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**Millimeter-Wave Measurements at 137 GHz
of DWPF Black Frit Glass Flow and Salt
Layer Pooling in a Pilot Scale Melter**

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Introduction

The vitrification of nuclear waste in joule-heated melters is a costly multi-decade national effort. Current operations at the Defense Waste Processing Facility (DWPF) and future operations at the Waste Treatment Plant (WTP), now under construction at Hanford, rely on predictive models because of the lack of on-line monitoring of most processing parameters. This necessitates conservative operation to take into account uncertainties in the modeling and the ability to control actual input parameters. It also makes the operations susceptible to unpredictable anomalies such as foaming, liquidus crystallization, noble metals build up, and salt layer formation. Predictive modeling will become increasingly more difficult in time as the waste glass chemistry evolves with changes in the waste feed compositions. Comprehensive on-line monitoring of the vitrification process would alleviate the uncertainties, improve processing efficiencies (waste loading and throughput), and safeguard the facilities from anomalies.

The millimeter-wave (MMW) range (0.1-10 mm) of the electromagnetic spectrum is ideally suited for on-line monitoring applications in the harsh, high-temperature environment of a nuclear waste glass melter. These wavelengths are long enough to propagate through optically/infrared obscure views, yet short enough to provide useful spatial resolution. MMW waveguide/mirror components that go into the melter can be fabricated from the same ceramic and refractory materials from which the melter is constructed for long life survivability. Laboratory experiments have established the basis for MMW temperature, emissivity, viscosity, and specific gravity measurements [1]. A field test has demonstrated the capability for MMW cold cap 2-D profile temperature measurements important for monitoring the surface dynamics of a melter that affect the plenum off gases and melt rate [2]. In the work reported here, we extend the field testing to include the first application of MMW techniques to real-time measurements of glass flow and salt layer formation in a pilot scale melter.

Experiment

Tests were carried out on the EV-16 joule-heated melter at Clemson Environmental Technology Laboratory. The 18" (45.7 cm) square melt pool was filled to a depth of about 8" (20.3 cm) with 245 lbs (111 kg) of DWPF black frit #265 glass. The 137 GHz millimeter-wave receiver was installed on a vertically movable platform above the melter. The receiver field-of-view was directed through a 1.13" (2.86 cm) internal diameter, 21" (53.3 cm) long mullite ceramic waveguide that extended down from the receiver and into the melter through a 2" (5.1 cm) diameter hole in the melter cover

above the center of the melt pool. The end of the waveguide could be vertically positioned at any height from a few inches above the melt surface to the bottom of the melt pool.

The receiver detected two raw signals in separate channels; the reflected leaked local oscillator (LO) signal and a 3 GHz broadband thermal emission signal. The coherent LO reflection provided interferometric information of the distance to the glass surface and the thermal signal provided temperature and emissivity information. The emissivity could be obtained by either referencing the MMW thermal emission to a thermocouple or by a thermal return reflection method (TRR) [3]. For glass melt flow measurements the immersed waveguide was pressurized with nitrogen gas to displace the melt. The melt flow velocity was recorded as the melt refilled the waveguide after a computer controlled solenoid valve suddenly released the pressure. This is analogous to the standard laboratory capillary flow technique for viscosity measurement.

Glass Flow Measurements

Glass flow was measured over a 1" to 8" (2.5 to 20.3 cm) waveguide immersion range for a series of pressurizations from 0.5" (0.9 mm Hg) to a maximum of 12" (22.4 mm Hg) water pressure at the deepest immersion. A sample set of the reflection signal data for 4" (5.6 mm Hg) pressurization is shown in Figure 1 for six immersion depths. The reflection signals appear as a series of fringes as the melt flows toward the receiver, one fringe for each $\frac{1}{4}$ wavelength (0.55 mm) movement. Not all fringes are resolved at the fastest flow speeds, particularly at the shallower depths. However, the total glass displacement for the applied pressure can be resolved from the slowest flow trace at 7" (17.8 cm) depth and used with the total flow time at each dept to obtain an average flow velocity.

The behavior of the melt flow velocity in the waveguide can be modeled by a simplified laminar flow in a pipe model. The resulting relationship between the average flow velocity, v_{ave} , and the viscosity, η , is given by:

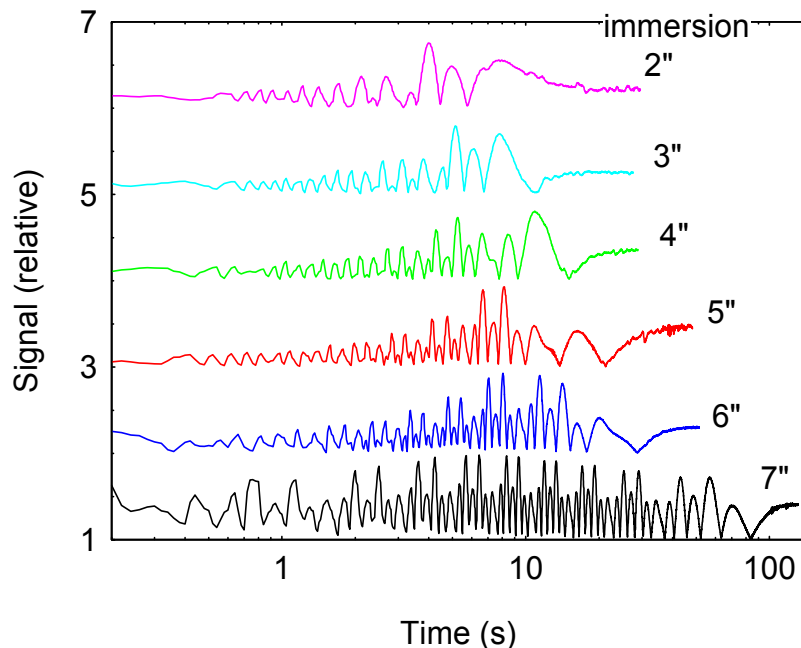


Figure 1. Glass flow reflection data for 4" wg pressurization

$$v_{ave} = A\rho \frac{z_o}{(l-z_o)\eta} \quad (1)$$

where A is an instrumentation constant, ρ is the melt density, l is the waveguide immersion depth, and z_o is the initial pressurization displacement. This relationship has been applied to the data of Figure 1 and to another similar data set for 2" (2.8 mm Hg) pressurization. In Figure 2 the measured average flows are plotted as data points along

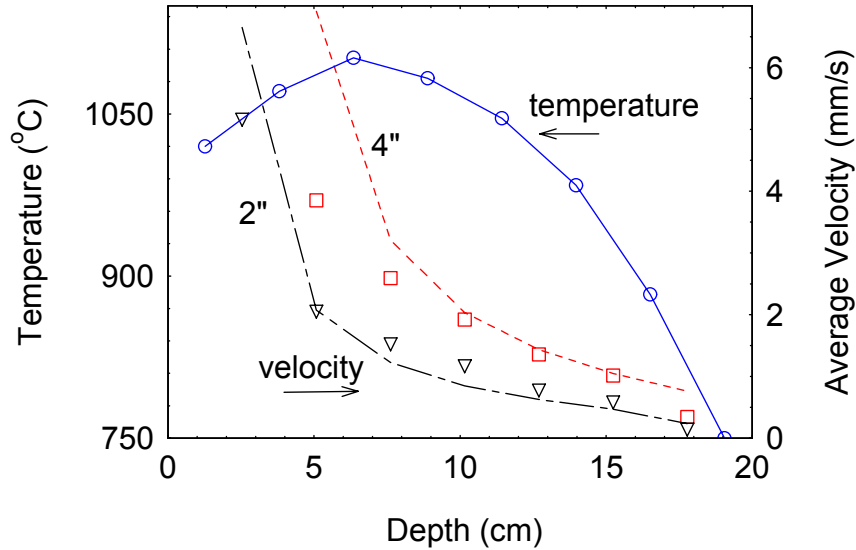


Figure 2. Temperature and melt flow velocity measurements and comparison with simple viscosity theory (dashed curves).

with a measured vertical temperature profile obtained by lowering a thermocouple through the melt. Eq. (1) is plotted as the two dashed curves assuming viscosity is inversely proportional to temperature ($\eta \propto 1/T$). Both calculated plots were fit to the same point at 2" (2.8 mm Hg) pressurization at a depth of 2" (5 cm) to obtain the instrumentation constant. The general trends of the measured flow velocities with depth and pressurization are reproduced reasonably well by the assumed model. The discrepancies could be explained by effects not modeled such as temperature gradients, drifts in temperature during the course of the measurements, and deviations from the laminar flow assumption, particularly at the shallow depths when immersion depth and displacement are similar.

Since immersion depth can be independently obtained, this result demonstrates for the first time in a glass melter a capability for not only on-line viscosity monitoring, but also for obtaining viscosity profiles within the melt.

Salt Measurements

The presence of salt in the melt was monitored by viewing the surface thermal emission. The end of the waveguide was positioned 5/8" (15.6 mm) above the glass surface. Over a 5-hour monitoring period, 1.4 lbs (0.64 kg) of NaSO_4 salt was added at three different times to slowly build up the content of salt in the melt. The resulting MMW thermal emission record is shown in Figure 3. The thermal emission signal, as

for all pyrometers, is a measure of the emissivity-temperature product, ϵT . Except for the brief times when the salt was added, which resulted in sharp cool downs, the melt temperature as recorded by a thermocouple slowly drifted in the range of 950 to 1050 °C.

The MMW temperature record displays two distinct temperatures outside the times that the salt was added and the upward TRR [3] measurement spikes. When the melt was all black frit glass, the MMW temperature was about 570 °C. After the first salt addition it decreased slightly. After the second salt addition it fluctuated rapidly between a high and low value. Finally, after the third salt addition it settled to a lower MMW

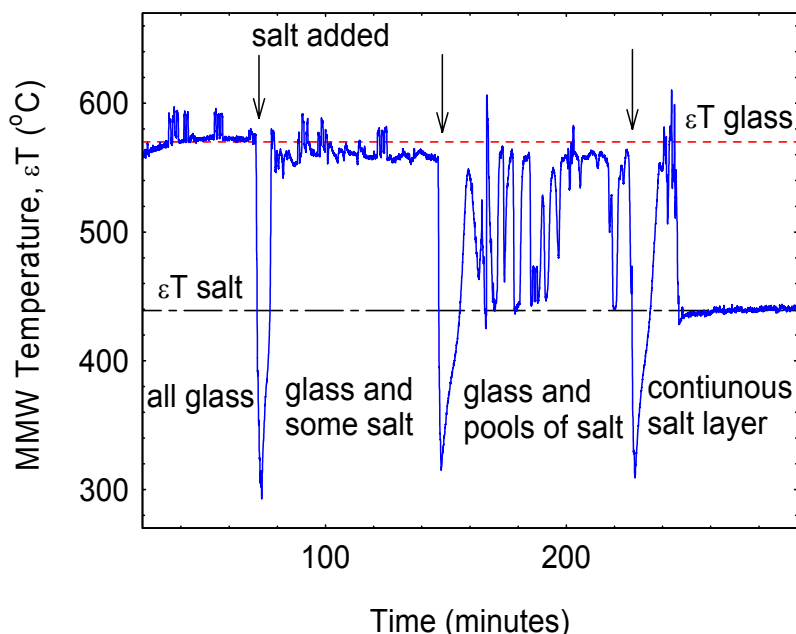


Figure 3. MMW temperature as NaSO₄ salt was added to melter

temperature of about 440 °C. We interpret this signal record as showing the presence of two different materials in the melter, the glass and the salt. A continuous molten salt layer is not formed until the last salt addition. Interesting dynamics in the formation of a salt layer are revealed here for the first time as the formation of a smaller pool or pools of salt that flow around the surface before a continuous layer is established.

The large change in surface emissivity of about 30% demonstrates the capability of MMW technology to monitor salt pool formation in a melter.

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References

1. P. P. Woskov, J. S. Machuzak, P. Thomas, S. K. Sundaram, and W. E. Daniels, Jr., "Millimeter-Wave Monitoring of Nuclear Waste Glass Melts – An Overview", in Environmental Issues and Waste Management Technologies VII, Editors: G. L. Smith, S. K. Sundaram, and D. R. Spearing (Ceramic Transactions, Volume 132) pp. 189-201, 2002.

2. S. K. Sundaram, P.P. Woskov, J.S. Machuzak, and W.E. Daniel, Jr., "Cold Cap Monitoring using Millimeter Wave Technology", *ibid.* pp. 203-213.
3. P. P. Woskov and S. K. Sundaram, "Thermal return reflection method for resolving emissivity and temperature in radiometric measurements", *J. Appl. Phys.*, 92(10), 6302-6310, 2002.