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COMSOL Multiphysics Model for HLW Canister Filling

M. R. Kesterson

March 2016

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EXECUTIVE SUMMARY

The U.S. Department of Energy (DOE) is building a Tank Waste Treatment and Immobilization Plant (WTP) at the Hanford Site in Washington to remediate 55 million gallons of radioactive waste that is being temporarily stored in 177 underground tanks. Efforts are being made to increase the loading of Hanford tank wastes in glass while meeting melter lifetime expectancies and process, regulatory, and product quality requirements. Wastes containing high concentrations of Al_2O_3 and Na_2O can contribute to nepheline (generally $NaAlSiO_4$) crystallization, which can sharply reduce the chemical durability of high level waste (HLW) glass. Nepheline crystallization can occur during slow cooling of the glass within the stainless steel canister.

The purpose of this work was to develop a model that can be used to predict temperatures of the glass in a WTP HLW canister during filling and cooling. The intent of the model is to support scoping work in the laboratory. It is not intended to provide precise predictions of temperature profiles, but rather to provide a simplified representation of glass cooling profiles within a full scale, WTP HLW canister under various glass pouring rates. These data will be used to support laboratory studies for an improved understanding of the mechanisms of nepheline crystallization.

The model was created using COMSOL Multiphysics, a commercially available software. The model results were compared to available experimental data, TRR-PLT-080 [1], and were found to yield sufficient results for the scoping nature of the study. The simulated temperatures were within 60 ºC for the centerline, 0.0762m (3 inch) from centerline, and 0.2286m (9 inch) from centerline thermocouples once the thermocouples were covered with glass. The temperature difference between the experimental and simulated values reduced to 40 ºC, 4 hours after the thermocouple was covered, and down to 20 ºC, 6 hours after the thermocouple was covered. This level of precision is considered acceptable for the scoping nature of the model and the subsequent laboratory glass studies

Using the model, two additional glass pouring cycles were conducted. Representative thermocouple data were plotted to show the variations between the two cycles. This provides preliminary data that will be used in laboratory experiments to determine the potential for controlling nepheline crystallization in glass by varying the glass pouring conditions.

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LIST OF ABBREVIATIONS

DOE	Department of Energy
DWPF	Defense Waste Processing Facility
HLW	High Level Waste
LAW	Low Activity Waste
SRNL	Savannah River National Laboratory
WTP	Waste Treatment and Immobilization Plant

1.0 Introduction and Background

The U.S. Department of Energy (DOE) is building a Tank Waste Treatment and Immobilization Plant (WTP) at the Hanford Site in Washington to remediate 55 million gallons of radioactive waste that is being temporarily stored in 177 underground tanks. The low-activity waste (LAW) fraction will be partitioned from the high-level waste (HLW). Both the LAW and HLW will be vitrified in borosilicate glass with Joule-heated ceramic melters. The glass will be poured into stainless steel canisters. The immobilized LAW will be permanently dispositioned at the Hanford site, and the immobilized HLW will be placed in a geologic repository.

Efforts are being made to increase the loading of Hanford tank wastes in glass while meeting melter lifetime expectancies as well as process, regulatory, and product quality requirements. Higher loading of wastes in glass will increase facility throughput, reducing cost and mission duration. Wastes containing high concentrations of Al_2O_3 and Na_2O can contribute to nepheline (generally $NaAlSiO_4$) crystallization, which can sharply reduce the chemical durability of HLW glass. Nepheline crystallization can occur during slow cooling of the glass within the stainless steel canister. In order to maximize waste loading for compositions high in Al_2O_3 and Na_2O, nepheline formation must be better understood and controlled. Knowledge of the chemical, thermal, and kinetic drivers for nepheline formation in complex glass systems must be expanded.

The purpose of this work was to develop a model that can be used to predict temperatures of the glass in the HLW canisters during filling and cooling. The intent of the model is to support scoping work in the laboratory. It is not intended to provide precise predictions of temperature profiles, but rather to provide a simplified representation of glass cooling profiles within a full scale, WTP HLW canister under various glass pouring rates. To accomplish these objectives, a simplified model was created using the finite element modeling software COMSOL Multiphysics [2], which accepts user defined constants or expressions to describe material properties. Empirical from data a full scale, instrumented WTP canister filling test [1] are used to develop and validate the model parameters. Simplifications of several material properties and boundary conditions were considered acceptable for the purposes of this model as a scoping tool. For example, no attempt is made to model the changes in the density of the glass as a function of temperature. No attempt is made to model the changes in thermal or physical properties as a function of composition. The viscosity of the glass is not included in the model, and two-phase flow in the canister is not considered. These factors (among others) can be incorporated in a more detailed, future model.

The data generated by the simplified model described in this report will be used to support laboratory studies of multiple glass compositions, melted and then cooled following various profiles predicted by the model. The cooled glasses will be characterized to determine the types and amounts of crystalline phases formed. Should the results of this work prove to be useful in improving the understanding of the mechanisms of nepheline crystallization, then the development of a more detailed, future model will be of benefit in more precisely predicting the temperature profiles experienced by the glass.

1.1 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in Savannah River Site Manual E7, Procedure 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.

2.0 Model Description

The mathematical equations describing the thermal models are solved by numerical methods. The heat transfer module in the general purpose computer code COMSOL® Multiphysics was used to perform the computations [2]. This computer code meets SRNL nuclear safety QA requirements [3, 4]. A COMSOL model previously developed [5] was modified for use in the Defense Waste Processing Facility (DWPF) canister simulations.

The WTP HLW canister is 0.609m (24 inches) in diameter by 4.480m (14.7 feet) high, constructed of 0.00953m (0.375 inch) thick stainless steel [1]. The COMSOL geometry was built from the HLW drawing in the pilot melter report [1]. Figure 1 shows a material representation of the canister model. The model includes heat transfer by conduction, convection, and radiation. The conductive terms are applied to all domains in the model. Convective heat transfer is modeled as glass flow from the centerline outward in the radial direction as well as a convective cooling in the enclosure surrounding the canister based on the air flowrate through the enclosure. Radiative heat transfer is applied on the outer surface of the canister to allow heat loss to the enclosure wall. Heat transfer is also applied as surface-to-surface radiation between the top surface of the poured glass and canister wall.

® COMSOL Multiphysics is a registered tradename of COMSOL, Inc., of Burlingame, Massachusetts.

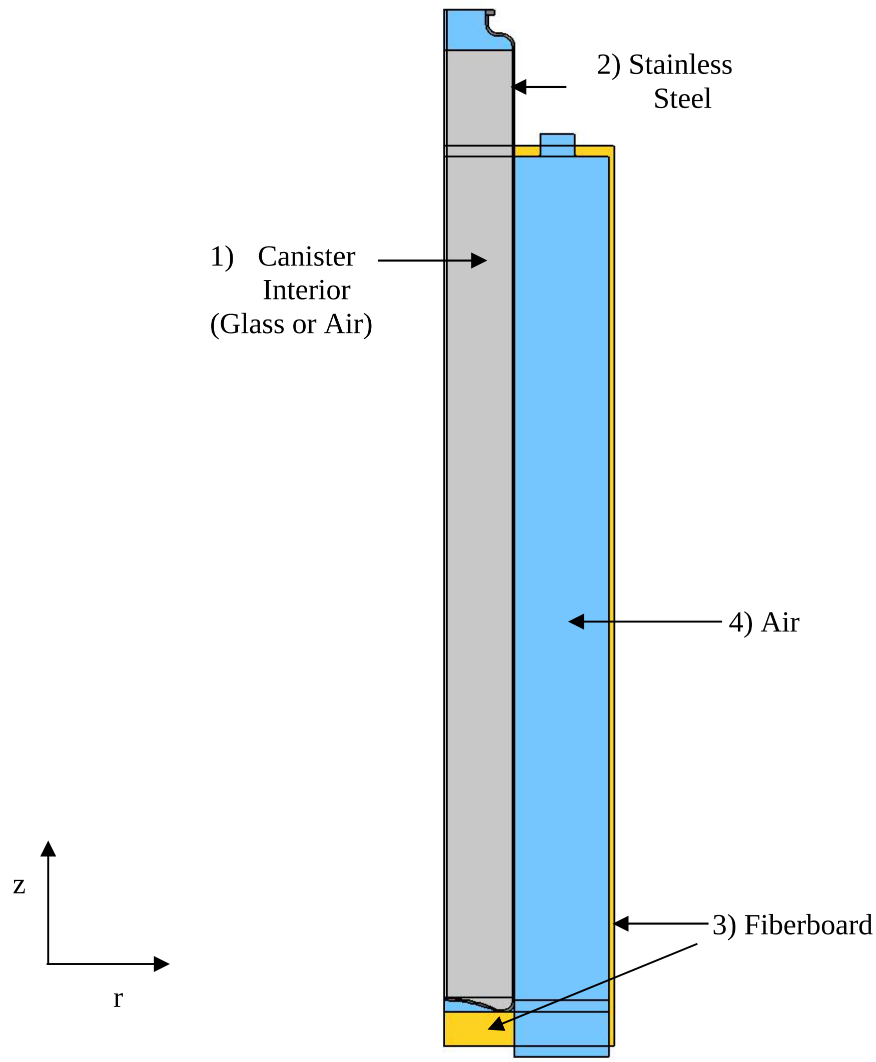

Figure 1. COMSOL Multiphysics model of the HLW canister.

A two-step method was used to model convective heat transfer in the glass phase. The first step was to model the glass stream along the centerline of the container and above the current fluid height. A variable named *glass_r* was used to calculate the radius of the glass stream as a function of pour rate and vertical position within the container. It was assumed that the glass was poured from a spout approximately 0.3048m (1 foot) above the entrance of the canister, giving an initial velocity of the glass.

For a system with an initial velocity of 0, the z-displacement is equal to:

$$D = \frac{a}{2} \cdot t^2 \quad (1)$$

Knowing a = 9.8 m/s^2, and D = 0.3048 m, the time to reach the entrance to the canister was calculated. Next, the velocity of the glass when it reaches the canister opening was calculated by:

$$v = a \cdot t \tag{2}$$

With the calculated velocity, glass density, and using a mass flowrate of 8.346 kg/min [1], the cross-sectional area of the glass can be obtained, and therefore the radius of the glass at the entrance.

$$A = \frac{V}{v} \tag{3}$$

$$t = \sqrt{\frac{2 \cdot (4.79 - z)}{a}} \tag{4}$$

$$v = \sqrt{2 \cdot a \cdot (4.79 - z)} \tag{5}$$

$$r_{glass\_inlet} = \sqrt{\frac{A}{\pi}} \tag{6}$$

Where z = height (m), and the 4.79 term is the combination of the canister height plus 0.3048 m above for the assumed pour height.

Applying a velocity increase due to gravity, the radius of the glass column was obtained as a function of the vertical height.

$$r_{glass} = \sqrt{\frac{V}{\pi \cdot \sqrt{2 \cdot a \cdot (4.79 - z)}}} \tag{7}$$

where V = volumetric flowrate (m^3/s); a = acceleration (m/s^2), and z = height (m).

Once the falling glass reached the current fill height, the convective heat transfer term was changed from a vertical flow to a radial flow. The maximum z velocity (calculated at the bottom of the canister) was used as the base value, multiplied by a power function, decreasing the velocity of the glass as it flows radially outward. The general form of the equation is:

$$v_r = V / (2 * \pi * r * t_h) * flc2hs(0.282 - r, 0.02) \tag{8}$$

where

- v_r= velocity of the glass flowing in the radial direction (m/s)
- r = radius at which the equation is evaluated (m)
- t_h = 0.02m, thickness of pour region. (an adjustable parameter, but works well for this model using a simulation timestep of 5 seconds or greater.

This equation for the radial velocity yields a decreasing glass velocity as the glass approaches the canister wall. A function to decrease the velocity of the glass as it approaches the canister wall is applied over the radial region 0.262m < r < 0.302m to ensure a zero radial velocity at the canister inner surface.

3.0 Experimental Data

Experimental data referenced throughout this report was obtained from “RPP Pilot Melter Prototypic LAW Container and HLW Canister Glass Fill Test Results Report”, reference [1]. The HLW tests, conducted in September 2003, utilized full scale HLW canisters being filled with simulated HLW glass. The canisters were outfitted with a thermocouple tree which allowed the recording of temperatures within the canister at the centerline, 0.0762m, and 0.2286m radial locations at various heights within the canister. Additional data from the report include canister surface temperature, air flow rates, ambient air temperature, and canister weight during the glass pouring process. Specifically, the data reported from HT001 was used in benchmarking the COMSOL model as described in section 5.1.

4.0 Inputs and Assumptions

Below are the inputs and assumptions that are used in the COMSOL model. Appendix A contains COMSOL parameter and variable lists for canister dimensions, constants, and the previously derived velocity equations. All units reported below are used in the model. The COMSOL model has a base unit system of SI, but the user is allowed to input properties in any units and COMSOL converts them to the base unit system for calculations.

When referring to experimental data in Ref 1, the experimental data corresponds to the HT001 experiment.

Inputs for benchmarking model:

1) The experimental data [1] show that the airflow temperature varied between 15 ºC and 40 ºC. A time dependent temperature curve was input to the model to simulate the experimental conditions.
2) The experimental canister glass thermocouple data used were based on a scale glass melter run with a nominal pour rate [1].
3) Thermal radiation from the surface of the canister was applied by the use of the Surface-to-Surface radiation group with the enclosure surface. The enclosure wall temperature was specified, below in Table 2, based on the average enclosure thermocouple readings presented in the experimental data [1].
4) Surface to Ambient radiation heat transfer was applied to the upper section of the canister outside of the enclosure.
5) The lower fiberboard boundary that the canister is placed on is set to a floor temperature of 21 ºC.
6) Thermal radiation from the poured glass surface to the canister internal wall was applied as a heat flux boundary condition based on the average glass surface temperature and the average canister interior wall temperature.
7) The glass temperature at the canister inlet was assumed to be 1000 ºC.
8) Canister dimensions are supplied in TRR-PLT-080 [1]
9) Material properties are presented in Table 1.
10) 15 glass pours were simulated within the canister. A glass pour lasts for 27 minutes, with a subsequent 60 minute non-pour time, yielding 87 minutes between each start of pour cycle.

Table 1 - Material Properties

Material	Thermal Conductivity		Density (kg/m^3)	Specific Heat		Emissivity
Stainless Steel Canister [6,12]	W/m/K	Temperature (ºC)	8030	kJ/kg/K	Temperature (ºC)	0.4
	15	0		0.480	0	
	16.3	100		0.480	100	
	18.9	300		0.528	300	
	21.4	500		0.575	500	
Glass [7,11]	0.92 1.1 1.465 1.513 2.288 4.126 5.725	25 600 800 900 1000 1100 1150	2499 (156 lb/ft^3)	1.00 1.00 1.095 1.46 1.5404 1.504	25 300 400 500 600 1070	0.8
Air [8]#	0.0262 0.0333 0.0397 0.0457 0.0821	27 127 227 327 1000	1.161 @27 C 0.696 @227 C 0.28 @ 1000C	1.007 1.030 1.141 1.200	27 227 727 1000	NA
Fiberboard [9,10]	0.0764 0.1038 0.1428	204 427 649	448	1.13	1093	0.8

values at 1000C were obtained from the COMSOL internal materials library.

Table 2 – Average Enclosure Wall Temperature

Time (hr)	Average Wall Temperature (ºC)
0	18
17	55
19	55
34	25
44	32
50	25

5.0 Results

5.1 Model Benchmark

The COMSOL model was run with parameters to mimic the conditions used in generating the full scale data [1]. For this benchmark model, the inlet glass temperature at the top of the canister was assumed to be 1000 ºC. The glass temperature in the melter was measured to be around 1170 ºC [1], whereas the maximum temperature measured by the thermocouples in the canister was 1000 ºC. For this reason, the glass pour steam was set to 1000 ºC.

As a bounding calculation, the glass pour stream along the centerline of the canister was allowed to radiation heat to the ambient using an emissivity of 1.0. Based on the time required for the glass to travel from the canister entrance to the bottom of the canister, ~1.7 seconds, a glass inlet temperature of 1000°C and an ambient temperature of 25°C, the average, the glass will lose approximately 0.03°C. Therefore, as a simplification thermal radiation for the pour stream was not included in this model. The airflow and air temperature around the canister were varied based on experimental measurements [1]. The enclosure outside boundary was set to the average enclosure wall temperature. Additionally, the glass was added to the canister following the pour rate schedule [1].

The temperature data is pulled from the simulations to correspond to the thermocouple locations from the HLW canister test, HT001. For the experiment, and simulation output, four thermocouples were located at the centerline of the canister at heights of 0.9144m, 1.6764m, 2.4384m, and 3.3528m. Similar sets of four thermocouples were located radially in the canister at 0.0762m and 0.2286m. As seen in Figure 2 through Figure 5, the COMSOL model predicts the trends reported in the experimental data. Simulated temperatures are only reported in the figures below once the glass height in the canister reaches a level to completely cover the thermocouple.

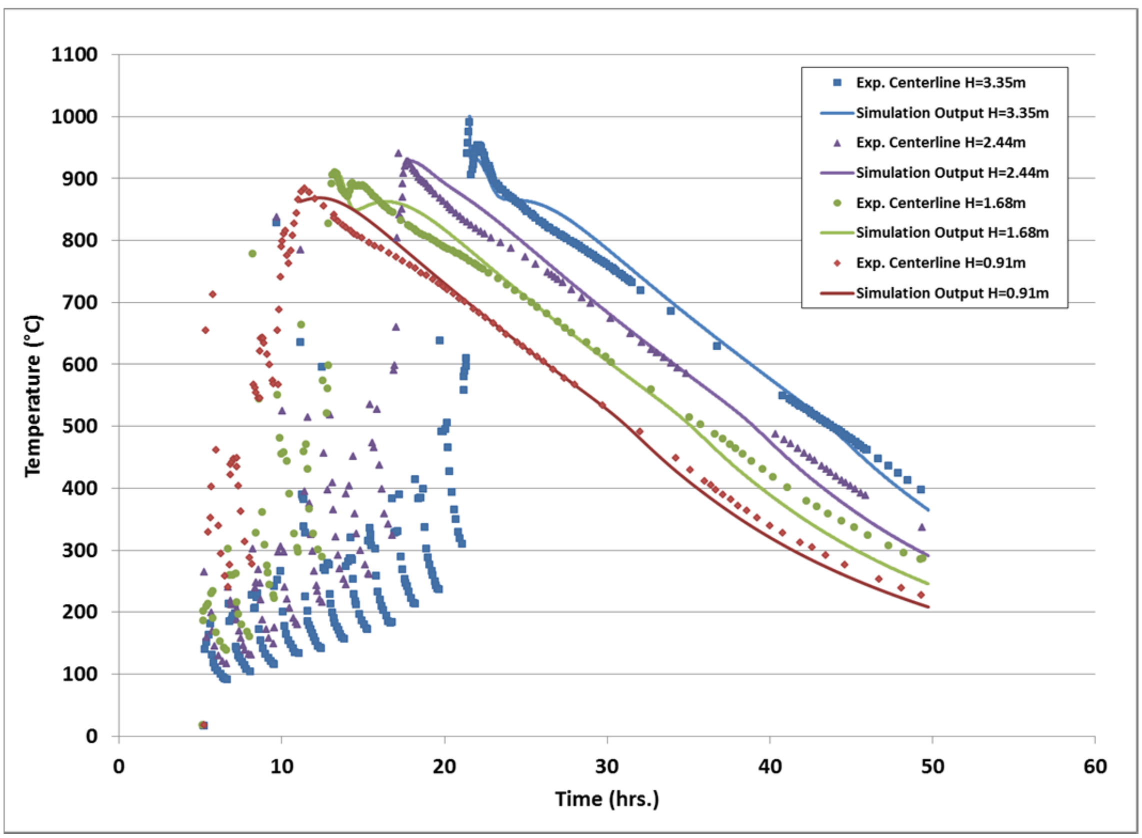

Figure 2. Comparison of canister centerline thermocouple temperatures from experimental and simulation data.

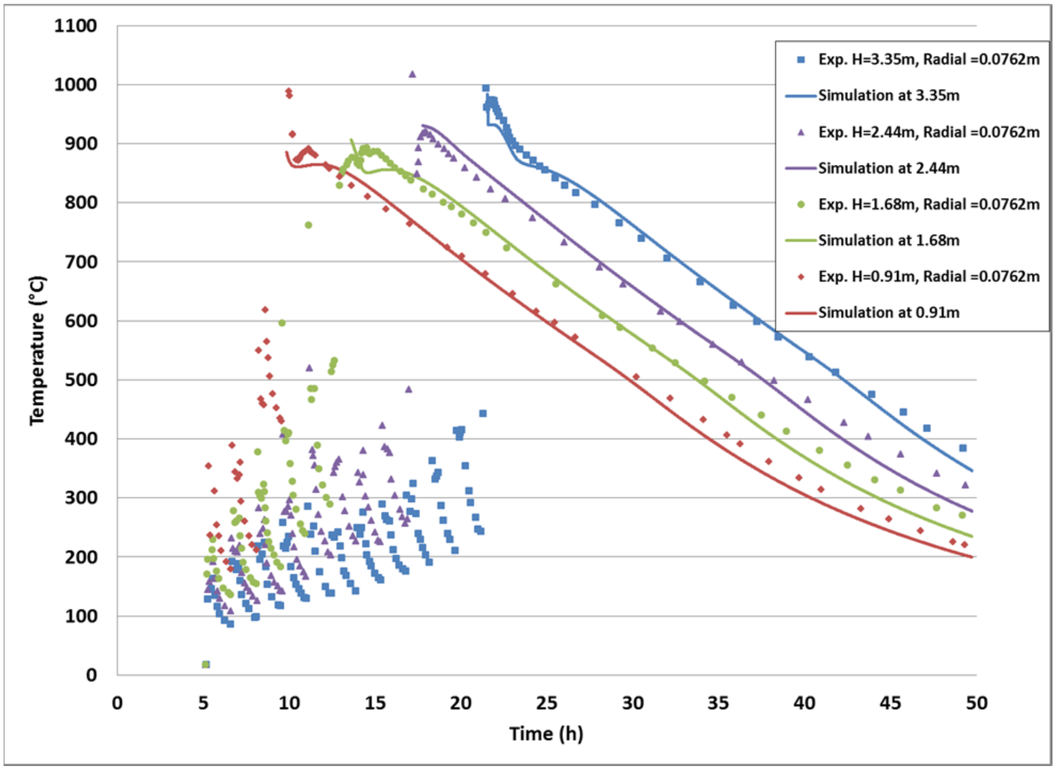

Figure 3. Comparison of canister thermocouple data from experimental and simulation data at the 0.0762m radial location.

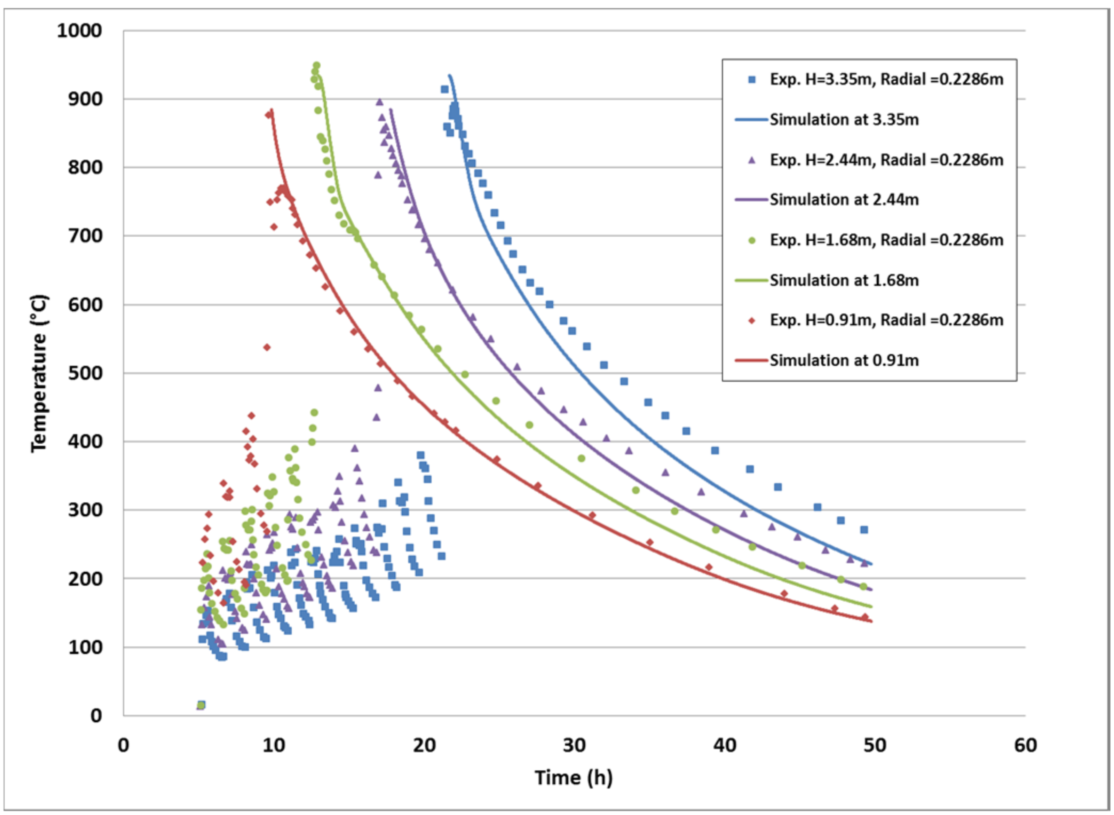

Figure 4. Comparison of canister thermocouple data from experimental and simulation data at the 0.2286m radial location.

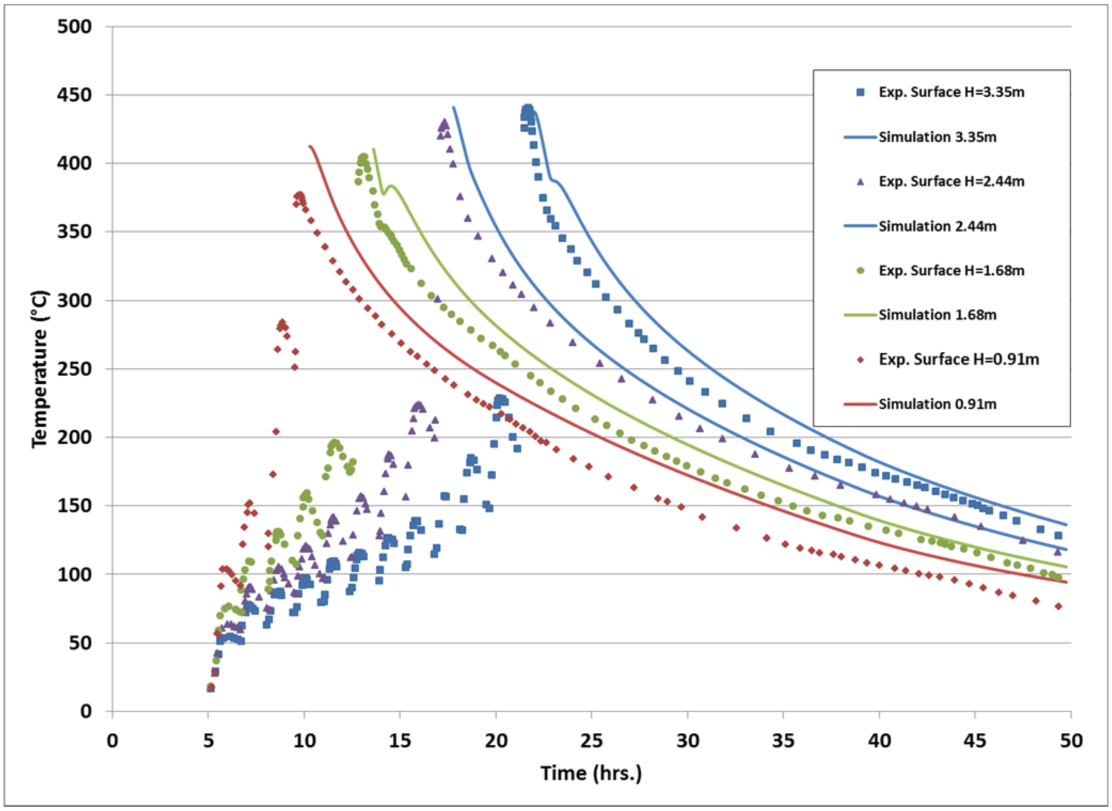

Figure 5. Comparison of canister surface thermocouple data temperature from experimental and simulation data.

The percent deviation of the simulated values compared to the experimental values was determined for the time region after the thermocouples were covered with glass. The results are shown below in Figure 6. The simulated temperature deviates up to 13% from the experimental values. As seen in Figure 6, approximately 50% of the values analyzed have a deviation of 4% or less, 75% have a deviation of 6% or less, and 90% of the points analyzed show a deviation of 9% or less compared to the experimental values. This level of precision is considered acceptable for the scoping nature of the model and the subsequent laboratory glass studies.

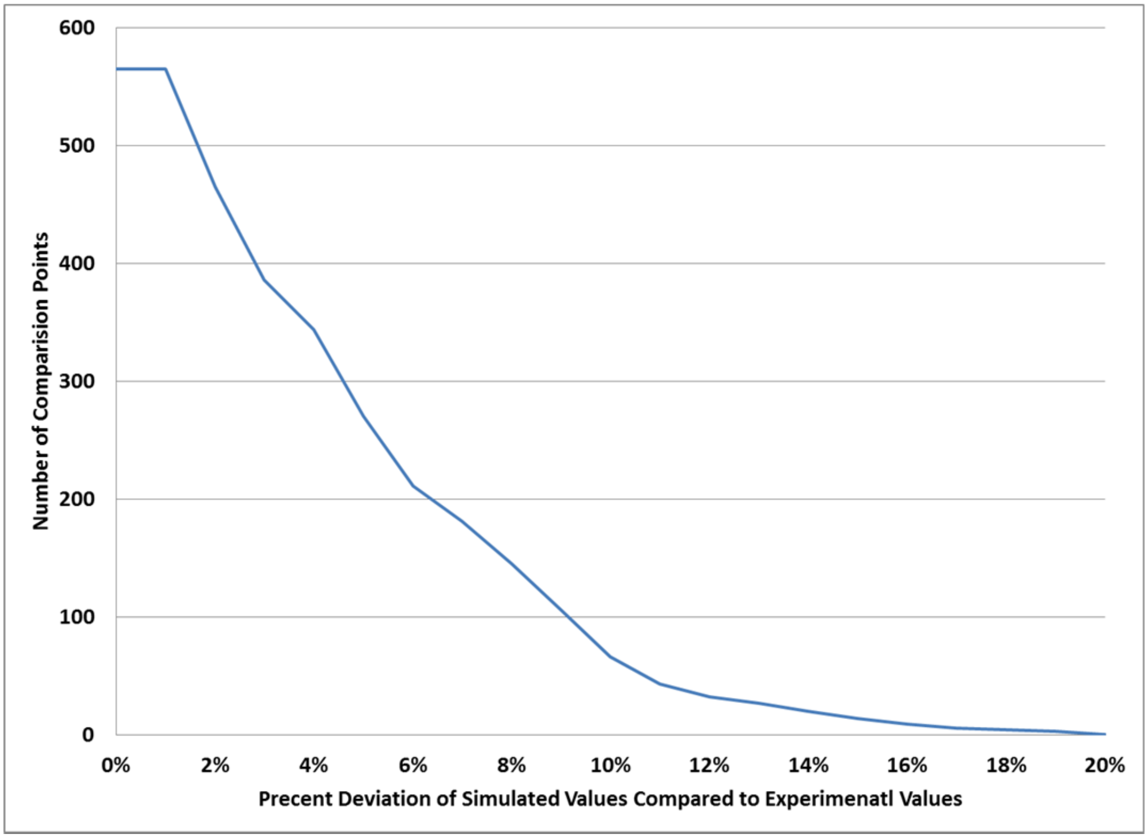

Figure 6. Percent deviation for the simulation compared to the experimental values.

5.2 Results from Flowrate Variations

After the model was developed and validated as described in Section 5.1, the model was modified for two different periodic flow rate scenarios in order to provide time and temperature data of interest to support laboratory crystallization experiments. The flow rate scenarios were selected starting from the WTP targeted conditions described in Reference [1] and shown in the first row of Table 3. For Task 1, the pour frequency time was increased by a factor of 1.33 to simulate slower melter throughput. For Task 2, the pour time was reduced by 33% to simulate a potential change in canister cooling conditions. The ambient air temperature was fixed at 26 ºC and the airflow rate was also fixed at 5.3 m/s. For each pour in Task 1 and Task 2, 212.7 kg of glass was used. As an example, for Task 1, the 212.7 kg of glass is poured during the time period of 2-27 minutes (corresponds to Pour 1). No flow of glass happens between 27-116 minutes. Pour 2 runs from 116-143 minutes, with a no pour time period of 143-232 minutes. For these parameters the glass fills the canister approximately up to the 4.27 meter fill level. The inlet glass temperature was assumed to be 1000 ºC for these scenarios.

Table 3 - Pour Rate Schedules

	Pour Time (min)	Frequency (min)	Pour Rate (kg/min)
Targeted [1]	27	87	7.78*
Task 1	27	116	7.88
Task 2	18	87	11.82

** The difference in pour rate between the Targeted and Task 1 cases is attributed to Task 1 using equal pours of 212.7 kg of glass per cycle. The Targeted case fluctuated around 209 kg of glass per pour cycle.*

Data from the simulations were collected at multiple points in the simulation corresponding to thermocouple points located at 0.00635m (¼ inch) above and below each pour level as well as the middle of each pour in the vertical direction. Radially, temperatures were obtained at 0.0254m and 0.1524m from the centerline and also at the surface of the canister.

The COMSOL model can report temperature profile for any point in the model as a post processing routine. Thermocouple data for the probes which get covered by glass at heights of 0.081m, 1.74m, 2.67m, and 3.61m (corresponding to pours 3, 6, 9, and 12) from Task 1 are plotted in Figure 7 and Figure 8, respectively, while thermocouple data from Task 2 for the same pours are plotted in Figure 9 and Figure 10. For Tasks 1 and 2, the same amount of glass is poured, but Task 2 pours at a faster rate and has less time to cool down between pours compared to Task 1. The resulting time and temperature conditions experienced by the glass in the canister will be compared via laboratory experiments to determine impacts on the potential for nepheline crystallization.

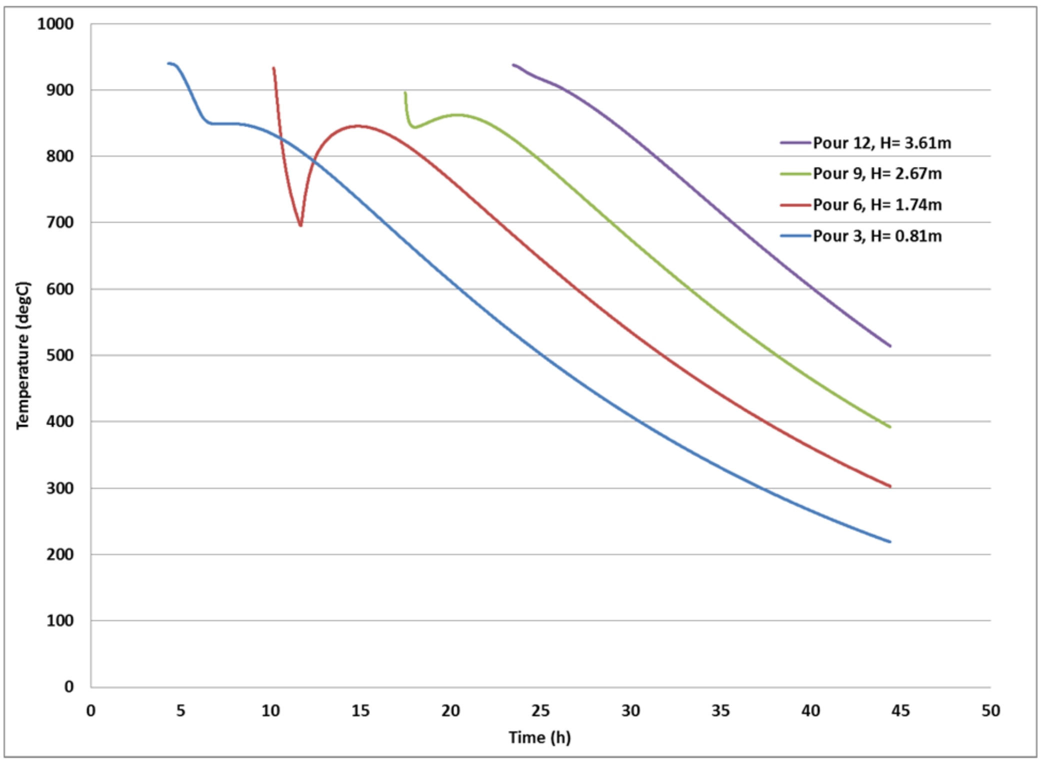

Figure 7. Simulated temperature profiles for Task 1 taken at the 0.0254m radial location for various heights.

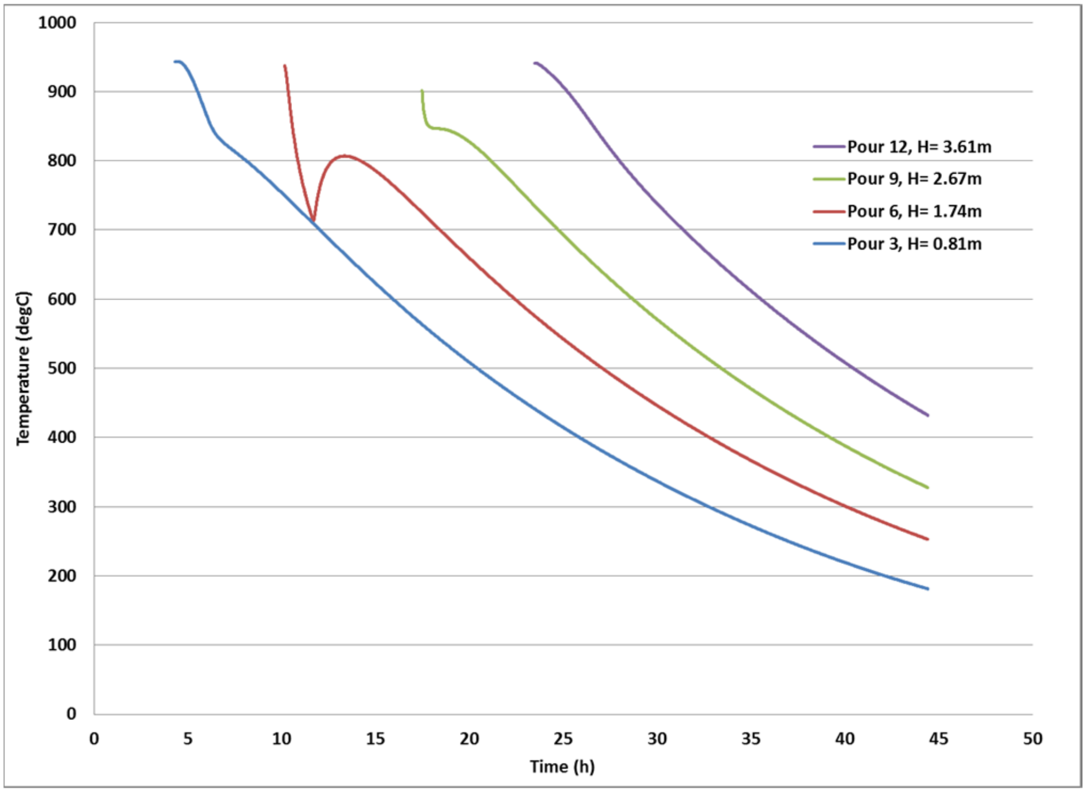

Figure 8. Simulated temperature profiles for Task 1 taken at the 0.1524m radial location for various heights.

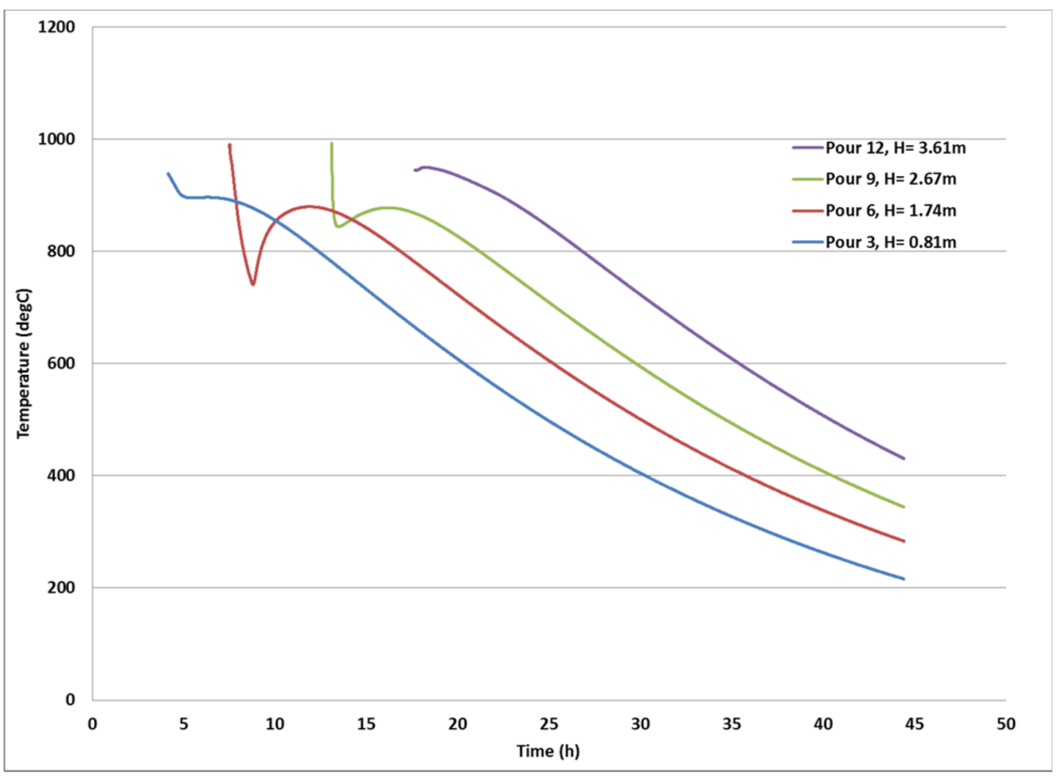

Figure 9. Simulated temperature profiles for Task 2 taken at the 0.0254m radial location for various heights.

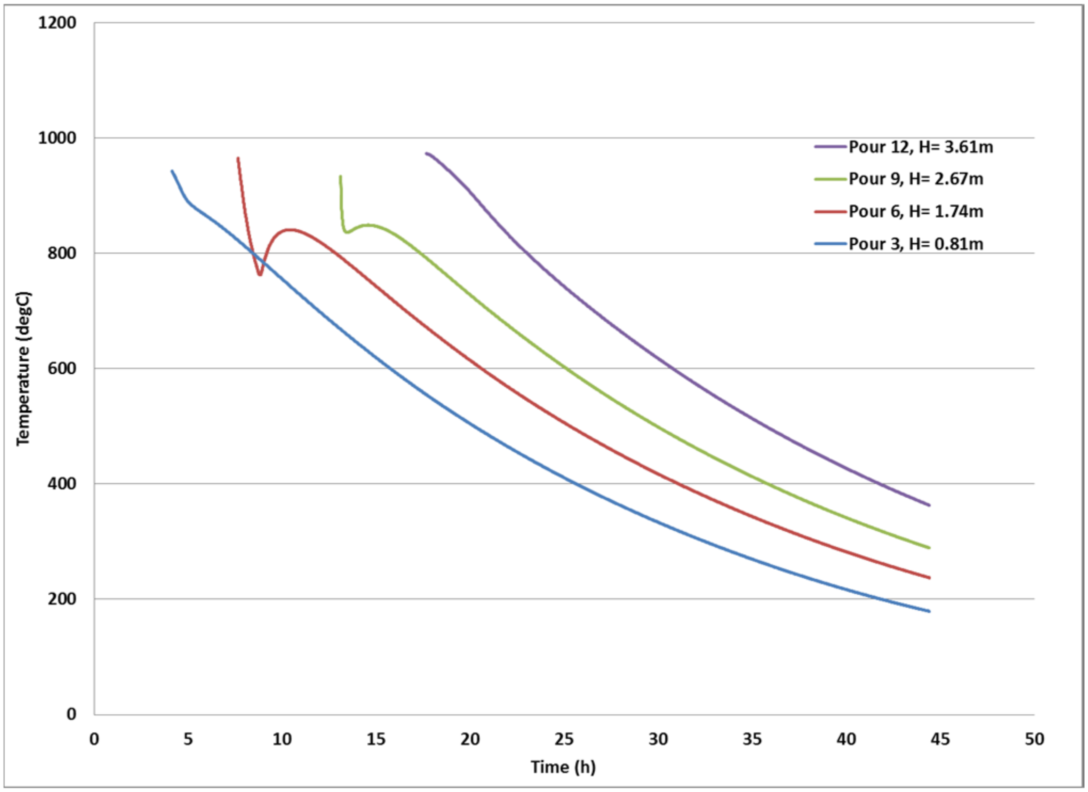

Figure 10. Simulated temperature profiles for Task 2 taken at the 0.1524m radial location for various heights.

6.0 Conclusions

A COMSOL Multiphysics model was developed to predict the glass temperatures in a WTP HLW canister during glass pouring and subsequent cooling. The intent of the model is to support scoping work in the laboratory. It is not intended to provide precise predictions of temperature profiles, but rather to provide a simplified representation of glass cooling profiles within a full scale, WTP HLW canister under various glass pouring rates. Simplifications of several material properties and boundary conditions were considered acceptable for the purposes of this model as a scoping tool. These factors (among others) can be incorporated in a more detailed, future model.

The model results were compared to available experimental data and were found to yield sufficient results for the scoping nature of the study. Initial dips, around 100ºC in the thermocouple data after being covered with glass is attributed to the deviation from the actual glass height in the canister. The model uses a constant glass density and a specific pour rate. The simulated temperatures were within 60ºC for the centerline, 0.0762m, and 0.2286m radial thermocouples once the thermocouples were covered with glass. The temperature difference between the experimental and simulated values reduced to 40 ºC, 4 hours after the thermocouple was covered, and down to 20 ºC, 6 hours after the thermocouple was covered. This level of precision is considered acceptable to support laboratory scale testing of the glass cooling profiles.

Using the model, two additional glass pouring cycles were conducted. Representative thermocouple data were plotted to show the variation between the two cycles. This provides preliminary data that will be used in laboratory experiments to determine the potential for controlling nepheline crystallization in glass by varying the glass pouring conditions.

7.0 References

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8.0 Appendix A - Comsol Constants and Expressions

The following tables are the constants and global expressions directly exported from COMSOL.

COMSOL Constants

gravity	9.8[m/s^2]
R_ig	.08206[L*atm/mol/K]
inlet_area	0.031416[m^2]
Lower_area	pi*(24[in]/2)^2
sigma	5.67e-8[W/m^2/K^4]
ContainerHeight	4.5[m]
P_atm	1[atm]
Wall_th	(3/8)[in]
CanisterR	(24/2)[in]
CrossArea	pi*CanisterR^2
Glass_T_init	1000[degC]
glass_emiss	0.8

COMSOL Global Expressions

Cp_eff	air_cp*(z>FluidHeight)+glass_cp*(z<=FluidHeight)
Entrance_Temp	(glass_T_init)*Filling+T_amb*(1-Filling)
Filling	flc2hs((LiftTime+TotalLiftTime*(FlowSchedule(t)-1))*60-t[1/s],5)*flc2hs(t[1/s]-TotalLiftTime*(FlowSchedule(t)-1)*60,5)
FluidHeight	TotalMassFunction(t)/glass_rho/CrossArea+0.027
glass_r	sqrt(PourRate/glass_rho/pi/sqrt(2*gravity*(4.49-z)*Filling
k_eff	((((10*(z[1/m]>FluidHeight[1/m]))*air_k+glass_k*(r[1/m]<glass_r[1/m]+0.03))*(z>FluidHeight))*Filling)+(1*air_k*(z>FluidHeight))*(1-Filling)+glass_k*(z[1/m]<=FluidHeight[1/m])
Q_rad1	glass_emiss*view_factor*sigma*((200[degC])^4-(mod1.T)^4)
r_glass_vel	PourRate/glass_rho/(2*pi*r*0.02[m])*Filling*(Filling>0.3)*flc2hs(0.282-r[1/m],0.02)*flc2hs(r[1/m]-0.01,0.01)*r_vel_fac*(z[1/m]>(FluidHeight[1/m]-0.02))
r_vel_air	-0.04*(z>FluidHeight)*r_vel_air_base*(1-flc2hs(z[1/m]-4.3,0.1))*(r>glass_r)*Filling
r_vel_air_base	AirMaxFlow*(-44.444*(r[1/m])^2+13.333333*r[1/m])
rho_eff	air_rho*(z>FluidHeight)+glass_rho*(z<=FluidHeight)+(glass_rho-air_rho)*(r[1/m]<=glass_r[1/m])*(z>FluidHeight)*Filling
view_factor	1[1]
z_vel_glass	z_vel_glass_base*flc2hs(glass_r[1/m]-r[1/m],0.0001)*(z[1/m]>=FluidHeight[1/m])*Filling*flc2hs(z[1/m]-0.08,0.02)
z_vel_glass_base	-(9.8*((4.6-z[1/m])/4.9)^(0.5))[m/s]

Distribution:

N. M. Askew, 703-41A
J. W. Amoroso, 999-W
T. B. Brown, 773-A
H. H. Burns, 773-41A
A. D. Cozzi, 999-W
D. A. Crowley, 773-42A
C. S. Dandeneau, 999-W
D. E. Dooley, 773-A
A. P. Fellinger, 773-42A
S. D. Fink, 773-A
G. P. Flach, 773-42A
K. M. Fox, 999-W
J. C. Griffin, 773-A
T. Hang, 703-41A
E. K. Hansen, 999-W
C. C. Herman, 773-A
D. T. Hobbs, 773-A
E. N. Hoffman, 999-W
J. E. Hyatt, 773-A
C. M. Jantzen,
F. C. Johnson, 999-W
M. R. Kesterson, 703-41A
K. M. Kostelnik, 773-42A
J. E. Laurinat, 703-41A
S. Y. Lee, 703-41A
B. B. Looney, 773-42A
D. J. McCabe, 773-42A
D. L. McClane, 999-W
T. O. Oliver, 773-42A
F. M. Pennebaker, 773-42A
J. A. Pike, 703-41A
M. R. Poirier, 773-42A
F. G. Smith, 773-42A
M. E. Stone, 999-W
C. L. Trivelpiece, 999-W
B. J. Wiedenman, 773-42A
W. R. Wilmarth, 773-A
Records Administration (EDWS)

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J. Matyáš, PNNL
D. K. Peeler, PNNL
M. J. Schweiger, PNNL
J. D. Vienna, PNNL