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CBU-PIT-2005-00195 REV. 0
August 9, 2005

KEYWORDS:
Salt Disposition
Radionuclide
Sr-90, Cs-137, TRU
DDA, ARP, MCU, SWPF
Removal Efficiency

RETENTION: PERMANENT
CLASSIFICATION: NA

Radionuclides in SRS Salt Waste

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Prepared for U.S. Department of Energy Under Contract No. DE-AC09-96S

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Radionuclides in SRS Salt Waste

Executive Summary

In this paper, inventories of radionuclides in untreated salt waste (without salt-based nuclide removal treatments) were compared against inventory limits based on Nuclear Regulatory Commission (NRC) Class A concentration limits and the dose limits for radiation protection in the performance objectives of 10 CFR 61 Subpart C. This approach shows that Sr-90, Cs-137, and four alpha-emitting transuranic (TRU) nuclides (Pu-238, Am-241, Cm-244, and Pu-239) contribute most significantly to the radiological risks to the workers, the public, and the environment for the reasons summarized below. Sr-90, Cs-137, and the alpha-emitting TRU nuclides are the only radionuclides in the solidified untreated salt waste¹ having total inventories exceeding the Saltstone Disposal Facility (SDF) inventory limits based on NRC Class A concentration limits (10 CFR 61.55).² No radionuclides (individually or cumulatively) in solidified untreated salt waste have average inventories in an SDF vault that would result in an “all pathways” dose exceeding 10%³ of the maximum annual public dose of 25 mrem specified in 10 CFR 61.41. Cs-137 is the only radionuclide in solidified untreated salt waste with an average inventory in an SDF vault which would result in an intruder dose exceeding 10% of the maximum recommended annual intruder dose of 500 mrem (See 10 CFR 61.42 and Section 5.2 of NUREG-0945, Vol. 1). Cs-137 is the only radionuclide with a total inventory in untreated salt waste exceeding 10% of the SDF inventory limit⁴ based on a maximum allowable annual worker gamma dose of 5.0 rem (See 10 CFR 61.43).⁵ Additionally, Sr-90, Cs-137, and the alpha-emitting TRU nuclides are the radionuclides driving worker inhalation dose.

¹ The comparisons were made as if the waste were solidified as grout, using the grout quantity and grout composition which will be used in the SDF, but without first treating the salt waste through DDA, ARP/MCU or SWPF, which are discussed later in this paper. The solidified waste form was used because it will affect the availability of the radionuclides to the environment, human intruder and the public after disposal.

² Reference to Class A limits is intended only as a tool to assist in screening nuclides for consideration as “highly radioactive.” It does not mean that all nuclides that exceed Class A are highly radioactive radionuclides, *per se*.

³ Use of 10% in this context is not inconsistent with the position adopted by the NRC in another context (decommissioning). Specifically, in that context, the NRC has stated: “NRC staff considers radionuclides and exposure pathways that contribute no greater than 10 percent of the dose criteria to be insignificant contributors” (NUREG-1757, *Consolidated NMSS Decommissioning Guidance*, Vol. 2, Sec. 3.3, p. 3-4).

⁴ For conservatism in this analysis, the inventory limit for liquid waste is used instead of the inventory limit for solidified waste. The inventory limit for liquid waste is lower than for solidified waste because addition of grout materials reduce the radionuclide concentrations and provide additional radiation shielding.

⁵ The maximum dose in this analysis is used for comparison purposes only. The worker dose rate during disposal operations will be controlled to as low as reasonably achievable (ALARA), and hence will be lower than the 5.0 rem used for comparison in this analysis.

Removal of the radionuclides which contribute most significantly to the radiological risk to the workers, the public, and the environment will be accomplished using a combination of the following five treatment processes: 1) deliquification, dissolution, and adjustment (DDA); 2) actinide removal process (ARP) without monosodium titanate (MST) sorption; 3) ARP with MST sorption; 4) modular caustic side solvent extraction unit (MCU); and 5) Salt Waste Processing Facility (SWPF) treatments. Removal efficiencies for each of the planned treatment processes are identified in the table below.

Treatment Process	Removal Efficiency, %								
	Sr-90			Cs-137			α -emitting TRU		
	Nom	Low	High	Nom	Low	High	Nom	Low	High
DDA	66	46	86	50	30	70	63	43	83
ARP without MST	99.6	98.0	99.9	~ 0	~ 0	~ 0	78	50	93
ARP with MST	99.997	99.4	99.999	~ 0	~ 0	~ 0	98	90	99.9
MCU	0	0	0	91	90	92	0	0	0
SWPF	99.98	99.4	99.995	99.998	99.99	99.998	96	90	99.5

For removal of strontium and alpha-emitting TRU nuclides, the relative treatment efficacies are:
 ARP with MST \approx SWPF > ARP without MST > DDA >> MCU

For removal of cesium, the relative treatment efficacies are:
 SWPF > MCU > DDA >> ARP

Introduction

This document compares the radionuclide inventories in the untreated salt waste to the inventory limits based on the Class A concentration limits (10 CFR 61.55) and on the dose limits addressed in the performance objectives (*See* 10 CFR Part 61, subpart C). This document also provides the technical basis for quantifying treatment removal efficiencies for those radionuclides which contribute most significantly to the radiological risk to the workers, the public, and the environment.

The treatment processes addressed in this document include:

- Deliquification, Dissolution, and Adjustment (DDA)
- Actinide Removal Process (ARP) without Monosodium Titanate (MST) Sorption
- ARP with MST Sorption
- Modular Caustic Side Solvent Extraction Unit (MCU)
- Salt Waste Processing Facility (SWPF)

Methodology

Background

Approximately thirty-six million gallons of radioactive waste are currently stored in 49 underground tanks at the Savannah River Site (SRS). Most of this material was generated through defense-related reprocessing of spent nuclear fuel. By volume, approximately 93% of the material is “salt waste” consisting of supernatant salt solution and solid precipitated salts. Disposition of the salt waste will be accomplished through a three-part approach. First, the salt waste will be treated to remove to the maximum extent practical those radionuclides which contribute most significantly to the radiological risk to the workers, the public, and the environment. Second, the treated salt waste will be stabilized in a grout matrix. Lastly, the stabilized waste will be disposed in vaults at the SRS Saltstone Disposal Facility (SDF).

Note that in this document the term “treatment” applies solely to the processes performed for the purpose of removing radionuclides from salt waste. This includes DDA, ARP, MCU, and SWPF treatments. In this document, “treatment” does not refer to processes performed: a) before waste was received into the underground storage tanks (i.e., segregation of waste and pH neutralization); b) for the purpose of maximizing tank space (i.e., evaporation); or c) for the purpose of stabilization at SDF (i.e., grouting). Consistent with this approach, the term “untreated waste” refers solely to waste that has not been treated by DDA, ARP, MCU, or SWPF.

Approach for Determining Radionuclides Which Pose a Significant Risk to the Public, Workers, or the Environment

From a technical perspective, the approach should be based on quantification of the potential risks impacting public, worker, and environmental health. For radionuclides, this includes consideration of the potential internal and external radiation doses associated with near proximity exposure to the waste, as well as remote exposures associated with inhalation and/or ingestion of material transported via environmental media.

An approach was taken that is sufficiently conservative to take variabilities and uncertainties into account. The approach provides a basis for identifying the radionuclides which pose a significant risk from disposal of the solidified salt waste to the public and the environment, and to workers during disposal operations, as well as radionuclide removal needs. As such, the approach is based on comparisons of quantities of radionuclides in untreated waste versus radionuclide limits based on NRC Class A concentration limits (10 CFR 61.55) and performance objective dose limits (*See* 10 CFR Part 61, Subpart C). An observation that the quantity of a nuclide is an insignificant fraction of the limits is consistent with a conclusion that the given radionuclide does not pose a significant risk to the public, workers or the environment.

The first comparison is based on the NRC Class A concentration limits, which are significantly lower than the Class C concentration limits. In the case of the long-lived nuclides, the Class A limits are 10% of the Class C limits. In the case of most of the short-lived nuclides, the Class A limits are vanishingly small fractions of the Class C limits (from 6E-4% to 5E-1% of the Class C limits). The rationale for using Class A limits rather than a predefined fraction of the Class C limits is based on the fact that, under the NRC waste classification scheme in 10 CFR 61.55 for disposal of low level waste, waste with concentrations below Class A limits have less rigorous disposal requirements and, if those requirements are met, pose an insignificant risk to the public and to workers.

The second, third, and fourth measures of radiological risk address the dose limits in the NRC performance objectives pertaining to public, environmental, intruder, and worker safety. In these cases, the bases for comparisons are set at 10% of the performance objectives dose limits, namely 10% of the public and environmental annual dose limit of 25 mrem (10 CFR 61.41); 10% of the recommended intruder annual dose limit of 500 mrem (*See* 10 CFR 61.42 and Section 5.2 of NUREG-0945, Vol. 1); and 10% of the worker annual dose limit of 5.0 rem (10 CFR 61.43).

In this analysis, comparisons were made between the total radionuclide inventories in untreated salt waste (or some fraction of the total inventories, when applicable and as explained below) and the inventory limits derived for each radiological measure identified above.

It is recognized that using the total radionuclide inventories as bases for the risk-based decisions produces conclusions reflective of average waste compositions, rather than the full range of waste compositions. Although this approach may seem non-conservative, it is assumed appropriate for this analysis for the following reasons: 1) the uncertainties of the total radionuclide inventories are significantly lower than the uncertainties of the inventories in individual waste tanks; 2) the total radionuclide inventories provide a good indication of the relative impacts of the various radionuclides; and 3) the risk measures are sufficiently conservative to accommodate most concentration fluctuations.

The methods for deriving the radionuclide inventory limits are provided in the section entitled Calculation of Inventory Limits.

Radionuclide Inventories in Untreated Salt Waste

Soluble and insoluble radionuclide inventories in untreated salt waste were identified based on the characterization data reported by Tran (2005). Soluble inventories of C-14, Na-22, and Al-26 were determined by summing the inventories reported for the precipitated salt phase (referred to by Tran as the insoluble salt phase) and the total free supernatant phase. Soluble inventories of all other radionuclides were those reported for the total free supernatant phase.

Insoluble inventories of all radionuclides were calculated based on the assumption that entrained sludge exists in salt feed at a concentration of 600 mg/L (600 mg/L is the design basis for the various salt treatment processes (d'Entremont and Drumm, 2005)). Based on a total projected salt feed volume of 107 Mgal (DOE, 2005) and a total estimated dry sludge mass of 2.9E+6 kg (WCS1.5, 2005), the

entrained sludge represents 8.4% of the total sludge. Consequently, the insoluble radionuclide inventories in salt were determined by multiplying 0.084 by the total dry sludge radionuclide inventories.

Soluble and insoluble inventories of alpha-emitting transuranic (TRU) nuclides were calculated by summing the respective soluble and insoluble phase inventories of Np-237, Pu-238, Pu-239, Pu-240, Pu-242, Am-241, Am-243, Cm-242, Cm-243, Cm-244, and Cm-245.

Total inventories of radionuclides were computed by the summing the soluble and insoluble inventories.

Calculation of Inventory Limits and Comparisons Against Radionuclide Inventories

Comparisons were made between the total radionuclide inventories in untreated salt waste and the total curie limits for the SDF as if NRC Class A concentration limits applied and under the assumption that radionuclide inventories were evenly distributed across the total volume of grout at the SDF. Specifically, the total curie limits for the SDF were determined by multiplying the Class A concentration limits by: a) the total projected grout volume ($6.4E+5$ cubic meters (DOE, 2005)) when the Class A concentration limit was given in units of curies per cubic meter; and b) the product of the total projected grout mass ($1.1E+12$ g, which assumes a grout density of 1.7 g/mL) and the activity conversion factor ($1E-9$ Ci/nCi), when the Class A concentration limit was given in units of nCi/g. Ratios of “the total curies in untreated salt waste” to “the Class A total curie limit” were then calculated and reported.

To indicate public, environmental, and intruder risk, comparisons were made between the average untreated radionuclide inventories in 9.6 Mgal of grout (Vault 4 capacity) and the applicable inventory limits for Vault 4. This was done under the assumption that the Vault 4 limits would be representative of the limits of other vaults of the same size. The average untreated inventories in 9.6 Mgal of grout were determined by multiplying the total radionuclide inventories by a factor of 0.057. This factor represents the proportion of the waste in 9.6 Mgal grout and is computed by dividing the Vault 4 volume (9.6 Mgal) by the total projected grout volume (168 Mgal, as reported by DOE in 2005). For the public and environmental risk, the inventory limits for a 9.6 Mgal vault were those values reported by Cook et. al. (2005) under the “all pathways” scenario, which assumes a public dose rate of 25 mrem/yr. Ratios of the “average radionuclide inventories in 9.6 Mgal grout” to the “Vault 4 public dose inventory limits” were then calculated and reported.

For the intruder risk, comparisons were made between the average untreated inventories in 9.6 Mgal of grout and the inventory limits for Vault 4 assuming a 500 mrem maximum annual intruder dose. In the intruder case, the inventory limits for Vault 4 were computed by multiplying by five the inventory limits reported by Cook et. al. (2005) under the “intruder” scenario, which assumes an intruder dose rate of 100 mrem/yr. Ratios of the “average radionuclide inventories in 9.6 Mgal grout” to the “Vault 4 intruder dose inventory limits” were then calculated and reported.

For the worker dose risk, two scenarios were considered, one focusing on worker gamma dose and the other focusing on radionuclides driving potential inhalation dose. For the worker gamma dose risk measure, comparisons were made between the total radionuclide inventories in untreated salt waste

(normalized for Cs-137/Ba-137m dose) and the SDF Cs-137/Ba-137m inventory limit based on a 5.0 rem maximum annual worker gamma dose. Note that the SDF worker gamma limit is defined as 0.2 curies Cs-137 per gallon of salt solution (Howell, 2005), based on a maximum annual worker gamma dose of 1.0 rem/yr. To convert this to an inventory limit representing a 5.0 rem maximum annual worker gamma dose, the facility concentration limit was multiplied by the total projected volume of salt feed (107 Mgal) and a factor of five (5.0 rem/1.0 rem). This results in a total facility limit of 1.1E+8 curies Cs-137. For comparisons against this limit, the inventories of photon-emitting radionuclides in the untreated salt waste were normalized for Cs-137/Ba-137m dose, by multiplying each radionuclide inventory by the ratio of the radionuclide's photon dose constant to the Cs-137/Ba-137m photon dose constant (this ratio is referred to as the dose normalization factor). Photon dose constants used in these calculations were those reported by Unger and Trubey (1982). In cases where the nuclide had short-lived decay products (cases such as Ru-106, Sn-126, Sb-125, Ce-144, Np-237), the photon dose contributions of the decay products were taken into account. For example, in the case of Sn-126, the overall photon dose constant was the sum of the Sn-126 constant, the Sb-126m dose constant, and 14% of the Sb-126 constant (this takes into account that 100% of the Sn-126 disintegrations produce Sb-126m and 14% of the Sb-126m disintegrations produce Sb-126). Ratios of the "normalized radionuclide inventories" to the "worker gamma dose inventory limit" were then calculated and reported.

For the worker inhalation dose, the primary radionuclides contributing to the total potential inhalation dose were identified. This was accomplished by: a) multiplying the radionuclide inventories in untreated salt waste by the nuclide-specific inhalation dose factors to identify the total potential inhalation dose associated with each nuclide; b) summing the contributions of all the total potential inhalation doses; and c) computing the percentage of the cumulative potential inhalation dose associated with each nuclide. Radionuclides contributing one percent or more of the cumulative potential inhalation dose were considered to be the primary drivers of the potential inhalation dose. The inhalation dose factors used in these calculations were those reported by EPA (1988), converted from units of Sv/Bq to units of rem/Ci by multiplying the Sv/Bq values by factors of $3.7E+12$ rem-Bq/Sv-Ci. Note that this risk measure does not address the significance of worker inhalation dose, however, it assures that the primary worker inhalation risk drivers are identified.

Calculation of Radionuclide Removal Efficiencies

Assumptions regarding removal efficacies of the various processes are given in the Results and Discussion section, along with flow schemes and process descriptions for each treatment scheme. In every case, removal efficiencies were calculated by applying the appropriate treatment assumptions to the expected soluble and insoluble constituent phases. The bases for the soluble-insoluble distributions are the soluble and insoluble inventories identified for the untreated salt waste (Table 1).

Nominal⁶ removal efficiencies were calculated based on the exact soluble-insoluble distributions given in Table 1 and the nominal decontamination assumptions. Lower and upper bounds of the removal

⁶ Nominal values throughout this report are those approximate values normally expected to be achieved. Actual values may vary around this nominal value depending upon the unique characteristics of each specific batch.

efficiencies were calculated based on conditions where the soluble-insoluble distributions increased and decreased by a factor of four (a total factor of sixteen), and lower and upper bounds of the decontamination assumptions were applied, when available. In cases where bounding decontamination assumptions were not available, lower and upper bounding removal efficiencies were calculated based on nominal decontamination assumptions applied to lower and upper bounding soluble-insoluble distributions. Note that in several cases, the nominal decontamination factors were chosen conservatively and actually represent lower bounding decontamination factors.

Removal efficiencies for alpha-emitting TRU were identified based on weighted averages of the individual removal efficiencies of Pu-238, Am-241, Cm-244, and Pu-239. These four nuclides contribute approximately 99% of the TRU alpha activity and therefore are considered the primary nuclides impacting TRU alpha removal.

Results and Discussion

Radionuclide Inventories in Untreated Salt Waste

Soluble, insoluble, and total radionuclide inventories in untreated salt waste are given in Table 1. Soluble radionuclides are those radionuclides present as dissolved ions in supernatant solution or as precipitated salts that will dissolve upon addition of water. Insoluble radionuclides are those radionuclides present in the form of entrained sludge. Note that for a given radionuclide, the soluble-insoluble distribution is an important factor determining treatment requirements for effective removal. Soluble constituents typically require chemically-specific removal technologies tailored to the particular constituent. Insoluble constituents are typically removed through physical methods such as filtration or centrifugation.

Based on the information in Table 1, it can be seen that the soluble-insoluble distribution varies considerably as a function of radionuclide, as would be expected based upon the chemical differences of the various constituents. About 20% of the radionuclides exist primarily as soluble constituents, 40% exist primarily as insoluble constituents, and 40% exist with soluble and insoluble contributions of the same order of magnitude. This suggests about 20% of the isotopes would be relatively unaffected by a physical separation process (such as filtration), 40% would be highly affected by physical separation, and 40% would be moderately affected by physical separation.

Based on the total radionuclide inventories, Cs-137 is clearly the predominant radionuclide from an activity perspective. (It should be noted that Ba-137m, the short-lived decay product of Cs-137, is also a predominant radionuclide. Ba-137m is not included in Table 1, but is taken into account when calculating inventory limits for Cs-137 shown in Tables 2 through 5). As seen in the table, activities of all other radionuclides (except for Ba-137m) are one or more orders of magnitude below that of Cs-137.

Also of note are the primary radionuclides contributing to alpha-emitting TRU activity. Approximately 82% of the alpha-emitting TRU inventory is Pu-238, 11% is Am-241, 4% is Cm-244, and 3% is Pu-239. All other alpha-emitting TRU nuclides contribute minimally to the TRU inventory.

Comparisons Based on Class A Concentration Limits

Table 2 gives: a) the radionuclide inventories in untreated salt waste, b) the NRC Class A inventory limits for the total projected grout volume, and c) the ratios of the inventories to the limits. Based on the ratios, it is clear that the inventories of Sr-90, Cs-137, and alpha-emitting TRU in untreated salt waste are significantly greater than the inventory limits. Specifically, the inventory of Sr-90 is 290 times the limit, the inventory of Cs-137 is 170 times the limit, and the inventory of alpha-emitting TRU is 25 times the limit. In contrast, the inventories of all other nuclides are clearly less than the inventory limits (by a factor of three or more).

Comparisons Based on the Public Dose Limit

Table 3 gives: a) the average radionuclide inventories projected for a 9.6 Mgal volume of grout (the Vault 4 grout volume); b) the Vault 4 inventory limits based on a 25 mrem/yr public dose limit, and c) the ratios of the average inventories to the inventory limits. For all nuclides, the ratios of the average inventories to the inventory limits are significantly less than 0.10 (by one or more orders of magnitude), suggesting no nuclides pose a significant risk from the public dose perspective.

Comparisons Based on Intruder Dose

Table 4 gives: a) the average radionuclide inventories projected for a 9.6 Mgal volume of grout (the Vault 4 grout volume); b) the Vault 4 inventory limits based on a 500 mrem/yr intruder dose limit, and c) the ratios of the average inventories to the inventory limits. Based on the results, it is clear that Cs-137 is the only nuclide with a ratio greater than 0.10. Specifically, the ratio for Cs-137 is 0.21, approximately twice the intruder dose risk measure, thus suggesting that without treatment, Cs-137 may pose a significant risk to a hypothetical intruder. All other nuclides have ratios significantly less than 0.10 (by two or more orders of magnitude).

Comparisons Based on Worker Dose

Results of the comparisons based on the gamma exposure and inhalation dose risk measures are given in Table 5A and 5B, respectively.

Table 5A gives: a) the radionuclide inventories in untreated salt waste, b) the Cs-137/Ba-137m dose normalization factors, and c) the ratios of the normalized inventories to the Cs-137/Ba-137m inventory limit, based on a worker gamma dose rate of 5.0 rem/yr. From the results, it is clear that Cs-137 is the only nuclide with a ratio greater than 0.10. Specifically, the ratio for Cs-137 is 1.0, tens times the worker gamma dose risk measure. All other nuclides have ratios significantly less than 0.10 (by one or more orders of magnitude).

Table 5B gives: a) the radionuclide inventories in untreated salt waste, b) the inhalation dose factors, and c) the percentages of the total potential inhalation dose due to each nuclide. From the results, it is clear that Sr-90, Cs-137, and the four primary alpha-emitting TRU nuclides (Pu-238, Pu-239, Am-241, and Cm-244) contribute the majority of the potential inhalation dose. Specifically, the percentages of the inhalation doses contributed by these nuclides are approximately 7, 3, 73, 3, 10, and 2, respectively. Collectively, these nuclides contribute approximately 98% of the total potential inhalation dose. Each other nuclide contributes less than one percent of the total potential inhalation dose, with contributions from most of the nuclides being two or more orders of magnitudes lower than one percent. Clearly, Pu-238 is the largest potential contributor of inhalation dose (73% of the total potential inhalation dose), with Am-241 and Sr-90 being the second and third largest contributors (10 and approximately 7%, respectively), and Cs-137, Pu-239, and Cm-244 being minor contributors (approximately 3, 3, and 2%, respectively). Based on the results, it is clear that the nuclides most important from a worker inhalation dose perspective are the same nuclides identified by the other radiological risk measures.

Summary

In summary and as reflected in Table 6, Sr-90 and four alpha-emitting TRU nuclides (Pu-238, Am-241, Cm-244, and Pu-239) have inventories exceeding the inventory limits based on NRC Class A concentration limits and are important worker inhalation dose drivers. Cs-137 has an inventory exceeding the inventory limits based on the NRC Class A concentration limit, 10% of the intruder dose limit, 10% of the worker gamma dose limit, and is an important worker inhalation dose driver. All other radionuclides in untreated salt waste have inventories below the inventory limits based on the NRC Class A concentration limits, 10% of the recommended intruder dose limit, 10% of the worker gamma dose limit, and are not an important worker inhalation dose drivers

Table 1
Soluble, Insoluble, and Total Curie Inventories in Untreated Salt Waste

Radionuclide	Soluble Curies in Untreated Salt Waste	Insoluble Curies in Untreated Salt Waste	Total Curies in Untreated Salt Waste
H-3	9.4E+3	Negligible	9.4E+3
C-14	5.2E+2	2.8E-1	5.2E+2
Na-22	5.1E+3	9.3E-1	5.1E+3
Al-26	2.4E+1	4.9E-1	2.4E+1
Co-60	8.6E+1	2.9E+4	2.9E+4
Ni-59	2.4E0	2.2E+2	2.2E+2
Ni-63	2.1E+2	1.9E+4	1.9E+4
Se-79	8.9E+1	1.3E+2	2.2E+2
Sr-90	2.8E+4	7.3E+6	7.3E+6
Nb-94	7.0E-4	5.9E-2	6.0E-2
Tc-99	3.3E+4	2.2E+3	3.5E+4
Ru-106	2.3E+3	5.7E+2	2.9E+3
Sn-126	4.5E+2	1.7E+2	6.2E+2
Sb-125	9.2E+3	1.5E+4	2.4E+4
I-129	1.8E+1	9.0E-3	1.8E+1
Cs-134	2.3E+5	8.8E+2	2.3E+5
Cs-135	3.9E+2	1.5E0	3.9E+2
Cs-137	1.1E+8	4.4E+5	1.1E+8
Ce-144	5.9E0	5.0E+2	5.1E+2
Pm-147	3.8E+3	3.2E+5	3.2E+5
Sm-151	4.3E+3	3.6E+5	3.6E+5
Eu-152	2.1E+1	1.7E+3	1.7E+3
Eu-154	9.1E+2	7.6E+4	7.7E+4
Eu-155	2.4E+2	2.0E+4	2.0E+4
Th-232	1.0E-1	2.4E-1	3.4E-1
U-232	2.9E-2	4.7E-2	7.6E-2
U-233	2.7E0	8.3E0	1.1E+1
U-234	4.2E0	3.2E0	7.4E0
U-235	8.4E-2	1.3E-1	2.1E-1
U-236	3.6E-1	5.2E-1	8.8E-1
U-238	6.8E0	5.5E0	1.2E+1
Np-237	4.2E0	8.3E0	1.3E+1
Pu-238	5.7E+4	1.7E+5	2.3E+5
Pu-239	3.4E+3	4.0E+3	7.4E+3
Pu-240	9.1E+2	1.7E+3	2.6E+3
Pu-241	3.8E+4	1.0E+5	1.4E+5
Pu-242	9.4E-1	2.8E0	3.7E0
Am-241	3.6E+2	3.0E+4	3.0E+4
Am-242m	2.1E-1	1.8E+1	1.8E+1
Am-243	7.0E-2	5.8E0	5.9E0
Cm-242	1.7E-1	1.5E+1	1.5E+1
Cm-243	4.1E-2	3.4E0	3.4E0
Cm-244	1.5E+2	1.2E+4	1.2E+4
Cm-245	1.4E-2	1.2E0	1.2E0
α-emitting TRU	6.2E+4	2.2E+5	2.8E+5

Table 2
 Comparison of Inventories in Solidified Untreated Salt Waste
 With Inventory Limits Based on NRC Class A Concentration Limits

Radionuclide	Total Curies in Untreated Salt Waste	Class A Limit, Curies Per Total Projected Grout Volume	Ratio of "Total Curies in Untreated Salt Waste" to "Class A Limit"
H-3	9.4E+3	2.5E+7	3.8E-4
C-14	5.2E+2	5.1E+5	1.0E-3
Co-60	2.9E+4	4.5E+8	6.4E-5
Ni-63	1.9E+4	2.2E+6	8.6E-3
Sr-90	7.3E+6	2.5E+4	2.9E+2
Tc-99	3.5E+4	1.9E+5	1.8E-1
I-129	1.8E+1	5.1E+3	3.3E-3
Cs-137	1.1E+8	6.4E+5	1.7E+2
Pu-241	1.4E+5	3.8E+5	3.7E-1
Cm-242	1.5E+1	2.2E+6	6.8E-6
α-emitting TRU	2.8E+5	1.1E+4	2.5E+1

Table 3
Comparison of Inventories in Solidified Untreated Salt Waste and Inventory Limits
Based on a Public Dose Rate Limit of 25 mrem/yr

Radionuclide	Average Untreated Curies in 9.6 Mgal of Grout (Vault 4 Volume)	Vault 4 Curie Limit Based on a Maximum Public Dose Rate of 25 mrem/yr	Ratio of "Average Untreated Curies in 9.6 Mgal Grout" to "Vault 4 Curie Limit Based on a 25 mrem/yr Public Dose Rate"
H-3	5.4E+2	1.3E+12	4.2E-10
C-14	3.0E+1	1.1E+8	2.7E-7
Al-26	1.4E0	2.3E+10	6.1E-11
Co-60	1.7E+3	> 1.0E+20	< 1.7E-17
Ni-59	1.3E+1	1.6E+19	8.1E-19
Se-79	1.3E+1	1.0E+3	1.3E-2
Sr-90	4.2E+5	1.4E+17	3.0E-12
Nb-94	3.4E-3	7.0E+17	4.9E-21
Tc-99	2.0E+3	1.1E+17	1.8E-14
Sn-126	3.5E+1	2.9E+19	1.2E-18
Sb-125	1.4E+3	> 1.0E+20	< 1.4E-17
I-129	1.0E0	4.0E+3	2.5E-4
Cs-134	1.3E+4	> 1.0E+20	< 1.3E-16
Cs-135	2.2E+1	> 1.0E+20	< 2.2E-19
Cs-137	6.3E+6	> 1.0E+20	< 6.3E-14
Eu-152	9.7E+1	> 1.0E+20	< 9.7E-19
Eu-154	4.4E+3	> 1.0E+20	< 4.4E-17
Eu-155	1.1E+3	> 1.0E+20	< 1.1E-17
Th-232	1.9E-2	> 1.0E+20	< 1.9E-22
U-232	4.3E-3	> 1.0E+20	< 4.3E-23
U-233	6.3E-1	> 1.0E+20	< 6.3E-21
U-234	4.2E-1	> 1.0E+20	< 4.2E-21
U-235	1.2E-2	> 1.0E+20	< 1.2E-22
U-236	5.0E-2	> 1.0E+20	< 5.0E-22
U-238	6.9E-1	> 1.0E+20	< 6.9E-21
Np-237	7.4E-1	8.9E+18	8.3E-20
Pu-238	1.3E+4	> 1.0E+20	< 1.3E-16
Pu-239	4.2E+2	> 1.0E+20	< 4.2E-18
Pu-240	1.5E+2	> 1.0E+20	< 1.5E-18
Pu-241	8.0E+3	> 1.0E+20	< 8.0E-17
Pu-242	2.1E-1	> 1.0E+20	< 2.1E-21
Am-241	1.7E+3	> 1.0E+20	< 1.7E-17
Am-242m	1.0E0	> 1.0E+20	< 1.0E-20
Am-243	3.4E-1	> 1.0E+20	< 3.4E-21
Cm-243	1.9E-1	> 1.0E+20	< 1.9E-21
Cm-244	6.9E+2	> 1.0E+20	< 6.9E-18
Cm-245	6.9E-2	> 1.0E+20	< 6.9E-22

Table 4
 Comparison of Inventories in Solidified Untreated Salt Waste and Inventory Limits
 Based on a Resident Intruder Dose Rate Limit of 500 mrem/yr

Radionuclide	Average Untreated Curies in 9.6 Mgal of Grout (Vault 4 Volume)	Vault 4 Curie Limit Based on an Intruder Dose Rate of 500 mrem/yr	Ratio of "Average Untreated Curies in 9.6 Mgal Grout" to "Vault 4 Curie Limit Based on a 500 mrem/yr Intruder Dose Rate"
H-3	5.4E+2	> 5.0E+20	< 1.1E-18
C-14	3.0E+1	> 5.0E+20	< 6.0E-20
Al-26	1.4E0	8.0E+2	1.8E-3
Co-60	1.7E+3	2.9E+10	5.9E-8
Ni-59	1.3E+1	> 5.0E+20	< 2.6E-20
Se-79	1.3E+1	> 5.0E+20	< 2.6E-20
Sr-90	4.2E+5	> 5.0E+20	< 8.4E-16
Nb-94	3.4E-3	5.0E+3	6.8E-7
Tc-99	2.0E+3	1.9E+14	1.1E-11
Sn-126	3.5E+1	6.0E+3	5.8E-3
Sb-125	1.4E+3	7.0E+17	2.0E-15
I-129	1.0E0	> 5.0E+20	< 2.0E-21
Cs-134	1.3E+4	2.1E+20	6.2E-17
Cs-135	2.2E+1	> 5.0E+20	< 4.4E-20
Cs-137	6.3E+6	3.0E+7	2.1E-1
Eu-152	9.7E+1	3.2E+7	3.0E-6
Eu-154	4.4E+3	6.0E+8	7.3E-6
Eu-155	1.1E+3	5.5E+19	2.0E-17
Th-232	1.9E-2	8.0E+2	2.4E-5
U-232	4.3E-3	4.5E+4	9.6E-8
U-233	6.3E-1	7.0E+4	9.0E-6
U-234	4.2E-1	2.3E+4	1.8E-5
U-235	1.2E-2	5.0E+5	2.4E-8
U-236	5.0E-2	1.6E+9	3.1E-11
U-238	6.9E-1	3.3E+5	2.1E-6
Np-237	7.4E-1	3.4E+5	2.2E-6
Pu-238	1.3E+4	6.5E+7	2.0E-4
Pu-239	4.2E+2	7.0E+10	6.0E-9
Pu-240	1.5E+2	1.5E+13	1.0E-11
Pu-241	8.0E+3	5.0E+10	1.6E-7
Pu-242	2.1E-1	2.5E+11	8.4E-13
Am-241	1.7E+3	1.7E+9	1.0E-6
Am-242m	1.0E0	4.9E+7	2.0E-8
Am-243	3.4E-1	1.5E+6	2.3E-7
Cm-243	1.9E-1	3.5E+10	5.4E-12
Cm-244	6.9E+2	5.5E+15	1.3E-13
Cm-245	6.9E-2	4.2E+7	1.6E-9

Table 5A
 Comparison of Inventories in Untreated Salt Waste and Facility Curie Limit
 Based on a Worker Gamma Dose Rate of 5.0 rem/yr

Radionuclide	Total Curies in Untreated Salt Waste	Cs-137/Ba-137m Dose Normalization Factor	Ratio of "Normalized Total Curies in Untreated Salt Waste" to "Cs-137 Curie Limit Based on a 5.0 rem/yr Worker Gamma Dose Rate"
Al-26	2.4E+1	3.9E0	8.7E-7
Co-60	2.9E+4	3.6E0	9.8E-4
Nb-94	6.0E-2	2.6E0	1.5E-9
Tc-99	3.5E+4	1.2E-6	3.9E-10
Ru-106	2.9E+3	3.6E-1	9.8E-6
Sn-126	6.2E+2	3.3E-1	1.9E-6
Sb-125	2.4E+4	9.9E-1	2.2E-4
I-129	1.8E+1	3.3E-1	5.6E-8
Cs-134	2.3E+5	2.6E0	5.6E-3
Cs-137	1.1E+8	1.0E0	1.0E0
Ce-144	5.1E+2	6.1E-2	2.9E-7
Pm-147	3.2E+5	7.0E-6	2.1E-8
Sm-151	3.6E+5	2.4E-4	8.1E-7
Eu-154	7.7E+4	2.0E0	1.4E-3
Eu-155	2.0E+4	1.7E-1	3.2E-5
Th-232	3.4E-1	1.8E-1	5.7E-10
U-232	7.6E-2	2.3E-1	1.6E-10
U-233	1.1E+1	7.6E-2	7.8E-9
U-234	7.4E0	2.0E-1	1.4E-8
U-235	2.1E-1	8.9E-1	1.7E-9
U-236	8.8E-1	1.9E-1	1.6E-9
U-238	1.2E+1	1.7E-1	1.9E-8
Np-237	1.3E+1	1.2E0	1.5E-7
Pu-238	2.3E+5	2.1E-1	4.5E-4
Pu-239	7.4E+3	7.9E-2	5.5E-6
Pu-240	2.6E+3	2.0E-1	4.9E-6
Pu-242	3.7E0	1.6E-1	5.5E-9
Am-241	3.0E+4	8.2E-1	2.3E-4
Am-242m	1.8E+1	4.8E-1	8.1E-8
Am-243	5.9E0	8.2E-1	4.5E-8
Cm-242	1.5E+1	2.4E-3	3.4E-10
Cm-244	1.2E+4	1.7E-1	1.9E-5
Cm-245	1.2E0	1.2E0	1.3E-8

Table 5B
Inhalation Dose Contributions in Untreated Salt Waste

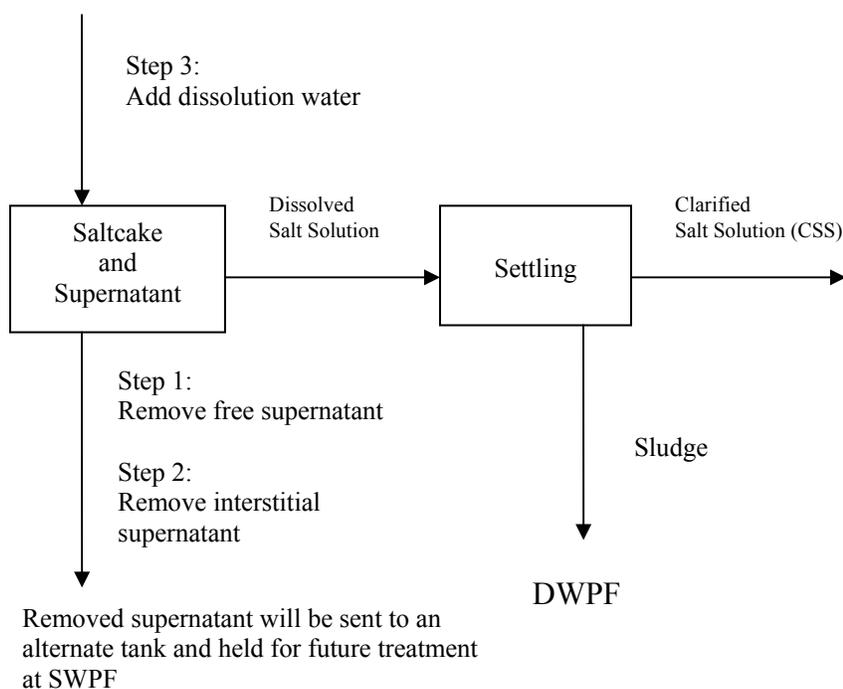
Radionuclide	Total Curies in Untreated Salt Waste	Inhalation Dose Factor, rem/Ci	% of Total Potential Inhalation Dose in Untreated Salt Waste
H-3	9.4E+3	6.3E+1	4.6E-7
C-14	5.2E+2	2.4E+1	9.3E-9
Na-22	5.1E+3	7.8E+3	3.1E-5
Al-26	2.4E+1	8.1E+4	1.5E-6
Co-60	2.9E+4	2.2E+5	5.0E-3
Ni-59	2.2E+2	2.7E+3	4.6E-7
Ni-63	1.9E+4	6.3E+3	9.3E-5
Se-79	2.2E+2	1.0E+4	1.7E-6
Sr-90	7.3E+6	1.3E+6	7.4E0
Nb-94	6.0E-2	4.1E+5	1.9E-8
Tc-99	3.5E+4	8.5E+3	2.3E-4
Ru-106	2.9E+3	4.8E+5	1.1E-3
Sn-126	6.2E+2	1.0E+5	4.8E-5
Sb-125	2.4E+4	1.2E+4	2.2E-4
I-129	1.8E+1	1.7E+5	2.4E-6
Cs-134	2.3E+5	4.8E+4	8.5E-3
Cs-135	3.9E+2	4.4E+3	1.3E-6
Cs-137	1.1E+8	3.2E+4	2.7E0
Ce-144	5.1E+2	3.7E+5	1.5E-4
Pm-147	3.2E+5	4.1E+4	1.0E-2
Sm-151	3.6E+5	3.0E+4	8.5E-3
Eu-152	1.7E+3	2.2E+5	2.9E-4
Eu-154	7.7E+4	2.8E+5	1.7E-2
Eu-155	2.0E+4	4.1E+4	6.4E-4
Th-232	3.4E-1	1.6E+9	4.2E-4
U-232	7.6E-2	6.7E+8	4.0E-5
U-233	1.1E+1	1.4E+8	1.2E-3
U-234	7.4E0	1.3E+8	7.4E-4
U-235	2.1E-1	1.2E+8	1.9E-5
U-236	8.8E-1	1.2E+8	8.5E-5
U-238	1.2E+1	1.2E+8	1.1E-3
Np-237	1.3E+1	5.6E+8	5.7E-3
Pu-238	2.3E+5	4.1E+8	7.3E+1
Pu-239	7.4E+3	4.4E+8	2.6E0
Pu-240	2.6E+3	4.4E+8	8.5E-1
Pu-241	1.4E+5	8.1E+6	8.5E-1
Pu-242	3.7E0	4.1E+8	1.2E-3
Am-241	3.0E+4	4.4E+8	1.0E+1
Am-242m	1.8E+1	4.4E+8	6.1E-3
Am-243	5.9E0	4.4E+8	2.0E-3
Cm-242	1.5E+1	1.7E+7	2.0E-4
Cm-243	3.4E0	3.1E+8	8.5E-4
Cm-244	1.2E+4	2.5E+8	2.3E0
Cm-245	1.2E0	4.4E+8	4.1E-4

Table 6
Summary of Risk Comparison for Radionuclides in Untreated Salt Waste

Radionuclide	Technical Basis			
	Exceeds Class A	> 10% Intruder Limit	> 10% Worker Gamma Limit	Drives Worker Inhalation Dose
Sr-90	X			X
Cs-137	X	X	X	X
Alpha-emitting TRU ⁷	X			X

Planned Treatment Processes and Projected Removal Efficiencies

Deliquification, Dissolution, and Adjustment (DDA):



The DDA process relies on two removal mechanisms, removal of supernatant fluid through pumping/draining and removal of suspended solids (sludge) through gravity settling/clarification. As shown in Step 1 of the flow diagram, the DDA process is initiated when free supernatant solution (supernatant above saltcake) is pumped from the tank. During Step 2, interstitial supernatant fluid is drained/removed from the saltcake after a well is generated through the saltcake. All fluid removed is sent to an alternate tank for future treatment at the SWPF. In Step 3, the saltcake is dissolved and transferred to a settling tank. Following a settling period, the clarified salt solution (CSS) is decanted

⁷ Pu-238, Am-241, Cm-244, and Pu-239.

out of the tank and dispositioned to SPF. In the future, the settled solids will ultimately be removed from the tank and dispositioned to DWPF. Note that early batches of CSS containing elevated Cs-137 concentrations will undergo further treatment via ARP/MCU before being dispositioned to SPF.

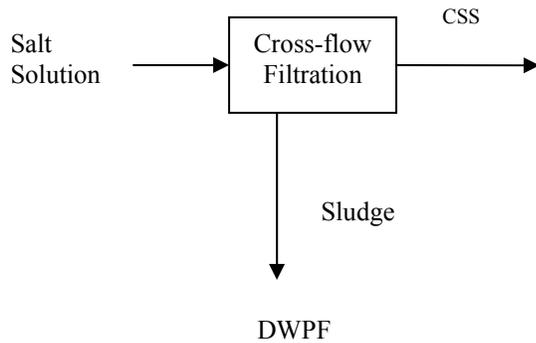
In determining the overall removal efficiencies of DDA, the following individual removal efficiencies are assumed. Deliquification typically removes 50% of the supernatant solution (Shah and Hopkins, 2004), with a lower bound of 30% and an upper bound of 70% (these assumptions incorporate the lessons learned from Tank 41 salt). For a thirty day period, gravity settling typically removes two-thirds of the suspended solids (Gillam, 2005), with a lower bound of 50% and an upper bound of 80%. Given the magnitude of these variabilities, the uncertainty of the overall removal efficiency is typically $\pm 20\%$, regardless of the soluble/insoluble distribution.

Radionuclide removal efficiencies are given in Table 7. Nominal removal efficiencies range from 50% to 66%, depending on the solubility of the constituent (50% for highly soluble constituents; 66% for highly insoluble constituents). For Sr-90, the nominal removal efficiency is 66%, with a lower bound of 46% and an upper bound of 86%. For Cs-137, the nominal removal efficiency is 50%, with a lower bound of 30% and an upper bound of 70%. For Pu-238, the nominal removal efficiency is 63%, with a lower bound of 43% and an upper bound of 83%. For Am-241, the nominal removal efficiency is 66%, with a lower bound of 46% and an upper bound of 86%. For Cm-244, the nominal removal efficiency is 66%, with a lower bound of 46% and an upper bound of 86%. For Pu-239, the nominal removal efficiency is 59%, with a lower bound of 39% and an upper bound of 79%. For alpha-emitting TRU, the nominal removal efficiency is 63%, with a lower bound of 43% and an upper bound of 83%.

Table 7
Radionuclide Removal Using DDA

Radionuclide	Projected Removal Efficiency, %		
	Nominal	Lower Bound	Upper Bound
Sr-90	66	46	86
Cs-137	50	30	70
Pu-238	63	43	83
Am-241	66	46	86
Cm-244	66	46	86
Pu-239	59	39	79
α -emitting TRU	63	43	83

Actinide Removal Process (ARP) without Monosodium Titanate (MST):



The ARP process (without MST sorption) relies on one removal mechanism, removal of suspended solids (sludge) by cross-flow filtration. Removed solids are dispositioned to DWPf. CSS is dispositioned to SPF.

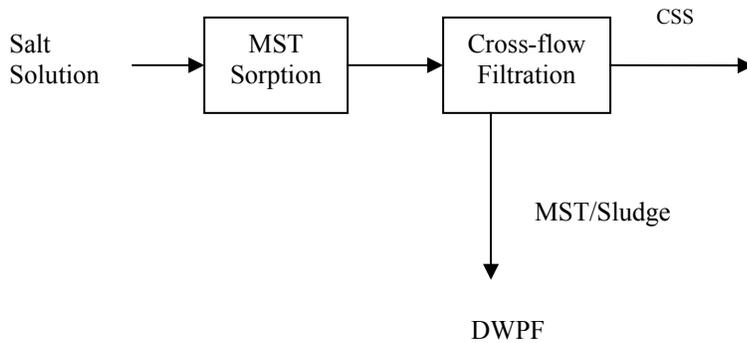
Cross-flow filtration is assumed to nominally remove 100% of the suspended solids, although it is recognized that actual removal will be slightly lower. A lower bound of 99.5% removal is assumed, based on industrial filtration experience.

Radionuclide removal efficiencies are given in Table 8. For Sr-90, the removal efficiencies are high (98.0 – 99.9%), due to the low solubility which makes particulate removal significant. In contrast, for Cs-137, the removal efficiencies are negligible, due to the high solubility which makes particulate removal insignificant. For alpha-emitting TRU, the range of removal efficiencies is relatively broad (51 – 93%), reflecting the wide variation of solubility for the individual radionuclides comprising the alpha-emitting TRU category.

Table 8
Radionuclide Removal Using ARP without MST

Radionuclide	Projected Removal Efficiency, %		
	Nominal	Lower Bound	Upper Bound
Sr-90	99.6	98.0	99.9
Cs-137	~ 0	~ 0	~ 0
Pu-238	75	43	92
Am-241	98.8	94.9	99.7
Cm-244	98.8	94.8	99.7
Pu-239	54	23	82
α-emitting TRU	78	50	93

ARP with MST:



The ARP process (with MST sorption) relies on two removal mechanisms, removal of soluble constituents by MST sorption and removal of suspended solids (MST and sludge) by cross-flow filtration. Removed solids are dispositioned to DWPF. CSS is dispositioned to SPF.

Duration of the MST strike is assumed to be 24 hours. This is the same assumption used in a separate analysis performed for the SWPF and allows for consistent comparison. Assumed MST decontamination factors (DFs) are given in the table below. Nominal DFs are those reported by d’Entremont (2005) for a twenty four hour duration strike. Lower and upper bounding DFs are those reported by Le (2005) under conditions of four to twenty four hour duration strikes. Assumptions regarding efficiency of the cross-flow filter are the same as in the previous case (ARP without MST).

Constituent	ARP MST Soluble Phase Decontamination Factor		
	Nominal	Lower Bound	Upper Bound
Strontium	130	20	130
Cesium	0	0	0
Plutonium	13	5.5	13
Americium	1.7	1.0	4.6
Curium	1.7	1.0	1.7

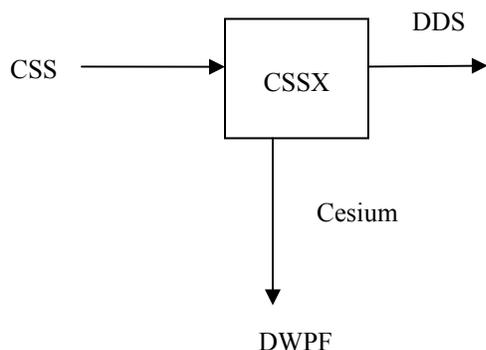
Radionuclide removal efficiencies are given in Table 9. For Sr-90, the removal efficiencies are extremely high (99.4 – 99.999%), due to a) the very low solubility of strontium that makes particulate removal significant and b) the very high removal efficiency of MST for soluble phase strontium. For Cs-137, the removal efficiencies are negligible due to a) the high solubility of cesium that makes particulate removal insignificant and b) the inability of MST to sorb soluble phase cesium. For alpha-emitting TRU, the removal efficiencies are relatively high (96 – 99%), due to the combination of low

solubility and reasonably high soluble phase removal. Clearly, the combination of MST and cross-flow filtration is an effective treatment for Sr-90 and alpha-emitting TRU nuclides.

Table 9
Radionuclide Removal Using ARP with MST

Radionuclide	ARP with MST Projected Removal Efficiency, %		
	Nominal	Lower Bound	Upper Bound
Sr-90	99.997	99.4	99.999
Cs-137	~ 0	~ 0	~ 0
Pu-238	98.1	89.4	99.9
Am-241	99.3	94.9	99.9
Cm-244	99.3	94.8	99.8
Pu-239	96.4	85.8	98.6
α -emitting TRU	98.1	90.1	99.9

Modular Caustic Side Solvent Extraction Unit (MCU):

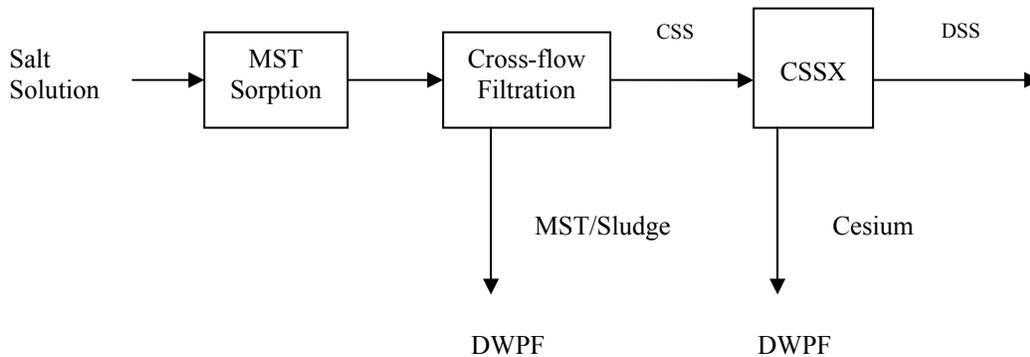


The MCU process relies on one removal mechanism, removal of soluble phase cesium by liquid-liquid extraction utilizing the Caustic Side Solvent Extraction (CSSX) technology. In this process, CSS is the feed stream and the effluents include a concentrated cesium stream that is dispositioned to DWPF and a decontaminated salt solution (DSS) that is dispositioned to SPF.

For MCU, a DF of 12 is assumed for soluble phase cesium (d’Entremont, 2005). For Sr-90 and alpha-emitting TRU nuclides, the MCU removal efficiency is assumed to be zero.

The nominal removal efficiency for Cs-137 is 91%, with a lower bound of 90% and an upper bound of 92%.

Salt Waste Processing Facility (SWPF) Treatment:



The SWPF treatment process relies on three removal mechanisms: 1) removal of soluble constituents by MST sorption; 2) removal of suspended solids by cross-flow filtration; and 3) removal of cesium by liquid-liquid extraction utilizing CSSX. In this process, salt solution is first treated with MST and then filtered to produce a CSS that is subsequently treated with CSSX. The removed solids and the concentrated cesium streams are dispositioned to DWPf, and the DSS stream is dispositioned to SPF.

Duration of the MST strike is assumed to be 12 hours (Parsons, 2004). Assumed DFs for the MST treatment are given in the table below. Nominal MST DFs are those reported by d’Entremont (2005) for a twelve hour duration strike. Lower and upper bounding MST DFs are those reported by Le (2005) under conditions of four to twenty four hour duration strikes. Assumptions regarding efficiency of the cross-flow filter are the same as in the previous ARP cases. For SWPF, the CSSX DF for soluble phase cesium is assumed to be 40,000 (d’Entremont, 2005).

Constituent	SWPF MST Soluble Phase Decontamination Factor		
	Nominal	Lower Bound	Upper Bound
Strontium	20	20	130
Cesium	0	0	0
Plutonium	5.5	5.5	13
Americium	4.6	1.0	4.6
Curium	1.0	1.0	1.7

Radionuclide removal efficiencies are given in Table 10. For Sr-90, the removal efficiencies are very high (99.4 to 99.999%), due to the combination of effective particulate removal and high soluble phase decontamination. For Cs-137, the removal efficiencies are extremely high (99.990 to 99.998%, respectively), due to the extremely high removal efficiency of CSSX for soluble phase cesium. For alpha-emitting TRU, the removal efficiencies are high (91 to 99%), although lower than those of Sr-90

and Cs-137. Clearly, the SWPF treatments offer an effective means of removing Sr-90, Cs-137, and alpha-emitting TRU nuclides.

Table 10
Radionuclide Removal Using SWPF

Radionuclide	SWPF Projected Removal Efficiency, %		
	Nominal	Lower Bound	Upper Bound
Sr-90	99.98	99.4	99.999
Cs-137	99.998	99.990	99.998
Pu-238	95.5	89.4	99.4
Am-241	99.7	94.9	99.94
Cm-244	98.8	94.8	99.8
Pu-239	91.6	85.8	98.6
α -emitting TRU	96	90	99.5

Conclusions

1) Cs-137 in solidified untreated salt waste is a primary driver of intruder, worker gamma, and worker inhalation dose risks.

2) Sr-90 and four alpha-emitting TRU nuclides (Pu-238, Am-241, Cm-244, and Pu-239) in solidified untreated salt waste are primary drivers of worker inhalation dose risk.

3) Other than Sr-90, Cs-137 and the four alpha-emitting TRU nuclides, no nuclides in solidified untreated salt waste pose a significant risk to the public, worker, or environment.

4) For removal of strontium and the alpha-emitting TRU nuclides, the relative efficacies of the planned treatments are:

ARP with MST \approx SWPF > ARP without MST > DDA >> MCU

5) For removal of cesium, the relative efficacies of the planned treatments are:

SWPF > MCU > DDA >> ARP

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