



**Pacific Northwest
National Laboratory**
Operated by Battelle for the
U.S. Department of Energy



Millimeter-Wave High Level and Low Activity Waste Glass Research

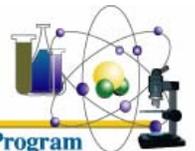
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EMSP HLW Workshop, Aiken, SC
January 18-21, 2005



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Problem Statement

- Current and planned waste vitrification operations use predictive models without on-line monitoring
- Vitrification efficiencies are limited by uncertainties in models and operating parameters
- Melter operations are vulnerable to disruptive anomalies that cannot be predicted
- Identified monitoring needs:
 - Viscosity
 - Salt layer formation
 - Foaming
 - Liquidus crystals
 - Noble metals buildup
 - Pour spout flow

Research Objectives

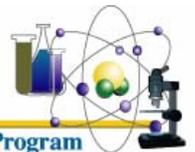
Reduce Immobilization Costs and Risks

- Develop new robust diagnostic tools for glass melt monitoring in high temperature and radioactive environments
- Exploit these new tools to advance vitrification materials science as it relates to nuclear waste and glass melt property modeling
- Progress toward a goal to modernize nuclear waste glass vitrification operations\control to industrial standards

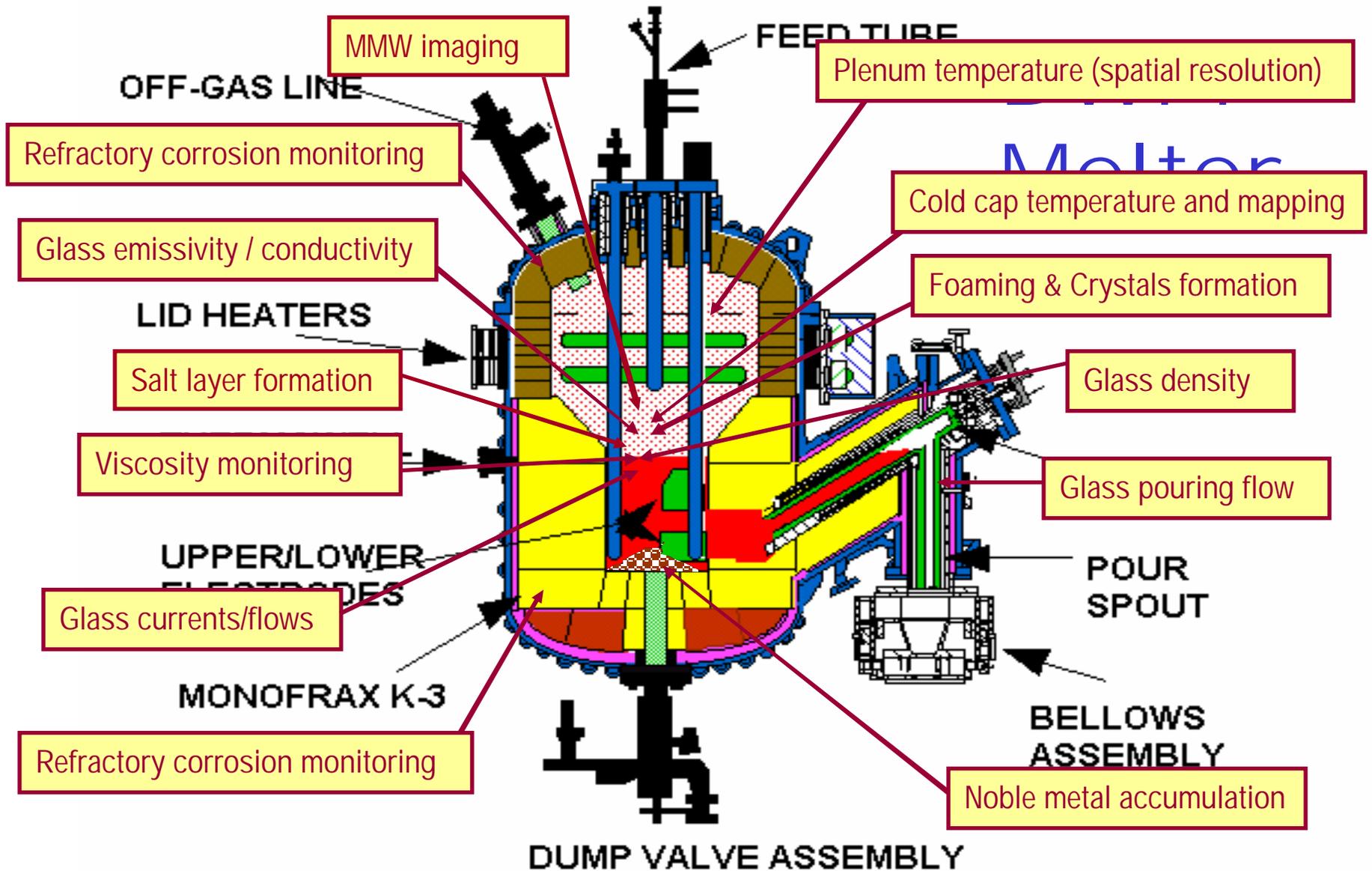


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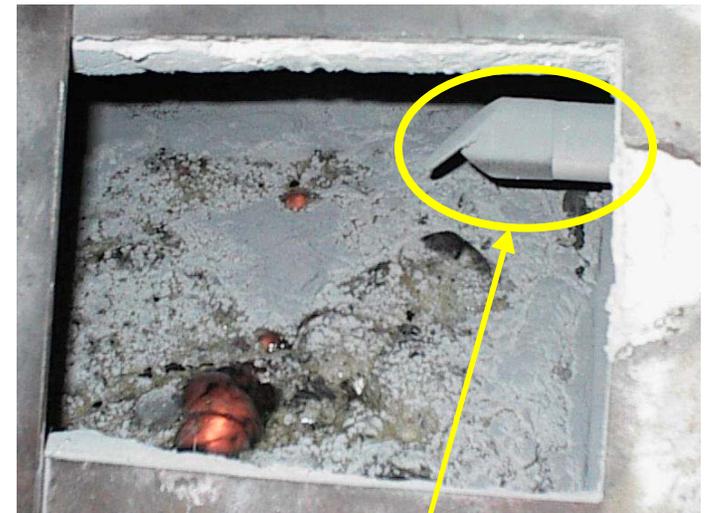
Long Range Vision of Research



Approach

Millimeter-Wave Technology (10-0.3 mm, 30-1000 GHz)

- Wavelengths are long enough to penetrate optical/infrared obscure viewing paths through dust, smoke, and debris
- Wavelengths are short enough for spatially resolved point measurements and profiles
- Waveguide/mirror components can be fabricated from the same refractory materials as the melter



MMW Inconel mirror and mullite guide



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Summary of Achieved Development

New Millimeter-Wave (MMW) Techniques and Technologies Developed for Melter Monitoring

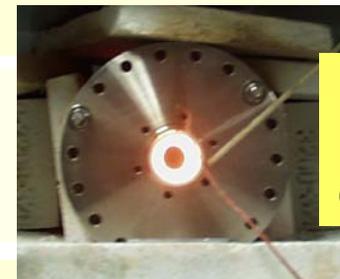
- Thermal Return Reflection (TRR)

Use thermal emission as a probe to resolve emissivity and temperature

- Reflective Interferometry

Monitor fluid displacement, fluctuations, and flow (viscosity)

- High Temperature ($> 1000^{\circ}\text{C}$)
Efficient Waveguides



1200 °C
mullite
guide

- Efficient Quasi-Optical MMW Signal
Control components

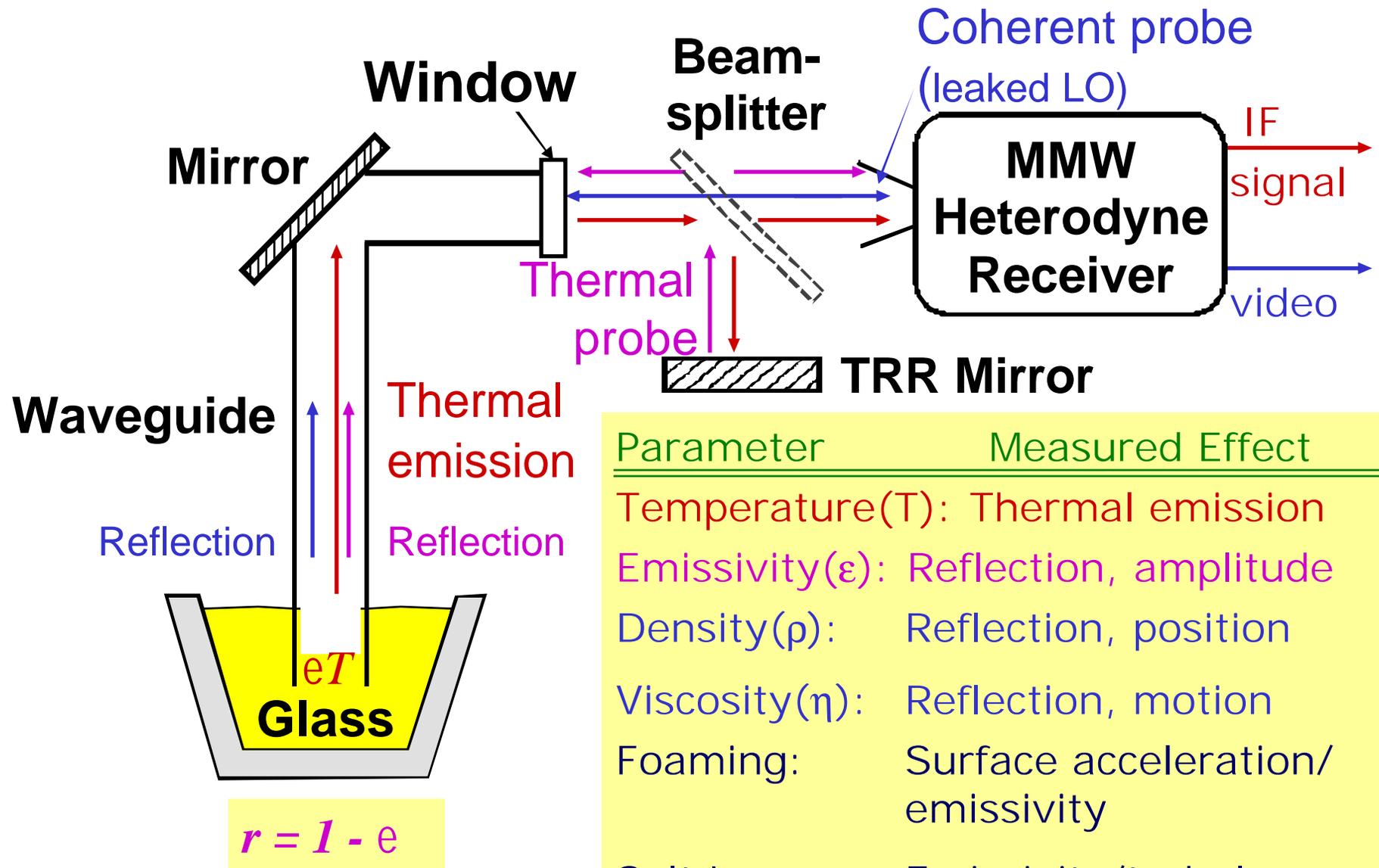


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MMW Sensor Configuration



Parameter	Measured Effect
Temperature(T):	Thermal emission
Emissivity(ϵ):	Reflection, amplitude
Density(ρ):	Reflection, position
Viscosity(η):	Reflection, motion
Foaming:	Surface acceleration/emissivity
Salt Layer:	Emissivity/turbulence



Summary of Accomplished Research

In the Laboratory

- High Temp. MMW properties of materials investigated (chromium oxide (Cr_2O_3), alumina-zirconia-silica (AZS), Monofax K3, NaSO_4 salt)
- Melt flow and MMW fluctuations investigated for viscosity (Hanford #7 & #8, DWPF black frit #265, commercial E glass)
- MMW signature of foaming event observed

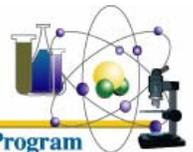
In the Field (Clemson EV-16 Melter)

- 2-D cold cap temperature profiles measurements
- Viscosity depth profile measurements
- Salt layer formation studied



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Viscosity and Salt Field Test

Clemson Environmental Technology Lab (CETL)

- EV-16 Joule Heated Glass Melter (up to 1150 °C)
 - 45.7 cm (18 inch) square melt pool,
filled to 20.3 cm (8 inch) depth
- Defense Waste Processing Facility (DWPF) black frit #265 glass --- 111 kg (245 lbs)

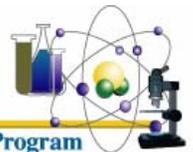
MMW Measurements:

- Glass flow versus depth and temperature
- Salt layer formation and emissivity of glass and salt --- 1.9 kg (4.2 lbs) sodium sulfate added

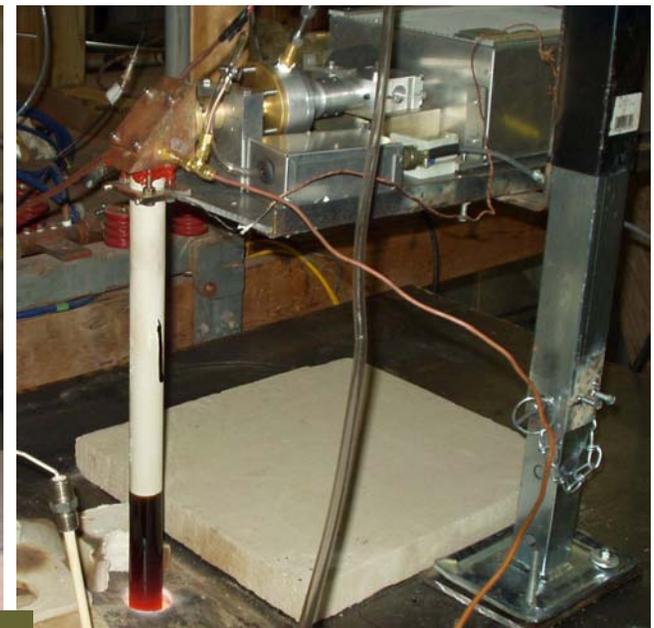
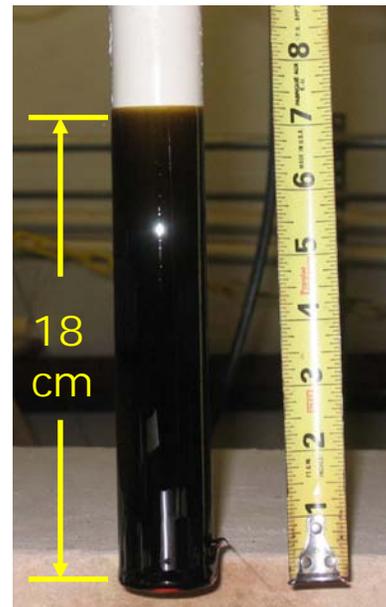
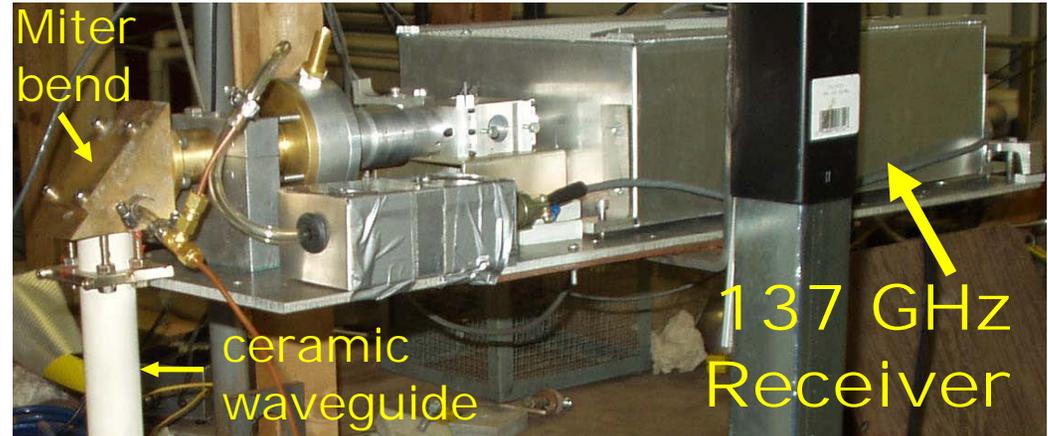


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Experimental Setup



Waveguide after immersion



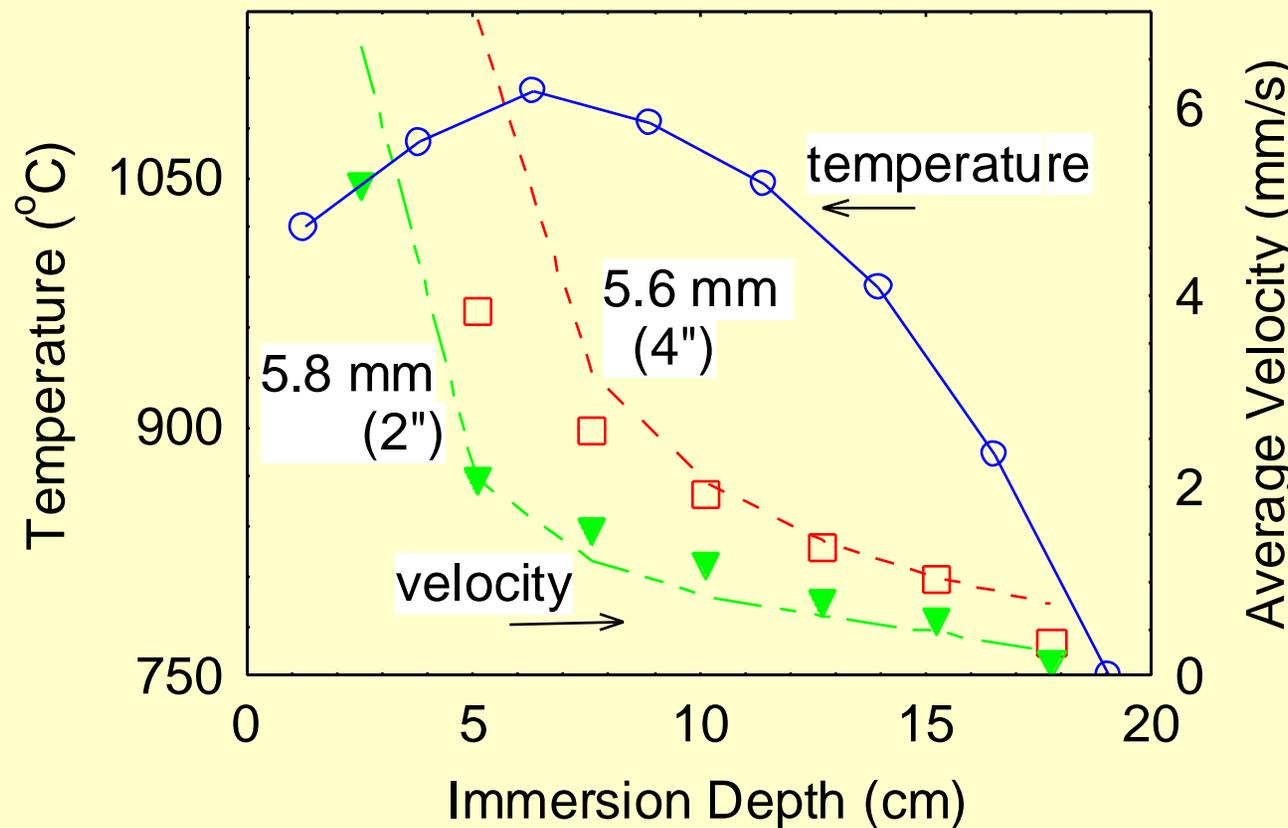
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Glass Flow Measurements

- Waveguide flow measured as a function of immersion depth for 2.8 mm and 5.6 mm Hg pressurizations



Points are measurements

Dashed lines are theory

$$v_{ave} = Ar \frac{z_0}{(L - z_0)h}$$

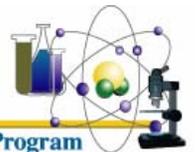
1/T scaling of viscosity (η) assumed

Good agreement between theory and experiment for large depths



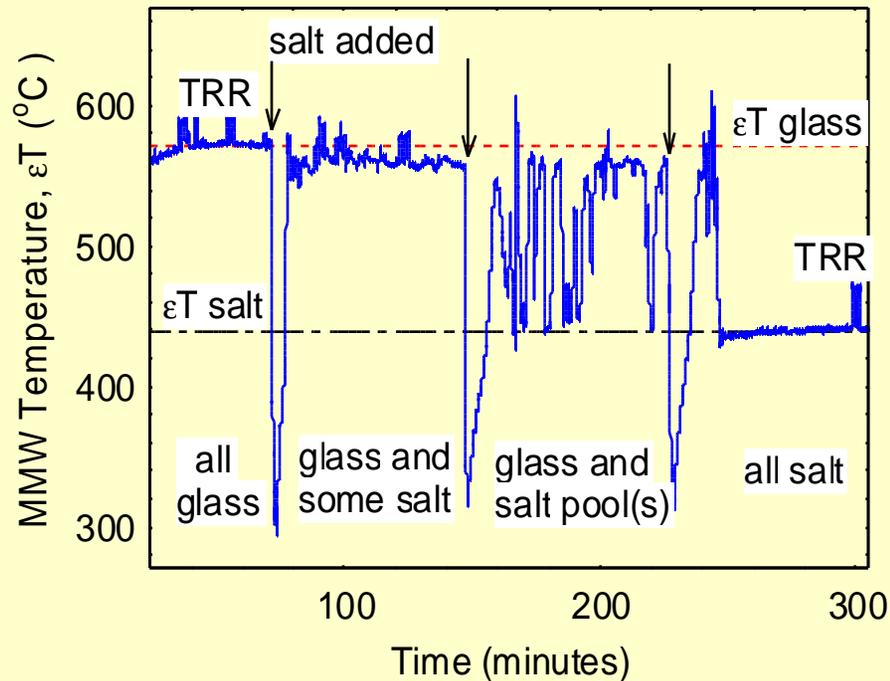
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Salt Experiments

MMW Temperature Signal
proportional to ϵT



- Salt (NaSO_4) added at 3 times shown
- Thermocouple temperature steady between additions



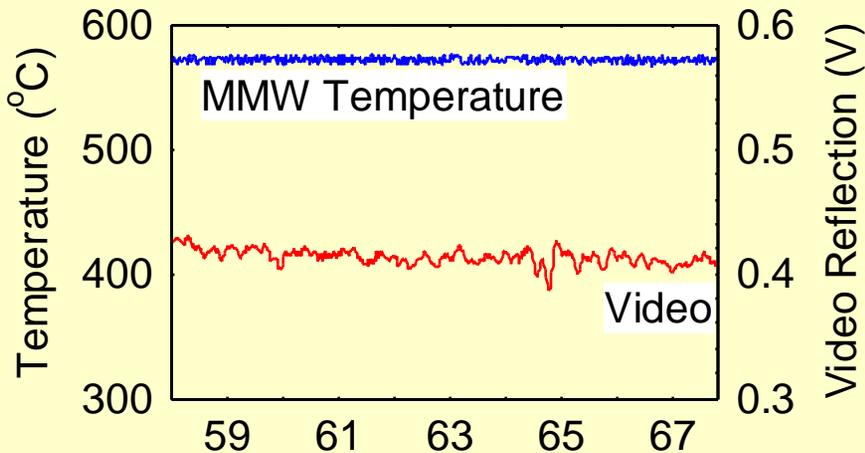
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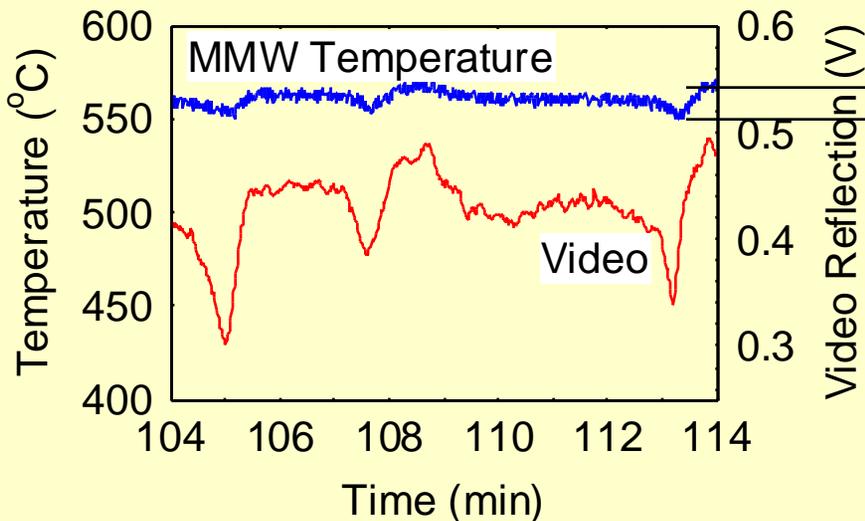


Salt Data Analysis

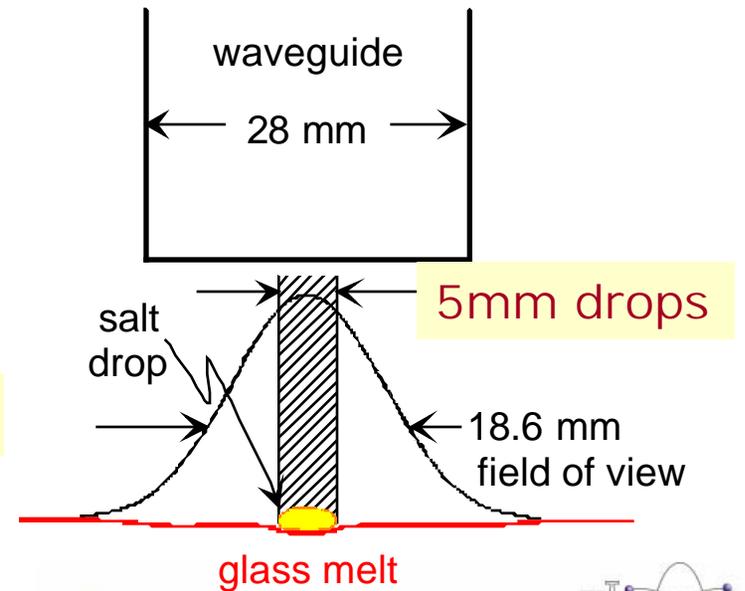
All Glass



Some Salt

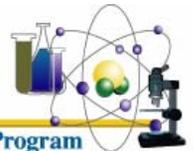


- MMW surface fluctuations occur with salt
- Salt in discrete drops
- Drop size can be estimated from data

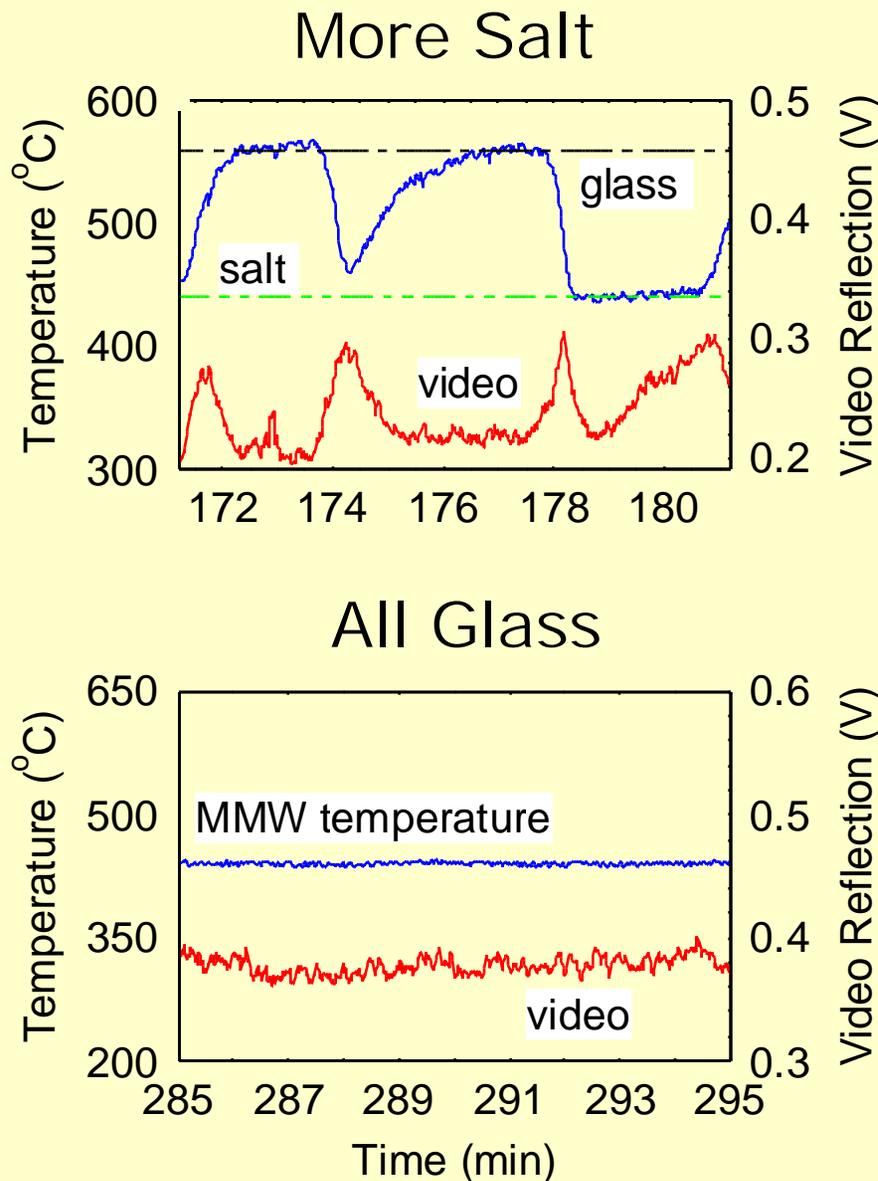


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Salt Data Analysis

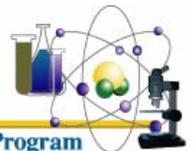


- With more salt drops grow into larger pools > 28 mm diameter
- Salt pools are in constant flow on melt surface
- 4.2 (1.9 kG) of NaSO_4 established continuous layer on 18^2 in^2 (46^2 cm^2) glass surface --- layer depth ~3.3 mm



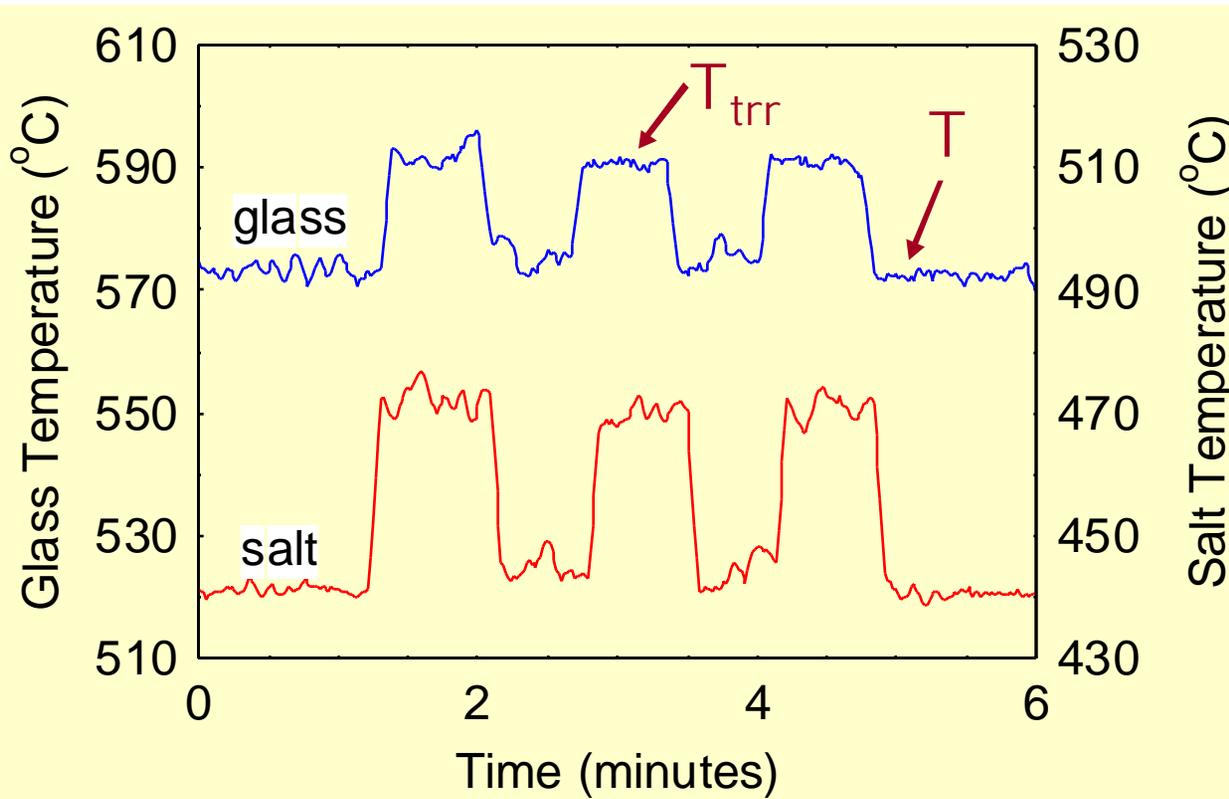
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Salt and Glass Emissivity

- Emissivity and surface figure determined by thermal return reflection method and thermocouple



At 950°C &
137 GHz

DWPF Glass:

$$\epsilon = 0.64 \pm 0.05$$

$$\kappa = 0.46 \pm 0.05$$

NaSO₄ Salt:

$$\epsilon = 0.44 \pm 0.05$$

$$\kappa = 0.60 \pm 0.05$$

$$e = 1 - \frac{1}{kr_{bs}^2 t_{wg}^2} \left(1 - \frac{T}{T_{trr}} \right)$$

Woskov & Sundaram,
J. Appl. Physics, **92**,
6302, 2002.



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Planned Research

- Obtain more data on salt layer formation with future planned waste glasses (regular and bulk vitrification processes)
- Compare MMW glass flow measurements with other viscosity data, improve modeling, and research limits of melt viscosity and specific gravity measurements
- Obtain more data and model foaming of melts and correlate foaming to processing conditions and glass chemistry
- Research measurements and interpretation of melt surface fluctuations (Maragoni force)



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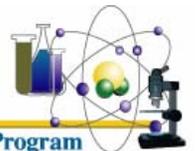
Planned Research continued

- Investigate more sensitive emissivity monitoring for melt parameters such as liquidus (dual receiver system, in situ reference)
- Implement MMW receiver system for more laboratory research at ACTL (run melt tests at ACTL)
- Carry out additional tests relevant to DWPF/WTP



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Further Reading

“Millimeter-Wave Monitoring of Nuclear Waste Glass Melts – An Overview”, *P. P. Woskov, J. S. Machuzak, P. Thomas, S. K. Sundaram, and W. E. Daniels, Jr.*, in Environmental Issues and Waste Management Technologies VII (*Ceramic Transactions, Volume 132*) pp. 189-201, 2002.

“Cold Cap Monitoring using Millimeter Wave Technology”, *S. K. Sundaram, P.P. Woskov, J.S. Machuzak, and W.E. Daniel, Jr.*, in Environmental Issues and Waste Management Technologies VII, Editors: G. L. Smith, S. K. Sundaram, and D. R. Spearing (*Ceramic Transactions, Volume 132*) pp. 203-213, 2002.

“Thermal return reflection method for resolving emissivity and temperature in radiometric measurements”, *P. P. Woskov and S. K. Sundaram*, *J. Appl. Physics*, vol. 92, 6302-6310, 2002.

“Molten salt dynamics on glass melt using millimeter-wave emissivity measurements”, *P. P. Woskov, S. K. Sundaram, W. E. Daniel, Jr., D. Miller*, *J. of Non Crystalline Solids*, vol. 341/1-3, 21-25, 2004.



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