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using millimeter-wave emissivity measurements**

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Molten salt dynamics in glass melts using millimeter-wave emissivity measurements

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Abstract

A novel application of millimeter-wave radiometry has been made for the first time to non-contact detection and monitoring of molten salt layer formation on a nuclear waste glass melt (without the nuclear waste) in a joule-heated melter, which could eventually be implemented for on-line monitoring in nuclear waste vitrification facilities. The experiments were carried out at a frequency of 137 GHz in the EV-16 melter at Clemson Environmental Technology Laboratory (CETL) with 245 lbs. (111 kG) of glass and a total of 4.2 lbs. (1.9 kG) of added salt. The dynamics of salt layer build up were observed from the initial formation of small drops of about 5 mm diameter or less to larger pools > 28 mm cross-section that were coincident with the increase in millimeter-wave surface level fluctuations causing the salt to flow back and forth until a continuous layer was formed. The millimeter-wave emissivity at 137 GHz of DWPF black frit glass melt and molten sodium sulfate salt at 950°C was determined to be 0.64 ± 0.05 and 0.44 ± 0.05 , respectively.

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1. Introduction

Nuclear wastes contain salts [1, 2]. In particular, sodium sulfate (NaSO_4) is an unavoidable component of nuclear waste feed compositions. Generally, salts (especially sulfates) have poor solubility in glass melts and often limit the waste loading of the glass. Formation of salt pools in nuclear waste glass joule-heated melter is of considerable concern. Supersaturated salt coming out of the glass solution can compromise the integrity of the glass as a permanent nuclear waste storage medium (waste form), increase corrosion in the melter (due to corrosive nature of the molten salt), introduce power instabilities in joule-heated melter, and pose a serious hazard to the melter facility if a continuous layer is formed to short out the electrodes. A nearly completed vitrification effort at the West Valley Demonstration Project (WVDP) was limited to a very low level of sulfate (0.3 wt %) in the waste glass [3]. Increased quantities of sodium sulfate will be encountered in future waste compositions. At the Defense Waste Processing Facility (DPWF) and the Waste Treatment Plant (WTP), now under construction in Hanford, extensive modeling [4] and laboratory testing [5] are being performed to optimize waste glass chemistries. Until now there has been no experimental data of the actual real-time dynamics of salt-layer build up in a glass melter or the demonstration of suitable instrumentation for making such measurements. Four probe electrode techniques requiring glass contact for conductivity measurements [6] are not reliable in a plant environment. In this report, we demonstrate the novel adaptation of active millimeter-wave radiometry [7] to non-contact detection and monitoring of salt pool formation and its dynamics in a pilot scale joule-heated test melter.

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, and viscosity measurements [8]. A field test has demonstrated the capability for MMW cold cap two-dimensional profile temperature measurements important for monitoring the surface dynamics of a melter that affect the plenum temperature, evolution of off-gases, and melt rate [9]. In the work reported here, we used the MMW capability to simultaneously measure emissivity, temperature, and surface position to observe for the first time the dynamics of molten salt build up from small flowing droplets to larger pools on the undulating surface of molten glass in a pilot scale melter.

1. Experiment

The experiments 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 # 165 glass (composition by weight: SiO₂=55%, B₂O₃=7.2%, Li₂O=5%, Na₂O=11%, MgO=0.7%, CaO=1.4%, Fe₂O₃=11.3%, Al₂O₃=4.3%, MnO₂=2.5%, NiO=0.9%) [10]. A 137 GHz (λ = 2.2 mm) millimeter-wave heterodyne 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. For the experiments described here, the waveguide end was positioned 5/8" (15.6 mm) above the glass surface, which was located accurately by previous MMW measurements. The set up is shown in Figure 1.

The receiver detected two signals in separate channels; the reflected leaked local oscillator (LO) signal and a 3 GHz wide double sideband thermal emission signal. The phase of the coherent LO reflection depends on the path length between the receiver and melt surface. Consequently, the reflection signal level when heterodyned with the receiver LO varied between a maximum (in phase) and minimum (out of phase) for every $\frac{1}{4}\lambda$ (0.55 mm) change in the relative distance between the receiver and melt surface. Thus, vertical melt surface movements were observed with submillimeter precision.

Temperature and emissivity information were obtained from the broadband MMW thermal signal, which is proportional to the product of emissivity and temperature (ϵT). At constant temperature and melt surface figure, changes in the MMW thermal signal provide an indication of changes in emissivity. Since glass and salt have significantly different MMW emissivities, the MMW thermal signal provided a clear response to the growing and flowing salt pools on the surface of the molten glass below the waveguide view.

Thermal return reflection (TRR) [11] measurements were taken to obtain a quantitative measure of the surface emissivity. In the TRR method a beamsplitter at the receiver redirects a portion of the thermal signal back to the melt as a probe of its reflectivity. The resulting increase in thermal signal depends on the emissivity (ϵ) and

surface figure for coupling (τ_k) the radiation back to the receiver, were the surface figure depends on the surface roughness, curvature, and alignment. In the present experiment the instrumentation layout is the same as in [11] except that the positions of the receiver and return mirror are interchanged relative to the beamsplitter so that the beamsplitter reflectivity, r_{bs} and transmission, τ_{bs} are also interchanged in the equations of [11] for analysis of the data here.

2. Results

The MMW thermal emission over a 5-hour monitoring period was recorded as shown in Figure 2. During this time 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 salt was spooned in through the gap beside the waveguide in the melter cover hole. 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. It was steady at 950°C for the TRR measurements at the beginning and end of the plot.

The MMW temperature record displays two distinct temperatures outside the times that the salt was added and the upward TRR 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 temperature of about 440°C. We interpret this signal record as showing the presence of two different materials on the melt surface, the glass melt and the molten salt. A continuous molten salt layer was not formed until the last salt addition.

The dynamics of the molten salt build up are made evident by also considering the coherent reflection signal, which is sensitive to vertical surface position. Figure 3 shows both the MMW temperature and the coherent reflection (video) signals for ten-minute segments in each of the four time periods between salt additions. The interpretation of the video signal as related to melt surface displacement is illustrated in Figure 4. For every $\frac{1}{4} \lambda$ displacement of the melt level, the video signal will cycle through a minimum–maximum period (fringe) corresponding to a round trip reflection phase shift of π . For a linear displacement the video signal will appear as a rectified cosine function as shown in the beginning of the plot in Figure 4. For a non-linear level change the video signal fluctuations will be distorted from a cosine function as shown in the latter part of the plot in Figure 4. The video fluctuations will appear the same whether the surface level is rising or falling, so there is an ambiguity as to which way the surface is moving, however the magnitude of the surface fluctuations can be estimated.

In Figure 3a, the viewed melt surface is all glass. The MMW temperature is steady at 570°C and the video signal shows steady minor surface turbulence with small displacements a fraction of a fringe < 0.1 mm. In Figure 3b, after the first addition of salt, there is a slight lowering of the MMW temperature to 560°C on average with minor fluctuations over the 570 – 550°C range. The video signal indicates larger maximum surface displacements closer to one fringe ~ 0.5 mm in coincidence with the MMW temperature bumps. In Figure 3c, with more salt added the MMW temperature now shows large transitions between two distinct levels of 560 and 440°C. The corresponding video signal shows sharp surface position shifts also of the order of one fringe ~ 0.5 mm coincident with most temperature shifts as well as more gradual position shifts as seen

during the salt phase toward the end of the record in Figure 3c. In Figure 3d, after the final addition of salt, the MMW temperature becomes steady at 440°C corresponding to an all salt view and the video signal returns to a steady minor turbulent one similar to that in Figure 3a.

We interpret the data in Figures 3b and 3c as observations of salt drops flowing around on the fluctuating melt surface level that grow to pools as more salt is added. It is possible to estimate the size of the largest drop in Figure 3b by knowing that the receiver field-of-view has a Gaussian profile (HE_{11} mode waveguide propagation) with a spot size of 18.4 mm ($1/e^2$ diameter). The maximum MMW temperature fluctuation in Figure 3b is 13.8% of the temperature between 570 and 440°C. Assuming the drop flows through the center of the field-of-view, the drop diameter would be about 5 mm. This case is illustrated in Figure 5. The data suggests that there may also be numbers of smaller drops in the regions where the temperature is slightly less than 570°C. In Figure 3c, the size of salt pools need to be larger than the 28 mm diameter of the waveguide to completely cover its field-of-view.

A quantitative measure of the emissivity of the molten glass and salt at 137 GHz was obtained with the aid of the TRR method. Figure 6 shows the TRR data from Figure 2 expanded and overlapped to the same time scale. Not only is the MMW temperature of the glass melt higher than the molten salt, but also the magnitude of the temperature modulation by the thermal return reflection is smaller for the glass melt as expected for a material having a higher emissivity. The equations for interpreting this data need to include the emission and absorption by the waveguide. From Reference [11] they are:

$$T_{eff} = \varepsilon_{wg} T_{wg} + \tau_{wg} \varepsilon_s T_s + r_s \tau_\kappa \tau_{wg} \varepsilon_{wg} T_{wg} \quad (1)$$

$$T'_{eff} = \frac{T_{eff}}{1 - r_s \tau_\kappa r_{bs}^2 \tau_{wg}^2} \quad (2)$$

where T_{eff} and T'_{eff} are the MMW temperatures measured at the receiver without and with the thermal return reflection, respectively, T_{wg} and T_s are the waveguide and melt surface temperatures, ε_{wg} and τ_{wg} are the waveguide emissivity and transmission related by $\varepsilon_{wg} = (1 - \tau_{wg})$, ε_s and r_s are the emissivity and reflectivity of the viewed surface related by $\varepsilon_s = (1 - r_s)$, r_{bs} is the beamsplitter reflectivity, and τ_κ is the viewed surface coupling factor as previously defined.

Using the measured TRR temperature ratios from Figure 6 of $T_{eff} / T'_{eff} = 0.9691$ for the glass and $T_{eff} / T'_{eff} = 0.9368$ for the salt, and the other experimental values: surface temperature, $T_s = 950^\circ\text{C}$, waveguide temperature $T_{wg} = 360^\circ\text{C}$, waveguide transmission $\tau_{wg} = 0.80$, and beamsplitter reflectivity $r_{bs} = 0.54$ to solve Equations 1 and 2 for the emissivity and coupling factor results in $\varepsilon = 0.64 \pm 0.05$, $\tau_\kappa = 0.46 \pm 0.05$ for the glass and $\varepsilon = 0.44 \pm 0.05$, $\tau_\kappa = 0.60 \pm 0.05$ for the salt. The MMW emissivity of the molten salt is 32% lower than that for the molten glass. The difference in coupling factor suggests that the molten salt surface is a smoother, flatter surface (due to its low viscosity hence smooth covering) than the glass melt at these wavelengths.

3. Conclusions

Non-contact millimeter-wave measurements at a frequency of 137 GHz were used for the first time to detect the thermal emission and reflectivity from the molten surface

of Defense Waste Processing Facility black frit glass as sodium sulfate salt was added. A large difference in the MMW surface emissivity of 32% between that of the glass melt and molten salt has been observed. Interesting dynamics in the formation of a salt layer are also revealed as the formation of smaller drops of molten salt that grow to larger pools which flow around the surface of the melt before a continuous layer is established. Partial coverage of the glass melt surface with molten salt causes a noticeable increase in the amplitude of melt level fluctuations on the millimeter scale. This increase in fluctuations could be used as an indicator of the onset of molten salt pooling in a melter long before a continuous layer is formed, if the mechanisms for formation are the same for salt coming out of solution as for salt added from the top.

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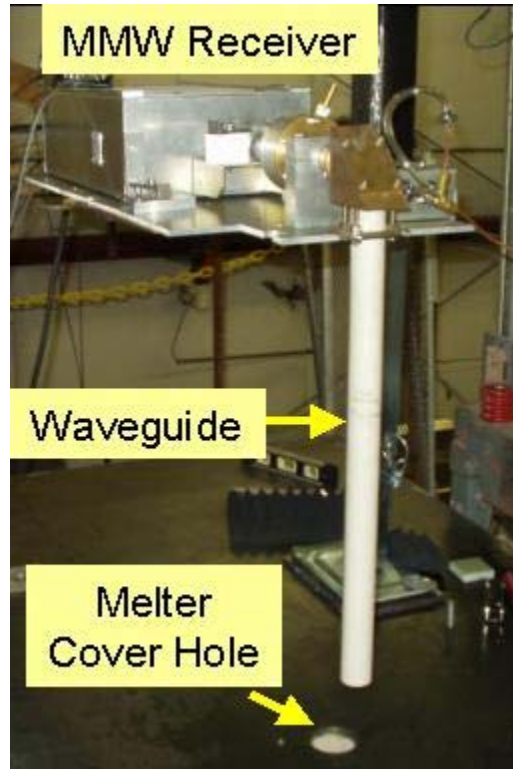


Figure 1. Millimeter-wave receiver and waveguide setup above the EV-16 joule-heated melter at CETL. The receiver is on a vertically translatable platform for lowering the waveguide through the cover to the melt surface.

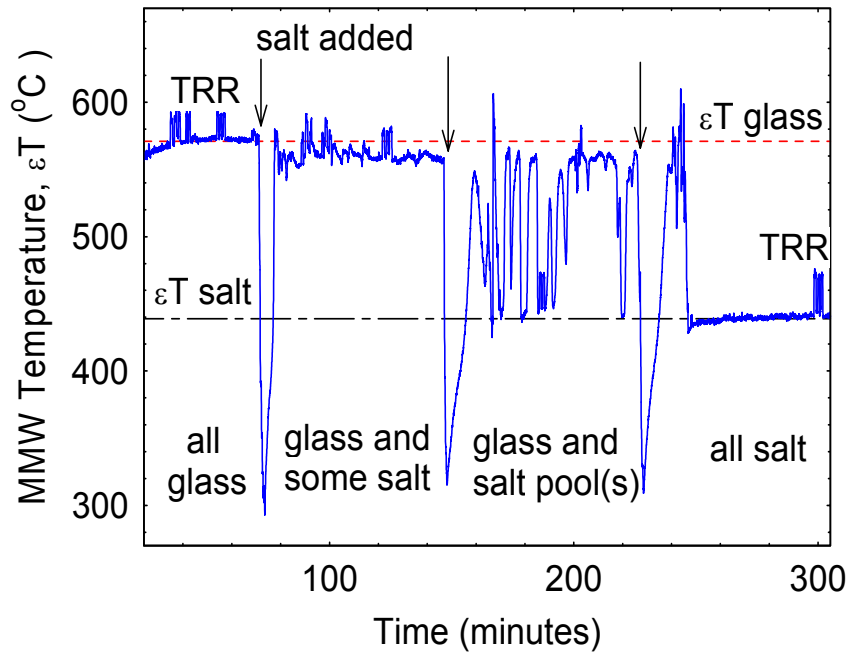


Figure 2. MMW thermal signal as sodium sulfate salt is added to DWPF black frit molten glass at three different times.

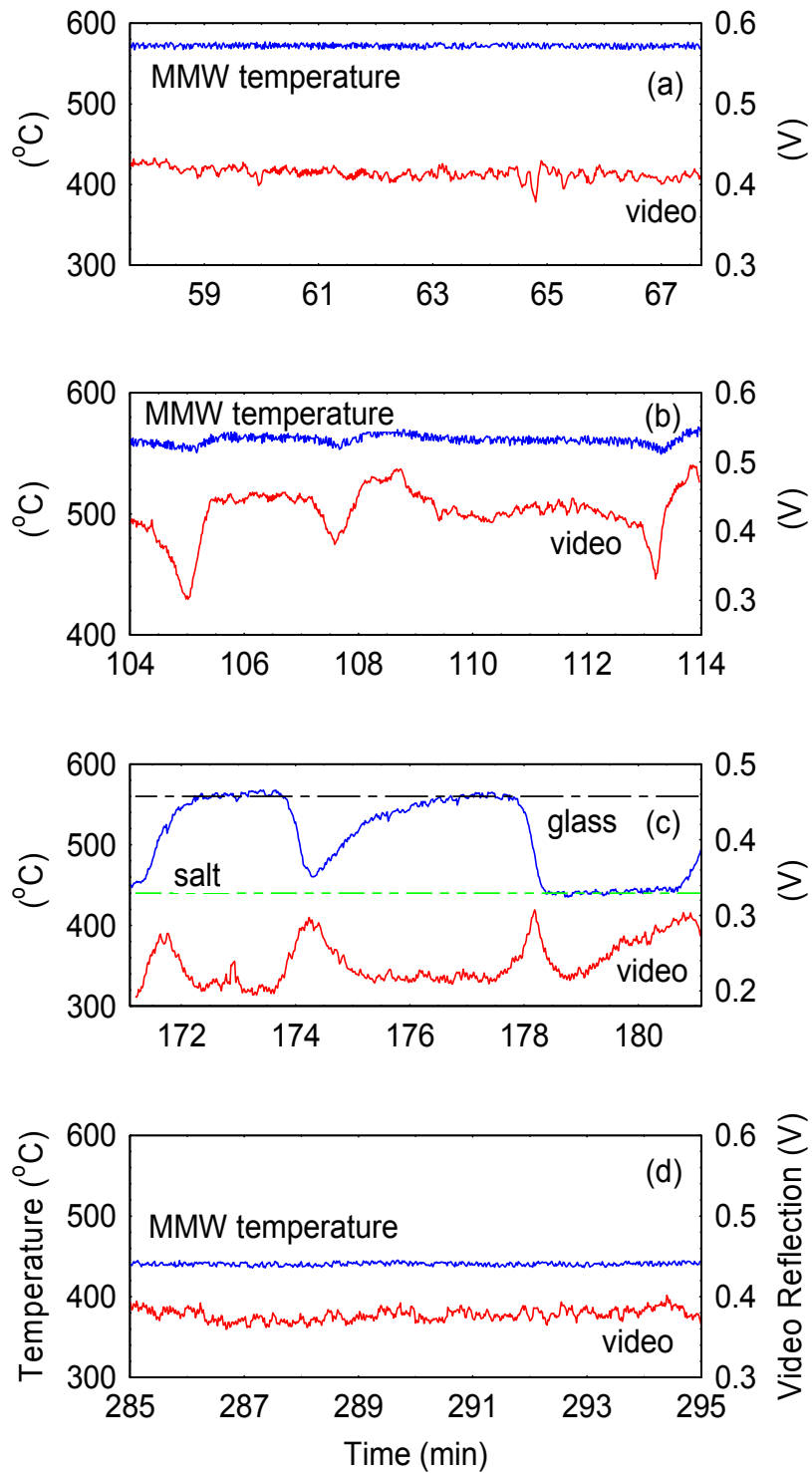


Figure 3. Expanded ten-minute time segments of MMW temperature and video reflection for the four periods between salt additions of Figure 2, a) all glass surface, b) some salt, c) more salt, d) all molten salt surface.

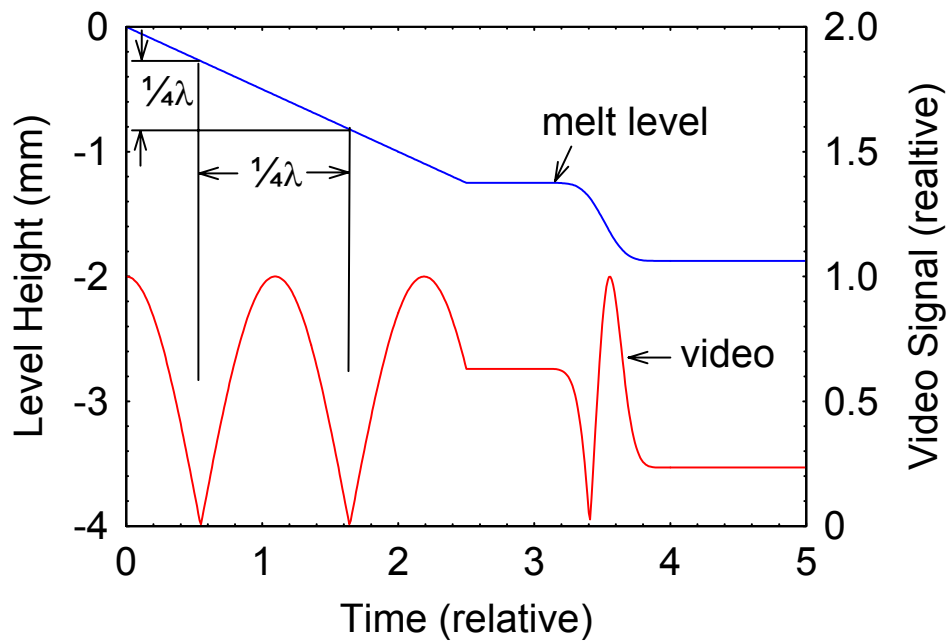


Figure 4. Calculated illustration of the relationship between melt level height change and a millimeter-wave video reflection signal at a frequency of 137 GHz ($\lambda = 2.2$ mm).

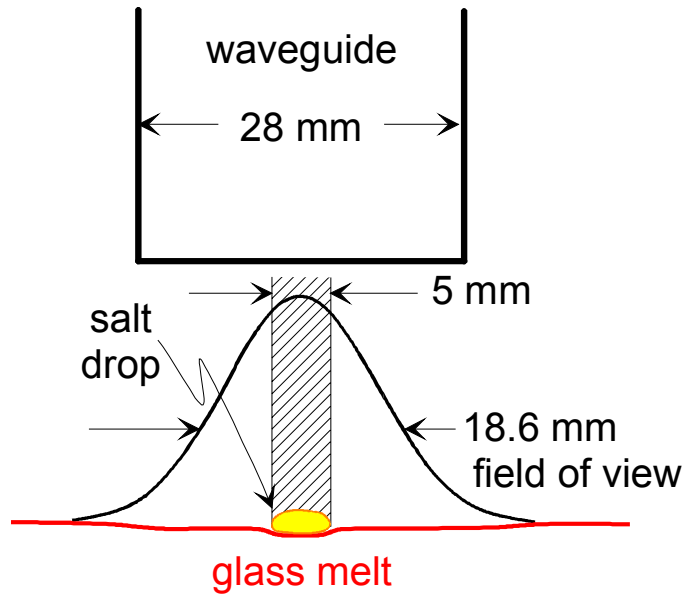


Figure 5. Illustration of 13.8% occultation of the receiver Gaussian field-of-view by a 5 mm diameter salt drop as suggested by the data in Figure 3b. Distance of the waveguide from the melt surface is exaggerated.

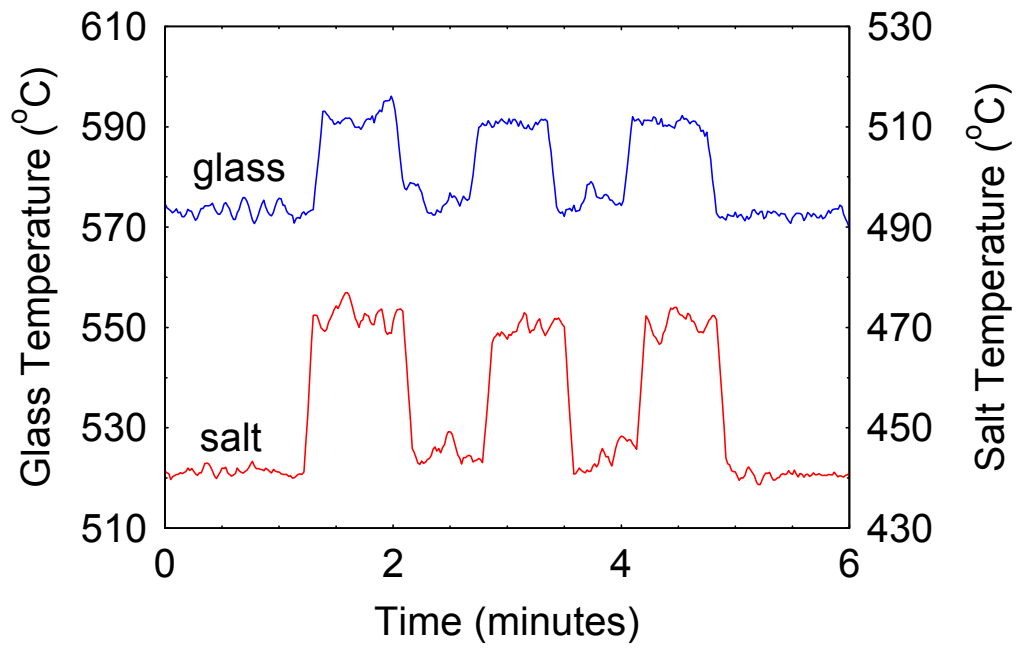


Figure 6. MMW thermal return reflection measurements through an 80% transmitting warm waveguide ($\sim 360^{\circ}\text{C}$) of molten glass and salt at a thermocouple temperature of 950°C .