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by

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F. Houston Brown, Chris T. Randall, Marion B. Cosper, and Jon P. Moseley

Savannah River Laboratory
E. I. du Pont de Nemours & Co.
Aiken, South Carolina 29808

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F. Houston Brown, Chris T. Randall, Marion B. Cosper, and Jon P. Moseley
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ABSTRACT

The Department of Energy, with recommendations from the Du Pont Company, is proposing that a Defense Waste Processing Facility be constructed at the Savannah River Plant to immobilize radioactive waste. The immobilization process[1] is designed around the solidification of waste sludge in borosilicate glass. The Savannah River Laboratory, who is responsible for the solidification process development program, has completed an experimental program with one large-scale glass melter and just started up another melter. Experimental data indicate that process requirements can easily be met with the current design.

INTRODUCTION

At the request of the U. S. Atomic Energy Commission the Du Pont Company constructed the Savannah River Plant (SRP) in the early 1950's, and the plant is still operated by the Company for the U. S. Department of Energy. The plant is comprised of a large, remote land area in South Carolina with extensive support facilities and a single operating contractor. SRP is presently the nation's primary source of tritium, weapons plutonium, heat source plutonium, and several other radionuclides for defense, space, medical, and energy applications.

The radioisotopes are produced in three nuclear reactors by irradiating appropriate target materials with neutrons from uranium fuels. The irradiated products are chemically separated in one of two remotely operated canyon buildings. In the approximately 30 years of operation, the chemical separation process has generated 72 million gallons of high level waste, and waste is expected to be generated

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continually at the rate of 1.5 million to 4 million gallons a year depending on operating schedules. The waste has been evaporated to about 25 million gallons which is now stored in large carbon steel, underground tanks. The principle radionuclides in the waste are strontium-90 and cesium-137. Approximately 10% of the waste is a sludge in the form of iron, manganese, and alumium hydroxides; the sludge contains most of the strontium and small amounts of actinides not recovered in the reprocessing plants. The remainder of the waste is liquid and salt cake, which contains primarily sodium nitrate, sodium nitrite, sodium aluminate, and sodium hydroxide; this waste fraction contains most of the cesium.

The Department of Energy, with recommendations from the Du Pont Company, is proposing that a Defense Waste Processing Facility (DWPF) be constructed at SRP to immobilize this large quantity of radioactive waste. The DWPF will provide a timely first step toward solving the problem of nuclear waste disposal and will assist in the development of a rational nuclear waste disposal policy for the United States.

The DWPF is ready for authorization of Construction in 1983 and can be in full production in 1988. The current plan is to implement the DWPF in two stages. The first stage will immobilize the sludge component of the waste which contains 60% of the total radioactivity in the high level waste and almost all of the long-lived radionuclides.[1] A salt processing facility (second stage) will remove cesium-137 and other radionuclides from the salt component of the waste and will immobilize the decontaminated salt in concrete. The radionuclides removed from the salt will be returned to the sludge processing facility, or packaged for use as a byproduct.[2]

The reference process is designed around the solidification of the waste sludge component in borosilicate glass. Initially the alkaline sludge is mixed with formic acid where the mercury components are reduced to the metal and removed. Glass frit is mixed with the formated sludge, and the 40 wt % solid slurry is delivered to the melter which is maintained at 1150°C. Water is evaporated and the sludge is dissolved into the glass. The molten glass is poured into a 2-foot diameter by 10-foot tall stainless steel canister. A five-inch diameter cap is welded into the nozzle of the canister, after which it is decontaminated and removed to an interim storage area. The off-gas from the melter is purified by scrubbing and filtering.

Presently, the Savannah River Laboratory is involved in research and development work in all areas of the DWPF process. Much of the effort is centered around the design, operation, and testing of the glass melter.

Savannah River Laboratory originally began vitrification research and development work in 1974. Melter development began in 1977 (Figure 1). Initial work was done on small laboratory sized,

melters. Since PNL had already been in waste vitrification development work for several years, SRL subcontracted them to begin simultaneous efforts with larger melters. The first SRL moderate size melter - Small Cylindrical Melter (SCM) with a one square foot surface area - started operation in 1978. The first large melter, Project 1941 melter or CMOG, was started in 1980, and the second generation large melter, Large Slurry Fed Melter (LSFM), started up in January of this year. This program is directed to the experimental operation of the first DWPF melter in 1984 and DWPF startup in 1988.

VITRIFICATION PROCESS

A slurry-fed, joule-heated ceramic melter has been chosen for DWPF design. The melter for DWPF is cylindrical with a 28 square foot surface area. The glass contact refractory is Monofrax K-3® (trademark of the Carborundum Company) and the outside stainless steel surface is water cooled. Water cooling serves two purposes - 1) it freezes the molten glass, which might have penetrated cracks and joints in the refractory, before it reaches the shell and grounds the electrodes, and 2) it reduces the thermal air currents in the process cell by maintaining the melter surface below 50°C. Thermal air currents can quickly spread radioactivity and grossly contaminate the process crane. The joule heating electrodes, as well as all other metal components placed inside the melter, are fabricated from Inconel 690® (trademark of Huntington Alloys, Inc.). Glass can be poured from the melter either by an overflow method or by creating a differential pressure between the main melt chamber and the pour trough. A bottom drain/freeze valve is provided to completely drain the contents of the melter.

In addition to the main electrodes, other heating systems are required inside the melter. Vapor space or lid heaters are required to bring the melt body to a sufficient temperature to initiate the joule effect within the melt pool and to restart the melter if power is lost during normal production. Also, the lid heaters will supply some of the energy required to evaporate the water from the slurry and raise the temperature of the cold cap during normal operation. Second, riser heaters, designed as joule heating elements, are required to maintain the glass in the riser at or near the bulk glass temperature. Third, a resistance heater is required in the pour spout to ensure that glass is not prematurely chilled before it enters the canister and flows freely to form a solid mass of glass. Last, the drain valve requires a heater to melt a glass plug within the valve so the contents of the melter will be free to drain. If the valve is also to be used as a freeze valve, the capability to add a cooling medium to the valve must be supplied. This would allow the valve to be cycled so only a portion of the melter contents would be drained at a time.

The slurry fed to the melter will consist of 28 wt % waste sludge and 72 wt % frit (on an oxide basis); total solids will be 40 wt %. As the slurry is introduced into the melter through a water cooled nozzle, it will cover most of the melt surface forming a cold cap. The cold cap will insulate the bulk glass, and also significantly reduce the amount of semivolatiles released from the melt surface. Slurry feeding has proven to be a reliable process.

The original concept was to dry the waste sludge before it was introduced into the melter. This was accomplished in an induction-heated calciner (Figure 2). The sludge was first atomized, dried by heating to 550-900°C, and mixed with frit. Off-gas from the calciner was exhausted through a set of sintered metal filters. The filters removed most of the entrained particulates and allowed the gas to pass into the off-gas system. The calciner, which was 30 feet tall, was removed from the DWPF design to save approximately $75 million. The height of the calciner controlled the height of the entire canyon building; thus, the total building height was reduced resulting in the cost savings.

FIRST GENERATION LARGE-SCALE MELTER

The first large melter operated by SRL, referred to as the Project 1941 melter (Figure 3), was very similar to the proposed DWPF melter. It had Monofrax K-3® refractory enclosed in a water-cooled, stainless steel shell. The melt surface area was 11.75 square feet. This melter was heated by rod-type electrodes instead of plates. The riser contained a resistant heater instead of joule heating elements.

The Project 1941 melter was maintained at a glass temperature of 1130°C for 398 days. During this period it was operated in both the originally designed calcine-fed mode and the modified slurry-fed mode. In either mode the DWPF reference melt flux rate was easily achieved (Table 1).

One of the major objectives of the experimental program was to characterize the corrosion rates of the materials which contact glass. The Inconel 690® electrodes and thermowells exhibited an average corrosion rate of 0.0005 inches per day. This rate agreed with earlier data if the idling and running periods are considered separately. The melter was idled 90% of the time, thus: Idle Corrosion (90%) + Run Time Corrosion (10%) = Total Corrosion

3/8 mil/day (.9) + 1-1/4 mil/day (.1) = 1/2 mil/day

This correlation makes the assumption that the melter is maintained at approximately 1150°C. The general design concept is that all Inconel®

components except thermowells are designed to last at least 2 years, and this concept was proven to be viable.

The Monofrax K-3® refractory was in exceptionally fine shape after the experimental program. Several consultants from the glass and refractory industry confirmed that the refractories would easily last the required two years. The maximum measured corrosion rate was 1 inch per year, about half the predicted rate. As expected, the maximum corrosion occurred in the throat. Since glass industry experts agree that the refractory throat is the limited life component of a melter, the DWPF design will incorporate an Inconel® lined refractory throat.

Approximately 7 inches of spinel or slag formed on the melter floor because the melter bottom temperature was below the liquidus temperature of the glass. This slag formation can lead to a reduced melter volume, which affects glass residence time, and may reduce glass quality by introducing crystals into the final glass product. Spinel can be formed in the melter by any of three methods: 1) corrosion products of the refractory, 2) incorrect feed composition, or 3) zones in the melter maintained at a temperature below the liquidus of glass. Method 1 above can not be eliminated; it can be reduced by carefully choosing the refractory glass composition, and operating temperature. Method 2 can be controlled by a good quality assurance program. Method 3, the major cause of the spinel in the Project 1941 melter is a function of the melter design. The Project 1941 melter had a bottom temperature of 850°C while the bulk glass was 1150°C (Figure 4). To eliminate this problem in future melters either or both of the following solutions can be utilized: 1) insulate the bottom so that the heat transfer coefficient is less than that through the sidewalls, and 2) design the electrode configuration so that the power distribution can be varied throughout the melt tank. The ability to vary the power distribution during the life of the melter provides the flexibility to adjust operating conditions to account for feed changes, such as high iron versus high aluminum, and to reduce power at the bottom if a higher electrically conductive deposit or slag layer forms.

SECOND GENERATION LARGE-SCALE MELTER

The LSFM has just been placed into service by SRL (Figure 5). It offers some different design concepts but is a progressive step in reaching the final melter design for DWPF. The LSFM has a surface area of 12 square feet, is octagonal in shape, and is heated by plate-type, joule heating elements. These electrodes offer some ability to vary the power distribution in that the two electrode pairs are of different heights, and one pair has "feet" attached to the bottom which increases the surface area thus increasing the current density in the floor of the melter. The walls of the melter are made from several different layers of refractory instead of a single layer as in the Project 1941

melter. The temperature drop across the various layers of refractory will stop the flow of glass that may have penetrated any cracks or joints in the glass-contact, refractory layer. The surface temperature of this type of melter will be higher than one where water cooling is used. Temperatures of 75° to 150°C can be expected.

The vertical riser is heated by two joule heating elements. Power is regulated by thermocouples embedded in the electrodes. Silicon carbide elements are used for supplemental heat in both the vapor space and pour trough. A water spray system is being installed to reduce the vapor space temperature during idle conditions.

Initial operating results from the melter are very favorable. The maximum temperature differential within the glass pool is only about 30°C, thus, ensuring that all areas in the melter are above the liquidus of the glass and reducing the potential for spinel formation. These results compare well with data obtained from a scaled physical model of the LSFM.

Physical modeling has proven to be a valuable design and experimental tool. Initial results indicate that model temperature profiles agree closely with actual measured temperatures in the melter (Figure 6). Molten glass is modeled by a lithium chloride solution enabling the physical model to be operated at temperatures below 100°C. The walls are made from acrylic which are surrounded by temperature controlled water baths which enable the melter boundary conditions to be changed (Figure 7). Under these conditions it has proven to be much easier to measure temperature profiles, voltage and current distributions, and thermal convection currents within the glass pool.

The LSFM will be operated for about 18 to 24 months. The main objectives of the experimental program are to (1) evaluate the effect of feed composition, especially formated feeds, on melter performance, (2) support off-gas studies, (3) evaluate new design concepts, and (4) determine process variables which may affect the operation of the DWPF melter.

CONCLUSION

The technology and engineering are ready for construction of the DWPF for immobilizing high level radioactive waste at SRP. The development program underway at SRL has proven that all processes are viable and that DWPF can be safely operated with no danger to personnel or the public. Proceeding with this project will provide the public, and leadership of this country, with a crucial demonstration that a major quantity of existing high level waste can be safely and permanently immobilized. Early demonstration will both expedite and facilitate

rational decision making on this important aspect of the national nuclear program.

REFERENCES

1. "Preliminary Technical Data Summary for the Defense Waste Processing Facility, Stage 1," DPSTD-80-38-2, September 1981.

2. R. Maher, et al, "Solidification of Savannah River Plant High Level Waste," Paper for presentation at the American Nuclear Society, December 4, 1981.

TABLE 1. Summary of Project 1941 Melter Experimental Program

	Calcine-Fed	Slurry Fed
Total Operational Time (Months)	10	4
Experimental Runs	7	3
Days of Operation	34	21
Glass Produced (Tons)	54	20
Production Rates (lb/hr-ft^2):		
Without Supplemental Heat	20	6.4
With Supplemental Heat	35	11.4

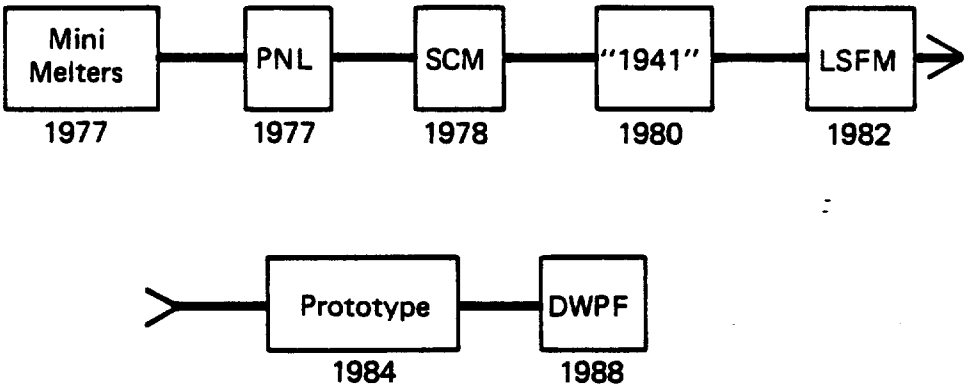

FIGURE 1. DWPF Melter Development

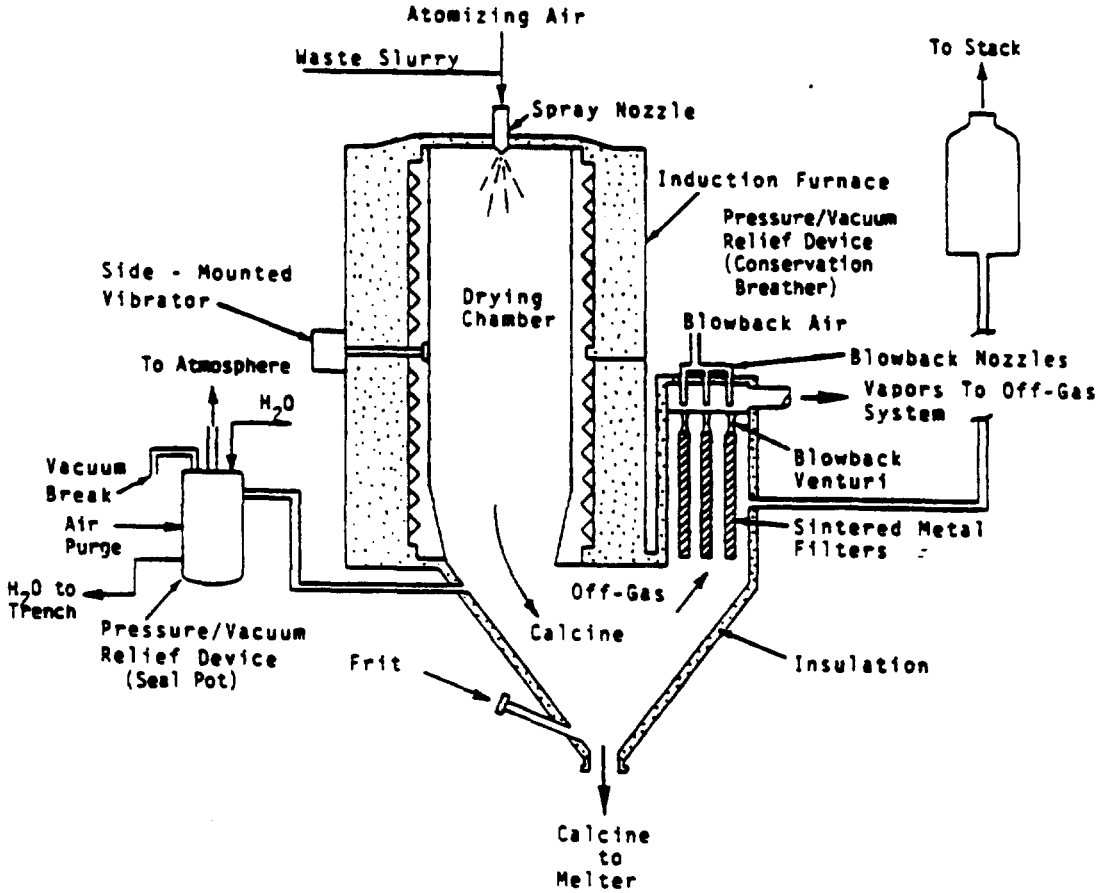

FIGURE 2. Development Spray Calciner

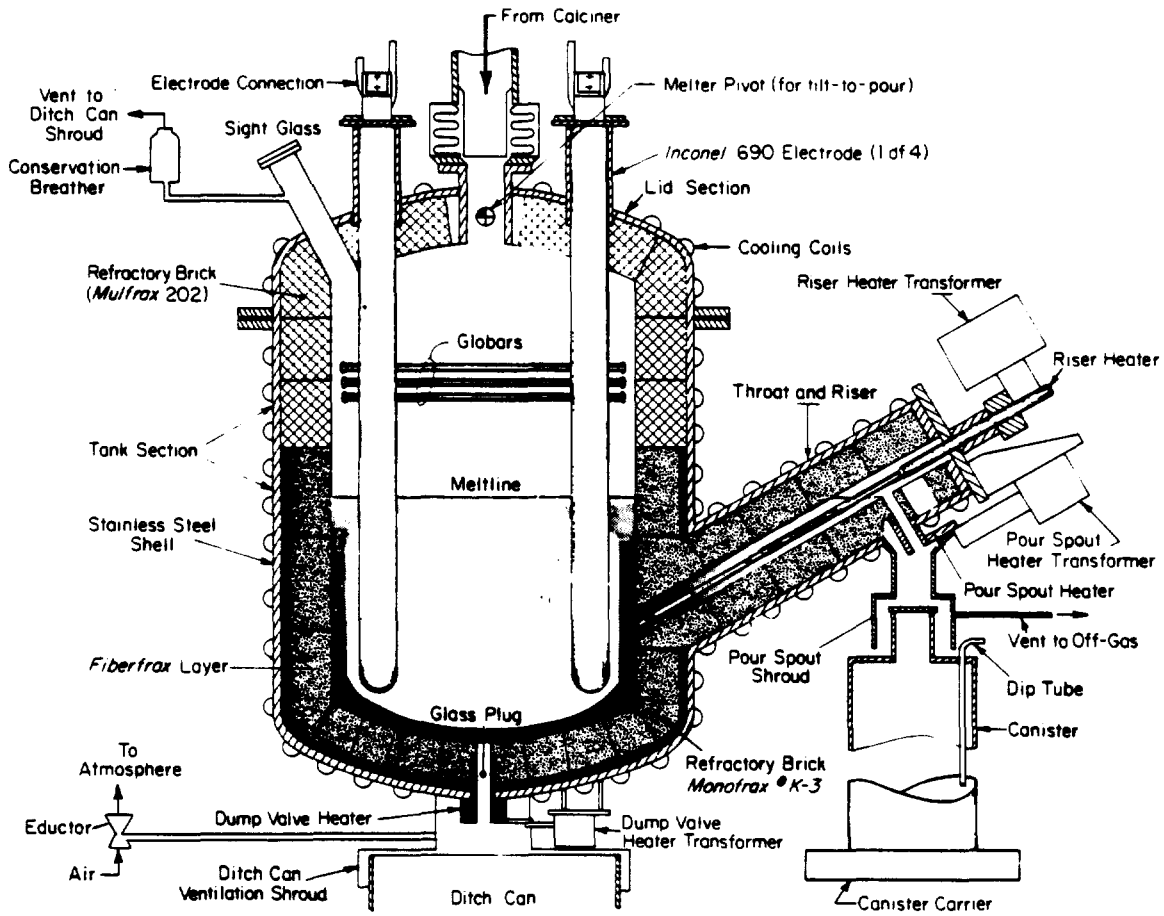

FIGURE 3. Large-Scale Melter (Project 1941)

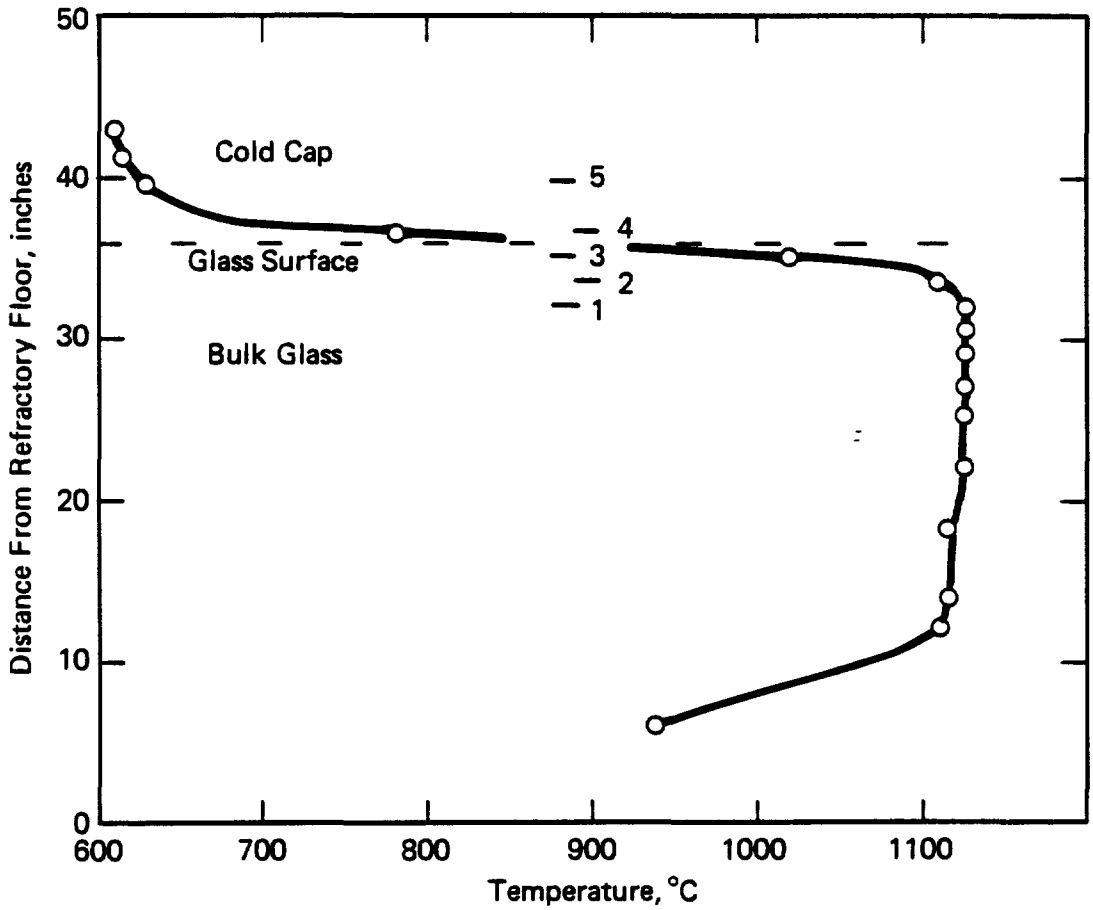

FIGURE 4. Axial Temperature Profile for Project 1941 Melter. Melt flux = 7.9 lb/hr-ft^2.

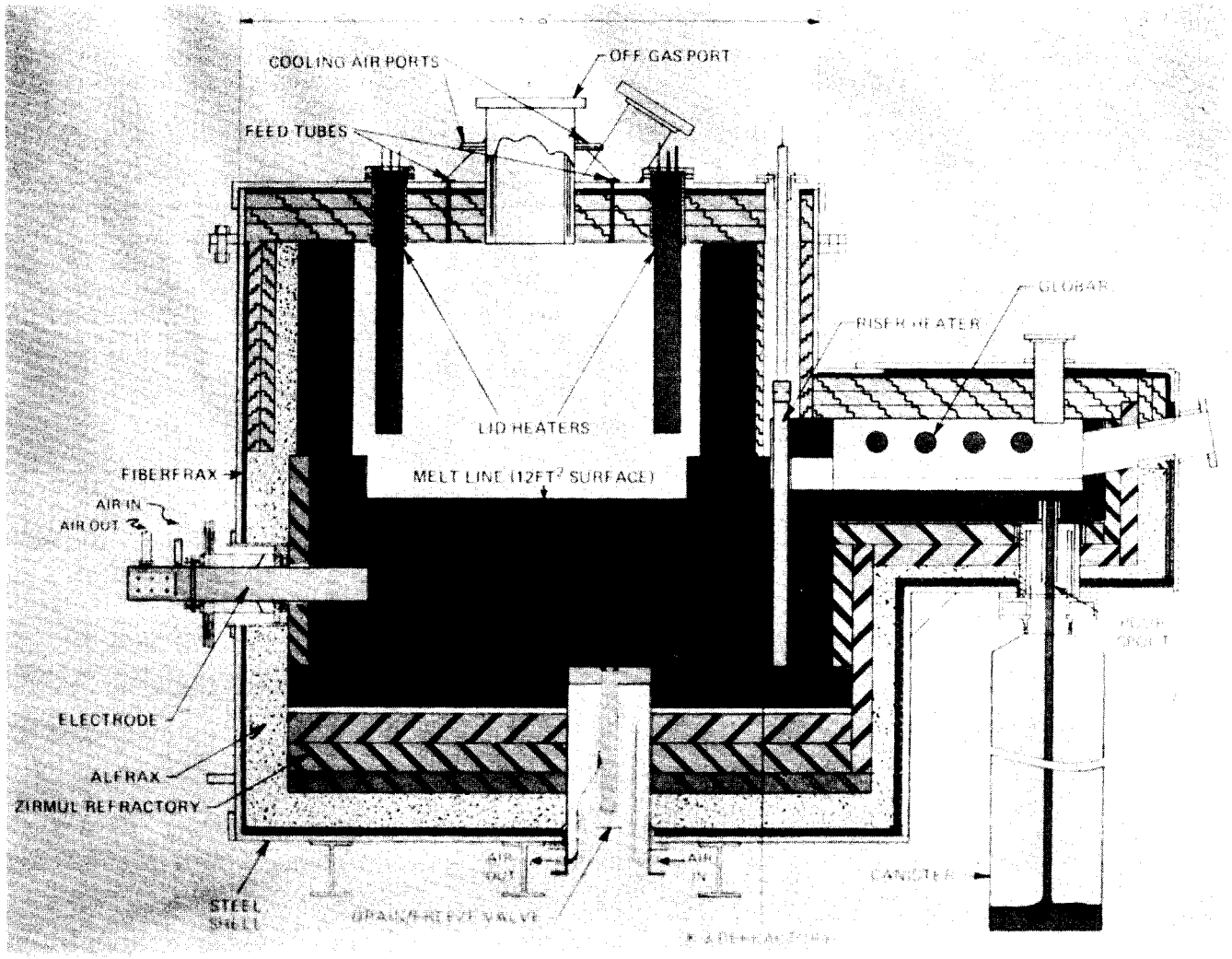

FIGURE 5. SRL Second Generation, Large-Scale Melter (LSFM)

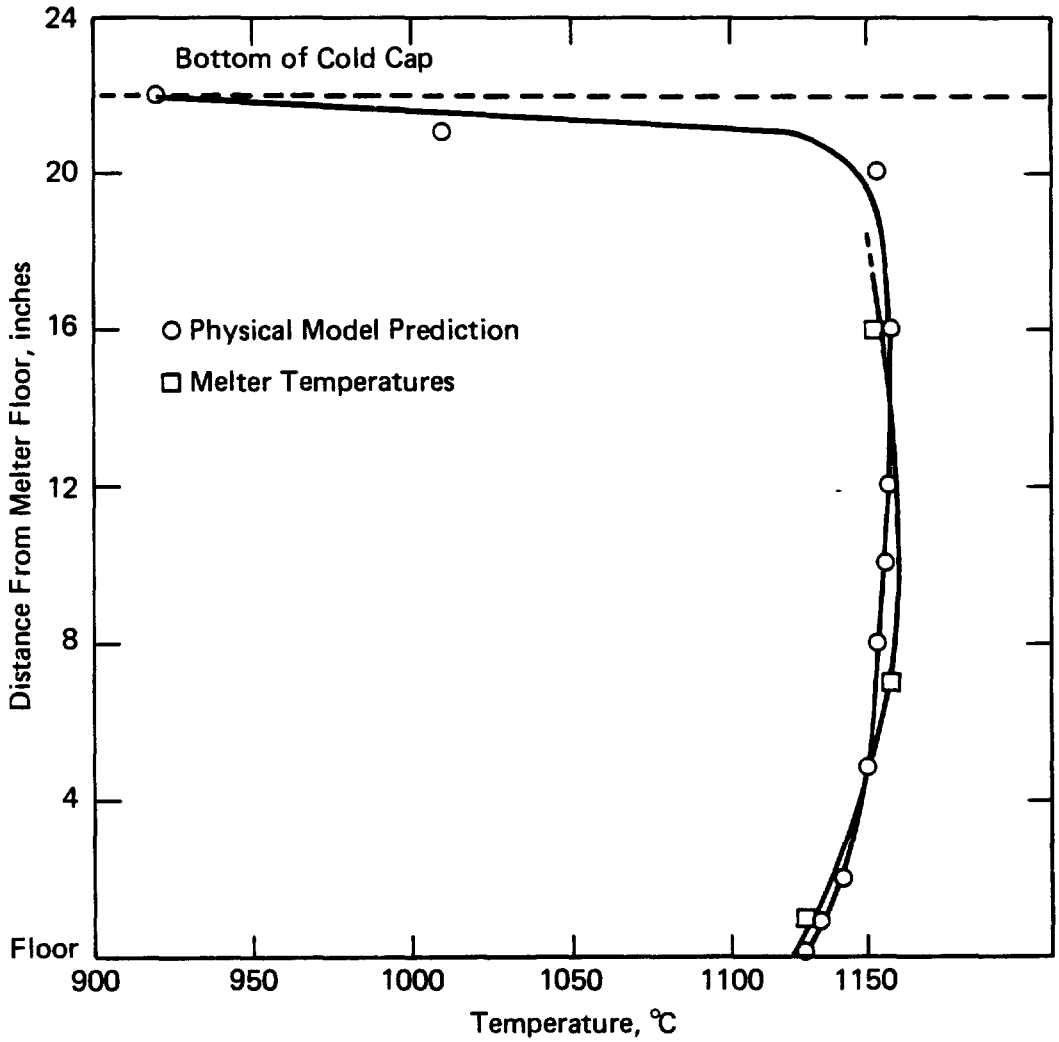

FIGURE 6. Comparison of Temperature Profile Between LSFM and Physical Model

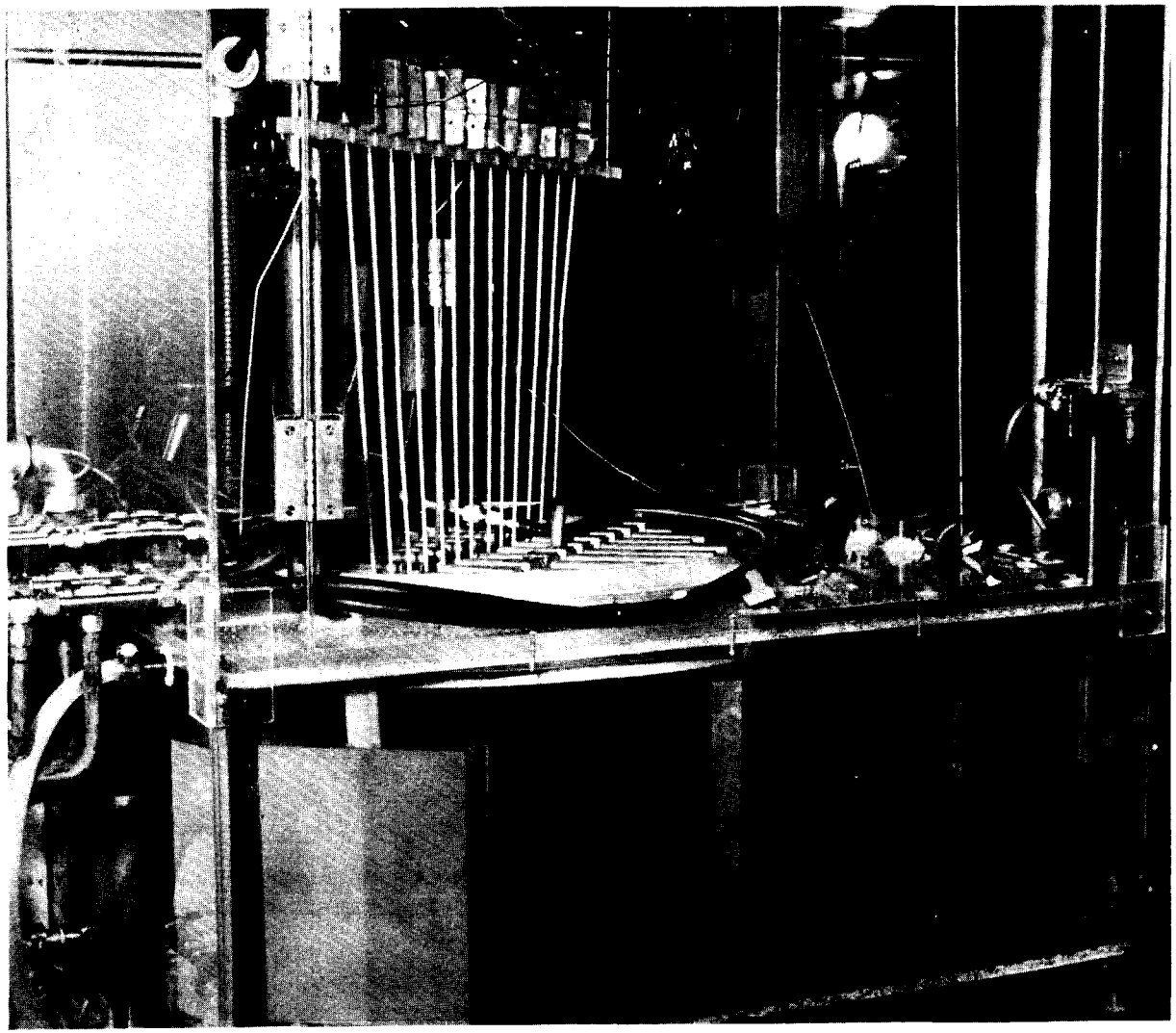

FIGURE 7. Physical Model for LSFM