Modeling of glass properties and their effects on glass production rate in an electric melter

Melting Rate Correlation and Primary Foam Stability

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Hanford waste lifecycle

The duration of **Hanford nuclear waste cleanup by vitrification** can be reduced by increasing

- the **waste loading** (the fraction of waste in the glass product): improved glass formulation models
- the **processing rate** (the rate at which the melters are producing the waste glass): improved understanding of the conversion process (received attention relatively recently)

Glass production rate

The **glass production rate** can be **maximized** through

- optimizing the **waste treatment**
- selecting **glass-forming and modifying additives**
- adjusting **melter operation parameters**

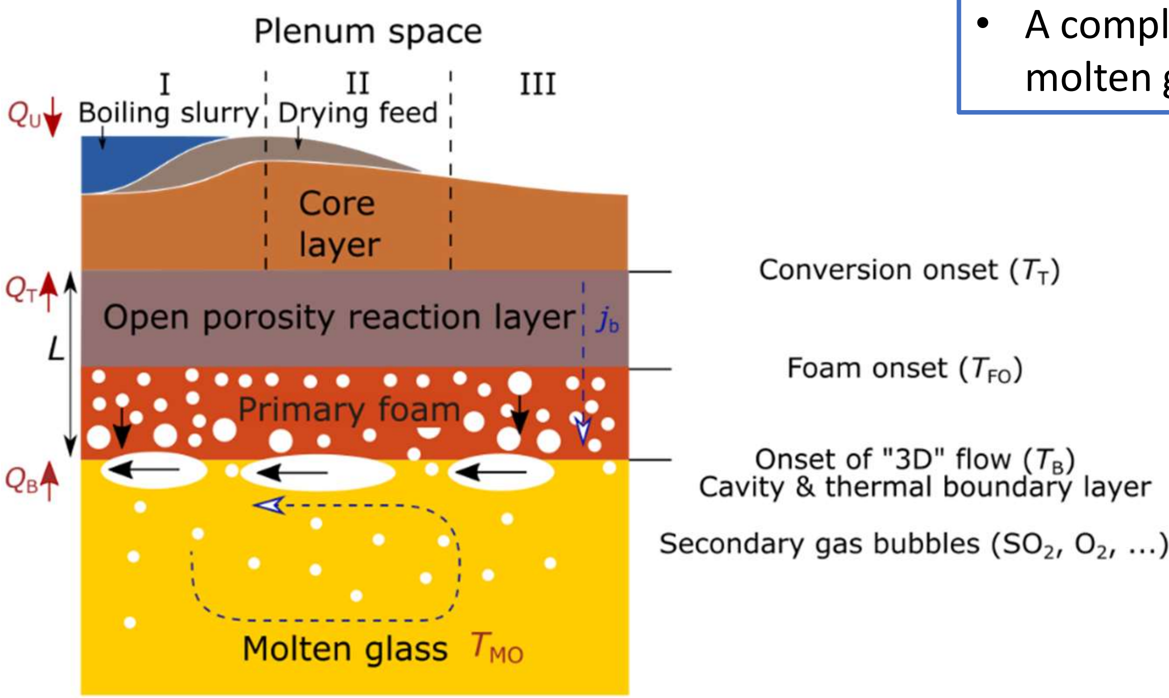
Approaches:

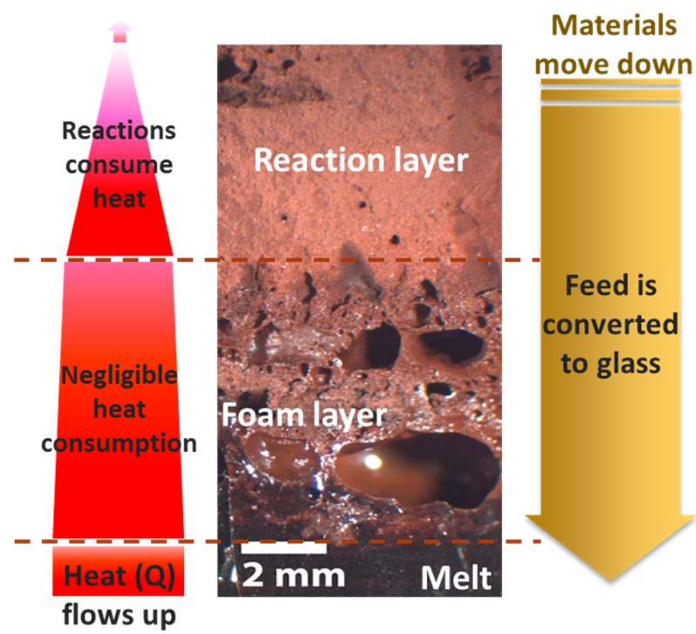
- **scaled melter runs** (expensive and cumbersome)
- **mathematical modeling** (extremely complex)
- **engineering approach**: **melting rate correlation (MRC)**

Cold Cap Structure I

Two major problems for Hanford:

- A large number (~55000) of waste compositions
- A complex process of melter feed conversion to molten glass

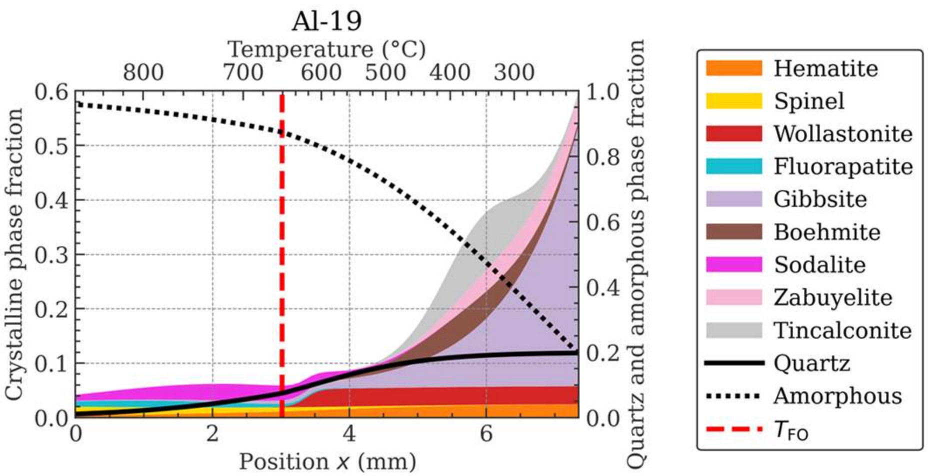


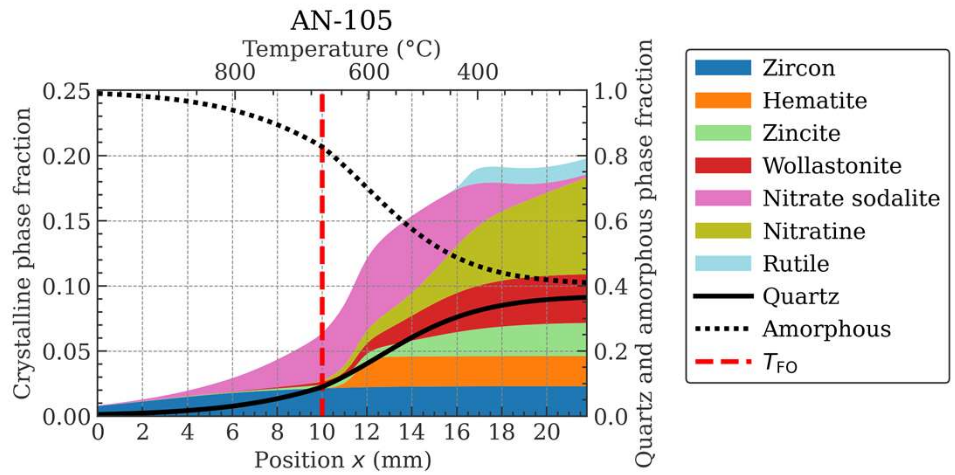


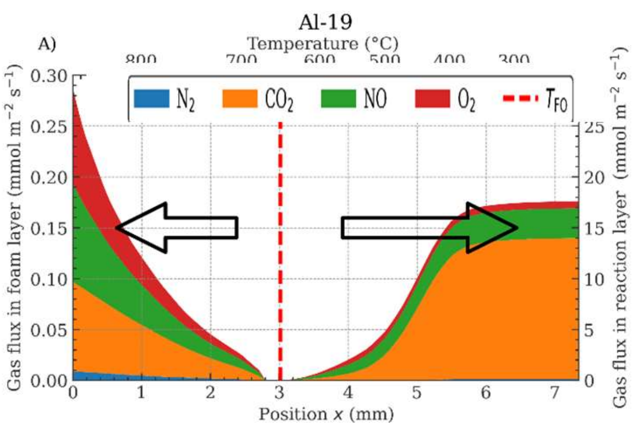
I, II, III	core layer variations
j_b	condensed (nonvolatile) mass flux
L	reaction zone thickness (open porosity and primary foam)
Q_U, Q_B, Q_T	top (upper), bottom, and conversion onset heat flux
T_T, T_{FO}, T_B, T_{MO}	temperature: conversion onset (T), foam onset (FO), cold cap bottom (B), and bulk (melter operating, MO)

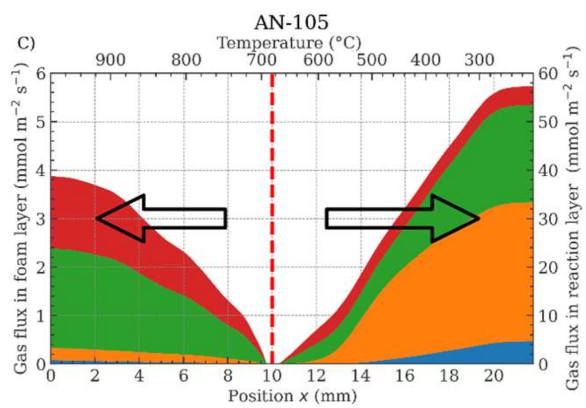
Cold cap structure II

(top) crystalline phases
(bottom) evolving gases









Melting Rate Correlation (MRC)

The **heat flux** from the melt pool to the cold cap:

$$Q_B = h(T_{MO} - T_B)$$

The **shear stress** imposed on the cold cap bottom:

$$\sigma_B = \eta_{MB} dv_x/dz|_B$$

The **glass production rate**:

$$j = Q_B/\Delta H$$

The **MRC relationship**:

$$j = \xi \mathrm{Re}^{\gamma}(T_{MO} - T_B)/\Delta H$$

Melter is operated at a **constant T_{MO}**

Melt pneumatically stirred by gas **bubbling:**

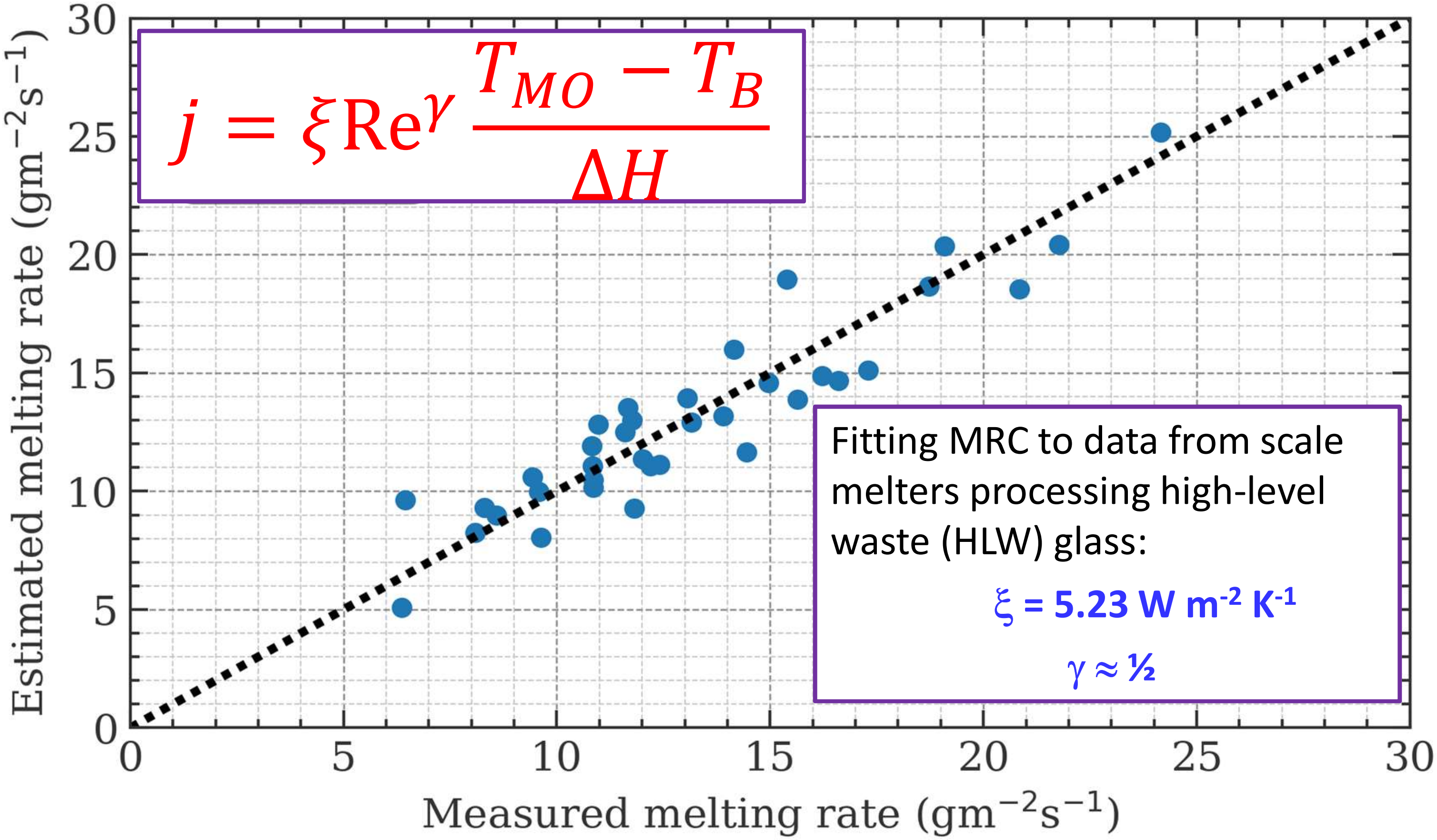
$$h = \xi \mathrm{Re}^{\gamma},$$
$$\mathrm{Re} = yu/\nu$$

h **heat transfer coefficient**
T_{MO} melter operating temperature
T_B cold cap bottom temperature
η_M glass melt viscosity
V_x glass melt velocity horizontal component
x horizontal coordinate
z vertical coordinate
B denotes the cold cap bottom
ξ and γ adjustable parameters
y melt pool dimension (depth or width)
u **bubbling gas flow rate**
ν kinematic viscosity
ΔH feed-to-melt conversion heat

Unknown parameters: T_B, ξ, and γ

Various methods to estimate T_B exist (none of them fully satisfactory).

Melting Rate Estimated with MRC



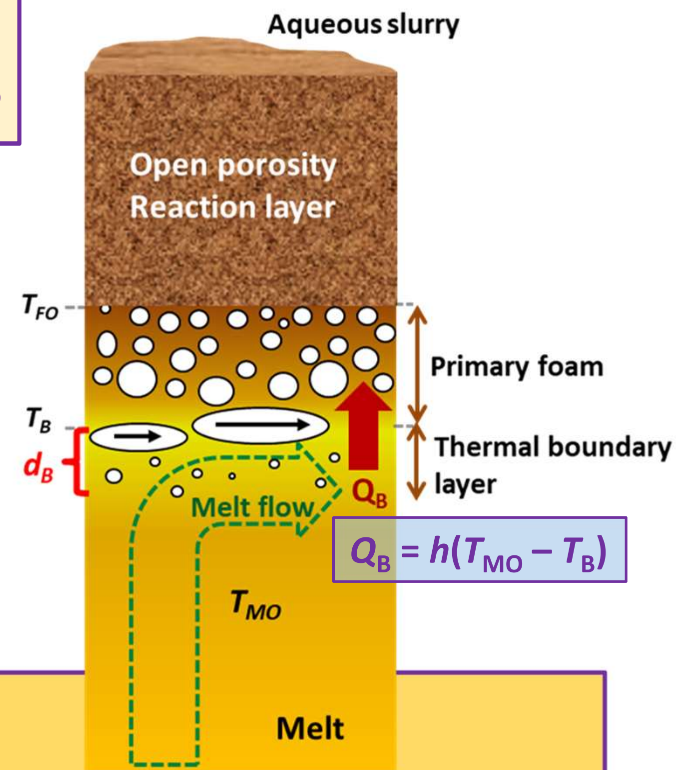
Cold cap bottom temperature, T_B

The reacting zone of the melter feed in the cold cap is separated from the melt below by the **primary foam layer**.

With other parameters fixed, melting rate, ***j*****, increases as T_B decreases**:

$$j = \xi \mathrm{Re}^{\gamma} \frac{T_{MO} - T_B}{\Delta H}$$

Ultimately, the melting rate is maximized when primary foam ceases to exist, i.e., when $T_B \rightarrow T_{FO}$.



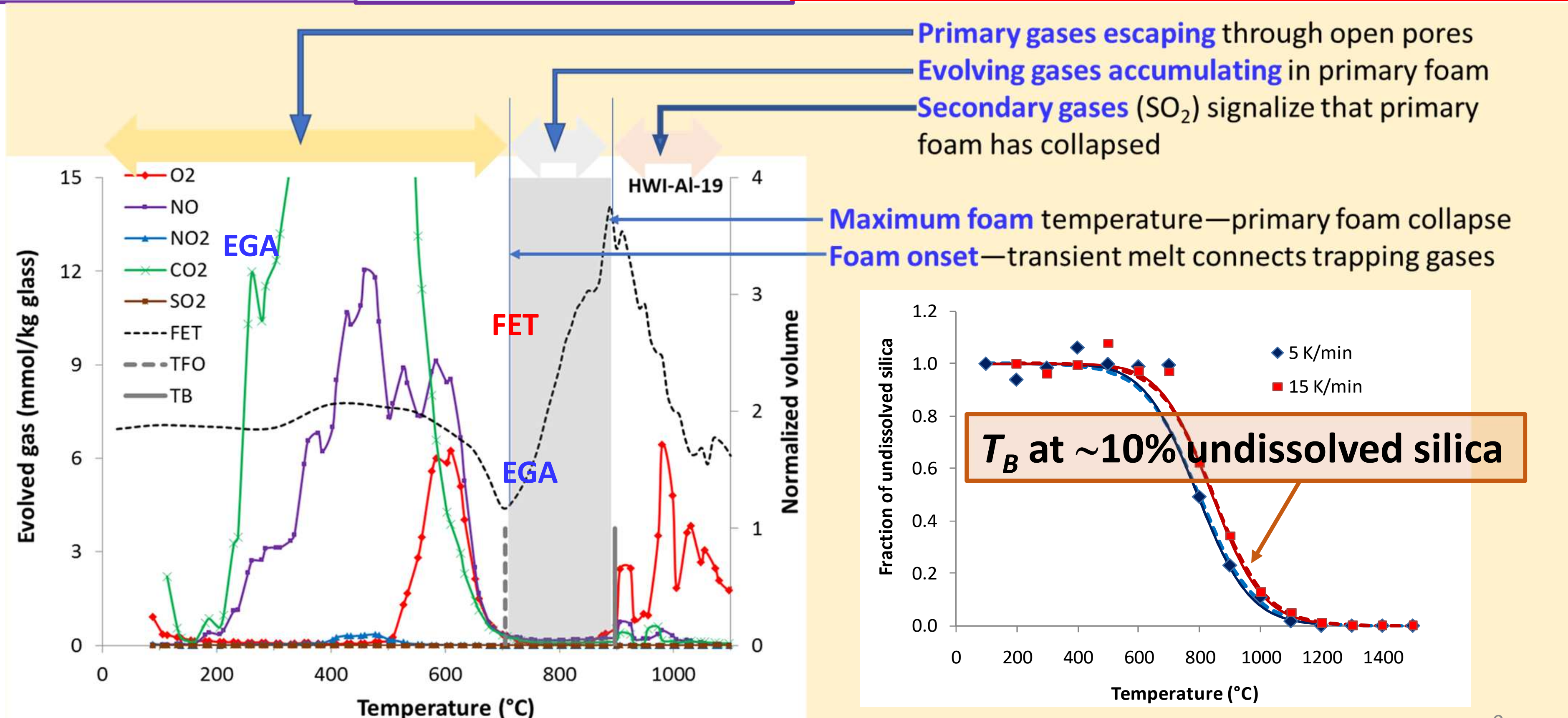
At the cold cap bottom

- **primary foam is collapsing: cell walls are breaking**
- **bubbles are coalescing into cavities** carried away by circulating melt
- downward motion in the cold cap turns into **horizontal motion in the melt pool**

Methods to estimate T_B:

- Feed expansion test (FET)
- Evolved gas analysis (EGA)
- Silica dissolution extent

For moderately foaming feeds $T_B \approx T_{FM}$.
For vigorously foaming feeds (mostly LAW) $T_B > T_{FM}$
(Intense gas evolution resembles a boiling liquid.
Cold cap bottom zone fluctuates.)

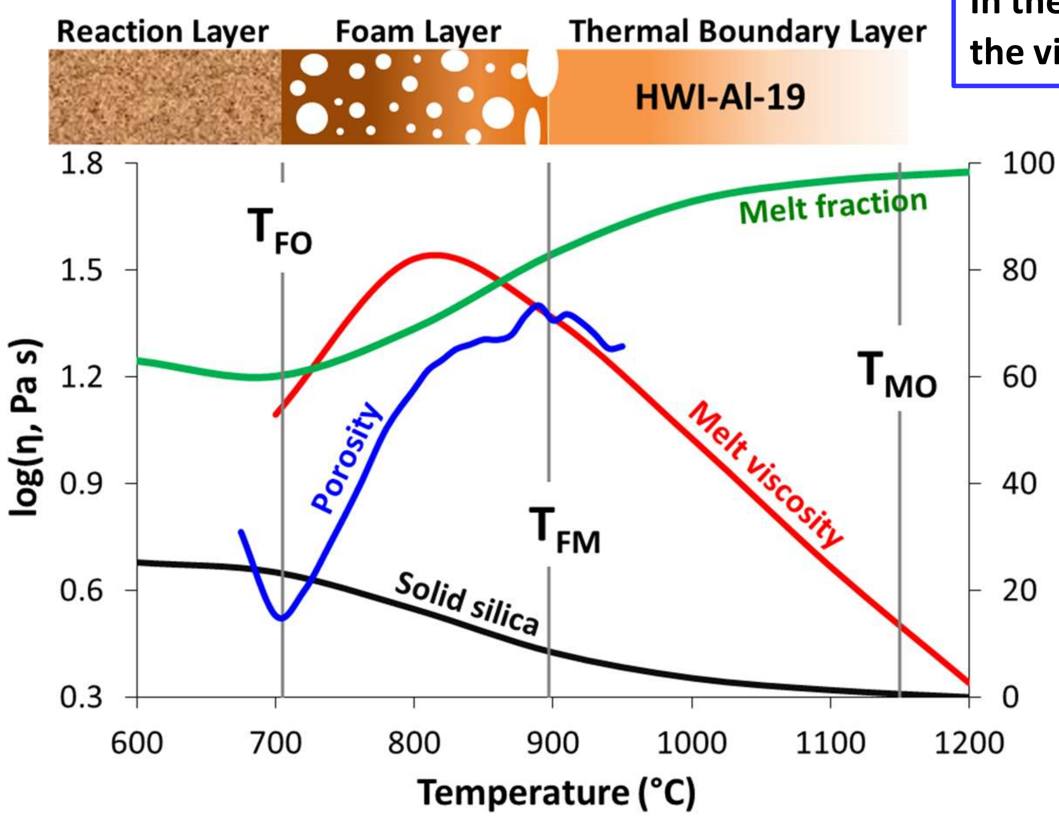


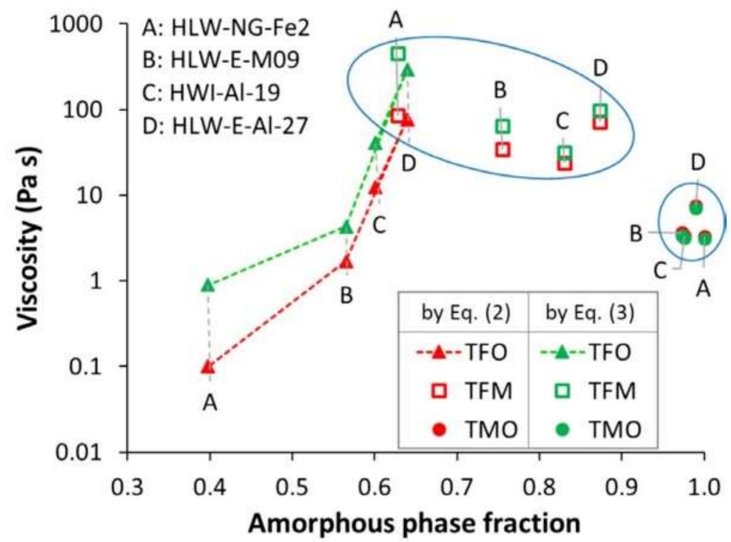
Transient glass melt

In the glass melt, foam stability is determined by the melt viscosity.

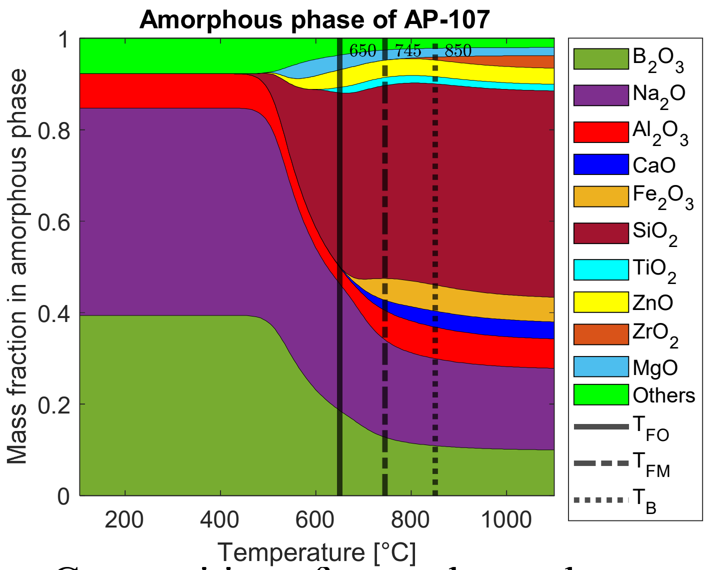
The transient melt viscosity is a function of temperature and composition.

In the primary foam, the temperature is highest and the viscosity lowest at the bottom.

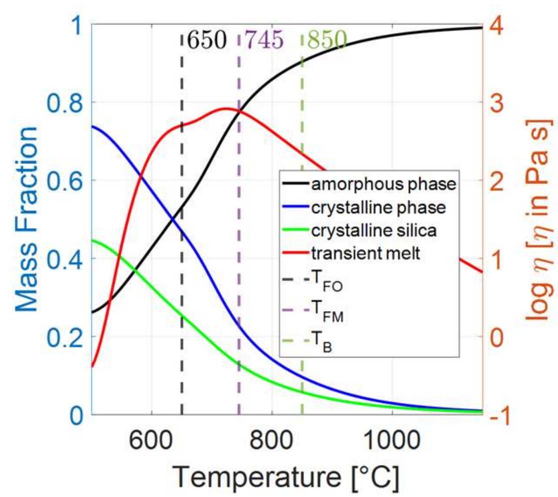




Primary foam stability: Transient glass melt composition and viscosity



Composition of amorphous phase versus temperature during melting of AP-107 feed.



Transient melt viscosity for AP-107 feed, calculated by Adam-Gibbs model.

Primary foam onset viscosity, typically 10^2 to 10^4 Pa s, depends on the transient melt fraction (the higher fraction, the higher η_{FO}, the lower T_{FO}).

Primary foam collapses at the melt viscosity η_B = 40 to 60 Pa s, depending on the shear stress and other factors, such as inclusions of solid particles.

Primary foam stability: Critical cell wall thickness

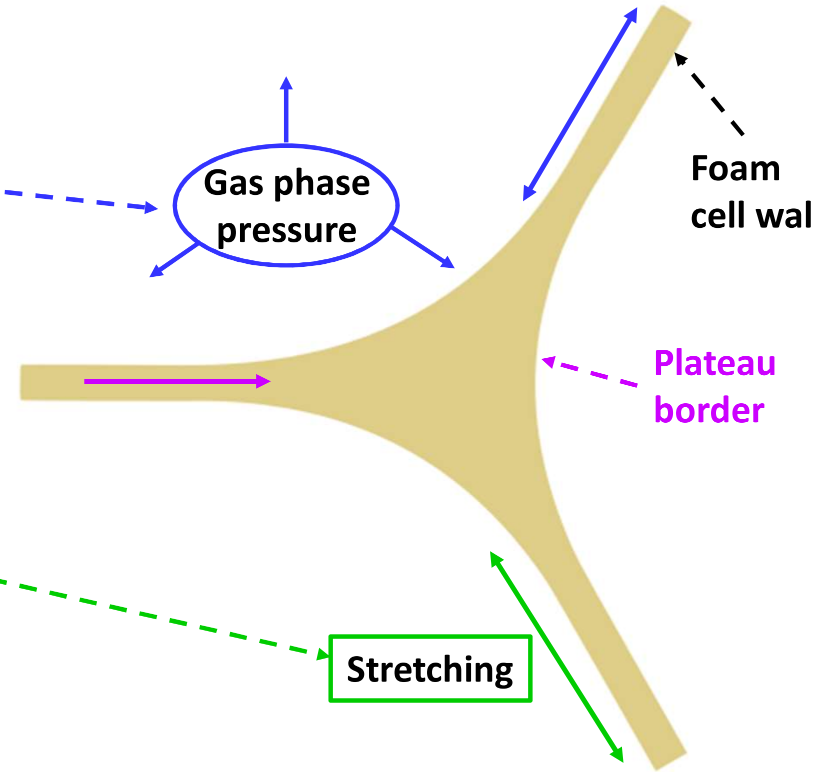
Thinning of foam cell walls occurs by three mechanisms:

gas phase volume in foam bubbles increases through thermal expansion and gas-evolving reactions

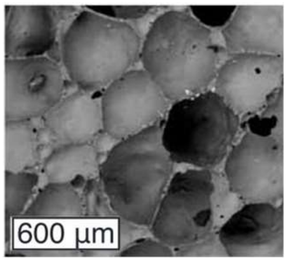
the surface curvature gradient drives the melt to the Plateau borders

the external shear stress imposed by the velocity gradient on the melt side induces strain on the foam cells

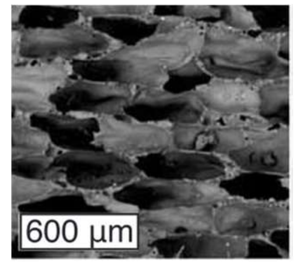
The **critical thickness** at which the cell walls break is affected by the dispersed solid particles.

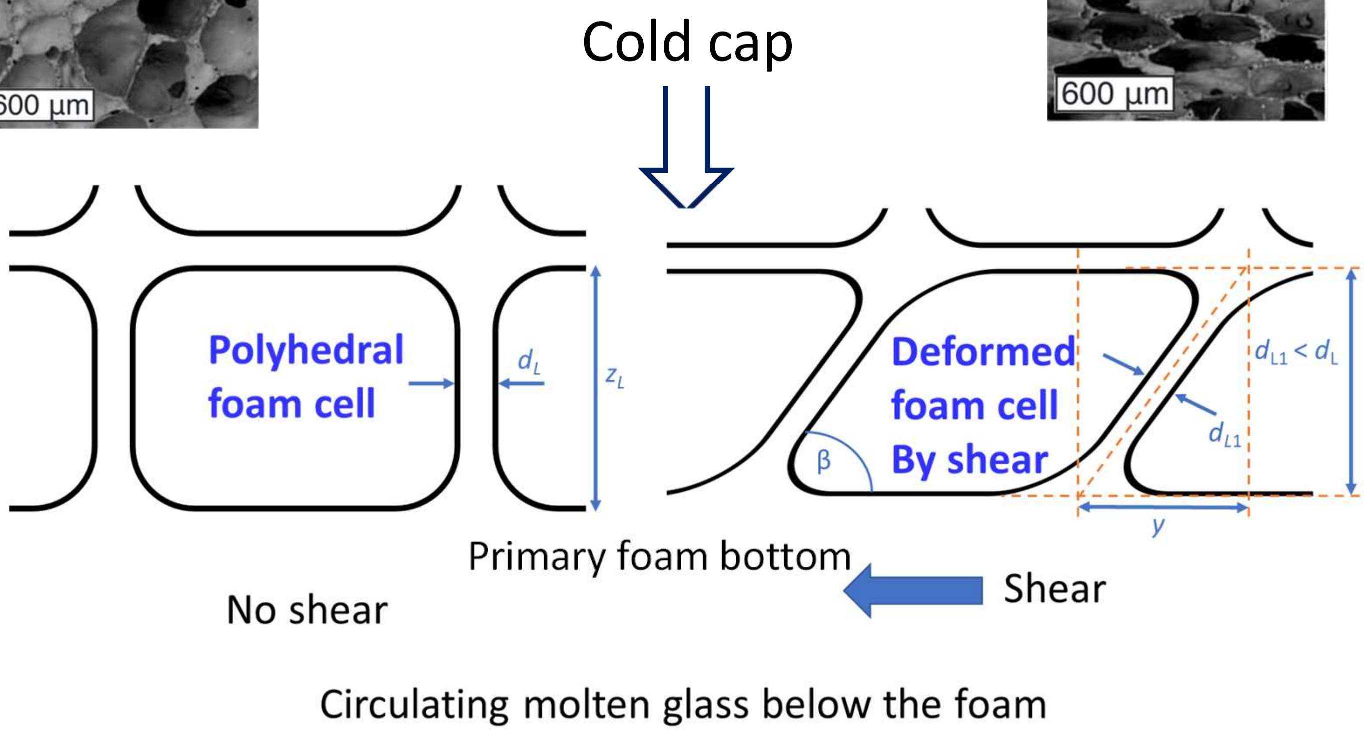


Primary foam stability: Shear-induced deformation of foam cells



In the melter, the shear rate producing convection is driven by melt bubbling.





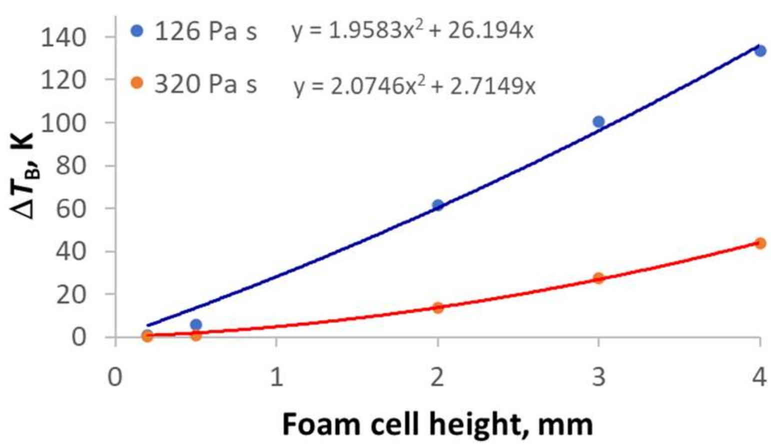
The original lamella thickness, d_L, is critical and breaks at the survivor time $t_L = z_L \rho_F / j$.

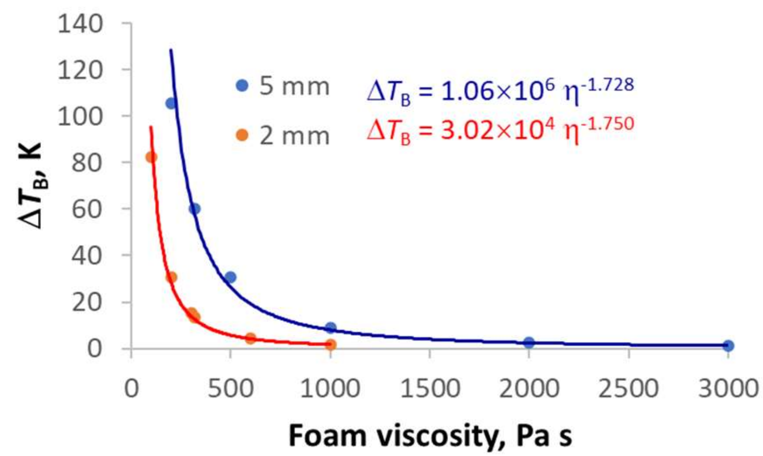
The thinner inclined lamella, $d_{L1} < d_L$, is subcritical and breaks at $t < t_L$.

With thinner lamellas, primary foam bottom stabilizes at a higher viscosity, which means at a lower cold cap bottom temperature.

Primary foam stability: Shear-induced cold-cap-bottom-temperature drop (ΔT_B)

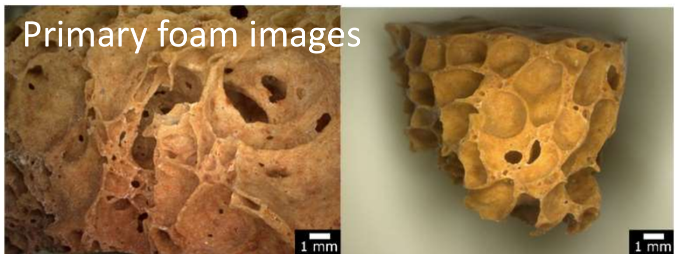
Small foam cells (small z_L) and stiff foam (high η_{FB}) resist the shear stress effect.



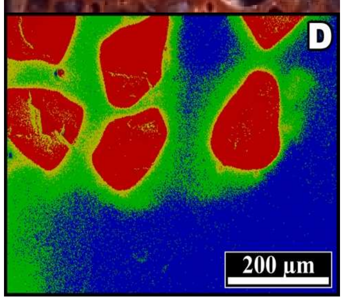


Cold cap bottom temperature drop (ΔT_B) versus (left) foam cell height (z_L) for two η_{FB} values and (right) foam viscosity (η_{FB}) for two z_L values.

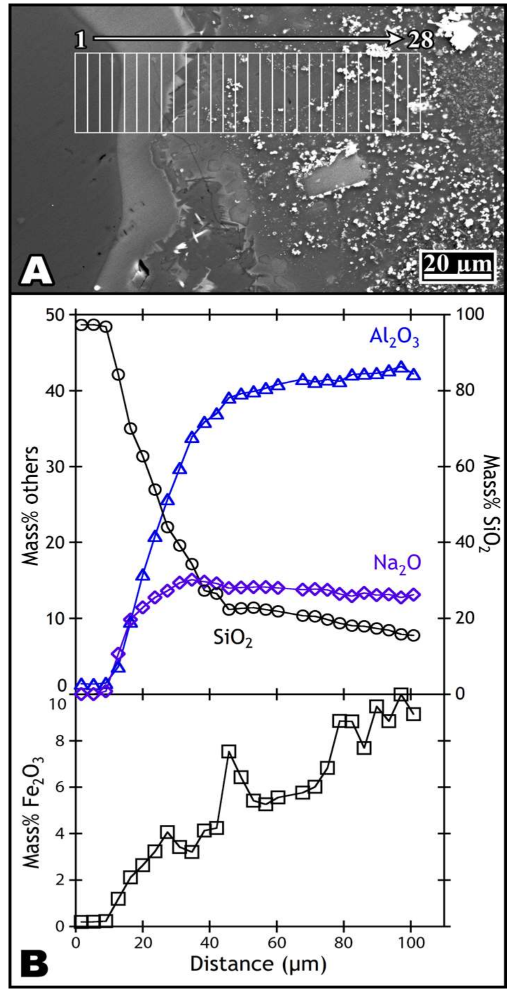
Dissolving quartz particles in foam

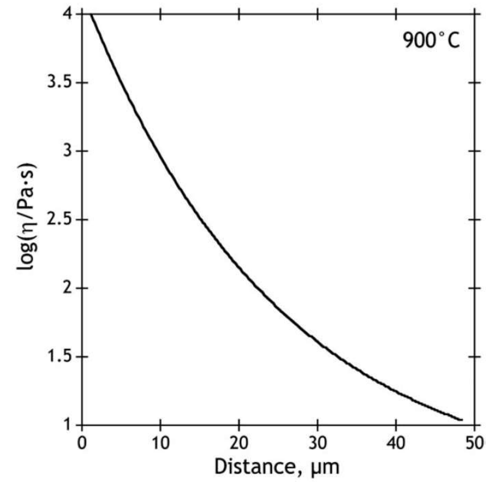


Silica particles (white) and trapped bubbles (black) in the connected glass-forming melt.



High-viscosity concertation layers (green) around dissolving silica particles (red) in transient melt (blue).





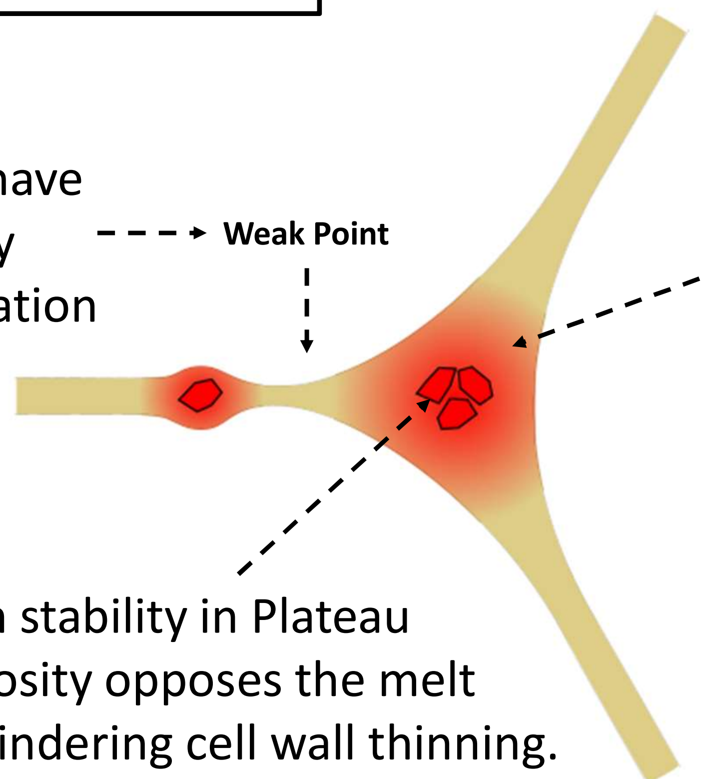
(above) Viscosity distribution at quartz-melt interface.

(left) Melt component fractions (SiO_2, Al_2O_3, Na_2O, Fe_2O_3) at quartz-melt interface. Note spinel crystals away from the silica-rich layer.

Primary foam stability: Effects of dissolving silica particles

Silica particles are the main solid phase that continues to dissolve at the cold cap bottom.

In the cell walls, silica particles have destabilizing effect: low viscosity melt outside the silica concentration layer are the weak points.



The viscosity of melt around dissolving silica particles (which has a high concentration of SiO_2) can be up two or more orders of magnitude higher than in the bulk melt.

Silica particles promote foam stability in Plateau borders, where the high viscosity opposes the melt inflow from cell walls, thus hindering cell wall thinning.

Effect of cold cap bottom shear stress

The cap bottom temperature approximation function:

$$T_{\mathrm{B}} = T_{B0} + p_{\mathrm{Q}}Q - p_{\mathrm{u}}u$$

Q heat flux to the cold cap

u bubbling gas flow rate

T_{B0}, p_{Q}, and p_{u} are empirical coefficients to be determined through computational fluid dynamics modeling.

The shear stress at the cold cap bottom is proportional to the velocity gradient.

The velocity gradient is proportional to the bubbling rate.

Summary

As the cold cap bottom temperature decreases, the melting rate increases.

The bottom cells of the primary foam continue to break until the transient melt viscosity is high enough to resist the wall thinning forces.

The wall thinning is promoted by foam gas expansion and the shear stress deformation induced by melt bubbling.

The transient melt viscosity and primary foam viscosity are functions of temperature, composition, and the presence of solid (silica) inclusions.

The heat flux and stress are controlled by forced convection produced by bubbling.

Postscript

Ultimately, the melting rate is maximized when primary foam is absent.

This option is likely achievable in glasses formulated for an elevated melting temperature and would be possible if LAW glass are processed in melters with molybdenum electrodes.