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Experience with a Joule-Heated Ceramic Melter While Converting Simulated High-Level Waste to Glass

by
C. C. Chapman

August 1976

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Battelle
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C. C. Chapman

August 1976

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Richland, Washington 99352

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EXPERIENCE WITH A JOULE-HEATED CERAMIC MELTER WHILE CONVERTING SIMULATED HIGH-LEVEL WASTE TO GLASS

C. C. Chapman

SUMMARY

Development of a joule-heated ceramic melter, sponsored by the Energy Research and Development Administration, has been progressing for nearly 3 years. In January 1975, a ceramic-lined, direct joule-heated glass melter was started up and operated continuously for nearly 11 months. During this period, process testing was completed both while feeding simulated high-level waste calcine and while feeding simulated high-level liquid waste. While feeding waste calcine and frit, the unit was demonstrated at a production rate in excess of 45 kg of glass/hour, which meets the needs of a reference 5 MTU/day reprocessing plant. When the simulated liquid waste and frit slurry were fed to the system, a 25-liter/hr process rate was demonstrated. This capacity is equivalent to the needs of a 1.5 MTU/day reprocessing plant. Evaluation of the melter after 10.8 months of operation suggests that a melter life in excess of 2 years is likely.

The operation of the engineering-scale ceramic melter has been encouraging. The high capacity of the melter with the capability for direct liquid feeding and a long operating life suggests that a joule-heated ceramic melter will play a major role in future waste solidification processes.

INTRODUCTION

There has been a continuing effort to convert high-level radioactive waste to a durable glass since the early 1950's in the United States and throughout the world. Several melters have been developed at Battelle, Pacific Northwest Laboratories (PNL).(1) A platinum melter was the primary melting technology that was evaluated during the Waste Solidification Engineering Prototype program(2) conducted at PNL. A less expensive metallic melter was also evaluated at the beginning of the Waste Fixation Program. This melter was made of a nickel-chrome alloy, but its operating life was somewhat limited. The current melting process is the In-Can Melter which assures a long operating life, but limits the selection of waste glasses because of its somewhat low operating temperature (1050°C). A review of current commercial glass industry melting technology suggested that a joule-heated ceramic melter or an all electric melter could be adapted to high-level waste (HLW) vitrification. This type of melter promised to provide a long operating life system while melting high temperature glasses. Thus, a development program was initiated.

ENGINEERING-SCALE CERAMIC MELTER

MELTER DESCRIPTION

After gaining operating experience and understanding of system operation with small laboratory melters, an engineering-scale ceramic melter was designed and constructed. A schematic cutaway view of the assembled melter is given in Figure 1 while a plan and section view of the melter is given in Figure 2. The dimensions of this unit are:

- Melting cavity - 14 in. wide, 30 in. long and 12 in. deep. The glass depth was maintained at 6 in. by overflow drain pipe.
- Overflow cavity - 6 in. wide, 7 in. long and 12-in. deep
- Electrodes in the melting cavity - 2 in. thick, 12 in. wide, 11-1/2 in. high and were suspended from the top of the refractories.

The overflow pipe had a 2-in. outside diameter and 1/2 in. inside diameter. The drain pipe was located in the overflow cavity which was positioned adjacent to, and centered on, one sidewall of the melting cavity. The overflow cavity and the melting cavity were powered by independent sets of electrodes. The overflow electrode was also 2 in. thick and was suspended from the top of the refractories. The bottom of this electrode was 2 in. above the floor. This allowed molten glass to flow from the melting cavity to the overflow cavity where it could flow out the drain pipe.

The glass contact refractory was Monofrax® E which is a 3-in. thick fused cast, chrome-spinel material composed primarily of chrome oxide (∿78%). These glass contact refractories were backed with zircon bricks also 3 in. thick.

Water-cooled stainless steel plates were placed on the outside and bottom of the refractories. The refractories were constrained by compression

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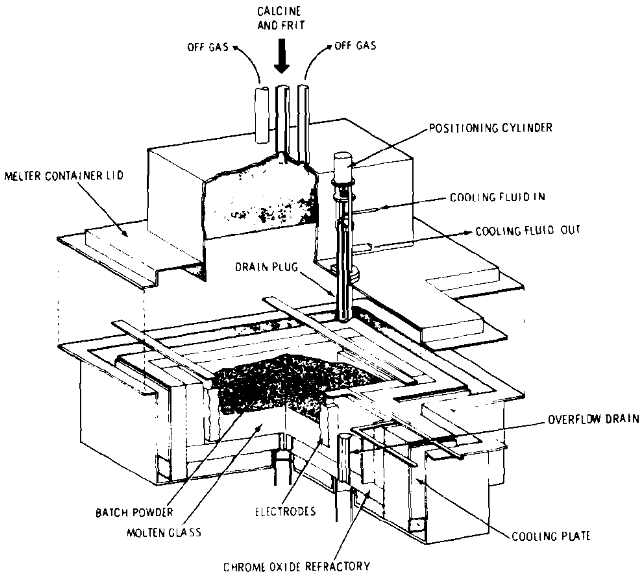

FIGURE 1. Schematic of Ceramic Melter

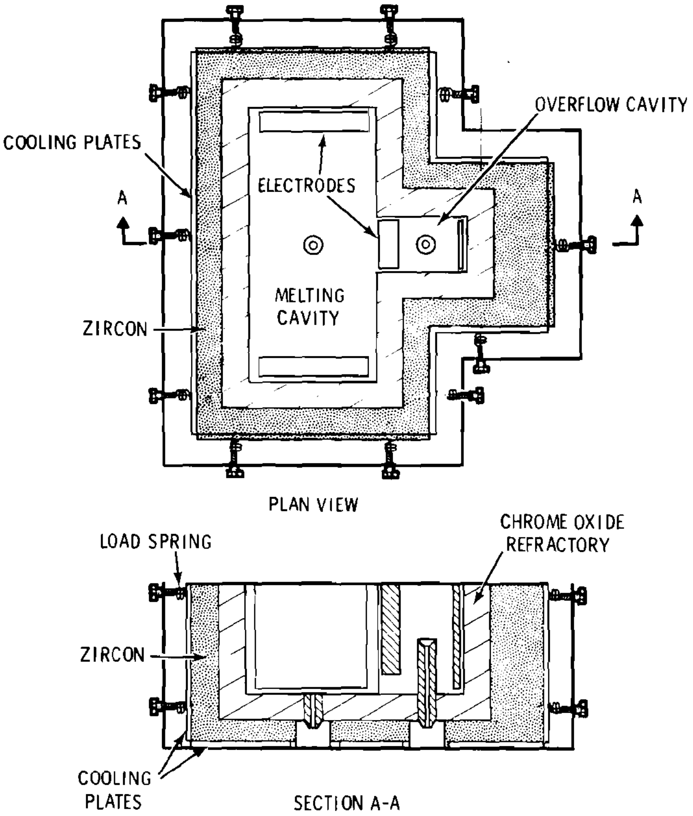

FIGURE 2. Plan and Section Views of Engineering Scale Melter

springs which could be adjusted by positioning screws on the outside of the box. The exterior dimensions of the entire melter were 4-ft wide, 4 1/2-ft long and 3-ft tall.

In operation, the simulated calcine and frit were fed proportionally into the melter through the centrally located nozzle in the container lid. After the batch was melted and refined in the melting cavity, it flowed under the overflow cavity electrode and out the drain pipe. A fluid-cooled plug valve was used to stop glass drainage. A picture of the melter during production melting is given in Figure 3.

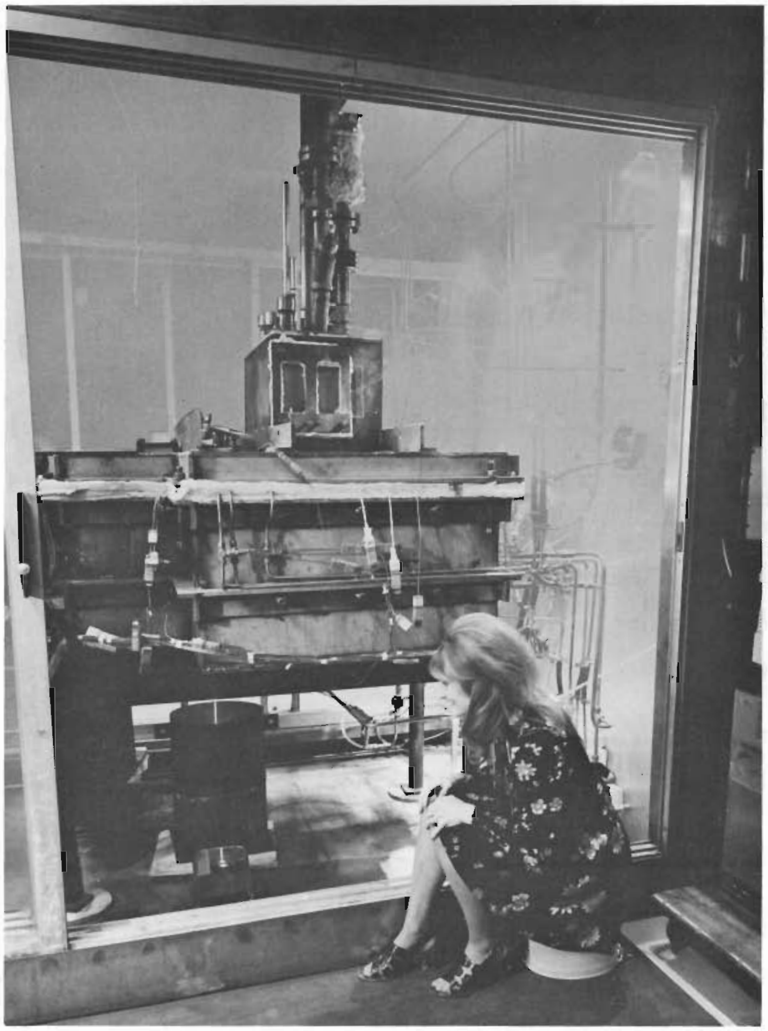

FIGURE 3. Engineering-Scale Ceramic Melter During a Melting Test (Note mirror in front of canister shows outlet of overflow pipe)

MELTER CONTROL

This melter was designed to be controlled either by a temperature feedback control loop or by a manually adjusted, constant current control. From earlier laboratory melters, it was verified that voltage control was unstable and therefore not suitable. Tests with this melter using glass temperature control were satisfactory but were not as suitable as constant current control. Temperature control was difficult to tune due to the thermal lag of the glass. Selecting the proper location for the thermocouple as well as achieving a suitable thermocouple life were also problems encountered using temperature control.

Power control is frequently used in the commercial glass industry, but has not been thoroughly tested. However, it appears from system analysis and favorable experimental results that current control is an appropriate control method for a nuclear waste melter. This control technique was used almost exclusively during the 10.8 months of continuous operation of the engineering-scale melter. The melter was idled (no glass produced but maintained at operating temperature) for weeks without operator action and frequently without being monitored.

SACRIFICIAL ELEMENT STARTUP TECHNIQUE

After testing several laboratory-scale melters, it was found that startup and restart of a melter with a frozen tank required resolution before it could be used in a radioactive facility. After several techniques were evaluated, the one which provided most flexibility was the sacrificial element startup technique.

To obtain an operating status with a joule-heated ceramic melter, it is necessary to heat the glass charge between the electrodes to a temperature at which the glass is conductive. In commercial glass tanks, gas firing is the common startup technique which is not well suited for starting up a radioactive waste melter for two reasons. First, the voluminous noncondensible off-gases might not be compatible with the reprocessing plant. Second, when restarting a frozen tank with submerged electrodes, gas firing might not be successful due to the opaque characteristics of the waste glasses.

To provide a method for initial startup and restart with a frozen tank, the sacrificial element technique was developed. This approach for startup of a joule-heated ceramic melter is illustrated in Figure 4. Because restart of a frozen tank with submerged electrodes is the most challenging situation, the startup procedure will be explained for this condition.

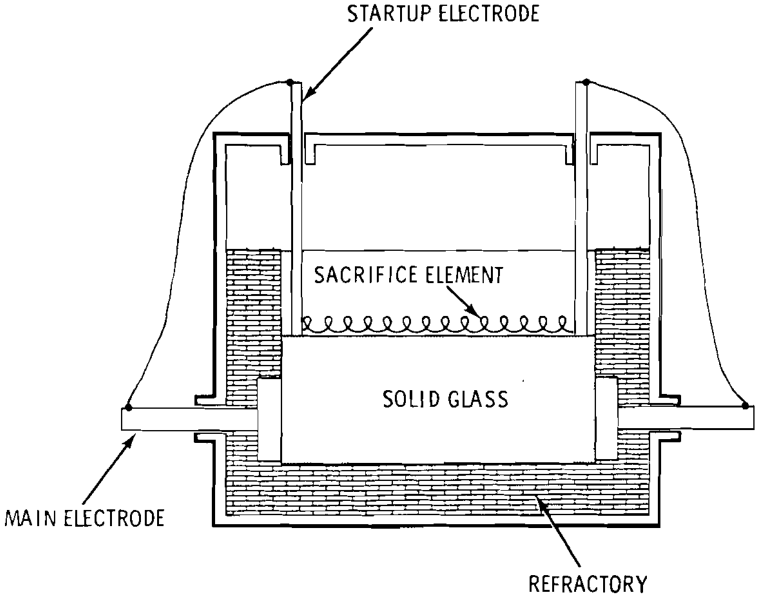

FIGURE 4. Schematic of Startup Technique

A set of startup electrodes with a resistance heating element between them is placed in a flange which fits in the cover plate of the melter. This assembly is lowered onto the mating opening on the melter shell and is then secured. Since the vertical startup electrodes are free to translate in the vertical direction, the electrodes settle to the top of the frozen glass when the assembly is lowered onto the roof. The startup electrodes are then attached to an independent power supply or in parallel with the main power electrodes. After glass frit is dropped into the cavity and the startup element is buried, the melter is ready for a programmed startup.

Using either a temperature feedback or power control system, energy is dissipated in the sacrificial element and heats the surrounding glass. As the power level increases, glass begins to melt adjacent to the heating wire. Further power input results in a molten zone between the startup electrodes. At this condition the use of the sacrificial element is complete. The sacrificial element used in all cases has been a common electrical resistance wire. However, other materials like molybdenum disilicide or silicon carbide may be more appropriate. The important characteristic of the sacrificial element is that it is readily dissolved by the glass at high temperatures.

As power input continues to increase, the molten zone expands and the startup electrodes settle into the frozen tank. Ultimately, the startup electrodes settle down to the elevation of the main electrodes and the molten zone expands to the main electrodes. At this stage, the main electrodes begin to pass current. Finally, the main electrodes assume entire control and the startup electrodes can be removed or allowed to dissolve in the molten glass.

This technique has been used for initial startup in two different melters and used to restart one melter with a frozen tank on three separate occasions. Heatup rates as low as 6°C/hr to as high as 300°C have been demonstrated. For a hot cell unit, the initial startup assembly would be an integral part of the melter and no remote operation would be required because both the startup electrodes and sacrificial elements would be consumed during startup.

Restart of a joule-heated ceramic melter would require the remote operations outlined above, however, it should be noted that shutdown of the melter would only be necessary in case of an abnormal condition in the melter. Should other equipment require shutdown for maintenance, a joule-heated ceramic melter would not require shutdown because it can operate for extended periods of time at a reduced glass temperature without significant loss of operating life.

PRODUCTION EXPERIENCE WHILE FEEDING CALCINE AND FRIT

In preliminary tests the simulated waste calcine and frit were intimately mixed prior to being fed to the melter. After experiencing underpowering in the initial test, smooth rapid melting of the batch was demonstrated. Glass production rates in excess of 45 kg/hr were frequently shown with no problems. Typical operating conditions during these tests were power input of 35 kW at an rms voltage of 160 between the melting cavity electrodes.

After a second solids feeder was purchased and installed, the calcine and frit were fed to the melting cavity through separate lines. Both material streams accumulated in the melter at approximately the same location, but little mixing was realized. In spite of this condition the batch did melt into a homogeneous glass in the melter. However, this method of feeding appeared to reduce the capacity of the unit by about 10%.

While melting a calcine with a high content of sodium nitrate, increased capacity was noted. The sodium nitrate decomposed beneath the batch cone, escaped to the periphery, and induced significant agitation in the molten glass. This phenomena appeared to increase the melting rate by about 15%.

DIRECT LIQUID WASTE FEEDING

The high capacity of this unit suggested that feeding the simulated high-level liquid to the melter might be practical. The first direct liquid feeding test, the simulated high-level liquid waste and the frit were fed through separate lines. However, after about 1 hour of feeding, it became obvious that this was not a suitable approach. When the liquid waste evaporated and calcined, no frit was present to flux it into the pool.

Instead, the calcine sintered, and formed cakes and crusts near the walls. To eliminate this problem, the frit was added to the acid waste to form a slurry. When this combined liquid waste and frit slurry was fed to the melting cavity, the calcined material rapidly reacted with the finely divided frit and melted into the pool.

Using this feeding method, early tests showed process rates of about 15 ℓ/hr. To boost the units capacity, an air bubbler pipe was submerged in the molten glass. This improved the capacity but would periodically cause molten glass to be splattered inside the melting cavity. After the initial tests it was felt that at least a portion of the glass surface needed to be exposed to allow rapid processing of the liquid waste. However, review of these tests suggested that insufficient power had been delivered to the molten glass which would obviously limit throughput capacity.

In the most recent tests, the entire glass surface has been purposely flooded (i.e., covered with from 1 to 2 in. of the slurry) to observe the system's operation. In these tests, additional power was introduced into the melter and process rates averaged about 25 ℓ/hr. Although a significant improvement in capacity was realized, the major contrast of flooded operation was the much lower entrainment of material in the off-gas stream. In one test less than 0.8 wt% of the oxides present in the slurry was entrained in the off-gas which was a significant improvement over liquid feeding with no flooding. Should this be characteristic of this process, simplificiation of the off-gas filtration equipment may result.

From the direct liquid feeding tests, confidence has been gained that this process is not only promising, but also practical. It would significantly reduce the complexity of waste solidification, improve process reliability, and reduce overall processing costs while providing the capability of producing homogeneous, highly durable waste glasses. For these reasons, development of a full-scale, remotely operable, direct liquid-fed ceramic melter will receive much emphasis in the future development program. It appears that a joule-heated ceramic melter with 12 square feet (ft^2) of surface area would be capable of processing the liquid waste from a 5 MTU/day reprocessing plant. A conceptual design with this amount of surface area would have maximum external dimensions of 6-1/2 ft long, 6 ft wide and 5 ft high. This scale up assumes a liquid waste concentration of 378 ℓ/MTU (metric ton uranium) reprocessed. If the liquid waste was more dilute, the size would be increased accordingly or a preconcentrator could be used such as a wiped film evaporator.

A summary of the test results with the engineering-scale melter is given in Table 1. The unit was started up in mid-January of 1975 and maintained at, or above, a molten glass temperature of 1150°C for 10.8 continuous months. During the nearly 11 months of operation, 20 day-long experiments were completed to test the unit while feeding day calcine powder and liquid waste. Over 3600 kg (8000 lb) of glass were produced by the unit. While feeding powdered calcine and frit, the unit demonstrated a maximum capacity of 60 kg/hr. The average capacity for all batch powder tests was over 45 kg/hr. Thus, this melter has been demonstrated at or above the requirement of a Reference 5 MTU/day reprocessing plant.

While feeding the simulated high-level liquid waste, the melter has been demonstrated at 29 ℓ/hr which is equivalent to the needs of a 1.75 MTU/day reprocessing plant.

TABLE 1. Performance of the Engineering-Scale Ceramic Melter(a)

Capacity		Equivalent Plant Capacity, MTU/day
Calcine Feeding		
Maximum	60 kg/hr	6.0(b)
Average All Tests	46 kg/hr	4.6
Liquid Waste Feeding		
Maximum	29 ℓ/hr	1.8(c)
Average All Tests	20 ℓ/hr	1.3
Restarted	1/15/76	

(a) Time at operating temperature, continuous - 10.8 months (1-14-75 to 12-11-75)

(b) Assuming a waste yielding 65.5 kg oxides/MTU and frit to calcine weight ratio of 2.7

(c) High-level liquid waste concentrated to 378 ℓ/MTU and 5% recycle from effluent treatment.

MELTER EVALUATION AFTER 10.8 MONTHS OF OPERATION

The engineering-scale melter was drained and shut down in mid-December of 1975 because the building electrical power was shut off for over 24 hr. This provided an opportunity to evaluate the condition of the melter.

ELECTRODES

The use of the nickel-chrome alloy as the electrodes and the drain pipe was anticipated to be the limiting factor for melter life when the engineering-scale ceramic melter was designed. Although this alloy had been used successfully in the metallic melter, prior to the ceramic melter startup, it was felt that the electrodes would last only 6 to 9 months due to high temperatures in this type of melter.

After the melter was shut down, the electrodes were removed and the adhering glass and metal oxides were removed by sand blasting; a photograph of the electrodes is given in Figure 5. During the 10.8 months of operation, the material lost by the electrodes was significantly less than anticipated; data on electrode weight loss are given in Table 2. It should be noted that the weight percent lost was computed for Table 2 by dividing the total weight lost by only the weight of the electrode which was immersed in the molten glass. Thus, any material loss due to oxidation or corrosion in the vapor space portion of the electrodes was included as weight lost due to reaction with the glass. Another fact should be noted about material lost from the melting cavity electrodes. It can be observed in Figure 5 that the corners of the electrodes have experienced significant loss. The majority of this material was lost during melter shutdown. The melting cavity electrodes, which were suspended from the top of the refractories and were 1/2-in. above the melter floor, experienced loss by arcing when the glass level fell below the electrode. Although current flow should have stopped, this flow continued by arcing from the electrodes to the glass. Before this condition was noted, a portion of the electrodes had melted. When the electrodes were removed from the melter, globs of metal were found

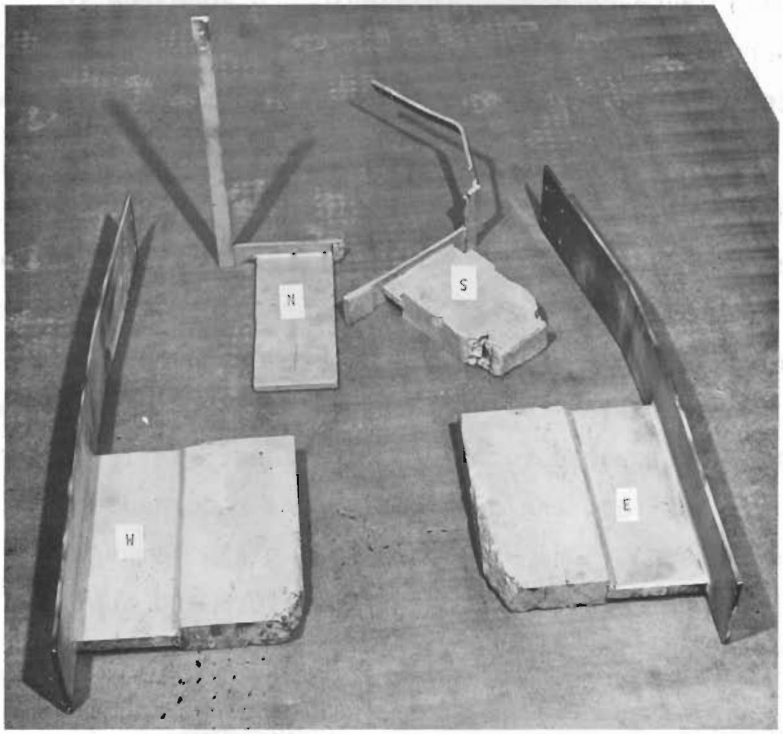

FIGURE 5. Melting Cavity Electrodes (Foreground) and Overflow Electrodes After 10.8 Months of Operation

TABLE 2. Weight Loss from Melter Electrodes After 10.8 Months of Operation

	Total Weight Lost, lb	Weight Lost, %(a)
Overflow Cavity Electrodes		
N(b)	0.19	5.2
S(b)	2.89	27.3
Melting Cavity Electrodes		
E(b)	1.37	3.9
W(b)	0.85	2.4

(a) Computed using only the weight exposed to molten glass. Oxidation and corrosion above glass included in total weight lost.

(b) See Figure 5.

directly beneath these corners. This evidence appeared to substantiate that the corners had melted off while arcing to the glass. Thus, the true amount of material lost due to corrosion by the molten glass was significantly lower than given in Table 2.

The surprisingly low amount of corrosion of the melting cavity electrodes can be readily explained by the data presented in Figure 6. Shortly after melter startup, thermowells were lowered into the molten pool to determine the glass temperature. To determine the electrode temperatures, 1/4-in. holes were drilled in the melting cavity electrodes which enabled thermocouples to be lowered into the electrodes. While idling, the temperatures within the pool and the electrodes were recorded and are given in Figure 6. As can be seen in Figure 6 the peak glass temperature is nearly 200°C higher than the peak electrode temperature and that the peak temperature of the electrode is only 1000°C.

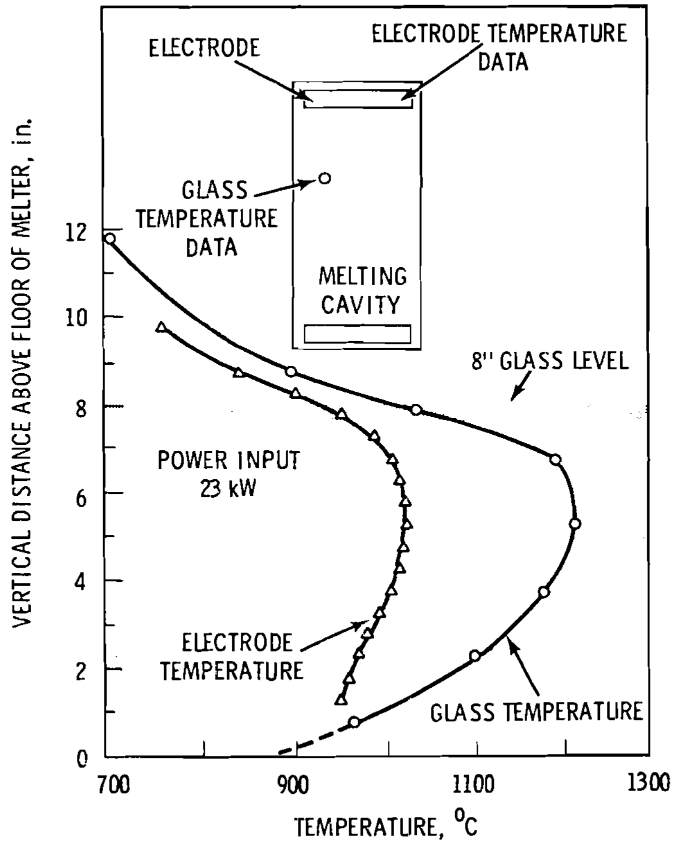

FIGURE 6. Temperatures in Melter While Idling

Except for a more uniformly high glass temperature, these data are typical of temperatures observed in the system during production melting. Thus, the low corrosion of this nickel-chrome alloy is explained by its low operating temperature. The low temperature in the electrode is due to vertical cooling to the vapor head space and horizontal cooling to the water-cooled refractory end wall.

The significant amount of attack to the overflow cavity electrode, S, is attributed to two factors. First, the electrode operated at a consistently higher temperature than the other electrodes because it was exposed to the hot glass on both faces and was not adjacent to a water-cooled refractory. Secondly, shorting along the refractory sidewall caused high current densities at the corners adjacent to this electrode which resulted in much higher temperatures in this area. (See discussion of shorting through refractory in a following paragraph.)

REFRACTORIES

After shutting down the melter, residual glass was chipped from the faces of the refractory to reveal the extent of refractory damage. The refractory blocks all exhibited cracking with a spacing of 2 to 4 in. between cracks. Although the blocks had cracked, there were no locations where pieces of the material had fallen into the cavity and the blocks still retained their integrity.

The most severe damage to the glass contact refractory occurred where it had been attached by molten metal. The reason for the presence of the molten metal is explained below.

USE OF A BATCH REDUCING AGENT

An important part of developing a waste solidification process is to make a durable waste product at a minimum cost. One way of reducing waste management costs is to minimize waste glass volume. This approach is limited by the chemical durability and the formation of nonvitreous phases (primarily alkali molybdates). The addition of a few weight percent of silicon metal to the batch had been successful in eliminating the molybdate phases even at low frit to calcine ratios in laboratory tests.

To evaluate the utility of this approach, a batch composed of one part calcine and two parts frit was thoroughly mixed with 3 wt% silicon metal and fed to the ceramic melter. During the test, it became apparent that an excess of silicon had been added to the batch which reduced several metal oxides in both the frit and the calcine. Metals with low boiling temperatures (zinc, cadmium, tellurium) volatilized from the molten glass and rapidly oxidized at the surface. However, the more refractory metals (primarily nickel) settled to the floor of the melter. The presence of this molten metal on the floor severely disrupted the operating characteristics of the melter because the electric current shorted through the molten metal. This caused temperatures at these locations to become very high (>1400°C). Shorting through the metal continued for approximately 2 weeks, but this gradually subsided and melting tests were reinitiated.

The molten metal apparently reacted with the refractories because only a moderate amount of metal was found in the floor when the melter was cleaned. The reaction of the metal with the refractory resulted in a 1/2 to 1-1/2 in. deep porous mass which was completely removed when the melter was cleaned. The location of the reaction zone is outlined in Figure 7. Note that a portion of the sidewall has been removed to accomplish the desired design modification explained in a following section.

Shorting through Refractory

The other damage to the refractory occurred adjacent to the overflow cavity. Under certain conditions a portion of the current passed through the refractory along the overflow cavity sidewall. At the interface between the refractory and the overflow electrode, high current densities (high temperatures) apparently existed. Although the damage to the refractory was moderate, it was a location of high wear. This high wear area corresponded to the portion of the overflow cavity electrode which experienced the greatest weight loss (see Figure 5).

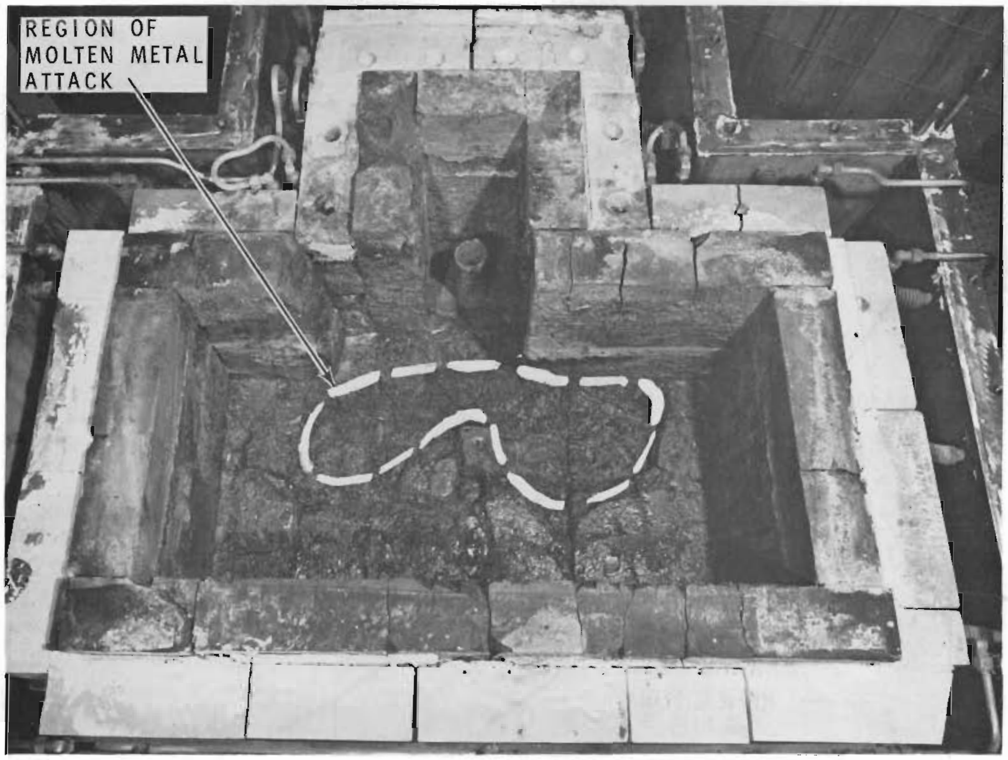

FIGURE 7. Engineering-Scale Ceramic Melter After 10.8 Months of Operation (Glass has been removed from surfaces.)

Prior to construction of this melter, information about the resistivity of the refractory was not available. This data did become available subsequent to startup of the unit. The resistivity of the refractory and of the waste glass is given in Figure 8. From this data, it is obvious that the refractory is less resistive or more conductive than the waste glass. Assuming the mean temperature of the refractory was 1000°C and the glass temperature was 1150°C, more than half of the current could flow through the refractories. This analysis assumes that the refractory is a continuous homogeneous block without cracks; thus, this prediction would be higher than actual. However, this analysis is indicative of what was experienced with this unit.

In one test, a glass composition was melted which had much higher resisitivity than the typical waste glass shown in Figure 8. The day following the test, it was found that nearly all of the current was flowing through

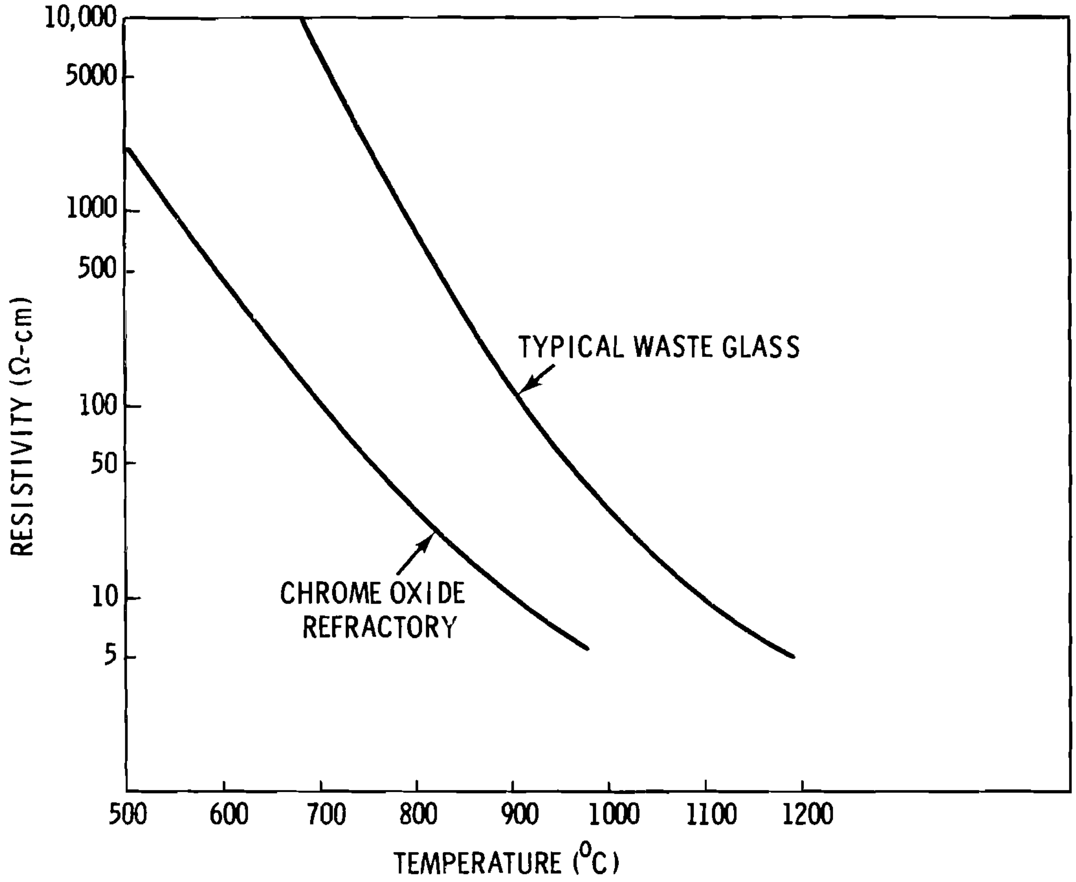

FIGURE 8. Resistivity of Refractory and Waste Glass Used in Engineering-Scale Ceramic Melter

the refractory and the majority of the glass had frozen. Recovery from this condition was achieved by adding sodium hydroxide to the melting cavity which decreased the resistivity of the glass.

The shorting problem has the effect of reducing the effectiveness of joule heating and significantly reducing the capacity of the unit. In spite of this adverse characteristic, the unit has performed acceptably. By using a refractory with much higher electrical resistivity in future designs, marked improvement in system operation is anticipated.

The condition of the engineering-scale ceramic melter after 10.8 months of operation was considerably better than anticipated. Although the melter did not produce glass continuously over the test period, the materials were continuously exposed to the molten glass typical of continuous operation. This is a necessary but not sufficient test for long melter life. Nonetheless, if an order of magnitude increase in corrosion rate would result from continuous glass melting, the anticipated life for this melter would still be more than the design objective of two years.

The severe conditions which this melter experienced demonstrates its ruggedness. It was exposed to extremely severe thermal shock during direct liquid feeding and was exposed to large variations in bulk glass temperature. Further, on two separate occasions, building power was interrupted for more than 25 min and the melter recovered on its own. Shorting through the sidewalls also exposed the refractory to severe thermal cycling. In spite of these adverse conditions, the unit was judged to be in good enough condition for continued use.

Design Modifications

To reduce shorting along the overflow cavity sidewall, the overflow cavity electrode was moved to the far sidewall and a block with a 6-in. wide by 3-in. high cutout replaced a portion of the sidewall and the electrode. A plan view of the melter after the modifications is given in Figure 9. This can be compared to the original design given in Figure 2. In the modified design, the overflow cavity is kept hot by firing between a set of electrodes across the melting cavity through the 3- by 6-in. cutout in the sidewall.

Except for the new larger overflow electrodes and the sidewall weir block, the melter was not repaired. In mid-January of 1976 the melter was again started up and three melting tests have been completed producing over 1000 lb of simulated waste glass.

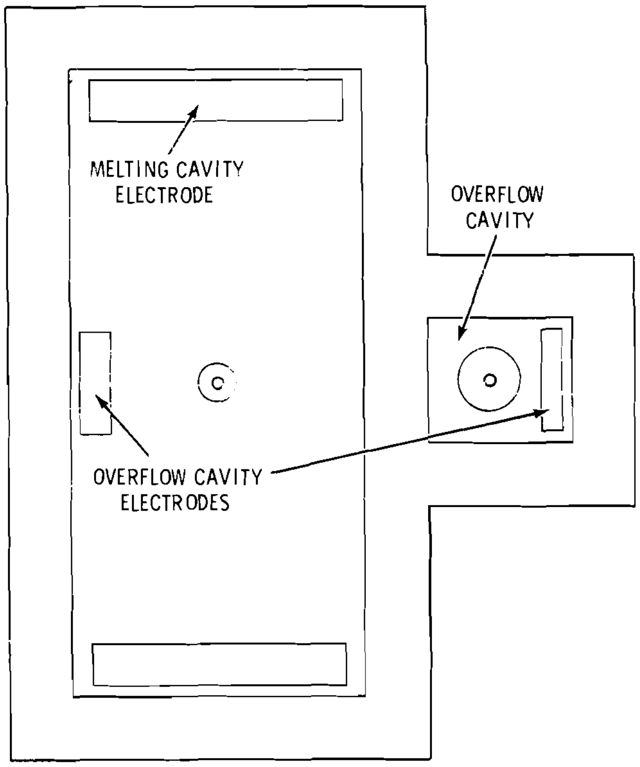

FIGURE 9. Schematic of Engineering-Scale Ceramic Melter After Modifications

Future Plans

Continued testing is planned of the engineering-scale ceramic melter. Major emphasis will be concentrated on testing a technique for boosting the liquid throughput capacity. A conceptual design for a full-scale liquid fed melter is complete. This design will also use a nickel-chrome alloy for electrodes but will be designed specifically to process liquid waste. A calcine fed melter will also be designed which will be tested under fully radioactive conditions.

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