

SMALL-COLUMN ION-EXCHANGE ALTERNATIVE TO REMOVE ^{137}Cs FROM LOW-CURIE SALT WASTE: SUMMARY OF PHASE 1

May 2004

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Nuclear Science & Technology Division

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LIST OF ACRONYMS AND ABBREVIATED TERMS

ANISN	Anisotropic S_n , code for shielding calculations (one dimensional)
ARP	Actinide Removal Process
CFD	computational fluid dynamics (code)
Ci	curie
CSSX	Caustic-Side Solvent Extraction
CST	crystalline silicotitanate
DABL69	photon library for shielding calculations
DOE	U.S. Department of Energy
DORT	Discrete Ordinates Radiation Transport, code for shielding calculations (two dimensional)
DWPF	Defense Waste Processing Facility
HLW	high-level waste
kVA	kilovolt-amps
M	molar, gmoles/L
LCS	low-curie salt
LLW	low-level waste
mrem	millirem or effective dose as thousandths of a rem
MST	monosodium titanate
MVST	Melton Valley Storage Tank
NETL	National Energy Technology Laboratory
OLI	Optimal Logic, Inc.
ORNL	Oak Ridge National Laboratory
R&D	research and development
SCIX	Small-Column Ion-Exchange
SRS	Savannah River Site
SRTC	Savannah River Technology Center
SWPF	Salt Waste Processing Facility
TRU	transuranic
WAC	waste acceptance criteria
WVDP	West Valley Demonstration Project
VERSE-LC	VERsatile Reaction SEparation for Liquid Chromatography (model)
ZAM	Zheng, Anthony, and Miller (model)

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EXECUTIVE SUMMARY

A Small-Column Ion-Exchange (SCIX) system is being evaluated for removing cesium from the Type 2 and/or Type 3 dissolved saltcake wastes at the Savannah River Site (SRS) to ensure that the dissolved saltcake meets the waste acceptance criteria at the Saltstone Facility. Both crystalline silicotitanate (CST) and IONSIV™ IE-96 zeolite were evaluated as the ion-exchange media. The accelerated alternative, using CST in the SCIX, could save as much as \$3 billion in operating and storage costs and ~20 years in processing time compared to the current baseline. With its proven high cesium-loading capacity for the expected dissolved saltcake compositions and temperatures, CST is the preferred sorbent for SCIX. The low-cost alternative sorbent, zeolite, greatly increases the volume of sorbent required because of its much lower cesium-loading capacity. Thus, zeolite greatly increases the cost for the alternative, mainly because of the increased number of Defense Waste Processing Facility canisters required to dispose of the loaded sorbent (potentially over 7000 for zeolite, compared with <500 for CST). The models previously developed for predicting cesium loading on CST compared favorably with laboratory measurements of equilibrium distribution ratios and column loading performance using dissolved saltcake simulants. These models predict that a column of 432 gal of CST can operate at 25 gal/min and treat 100,000 to 900,000 gal of dissolved saltcake, depending on the solution composition. An average value of 300,000 gal per column was used for the cost benefit analysis. Predicted cesium loading on the CST is normally below 300 Ci/L; however, solutions with low salt concentrations could potentially load the CST to 630 Ci/L. Heat transfer calculations predict nonboiling temperatures for the small columns with loadings <100 Ci/L with only natural convection cooling. For the loadings up to the maximum calculated for the tank farm (630 Ci/L), a water cooling system is required to ensure that no boiling occurs in the column if the process flow is stopped. Dose rate calculations indicate that the maximum dose rate above the tank riser is expected to be $\sim 10^{-2}$ mrem/h for a column loaded at 300 Ci/L in the riser. The risk analysis indicates a net beneficial impact with no major problems likely to prevent implementation or completion of saltcake treatment.

1. BACKGROUND

1.1 BASELINE APPROACH

High-level waste (HLW) at the SRS is stored in 49 carbon steel tanks. These tanks contain ~37 million gal of waste with a radioactive content of ~426 million curies. The HLW consists of 3 million gal of sludge, containing ~226 million curies, and 34 million gal of a salt component, containing ~200 million curies. Although the sludge waste contains less than 10% of the total volume of HLW, it contains ~53% of the total curies. The sludge generally contains insoluble radioactive elements including strontium, plutonium, americium, and curium in the form of metal hydroxides. Salt waste, the soluble portion of the HLW, contains most of the soluble radioactive cesium, with ~98% of the curies in the salt component being ¹³⁷Cs. Dissolution of the salt component with water will raise the total volume of HLW to be treated to ~83 million gal (of which ~80 million gal is supernate plus dissolved saltcake). The baseline plan calls for treatment of this total volume of waste in the Salt Waste Processing Facility (SWPF). The SWPF will utilize monosodium titanate (MST) sorption/cross-flow filtration for removal of strontium and actinides followed by Caustic-Side Solvent Extraction (CSSX) for removal of the cesium. The HLW from the SWPF would be vitrified in the Defense Waste Processing Facility (DWPF) and the treated liquid low-level waste (LLW) would be sent to the Saltstone Facility. The estimated life-cycle cost for the SWPF is \$2.2 billion and the unit cost for processing saltcake through the SWPF and the Saltstone Facility is \$104.06/gal of saltcake. [1]

1.2 ACCELERATED BASELINE APPROACH

Operational costs for storing saltcakes and supernates in the SRS tank farm are approximately \$100 million per year. In addition, an engineering study provided by SRS personnel indicates that the cost to process a gallon of saltcake through the SWPF into saltstone is significantly higher than the cost for disposal of saltcake directly into saltstone (\$104.06 vs \$17.48 per gal of saltcake). [1] As a result, SRS has formulated an accelerated baseline approach designed to reach closure of the tank farm more quickly than the baseline approach (the SWPF currently planned may have about 50% of the capacity originally used in the baseline) and to reduce the amount of material processed through the full-scale DWPF. Figure 1 compares the baseline approach with the accelerated baseline approach, as presented in the Request for Proposals. [2]

The salt waste portion of the HLW (~80 million gal of supernate plus dissolved saltcake) would be separated into four streams:

1. High-Curie Salt Supernate (Type 1 Waste): This stream is expected to contain approximately one-third of the total dissolved salt waste volume (~27 million gal). This waste stream would be treated in the SWPF currently planned, which is scheduled to be operational in 2010. The HLW stream from the SWPF would be vitrified in the DWPF, and the LLW would be grouted in the Saltstone Facility.
2. Low-Curie Salt (LCS) Supernate Not Requiring Radionuclide Removal (Type 2 Waste): This stream is expected to contain approximately one-third of the total dissolved salt waste volume (~27 million gal). This waste stream would be sent directly to the Saltstone Facility for grouting.
3. LCS Supernate Requiring Strontium or Actinide Removal (Type 3 Waste): This stream is expected to contain approximately one-third of the total dissolved salt waste volume (~27 million gal). This waste stream would be processed for removal of strontium and actinides by a large demonstration-scale process (>10% of the scale required for strontium and actinide removal in the SWPF), which is being constructed in existing facilities

(Building 512-S) at SRS. The HLW from the demonstration scale strontium and actinide process would be vitrified in the DWPF, and the LLW would be grouted in the Saltstone Facility.

4. Unique Wastes (Type 4 Waste): This would include wastes for which no technology is currently available for treatment. A primary example is Tank 48, which contains legacy tetraphenylborate organic wastes.

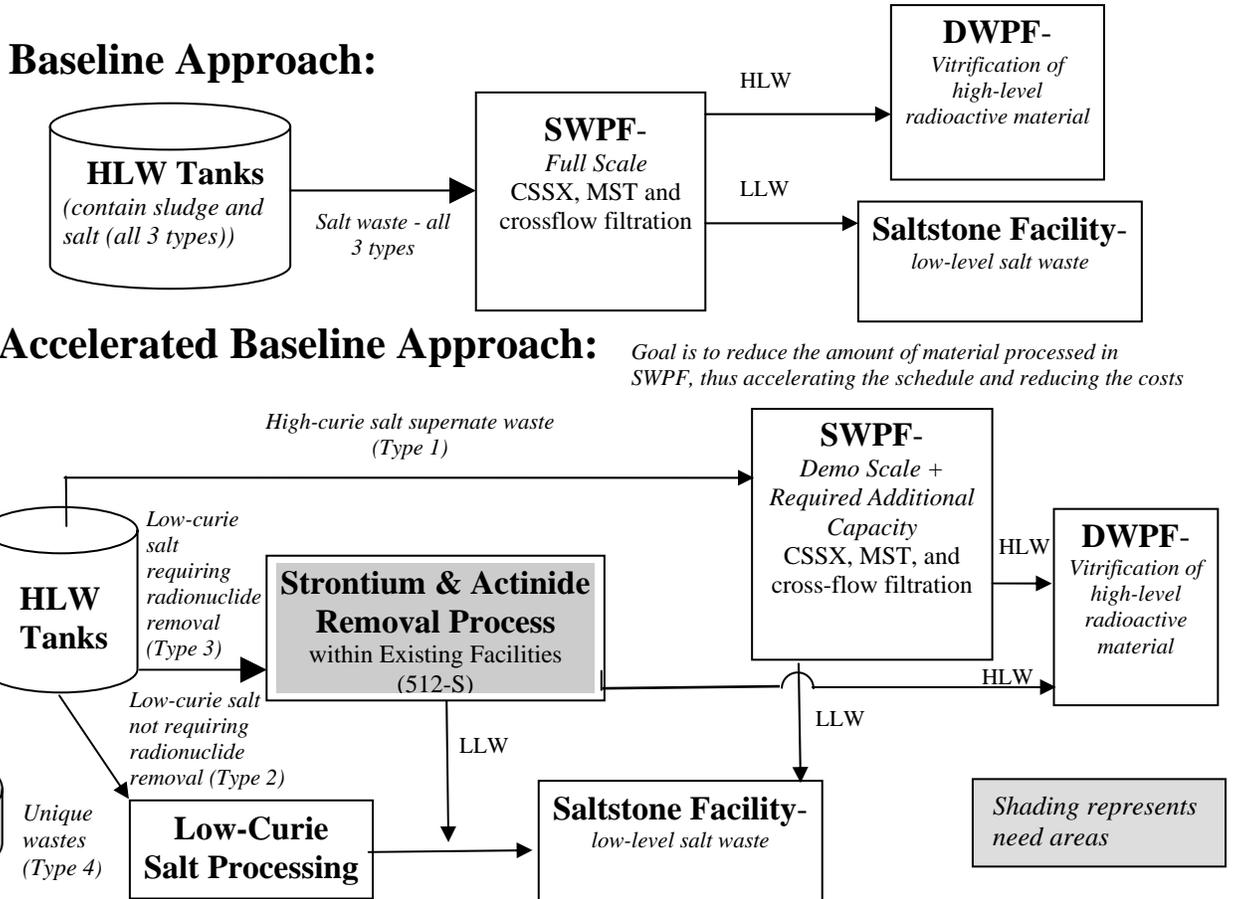


Figure 1. Generic flow path for the SRS HLW salt alternatives project as presented in the Request for Proposals (Ref. 2).

To maximize cost savings in the accelerated baseline approach it is necessary to:

1. utilize improved treatment approaches to maximize the volume of salt waste that can be sent to the Saltstone Facility so the waste does not have to be processed by the SWPF,
2. initiate treatment as soon as possible in order to accelerate closure of the tank farm, and
3. utilize existing SRS facilities in any improved treatment approaches.

1.3 CESIUM REMOVAL FROM LCS

The primary assumption that makes the accelerated baseline approach to waste segregation possible is that two-thirds of the waste, ~54 million gal of dissolved saltcake, will not require

cesium removal. Salt waste, the soluble portion of the HLW, contains most of the soluble radioactive ^{137}Cs . Salt crystallizes out of the salt wastes and settles, as saltcake, to the bottom of a waste tank. The concentrated supernate is present as a free liquid layer on top of the saltcake and in interstitial spaces within the saltcake. This concentrated supernate (~27 million gal of Type 1 Waste) contains most of the cesium, and the accelerated baseline approach calls for this waste to be treated in the SWPF. The accelerated baseline approach assumes that the total interstitial space within the saltcake is 22% [1] and that the interstitial space can be drained so that no more than 30% liquid supernate remains. [3] Laboratory tests on simulated saltcake have shown that such drainage is possible [4] and hydraulic modeling indicates the same may be possible at full scale. [5]

Waste acceptance criteria (WAC) for the Saltstone Facility currently require that the cesium and alpha components in the grout be kept below 45 nCi/g (0.0002 Ci/gal) and 20 nCi/g, respectively. The Saltstone Facility has shielding to handle solutions containing up to 0.05 Ci/gal, and a new WAC limit of 0.1 Ci/gal has been proposed, which would require additional shielding. If the interstitial concentrated supernate cannot be drained to required levels, the quantity of waste that could be processed by the accelerated baseline approach could be greatly reduced because the saltstone product would not meet the required cesium levels. Dissolution of the saltcake in Tank 41H produced a solution containing 0.377 Ci/gal of ^{137}Cs . [6] Therefore, the assumption that two-thirds of the dissolved salt waste can be sent to the Saltstone Facility without processing in the SWPF may not be valid and the current accelerated alternative approach has no provisions for removing cesium from the dissolved saltcake. If the cesium levels in the dissolved saltcake are too high for processing in the Saltstone Facility, the waste would have to be reevaporated for storage as saltcake or sent to the SWPF for cesium removal.

The proposed Small-Column Ion-Exchange (SCIX) system addresses this risk by providing a cesium-removal polishing operation utilizing an ion-exchange system with a cesium sorbent for Type 2 and/or Type 3 wastes to ensure that the dissolved saltcake does not exceed the proposed new WAC of 0.1 Ci/gal for the Saltstone Facility. (The more stringent WAC of 0.0002 Ci/gal can also be met using different processing conditions than those targeting 0.1 Ci/gal.) A schematic diagram of the proposed improved treatment approach is presented in Figure 2. Figure 3 illustrates the operational decision making that can be utilized to minimize saltcake processing while ensuring that the WAC for producing saltstone is met.

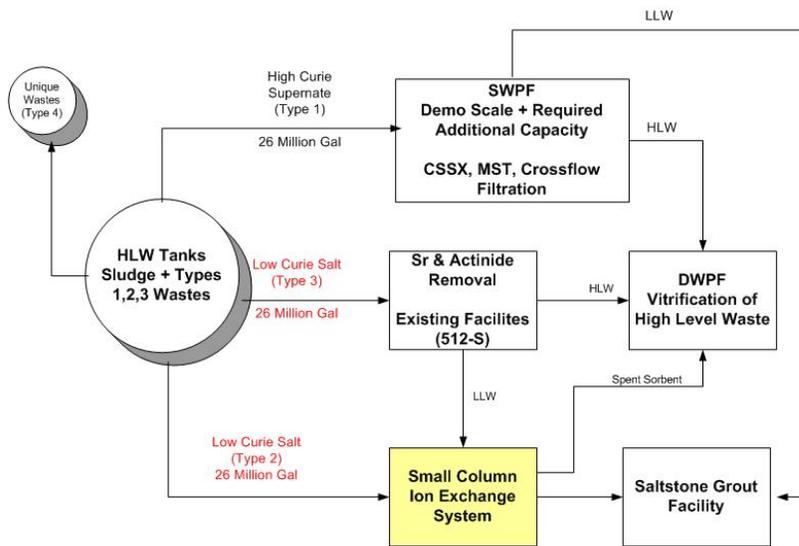


Figure 2. Schematic diagram using the SCIX process in the accelerated baseline approach.

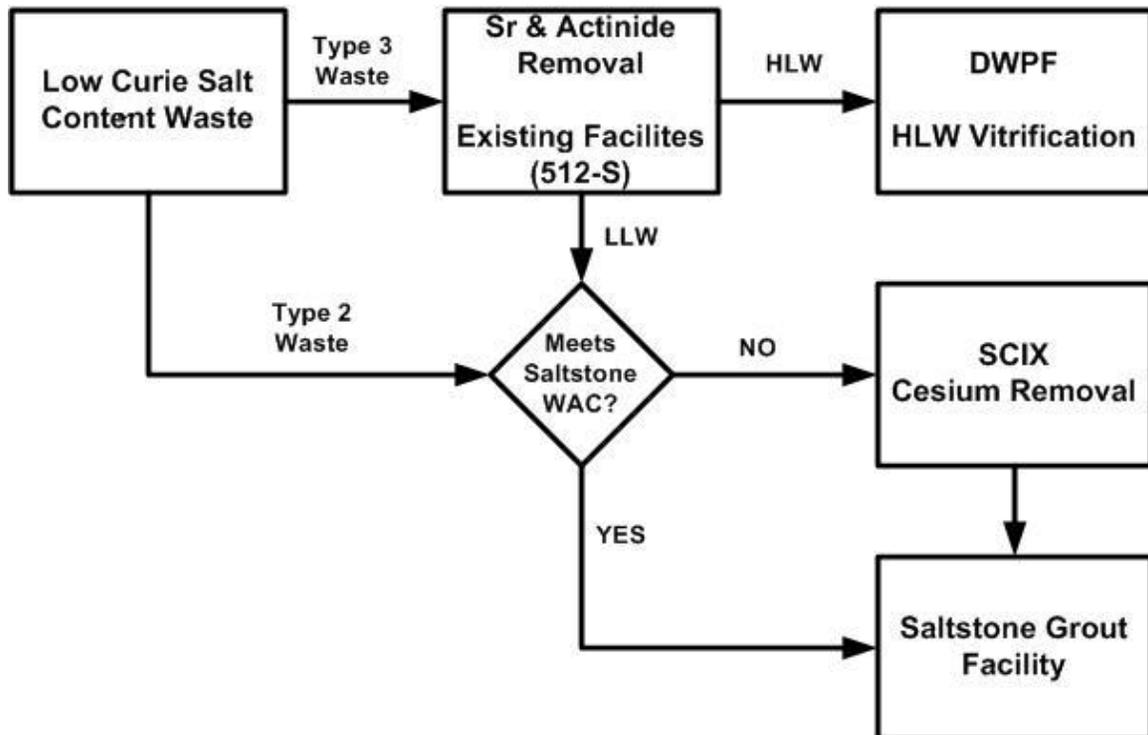


Figure 3. Schematic diagram illustrating the decision point about whether cesium-removal polishing is required to meet the Saltstone WAC.

1.4 SCIX SYSTEM

The proposed technology deploys a small ion-exchange system in existing facilities at SRS to remove cesium from LCS waste. Figure 4 presents schematically the deployment of small columns of ion-exchange particles in the risers of an HLW tank at SRS, the conceptual flow paths for removing cesium from the LCS solution, and the removal and disposition of the loaded sorbent. The following sections summarize the results for Phase 1 of a project funded by the National Energy Technology Laboratory (NETL) to evaluate the SCIX system, including sorbent testing, model predictions, heat transfer calculations, facility evaluation, disposition selection, cost benefits analysis, and risk analysis.

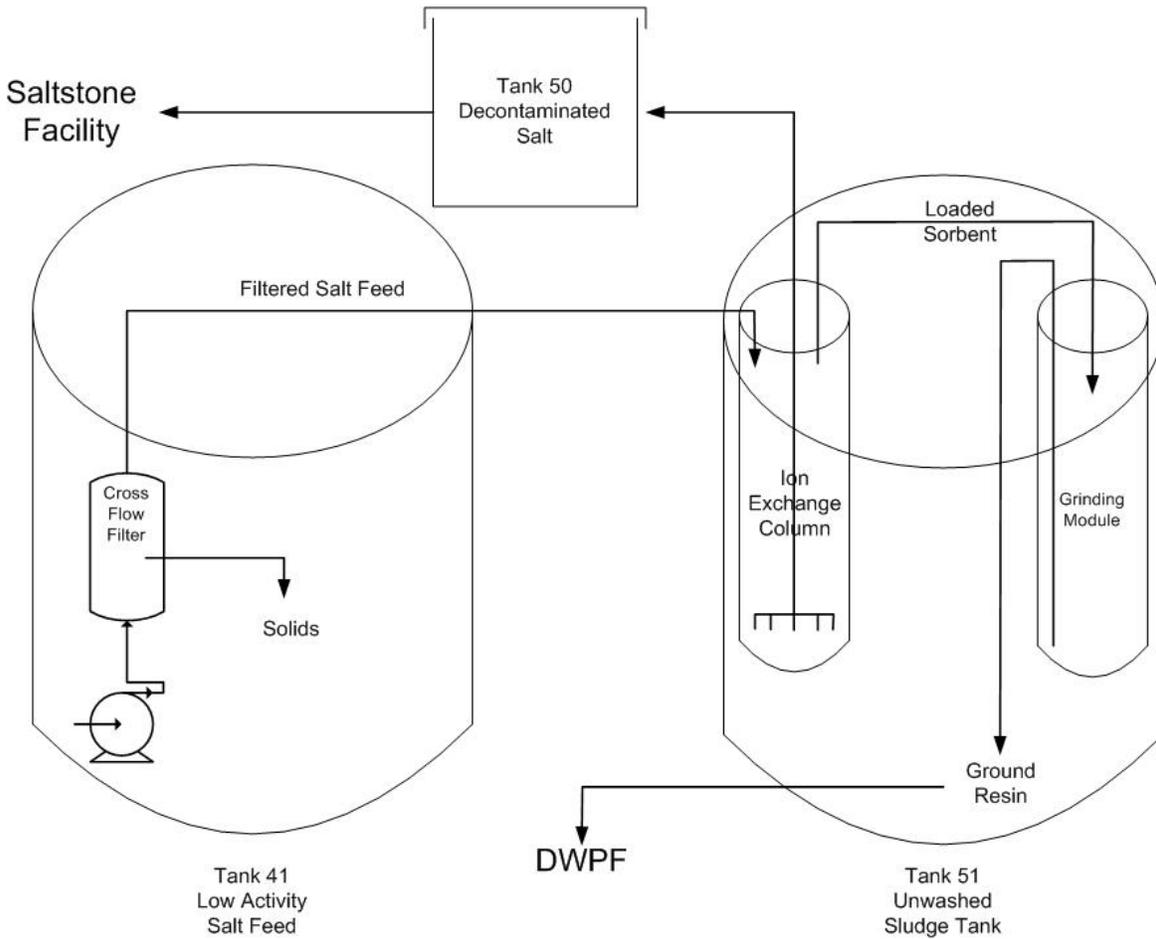


Figure 4. Schematic drawing of the small columns positioned in Tank 51 and the flow paths for SCIX.

1.5 EXPECTED BENEFITS AND PERFORMANCE

The SRS plan to segregate the waste and send only 33 vol % of the salt waste through the SWPF drastically cuts both the schedule and the costs for treating the waste from the HLW tanks. The primary problem with the plan to segregate the waste is that 70% of the interstitial supernate must be drained from the saltcake for this waste stream to meet the proposed WAC for cesium (0.1 Ci/gal) at the Saltstone Facility. Success of this approach is not assured unless it can be guaranteed that the cesium WAC for the Saltstone Facility can be met. Therefore, the SCIX is needed to remove the cesium from the LCS waste to meet this requirement.

The primary benefits of SCIX are as follows:

1. It can be deployed in the SRS tank farms in existing facilities or with minimal construction.
2. It is a proven technology that has been used on similar waste in baseline operations at Oak Ridge National Laboratory (ORNL) and at the West Valley Demonstration Project (WVDP).
3. Much of the research and development (R&D) necessary to apply this technology to the LCS waste at SRS has already been conducted.
4. The system can ensure that the cesium WAC (0.1 Ci/gal for the results presented, 0.0002 Ci/gal can be accommodated using different processing conditions) for the Saltstone Facility is met, capturing the benefits of the accelerated baseline for the maximum quantity of LCS waste.

2. SORBENT SELECTION AND TESTING

The selection of crystalline silicotitanate (CST) as a candidate sorbent is based on the following primary factors.

1. The high cesium loadings have been proven both in equilibrium distribution ratio measurements and in column testing.
2. A considerable amount of R&D has been completed with CST because it was one of the competing technologies for the full-scale SWPF. This R&D indicated that the CST would be very effective in removing cesium for this application. The R&D also indicated that CST would have minimal impact on the other facilities in the HLW system (i.e., DWPF and the Saltstone Facility).
3. A similar system utilizing CST was successfully developed, demonstrated, and deployed for removal of cesium from supernate at ORNL's Melton Valley Storage Tank (MVST) Facility.

Zeolite was used in the WVDP columns and is considered a widely available, low-cost alternative sorbent. For this reason, some experimental work was done on zeolite, and in the cost benefits analysis, zeolite was considered as a substitute for CST. The lower purchase price per pound for zeolite was offset to a large extent by the much greater quantity required to treat the LCS solution. However, the greatest impact on cost resulted from dispositioning all the loaded sorbent to the DWPF and the high cost of HLW vitrification, as is seen in Sect. 6.

2.1 MODEL CALCULATIONS

Modeling results indicate that a 432-gal column of CST can process 100,000 to 900,000 gal of waste at 25 gal/min before the column packing must be changed.[8] Average cesium loadings on the columns range from 50 to 630 Ci/L.

CST has received considerable attention for nuclear waste applications because it exhibits very high selectivity for cesium ions and also shows specificity for strontium. The engineered form of CST, known as IONSIV™ IE-911, has undergone a myriad of tests across the U.S. Department of Energy (DOE) complex because it was considered for the baseline SWPF at SRS. CST is unusual in that it exhibits the high selectivity for cesium ions in highly concentrated salt solutions and over a wide pH range. [9] Tests with IONSIV IE-911 on both simulants and with actual samples from the SRS HLW tanks clearly indicate that it can be very effective in the removal of cesium from SRS supernate. [10, 11, 12] Utilizing data from these tests has allowed development of models that are effective for the prediction of process performance for various SRS HLW compositions. [13] These models were used to size the ion-exchange columns, to set process parameters, and to predict process performance under different bounding conditions. These model calculations are summarized below. Tests at ORNL also investigated the effects of radiolytic gas formation on the loading of cesium and concluded that even in radiation fields 16 times that expected in the full-scale SWPF CST columns, no operational problems stemming from interparticle or intraparticle gas should be expected. [14]

The Zheng, Anthony, and Miller (ZAM) model predicted cesium equilibrium sorption isotherms for various waste compositions in contact with CST sorbent. The Versatile Reaction SEparation for Liquid Chromatography (VERSE-LC) uses the ZAM isotherms and predicts the cesium breakthrough curve for the ion-exchange column. The results of the VERSE-LC model show the volume of waste that can be processed by a given column design (i.e., column diameter and height) and operating parameters (e.g., flow rate, temperature, feed composition). In addition, the

modeling results provide the concentration of cesium on the sorbent, which is important in heat transfer calculations and in the safety analysis.

The ZAM code performs a simulated batch contact test for a given cesium concentration initially in solution to produce the final solution concentration in equilibrium with the cesium loaded on the CST. To construct an isotherm curve of loading versus cesium concentration, the code must be run for a series of starting cesium concentrations covering the concentration range expected for each situation. For each isotherm curve, the ZAM model calculated the equilibrium partitioning between solution and CST for 14 initial Cs⁺ concentrations. All calculations were performed at 30°C with a measured liquid solution density or a liquid density derived from an Optimal Logic, Inc., (OLI) Stream Analyzer computation.

The ion-exchange model VERSE-LC was used to predict the performance of columns loaded with IONSIV IE-911. The kinetic model includes axial dispersion, film diffusion, and pore diffusion within the IE-911 particles. Given column and operating parameters, the VERSE-LC code provides the cesium concentration in the column effluent as a function of the volume of waste processed (breakthrough curve).

Five waste compositions were investigated during the modeling effort to capture the variation in column performance expected from changes in the composition of the waste feed listed in Table 1. The LCS “Average” waste reflects the average composition of the eight LCS tanks (Tanks 9H, 10H, 29H, 31H, 36H, 37H, 38H, and 41H). The “Early,” “Middle,” and “Late” wastes are variations designed to reflect the expected composition trends during salt dissolution in a single saltcake tank. The Tank 41H composition reflects the actual composition of a batch recently dissolved in one of the LCS tanks.

Table 1. Waste compositions used in column performance modeling

Component	Concentration (M)				
	Early	Middle	Late	Average	Tank 41H
<u>Cations</u>					
Na ⁺	7.0	7.0	4.2	6.0	7.9
K ⁺	0.0070	0.0070	0.0042	0.006	0.0079
Cs ⁺	0.00020	0.00020	0.00020	0.000040	0.00038
<u>Anions</u>					
OH ⁻	1.90	1.00	0.30	1.60	0.85
NO ₃ ⁻	2.60	4.00	0.50	2.30	4.9
NO ₂ ⁻	0.90	0.10	0.020	0.71	0.24
AlO ₂ ⁻	1.20	0.40	0.010	0.35	0.45
CO ₃ ⁻²	0.13	0.45	1.08	0.12	0.45
SO ₄ ⁻²	0.050	0.30	0.55	0.16	0.23
PO ₄ ⁻³	0.0070	0.0020	0.010	0.13	0.035
Cl ⁻	0.035	0.035	0.035	0.033	0.001
F ⁻	0.033	0.033	0.033	0.031	0.001

Previous testing of saltcake dissolution showed the following trends.

1. The total sodium-ion concentration will decrease because the highly soluble compounds will be selectively dissolved in the early batches, leaving less soluble components in the later batches.
2. Sodium hydroxide and nitrite will be enriched in the early batches.
3. Maximum nitrate concentration will occur following the peak in hydroxide and nitrite.
4. Aluminate will decrease as the hydroxide decreases.
5. Sulfate and carbonate will increase as the nitrate and hydroxide decrease.

Figure 5 shows the loading isotherms for the five waste compositions. Four of the five load similarly. The low sodium ion concentration in the Late composition (4.2 *M*) allows much higher loading of cesium. Similarly, the high sodium-ion concentration of the Tank 41H waste (7.9 *M*) reduces the potential loading. Results for IE-910 (CST powder) in the LCS Middle simulant are shown for comparison. The ZAM model directly calculates the performance of the IE-910 powder, and the binder used to form the IE-911 granules is expected to lower loadings for IE-911 due to dilution of the CST in the engineered form. A dilution factor of 68% was used to calculate the IE-911 isotherms and in the VERSE-LC modeling to conservatively estimate the volume of salt solution that can be processed.

Figure 6 shows the cesium breakthrough curves for the five waste compositions. Assuming a process objective of reducing the cesium to below 0.1 Ci/gal, this graph shows that a 432-gal column of CST can process approximately 200,000 gal of the Tank 41H, LCS Early, or LCS Middle compositions but close to 1 million gal of the LCS Late or LCS Average waste. The column can process much higher volumes of LCS Average feed before breakthrough because of the lower cesium concentration in this feed, compared with that in the Early, Middle, and Tank 41 feeds. In preparing an average flow sheet and material balance, we have assumed the process will average 300,000 gal of waste before the column packing must be changed. The cesium concentration on the CST varies through the length of the column. The loading at the head of the column will be higher than the loading at the exit (see Figure 7). All of the average loadings calculated fell below 300 Ci/L except for the LCS Late case (627 Ci/L).

2.2 SORBENT TESTING

A small-column treatment system, using either CST or IE-96 zeolite sorbent, was evaluated for treating low activity dissolved saltcake from the HLW tanks at the SRS. Previously, ORNL had shown that CST could form clumps after long-term storage in the high-hydroxide, high-aluminum supernate from the SRS waste tanks. Nucleating precipitation in the ion-exchange bed can result in clumping of the CST particles. UOP Molecular Sieves LLC improved the formulation of the IONSIV™ IE-911, and, with the improved sorbent, clumping does not take place until after ~60 days of operation. [7] No clumping was observed during exposure of CST to simulated waste solutions for the expected operation time of a given column (approximately 1–2 weeks) for work conducted during this project.

The proposed small-column treatment system would have processing times of less than 2 weeks between column change-outs. Column (see Figure 8) and batch tests were conducted for over a month to examine potential clumping problems in dissolved saltcake simulants that bound the expected compositions of the actual waste streams. Figure 9 shows experimental results for cesium breakthrough using the Middle simulant and predicted breakthrough results using the VERSE-LC model for the Middle and Early simulant compositions at the same conditions. In batch tests, no clumping was detected during the first two weeks. After 3 weeks, both CST and zeolite

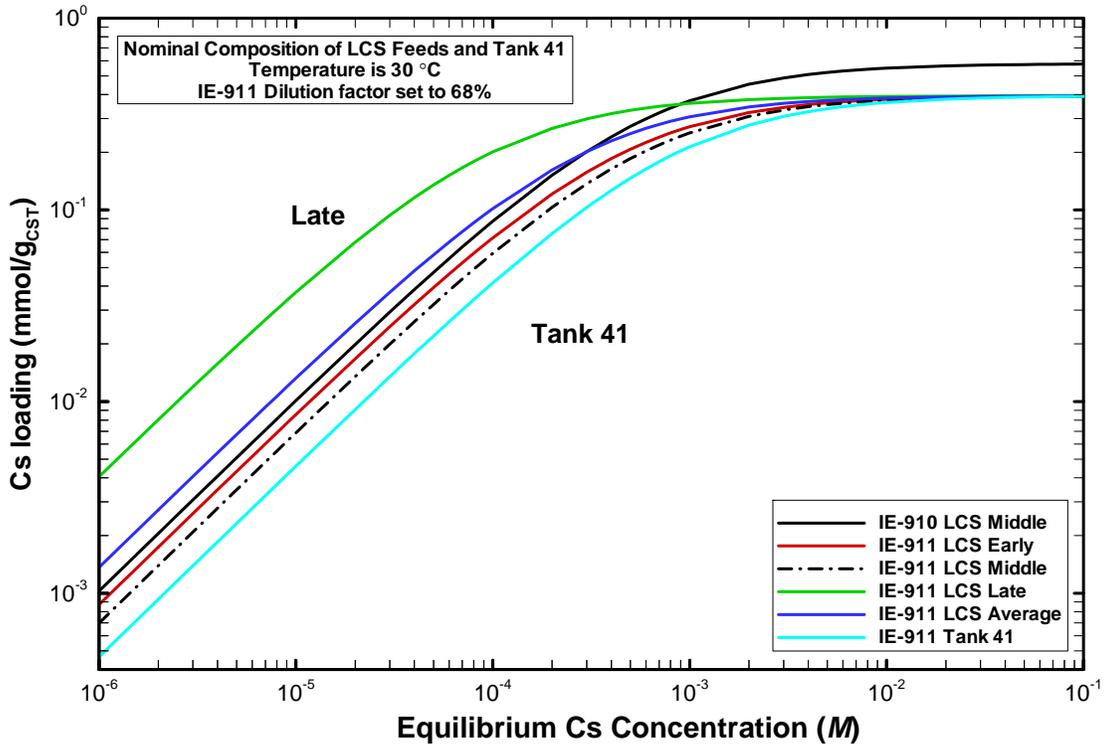


Figure 5. Cesium isotherms for LCS feeds and Tank 41 at 30°C.

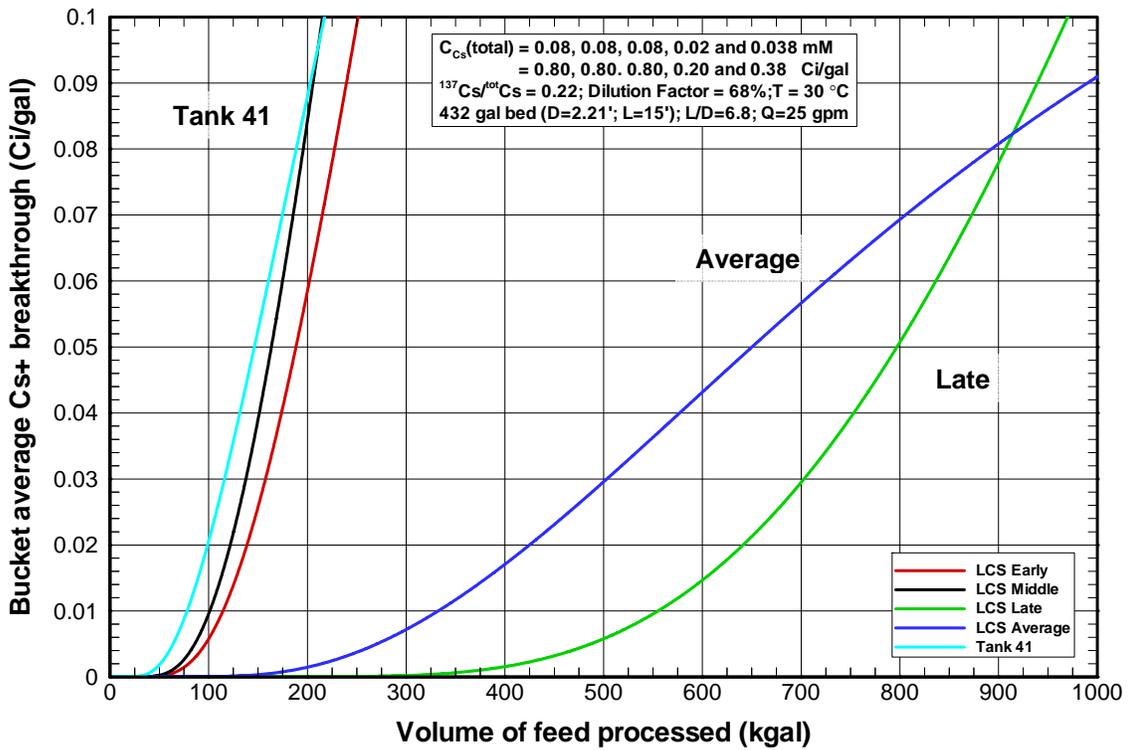


Figure 6. Bucket average cesium breakthrough of LCS feeds and Tank 41.

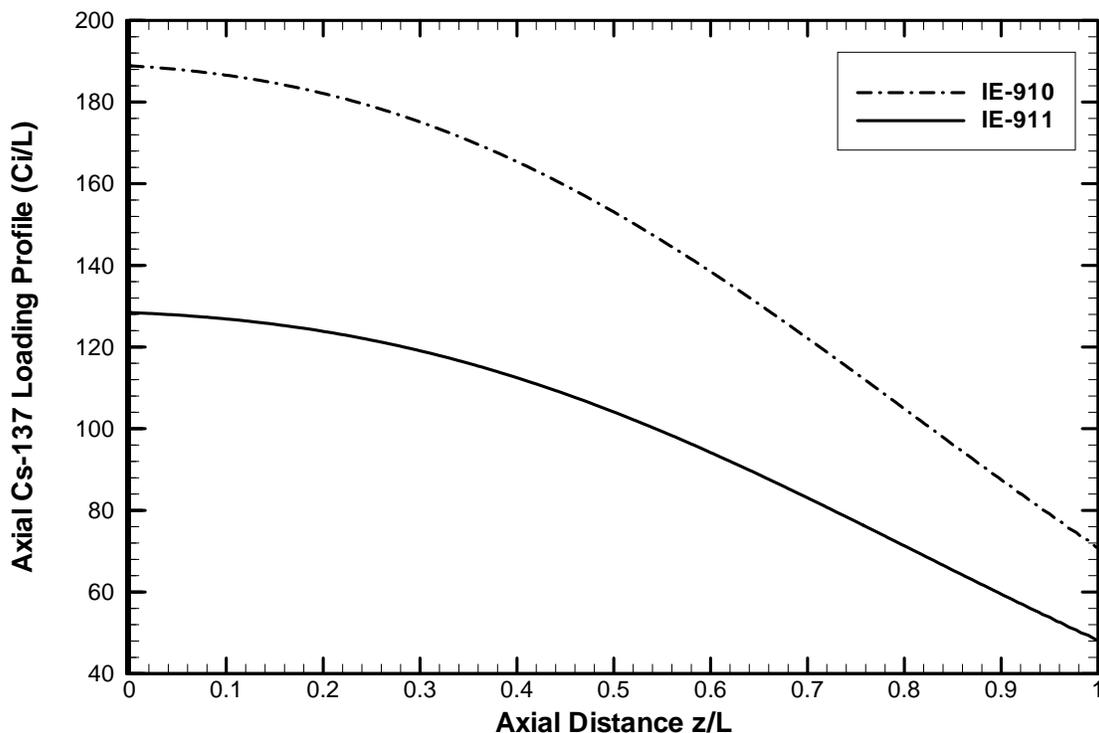


Figure 7. Axial ¹³⁷Cs CST loading profile for LCS Middle tank feed.

showed very weak clumps in Average simulant at 35°C. After 4 weeks, the zeolite in the Middle simulant at 35°C also showed very weak clumps. After 7 weeks, zeolite showed weak clumps in Early, Middle, and Average simulants and CST showed weak clumps in Average simulant at 35°C. No clumps were detected in the other simulants at 35°C or in any of the simulants at 23°C. The clumps that were formed were very weak and would not be expected to interfere with sluicing the sorbent from a column. Column tests using CST and two simulant compositions showed a small clump in the bottom of one column after 38 days. Cesium-loading tests were also conducted in the bounding waste compositions to verify model predictions for column design. Results from the cesium-loading tests using CST matched the ZAM model predictions for all of the simulant compositions tested.

Zeolite had much lower cesium capacity in these simulants. Some of the zeolite particles broke apart in the Early simulant, which had the highest concentration of sodium hydroxide. Figures 10 and 11 show photographs of zeolite and CST after exposure to simulated dissolved saltcake.



Figure 8. Photograph of CST column test equipment.

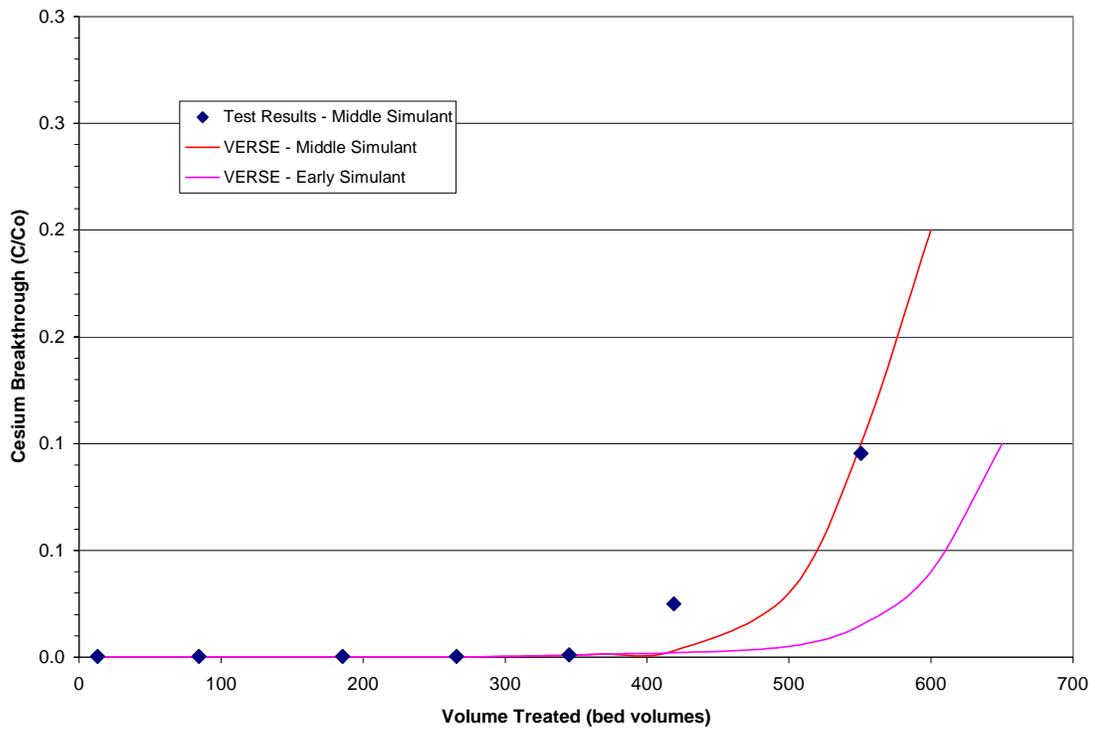


Figure 9. Cesium breakthrough results for CST column test and VERSE model predictions.

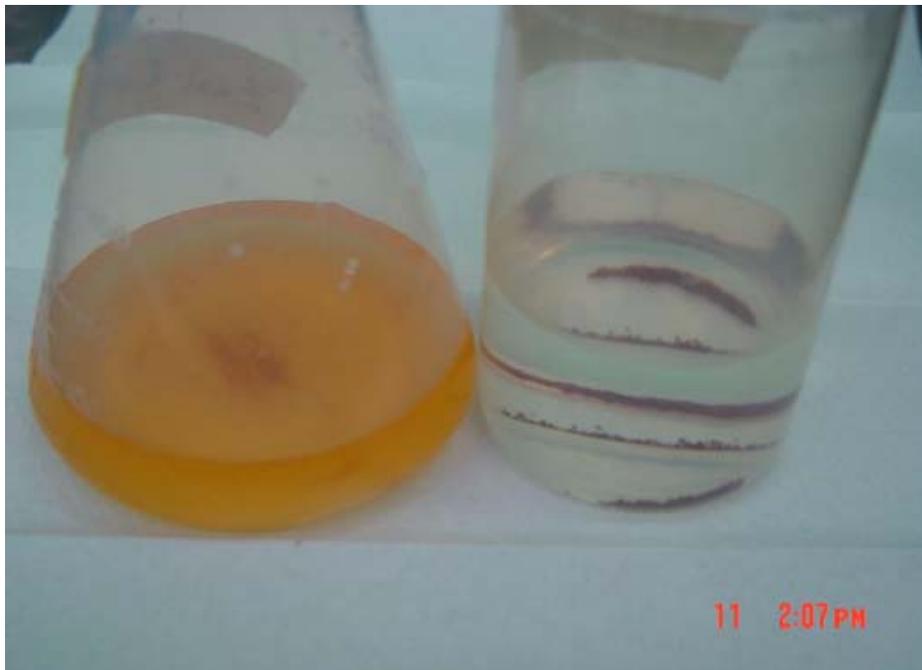


Figure 10. Zeolite in Early (left) and Late (right) simulants, after 7 days of mixing.



Figure 11. CST in Early (left) and Late (right) simulants, after 7 days of mixing.

3. FACILITY EVALUATION AND DISPOSITION OF SORBENT

Several options for the deployment and dispositioning of a small-column system within the SRS tank farms have been identified, including the following:

- Location within tank farm: salt dissolution tank, feed tank to the Saltstone Facility (Tank 50), or pump pit;
- Location at tank: hanging within a riser, shielded skid-mount; and
- CST disposition: to DWPF or to Class A burial ground.

These options were evaluated in the proposal for this work, and a preferred option was selected. This evaluation is summarized in this section. Two examples of possible deployment options follow.

1. Skid-mounted module: The small system for this project could be similar to that used at ORNL for processing radioactive supernate from the MVSTs. Figures 12 and 13, respectively, show photographs of the skid-mounted ion-exchange system before and after installation at ORNL. This modular system used IONSIV™ IE-911 to successfully remove cesium from ~30,000 gal of MVST supernate during a 1997 demonstration. After the demonstration, the skid-mounted ion-exchange system was modified and operated from 1997 through 2000 and was successfully deployed for 14 operational campaigns. During this period, the system processed more than 215,000 gal of radioactive supernate and removed ~9000 Ci of ¹³⁷Cs from the supernate. The deployment of the skid-mounted system within existing containment facilities at ORNL was an important concept that was successfully demonstrated. The ability to use existing facilities near the waste tanks and have the treatment unit designed and fabricated off-site translated into significant cost savings compared with on-site “greenfield” construction of large processing facilities. Modular shield walls were used to reduce the general area radiation doses within the facility. Hands-on maintenance and repairs were successfully performed on the unit without exceeding the planned radiation dose limits for the workers. The ability to drain and flush the system and to shield individual components within the system was a key factor in facilitating hands-on maintenance and reducing radiation exposure to personnel. [15] During processing at ORNL, the IONSIV IE-911 typically removed 90% of the cesium from the radioactive supernate. The cesium-loaded IONSIV IE-911 from the ion-exchange columns was sluiced into shielded high-integrity containers, which were successfully dewatered and packaged to meet the WAC for the Nevada Test Site.

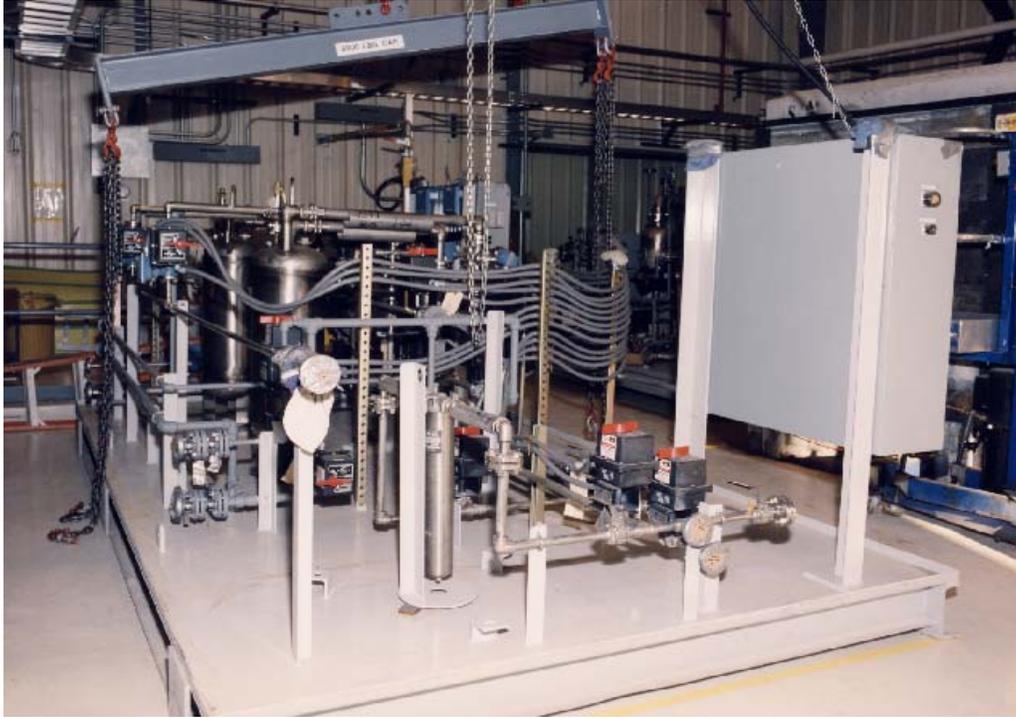


Figure 12. ORNL skid-mounted CST column system before installation.

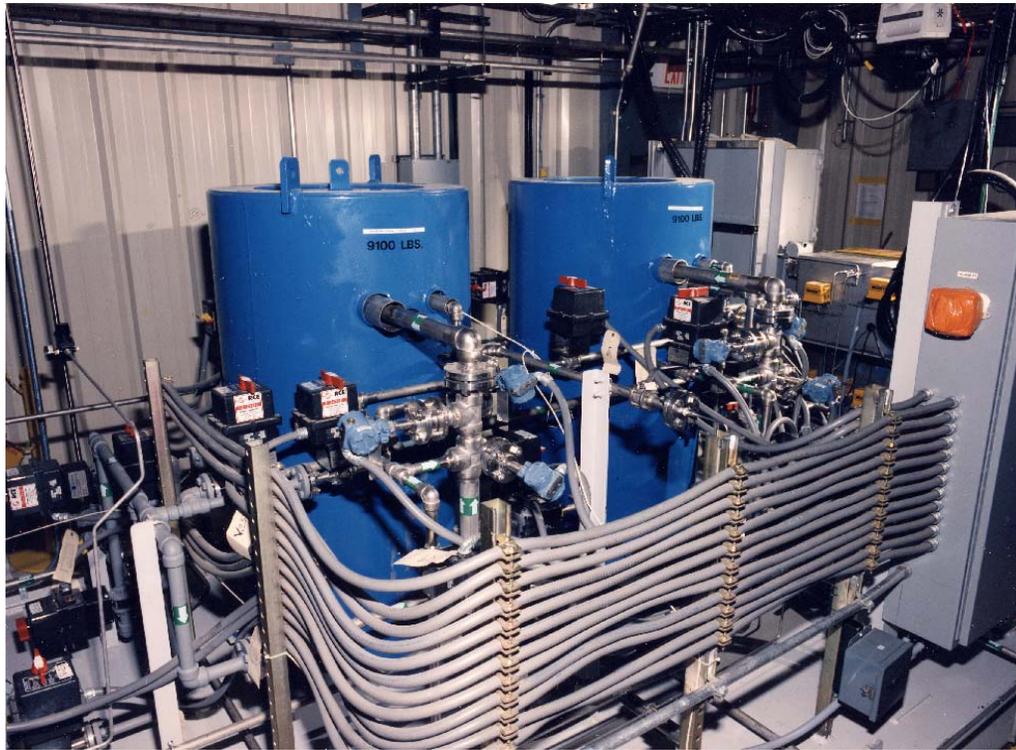


Figure 13. ORNL skid-mounted CST column system after installation.

2. In-tank columns: Alternatively, the small-column system for this project could be similar to the WVDP zeolite column. [16] Figure 14 indicates the general equipment arrangement and process flow for the WVDP system. The existing riser plug locations in the SRS F or H Area HLW Tanks could be used for the SRS application. The process design is compatible with existing waste tanks, transfer lines, diversion boxes, pump pits, etc. Although the system is intended to be run with two stations, each with two columns in parallel, a simple jumper change would allow operation of beds in series. Dissolved salt would be prefiltered and pumped through ion-exchange beds located in the tank riser, or alternatively near the diversion box. Figure 15 shows a conceptual drawing of an ion-exchange bed that could be used at SRS. Connections are made through small Hanford or transuranic (TRU) connectors. The overall length is 10 ft, with a 2-ft diam. The canister is 3/8-in.-thick stainless steel with a downflow bed configuration. A removable screen retains the media during column fluffing or backwashing. The screen can be removed and the spent resin transported by high-flow backwashing. In normal column changeout the suspended spent resin is passed through an in-line grinder before being dumped into the waste tank. A tank containing washed sludge is most suitable since it may help to suspend the spent resin and avoid clumping. Washed sludge also subjects the resin to long term storage under lower alkalinity levels than unwashed sludge does.

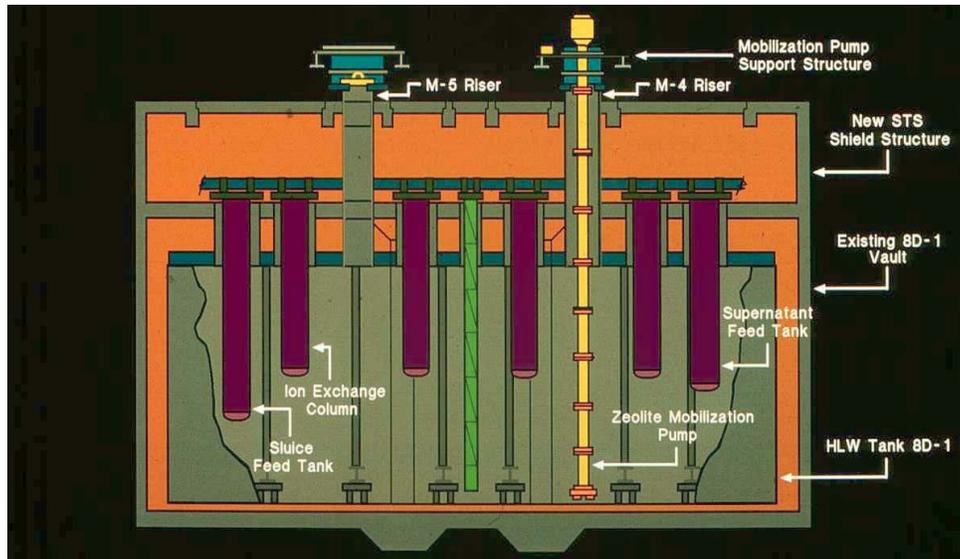


Figure 14. General equipment arrangement and process flow for the zeolite columns used in-tank at the WVDP.



Figure 15. Conceptual drawing of an ion-exchange column.

A portion of the cesium-loaded sorbent from ORNL was also shipped to the Savannah River Technology Center (SRTC) for use in vitrification demonstrations.[17] The SRTC developed a glass formulation for the IE-911 that could incorporate 65 wt % of the cesium-loaded sorbent into the glass matrix without crystal formation. [18] Past vitrification studies conducted by SRTC indicate that the CST is a candidate for vitrification in the DWPF along with the high-curie salt supernate waste. [19, 20] Thus, two options for disposition are as follows:

1. store loaded sorbent in the column or a container at a burial site, or
2. send the loaded sorbent to DWPF to be vitrified into HLW glass.

On the basis of cost and safety evaluations conducted during this phase of the project the use of in-tank columns similar to those at WVDP with final disposition to DWPF was selected as the preferred option for SCIX.

Figure 16 is a conceptual drawing of two of the resin beds contained in a modified SRS waste tank riser plug. A remote manipulator or epoxy-coated industrial-grade gantry crane with an impact wrench are used to connect and disconnect the mini Hanford or TRU connectors between the bed and solution lines. Valving and pump flows are controlled at a diversion box. Column operation is in the normal manner with the columns either in series or parallel depending on the particular salt requirements. A wash-down ring allows removal of the bed by an overhead crane (or DWPF transporter if the column is located at the diversion box) if necessary. Column leakage or inhibited wash-down water flows by gravity through seal legs directly into the tank below. Spent resin is back-washed through an in-line grinder similar to the one used at the WVDP, and is then discharged through a seal leg into the tank below. Plugged beds or cemented beds are disconnected and sealed by a remote manipulator or crane, to be removed by crane or by DWPF transporter if adequate transporter support is provided.

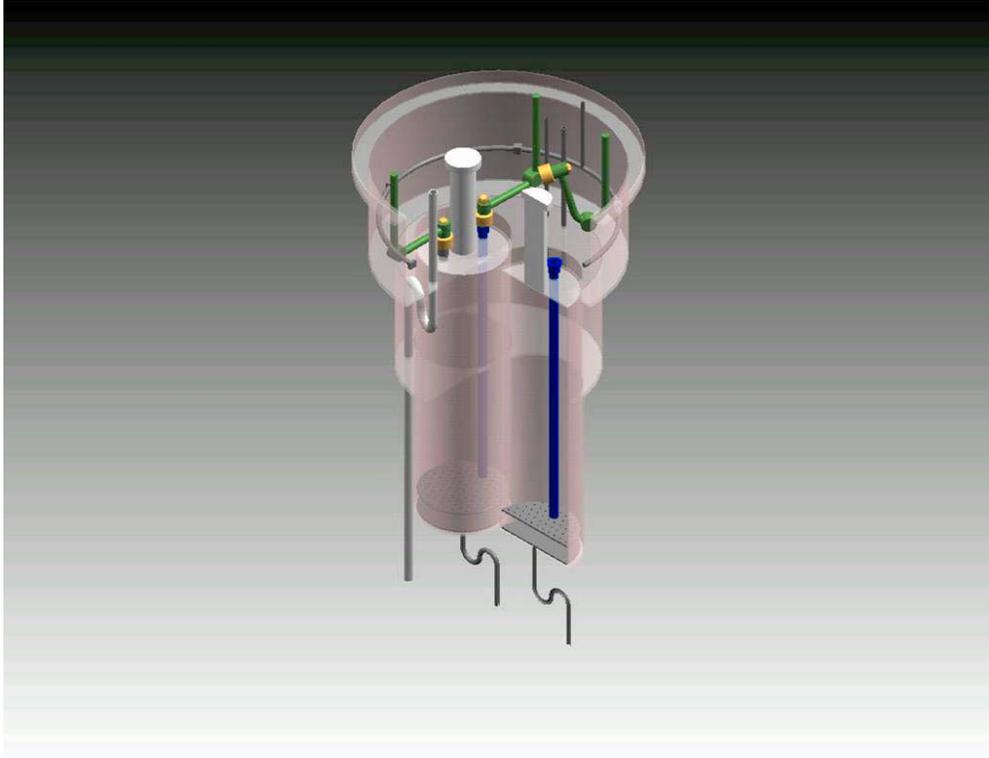


Figure 16. Conceptual drawing of two resin beds contained in a modified SRS waste tank riser plug.

4. SYSTEM DESIGN

4.1 HEAT TRANSFER CALCULATIONS

In the large CST columns (16 ft high with a diameter of 5 ft) evaluated for SWPF, the large ^{137}Cs source (up to 5.8 million curies) would generate enough heat to boil the salt solution within the column in about 33 h should loss of flow occur. In the small system considered for this proposal, solution boiling is prevented by keeping the diameter of the ion-exchange column below 32 in., limiting ^{137}Cs loading, and cooling by ambient air and cooling water. Calculations of the heat evolved resulted in the design of a cooling water system to maintain the SCIX columns within safe operating temperatures if the flow of dissolved saltcake is stopped in a fully loaded column. The conclusions of these calculations are presented below.[21]

Transient two-dimensional heat conduction calculations with modeling assumptions have been performed to assess how rapidly a CST ion-exchange column heats up on loss of flow under the potential operating conditions and design geometries. In the present analysis, convection and radiation transport processes inside the CST column were assumed to be negligible compared with conduction heat transfer under no-process-flow conditions. The CST-salt solution column was assumed to be cooled by natural convection with no process flow and a mixture of natural and forced-flow convection with process flow. The use of forced-flow cooling water in a conduit in the center of the resin bed and in tubing placed against the outside of the resin bed increased the driving force and rate of heat transfer but did not provide forced convective flow through the bed itself. Thus, the main mechanism for heat transport remained conduction, with the column cooled by natural convection with or without the forced flow convection of process flow. A computational heat transfer approach was taken using Fluent™, a commercial computational fluid dynamics (CFD) code. In addition, the results computed by the present model were verified by the theoretical results.

Cases I and II investigated steady-state and transient temperature profiles of the CST-salt solution system due to natural convection cooling and quantified the CST temperatures inside the cylindrical packed column containing a decay heat source for the no-process-flow situation. Because the modeling results of the Case I and Case II designs show that peak temperature of the CST-salt solution system reaches an undesirable boiling condition in case of no process flow, Cases III and IV quantified the transient temperature responses of the system heatup for various design conditions and different cooling mechanisms. The transient results for the column cooled by mixed convection showed that the 130°C maximum temperature of the column was reached about 10 days after the transient initiation of no-flow conditions.

A maximum temperature of 100°C is reached about 56 hours after losing solution flow through the column if the column is loaded at 300 Ci/liter and the emergency cooling water system is activated about 12 hours after loss of solution flow. Sensitivity analysis for the CST column quantified the impact of the parameters on the system cooling capability. The parameters studied were the efficient column geometry, heat load, and cooling mechanism of the CST column with no process flow.

The main conclusions of this analysis follow:

- Under the no-process-flow condition, the transient temperature response of the CST-salt column loaded with 300 Ci/L decay heat is slow. It takes about 4 days for a 20-in. diam

column to reach boiling conditions with no process flow and no emergency cooling water system.

- The water coolant system along the central axis of the column is most effective in removing the column heat load under no-flow conditions, since an effective thermal boundary layer adjacent to the potential location of the peak temperature is formed by convective fluid motion.
- The maximum column temperature was affected more by the cooling mechanism at the column center than by the cooling mechanism at the column boundary. Thus, the temperature increase of the column is reduced rapidly as cooling capability at the center of the column is enhanced.

The modeling analysis has been performed to compute temperature distributions within the modeling boundary shown in Figure 17 and to investigate transient temperature response to the decay heat load of the CST column with no process flow. In this case, a non-boiling temperature criterion (130°C) was used as one thermal acceptance criteria for the potential CST column designs.

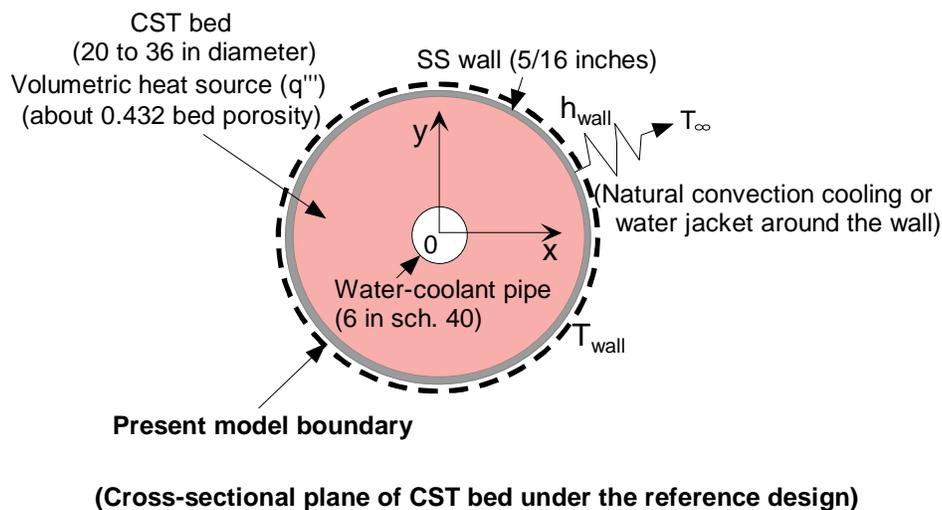


Figure 17. Present model boundary for the heat transfer analysis of the CST bed.

Figure 18 shows the results of calculations investigating the effect of column diameter on the maximum temperature reached by a column cooled by convective heat transfer to air at the outside surface. The graph clearly shows that the cesium loading must be restricted to less than 100 to 200 Ci/L for the required range of column diameters to avoid exceeding the boiling point of the waste solution (130 °C). The capability of CST to exceed this limit suggests additional cooling must be designed into the columns. We investigated the effect of cooling fins on the outside of the column. However, these fins had little impact on the temperature, presumably because of slow heat transfer within the column rather than at the surface.

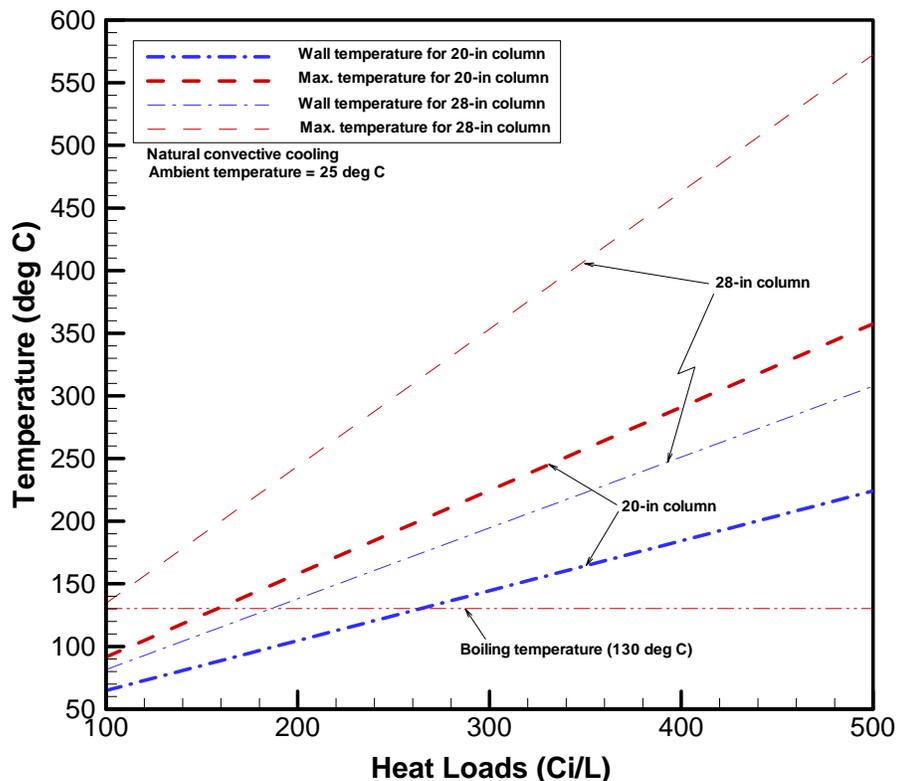


Figure 18. Temperature (at wall and maximum internal) for columns with various heat loads following loss of flow. No cooling occurs except for ambient air at outside surface of column.

Since passive cooling methods appear inadequate, we considered using water cooling jackets. Calculations examined a central cooling tube, outside cooling strips, and a combination of both. The combination of the two proved most suitable for the cesium loading range of the sorbent (up to 600 Ci/L). Figure 19 shows the proposed cooling system containing a 6-in.-diam central cooling tube and either four (Case a) or eight (Case a1) cooling strips on the outside surface of a 28-in.-diam column. Addition of more cooling lines on the outside of the column reduces the maximum temperature. As shown in Figure 20, doubling the number of strips from four to eight reduces the maximum column temperature by about 10°C. Increasing the internal pipe diameter reduces the maximum temperature, although little gain resulted for diameters greater than a few inches. The reference case uses a 6-in.-diam tube, based on the ready availability of this size.

Changing the temperature of the cooling water directly affects the maximum temperature. Most calculations assumed that a dedicated chiller delivers 20°C cooling water. However, use of 30°C water raises the maximum temperature by about 10°.

Figure 21 shows the change in column temperature during a loss-of-flow/loss-of-cooling accident. The initial column temperature of 30°C increases to 100°C in 56 hours. At that time, cooling water flow at 16 gal/min and 30°C is restored. The column

temperature continues to increase by 5° over 12 h and then decreases to approach a steady state value of 91°C.

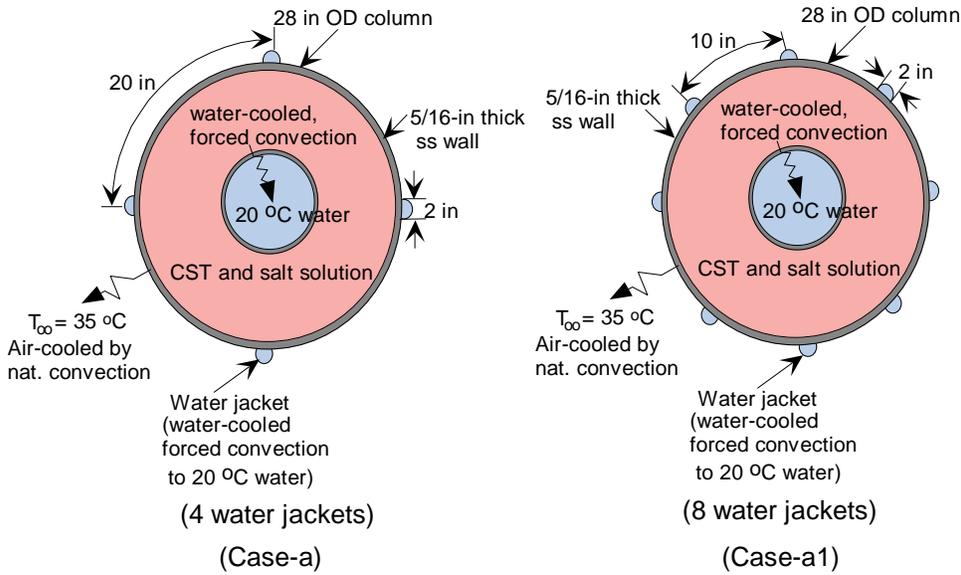


Figure 19. Model boundary of CST bed as an annulus with a central cooling core and cooling channels on the outer perimeter.

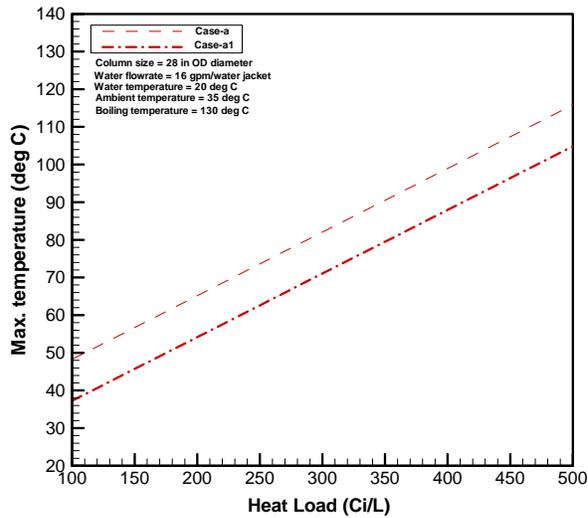


Figure 20. Effect of cooling on maximum temperature at various heat loads during loss of feed flow to column. Cooling is provided by 16 gal/min water flow through a central cooling tube (6-in.-diam) and 4 (upper curve) or 8 (lower curve) cooling lines on the outside of the column.

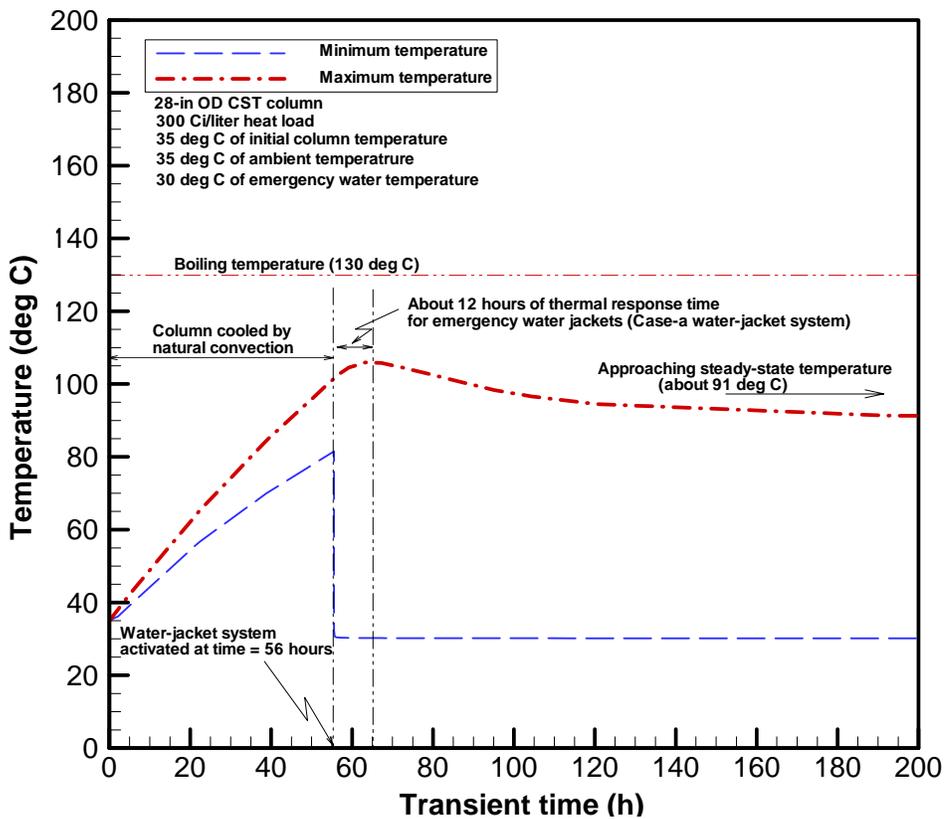


Figure 21. Temperature changes following loss of flow and loss of cooling in a column loaded with sorbent at 300 Ci/L.

4.2 SHIELDING CALCULATIONS

Dose rates for 30 scenarios were calculated using the Anisotropic S_n (ANISN) one-dimensional discrete-ordinates radiation transport code. The cylindrical geometry option was used along with an S_{16} quadrature and a P_5 Legendre polynomial expansion of the scattering cross-section data. For the two source strengths in supernate, a calculation was performed for 1.0 Ci/gal, and the results for 0.8 and 2.0 Ci/gal were obtained by scaling those results during plotting. Figures 22 and 23 illustrate the estimated dose rates using dissolved saltcake at 0.8 Ci/gal and resin loaded at 300 Ci/L in a 1.5-in.-diam Schedule 40 stainless steel pipe, respectively.

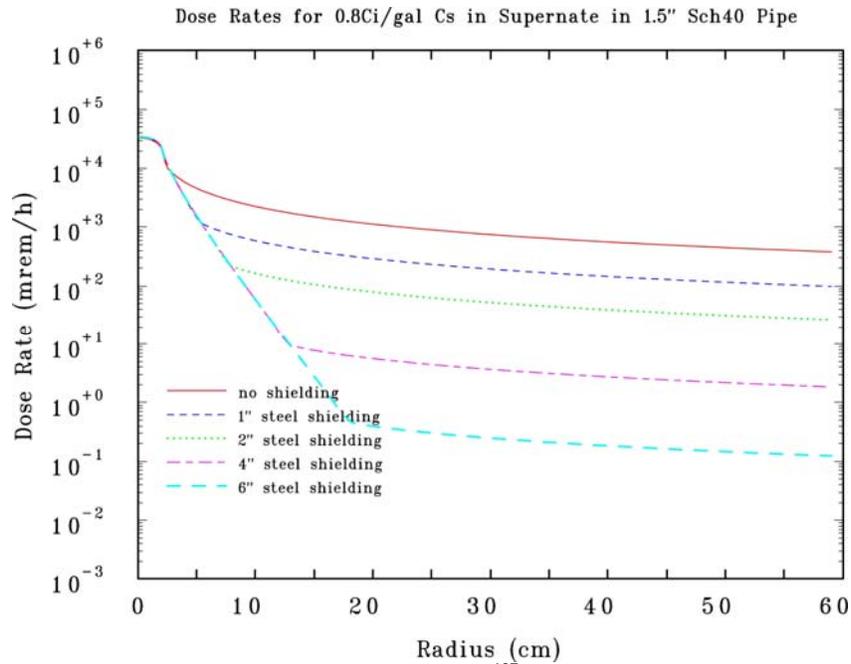


Figure 22. Estimated dose rates for 0.8 Ci/gal of ^{137}Cs in a nominal 1.5-in.-diam Schedule 40 stainless steel pipe.

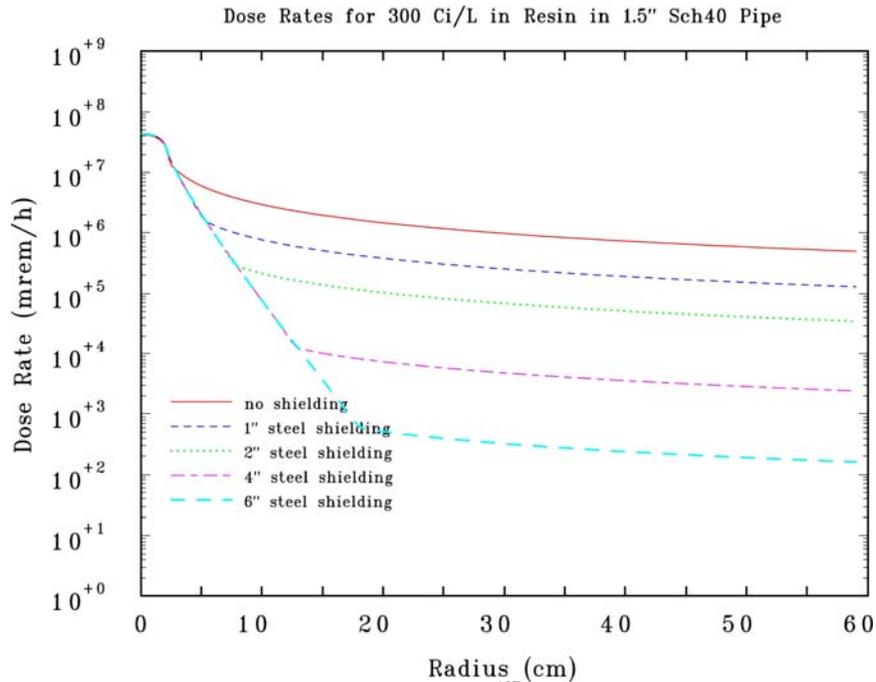


Figure 23. Estimated dose rates for 300 Ci/L of ^{137}Cs loaded on resin in a nominal 1.5-in.-diam Schedule 40 stainless steel pipe.

Calculations for the waste tanks with revised geometries were performed using the Discrete Ordinates Radiation Transport (DORT) two-dimensional radiation transport computer code. Nine cases were considered. A plot of the zone geometry is shown in Figure 24. There is a 2-in. steel plate just above the column, 38 in. of steel shot above the plate, and a 6-in. steel plate at the top of

the riser plug for shielding. The DORT calculations used cylindrical (R-Z) geometry, cross-section data from the DABL69 library (23-group photon-only) with scattering moments expanded to fifth order in Legendre polynomials and a symmetric S_{16} quadrature (160 directions). Eleven mixtures and void were present in the models. For the steel shot, a volume fraction of 0.64 was used because this was said to be the maximum volume fraction for random compaction of equal-sized spheres. The source energy spectrum was that of ^{137}Cs in equilibrium with ^{137m}Ba (a 23-group spectrum). Figures 25 and 26 illustrate two of these calculations. Figure 25 shows the dose rates with air in the riser (radiation from the waste in the tank), and Figure 26 shows the results with loaded resin (300 Ci/L) in the riser. Dose rates in the riser are increased two to four orders of magnitude when compared to the case of just air in the riser.

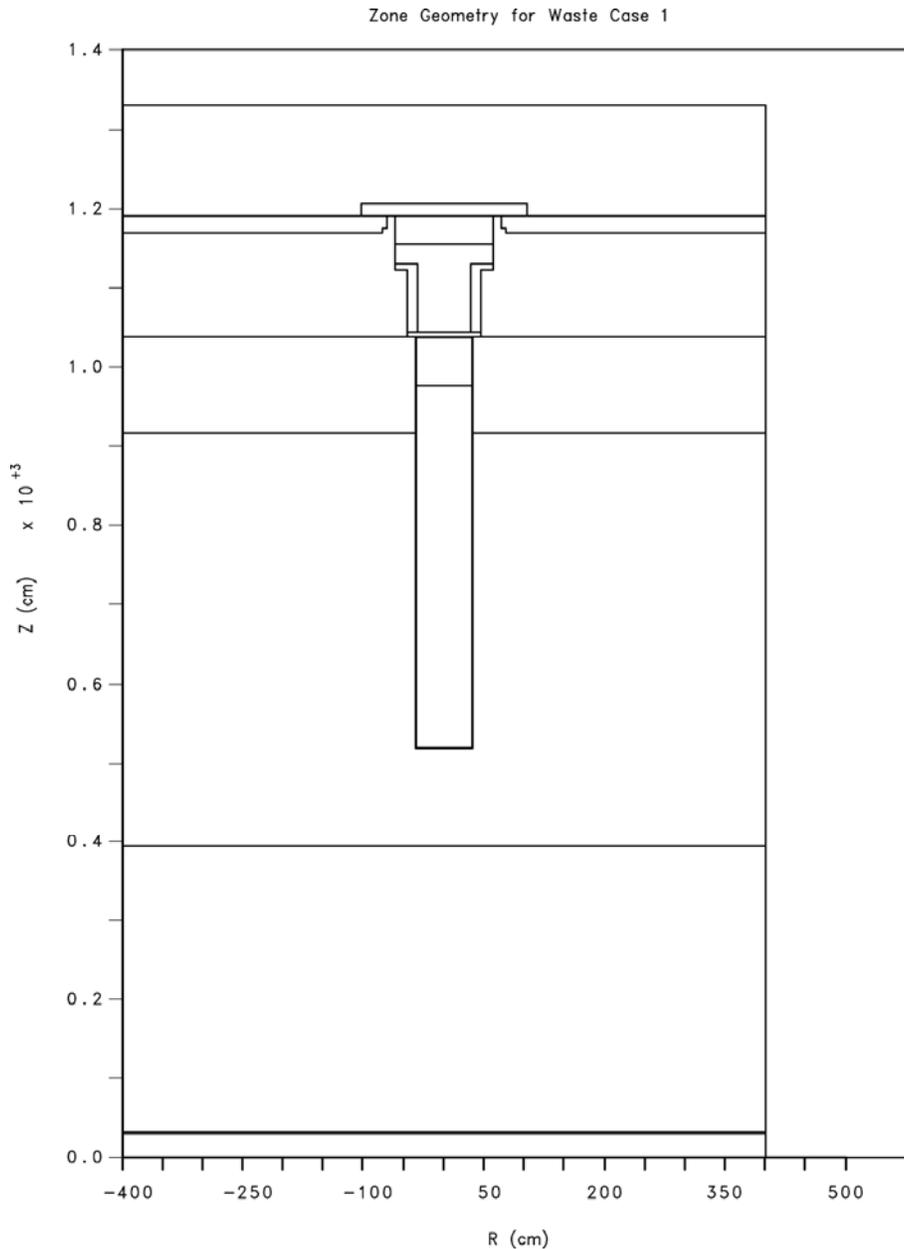


Figure 24. Zone geometry model for waste tank cases.

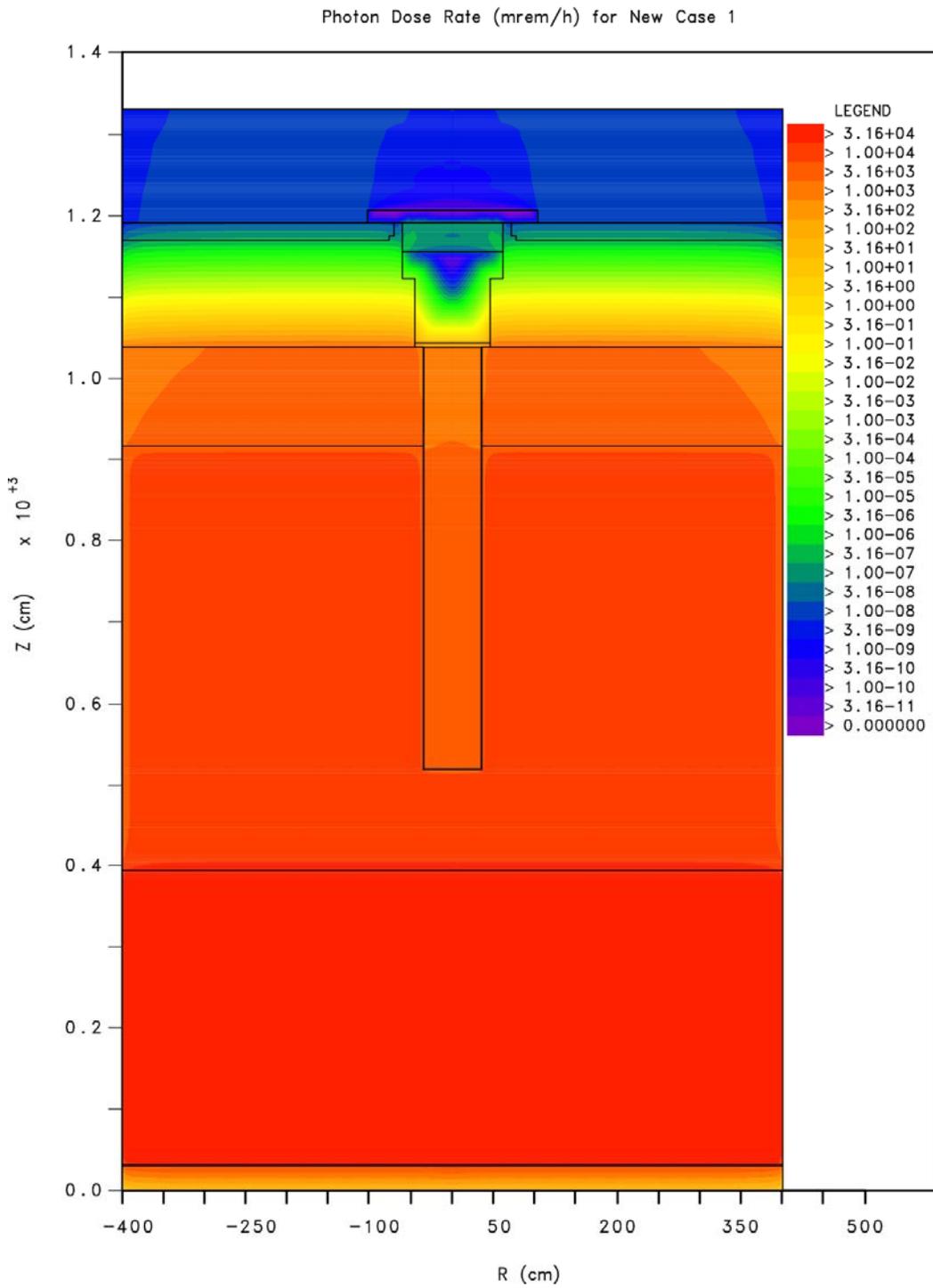


Figure 25. Dose rates (mrem/h) for a waste tank model having air in the riser with separated sludge and supernate regions.

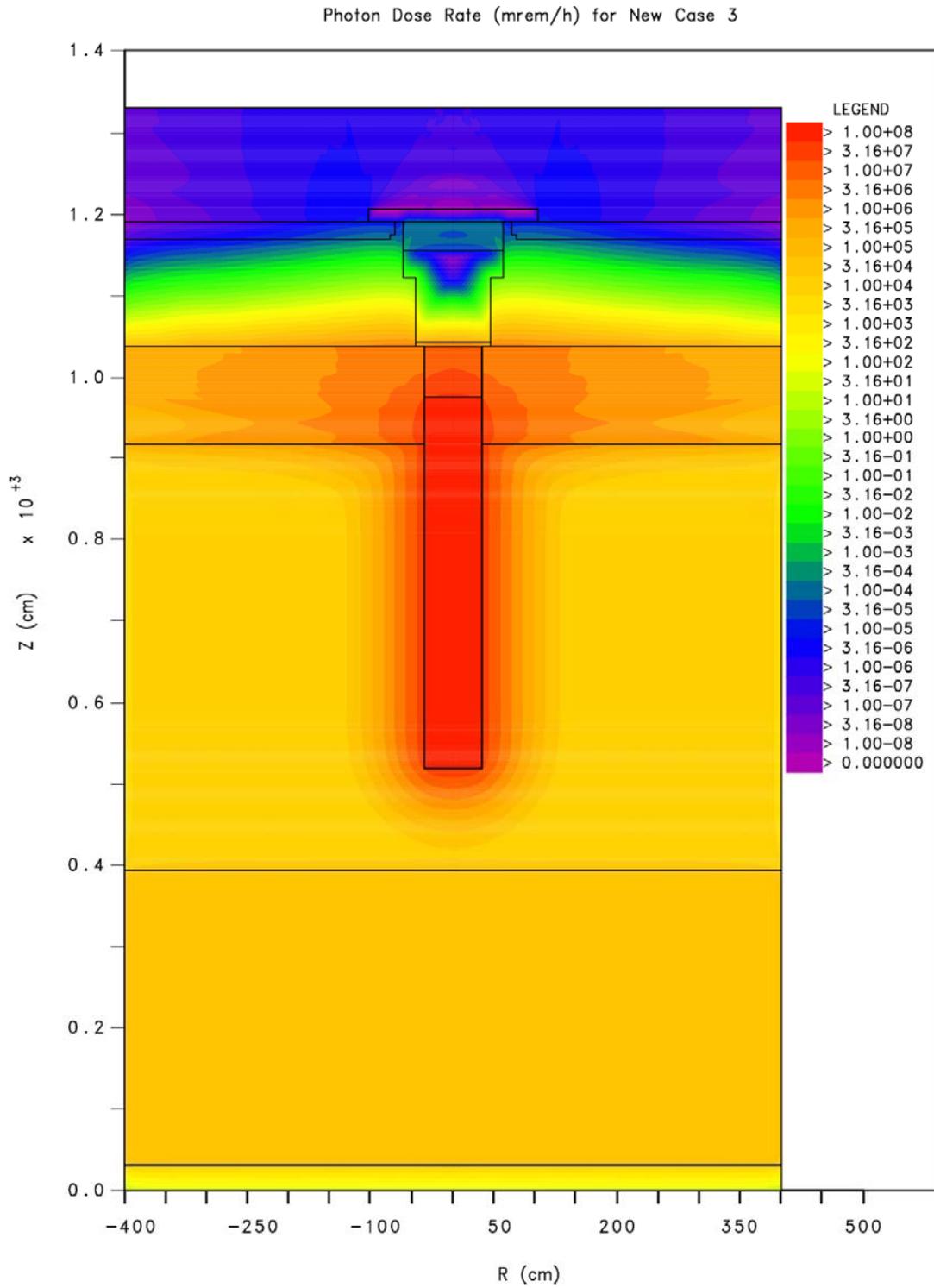


Figure 26. Dose rates (mrem/h) for a waste tank model having 300 Ci/L of ^{137}Cs loaded on resin within the riser and separated sludge and supernate regions.

5. COST BENEFIT ANALYSIS

The baseline treatment of the HLW saltcake at the SRS is projected to be completed in 2039 [2] at an estimated cost of about \$6 billion (total cost for HLW tank storage and to process dissolved saltcake materials through the following facilities SWPF, DWPF, and Saltstone) [22]. SRS devised an accelerated alternative to process only one-third of the dissolved saltcake through SWPF. The accelerated alternative is projected to save up to \$3 billion and to process the waste in less than one-half the time compared with the baseline, when CST is used to remove ^{137}Cs from liquid LLW in order to meet the proposed Saltstone Facility WAC. The unit cost was not known for processing Type 3 waste through the Actinide Removal Process (ARP). Therefore, this cost was conservatively assumed to be 50% of the SWPF (ARP + CSSX) cost per gallon of solution. The actual ARP cost is expected to be lower, increasing the projected saving over the baseline. Substituting zeolite for CST practically eliminates the conservative cost advantage, although actual ARP costs lower than that assumed could still result in savings that approach \$1 billion.

Figures 27 and 28 compare the estimated costs for the baseline with the accelerated alternatives for Types 1, 2, and 3 wastes, either using CST or zeolite. For Figure 27, all of the Type 2 and 3 wastes were assumed to be processed through the CSIX. For Figure 28, only Type 2 waste (none of Type 3) was assumed processed through CSIX. This latter case made processing Type 3 waste independent of CSIX, or which of the two sorbents is used. With the assumptions made in this evaluation, only the rates and costs for Types 2 and 3 were affected for these cases. For this evaluation, the HLW tank farm storage cost was treated as a fixed cost (\$100 M/yr) for the accelerated baseline. Since the dissolved saltcake volume was divided equally among the three waste types, this storage cost was also divided equally among these three liquid waste streams. The full cost for each stream was assessed each year during its processing but ceased when processing was completed for a given waste stream. (For example, if processing was completed on Type 2 waste, but not Types 1 or 3; then, in the following years, storage cost ceased for Type 2, but not Types 1 and 3.) The liquids added to the LLW while changing the sorbent bed in the SCIX system, and the loaded sorbent dumped into the tanks as HLW result in higher costs for the Saltstone Facility and DWPF, respectively. Table 2 lists other assumptions used in this analysis. In addition, the cost per gallon for ARP was conservatively assumed to be 50% of the SWPF cost per gallon. The cost per gallon of liquid waste was used to calculate total cost for a given alternative with no correction for fixed costs. Life cycle costs calculated by SRS divided by the total gallons expected to be processed are the source of these cost per gallon factors, but no breakdown of these lifecycle costs were available to allow estimation of the % fixed cost and % processing cost. Extrapolation of these numbers beyond the waste volumes that were used to estimate the original life cycle costs led to error and deviation from the original estimates. Thus, the costs estimated in this analysis roughly illustrate whether a benefit may be expected, but the actual dollar amounts may deviate greatly from those actually incurred when and if these plants are built and waste is actually processed. For comparative purposes, the analysis indicates a significant cost benefit can be expected if the accelerated alternative is implemented. The SCIX helps ensure that this alternative can be safely implemented without adversely impacting system criteria and protocols. Thus, the probability is significantly greater with SCIX supporting the accelerated alternative baseline of treating the saltcake from the SRS HLW tanks quicker and at less cost.

Zeolite is a lower-cost sorbent that is widely available commercially. However, its reduced capacity for loading offsets much of the lower purchase cost because of the much greater quantity of sorbent required. In addition, the extra mass of sorbent sent to DWPF results in costs far greater than any savings from the purchase price for zeolite. The main difference between Figures 27 and 28 is the much greater DWPF cost for the alternative using zeolite in Figure 27, because processing all of the Type 3 waste through SCIX doubles the amount of zeolite loaded and sent to DWPF. Processing all of this zeolite could potentially produce over 7000 additional glass canisters, while CST would produce <500.

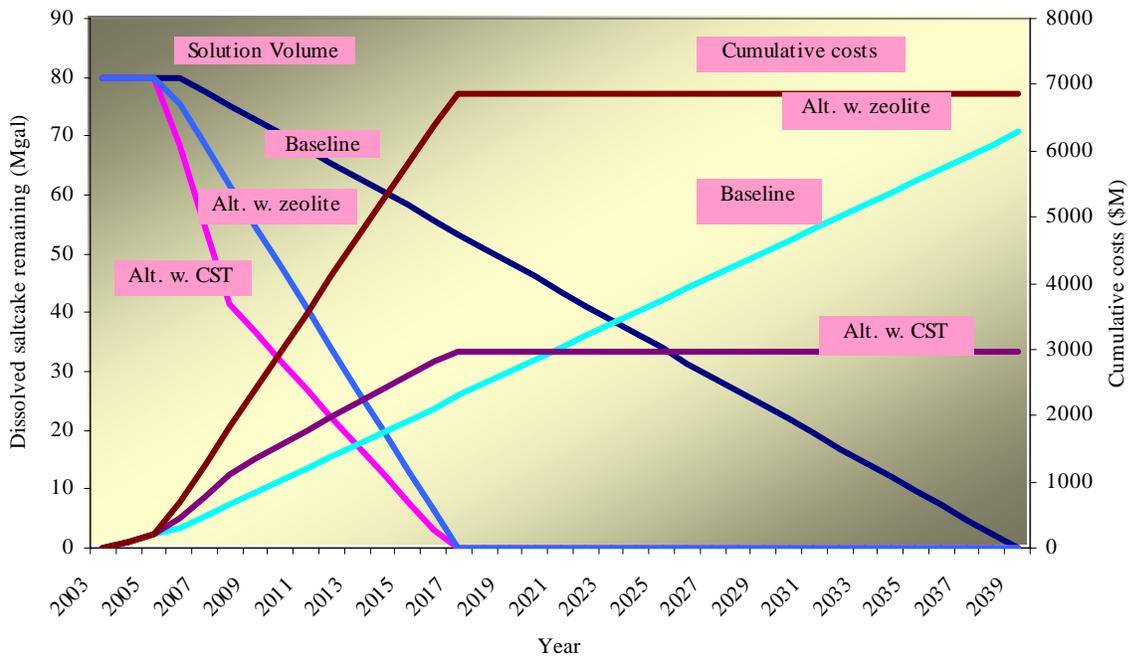


Figure 27. Comparison of saltcake solution processing and costs for the baseline and the accelerated alternative (using either CST or zeolite) when processing all Type 2 and 3 wastes through the SCIX (Case 1).

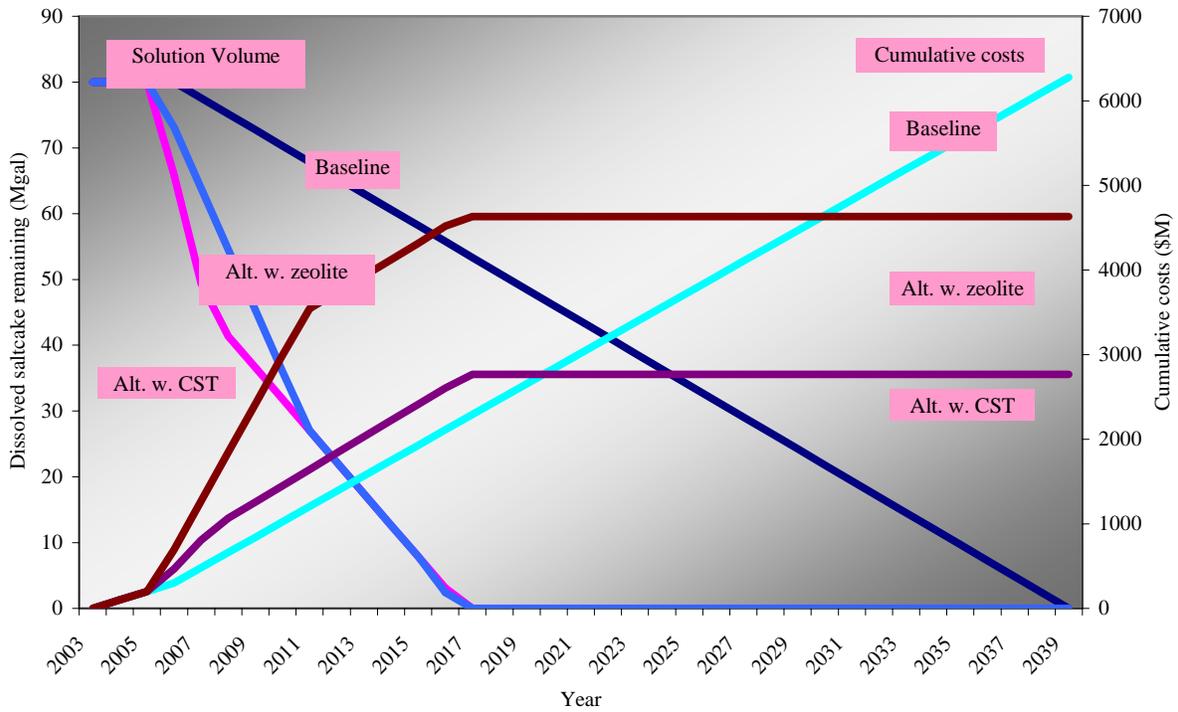


Figure 28. Comparison of saltcake solution processing and costs for the baseline and the accelerated alternative (using either CST or zeolite) when processing all Type 2 (but not Type 3) waste through the SCIX (Case 2).

Table 2. List of assumptions used in cost benefit analysis

<u>Some SRS costs</u>	
Cost of SWPF/gal solution	\$29.45
Cost of Saltstone/gal solution	\$4.01
(Cost of SWPF + Saltstone)/gal solution	\$33.46
Cost of SRS tank farm/yr	\$100,000,000
<u>DWPF cost</u>	
Assumed cost of one "glass log" (\$/canister)	\$500,000
Estimated log weight (lbs/canister)	4,000
Assumed waste oxide loading	35%
Waste oxide (lbs/canister)	1,400
Assumed cost of DWPF (\$/lb waste oxide)	\$357
<u>SRS waste solution volumes (supernate + dissolved saltcake)</u>	
Total vol, gal	80,000,000
Vol of Type 1, gal	26,666,667
Vol of Type 2, gal	26,666,667
Vol of Type 3, gal	26,666,667
Vol per HLW tank, gal/tank	4,000,000
<u>Manpower costs & workdays/y basis</u>	
Operator, unburdened, FY2003 (\$/h)	\$53.00
Operations engineer, unburdened, FY2003 (\$/h)	\$70.00
Overhead burden FY2003	35%
Inflation factor to adjust manpower to FY2004 basis	3%
Operator (\$/h)	\$73.70
Engineer (\$/h)	\$97.34
Assumed % of calendar days/yr processing	100%
Assumed # 24-h workdays/y	365
<u>Assumed timeline years</u>	
Zero year, times est'd from this year	2003
Year baseline scheduled for completion	2039
Year Alt processing starts for Type 2 (CST cols)	2006
Year Alt processing starts for ARP	2006
Assumed start of SWPF processing (baseline and Type 1)	2007
<u>Process rate (gal/y)</u>	
Assumed Saltstone process rate for 40 h/week (gal saltcake/y)	6,000,000

6. IMPACTS AND RISK ASSESMENT OF SCIX ON HLW SYSTEM

The team members performed a risk assessment of the SCIX process. This risk assessment is only summarized here as the risk assessment matrix was too large for this summary report. A total of 73 risk statements were identified and evaluated based on the preliminary design. The evaluation yielded 3 high, 33 moderate, and 37 low risks before mitigation. After mitigation, there were no high risks and two moderate risks, with the remainder categorized as low risks. The unmitigated high risks included high column temperature, hydrogen accumulation in the column, and process design uncertainty. After mitigation, the two remaining moderate risks were pressure relief in the column and adequacy of the grinder size reduction of the sorbent. The evaluation generated a few minor design changes and provides guidance concerning where to focus future efforts to reduce process risks.

In addition, team members performed an assessment of the impact of the SCIX on HLW processing, using quantitative ratings ranging from “---” for a major negative impact to “+++” for a major positive impact. Table 3 summarizes this quantitative assessment of the SCIX impacts.

Table 3. Quantitative assessment of the impacts of the SCIX on HLW processing at SRS

Title	Explanation	Impact^a
High-cesium LCS waste	Allows processing of LCS waste that does not meet cesium requirement for Saltstone. Avoids recycling dissolved salt waste to evaporators.	+++
High-alpha LCS waste	Can be applied to Sr and Actinide removal with mixed bed or alpha absorbent. May require columns in series	++
High-activity salt	Upon demonstrated use might be applied to high activity salt. Would require multiple columns and reevaluation of shielding and maintenance based on operating experience. May require pH adjustment for some absorbents.	++
Evaporators	Reduces load on evaporators by processing LCS and ARP waste that does not meet Saltstone limit.	++
Flexibility	Improves tank farm flexibility for blending wastes by emptying tanks sooner.	+++
Transfer frequency	Process requires 42 intertank transfers per year, thereby increasing complexity of tank farm operations.	-
Transport and storage of chemicals	Purchasing, handling, storage, pH adjustment, and fines removal of ion-exchange sorbent required in or near tank farm. Increases usage of inhibited water. Requires use of 1 M NaOH.	-
Ion-exchange feed tank	A dedicated feed tank for the ion-exchange process is required to maximize the processing rate. If Tank 41 is made the feed tank, an additional tank must be identified for the point of compliance for Saltstone. Process throughput is reduced by 50% if a tank is not available.	--
High-cesium ARP waste	Allows processing of ARP waste that does not meet cesium requirement for Saltstone. Avoids recycling dissolved salt waste to evaporators.	+++

Title	Explanation	Impact^a
Transfer line to ion-exchange process and back	For the ion-exchange process to handle noncompliant ARP waste, transfer lines to and from ARP must be installed if SCIX is placed after ARP. Adds complexity to ARP facility. Placing SCIX prior to ARP eliminates the need for this line.	-
Efficiency	Improves efficiency of Saltstone process by ensuring feed availability.	+++
Tank usage versatility	Limits sludge washing to Tank 51. Tank 40 and Tank 51 no longer interchangeable for sludge washing/DWPF feed tank. Note: this point may be moot because of americium/curium additions.	-
Sludge washing rate	Periodic transfers of ion-exchange streams to Tank 51 change tank composition, thus complicating and extending the washing process. Transfers during settling may resuspend sludge.	-
Life cycle costs	Processes saltcake sooner than SWPF, thus reducing amount of cesium-only glass produced at end of DWPF lifetime.	++
Waste Solids Loading	CST additions increase percent waste loading while adding silica	++
Waste composition	Sorbent added to sludge will change composition of washed sludge requiring change in composition and glass models.	-
Number of canisters	Addition of sorbent to sludge waste stream may increase number of glass canisters or sludge work-off rate.	--
Sampler	Addition of sorbent to washed sludge stream may require recalibration of sampler; successful grinding should obviate.	-
New facilities	No new facilities required.	+++
Footprint in existing facilities	Compact. Resin prep: 600 sq ft, one salt supply tank riser, two sludge tank risers with clear path in tank between risers. Temporary trailer. Intertank transfer lines.	+++
Utilities	No major changes. 30 kVA 110/220 V. 50 kVA 440 3-phase. Inhibited water 30 gal/min at 60 psi. Air: 50 scfm at 90 psi.	++
Services	Slight increase in rad control and patrol services.	-

^a

+++	=	major positive impact
++	=	moderate positive impact
+	=	slight positive impact
-	=	slight negative impact
--	=	moderate negative impact
---	=	major negative impact

7. SUMMARY

The results and conclusions of Phase 1 of the SCIX alternative to remove ^{137}Cs from LCS waste at SRS follow.

1. SCIX ensures that the low-curie dissolved saltcake meets the proposed Saltstone WAC (for the accelerated alternative baseline. (Current SCIX design is for WAC of 0.1 Ci/gal but could be redesigned for 0.0002 Ci/gal.)
2. The accelerated alternative, using CST in SCIX, saves ~\$3 billion in operating and storage costs and ~20 years in processing time compared with the current baseline, using a conservative estimate for the ARP cost.
3. CST is the preferred sorbent for SCIX, with its proven high cesium loading capacity for the expected dissolved saltcake solution and temperature.
4. Because of its much lower cesium loading capacity, the low-cost alternative sorbent, zeolite, greatly increases the volume of sorbent required. Thus, zeolite greatly increases the cost for the alternative, mainly because of the increased number of DWPF canisters required to dispose of the loaded sorbent (potentially over 7000 additional canisters for zeolite, compared with <500 for CST).
5. The models previously developed for loading cesium on CST compared favorably with laboratory measurements of equilibrium distribution ratios and column-loading performance.
6. These models predict that a column of 432 gal of CST can operate at 25 gal/min and treat 100,000 to 900,000 gal of dissolved saltcake depending on the solution composition. An average value of 300,000 gal per column was used for the cost benefit analysis.
7. Heat transfer calculations predict nonboiling temperatures for the small-columns with loadings <100 Ci/L with only natural convection cooling. For the loadings up to the maximum calculated for the tank farm (630 Ci/L), a water-cooling system is required to ensure that no boiling occurs in the column.
8. Dose calculations indicate that the maximum dose above the risers is expected to be $\sim 10^{-8}$ and $\sim 10^{-2}$ mrem/h for air and a column loaded at 300 Ci/L in the riser, respectively.
9. The impact analysis indicates a net beneficial impact with no major problems likely to prevent implementation or completion of saltcake treatment.

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