Journal of Nuclear Materials 496 (2017) 54–65

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Effect of melter feed foaming on heat flux to the cold cap

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HIGHLIGHTS

- The heat flow and the conversion heat determine the glass production rate.
- The foam layer hinders the heat flow and the foam layer thickness can be estimated.
- The melting rate was slower for the feed with corundum producing excessive foam.
- The melting rate was faster for the feed with boehmite producing less stable foam.

ARTICLE INFO

Article history:
Received 16 March 2017
Received in revised form
18 July 2017
Accepted 12 September 2017
Available online 15 September 2017

Keywords:
Cold cap
Foam layer
Heat flux
Heat conductivity
Evolved gases

ABSTRACT

The glass production rate, which is crucial for the nuclear waste cleanup lifecycle, is influenced by the chemical and mineralogical nature of melter feed constituents. The choice of feed materials affects both the conversion heat and the thickness of the foam layer that forms at the bottom of the cold cap and controls the heat flow from molten glass. We demonstrate this by varying the alumina source, namely, substituting boehmite or corundum for gibbsite, in a high-alumina high-level-waste melter feed. The extent of foaming was determined using the volume expansion test and the conversion heat with differential scanning calorimetry. Evolved gas analysis was used to identify gases responsible for the formation of primary and secondary foam. The foam thickness, a critical factor in the rate of melting, was estimated using known values of heat conductivities and melting rates. The result was in reasonable agreement with the foam thickness experimentally observed in quenched cold caps from the laboratory-scale melter.

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1. Introduction

Foaming is an unavoidable part of the batch melting process [1,2]. In gas-heated as well as electric continuous glass melting furnaces, glass batch is charged on top of molten glass. Foaming occurs at the interface between the batch and molten glass when glass-forming melt becomes continuous while batch gases still evolve. This phenomenon is often ignored even though foaming influences heat transfer and thus affects the rate of melting. Commercial glass batches are usually formulated to minimize foam formation. This is not always possible in case of melter feeds for nuclear waste vitrification, where some major waste components are prone to foaming. However, as the present paper demonstrates, even here foaming can be reduced, and thus the rate of melting increased, by proper selection of glass foaming additives and possibly waste pretreatment.

The Hanford Tank Waste Treatment and Immobilization Plant will vitrify over 0.2 million m^3 of radioactive waste now stored in underground tanks at the Hanford Site in Washington State. The tank waste will be treated as high-level waste (HLW) (comprising >95% of the waste radioactivity) and low-activity waste (LAW) (comprising ~90% of the waste mass) [3]. Both the HLW and LAW will be mixed with glass-forming and glass-modifying additives to produce slurry feed to be charged into an electric melter [3–5] in

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http://dx.doi.org/10.1016/j.jnucmat.2017.09.016
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which the feed is converted to glass [6–13].

The conversion occurs in the cold cap that floats on the molten glass. As illustrated in Fig. 1, the feed moves down through the cold cap, while the heat needed for conversion flows up into the cold cap from the bottom. The heat flux, Q, and the melting rate, j, are related as

$$Q = \Delta H j \quad (1)$$

where $\Delta H = \Delta H_W + \Delta H_C$ is the feed melting enthalpy per unit mass of glass; ΔH_W is water evaporation enthalpy and ΔH_C is the feed-to-glass conversion enthalpy. The melting rate, also called the glass production rate, is the mass of glass produced per unit cold cap area per unit time.

The cold cap is composed of a reaction layer and a foam layer (Fig. 1). Most of the melting reaction heat is consumed in the reaction layer [7–11]. The foam layer, which forms at the cold-cap bottom where the glass-forming melt becomes continuous, hinders the heat flow and thus limits the rate of melting [5–11]. A more detailed description of the foam layer structure and the effects of melt bubbling are provided in Section 4.

On the melt side, heat transfer is controlled by convection and conduction, thermal radiation being negligible in the opaque waste glass. On the cold-cap side, heat transfer is controlled by conduction together with the kinetics of chemical reactions and phase transitions. The heat conductivity in foam, discussed in Section 5, is affected by both conduction in the melt and thermal radiation in the gas, and hence is a function of porosity (the gas phase fraction), pore size distribution, and temperature. Local currents induced by surface forces can also influence the effective heat conductivity.

To enhance heat transfer to the cold cap, forced convection is induced by bubbling gas into the melt. As discussed in Section 6, bubbling suppresses the thermal boundary layer in the melt under the cold cap and washes away a bottom part of the foam layer, especially when the melt viscosity is low.

The response of the melter feed to heating is influenced by the choice of feed materials, including the particle size of chemicals and minerals [4,5,11]. Typically, about half of the feed comes from nuclear waste and the other half comes from glass-forming and -modifying additives. Adjustment of the chemical form of the waste component is limited, but the chemical and mineralogical forms of additive components can be chosen without affecting the composition of the glass product.

The effect of the feed chemistry on the ease of melting has previously been demonstrated by varying the alumina source in feeds designed for high-alumina HLW vitrification [14–16]. About 46% of the Hanford HLW contains high fractions of alumina [4]. Even though alumina was not an additive constituent but a waste component, this well-documented case provides an excellent example of the effect of a major feed constituent on melting behavior. The Vitreous State Laboratory (VSL) tested feeds to make glasses containing 23–24 mass% Al_2O_3 in the DM100 melter of the 0.108 m^2 melting area operated at 1150 °C with 9 l min^{-1} bubbling rate and the solids content equivalent to 500 g glass per liter of slurry. The alumina source substantially affected the steady-state melting rates: 950 kg m^{-2} d^{-1} (d = 24 h) is reported for the feed with gibbsite [$Al(OH)_3$], 1200 kg m^{-2} d^{-1} for feed with boehmite [AlO(OH)], and 700 kg m^{-2} d^{-1} for feed with corundum [Al_2O_3] [14,15].

Pierce et al. [16] showed that the slow-melting feed with corundum foamed extensively and Pokorny et al. [7] pointed out that the fast-melting feed with boehmite produced less stable foam and had a low conversion heat. In this study, we focus on the foam layer and its effect on the heat flow from the molten glass to the cold cap.

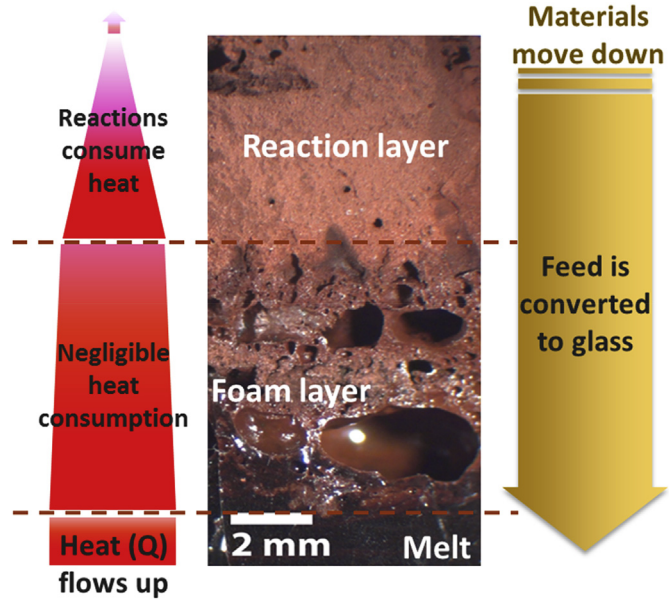

Fig. 1. Schematic of heat flux and feed movement in the cold cap (the cold-cap cross section was obtained from a laboratory-scale melter [12,13]).

To determine the extent of foaming, we measured feed volume expansion in response to increasing temperature by heating a dry-feed pellet at a rate similar to that which feeds experience within the cold cap [5,16,17]. A thermogravimetric analyzer (TGA) equipped with differential scanning calorimetry (DSC) was used to measure the reaction heat. Evolved gas analysis (EGA) was used to identify gases released from collapsing foam. Regarding the heat conductivity, we used existing data [6,18].

2. Experimental procedure

Tables 1 and 2 show compositions of simplified versions of the glass and feed containing gibbsite that were designed by VSL for a high-alumina HLW [14,15]. For feeds containing boehmite and corundum, the equivalent masses are 71.49 g and 60.75 g, respectively. The feed chemicals were mixed in 400 ml of deionized water to prepare slurry that was dried and milled to powder as described in Refs. [5,16,19].

The volume expansion test [5,17] was performed using cylindrical pellets ~13 mm in diameter and ~6 mm thick prepared by pressing 1.5 g feed at 168 MPa. Each pellet was heated at 5 or 10 K min^{-1} to 1100 °C. The pellet profile was monitored with a camera (Fig. 2). The Photoshop®[1] software and MATLAB®[2] program were used for numerical integration to calculate the volume [17,20].

For thermal analysis [21,22], ~60 mg feed was heated to 650 °C at 10 K min^{-1} in a Pt crucible in the TGA/DSC analyzer (Netzsch STA 449 F1). The reaction heat was determined using the run–rerun method described by Chun et al. [22], as shown in Fig. 3. Samples were heated to the set temperature (650 °C), cooled down to room temperature, and then heated again to the same set temperature. To distinguish the reaction heat from the sensible heat, the rerun data were subtracted from the run data.

Evolved gas analysis was performed using a gas chromatograph

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