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HEAT TRANSFER ANALYSIS FOR ION-EXCHANGE COLUMN SYSTEM

Si Y. Lee* and William D. King
Savannah River National Laboratory
Savannah River Site
Aiken, SC 29808
Phone: *(803) 725-8462; (803) 725-7556
*si.lee@srnl.doe.gov; william02.king@srnl.doe.gov

ABSTRACT

Models have been developed to simulate the thermal characteristics of Crystalline Silicotitanate (CST) ion exchange media fully loaded with radioactive cesium in a column configuration and distributed within a waste storage tank. This work was conducted to support the Small Column Ion Exchange (SCIX) program which is focused on processing dissolved, high-sodium salt waste for the removal of specific radionuclides (including Cs-137, Sr-90, and actinides) within a High Level Waste (HLW) storage tank at the Savannah River Site. The SCIX design includes CST columns inserted and supported in the tank top risers for cesium removal. Temperature distributions and maximum temperatures across the column were calculated with a focus on process upset conditions. A two-dimensional computational modeling approach for the in-column ion-exchange domain was taken to include conservative, bounding estimates for key parameters such that the results would provide the maximum centerline temperatures achievable under the design configurations using a feed composition known to promote high cesium loading on CST.

The current full-scale design for the CST column includes one central cooling pipe and four outer cooling tubes. Most calculations assumed that the fluid within the column was stagnant (i.e. no buoyancy-induced flow) for a conservative estimate. A primary objective of these calculations was to estimate temperature distributions across packed CST beds immersed in waste supernate or filled with dry air under various accident scenarios. Accident scenarios evaluated included loss of salt solution flow through the bed, inadvertent column drainage, and loss of active cooling in the column.

The modeling results demonstrate that the baseline design using one central and four outer cooling tubes provides a highly efficient cooling mechanism for reducing the maximum column temperature.

Keywords: Ion Exchange Column, Computational Heat Transfer, Natural Convection, Thermal Performance

INTRODUCTION

The Small Column Ion Exchange (SCIX) project is designed to accelerate closure of High Level Waste (HLW) tanks at the Savannah River Site (SRS). The SRS tanks store HLW in three forms: sludge, saltcake, and supernate. An in-tank ion exchange process is being designed to treat supernate and dissolved saltcake waste. Through this process, radioactive cesium from the salt solution is adsorbed into the ion exchange media (Crystalline Silicotitanate - CST) which is packed within a flow-through column. A packed column loaded with radioactive cesium generates significant heat from radiolytic decay. If engineering designs cannot handle this thermal load, hot spots may develop locally within the packed bed which could degrade the performance of the ion-exchange media. Performance degradation with regard to cesium removal has been observed between 50 and 80°C for CST [1]. In addition, the waste supernate solution will boil around 130°C. If the columns boiled dry, the sorbent material could plug the column and lead to replacement of the entire column module.

The objective of the present work is to compute temperature distributions across a CST-packed bed immersed in waste supernate and a dry, air-filled CST column under accident scenarios including loss of salt solution flow through the bed, complete loss of fluid inside the bed, and loss of coolant system flow. In addition, temperature distributions will be evaluated for a spent CST mound located on the tank floor and a layer of CST dispersed evenly across the tank floor. This is a potential fate of the spent CST after removal from the column. The spent CST will be ground prior to transfer to the tank in preparation for vitrification processing in the SRS Defense Waste Processing Facility (DWPF). Thermal evaluations were performed for the baseline design conditions of the ion-exchange column and the in-tank regions as shown in Figs. 1 and 2. The customer requested that calculations be conducted in such a manner as to ensure conservative and bounding results for the maximum temperatures achievable using the current baseline design.

The current thermal modeling evaluations assumed the maximum bounding cesium loading considered possible based on current knowledge regarding CST media and assumed project controls with regard to feed qualification. Since this cesium loading was considerably higher than the nominal loading conditions in SRS waste, fractionally-decreased thermal loading cases were also evaluated. The baseline design for the CST column was used for the initial calculations as shown in Fig. 1. Detailed sensitivity analysis with respect to the initial baseline results were performed in order to identify key parameters that significantly impact the thermal performance. A temperature limit of 130 °C based on the salt solution boiling point was used as a measure for the evaluation of the in-column cases, although boiling cannot occur for the air-filled column case since no liquid is present.

For in-tank evaluations, the equipment configuration shown in Fig. 2 involving a typical SRS Type-IIIA tank, Tank 41, as the baseline configuration. The location of the heat source region on the tank floor due to the accumulation of CST material was assumed to be just under the grinder. The shape of the CST heat source was assumed to be hemi-spherical for the most conservative evaluations. Selected alternative configurations involving other geometrical shapes for the CST mound were evaluated to ensure that the most conservative shape was used. In addition, evenly distributed layers of CST media were evaluated to simulate an ideally mixed tank. A tank wall temperature limit criterion of 100 ºC was used for in-tank evaluations based on current SRS tank structural integrity temperature limits [5]. Sensitivity analysis for the in-tank region was performed for different amounts of CST and combinations of CST with loaded Monosodium Titanate (MST) and sludge materials.

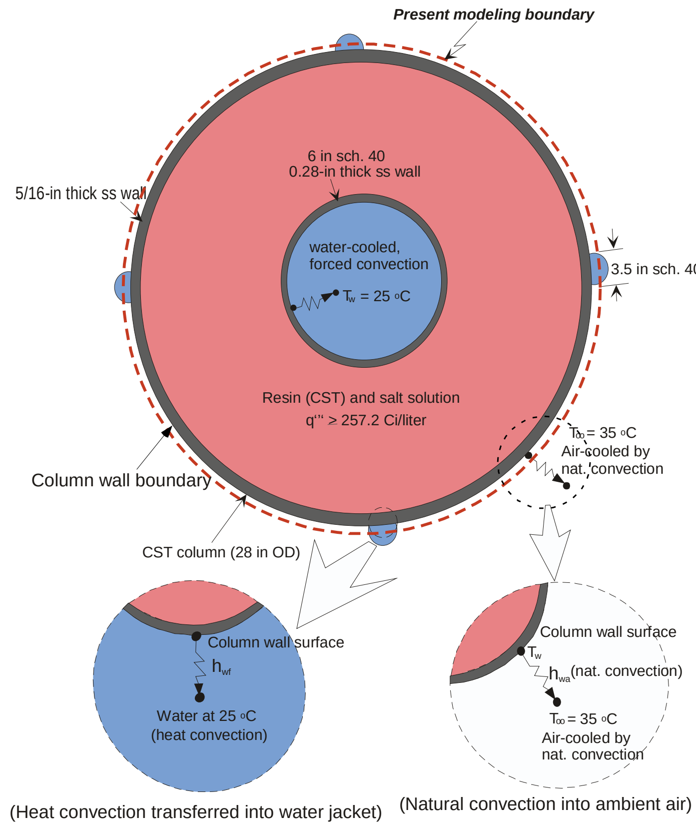


Figure 1. Baseline modeling domain for the ion-exchange column with CST media

NOMENCLATURE

A	Area (m^2)
°C	Degree Centigrade (or Celsius)
C	Coefficient
Ci	Curie (= 3.7 x 10^{10} disintegrations/sec)
C_p	Specific heat (J/kg-K)
d_h	Hydraulic diameter (m)
ft	Foot (=0.3048m)
gallon	3.7854x10^{-3} m^3
Gr_L	Grashof number based on length scale L
h_w	Wall heat transfer coefficient (W/m^2-K)
h_{wf}	Forced convective heat transfer coefficient (W/m^2-K)
in	Inch (=0.0254m)
J	Energy unit (Joule)
k	Thermal conductivity (W/m-K)
K	Absolute temperature (=273.15+ °C)
kg	Kilogram
L	Length (m) or liter (0.001m^3)
m	Meter or coefficient
Pr_f	Prandtl number
q'''	Volumetric heat source (W/m^3)
q_w''	Wall heat flux (W/m^2)
R	Energy residual (W)
Re	Reynolds number ($d\rho u/\mu$)

s or sec	Second
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
t	Time (second) or degree Centigrade
T_∞	Ambient temperature (K)
V_F	Cement volume containing the hydration heat (m^3)
W	Watts (J/sec)
x, y	The coordinate system for the two-dimensional domain as shown Fig. 1
α	Thermal diffusivity (m^2/sec)
ρ	Density (kg/m^3)
β	Thermal expansion coefficient (K^{-1})
μ	Dynamic viscosity (kg/m-s)
ν_f	Kinematic viscosity (m^2/s)

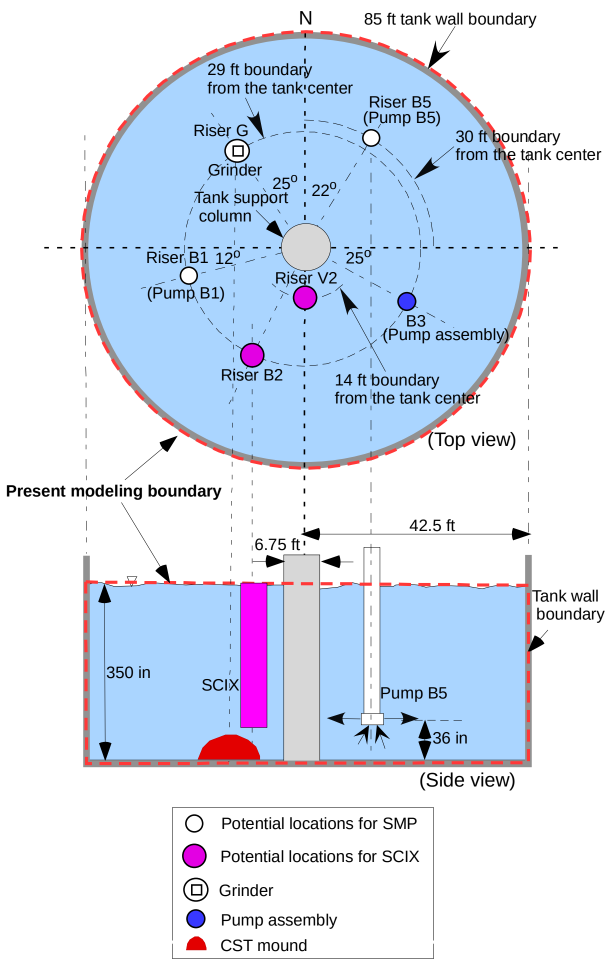


Figure 2. Initial baseline three-dimensional modeling boundary for in-tank calculations in the Tank 41 facility

MODELING APPROACH AND SOLUTION METHOD

The SCIX modeling and analysis scope included two main domain areas. One involves the in-column heat transfer analysis for an ion exchange column containing CST and either salt solution or air. The other is an in-tank domain which includes the entire waste tank with accumulated spent CST materials on the floor.

The SCIX in-tank cesium-removal system contains two ion-exchange column modules and one IX media grinder inside an 85-ft diameter SRS Type-IIIA tank. The column module is designed for cesium removal from an SRS High-Level Waste (HLW) salt solution containing numerous radioactive species. The columns are packed with CST ion exchange media. The baseline design includes a 15 foot tall column with an annular design which contains 450 gallons of CST media. The supernate is an alkaline, concentrated sodium salt solution (nominally 6 M Na^+). Through this process, radioactive cesium from the salt solution is adsorbed onto the ion exchange media, which is packed within the flow-through column. The packed ion exchange column loaded with radioactive cesium (~5 x 10^5 Ci) generates significant heat from radiolytic decay. Under normal operating conditions, process fluid flow through the column can provide adequate heat removal from the system through a coupled conduction and convection heat transfer mechanism. However, in the case of loss of fluid flow or inadvertent solution leakage from the column, there are safety concerns about the thermal response rate of the fully-loaded column and the effectiveness of the column cooling system. If engineering designs cannot handle this thermal load, hot spots may develop locally within the bed which could degrade the performance of the ion-exchange media. The waste supernate solution will also boil around 130^{o}C. If the columns boiled dry, the resulting solid sodium salts could foul and plug the column. The baseline design for the column module shown in Fig. 1 is used as the calculation domain. The baseline modeling conditions used for the in-column analysis are provided in Table 1.

For computational modeling purposes, a conservative approach was taken by assuming that the primary cooling mechanisms inside and outside of the column are conduction and natural convection, respectively, and axial heat removal effects from the column are negligible compared to radial heat transfer. A two-dimensional transient heat conduction model was developed to assess the thermal performance of the CST column with loss of flow using the prototypic geometry. Heat transfer calculations of the CST column were performed for a given boundary condition by using a computational heat transfer approach on a Cartesian x-y grid under a commercial CFD code, FLUENT. For the computational domain, about 8,000 mesh nodes for the in-column thermal analysis were established by the mesh sensitivity analysis.

Table 1. Baseline modeling conditions used for the heat transfer analysis of the ion exchange column.

Models	Conditions for the baseline model
Column heat load	257.22 Ci/liter (1273.24 W/m^3) [8]
CST material porosity	24.0% [7]
Column hydraulic conditions	no flow, or 5 gpm flow*
Column media	wet or dry column*
Granular bed conditions	fixed bed
Initial temperature	35 ^{o}C for the entire computational domain
Ambient temperature	35 ^{o}C (55 ^{o}C)*
Heat transfer coefficient at wall, h_w(W/m^{2}sec)	238 (for 6-in water pipe wall), and 620 (for the column wall surface attached to the water jacket),1.5 W/m^{2}sec (typical natural convection) [10]**
Coolant water flowrate in cooling jackets	6.25 gpm each side jacket, 12.5 for annular central coolant pipe
Coolant water temperature	no forced circulation or 25 ^{o}C (35 ^{o}C)* fixed by forced circulation
Bed porosity	43.2% [7,10,11]

#All Curies assumed converted to heat load wattage
*Conditions to be evaluated by sensitivity analysis
** Heat transfer coefficient at the exterior wall of the CST column

The model considers two basic process scenarios with no fluid flow. One case involves a packed CST bed filled with salt solution, while the other involves a packed CST bed filled with air and no salt solution. The dry column could potentially result from processing accidents such as inadvertent fluid drainage resulting from incorrect valve operations or column overheating and solution boiling. Spherical CST particles are assumed to be homogeneously packed inside a stainless steel cylinder that is 28 inches in diameter with a 0.5 inch thick wall. Detailed material and thermal properties for the wet and dry CST columns are summarized in Table 2. The CST packed bed porosity was estimated to be about 43.2 % based on ORNL measurements [7]. The void volume fraction of the packed bed has a substantial impact on estimations of the thermal conductivity of a composite mixture. In the ORNL work, the bulk density of the CST column filled with air was estimated to be about 1,168 kg/m^3 assuming that the density of CST solid is 2,056 kg/m^3. Modeling calculations for the in-column analysis used the following assumptions (unless otherwise indicated) in order to ensure conservative results for the maximum temperatures.

- The column is filled with a fixed, packed bed of CST particles with homogeneous packing.
- The CST bed is immersed in salt solution or air with no active or convective fluid flow through the bed.
- The CST particle and salt solution (or air for the dry bed case) are in local thermal equilibrium so that an average effective thermal conductivity can be assumed for the packed bed.
- The column is suspended in unventilated dry air at 35 ºC rather than salt solution within the High Level Waste tank head space.
- The initial heat source term used of 257 Ci/L of packed bed is 115% of the maximum cesium loading of 223 Ci/L predicted for the various SRS waste compositions previously considered for SCIX processing [8]. The heat source was calculated assuming secular equilibrium involving ^{137}Cs and ^{137m}Ba decay. The heat source is assumed to be uniformly distributed throughout the entire packed column as would be expected for cesium-saturated media. This Curie loading corresponds to 1.273 kW/liter.
- Outside the column there is no forced convective airflow, so natural convection is the primary heat transfer mechanism from the exterior column wall. Radiative cooling contributions at the outer wall surfaces of the column are also considered.
- A typical natural convective heat transfer coefficient (h_w) of 1.5 W/m^{2}K was used as an external wall boundary condition based on previous analysis [3].

Table 2. Material and thermal properties for heat transfer calculations of the CST, column, tank and soil

Material	Thermal conductivity (W/mK)	Density (kg/m3)	Specific heat (J/kgK)
CST [7]	0.1617	2056.3**	1052.3
Salt Solution [8]	0.68	1232.0	3630.0
CST-Salt Solution	0.4125#	1587.8# (from eqn. 8)	2517.3# (from eqn. 9)
Ground CST-Salt Solution	0.3386#	1723.2# (from eqn. 8)	2094.1# (from eqn. 9)
CST-Air##	-1.0922x10^{-2}+4.0960x10^{-4}T*	1168.0***	1031.9***
Stainless steel	17.30	7800.0	486.0
Concrete	1.5	2400	750
Ceramic	18.0	3690	880
soil	1.25	2000	1450

based on non-linear empirical correlation of Krupiczka at 25 ºC [15] considering particle porosities ($\varepsilon_{CST,\ particle}$ = 24%, $\varepsilon_{RF,\ particle}$ = 65.79%) and the volume fractions of air or fluid in the packed beds (0.432 for CST bed), giving total bed porosities of 0.57 for CST (total porosity evaluated considering bead and bed porosity.). In case of ground CST for the in-tank modeling analysis, porosity is assumed to be reduced by 50% from the void filling with smaller ones.
* T is absolute temperature in K [16]
** based on material density (not bulk density)
*** based on the condition that volume fraction of fluid or air in packed bed is 0.432 at 25ºC temperature

When the column becomes dry as a result of accidental drainage or solution boiling, the following additional assumptions were used.

- The CST material is completely dry throughout the bed and remains homogeneously packed.
- The air-packed column volume remains fixed relative to the initial packed configuration.

- Chemical reactions of the dried CST media material that could lead to changes in the thermal or physical properties of the packed bed are neglected.
- Air convection inside the column is conservatively neglected and only conductive heat transfer is considered.
- Radiative cooling contributions to the heat transfer at the inner column wall surfaces are neglected.

Using the modeling boundary shown in Fig. 1, the in-column modeling calculations were performed for a range of conditions to estimate maximum bed temperatures in a conservative way. The in-column modeling conditions used for the present analysis are summarized in Table 3.

Table 3. Modeling cases used for the in-column analysis

CST loading (Curie/liter)	Column Hydraulic conditions	Engineered cooling system	
		Central cooling system	External cooling system
257 (Baseline loading)	5 gpm flow	Active	Active
	Stagnant (Wet)	Active	Active
	Stagnant (Wet)	Inactive	Inactive
	Stagnant (Dry)	Active	Active
	Stagnant (Dry)	Inactive	Inactive
300	Stagnant (Wet)	Active	Active
	Stagnant (Wet)	Inactive	Inactive
	Stagnant (Dry)	Active	Active
	Stagnant (Dry)	Inactive	Inactive

For a conservative calculation, a low temperature gradient at the wall boundary layer was used to estimate the natural convection capability for the present geometrical configurations. The heat transfer coefficient (h_w) for natural convective cooling under a turbulent flow regime ($Ra_f = Gr_L Pr_f > 10^9$) is given in terms of non-dimensional numbers empirically.

$$Nu_L = \frac{h_w L}{k_w} = C(Gr_L Pr_f)^m \quad \text{for } Gr_L Pr_f < 10^{12} \qquad (1)$$

where C and m are the coefficients determined from literature data and L is the characteristic length of the CST column.

For the present geometrical configuration, C=0.10 and m=0.333 are given by Warner and Arpaci using the experimental data [12]. From eq. (1), the heat transfer coefficient (h_w) is about 1.5 W/m^{2}K corresponding to $Nu_L \approx 254$ conservatively under the present conditions. Heat transfer coefficients (h_{wf}) for forced convective heat transfer mechanisms through the column wall attached to the water jackets and through the inner surface of the coolant pipe at the column center were estimated by Dittus-Boelter's correlation [13]. That is,

$$Nu_d = \frac{h_{wf} d_h}{k_{wf}} = 0.023 (\mathrm{Re}_d)^{0.8} (\mathrm{Pr}_{wf})^a \quad \text{for } Re_d > 2000 \qquad (2)$$

Equation (2) is applicable to turbulent flow when the Reynolds number is larger than 2,000 in terms of the hydraulic diameter d_h, and the parameter a in eq. (2) is 0.4 when the fluid is heated as modeled in the present work. The Reynolds number for the present study is about 7,000 when 6.25 gpm flows through the 3.5-in half-moon coolant tubes, which corresponds to 0.25 m/sec flow velocity. In the present work, some modeling cases include active engineered cooling systems with a forced convection mechanism as shown in Table 3. Forced convection heat transfer coefficients at the water jackets (h_{wf}) attached to the exterior of the column wall and at the inner surface of 6-in water pipe were estimated by eq. (2). From the baseline modeling conditions, the wall heat transfer coefficient governed by a forced convection mechanism was estimated as h_{wf} = 238 (W/m^{2}K) for the wall surface of the 6-in central coolant pipe and h_{wf} = 620 (W/m^{2}K) for the wall of 3.5-in water jacket. Table 3 presents the modeling conditions for the baseline design of the 28-in cesium-saturated CST column. Table 4 shows a range of total heat loads generated by the SCIX column. These heat loads were used as the volumetric heat source term q''' for the modeling calculations.

The solution method has been established to calculate steady-state and transient temperature responses of the column system to the heat load q'''. The transient calculations were continued until maximum temperatures for the components were reached. In this work, two temperature limits were used for the operation and safety criteria in the thermal evaluation of the SCIX system. One was an operating temperature limit to prevent overheating of tank supernate, which is the 55^{o}C liquid temperature limit for the entire liquid domain of Tank 41 containing the CST column modules and spent CST mound. The other is used as a safety limit, which is the 100^{o}C temperature limit for the corrosion control of the tank wall material [5].

Complete setup of the modeling calculations requires the input parameters such as thermal and material properties of the components, heat source term, and initial boundary conditions along with the established modeling domain. For the heat transfer analysis of the CST column, the energy balance equation is applied to the two-dimensional computation domain as shown in Fig. 1, assuming that the axial heat transfer of the column is negligible. For conservative heat transfer calculations, the heat source was estimated for a fully-loaded and uniformly-distributed bed packed with CST solid material. The initial calculations used 257 Ci/liter for CST, corresponding to 1.273 watts/liter, as volumetric heat source q''' as shown in Table 4. The transient model considered temperature-dependent thermal properties to predict transient thermal responses of the fixed bed region in the case of loss of solution flow.

Table 4. Heat source terms for the baseline column shown in Fig. 1.

Column height (ft)	Total column vol. (liters)	Volumetric heat load, q''' Ci/liter [watts/liter]*	Total heat sources generated by column loading (watts)
10	1154.7	257 [1.273]**	1470
		300 [1.485]	1715
15	1732.0	257 [1.273] **	2205
		300 [1.485]	2572
25	2886.6	257 [1.273] **	3675
		300 [1.485]	4287

*Conversion factor for Cs-137 decay heat is 0.00495 watts/Ci.
**Baseline loading.

For computational efficiency, an effective thermal conductivity for the composite column region was used. The effective thermal conductivity of the CST bed region was estimated by a literature correlation [7]. That is, the effective thermal conductivity of the bed ($k_{b,eff}$) was developed as a function of the bed porosity, ε, in SI units (W/mK) using the literature experimental data.

$$k_{b,eff} = k_f \left(\frac{k_{peff}}{k_f} \right)^{A + B \log\left(\frac{k_{peff}}{k_f} \right)} \quad (3)$$

where

$A = 0.280 - 0.757\log\varepsilon$ and $B = -0.057$. (4)

$$k_{peff} = \varepsilon_p k_f + (1-\varepsilon_p) k_p \quad (5)$$

In eq. (3), k_{peff} is the effective thermal conductivity of a CST particle considering particle porosity, ε_p. k_f in eq. (3) is the thermal conductivity of the stagnant fluid trapped inside the porous CST particle. Coefficient A is a function of the bed porosity, ε. The thermal conductivity of the CST particle (k_p) is assumed to be constant for computational efficiency.

Effective material properties of the CST column are computed in terms of the bed porosity of the packed column, □. Effective density, $\rho_{b,eff}$, and specific heat, $Cp_{b,eff}$, of the bed column are based on a homogeneous assumption. That is,

$$\rho_{b,eff} = \varepsilon \rho_f + (1-\varepsilon)\rho_{peff} \quad (6)$$

Effective particle density, ρ_{peff}, is given by the particle porosity, ε_p.

$$\rho_{peff} = \varepsilon_p \rho_f + (1-\varepsilon_p)\rho_p \quad (7)$$

$$Cp_{b,eff} = \varepsilon Cp_f + (1-\varepsilon) Cp_{peff} \quad (8)$$

Effective particle specific heat, Cp_{peff}, is given by the particle porosity, ε_p.

$$Cp_{peff} = \varepsilon_p Cp_f + (1-\varepsilon_p) Cp_p \quad (9)$$

In eqs. (6) and (7), subscripts f and p refer to the fluid and particle materials within the packed bed, respectively. Computational time can be reduced by modeling a single-material region with the effective thermal conductivity instead of modeling a multi-material region composed of two different materials.

Thermal performance calculations were performed by employing two temperature limits as discussed earlier. Safety criteria limits for the column solution and tank wall temperatures are assumed to be 130 ºC and 100 ºC, respectively. These criteria were selected to prevent waste supernate boiling and to avoid structural damage to the tanks. A temperature limit of 55 ºC for the in-tank solution outside the CST column is also assumed for operational control. Using these temperature criteria, various thermal calculations for the in-column module and the in-tank domain were made to quantify key design and operating parameters and evaluate performance with and without engineered cooling systems. For the case of the air-filled column, a series of transient modeling calculations were conducted to determine the maximum bed temperature as a function of time.

This analysis is conservative by nature and gives bounding temperature data. Only conductive heat transfer was considered and it was assumed that the thermal conductivity of the CST material was constant with temperature. Additional transient calculations were conducted under the wet and dry column conditions using a cesium loading of 300 Ci/liter. The modeling results provide quantitative information associated with process heat control and management of the SCIX design.

The energy balance equation defined by the computational grid was solved by an iterative solution method. The detailed solution method was described in the previous work [2]. The overall energy balance should be checked to demonstrate the adequacy of the grid fineness used. This was done by using eq. (10).

$$R = -\int_{A_W} q_w'' dA + q''' V_b \quad (10)$$

The volumetric heat source term, q''', in eq. (10) is given by the code input. For all the cases considered here, the absolute value of the energy residual (R) was maintained at a value less than 0.5 watts. For instance, the residual results for the wet column model with active central and external cooling systems are shown as function of the grid number in Fig. 3. For the in-column analysis, an optimum grid of about 8,100 cells for the 28-in column was established from the grid sensitivity analysis under the Linux high performance platform.

The baseline configuration for the in-tank thermal analysis is shown in Fig. 2. For the in-tank analysis, there are safety concerns about reaching the maximum allowable temperature at the tank wall region under the CST mound since the spent CST material is dropped to the treatment tank floor for interim storage and the tank wall temperature cannot be higher than 100 ºC for preventing the tank wall corrosion.

Three-dimensional in-tank heat transfer models were developed to estimate the maximum temperatures for the liquid and for hot spots on the tank floor under conservative and bounding assumptions. The initial thermal calculations for the entire in-tank domain considered two geometrical cases involving 6,000 gallons of unground spent CST present on the tank floor as a hemispherical mound or as evenly flat layer. A volume of 6,000 gallons was selected by the customer based on the maximum amount of spent CST that might accumulate on the tank floor during processing. As shown in Fig. 4, the location of the CST mound for Case 1 is just under the grinder region while the flat layer for Case 2 covers the entire bottom tank surface. When the cooling capability is assumed to be uniform over the entire surface area of the 6000-gallon CST mound with its adiabatic bottom surface, heat flow per unit surface area for the flat layer is about 17 times less than that of the hemispherical mound as compared in Table 5. As shown in Fig. 5, about 2,000,000 computational nodes are established for the in-tank calculations with 6000-gallon hemispherical CST mound. Material and thermal properties assumed for the in-tank calculations are provided in Table 2.

RESULTS AND DISCUSSIONS

The present thermal modeling calculations primarily consist of two modeling domains. One domain involves the in-column heat transfer analysis case shown in Fig. 1. The other domain involves the in-tank analysis with CST media on the tank floor shown in Figs. 2 and 4. For in-column cases involving stagnant liquid, convective heat transfer mechanisms associated with movement of the mobile liquid phase within the column were assumed to be negligible as a conservative estimate of the maximum column temperature. The external wall surfaces of the wet and dry columns were cooled by natural convection coupled with radiation.

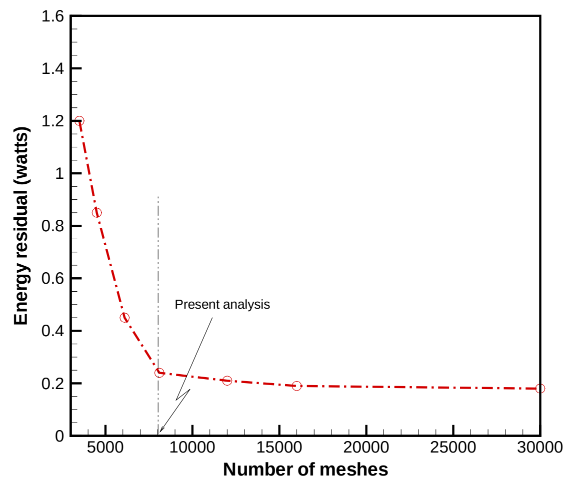
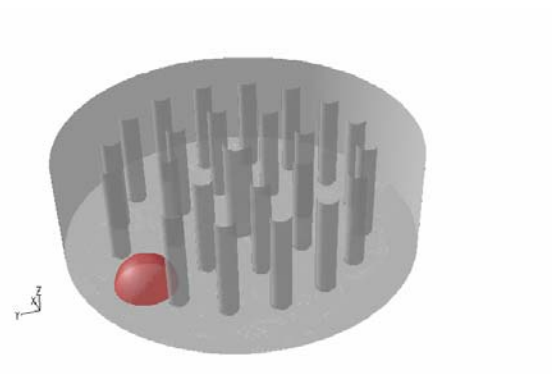
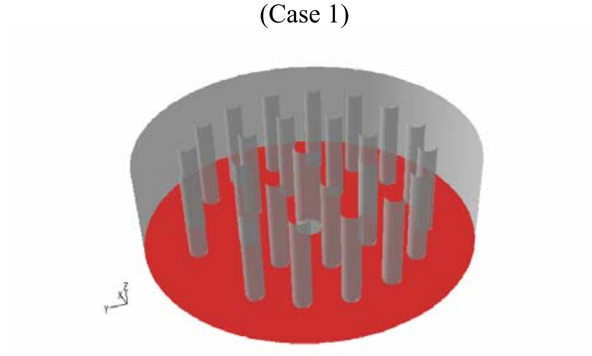


Figure 3. Sensitivity results associated with numerical energy residual showing that approximately 8,100 meshes are sufficient for the present analysis



(Case 1)



(Case 2)

Figure 4. Computational domains for the hemispherical mound (Case 1) and the flat layer (Case 2) formed by CST accumulation on the tank floor (red indicates CST media)

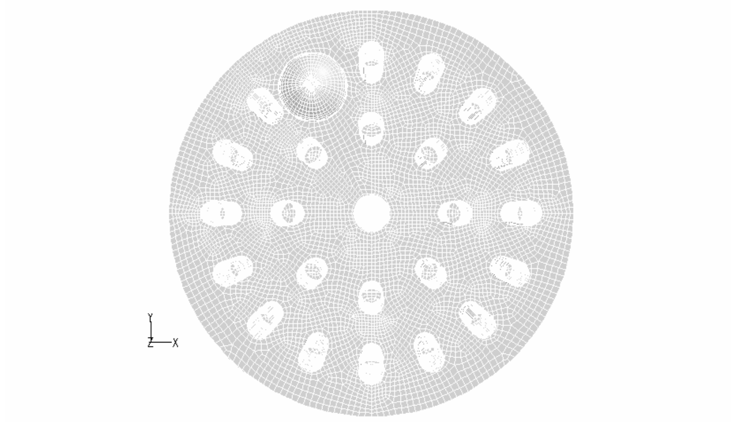


Figure 5. Tank interior computational meshes used for the in-tank modeling

For the baseline in-column case, the column was assumed to be cooled by forced convective cooling through the central cooling tube and the four external cooling tubes with natural convection cooling of the remaining column wall portions. Detailed cases for the in-column evaluations are shown in Table 3. A constant ambient temperature of 35 ^oC was assumed for all modeling cases. For both in-column and in-tank modeling, cesium loading on CST was assumed to be 257 Ci/L (1273 watts/m^3) based on previous analysis which indicated that this was the highest loading for the anticipated feeds.

The performance model was benchmarked against the theoretical results to verify the computational results against the previous work [3]. The verified model was used for the thermal calculations for the in-column and in-tank models. The thermal analysis and evaluation were made by applying two temperature limits to the modeling domain as safety criteria. The safety criteria for the column and tank wall temperatures are assumed to be 130 and 100 ºC, respectively. A temperature limit of 55 ºC for the tank liquid containing the spent CST is also assumed for operational control.

In-Column Thermal Modeling Results

For computational modeling purposes, a conservative approach was taken by assuming that the primary cooling mechanisms inside and outside of the column were conduction and natural convection, respectively. Two-dimensional modeling calculations were conducted with the assumption that axial heat removal (end effects) from the column was negligible relative to radial heat transfer as previously discussed.

For the in-column thermal analysis of the 28-in CST column, 100% cesium loading, one central cooling and four active external tubes, and 35 ºC ambient temperature were considered to be the baseline conditions. Table 6 shows quantitative comparisons of steady-state maximum temperatures for a range of column conditions. The results indicate that when both the internal and external engineered cooling systems are active and the CST bed is filled with stagnant liquid, the maximum temperature will reach about 63 ºC. With inactive central cooling and four active external cooling tubes, the peak temperature is about 114 ºC. On the other hand, when only the central cooling system is active, the peak temperature is about 80 ºC. When both of the engineered cooling systems are lost, the column temperature increases to about 156 ºC. This temperature would not be observed in practice for the wet column until complete supernate volatilization occurs as a result of boiling at a temperature near 130 ^oC. Radial steady-state temperature distributions for these cases are compared in Fig. 6. It is noted that the central cooling tube is particularly effective at cooling the column and results in a decrease in the peak temperature of 76 ºC as compared to the case with no active cooling.

When the CST column loaded with 100% cesium has 5 gpm solution flow without active cooling, the maximum column temperatures increase only by about 1 to 2 ºC across the column length, depending on the column height (Table 6). Therefore, 5 gpm process fluid flow through the column provides adequate heat removal from the column even with no active cooling.

Figure 7 compares transient calculation results for the stagnant, wet CST columns with and without active cooling systems. With both internal and external engineered cooling systems inactive, the maximum column temperature increases to above 75 ^oC within two days after cooling system loss. The steady-state boiling temperature of 130 ºC is reached in about 6 days. The calculation results are meaningless beyond the supernate boiling temperature. With active cooling the maximum column temperature approaches 60 ºC within 48 hours and the maximum steady-state temperature only reaches 63 ºC, as discussed above.

In the case of inadvertent solution leakage from the CST column or bed dryout due to insufficient cooling, there are safety concerns about the rate of transient thermal response of the fully-loaded column and the effectiveness of the cooling system at maintaining the temperature of the dry column. Transient results for the dry column are compared for two different thermal loadings in Fig. 8. With a dry column containing a loading of 257 Ci/liter at an initial temperature of 35 ºC and active engineered cooling, a steady-state maximum temperature of 122 ºC is reached in about 3 days. The results show that when the thermal loading is increased by about 17% (from 257 to 300 Ci/liter), the maximum column temperature increases by about 12%. It is noted that the transient thermal response time of the dry column is much more rapid than that of the wet column (Fig. 7), as expected.

When the dry column has inactive internal and external cooling systems, the temperature reaches 130 ºC in about 24 hours under 300 Ci/liter thermal loading. The peak temperature for the wet column is about 110 ºC lower that that of the dry column. For the case where forced air flow through the dry bed is used to cool the column, the results show that air flow through the bed has a large impact on the maximum bed temperature. The maximum bed temperature is estimated to be less than 100 °C with 80 cubic feet per minute air flow (4 inch/sec velocity) through the column.

Sensitivity analysis was performed for different operating conditions. With active cooling, when the column heat load was increased by about 17% from the 257 Ci/liter baseline value, the maximum column temperature increased by about 14 °C. Increasing the ambient temperature from 35 to 55 °C resulted in small increases in the maximum column temperature of <4 °C. Maximum column temperatures were also estimated for different coolant water temperatures with active engineered cooling. The calculation results show that when the coolant temperature increases by 10 °C, maximum column temperature changes by about 6 °C. A 20 °C increase from the baseline ambient temperature of 35 °C, results in a maximum column temperature increase of only about 8 °C.

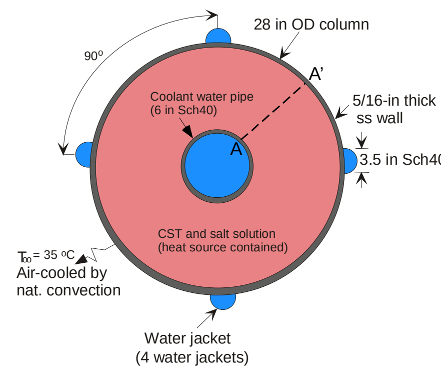
In-Tank Thermal Modeling Results

For the in-tank evaluations, the modeling approach changed as the work progressed based on the results. Initial modeling efforts involved an adiabatic tank floor with no heat transfer into the soil region and unground CST media with a loading of 257 Ci/L. Due to the high temperatures observed for various modeling cases assuming an adiabatic floor, a new modeling domain was developed which included the soil region below the tank. Subsequent analysis revealed that a significant amount of heat transfer occurred through the floor, which impacted the calculated maximum floor temperatures. When the cooling capability is assumed to be uniform over the entire surface area of the 6000-gallon CST mound with an adiabatic bottom surface, heat flux for the flat layer (Case 2) is 55 watts/m^2, which is about 17 times less than that of the hemispherical mound (Case 1). This indicates that the hemispherical mound shape provides a conservative estimate of the maximum temperature.

Table 6. Steady-state maximum column temperatures for various conditions for the wet column

Column Hydraulic conditions	Central cooling system	External cooling system	100% CST loading (Curie/liter)	Max. column temperature (°C)*
5 gpm flow	Inactive	Inactive	257	36.1 for 10 ft high col.
	Inactive	Inactive	257	36.7 for 15 ft high col.
	Inactive	Inactive	257	37.8 for 25 ft high col.
Stagnant	Active	Active	257	62.7
Stagnant	Active	Inactive	257	80.2
Stagnant	Inactive	Active	257	114.1
Stagnant	Inactive	Inactive	257	156.0

* based on 35 °C inlet temperature



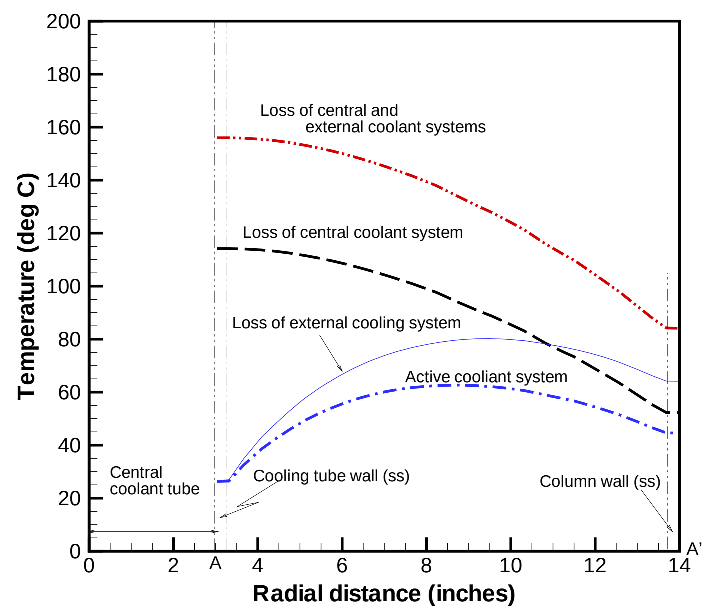


Figure 6. Steady-state column temperature profile along the radial line A-A' for stagnant wet CST media with active and inactive coolant systems (257 Ci/L)

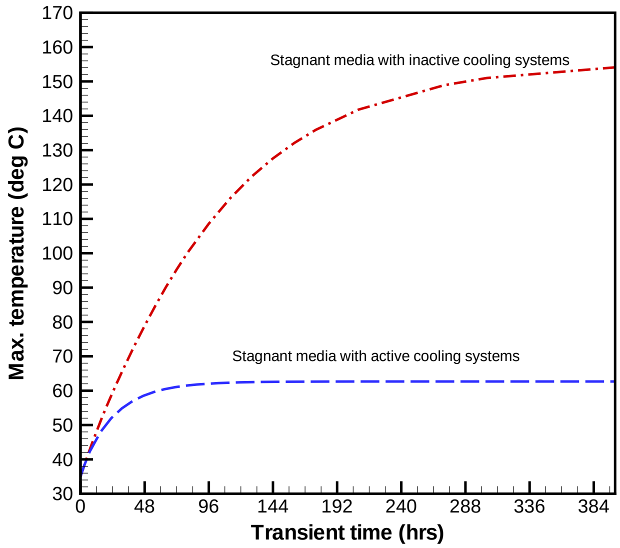


Figure 7. Transient maximum column temperatures for stagnant, wet CST media with active and inactive coolant systems

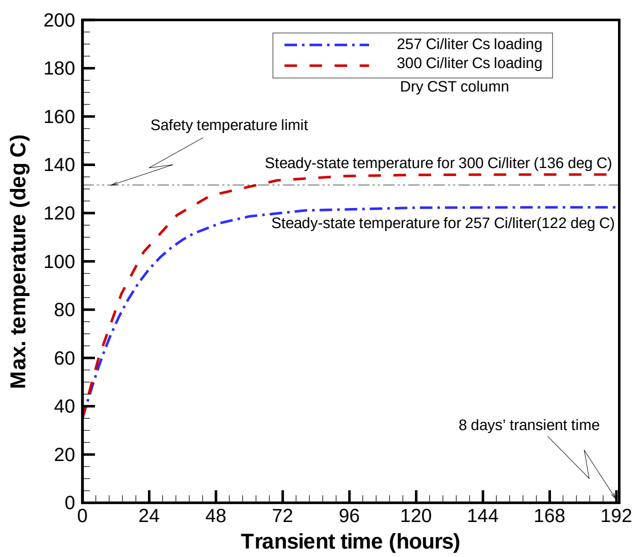


Figure 8. Transient responses of maximum column temperatures to the inadvertent loss of process fluid (dry column case) with active internal and external cooling systems (257 vs. 300 Ci/L)

The adiabatic in-tank modeling efforts assumed no heat transfer across the tank bottom surface as an initial conservative estimate. However, based on the results it was believed that the adiabatic heat transfer assumption for the tank bottom surface resulted in unrealistically conservative estimates of the maximum tank floor temperatures. The 0.5-in thick carbon steel tank bottom wall with multiple lower layers of ceramic and concrete were expected to provide significant heat transfer to the soil region which behaves as in infinite heat sink. Floor heat transfer was also expected to significantly impact the results because this heat transfer mechanism is operative in the exact location of interest as far as the maximum tank temperatures are concerned. Based on previous work [20], a 150-foot deep soil region below the tank bottom was expected to provide sufficient depth and heat transfer volume to reach thermal equilibrium at an assumed soil temperature of 20 ^{o}C. A schematic of the modified calculation domain including the various known material layers and a 150 foot soil region below the tank is shown along with results in Table 7. When heat transfer across the tank floor is allowed, significantly reduced maximum floor temperatures are observed. A quantitative comparison of maximum tank bottom surface temperatures is provided in Table 7 between the cases with and without floor heat transfer for three different cylindrical CST mound heights. With the floor heat transfer mechanism included, the temperature limit of 100 ^{o}C is not exceeded even for a 12 inch high cylindrical mound (900 gallon volume). As discussed above, it is expected that the total volume has little impact on the maximum temperature and the key parameter is the height of the mound. Therefore, larger mound volumes of this same height would also be expected to result in acceptable maximum temperatures. Figure 9 shows a comparison of vertical temperature profiles between the two models with and without heat transfer through the tank bottom for a 12-in high cylindrical mound located on the tank floor. This result graphically demonstrates the dramatic impact of floor heat transfer on the maximum wall temperature. It is also noted that when the heat transfer across the tank bottom is considered, the location of the maximum temperature within the mound changes from near the bottom of the mound to the center of the mound.

CONCLUSION

Models have been developed to simulate the thermal characteristics of Crystalline Silicotitanate (CST) ion exchange media fully loaded with radioactive cesium in a column configuration and distributed within a waste storage tank. This work was conducted to support the Small Column Ion Exchange (SCIX) program which is focused on processing dissolved, high-sodium salt waste for the removal of specific radionuclides (including Cs-137, Sr-90, and actinides) within a High Level Waste (HLW) storage tank at the Savannah River Site. Temperature distributions and maximum temperatures across the column were calculated with a focus on process upset conditions.

The main results are summarized as follows:

- With 5 gpm supernate flow through the column and without active engineered cooling the maximum column temperature should be below 40 ^oC.
- For a CST column filled with stagnant supernate and with active engineered cooling and 35 ^oC ambient external air, the peak temperature for the fully-loaded wet column is about 65 ^oC, which is well below the supernate boiling point. This maximum temperature is marginally acceptable with regard to the chemical and physical stability of the CST media.
- For the air-filled column case with active engineered cooling, the maximum temperature is expected to be below 140 ^oC.
- The column temperature exceeds 100 ^oC within 24 hours for the air-filled column with or without active engineered cooling.
- Active air flow through the dry column at 80 SCFM effectively maintains the maximum column temperature below 100 ^oC.
- The impact of the central cooling tube is very large under wet and dry column conditions since the cooling region is located at the hottest spot in the column.
- In-tank CST modeling results revealed that a hemispherical shape is the worst case mound geometry and leads to the highest tank floor temperatures. In contrast, even large volumes of CST distributed in a flat layer do not result in significant floor heating.

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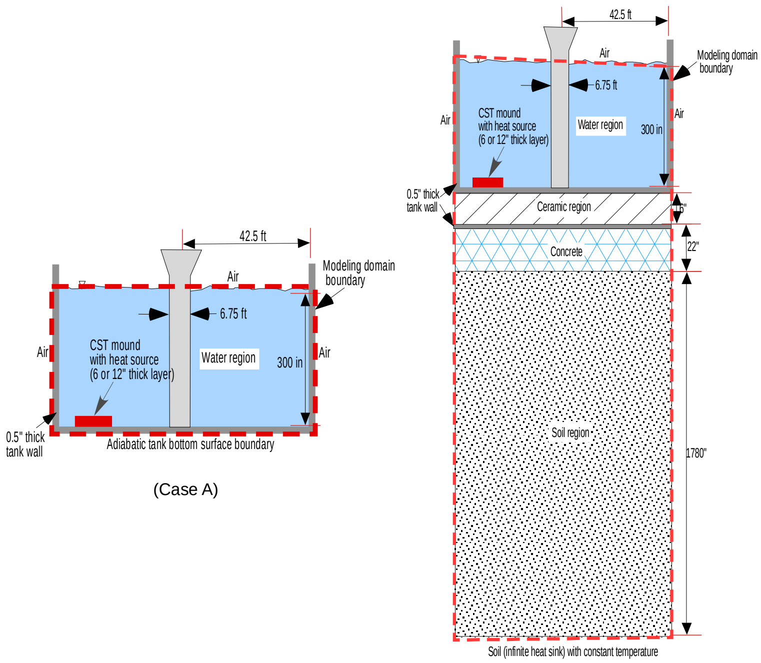
Table 7. Quantitative comparison of maximum tank bottom surface temperatures with and without heat transfer for different cylindrical, unground CST mound heights



(Case A)

(Case B)

Volume of the CST mound located at tank floor	CST loading (257 Ci/liter)*	No heat transfer allowed at tank bottom (Case A: Baseline model)	Heat transfer allowed at tank bottom (Case B)
12-in high cylindrical (900 gallons)	100%	170.3 °C	81.2 °C
9-in high cylindrical (675 gallons)	100%	136.8 °C	68.8 °C
6-in high cylindrical (450 gallons)	100%	81.2 °C	54.8 °C

* based on unground CST particulate

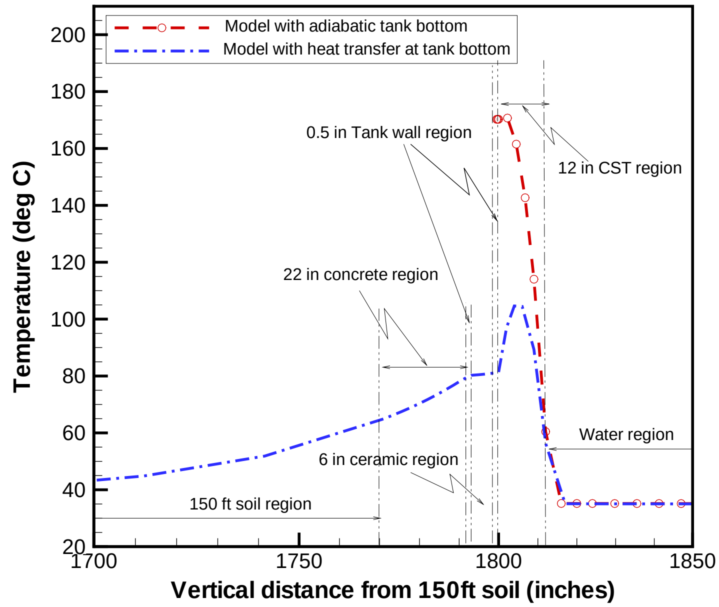


Figure 9. Comparison of temperatures between the models with and without heat transfer through the tank bottom for 12-in cylindrical pancake mound (unground CST)