

Characterization of a High-Level Waste Cold Cap in a Laboratory-Scale Melter

Derek R Dixon^a, Michael J Schweiger^a, and Pavel Hрма^{a,b*}

^aPacific Northwest National Laboratory, Richland, WA 99354

^bDivision of Advanced Nuclear Engineering, Pohang University of Science and Technology, Pohang, Republic of Korea

*Corresponding author: pavelhrma@postech.ac.kr

1. Introduction

High-level nuclear waste is immobilized by converting it into a durable glass form. The vitrification process takes place in a melter, where the pool of molten glass is maintained at a high temperature (1150°C or higher) by Joule or induction heating. The feed, slurry or calcine, is charged to the melter from above. The conversion of the melter feed to molten glass occurs within the cold cap, a several centimeters thin layer of the reacting material blanketing the surface of the melt (Fig. 1) [1-3].

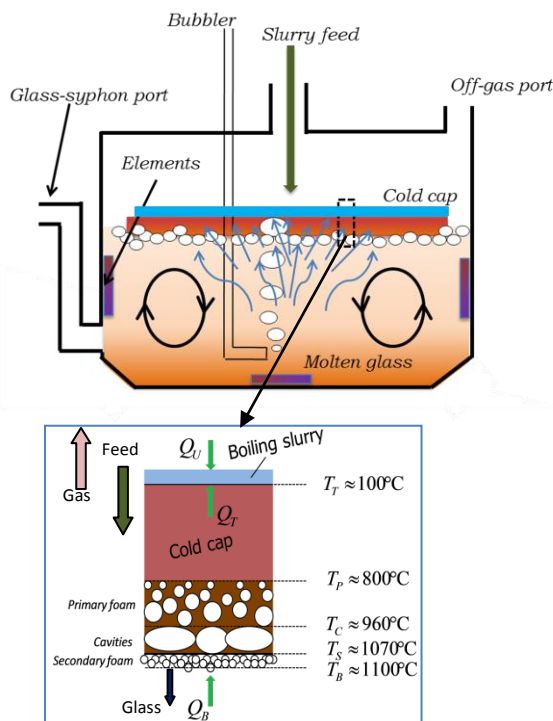


Fig. 1. Schematic of waste-glass melter [4,5] and the cold cap as anticipated via mathematical modeling [1]

Between the cold-cap top, which is covered by boiling slurry, and its bottom, where bubbles separate it from molten glass, the temperature changes by ~900°C [1]. The heat is delivered to the cold cap from the melt that is stirred mainly by bubbling. The feed contains oxides, hydroxides, acids, inorganic

salts and organic materials. On heating, these components react, releasing copious amounts of gases, while molten salts decompose, glass-forming melt is generated, and crystalline phases precipitate and dissolve in the melt [1-3].

Most of these processes have been studied in detail and became sufficiently understood for a mathematical model to represent the heat and mass transfer within the cold cap [1]. This allows us to relate the rate of melting to the feed properties.

While the melting reactions can be studied, and feed properties, such as heat conductivity and density, measured in the laboratory, the actual cold-cap dynamics, as it evolves in the waste glass melter, is not accessible to direct investigation. Therefore, to bridge the gap between the laboratory crucible and the waste glass melter, we explored the cold cap formation in a laboratory-scale melter (LSM) and studied the structure of quenched cold caps.

2. Laboratory-scale melter [4,5]

The LSM is a closed silica-glass cylinder, ~100 mm in diameter and ~180 mm high, equipped with two ports, one for the feed and off gas (dual function) and the other for two thermocouples which measure the temperature in the melt and directly below the cold cap.

After the LSM, containing ~100-g glass, was equilibrated in the furnace preheated to 1200°C, the feed was charged onto the melt surface for 35 minutes, a time sufficient for the cold cap to establish. Experiments were run with the charging rates from 3 to 7 ml/min. The LSM was then removed from the furnace and quenched on a copper block. The cold cap was photographed and its structure was studied with optical and scanning-electron microscopy (SEM).

3. Cold-cap structure

The cold cap increased as the charging rate increased, approaching ~50% of the melt surface, a coverage lower than in large-scale melters (~90%). At higher charging rates, those above 5 ml/min, the

feed heating rate approached a constant value of ~15 K/min, a value comparable to that estimated by cold-cap modeling. The feed conversion rate per cold-cap area stabilized at ~800 kg/m²/day, a value typical for scaled melter tests conducted without bubbling [6].

As the fracture of a quenched cold cap reveals (Fig. 2) and crucible experiments and mathematical models suggest (Fig. 1), the cold cap consists of a top reacting layer and a bottom foam layer.

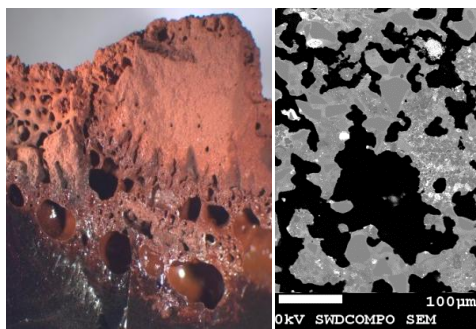


Fig. 2 Fractured cold cap showing porous top layer (SEM micrograph on right) and bubbly bottom layer

The top layer contains particles of quartz, alumina, hematite, and spinel that are dissolving in a sodium-calcium-alumino-borosilicate matrix (Fig. 3). While the porous top layer allows the reaction gases to escape through open channels (shafts), the bottom bubbles, which are 0.1–0.5 mm in diameter, coalesce into larger 3–8-mm cavities that move sideways and eventually open to the atmosphere.

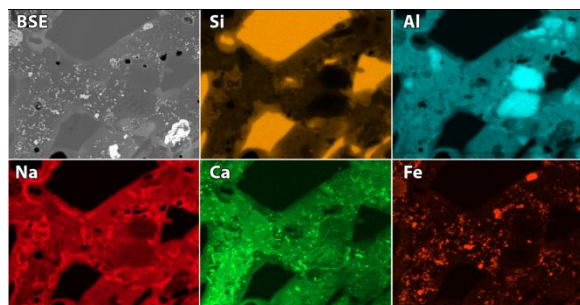


Fig. 3. SEM dot map of the reacting feed showing silica particles, alumina aggregates, and tiny crystals of hematite and spinel

Conclusions

The LSM is a suitable tool for investigating the cold cap. The cold cap that formed in the LSM experiments exhibited macroscopic features observed in scaled melter tests, as well as microscopic features accessible through laboratory studies and mathematical modeling.

The cold cap consists of two main layers. The top layer contains solid particles dissolving in the glass-forming melt and open shafts through which gases are escaping. The bottom layer contains bubbly melt or foam where bubbles coalesce into larger cavities that move sideways and release the gas to the atmosphere.

It is plausible that the LSM could be used for assessing the rate of melting of feeds variously formulated and prepared, thus allowing an optimization of the feed makeup for maximum performance.

Acknowledgments

The authors gratefully acknowledge that this work was supported by the Department of Energy's Waste Treatment & Immobilization Plant Federal Project Office under the direction of Dr. Albert A. Kruger as well as the World Class University program through the National Research Foundation of Korea funded by the Ministry of Education, Science and Technology (R31-30005). Also, the authors thank Dong-Sang Kim, Jarrett Rice, Steven Luksic, Abby Winschell, Jose Marcial, Carmen Rodriguez, Jaehun Chun, Jesse Lang, and David Pierce for their suggestions and help.

REFERENCES

- [1] R. Pokorny and P. Hrma, Mathematical modeling of cold cap, *J. Nucl. Materials* 429, 245-256 (2012).
- [2] R. Pokorny, D. A. Pierce, P. Hrma, Kinetics of batch-melting reactions, *Thermochimica Acta* 541, 8-14, (2012).
- [3] J. Chun, D. A. Pierce, R. Pokorný, P. Hrma, Cold-cap reactions in vitrification of nuclear waste glass: experiments and modeling, *Thermochimica Acta*, accepted.
- [4] D. S. Kim, M. J. Schweiger, W. C. Buchmiller, J. Matyas, Laboratory-Scale Melter for Determination of Melting Rate of Waste Glass Feeds, PNNL-21005, Pacific Northwest National Laboratory, Richland, Washington, EMSP-RPT-012 (2012).
- [5] D. R. Dixon, M. J. Schweiger, P. Hrma, Effect of Feeding Rate on the Cold Cap Configuration in a Laboratory-Scale Melter, WM2013, Phoenix, Arizona USA, 2013.
- [6] K. S. Matlack, W. K. Kot, F. Perez-Cardenas, I. L. Pegg, Determination of Processing Rate of RPP-WTP HLW Simulants using a DuraMelter™ 1000 Vitrification System, VSL-00R2590-2, Vitreous State Laboratory, The Catholic University of America, Washington, D.C., 2000.