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In-situ visual observation of glass melting processes

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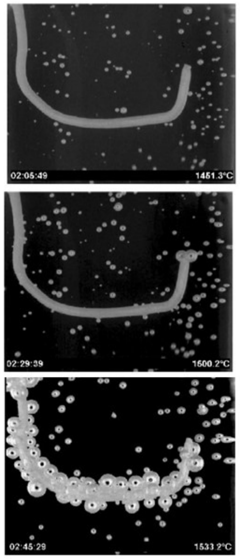
Motivation

- High-temperature visual observation (HTVO)
    - Used to understand processes during glass production + improve the overall process efficiency
- Due to experimental simplicity, HTVO has been used to investigate:
    - Bubble nucleation
    - Fining
    - Refractory corrosion



Bubble nucleation in commercial glassses

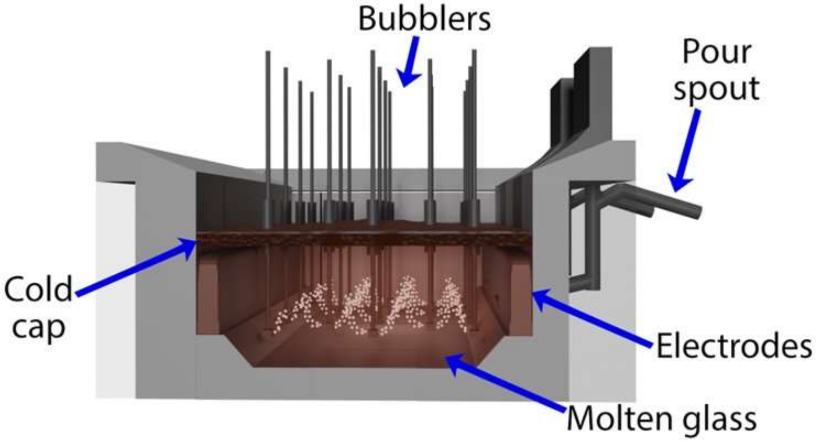
Klouzek, J., Arkosiova, M., and L. Nemec. Redox of Sulphur in glass melts (2006).

- For commercial batch melting and nuclear waste vitrification, HTVO is a cost-effective tool to study:
    - Influence of composition, atmosphere, and makeup (particle sizes, crystalline phase, loose batch vs pressed pellets or slurry) on conversion processes
    - Phenomena that influence the rate of glass production

History of batch observations

- Commercial glass furnaces
  - Viewports placed in furnace
  - Monitor batch blanket, fining, corrosion of refractory

- WTP melter
  - Remotely operated without visual feedback



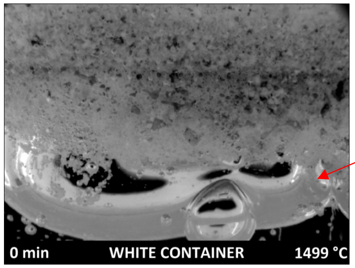
- Laboratory-scale experiments
  - Performed using batch samples in quartz glass crucibles

	25°C	750°C	800°C	850°C
VSL HAL-Na crucible				

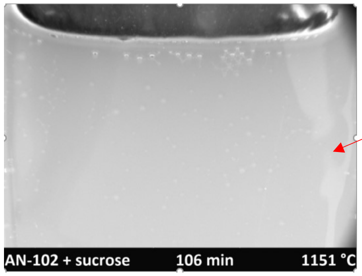
HTVO experimental setup

- Advantages
  - In-situ high-temperature imaging
    - Furnace construction and camera selection
  - Rapid (automated) image acquisition
  - Can control atmosphere or perform evolved gas analysis (using mass spectrometer)

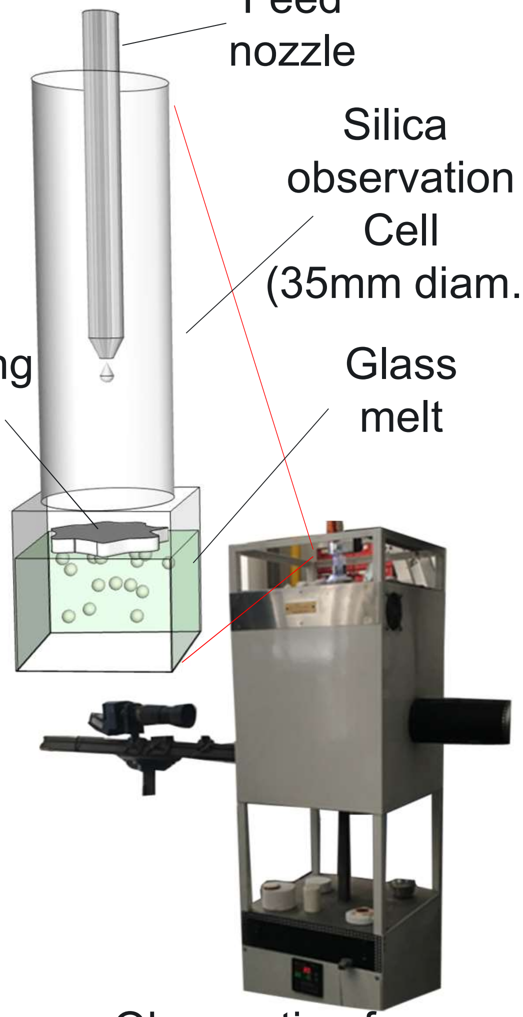
- Disadvantages
  - 2D imaging technique (stereographic limitations)
  - Imaging of reaction layer requires transparent melts
  - Foam bubbles adhere to walls; obstruct view



Large cavity



Opaque melt



Feed nozzle

Silica observation Cell (35mm diam.)

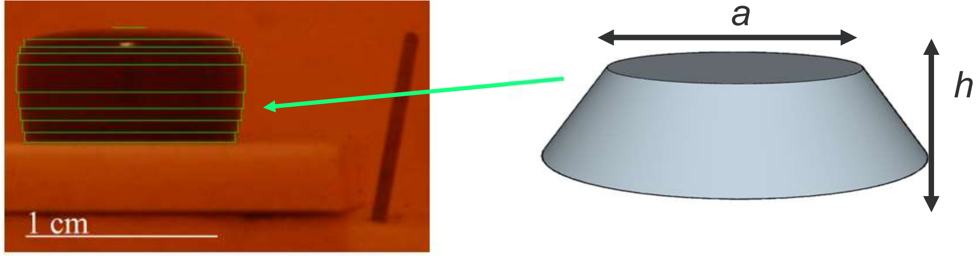
Reacting Feed

Glass melt

Observation furnace

HTVO: Feed expansion test (FET) volumetric expansion

- Using pellets allows for easy calculation of volume and porosity
  - Assume that feed pellets have rotational symmetry during melting
  - Approximate volume as stacked cylinders:

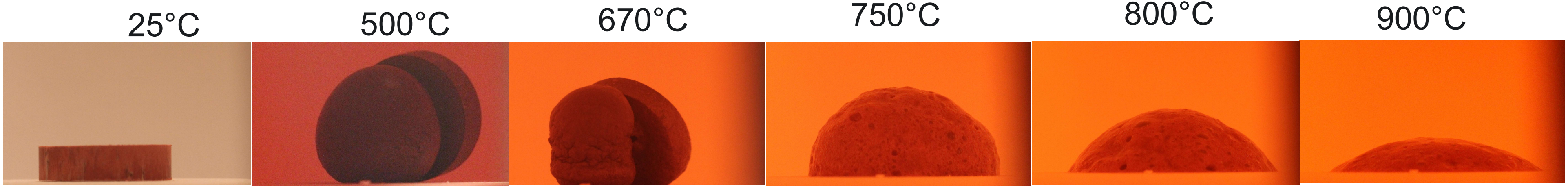


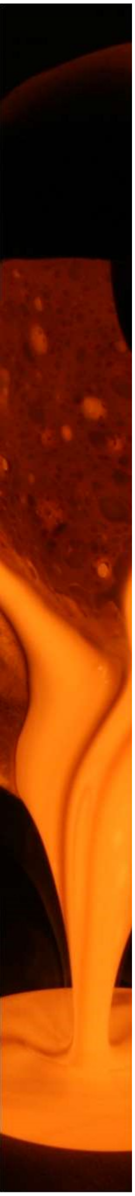
$$V_i = \frac{\pi}{12}\left(a_i^2 + a_i b_i + b_i^2\right) h_i$$

$$V = \delta^3 \sum_i V_i \qquad \delta = scale\ factor$$

Hilliard, Z. and P. Hrma. J. Am. Ceram. Soc. 1-8 (2015).

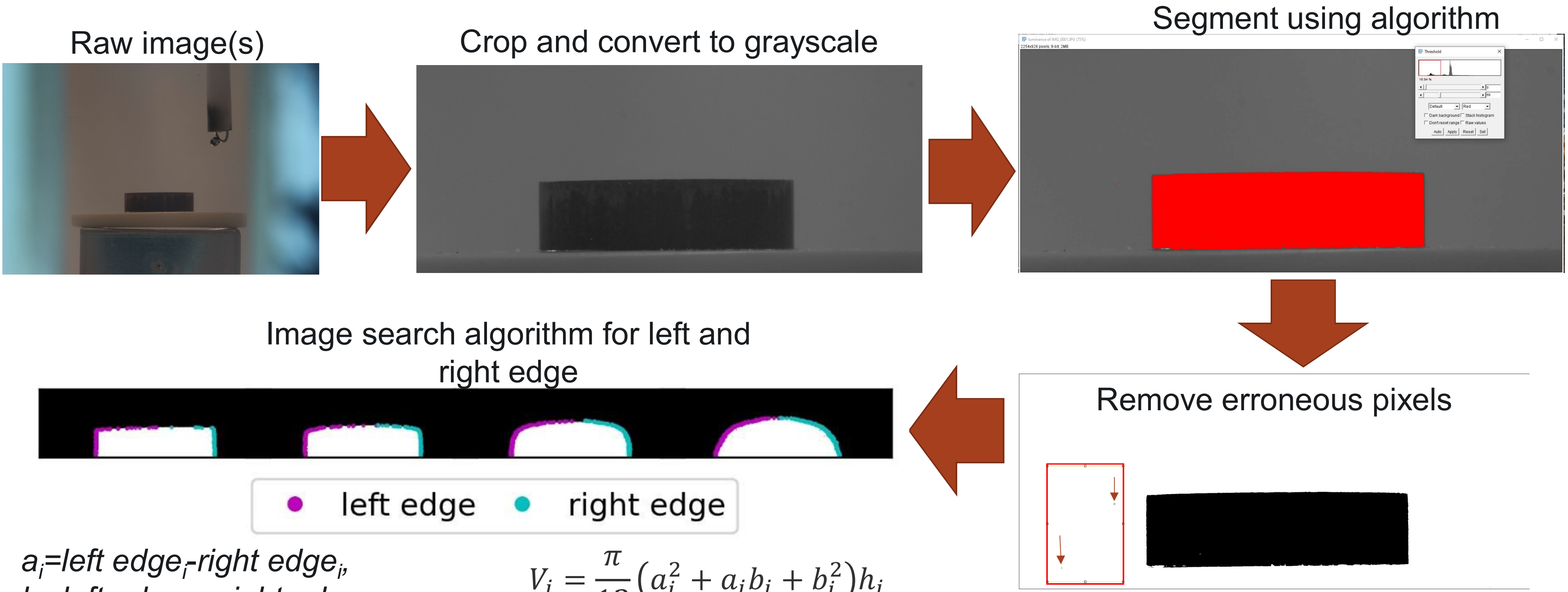
- Development of a batch processing approach reduces user variability and measurement time
- However, under certain cases rotational symmetry is broken (overcome using X-ray Computed Tomography, XCT)





HTVO: Feed expansion test (FET) volumetric expansion

- ***Batch image processing workflow:***



Raw image(s)

Crop and convert to grayscale

Segment using algorithm

Remove erroneous pixels

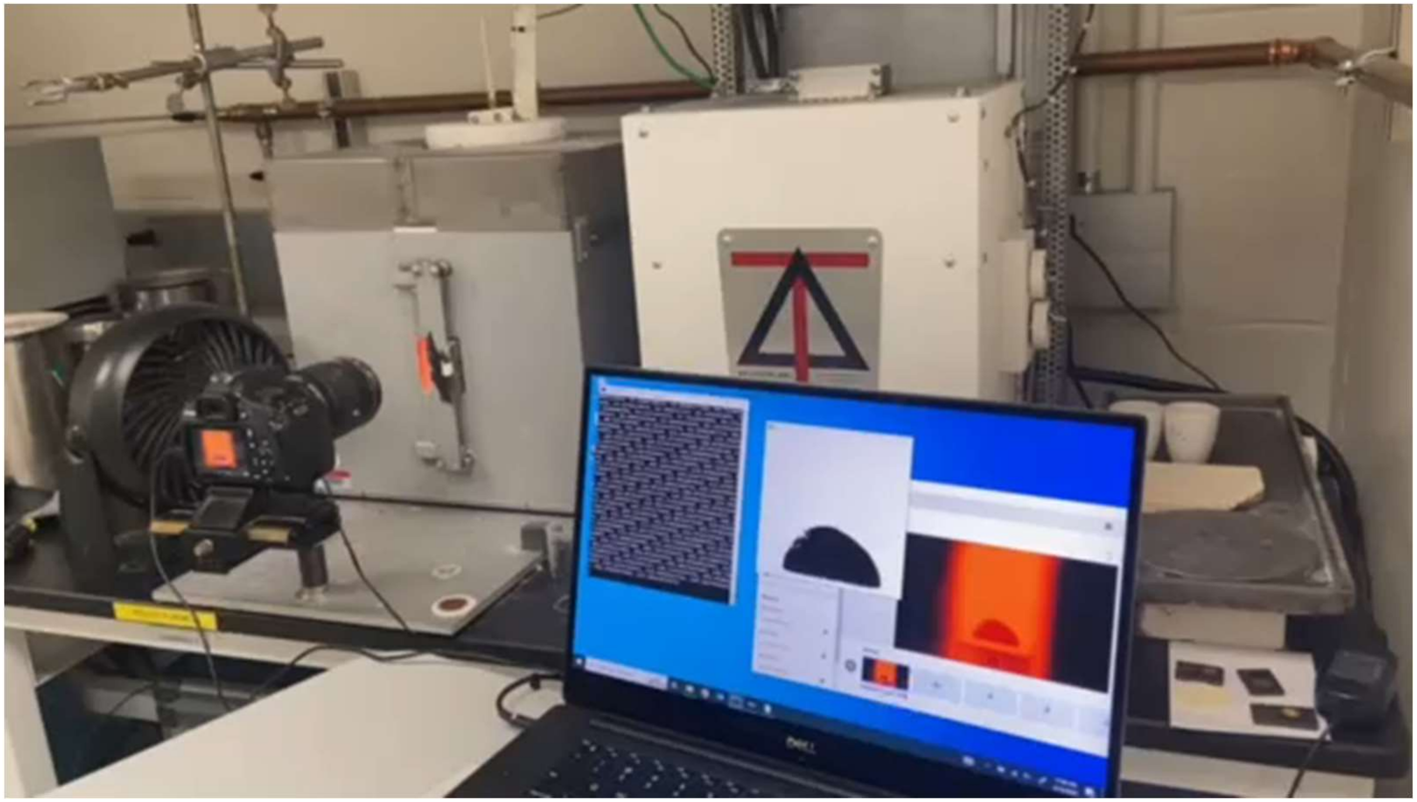
Image search algorithm for left and right edge

a_i=left edge$_i$-right edge$_i$,
b_i=left edge$_{i+1}$-right edge$_{i+1}$
h_i=1 pixel
i=pixel number

$$V_i = \frac{\pi}{12}\left(a_i^2 + a_i b_i + b_i^2\right)h_i$$

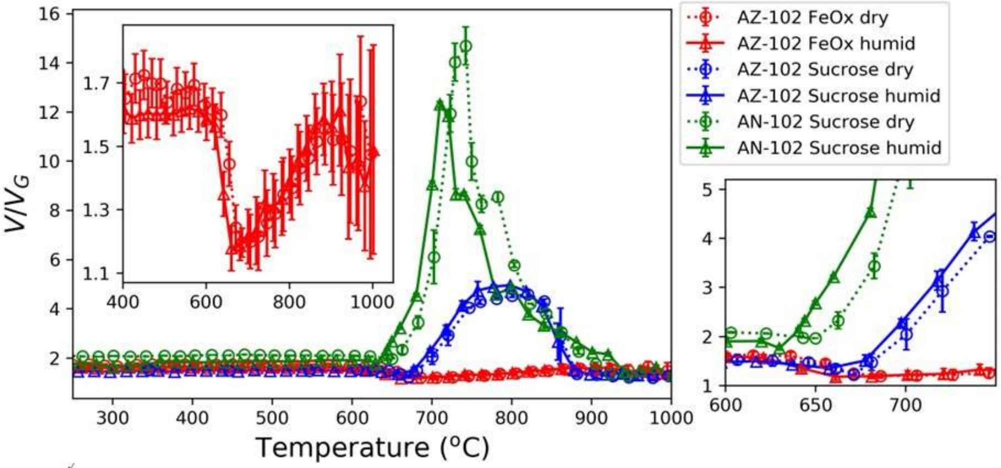
$$V = \delta^3 \sum_i V_i$$

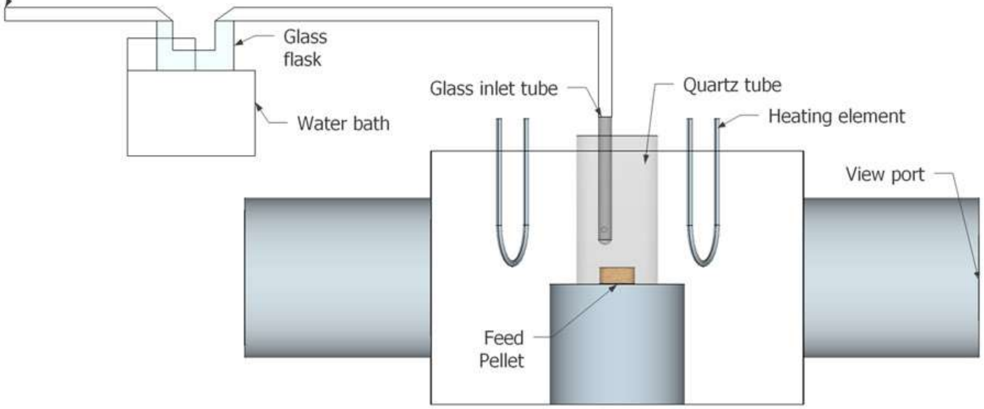
Real-time imaging and analysis

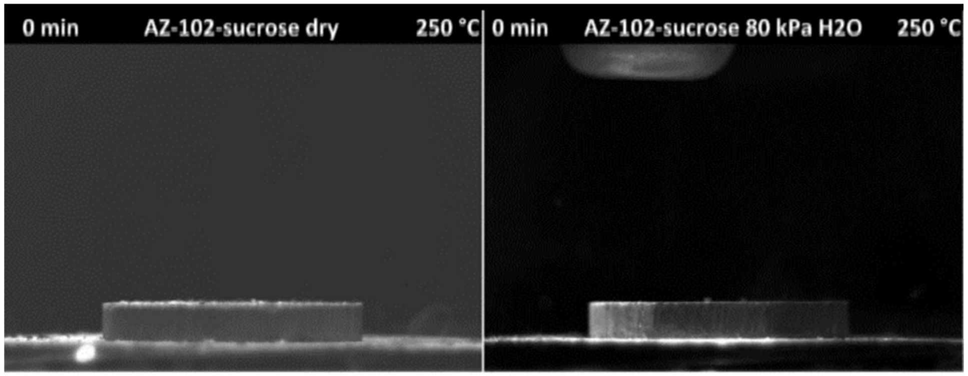


FET in water vapor atmospheres

- Capability of testing expansion under water vapor atmospheres
    - Water vapor is known to reduce surface tension and viscosity of melts
    - Closes pores and traps evolving gases
- Correlated to literature on commercial glass melting



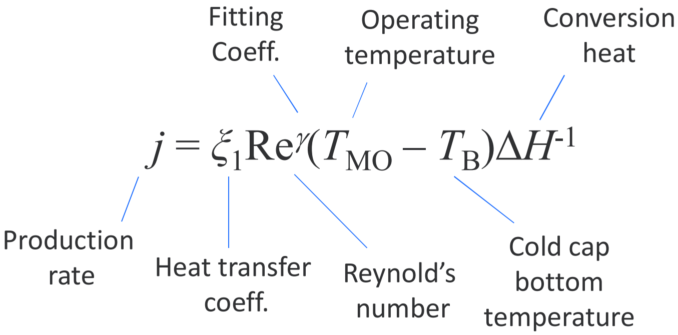




J. Marcial et al, Int. J. Appl. Glass. Sci. 12(1) 145-157 (2020)
https://doi.org/10.1111/ijag.15803

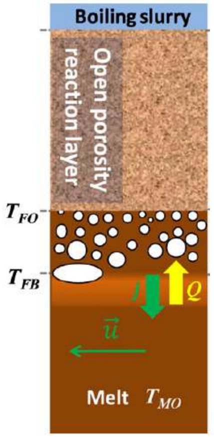
Correlation to large-scale melter

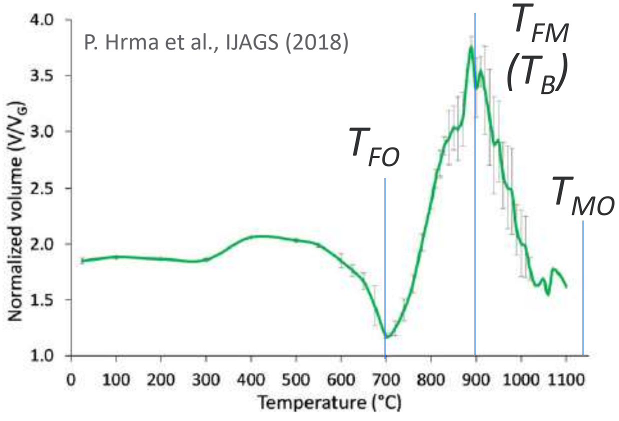
- Temperature of foaming maximum from pellet FET can be correlated to the temperature at the bottom of the cold cap
  - Originally tested using HLW feeds
- Developed for electric melters for nuclear waste vitrification with bubblers
  - Maybe applied to commercial glass melting furnaces



$$j = \xi_1 \mathrm{Re}^{\gamma}(T_{\mathrm{MO}} - T_{\mathrm{B}})\Delta H^{-1}$$

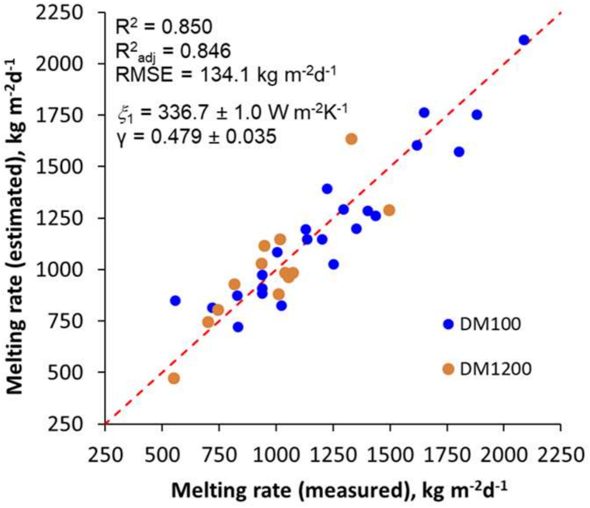
S M Lee et al., Simplified melting rate correlation for radioactive waste vitrification in electric furnaces. *Int. J. Appl. Glass Sci.* (2020) doi:10.1111/JACE.17281





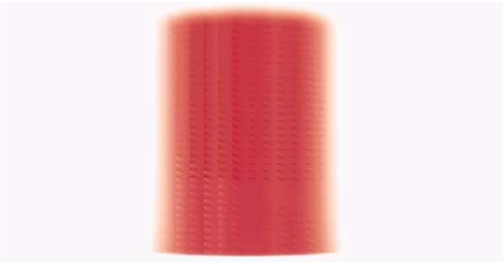
P. Hrma et al., IJAGS (2018)

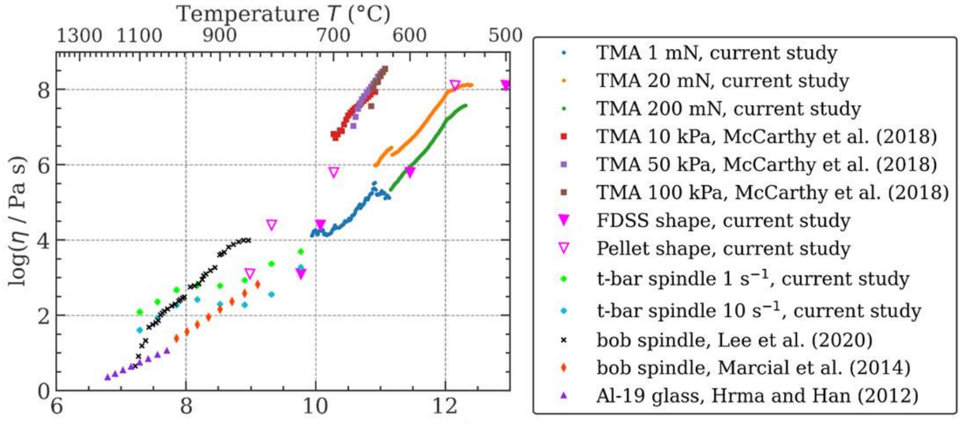
(b) Simplified MRC



Implementation of HTVO for viscosity measurements

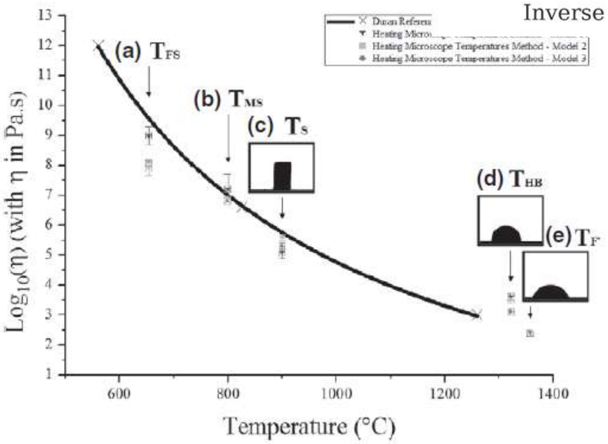
- Relationships identified among glass pellet shape and melt viscosity using “glass-slump-test”
- Could offer potential to understand transient viscosity under various conditions





Comparison of visual observation to measured apparent viscosity

J. George et al., subm. JACerS (2023).



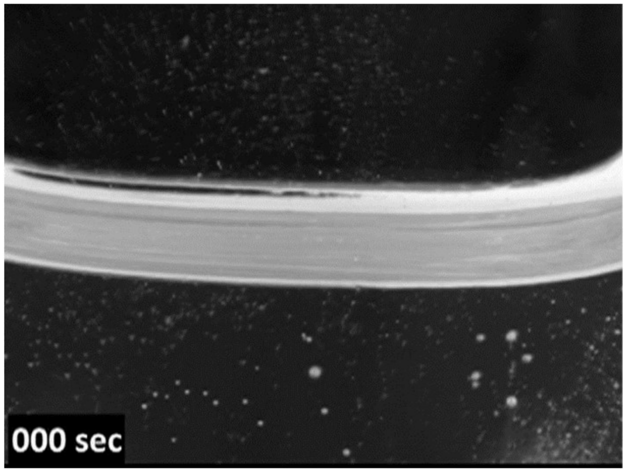
Geometric shapes corresponding to fixed viscosity points

F. Montanari et al., (2014).

Charging feed pellet onto melt

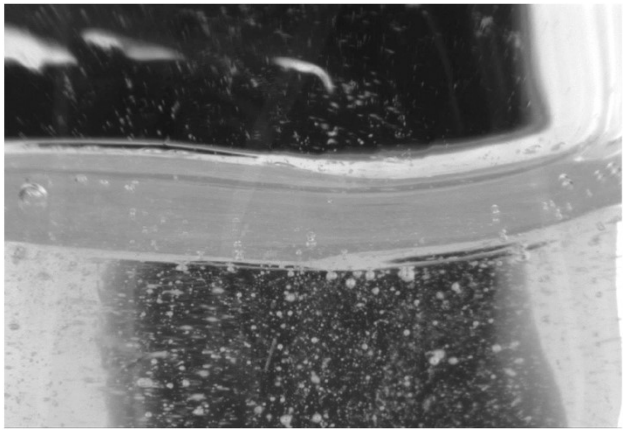
- Formulated transparent LAW glass melt by omitting transition metal glass-forming additives
- Melting was achieved at 1150°C
- Allows for real-time observation of interface between reacting feed and melt
  - Improved contrast using dark feed
- Further modifications to experimental protocol required
  - Feed pellets could not be placed flat on melt surface
  - Coalescence of bubbles to large cavities observed

White feed charged onto clear LAW melt



Observation cell

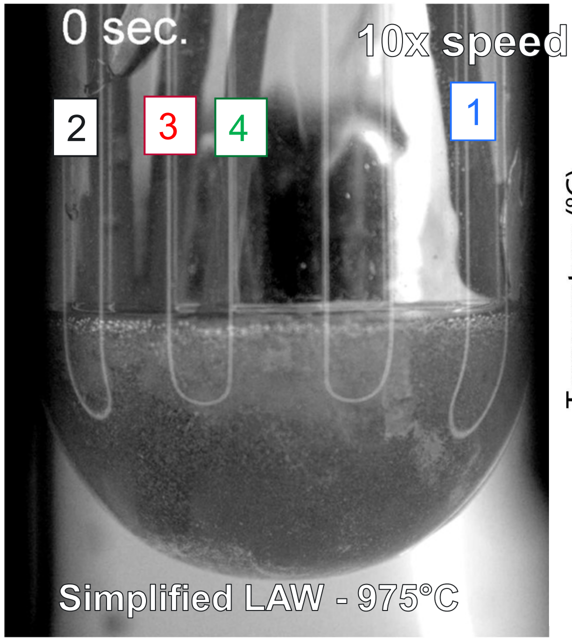
Dark feed charged onto clear LAW melt

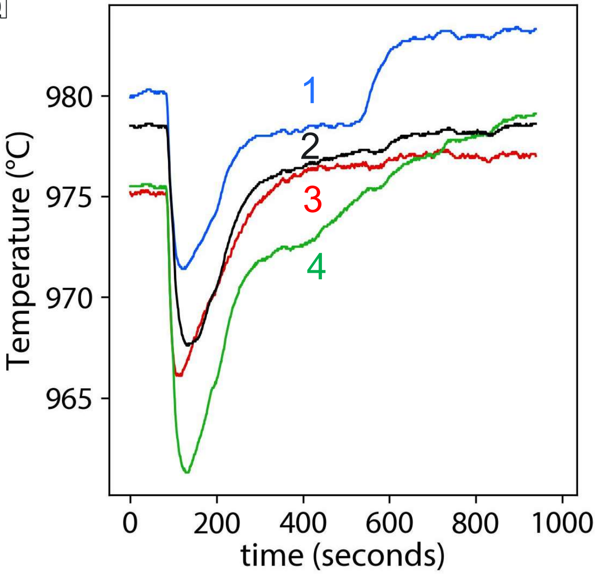


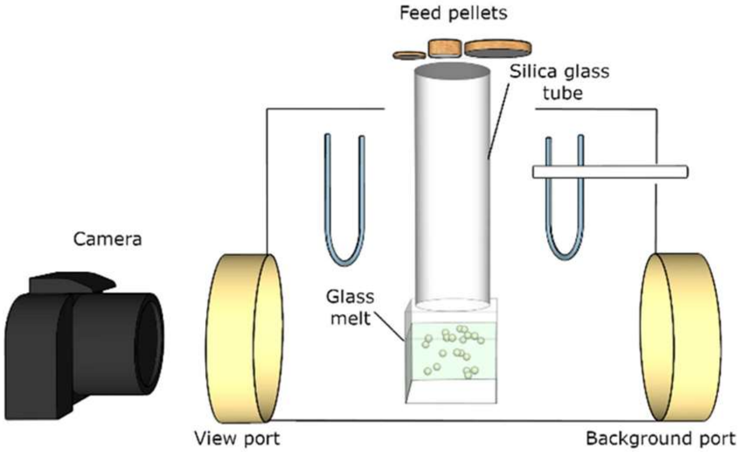
J. Marcial et al., Ceram. Int. (2022)

Charging feed pellet onto melt

- Immersing thermocouples in melt was performed as proof-of-concept to understand temperature profile
- Despite using transparent melt; the thermocouple placement was not easily visible
  - Metal leaching from thermocouples colored the melt while bubbles occluded view

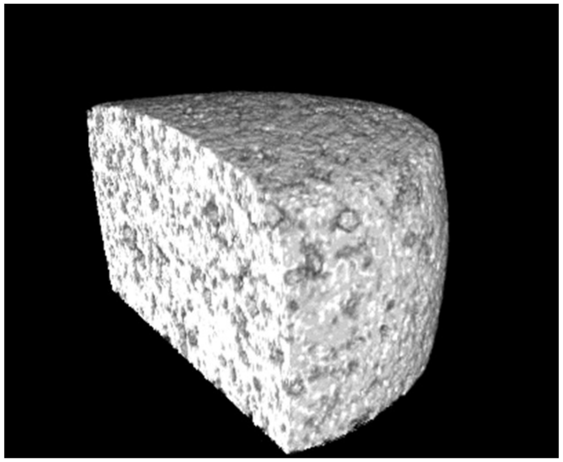






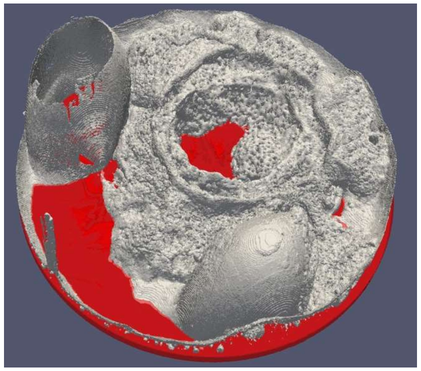
X-ray methods

- Traditionally feed volume has been determined by observing profiles
  - Assumes rotational symmetry of feed samples
- X-ray CT: no assumptions of the sample geometry
  - Directly observe pore structure within samples
  - Can measure multiple samples at once
- Utilizing 2D imaging, foaming interfaces can be observed (shortened time resolution)



X-ray CT 13mm diameter of HLW-AI19 pellet

Courtesy of S. Luksic

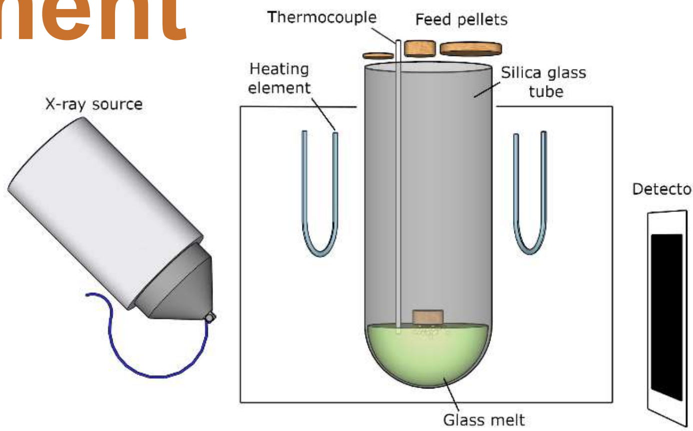


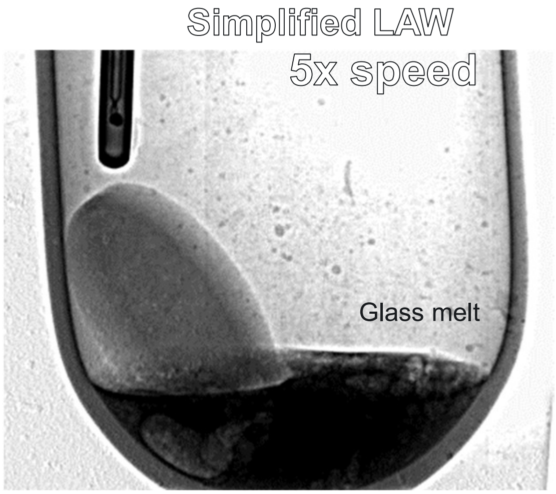
Colored X-ray CT reconstruction of cold cap on glass melt

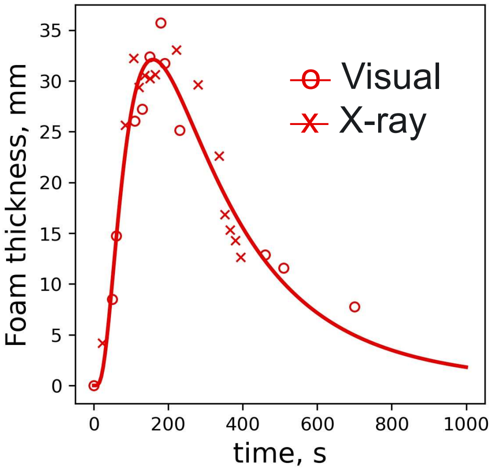
Courtesy of S. Luksic

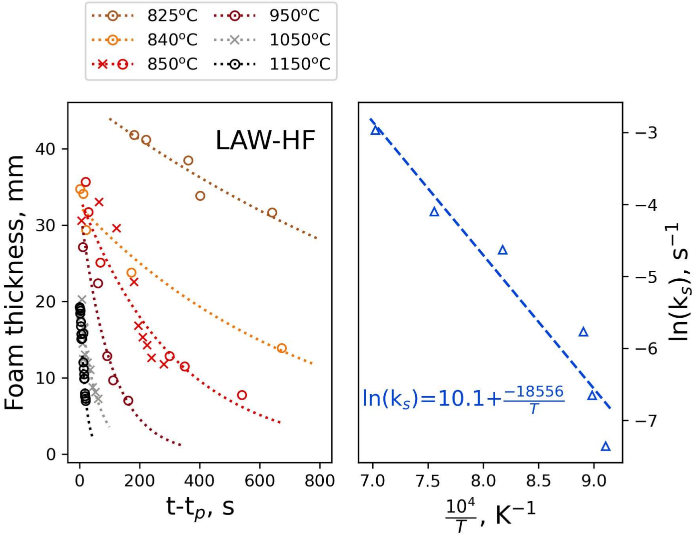
X-ray observation: pellet drop experiment

- Real-time observation of interface and surface foaming
- Close agreement to visual observation experiments
- Does not require removing components like Fe_2O_3
  - Feeds must be calcined to avoid excessive foaming from gases below T_{FO}
- Accurate visualization of foaming and foam decay





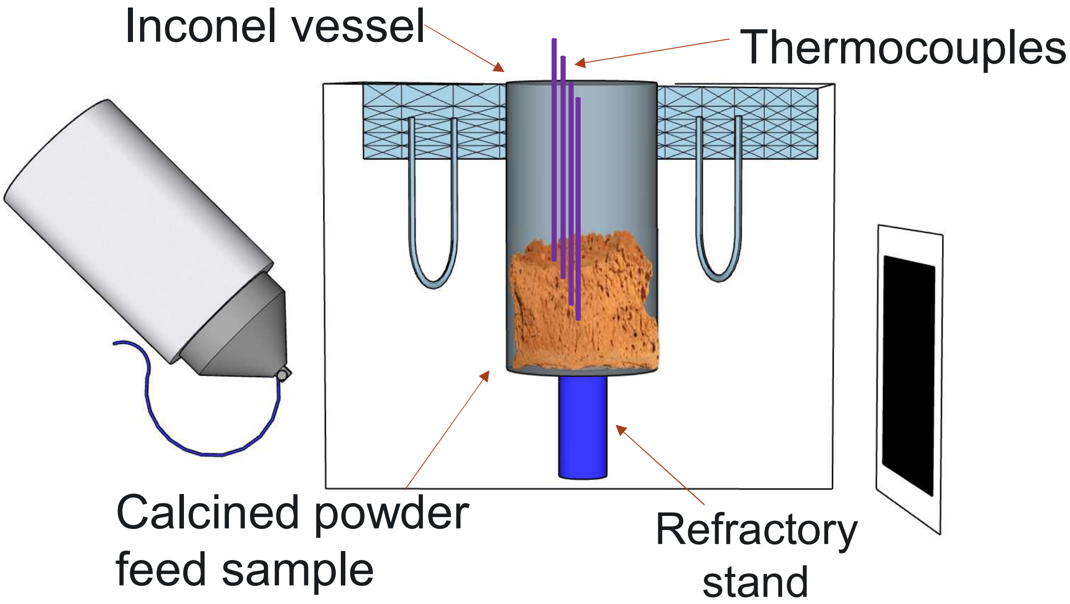


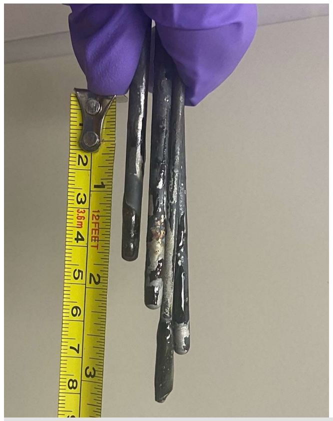


J. Marcial et al., Ceram. Int. (2022)

Feed-melt interfacial monitoring

- Feed sample was be prepared by heat treating A19 700°C at 10K/min
  - 4 thermocouples were placed in feed at 0.5” increments from bottom
    - Attempted to fix position using metal wire and silica wool
  - Inconel vessel was used; held near furnace top using refractory stand




Thermocouples


Furnace

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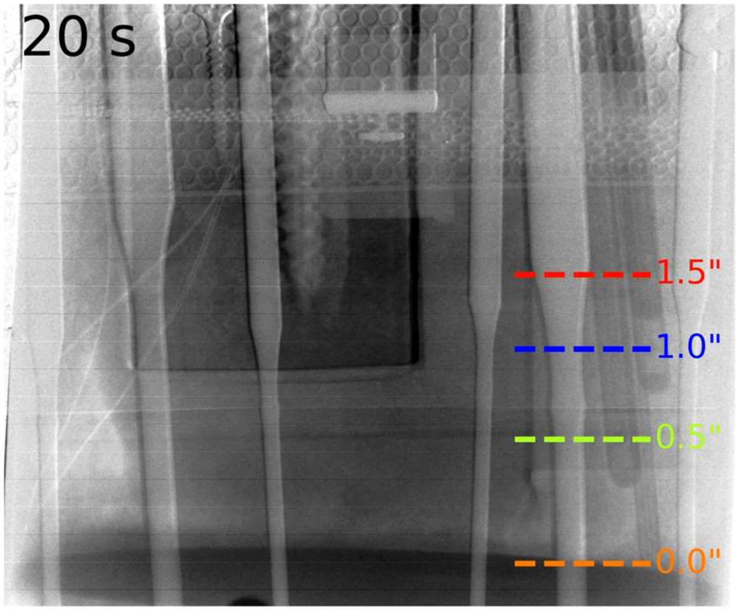
X-ray observation: temperature profile measurement

- Unlike visual methods; using X-rays we could observe thermocouple placement more clearly

stainless steel vessel



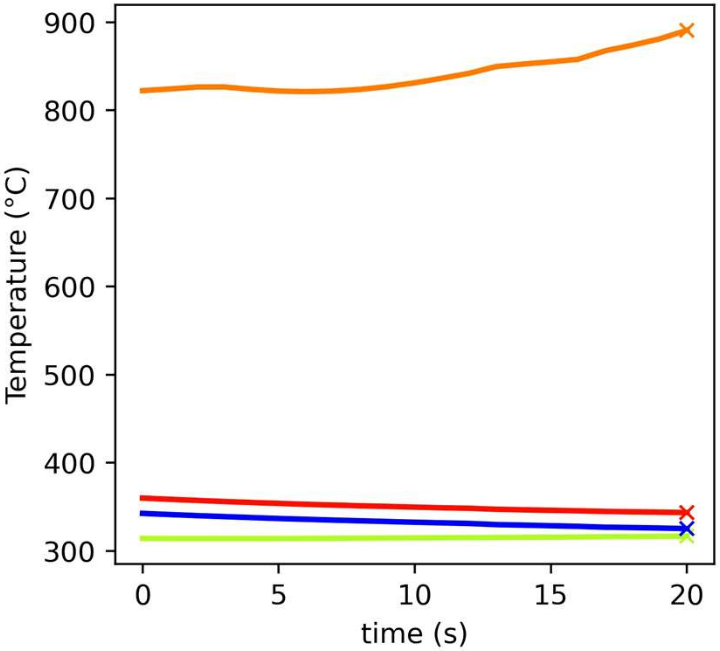
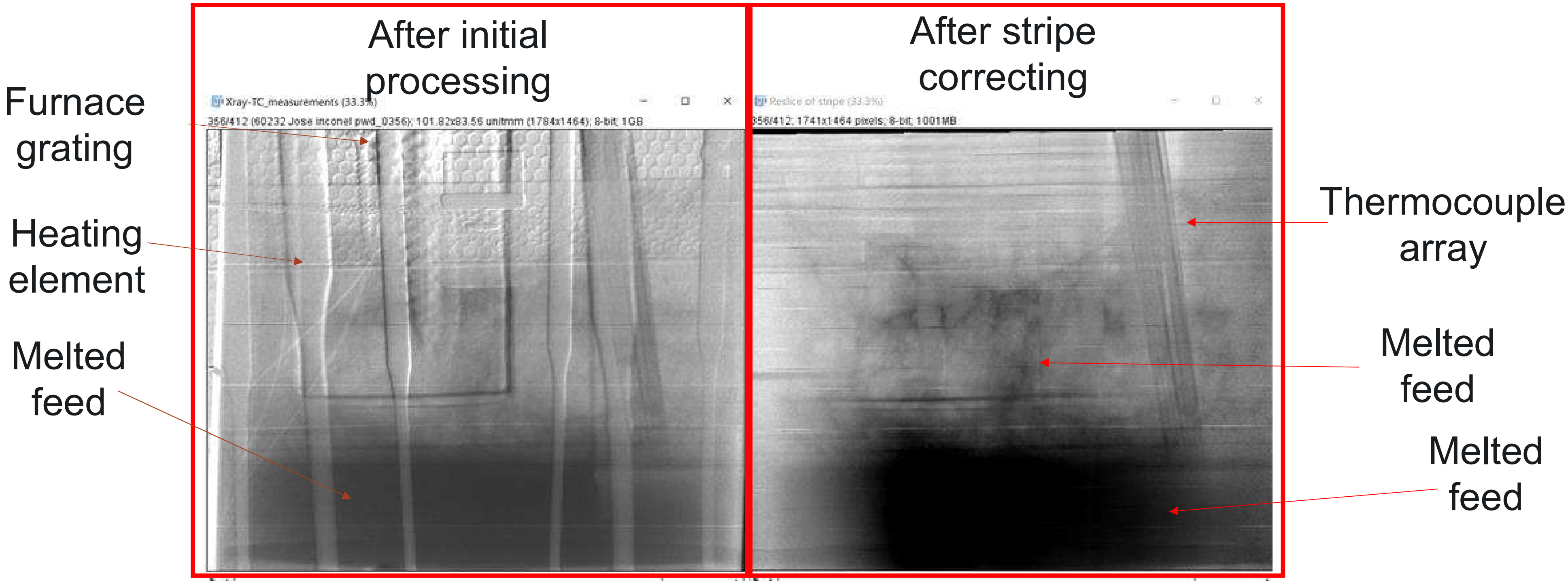


Image processing for improved resolution

- Effect of stripe correction, background correction, and contrast enhancement on image quality



After initial processing

After stripe correcting

Conclusions and Future work

- Applications of visual observation continue to inform emerging technologies
- For nuclear waste vitrification, it is used to interpret influence of composition and makeup on foaming
  - Results obtained then compared to large-scale melting characteristics using melting-rate correlation equation
- Future work seeks to better understand characteristics of reacting feed interface with glass melt
  - Preliminary data shows this can be achieved with modifications to feed compositions
  - Will provide complementary data for ongoing computational models and X-ray CT measurements

Acknowledgements

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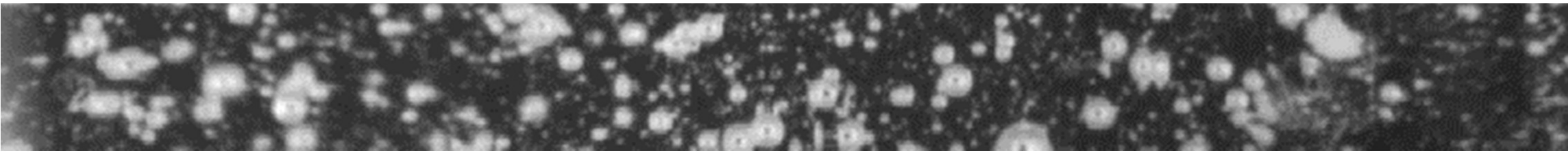

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Thank you