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VITRIFICATION OF SRP WASTE BY A SLURRY-FED CERAMIC MELTER

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ABSTRACT

Savannah River Laboratory (SRL) is testing the slurry feeding of a ceramic melter as a possible method to vitrify Savannah River Plant high-level radioactive waste. Feeding a liquid slurry requires simpler and less expensive equipment than feeding a powdered calcine. Experiments have progressed from manual feeding, to a semiautomatic system, to the present slurry-feed system, which is completely automatic. All experiments to date indicate that slurry feeding is a promising way of vitrifying waste. No safety hazards associated with feeding the slurry onto molten glass at 1150°C have been observed experimentally, even when the melter chamber was purposely flooded.

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INTRODUCTION

The concept of feeding a liquid slurry directly onto molten glass is not new. In commercial operation gas-fired liquid-fed melters have successfully produced sodium phosphate glass for 25 years, using a slurry containing about 28 weight percent water.

Liquid or slurry feeding for vitrifying radioactive waste has been studied for almost 20 years. Of particular interest are the British Harvest Process (pot vitrification) and the German Pamela Process (liqid feeding to a ceramic melter).

Over the past 5 years, the concept of liquid feeding for ceramic melters has been further developed in Germany, at the Karlsruhe Nuclear Center. Within the last several years, runs up to 500 hr have been successfully achieved using simulated high-level liquid waste (HLLW) in a slurry-fed ceramic melter.

In the United States, the liquid feeding concept was first introduced and developed at Battelle-Pacific Northwest Laboratory (PNL) in about 1975. A large-scale, direct-liquid-fed ceramic melter (LFCM) was constructed at PNL. Successful melts were made, although melting rates were reduced compared to melts in which dry powder feeds were used.

ADVANTAGES

Advantages of slurry feeding result from eliminating the drying operation. Figure 1 shows waste vitrification by (1) spray drying and (2) slurry feeding. Potential advantages of slurry feeding includes the following:

- Simplification of equipment. Elimination of spray calcination (SC) or drying would (1) simplify process operation and control, (2) simplify equipment replacement, and (3) reduce costs.
- Elimination of handling radioactive powder. The waste in slurry feeding would be charged to the melter in a liquid stream, thereby eliminating the need to handle finely divided, easily dispersed radioactive powder.
- Improved mixing. Slurry feeding would produce a relatively well-mixed system which would help minimize foaming and formation of insoluble precipitates in the melter.
- Off-gas reduction. The liquid component of the slurry would enable the feed batch to be more uniformly distributed over the surface of the melt. Uniform distribution would in turn, produce an effective cold cap which could reduce the volume of noncondensable off-gas produced during melting. Also, the large volume of atomizing air used in spray calcining operations would be eliminated.

UNCERTAINTIES

Although slurry feeding has clear advantages over calcine feeding there are some uncertainties about large-scale operation.

- Safety consideration - steam explosion. A steam explosion is a postulated event that could occur if slurry became trapped below the melt surface. No evidence of such occurence has been documented for compositions similar to that of SRP waste glass. Also, in experiments involving flooding, which may be considered a worst case from a safety standpoint, no explosions or even eructations were observed. Safety analyses of the liquid feeding system are now in progress.

- Reduced melting rate. Power density in the melter would increase because with slurry feeding the melter must evaporate water as well as make glass. Better batch distribution and reduced surface heat loss will partially compensate for the added load. Supplemental or booster power can also be used and may be needed in a large-scale melter.
- Thermal shock. The lifetime of electrode and refractory materials could be reduced due to the thermal shock these components sustain when room-temperature slurry contacts molten glass at 1150°C. However, since most melting occurs away from the walls and electrodes, thermal shock is minimal. Also, no thermal shock related degradation or failure of any melter material has been observed thus far.
- Line pluggage/valve malfunctions. Line pluggage and sticking valves are potential problems, but they can be minimized by proper design.

SRL EXPERIMENTAL PROGRAM

SRL's program to investigate slurry feeding has progressed in three stages. First, closely controlled manual feeding was studied to examine safety aspects. Next, a semiautomatic system was used to qualitatively examine the effects of continuous slurry feeding on glass melting. Finally, a completely automatic system was constructed to obtain quantitative data on melting rates and to compare these data with powder feeding.

In most of the experiments, an aqueous suspension containing 25 wt % simulated SRP waste designated TDS and 75 wt % ground Frit was used (Tables 1 and 2). The solids content of the slurry was varied, along with melting and feeding parameters. Continuous versus pulse feeding techniques were also studied. Pulse feeding was selected for further study because of easier feed control and better distribution of the batch over the melt surface and subsequent improved melting rate.

- Manual Feeding

  The manual feeding experiments consisted of adding small amounts of water onto the molten glass surface. The total amount of liquid was gradually increased: then, solids consisting of frit plus waste were added to the liquid. The solids content was then varied, and the amount of steam that evolved was observed. Large ports were left open on the lid of the melter to allow the steam to escape easily and thus avoid any possible pressure buildup within the melter.

  The reaction between the slurry feed and molten glass at 1150°C was vigorous, but not violent. When small amounts of slurry were added, the slurry danced over the surface of the melt, bounced off the walls, and deposited the powder towards the center of the melting chamber. The water was driven off in about 5 seconds in most runs, but before it disappeared it served as a flux to distribute the powder over the melt. Most of the melting occurred in the center of the melting chamber, away from the walls.

As the water content was decreased in the slurry, the melting rate improved, as long as enough water was present to distribute the batch over the melt surface. In addition, as expected, steam that was generated was quickly evolved, and none appeared to be trapped within the melt.

- Semiautomatic Feeding

A semiautomatic feed systeam was constructed to allow continuous experiments to be performed. The system consisted of a slurry tank with stirrer, peristaltic pump, and tubing with manual clamps to adjust the flow of the slurry to the melter and/or back to the tank.

Continuous as well as pulsed feeding techniques were studied. The continuous dripping of slurry into the melter produced a cold spot on the glass surface. Subsequent feed stayed in the cold region and built up an "iceberg," similar to that observed in powder feeding. Hence, a relatively long time was required to melt the feed. In pulse feeding, pulses of slurry were injected into the melter, which allowed the water to distribute the slurry. Then, the batch was allowed to completely melt, and the surface was allowed to regain some of the heat that was lost before another pulse was injected. Therefore, although additional energy is necessary to vaporize the water during slurry feeding, the increased surface area of the distributed powder for melting helps to offset the effect of increased energy demand.

To further study the safety aspects involving steam or popping of molten glass bubbles inside the melter, the melter cavity was intentionally flooded to various degrees. No significant effects were observed, and after a given time interval, the melter recovered without taking any corrective action and operated normally.

Some problems were identified with this system. Pluggage of the lines was observed as well as pluggage of the feed tube into the melter. The next feed system used a larger pump, larger feed lines, and a specially cooled inlet tube to avoid these difficulties.

- Automatic Feeding

An automatic feed system was constructed to obtain quantitative data on the melting rate and behavior of slurry feeding and to compare this with powder feeding. The system, (Figure 2) consists of a slurry tank with stirrer, peristaltic pump, slurry-cooled feed tube, and a heat exchanger to remove heat from the slurry cooling system. Two timers were placed in the feed lines to control the feed with a solenoid, i.e., to control the frequency of feed pulses and the duration of each pulse. By making calibration curves of slurry feed delivered as a function of time for a given slurry composition, the amount of solids delivered to the melter could be determined for any pulse frequency and duration. A manual override was also installed.

In one experiment, no difficulty was observed when glass was continuously melted in a 6-hour run using almost 200 pulses of feed. For a slurry containing 20 wt % solids, the maximum melting rate obtained was about 5 grams of solids per minute in an argon atmosphere. The optimum feeding technique involved charging the melter with slurry every two minutes with a pulse of 1-1/2 seconds and a surface coverage of 80 to 90%. Longer pulses flooded the surface and caused a limited amount of foaming, but 1-1/2 second pulses only produced a light froth over the melt surface. In addition, significant improvements in melting rates were observed by increasing the solids content of the slurry. The effects of these factors on melting rates are shown in Figure 3. The melting rates obtained with slurry feeding were comparable to feeding dry powder, in the laboratory-scale melting tests. Interactions of molten glass with the slurry are illustrated in Figures 4, 5 and 6.

The feed-line solenoid malfunctioned twice. In normal operation about 20-25 mL of slurry are fed to the melter. Due to the first malfunction, about 1500 mL were fed, and as a result of the second solenoid malfunction, more than 2000 mL of slurry flooded the melter. Even under these extreme conditions of high feed volumes, the melter recovered in less than an hour. No popping of glass or excessive buildup of steam was noted. No degradation of refractory bricks or electrode materials due to this event was detected. All experiments performed indicate that slurry feeding is a safe and effective technique for feeding ceramic melters.

FUTURE WORK

Slurry feeding studies will continue in larger-scale ceramic melters and prototypical units. In addition, vitrification of actual SRP waste using slurry feeding will be studied remotely. Methods of increasing melting rates and control of the environment over the melt will be examined in more detail. Investigations on materials of construction within the ceramic melter will be performed at the end of the melter campaigns.

ACKNOWLEDGEMENTS

The author is indebted to P. H. Chismar for his aid in constructing the slurry feed system and to J. L. Ehrhardt, Jr., for helping conduct many of the experiments.

Table 1

SLURRY COMPOSITION

75% Frit 21/25% Simulated SRP Waste (TDS) in 80% Water

Frit 21

SiO_2	52.5 wt %
Na_2O	18.5
B_2O_3	10.0
TiO_2	10.0
CaO	5.0
Li_2O	4.0

Table 2

SIMULATED SRP WASTE (TDS)

Fe_2O_3	45.5 wt %
MnO_2	12.0
$NaNO_3$	7.0
Na_2SO_4	3.7
Al_2SO_3	9.2
NiO	5.6
SiO_2	4.1
CaO	3.4
AW-500	9.5

LIST OF FIGURES

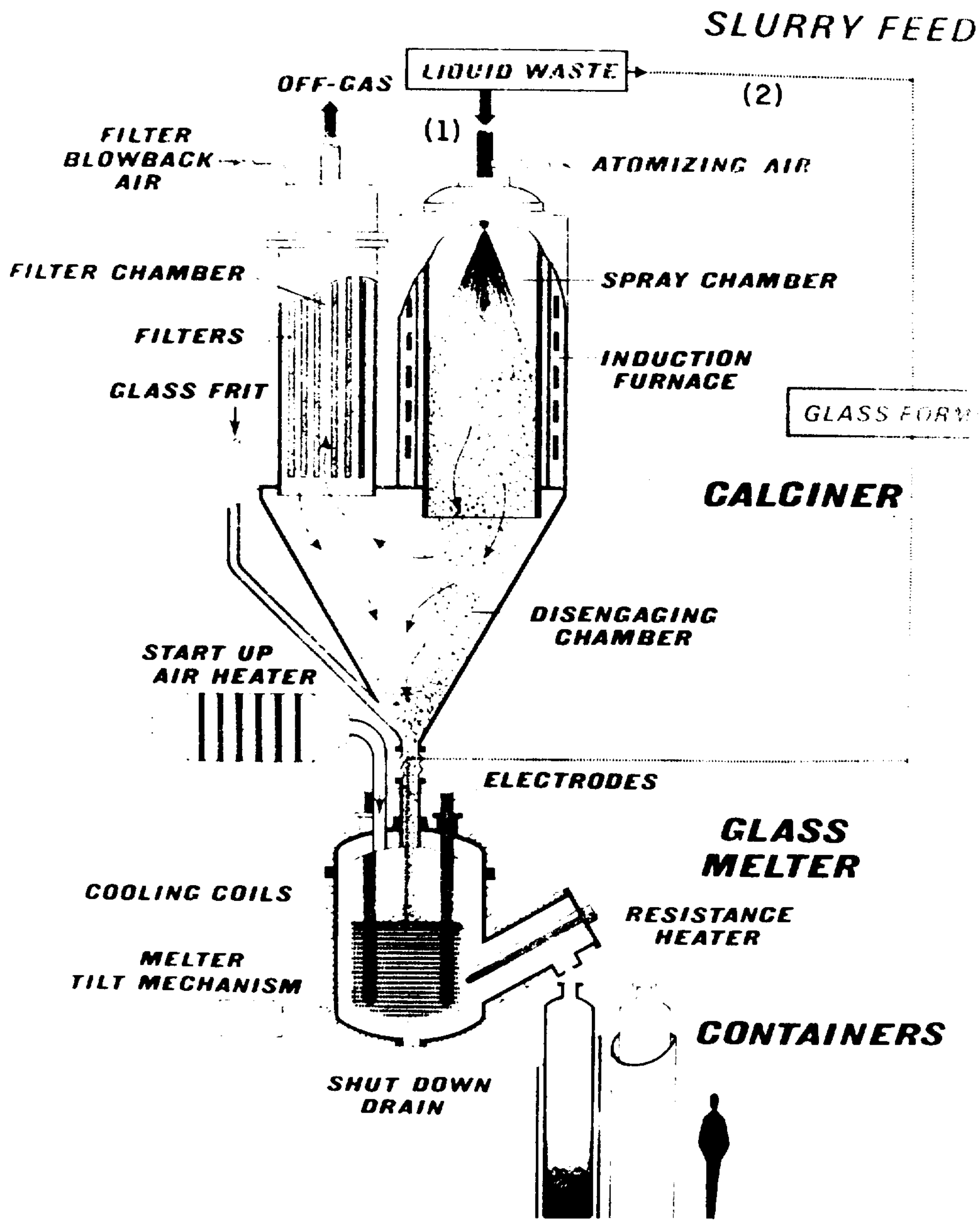

FIGURE 1. Slurry-Feed and Spray-Drying Systems

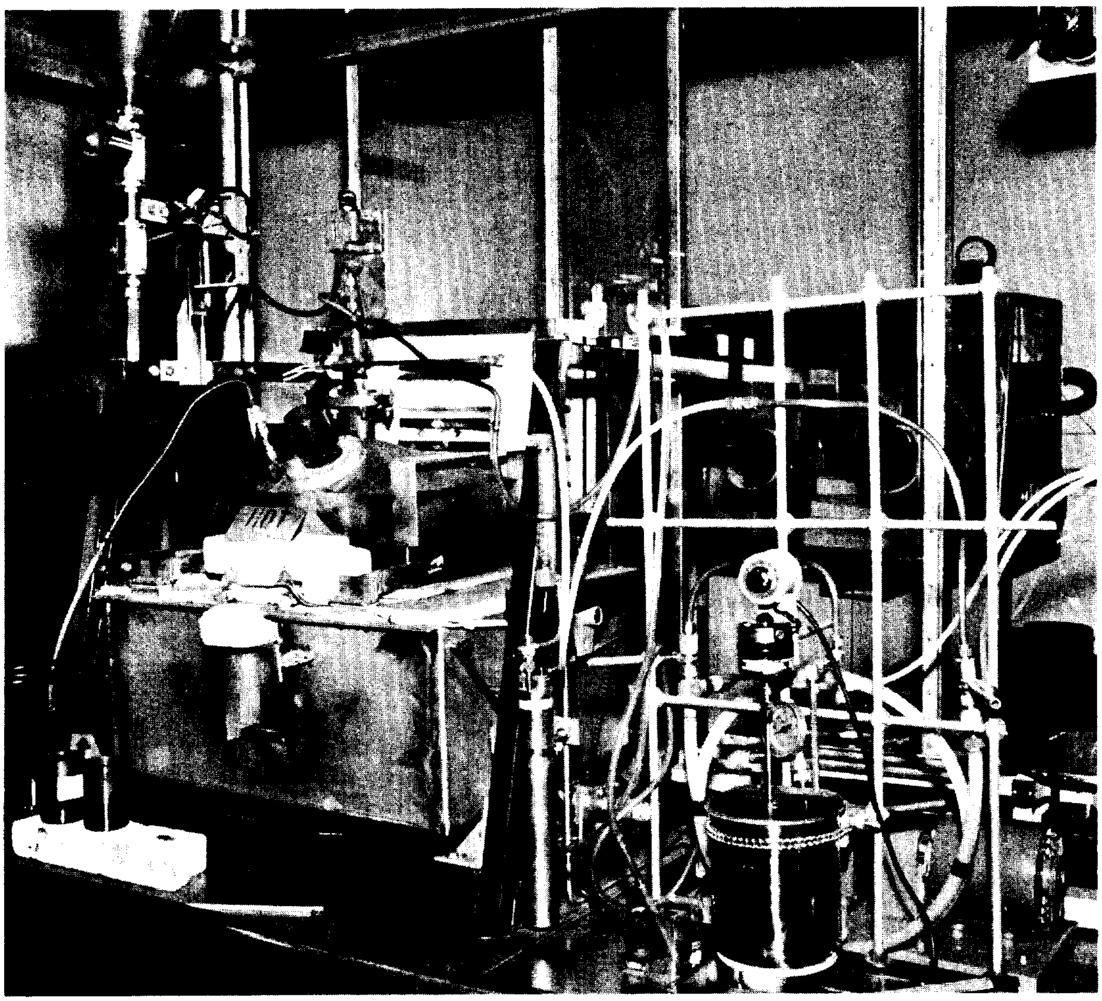

FIGURE 2. Slurry-Feed System

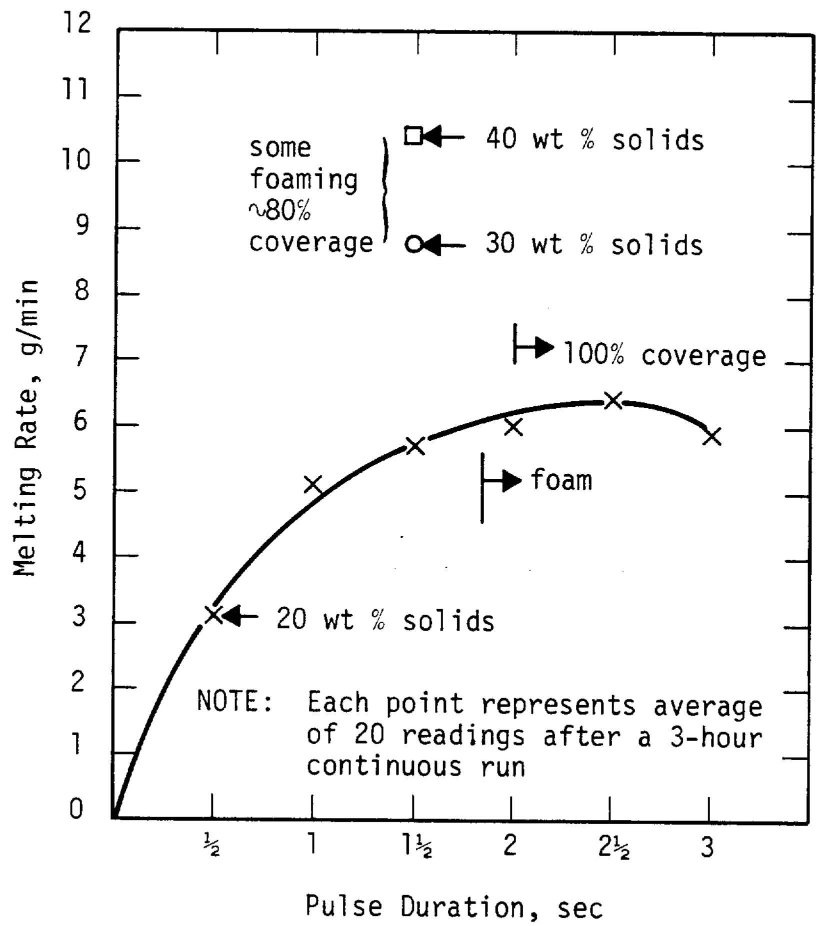

FIGURE 3. Melting Rates for Slurry Feeding

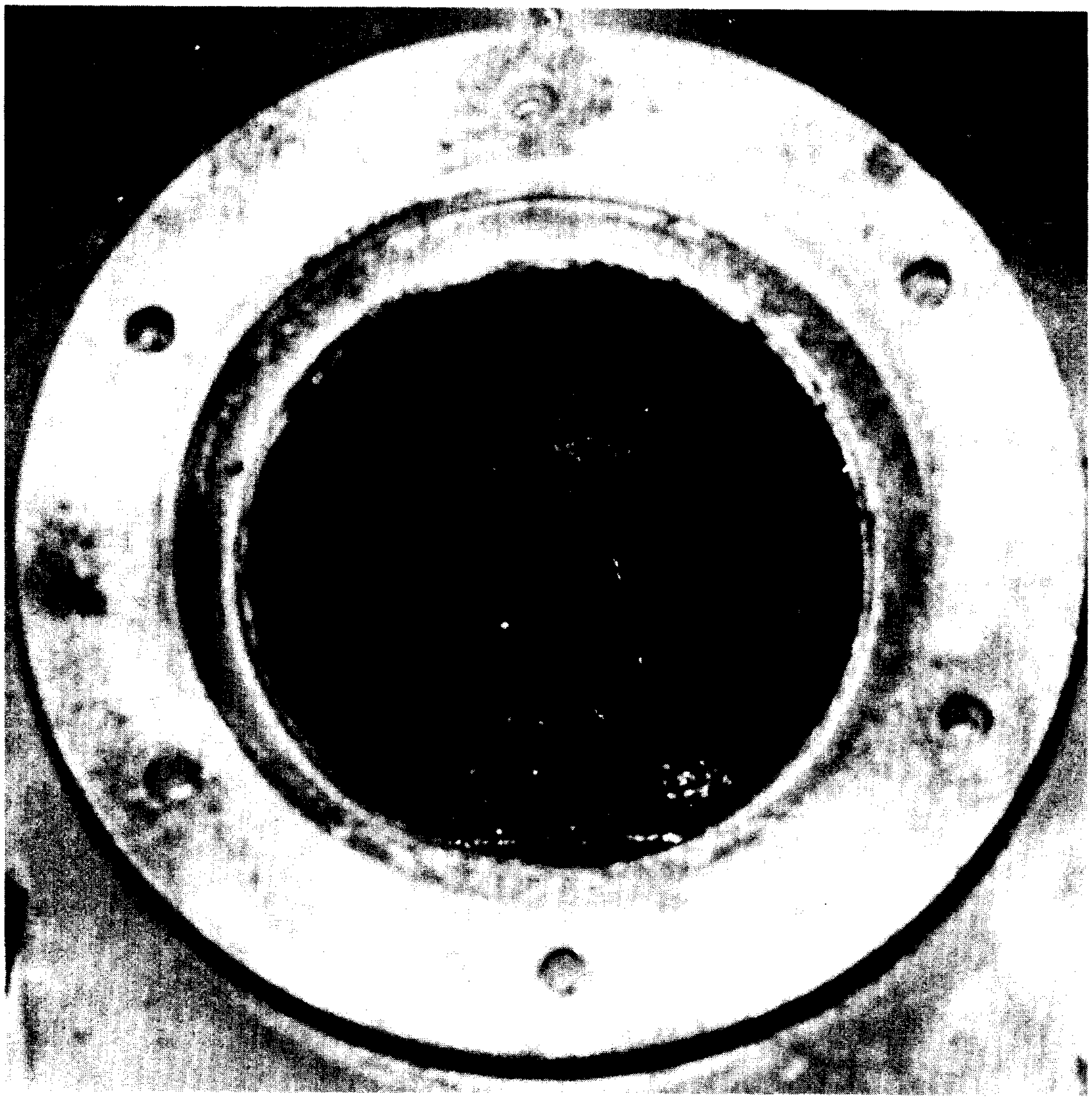

FIGURE 4. A Pulse of Slurry Feeding the Melter

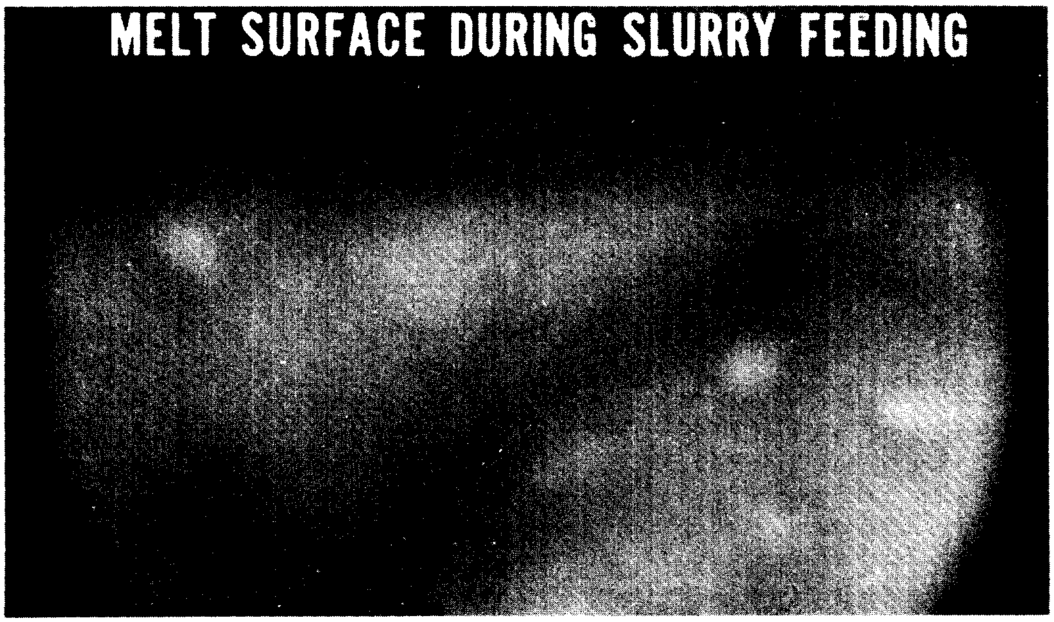

FIGURE 5. Melt Surface during Slurry Feeding

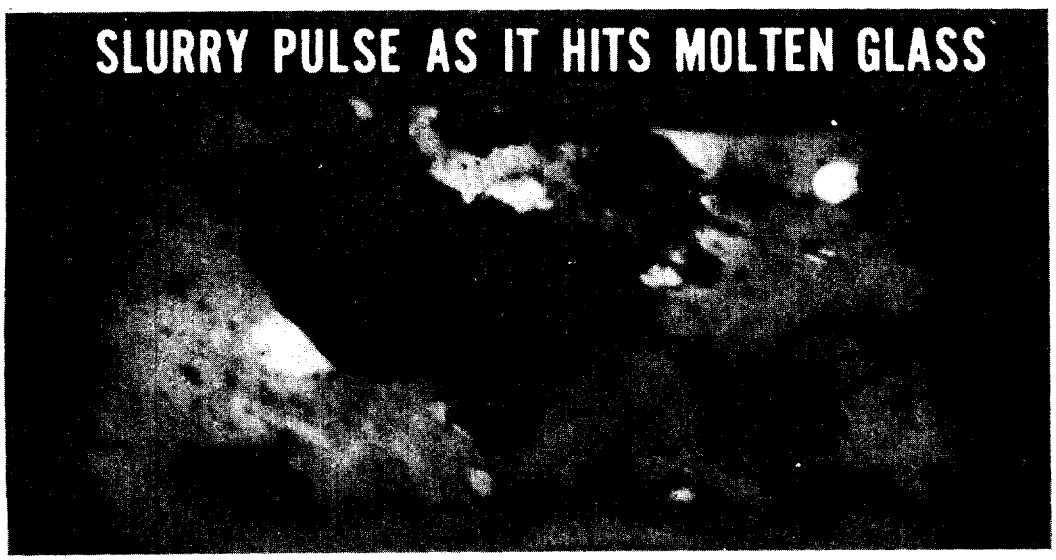

FIGURE 6. Slurry Pulse as it Hits Molten Glass