Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy.

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Thermal Performance Analysis for Small Ion-Exchange Cesium Removal Process

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INTRODUCTION

The In-Riser Ion Exchange program focuses on the development of in-tank systems to decontaminate high level waste (HLW) salt solutions at the Savannah River Site (SRS) and the Hanford Site. Small Column Ion Exchange (SCIX) treatment for cesium removal is a primary in-riser technology for decontamination prior to final waste immobilization in Saltstone. Through this process, radioactive cesium from the salt solution is adsorbed onto the ion exchange media which is packed within a flow-through column. Spherical Resorcinol-Formaldehyde (RF) is being considered as the ion exchange media for the application of this technology at both sites.

A packed column loaded with media containing radioactive cesium generates significant heat from radiolytic decay. Under normal operating conditions, process fluid flow through the column can provide adequate heat removal from the columns. However, in the unexpected event of loss of fluid flow or fluid drainage from the column, the design must be adequate to handle the thermal load to avoid unacceptable temperature excursions. Otherwise, hot spots may develop locally which could degrade the performance of the ion-exchange media or the temperature could rise above column safety limits. Data exists which indicates that performance degradation with regard to cesium removal occurs with RF at 65C. In addition, the waste supernate solution will boil around 130C [1]. As a result, two temperature limits have been assumed for this analysis. An additional upset scenario was considered involving the loss of the supernate solution due to inadvertent fluid drainage through the column boundary. In this case, the column containing the loaded media could be completely dry. This event is expected to result in high temperatures that could damage the column or cause the RF sorbent material to undergo undesired physical changes. One objective of these calculations is to determine the range of temperatures that should be evaluated during testing with the RF media. Although, the safety temperature limit is based on the salt solution boiling point which does not apply in the air-filled case (because there is no liquid), this same limit (130C) is used as a measure for the evaluation of this condition as well.

The primary objective of the present work is to develop models to simulate the thermal performance of the RF column design when the media is fully loaded with radioactive cesium and the central cooling tube is excluded. Previous analysis led to the consideration of this design simplification for RF, since the baseline column design with center cooling was developed assuming that CST media would be used for cesium removal which has a higher volumetric heat load [2]. Temperature distributions and maximum temperatures across the column during SCIX process operations and upset conditions were conducted with a focus on SCIX implementation at Hanford. However, a feed composition and cesium loading were assumed which were known to be considerably higher than would typically be observed at Hanford. In order to evaluate the impact of this potentially highly conservative assumption, fractionally-reduced loading cases were also considered. A computational modeling approach was taken to include conservative, bounding estimates for key parameters so that the results would provide the maximum temperatures achievable under the design configurations.

DESCRIPTION OF THE ACTUAL WORK

The baseline column design uses a 28-inch ID cylinder with four external cooling jackets prepared from 3.5-inch half-pipe tubes symmetrically distributed around the periphery of the column and oriented along the vertical column height of 15 feet. Coolant water at 25C is supplied to each of the outer jackets at 6.25 gpm. For each case the maximum cesium loading was assumed with initial internal column and external air temperatures of 45C.

Equilibrium cesium loadings for each of the RF media were calculated for the projected waste feed streams using the TMIXP code developed at SRNL [3]. Predicted cesium equilibrium loading levels for the most limiting waste type were 133 Ci Cs^{137}/liter packed bed, respectively. This highly concentrated radioactive source will generate a significant amount of heat in the column, which corresponds to about 0.661 watts/gallon of volumetric heat source [4]. Under normal operating conditions, process fluid flow through the column can provide adequate heat removal from the column through a coupled conduction and convection heat transfer mechanism. However, in the case of a loss of flow accident, there are concerns about the transient thermal response rates and the maximum steady-state temperatures reached for fully-loaded columns containing each ion exchange media. Fast thermal response and high peak temperature can lead to unacceptable consequences such as media degradation and solution boiling.

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, and that axial heat removal from the column was negligible relative to radial heat transfer. A two-dimensional transient heat conduction model was developed to assess the thermal performance of the packed column with loss of flow using the prototypic geometry.

The present work considers three basic cases. The first two cases are assumed to have no flow through the bed and involve internal heat transfer by conduction only for columns filled with salt solution. Heat transfer at the walls involves natural convection from the external wall boundary to the ambient air, with the assumption that the air serves as an infinite heat sink at constant temperature. For Case 1, the maximum column diameters will be determined with no active cooling to maintain the maximum temperature within the bed at or below each of the above limits. Case 2 is similar to Case 1 but with active cooling through the external cooling tubes. Transient calculations will also be conducted for an air-filled column (Case 3) to determine the maximum bed temperature as a function of time.

RESULTS

Transient two-dimensional models have been developed to simulate the thermal performance of the RF column baseline design fully loaded with radioactive cesium and to calculate temperature distributions and maximum temperatures across the column during SCIX upset conditions. All calculations assumed that the fluid within the column is stagnant. The baseline models were benchmarked against theoretical results.

In order to evaluate the impact of this potentially highly conservative assumption, fractionally-reduced loading cases were also considered. Steady state and transient modeling calculations were performed for three different conditions involving packed beds of RF resin which were saturated in cesium and immersed in waste supernate, as well as the case where the same, saturated bed had been inadvertently drained of liquid. In all cases, convection effects resulting from thermally induced motions of the fluid were assumed to be negligible to provide a conservative estimate of the maximum column temperatures. The column for Case 1 was cooled only by natural convection at the surface of the column, and the wet and drained columns for Cases 2 and 3 were cooled by both forced convective cooling through four external cooling tubes and natural convection at the remaining column wall sections. A "safety temperature limit" of 130C was assumed based on the calculated salt solution boiling point. An "operational temperature limit" of 65C was assumed based on the perceived temperature limit for the ion exchange media.

The main results are summarized as follows:

- Without any engineered cooling systems (Case 1) and assuming columns suspended in unventilated ambient air at 45C, the maximum diameter of the fully-loaded column expected to maintain the temperature below the assumed media and safety limits is 12 inches. In this case the maximum column diameter required to satisfy only the safety limit is 34 inches.
- For an RF column under Case 2 conditions with active cooling through four outer tubes and 45C ambient external air, the maximum column diameter expected to maintain the temperature below the assumed media and safety limits is 26 inches.
- Modeling analysis was conducted to predict the maximum column temperatures for the previously unevaluated accident scenario involving inadvertent drainage of a cesium-saturated column (Case 3). As expected, much higher maximum temperatures were observed in this case due to the poor heat transfer properties of air, compared to those of liquid. The results indicate that the maximum temperature within a 28 inch diameter RF column exceeds 250C within 2 days, while the maximum temperature of a 12 inch column never reaches 100C.
- The maximum column temperature decreases with higher porosity since a larger volume fraction of the column is air and because there is a smaller amount of cesium present within the column. Varying the porosity from the minimum value to the maximum theoretical value only resulted in a 14% decrease in the maximum column temperature after 96 hours with a 28 inch column under Case 3 conditions (air-filled, water cooled through 4 external tubes). In all cases the operational limit was reached within 3 hours and the safety limit was reached within 12 hours. The results indicate that the maximum column temperature is not highly impacted by porosity uncertainties.
- When cesium loading levels for the 28-in RF column under dry bed conditions are reduced from 100% to 50%, the time required to reach the safety and

operational temperature limits is approximately doubled.

- The impact of the active external cooling tubes for the air filled column is small because the active cooling area ratio of the four external tubes to external column wall surface is only about 18% for 28-in column, and the air medium of the drained column has low thermal conductivity. Under Case 3 conditions with 50% cesium saturation, it was shown that after 120 hours, the maximum column temperature predicted without active cooling (211C) was only 9% higher than the temperature reached with active cooling (192C). The results indicate that the external engineered cooling system is not highly effective at reducing the maximum column temperature for the air-filled column.

- When a 6 inch diameter cooling tube was inserted at the center of a 28-in air-filled column, the presence of the central cooling tube dramatically decreased the maximum predicted column temperature (from 331C to 134C). The shape of the temperature distribution also changed and the time required to reach the column safety limit increased from 12 to 35 hours.

- The modeling results demonstrate that although the lower cesium loading of spherical RF resin immersed in liquid seems to allow for the removal of the central cooling tube, the effectiveness of the central tube at cooling an air-filled column may prohibit this proposed design simplification.

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