sticking of hot particles, and development of a device called an Off-Gas Film Cooler (OGFC) to cool the entering gases and provide a protective clean gas boundary layer at the entrance.

System Description

The large-scale off-gas system associated with the LSFM is shown diagramatically in Fig. 4. This system is identical to the current DWPF design except the DWPF includes tandem (2-stage) High Efficiency Particulate Air (HEPA) filters and exhausts off-gases from the melter through an underground sand filter along with process vessel and building ventilation air.

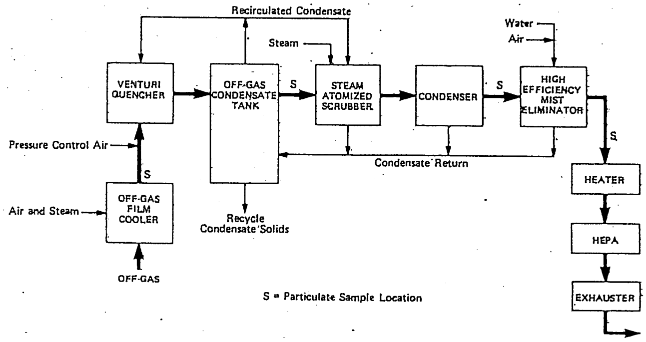

Fig. 4. Flow Diagram of the Large-Scale Off-Gas System at the Equipment Test Facility

The LSFM system consists of seven active components and a condensate receiving tank.

- Off-Gas Film Cooler (OGFC) and Brush Reamer Assembly
- Quencher

- Off-Gas Condensate Tank (OGCT)
- Two-stage Steam Atomized Scrubber (SAS)
- Condenser
- High Efficiency Mist Eliminator (HEME)
- Preheater and High Efficiency Particulate Air (HEPA) Filter Assembly
- Exhauster

In operation, off-gases at 700-750°C and particulates enter the OFGC (Fig. 5) where a mixture of air and steam is injected to form a cool protective boundary layer over the first few diameters of the off-gas line. The air to steam ratio is adjusted to cool the off-gases to 350-400°C and provide the required amount of dilution air. The quencher, a conventional jet venturi fume scrubber, removes most of the water vapor in the incoming off-gas and conditions it for high efficiency scrubbing by cooling to 40-45°C by direct contact with cooled recirculated condensate.

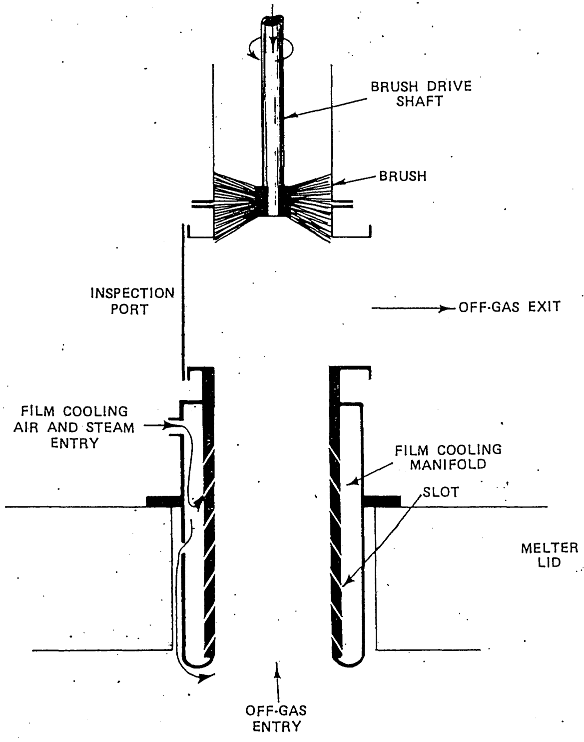

Fig. 5. Experimental Off-Gas Film Cooler with Brush Reamer

The quencher is located as near the melter as possible (about 6-m) to minimize the length of line subject to pluggage. The average velocity in this line is maintained at or above 15-m/s to prevent particle deposition. Off-gas velocity is measured by an S-type pitot tube. A variable quantity of additional air is injected immediately upstream of the quencher for melter pressure control.

Off-gases and condensate from the quencher pass through the Off-Gas Condensate Tank (OGCT) where the liquid and gas are separated and the gas routed to a high efficiency scrubber called a Steam Atomized Scrubber (SAS). The SAS consists of two stages of steam-driven, free jet Hydro-Sonic® Scrubbers (trademark of Hydro-Sonic Systems Company) (Fig. 6). Scrubbing is accomplished by recirculated condensate that has been finely atomized by a super-sonic steam jet. The condensate droplets are coalesced and then collected in a conventional reversing cyclone.

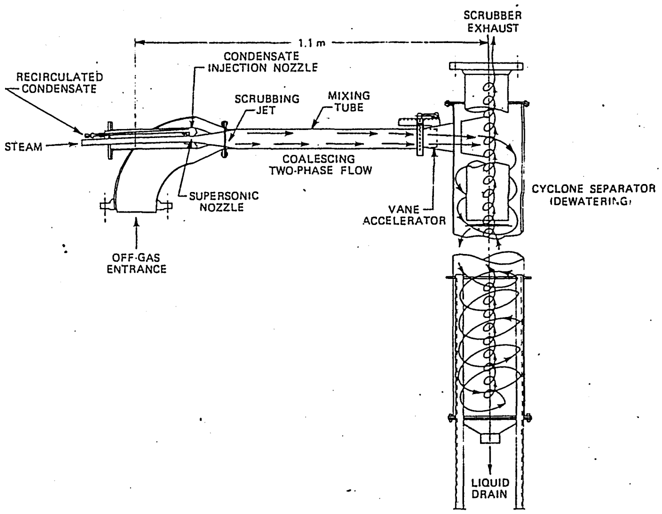

Fig. 6. Experimental Steam Atomized Scrubber (Single Stage)

Gases exit the SAS at 60-65°C. They are cooled to 10°C in a shell and tube heat exhcanger to condense and remove elemental mercury before being passed to the High Efficiency Mist Eliminator (HEME).

The HEME consists of a 7.6-cm thick cylindrical element of nominal 10-micrometer-diameter fiberglass packed to a density of 190-kg/m^3 (Fig. 7). Entering off-gas is assured to be wet as it passes through the element by spraying a small quantity of filtered, deionized water into the gas stream as it enters. Soluble contaminants removed from the off-gas are thereby washed from the element and returned to the condensate tank. The unit is sized for a 0.025-m/s superficial face velocity.

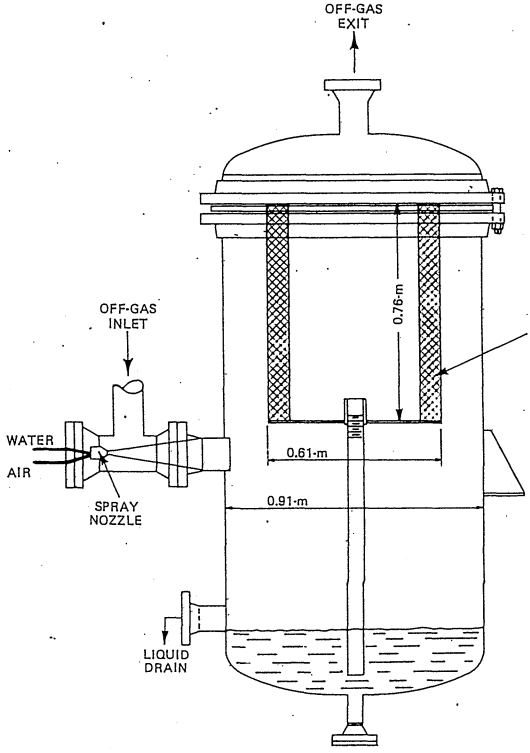

Fig. 7. Experimental High Efficiency Mist Eliminator

Gases exiting the HEME pass through a heater and a single stage HEPA filter before being exhausted to the atmosphere by a positive displacement exhauster.

Sampling. Particulate samples are obtained at locations marked with a "S" in Fig. 4. Samples are taken by EPA Method 5 procedures. A Mark V University of Washington impactor is used to determine size distributions. Decontamination factors for cesium are determined by analysis of filter collected samples by neutron activation methods.

Bomb samples are taken to determine non-condensible gases such as H_2, CO, CO_2. Percent LEL is monitored on-line by redundant sensors.

Performance

Decontamination. Decontamination factors (DF) for cesium for prototypical DWPF gas cleaning equipment items except the HEPA and sand filters have been measured on the integrated large-scale system using actual melter off-gas. In all cases, measured values exceed goal values (Table IV). Although HEPA and sand filter DFs have not yet been measured in an integrated melter/off-gas system, the technology is well established and goal DFs are shown to be conservative by work on other systems.[4,5,6] Assuming goal DFs for the HEPA and sand filters are realized, the overall forecast release of cesium-137 from the melter is 0.13-mCi/yr, a factor of 24 below the requirement of 3-mCi/yr.

TABLE IV

Off-Gas Decontamination Factors For Cesium
Kessler and Randall

	Goal	Demonstrated
Melter	10	40
Quencher	1	2
Steam Atomized Scrubber	50	60
High Efficiency Mist Eliminator	40	100
Tandem (HEPA)	2000	-
Sand Filter	200	-

Pressure Control. A primary goal of the large-scale off-gas program has been characterization of off-gas surges to enable adequate design of the DWPF off-gas system. Surges from the LSFM as high as seven times the calculated steady-state process off-gas rate have been measured. The magnitude and frequency of surges, which are primarily steam, have been found to depend heavily on feed composition and feed rate. Surge distribution data from the recent 63-day run at reference melt flux was extrapolated using an exponential statistical model. Surges greater than 7X were predicted to occur once every 200 days while surges greater than 10X were predicted once every 17 years (Fig. 8).

The rise in melter plenum pressure associated with the larger surges experienced on the LSFM has been less than 75-mm water. A computer simulation of the reference DWPF off-gas design is being used to assure the system will maintain a negative melter pressure during a design basis (7X) surge.

Prevention of Pluggage. The problem of pluggage of the entrance to the off-gas line has been solved by development of the Off-Gas Film Cooler and Brush Reamer. Prior to the addition of this type of cooling, pluggage of the off-gas line by a partially molten mass of desposits could be expected to occur on the order of once every 1 to 3 days. Since the OGFC was installed, no difficult-to-remove deposits have formed.

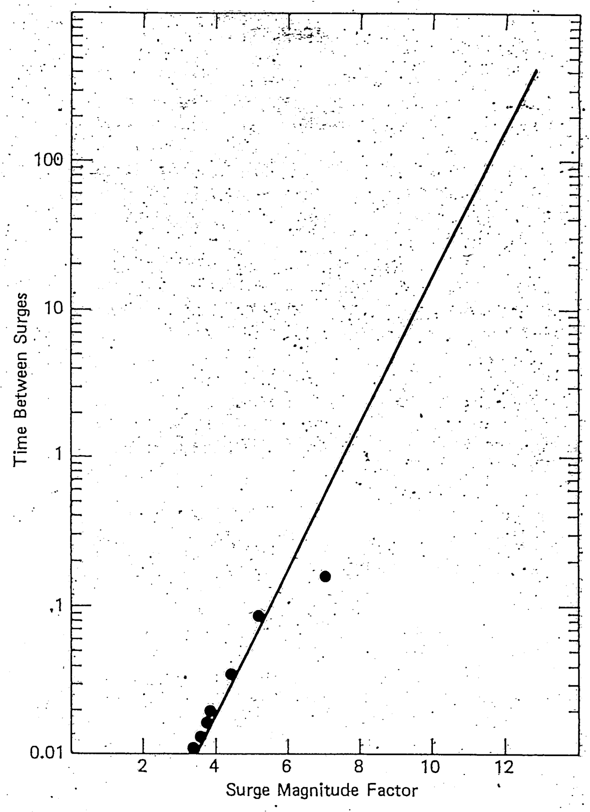

Fig. 8. Occurrence of Off-Gas Surges from LSFM

The key to the success of the OGFC design is cooling of the pipe wall by an internal flow of clean gas. Many of the large droplets of molten glass and slurry that are ejected from the melt surface are moving at velocities greater than 4-m/s when they enter the off-gas line. While many of these penetrate the clean gas boundary and stick to the wall of the OGFC, the internal cooling keeps the accumulating deposit cool maintaining it in a friable condition so it can be easily removed with a brush. The LSFM was run for 30 days during the recent 63-day run without cleaning. When necessary, cleaning is accomplished easily, generally being done without stopping melter feed. A remotely operated brush reamer is now being designed.

CONCLUSIONS

In the three years the large-scale melter/off-gas system has been in operation, several design concepts have been tested. The result of these tests and other tests on small scale equipment has been the definition of a workable process to vitrify SRP high level radioactive waste. This process has been demonstrated in long-term operation on the large-scale integrated system at the ETF showing that a good quality glass product can be produced while meeting process requirements for melt rate, melter lifetime, and atmospheric releases.

ACKNOWLEDGEMENT

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