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Cost Benefit of Caustic Recycle for Tank Waste Remediation at the Hanford and Savannah River Sites

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Title: *Cost Benefit of Caustic Recycle for Tank Waste
Remediation at the Hanford and Savannah
River Sites*

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SUMMARY

The potential cost savings due to the use of caustic recycle used in conjunction with remediation of radioactive underground storage tank waste, is shown in Figure 1 for the Hanford and Savannah River sites. As shown in Figure 1, two cost savings estimates for each case have been made for Hanford, and one cost savings estimate for each case has been made for the Savannah River site. This is due to the Hanford site remediation effort being less mature than that of Savannah River; and consequently, a range of cost savings being more appropriate for Hanford. This range of cost savings (rather than a single value) for each case at Hanford is due to cost uncertainties related to the LAW immobilization operation. Caustic recycle Case-1 has been defined as the sodium required to meet all identified caustic needs for the entire Site. Case-2 has been defined as the maximum sodium which can be separated from the low activity waste without precipitation of $\text{Al}(\text{OH})_3$. It has been determined that the potential cost savings at Hanford ranges from \$194M to \$215M for Case-1, and \$293M to \$324M for Case-2. The potential cost savings at Savannah River are \$186M for Case-1 and \$281M for Case-2. A discussion of the uncertainty associated with these cost savings estimates can be found in the Discussion and Conclusions section.

Figure 1. Potential cost savings due to the use of caustic recycle

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INTRODUCTION

The underground storage tank (UST) waste at the Hanford and Savannah River Sites (SRS) was derived primarily from processing of nuclear fuels. Much of this nuclear fuel processing waste originated as a nitric acid based liquid rich in radioactive fission products. In order to achieve some type of temporary storage while the Cold War raged on, the acid based liquid waste was neutralized with sodium hydroxide (i.e. caustic) to minimize corrosion of the carbon-steel storage tanks. Consequently, the vast majority of the waste consists of various sodium salts. The sodium will dictate the final disposal volume of low activity waste (LAW). One method to reduce the sodium in the HLW, currently in practice at SRS and planned for Hanford, is a caustic wash of the sludge. However, this practice is limited to the sludge, and only moves the separated sodium to the LAW. An electrochemical salt-splitting process has been proposed for separating some of the sodium from the LAW for recycle as a caustic (i.e. NaOH), to be used in unit operations such as the sludge wash, or other non UST related activities at the Sites.

The purpose of this study is to provide a preliminary estimate of the potential cost savings for the application of caustic recycle at the Hanford and Savannah River Sites for the remediation of the UST waste. To minimize the cost of this study, existing information and past related cost studies were used as much as possible. An effort was not made to reconcile inconsistencies between the assumptions used in past studies, rather these differences were used to bound the results of this analysis. In order to minimize the effect of uncertainties, only the relevant cost differences were considered rather than absolute costs. For example, retrieval and HLW vitrification costs were not considered since they are not impacted by caustic recycle. Major uncertainties in the cost estimates are identified so that future cost assessments may focus on reducing these uncertainties.

The method of caustic recycle under consideration involves electrochemical processing of the LAW. For the purpose of this study, two distinct levels of LAW reduction were considered. The first level referred to as Case 1 in this study, provides all the caustic required for the entire Site needs. This includes unit operations such as the sludge wash and ion exchange for the UST remediation, as well as non UST Site needs. The second level of waste reduction referred to as Case 2 in this study, separates all of the LAW sodium possible without causing $\text{Al}(\text{OH})_3$ precipitation. Disposition of the additional caustic (i.e. sodium) removed during Case 2 has not been identified by this study. The process flowsheet for Case 1 and Case 2 is shown in Figure 2.

BACKGROUND

Hanford

There are currently approximately 67 million gallons (2.54×10^8 L) of HLW residing in 177 single and double shell tanks at the Hanford Reservation awaiting treatment and disposal [Reference 3]. The waste is predominantly a liquid-based high-sodium alkaline mixture composed of supernate (liquid), slurry (solids suspended in supernate), sludge (settled solids), and salt cake (crystallized supernate). The primary contaminating radionuclides are cesium 137 (in the supernate), strontium 90 (in the sludge), and their daughter products. Other radionuclides that exist in significant quantities are Tc-99, Am-241, Np-237, and Pu-239, 240 and 241. Under the original treatment plan, the supernate was to be separated from the solids, and after an evaporation process, the cesium was to be removed in an ion exchange process. In this way, the volume of HLW would be significantly reduced, and the liquid remaining after ion exchange would be LAW. The HLW would be vitrified and ultimately disposed of in a repository to be constructed, and the LAW would be vitrified and disposed of on-site.

In an effort to reduce costs, DOE has begun a Privatization effort that is divided into two phases [Reference 4]. Phase 1 was initiated in September 1996, and consists of two parts. DOE chose two contractors to establish the technical, operational, regulatory, business, and financial elements that will be required to process and immobilize a limited quantity of waste. This was completed in January 1998 comprises part A. The DOE has selected one contractor to proceed with part B of Phase 1, which will result in between 4 and 13% of the UST waste being immobilized. Phase 2 will involve processing the remaining waste.

Under the current plan [Reference 6], the immobilization processes will generate an estimated 41,600 metric tons (MT) (or $15,700 \text{ m}^3$) of immobilized HLW product and 470,000 MT (or $188,000 \text{ m}^3$) of LAW product. During Phase 2 it is estimated that 11,300 MT of sodium will be added as sodium hydroxide (caustic). Since the tank wastes contain large amounts of excess caustic, it is possible to separate the caustic already in the waste and use this in the processing of the tank wastes instead of purchasing new caustic. Separation and recycle of the caustic offers a potential cost saving by avoiding the procurement of fresh caustic and reducing the volume of LAW that will be produced.

Savannah River

The SRS currently stores 34 million gallons (1.3×10^8 L) of HLW as a combination of supernatant liquid, salt cake and sludge. The liquid and salt cake primarily consist of soluble sodium salts that includes nitrate, nitrite, hydroxide, and aluminate. Greater than 99.9% of the soluble salts will be disposed of in saltstone after radioactive components such as Cs-137 and Sr-90 are removed. The separated radionuclides and the sludge are to be vitrified at the Defense Waste Processing Facility (DWPF).

CAUSTIC RECYCLE PROCESS DESCRIPTION

An electrochemical salt splitting process has been developed for caustic recycle at Hanford and Savannah River (Figure 3). Inorganic ceramic membranes developed by Ceramatec Inc. and organic based cation exchange membranes are being considered. In this process, the waste is added to the anode compartment, and an electrical potential is applied to the cell. This drives sodium ions through the membrane. The charge balance in the anode compartment is maintained by generating H^+ from the electrolysis of water. The charge balance in the cathode is maintained by generating OH^- , either from the electrolysis of water or from oxygen and water using an oxygen cathode. The normal gaseous products of the electrolysis of water are oxygen at the anode and hydrogen at the cathode. Potentially flammable gas mixtures can be prevented by (1) providing adequate volumes of a sweep gas, (2) utilizing an alternative reductant, or (3) destruction of the hydrogen as it is generated. As H^+ is generated in the anode compartment, the pH drops. The process may be operated with either an alkaline ($pH > 13.5$) or an acidic anolyte ($pH < 1$). Operation at the intermediate pH values is precluded by AlO_2^- in the waste which would precipitate as $Al(OH)_3$. Production of OH^- in the cathode compartment results in a rise in pH as the sodium hydroxide product is recovered.

Figure 3. Salt splitting process for caustic recycle

COST ASSESSMENT

Hanford Cost Background

The cost-effectiveness analysis has been performed by comparing the life cycle costs of implementing an innovative technology to those of using the current baseline technology. In the case of the Hanford UST remediation, the baseline has been complicated by the Privatization effort. Initially, the baseline was stipulated by the Hanford Tank Waste Remediation System (TWRS) Process Flowsheet [Reference 3], and by the Engineering Data Package (EDP) for the TWRS Environmental Impact Statement [Reference 1]. Tank inventories, process flow diagrams, mass balances, facility plans, and estimated costs were calculated under the assumption that construction would begin in 1998 and operations would continue until 2024 when the final waste immobilization would take place. The total estimated cost for pretreatment, vitrification of both HLW and LAW, D&D, transportation, and disposal was \$13.8 B in 1995 dollars. With the advent of Privatization, only Phase 1, Part A costs are known for certain (\$54 M total). One of the prospective contractors has been eliminated from consideration and negotiations are continuing with the other. Consequently, updated costs for treatment of the waste are not yet known.

The difficulty in obtaining a baseline cost to compare to caustic recycle costs is demonstrated in Table 1. Costs are given for pretreatment and vitrification (not including disposal) of all of the LAW from the original TWRS EDP as well as for Phase 1 waste quantities from the bids of the contractors, BNFL, Inc. and Lockheed Martin Advanced Engineering Systems (LMAES). These costs include design, construction, operation, and D&D. Retrieval and disposal costs are not included. The Phase 1 contractor unit costs are expressed in \$/MT-Na in the supplied waste stream, so that the TWRS unit costs are presented in that format as well. Obviously, there is a significant difference in the unit costs.

Table 1. Costs for pretreatment and vitrification of LAW

Source	total cost	unit cost (\$/MT Na)	Na in waste (MT)
TWRS EDP -- all waste	\$4.0 B	56,200	71,800
BNFL, Inc. -- Phase 1	\$2.9 B	1,040,000	2,800
LMAES -- Phase 1	\$2.1 B	767,200	2,800

The BNFL and LMAES costs are taken from Phase 1 bids (for the minimum waste order only). Phase 2 unit costs are expected to decrease significantly. The TWRS

EDP cost includes pretreatment and vitrification, along with half of the centralized facilities costs (shared between LAW and HLW costs). It is shown in 1995 dollars.

The difference in unit cost between the Phase 1 bids and the TWRS estimate is partially due to scaling. The cost for a large capacity facility compared to a smaller one scales according to a cost capacity factor, which is generally less than one. In order for Phase 2 costs to be in line with the TWRS EDP estimate, the cost capacity factor for scaling the Phase 1 sized facility to a Phase 2 sized facility must be small, on the order of 0.15 to 0.25 (this is an exponential factor). If the LMAES bid for Phase 1 is scaled by a cost capacity factor of 0.25, the cost to pretreat and vitrify 65,600 MT of Na bearing waste (the Phase 2 amount) is \$4.7 B, giving a unit cost of \$72,000 per MT-Na. This is much more in line with the original TWRS unit cost. However, the total cost for the remediation is much higher than the TWRS estimate (\$6.8 B compared to \$4.0 B), which brings into question, whether the original estimate was too low. In addition, there is uncertainty whether a cost capacity factor of 0.25 is realistic. If a cost capacity factor of 0.6 is used (a value appropriate for a chemical production facility), the cost of the Phase 2 facility would be \$14.3 B. Note that these costs do not include retrieval and disposal.

The TWRS process flowsheet material balances have been revised to take into account the Privatization effort and are contained in the TWRS Operation and Utilization Plan (TWRS OUP) document [Reference 6]. Waste retrieval and delivery sequencing is provided for Phase 1 and material balances and process diagrams are provided for Phase 2. The flowsheets also reflect the fact that the waste inventory has been revised, a process that is ongoing.

Because Phase 2 costs have not yet been defined as part of the Privatization effort, the LAW immobilization and disposal throughput dependent costs without caustic recycle, were determined by two independent methods. The first method utilized the TWRS EIS EDP immobilization cost estimate [Reference 1], and the Hanford Waste Disposal Integration Team (WIT) disposal cost estimate [Reference 7]. The second method utilized the Raytheon/BNFL immobilization cost estimate [Reference 2], and the WIT disposal cost estimate.

The maximum quantity for Phase 1 is waste containing 10,000 MT of sodium. The LAW will be vitrified and stored until it can be disposed of during Phase 2. The untreated waste remaining for Phase 2 will contain 47,300 MT of Na. Since caustic recycle will most likely be implemented after the completion of Phase 1, changes in the Phase 1 waste quantity will impact the amount of waste remaining to be processed in Phase 2. Included in the RFP for Phase 1 is a specification that the radionuclide loading of the LAW form must be less than the NRC limits for a Class C waste. In addition, the average concentrations of ^{137}Cs , ^{90}Sr , and ^{99}Tc are limited as follows: $^{137}\text{Cs} < 3 \text{ Ci/m}^3$, $^{90}\text{Sr} < 20 \text{ Ci/m}^3$, and $^{99}\text{Tc} < 0.3 \text{ Ci/m}^3$. It is assumed that this specification will also be valid for Phase 2 as well.

Hanford Case 1 and Case 2 Definitions for Caustic Recycle

Two distinct cases, reflecting different levels of caustic recycle at Hanford are considered in addition to the baseline, which does not contain a caustic recycle process. Case 1 would involve the recovery of caustic in sufficient amounts to meet all identified site needs, which at Hanford are all associated with UST waste remediation. Case 2 would involve the recovery of all recoverable caustic. In this case the term recoverable refers to all caustic that can be separated from the UST waste, while not precipitating solid $\text{Al}(\text{OH})_3$. This is assumed to occur at a pH of 13.5; $[\text{OH}^-] = 0.32 \text{ M}$. The process flowsheet for Hanford is shown as Figure 4.

The masses and volumes of selected processing streams are shown in Table 2. These values are based on information given in the TWRS Operation & Utilization Plan, (TWRS OUP) [Reference 6]. It is currently assumed that there is a total of 56,200 MT of sodium in the tanks. It is also assumed that 8900 MT of sodium will be processed for disposal in Phase 1 and 47,300 MT in Phase 2. Another 11,000 MT is added as caustic for sludge leaching and 125 MT are added as sodium nitrite. The total sodium content in the LAW vitrification feed for Phase II is 58,400 MT (with no caustic recycle) and results in a 396,000 MT of LAW glass. The waste loading is limited by Na_2O at 20 wt%, such that the fractional reduction of sodium in the waste proportionately reduces the amount of LAW glass. The glass volume is determined from the estimated mass assuming a glass density of 2.66 MT/m^3 . The volume of the LAW packages is determined on the basis of a 2.6 m^3 package that is filled 80% or 2.08 m^3 .

Table 2. Sodium mass balance at Hanford

Stream number		Baseline TWRS OUP [Ref. 6]	Case I: Caustic recycle for pretreatment	Case II: Recover available caustic
(1) Retrieved waste	Volume, L	4.60×10^8		
	Total Na+, MT	47,300		
	Na+ as free NaOH, MT	19,200		
	Na+ as recoverable NaOH, MT	16,170		
(2) Chemical additions	Na+ added as caustic, MT	11,000	0	0
	Na+ added as NaNO ₂	125		
(3) Cs IX Feed	Volume @ 5M Na, L	5.08×10^8		
	Total Na+, MT	58,400		
(4) Chemical additions	Na+ added as NaOH, MT	300		
(5) Salt Splitting Feed	Volume @ 5M Na, L	5.1×10^8		
	Total Na+, MT	58,700		
(6) Caustic product	Total Na+, MT	0	11,000	17,180
	NaOH, MT	0	19130	59756
	Volume, 50 wt%, L	0	25.1×10^6	39.2×10^6
	Volume, 23 wt%, (7 M) L	0	54.5×10^6	85.2×10^6
(7) LAW Feed	Volume @ 5M Na, L	5.1×10^8	4.11×10^8	3.61×10^8
	Total Na+, MT	58,700	47,300	41,520
(8) LAW Product	LAW, MT	396,000	319,000	279,000
	LAW, m3	148,700	119,800	105,200
	LAW package, m3	185,900	149,800	131,500

Figure 4. Hanford tank waste treatment flow diagram with caustic recycle

Caustic Recycle Costs

The electrochemical facility is assumed to be part of a waste treatment plant that is necessary for the treatment and immobilization of the wastes. The caustic required for treatment of the tank waste (Case 1) may be readily recycled within the plant. Caustic in excess of that required for pretreatment is assumed to be trucked elsewhere for use or disposal. For this study it is assumed that the excess is used in grout for backfilling tanks. The capital and operating costs are developed as incremental costs to this facility. A stand alone facility is usually more expensive.

It is assumed that the salt splitting process will be operated in a lightly shielded facility that will allow limited contact maintenance since the process is implemented after much of the radioactivity has been removed. The capital costs are based on cost information for nonradioactive electrochemical plant, and equipment with a cost multiplier of 5 used to estimate the cost of the radioactive facility of similar capacity. The plant size is based on the number of electrochemical cells required.

The number of electrochemical cells is determined by the rate at which sodium must be transported and the process operating characteristics as shown in Table 3. The process operating parameters are based on pilot-scale testing with simulants, bench-scale testing with simulants, and actual wastes. The total operating efficiency (TOE) of 60% is based on the historical performance of Hanford plants.

Table 3. Process operating parameters based on the Nafion Membrane

Current density, mA/cm ²	400
Voltage	8
Process operating time, yrs	12
Sodium transport efficiency, %	90
Total operating efficiency	60
% area available for ionic transport	100

The electrochemical cell requirements are shown in Table 4. The membrane area is based on the sodium transport requirements and the process operating parameters in Table 3. The ICI FM-21 cell has a projected membrane area of 0.42 m² per electrode pair. Each cell stack can contain up to 60 electrode pairs for a total membrane area of 25 m² per cell stack for Nafion.

Table 4. Electrochemical cell requirements for Hanford

ICI FM-21 with Nafion Membranes			
	Required membrane area, m ²	# of electrode pairs	# of cell stacks
Case 1: Caustic recycle for pretreatment	55	131	3
Case 1I: Recover available caustic	82.9	197	4

The electrochemical cell and plant costs for nonradioactive service (Table 5) were supplied by Dr. David Genders of the Electrosynthesis Co. The cell costs are based on the use of ICI FM21 cells containing Pt/Ti anodes (lantern blades), Ni cathodes (also lantern blades), a Nafion 350 membrane, EPDM gasketing, flexible electrode connections to the busbars and all cell busbars. The installed plant cost was estimated by applying a factor of 5 which is typical of small salt splitting applications. The installed plant cost includes any pretreatment, gas handling, caustic concentration and the rectifier. An additional factor of 5 was applied to the nonradioactive plant cost to obtain the cost of a similar plant for radioactive service. While these cost estimates have a large uncertainty associated with them they appear to be in line with a preconceptual design estimate of \$22.5M for an electrochemical plant containing 10, ICI FM 21 cells for the destruction of nitrates in Savannah River tank waste [Reference 8].

Table 5. Electrochemical plant capital costs at Hanford

	Electrode Area m ²	Unit Cost \$/m ²	Estimated Cell Cost	Estimated Installed Plant Cost (nonradioactive)	Estimated Installed Plant Cost (radioactive)
Case 1	55	17,750	\$ 976,250	\$4.88M	\$24.4M
Case 2	83	16,500	\$1,369,500	\$6.85M	\$34.2M

The operating costs are summarized in Table 6 for Case 1 and in Table 7 for Case 2. In general the most recent and reasonable estimates were used for the labor, utility and maintenance costs, and are based on the following assumptions.

- (1) The plant will operate for 12 years, which is the length of time currently allocated for operations in Phase 2 of the TWRS Privatization. One year is allowed for startup.
- (2) Operations will require a total of 5 shifts for complete coverage, and each shift will require 5 additional operating personnel each earning \$100k/yr in salary plus benefits.
- (3) Electric power is based on an operating voltage of 8 V with a current efficiency of 90%. The unit cost of power is 48 \$/MWh, and is the cost of power that will be supplied to TWRS Privatization contractor.
- (4) The required volume of raw water was obtained from Reference 2. The unit cost is 0.25 \$/m³, and is the cost of raw water that will be supplied to TWRS Privatization contractor.
- (5) The required volume of demineralized water was obtained from Reference 2. The unit cost is 9 \$/m³, and is the cost of demineralized water that will be supplied to TWRS Privatization contractor.
- (6) It is assumed that the membranes will require replacement once per year at a cost of \$800/m² based on a recent quote from the Electrosynthesis Company. Nafion membranes in nonradioactive commercial applications often last 5-10 years. However, these applications typically involve the use of very pure feeds and no radioactivity. Nafion membranes have been operated with SRS waste simulants for 1000 hours. Testing at SRS [Reference 9] has demonstrated that the Nafion membranes exhibit no deleterious effects when exposed to gamma radiation up to exposures of 3.8×10^5 Rads. This corresponds to an estimated operating time of 2 years.
- (7) The maintenance for the balance of process is assumed to require 2500 man-hours in addition to the normal operating staff at a cost of \$50/hr (salary + benefits). Equipment is estimated at 2.25% of capital/year which is the same assumption used in Reference 1.
- (8) The low level solid waste volume generation rate and disposal cost was obtained from Reference 2.
- (9) For Case 2 it is assumed that the caustic in excess of that required to treat the tank waste must be trucked to another location. It is assumed that the caustic is concentrated to 50 wt% and transported in a 5000 gallon cargo tank at a cost of \$4000/trip. These assumptions are similar to those used in a study of the disposition of sodium from the Hanford Fast Flux Test Facility [Reference 10].

- (10) The ratio of D&D to capital was assumed to be equal to that of the TWRS LAW immobilization facility [Reference 1]. This ratio was 0.227.

Table 6. Operating costs for Hanford Case 1

Case 1

	unit	unit cost, \$	number of units	annual O&M cost	yrs	total cost	Basis/comments
Operating years including 1 yr startup	yr				13		Baseline
Operating hours	hrs		5256		12	63,072	Raytheon/BNFL Trade study
Number of shifts			5				
Additional operating personnel	EA		5				Raytheon/BNFL Trade study
Staff- (base salary + 40% overhead)	M-yr	\$100,000	5	\$500,000	13	\$6,500,000	Typical salary + benefits
Working space	ft ² -yr	\$17	100	\$1,700	13	\$22,100	Raytheon/BNFL Trade study
Supplies and Misc Expenses @ 10%	M-Yr	\$7,143	5	\$35,714	13	\$464,286	Raytheon/BNFL Trade study
Electric Power	MWh	\$48	7013	\$336,625	12	\$4,039,505	Unit cost from Waste Integration Team
Raw water	m ³	\$0.25	18750	\$4,688	13	\$60,938	volume from trade study and unit cost from Waste Integration Team
Demineralized water	m ³	\$9	420	\$3,780	13	\$49,140	Unit cost and volume from Raytheon/BNFL trade study
Membrane replacement	m ²	\$800	66.7	\$53,372	13	\$693,831	unit cost per Electrosynthesis Co 1 yr lifetime assumed
Maintenance for balance of process							
-labor	M-hr	\$50	2500	\$125,000	13	\$1,625,000	Raytheon/BNFL Trade study
-equipment @ 2.25% of capital facility/ year)	\$/ yr			\$610,150	13	\$7,931,950	annual cost assumption, p G11-20, TWRS EDP
Low level solid waste	m ³	\$5,000	4	\$20,000	13	\$260,000	Raytheon/BNFL Trade study + PNNL disposal costs
Total				\$1,691,029		\$21,646,750	

Table 7. Operating costs for Hanford Case 2

Case 2

	unit	unit cost, \$	number of units	annual O&M cost	yr	total cost	Basis/comments
Operating years including 1 yr startup	yr				13		Baseline
Operating hours	hrs		5256		12	63,072	Raytheon/BNFL Trade study
Number of shifts			5				
Additional operating personnel	EA		5				Raytheon/BNFL Trade study
Staff- (base salary + 40% overhead)	M-yr	\$100,000	5	\$500,000	13	\$6,500,000	Typical salary + benefits
Working space	ft ² -yr	\$17	100	\$1,700	13	\$22,100	Raytheon/BNFL Trade study
Supplies and Misc Expenses @ 10%	M-Yr	\$7,143	5	\$35,714	13	\$464,286	Raytheon/BNFL Trade study
Electric Power	MWh	\$48	10467	\$502,437	12	\$6,029,240	Unit cost from Waste Integration Team
Raw water	m ³	\$0.25	18750	\$4,688	13	\$60,938	volume from Raytheon/BNFL trade study and unit cost from Waste Integration Team
Demineralized water	m ³	\$9	420	\$3,780	13	\$49,140	Unit cost and volume from trade study
Membrane replacement	m ²	\$800	99.6	\$79,661	13	\$1,035,591	unit cost per Electrosynthesis Co 1 yr lifetime assumed
Maintenance for balance of process							
-labor	M-hr	\$50	2500	\$125,000	13	\$1,625,000	Raytheon/BNFL Trade study
-equipment @ 2.25% of capital facility/year	\$/yr			\$855,950	13	\$11,127,350	annual cost assumption, p G11-20, TWRS EDP
Low level solid waste	m ³	\$5,000	4	\$20,000	13	\$260,000	Raytheon/BNFL Trade study + PNNL disposal costs
Transportation of excess caustic, 50 wt%	m ³	\$211	1173	\$247,503	12	\$2,970,036	Transportation and Packaging Evaluation
Total				\$2,128,929		\$30,143,680	

Potential UST Remediation Savings

The caustic recycle process provides a cost savings by reducing the volume of LAW that must be handled, reducing the amount of caustic that must be procured, and reducing the costs of vitrification. The cost of handling and disposal of the LAW packages after they are returned from the contractors is assumed to be \$3800/m³ per the Waste Integration Team [Reference 7]. The potential cost savings at Hanford for Cases 1 and 2 have been estimated as shown in Tables 8 and 9. The costs for LAW immobilization and disposal shown in Tables 8 and 9, are only those which are dependent on the total amount of LAW produced. It was assumed the LAW production rate would be constant, but the length of processing time would be reduced for Case 1 and Case 2. This implied the LAW immobilization and disposal capital costs would not change. Assuming caustic is selling for \$300/MT NaOH (the cost often fluctuates between \$200 and \$400/MT NaOH) the cost saving due to avoiding procurement of caustic is estimated at \$5.7M for the 19,130 MT of NaOH (11,000 MT of Na) that is required. This savings is identical for both cases since the caustic requirement is constant.

The TWRS EIS-EDP and Raytheon/BNFL immobilization costs were inflated from 1995 dollars to 1998 dollars by using a discount rate of 3.5%/year. Cost savings shown in Table 8 are based upon TWRS EIS-EDP immobilization costs; whereas, the cost savings shown in Table 9 are based upon Raytheon/BNFL immobilization costs. The TWRS EIS-EDP immobilization costs are based upon Table F-11 of Reference 2, and includes labor (\$624M-1995) and materials (\$176M-1995) for operations. Caustic recycle costs are based on Tables 3 through 5.

Table 8. Cost savings basis for Hanford using TWRS EIS-EDP immobilization costs

	TWRS EIS-EDP	WIT	Phase II	Case 1	Case 2
LAW immobilization (MT-glass)	n/a	n/a	396,000	319,000	279,000
LAW immobilization (1998 \$/MT-glass)	1609	no estimate	1609	1609	1609
LAW immobilization (1998 \$M)	n/a	n/a	637	513	449
LAW disposal (1998 \$/MT-glass)	no estimate	1784	1784	1784	1784
LAW disposal (1998 \$M)	n/a	n/a	706	569	500
Caustic Recycle capital (\$M)	n/a	n/a	0	24.4	34.2
Caustic Recycle operating (\$M)	n/a	n/a	0	21.6	33.6
Caustic Recycle D&D (\$M)	n/a	n/a	0	5.5	7.7
Caustic Recycle subtotal (\$M)	n/a	n/a	0	51.5	75.5
Caustic cost (\$M)	n/a	n/a	5.7	0	0
Total Cost (\$M)	n/a	n/a	1349	1134	1025
Caustic Recycle savings (\$M)	n/a	n/a	0	215	324

Table 9. Cost savings basis for Hanford using Raytheon/BNFL immobilization costs

	Raytheon & BNFL	WIT	Phase II	Case 1	Case 2
LAW immobilization (MT-glass)	n/a	n/a	396,000	319,000	279,000
LAW immobilization (1998 \$/MT-glass)	1344	no estimate	1344	1344	1344
LAW immobilization (1998 \$M)	n/a	n/a	532	429	375
LAW disposal (1998 \$/MT-glass)	no estimate	1784	1784	1784	1784
LAW disposal (1998 \$M)	n/a	n/a	706	569	500
Caustic Recycle capital (\$M)	n/a	n/a	0	24.4	34.2
Caustic Recycle operating (\$M)	n/a	n/a	0	21.6	33.6
Caustic Recycle D&D (\$M)	n/a	n/a	0	5.5	7.7
Caustic Recycle subtotal (\$M)	n/a	n/a	0	51.5	75.5
Caustic cost (\$M)	n/a	n/a	5.7	0	0
Total Cost (\$M)	n/a	n/a	1244	1050	951
Caustic Recycle savings (\$M)	n/a	n/a	0	194	293

SAVANNAH RIVER COST ASSESSMENT

Cost Background

Recovery of NaOH (caustic) from the salt solution could significantly reduce the volume of waste disposed in saltstone while reducing the quantity of new chemicals added to the HLW system at SRS. The recovered caustic could be used to neutralize fresh waste from the Separations canyons, Defense Waste Processing Facility, and the Effluent Treatment Facility, used as a corrosion inhibitor in the Tank Farm, and used to dissolve alumina in Extended Sludge Processing (ESP). The site requirement for fresh caustic is estimated at 93,900 gallons of 50 wt% solution per year. Over the course of the planned 24 year tank waste treatment and disposal period the total requirement is estimated at 2.25×10^6 gallons of 50 wt% caustic (containing 3740 MT of Na).

The average composition of the aqueous tank at SRS waste is given in Table 10 and includes the existing supernate, dissolved salt cake plus the estimated future salt additions [Reference 11]. Assuming that the radionuclide removal processes do not substantially change the composition, this is the average composition of the feed for a salt splitting process, or the saltstone feed in the absence of a caustic recycle process.

Table 10. Volume and average composition of SRS aqueous tank waste

Cation Component	Concentration, M	Anion Component	Concentration, M
Ca^{+2}	2.17×10^{-5}	$\text{Al}(\text{OH})_4^-$	0.237
K^+	0.0112	Cl^-	0.0188
Na^+	4.6	CO_3^{-2}	0.121
		F^-	0.0242
		NO_2^-	0.391
		NO_3^-	1.68
		OH^-	1.746
		PO_4^{-3}	0.006.1
		SO_4^{-2}	0.113
Total Volume = 4.35×10^8 L = 1.15×10^8 gallons			

Case 1 and Case 2 Definitions for Caustic Recycle

A simplified tank waste treatment diagram is shown in Figure 5. This processing diagram reflects the baseline approach except that the caustic recycle process is inserted after radionuclide removal. The radionuclides of interest for removal from the supernatant liquid fraction are Sr-90, TRU components and Cs-137. The method

for removing the Cs-137 components from the supernate is being re-evaluated due to high rates of benzene generation during the In-Tank precipitation process. It is assumed for the purpose of this study, that the Cs-137 removal process will not significantly alter the composition of the supernatant waste.

Two cases reflecting different levels of caustic recycle at Savannah River are considered in addition to the baseline which does not contain a caustic recycle process. Case 1 involves the recovery of caustic in sufficient amounts to meet the site needs. Case 2 involves the separation of all readily recoverable caustic which is defined as caustic that can be separated from the waste while not precipitating $(\text{Al}(\text{OH})_3)$. Precipitation generally occurs at a pH of 13.5; $[\text{OH}^-] = 0.32 \text{ M}$. The process flowsheet for SRS is shown as Figure 5.

A summary sodium balance is shown in Table 11. The existing inventory and future waste additions were obtained from the most recent High Level Waste System Plan, which is Revision 9 of Reference 11. The sodium concentration of 4.6 M corresponds to the estimated retrieved waste volume and assumes that supernatant treatment for radionuclide removal does not substantially change the composition or the volume. Evaporation of the saltstone feed is necessary in order to realize a reduction in the volume of saltstone. A sodium concentration of 6.8 M is used as the end point for evaporation, which is the approximate point at which salts start to precipitate. The saltstone volume is based on 1.77 times the saltstone feed volume and accounts for the addition of the saltstone forming materials. The number of saltstone vault cells is determined by the capacity of a vault cell which contains 1,782,000 gallons of cement waste form, or about 1 million gallons of feed solution.

Table 11. Sodium mass balance at Savannah River

Stream number		Baseline	Case 1 Caustic recycle to meet site needs	Case 2 Recover all available caustic
Existing Inventory in Supernate + Salt Cake (S+SC)	Volume, L (gallons)	4.35×10^8 (1.15×10^8)		
S+SC	Na+ concentration	4.6 M		
S+SC	Total Na+	1.81×10^9 moles (41,520 MT)		
S+SC	Na+ as free NaOH	5.96×10^8 moles (13710 MT)		
S+SC	Na+ as recoverable NaOH	4.71×10^8 moles (10,083 MT)		
Future Chemical additions (FCA)	Na+ added as caustic, moles (MT)	1.63×10^8 (3,740)	0	0
FCA	Na+ added as other compounds	3.18×10^7 moles (730 MT)		
Saltstone feed (SSF)	Total Na+, moles (MT)	2.0×10^9 45,980	1.84×10^9 42,250	1.37×10^9 31,420
SSF	Volume @ 4.6M Na, L	4.35×10^8	4.0×10^8	3.32×10^8
SSF	Volume @ 6.8 M Na, L	5.20×10^8	4.78×10^8	3.56×10^8
Saltstone product (SSP)	Volume with 4.6 M Na Feed, L (vault cells)	7.69×10^8 (115)	7.07×10^8 (105)	5.26×10^8 (78)
SSP	Volume with 6.8 M Na Feed, L (vault cells)	5.20×10^8 (78)	4.78×10^8 (71)	3.56×10^8 (53)

Figure 5. SRS Tank Waste Treatment Flow Diagram with Caustic Recycle

Caustic Recycle Costs

The electrochemical facility is assumed to be a stand-alone facility that is combined with an evaporator for concentrating the saltstone feed. The evaporator design and cost estimate is based on the information given in Reference 12. The evaporator consists of 3 skid mounted evaporators, each processing 8.3 gpm of distillate (total capacity of 25 gpm) from a total feed flow rate of 72 gpm (24 gpm per skid). The evaporator building was assumed to be 60' wide x 140' long with a ceiling height of 30' erected on a concrete slab. A total of four, 35,000 gallon underground tanks are included; two for feed to the evaporator and two for the concentrate. A single above ground 35,000 gallon tank is included for condensate storage. A rough order of magnitude cost estimate for this facility was \$35 million. This evaporation capability appears to be larger than required for the purposes of this study, in which a distillate rate of 4.9-8.2 gpm is sufficient with a total operating efficiency of 60%.

It is assumed that the salt splitting process will be operated in a lightly shielded facility that will allow limited contact maintenance, since the process is implemented after much of the radioactivity has been removed. The capital costs are based on cost information for nonradioactive electrochemical plant and equipment with a cost multiplier of 5 used to estimate the cost of the radioactive facility of similar capacity. The plant size is based on the number of electrochemical cells required. No credit is taken for any economy that might be achieved by combining the salt splitting process with the saltstone feed evaporator.

The number of electrochemical cells is determined by the rate at which sodium must be transported, and the process operating characteristics are shown in Table 12. The process operating parameters are based on pilot-scale testing with simulants, bench-scale testing with simulants, and actual wastes considering the projected performance of the ICI FM-21 cell. The total operating efficiency (TOE) of 60% is based on the historical performance of radioactive processing plants.

Table 12. Salt splitting process operating parameters

Current density, mA/cm ²	400
Voltage	8
Process operating time, yrs	25
Sodium transport efficiency, %	90
Total operating efficiency	60
% area available for ionic transport	100

The electrochemical cell requirements are shown in Table 13. The membrane area is based on the sodium transport requirements and the process operating parameters in Table 12. The ICI FM-21 cell has a membrane area of 0.42 m² per electrode pair. Each cell stack can contain up to 60 electrode pairs for a total membrane area of 25 m² per cell stack for Nafion. The fractional quantity of cells is determined by dividing the number of electrode pairs by the maximum number (60) of electrode pairs per cell stack. The number of cell stacks is determined by rounding this number up to the next greatest integer.

Table 13. Electrochemical cell requirements for Savannah River

ICI FM-21 with Nafion Membranes			
	Required membrane area, m ²	# of electrode pairs	# of cell stacks (fractional quantity)
Case 1: Caustic recycle for site needs	9.6	23	1 (0.4)
Case 2: Recover available caustic	37.4	80	2 (1.3)

The electrochemical cell and plant costs for nonradioactive service (Table 14), were supplied by Dr. David Genders of the Electrosynthesis Co. The cell costs are based on the use of ICI FM21 cells containing Pt/Ti anodes (lantern blades), Ni cathodes (also lantern blades), a Nafion 350 membrane, EPDM gasketing, flexible electrode connections to the busbars and all cell busbars. The installed plant cost was estimated by applying a factor of 5, which is typical of small salt splitting applications. The installed plant cost includes any pretreatment, gas handling, caustic concentration and the rectifier. An additional factor of 5 was applied to the nonradioactive plant cost to obtain the cost of a similar plant for radioactive service. While these cost estimates have a large uncertainty associated with them they appear to be in line with a preconceptual design estimate of \$22.5M for an electrochemical plant containing 10, ICI FM 21 cells for the destruction of nitrates in Savannah River tank waste [Reference 8].

Table 14. Electrochemical plant capital costs at Savannah River

	Electrode Area m ²	Unit cost, \$/m ²	Estimated Cell Cost	Estimated Installed Plant Cost nonradioactive	Estimated Installed Plant Cost radioactive
Case 1	9.6	19,950	\$191,500	\$0.96M	\$4.79M
Case 2	37.4	18,250	\$682,550	\$3.41M	\$17.1M

The operating costs are summarized in Table 15 for Case 1 and in Table 16 for Case 2. In general the most recent and reasonable estimates were used for the labor, utility and maintenance costs and are based on the following assumptions.

- (1) The plant will operate for a total of 25 years with one year allowed for startup.
- (2) The incremental labor costs for the salt splitting process [Reference 8] are based on the assumption that there are 5 shifts, each with 2 operators and one supervisor. The fully burdened cost for an operator is \$60,000/year and for a supervisor is \$75,000/yr.
- (3) The electric power costs are based on a cost of \$57/MWh with the power requirements being determined from the process operating parameters given in Table 13.
- (4) One lot of support utilities is included at an annual cost of \$60,000 [Reference 8]. This may overestimate the actual cost since it was not scaled from the 10 electrochemical cells to the smaller treatment plants required for caustic recycle in this study. In any case, the cost appears relatively insignificant.
- (5) The membranes will require replacement once per year. Nafion membranes in nonradioactive commercial applications often last 5-10 years. However, these applications typically involve the use of very pure feeds and no radioactivity. Nafion membranes have been operated with SRS waste simulants for 1000 hours. Testing at SRS [Reference 9] has demonstrated that the Nafion membranes exhibit no deleterious effects when exposed to gamma radiation up to exposures of 3.8×10^5 Rads. This corresponds to an estimated operating time of 2 years. The cost of new Nafion membranes is \$800/m² and was obtained from the Electrosynthesis Company. The purchased membrane area is 20% larger than the installed area to account for waste due to required cutting.

Table 15. Operating Costs for SRS Case 1

	unit	unit cost \$	number of units	annual O&M cost	years	total cost	Basis/comments
operating years including 1 yr startup	yr				25		High Level Waste System Plan, Rev 8
operating hours	hrs/yr		5256		25	131,400	60% TOE. Note that 70% is assumed in WSRC-OS-97-00008
number of shifts			5				4 shifts were assumed in WSRC-OS-97-00008. This is increased to 5 to ensure complete coverage due to vacations, illness, holidays ect.
Additional personnel - operators	EA		10				2 operators/shift assumed in WSRC-OS-97-00008
Additional personnel - supervisors	EA		5				1 supervisor/shift assumed in WSRC-OS-97-00008
Direct labor (fully burdened) - operators	M-yr	\$60,000	10	\$600,000	25	\$15,000,000	WSRC-OS-97-00008
Direct labor (fully burdened) - supervisors	M-yr	\$75,000	5	\$375,000	25	\$9,375,000	WSRC-OS-97-00008
Electric Power	MWh	\$57	1210	\$68,963	25	\$1,724,071	Unit cost from WSRC-OS-97-00008
Support utilities - HVAC, lighting, potable water, sewer	1 lot	\$60,000	1	\$60,000	25	\$1,500,000	Quantity and Unit cost from WSRC-OS-97-00008. May over estimate cost as the nitrate destruction plant in WSRC-OS-97-00008 is much larger (10 electrochemical cells vs 1).
Membrane replacement	m ²	\$800	11.5	\$9,208	25	\$230,189	unit cost per Electrosynthesis Co: 1 yr lifetime assumed, labor is included in process maintenance on next line.
Process maintenance @ 5% of capital facility/year)	\$/yr	\$1,989,400	1	\$1,989,400	25	\$49,735,000	5% Unit cost from WSRC-OS-97-00008
Low level solid waste	m ³	\$5,000	4	\$20,000	25	\$500,000	Raytheon/BNFL Trade study + PNNL disposal costs
Evaporator Operating Costs	\$/gallon	\$1.50	1.74x10 ⁶	\$2,607,600	25	\$65,190,000	Saltstone Feed Evaporator study, volume is distillate volume, not feed
Transportation of caustic	\$/m ³	\$211.00	2.97x10 ²	\$62,667	24	\$1,504,008	Assume all caustic is trucked as 50 wt% solution as there is no caustic return line.
Total				\$5,792,837		\$144,758,268	

Table 16. Operating Costs for SRS Case 2

	unit	unit cost \$	number of units	annual O&M cost	years	total cost	Basis/comments
operating years including 1 yr startup	yr				25		High Level Waste System Plan, Rev 8
operating hours	hrs		5256		25	131,400	60% TOE. Note that 70% is assumed in WSRC-OS-97-00008
number of shifts			5				4 shifts were assumed in WSRC-OS-97-00008. This is increased to 5 to ensure complete coverage due to vacations, illness, holidays ect.
Additional personnel - operators	EA		10				2 operators/shift assumed in WSRC-OS-97-00008
Additional personnel - supervisors	EA		5				1 supervisor/shift assumed in WSRC-OS-97-00008
Direct labor (fully burdened) - operators	M-yr	\$60,000	10	\$600,000	25	\$15,000,000	WSRC-OS-97-00008
Direct labor (fully burdened) - supervisors	M-yr	\$75,000	5	\$375,000	25	\$9,375,000	WSRC-OS-97-00008
Electric Power	MWh	\$57	6285	\$358,272	25	\$8,956,792	Unit cost from WSRC-OS-97-00008
Support utilities - HVAC, lighting, potable water, sewer	1 lot	\$60,000	1	\$60,000	25	\$1,500,000	Quantity and Unit cost from WSRC-OS-97-00008. Probably over estimates cost as the nitrate destruction plant in WSRC-OS-97-00008 is much larger (10 electrochemical cells vs 1). unit cost per Electrosynthesis Co: 1 yr lifetime assumed
Membrane replacement	m ²	\$800	44.8	\$35,876	25	\$896,899	5% Unit cost from WSRC-OS-97-00008
Process maintenance @ 5% of capital facility/year)	\$/yr	\$2,603,200	1	\$2,603,200	25	\$65,080,000	
Low level solid waste	m ³	\$5,000	4	\$20,000	25	\$500,000	Raytheon/BNFL Trade study + PNNL disposal costs
Evaporator Operating Costs	\$/gallon	\$1.50	2.47x10 ⁶	\$3,705,000	25	\$92,625,000	Saltstone Feed Evaporator study, volume is distillate volume, not feed
Transportation of caustic	\$/m ³	\$211.00	4.94x10 ²	\$104,234	24	\$2,501,616	Assume all caustic is trucked as 50 wt% solution as there is no caustic return line.
Total				\$7,861,582		\$196,435,307	

- (6) The annual process maintenance cost is estimated at 5% of the capital cost of the facility [Reference 8].
- (7) The cost of low level solid waste disposal is estimated to be \$5000/m³ and is based on disposal costs at a commercial site.
- (8) The evaporator operating costs of \$1.50 per gallon were taken from the preconceptual evaporator cost study [Reference 12].
- (9) For both Case 1 and 2 it is assumed that the caustic must be transported by truck to another location since there are no return lines to the tank farms. It is assumed that the caustic is concentrated to 50 wt% and transported in a 5000 gallon cargo tank at a cost of \$4000/trip. These assumptions are similar to those used in a study of the disposition of sodium from the Hanford Fast Flux Test Facility [Reference 10].
- (10) The ratio of D&D to capital was assumed to be equal to that of the TWRS LAW immobilization facility [Reference 1]. This ratio was 0.227.

Potential UST Remediation Savings

The caustic recycle process provides a cost savings by reducing the volume of Saltstone produced, and reducing the amount of caustic that must be procured. The variable cost of producing Saltstone has been estimated at \$4.88 per gallon (\$1.29 per L). The potential cost savings for Case 1 is therefore estimated, as shown in Table 17 to be \$186M. The potential cost savings for Case 2 is similarly estimated to be \$281M. . The costs for LAW immobilization and disposal shown in Table 18, are only those which are dependent on the total amount of LAW produced. It was assumed the LAW production rate would be constant, but the length of processing time would be reduced for Case 1 and Case 2. This implied the LAW immobilization and disposal capital costs would not change. Assuming caustic sells for \$300/MT NaOH (the cost often fluctuates between \$200 and \$400/MT NaOH), and a cost savings due to avoiding procurement of caustic is estimated at \$2.0M for the 6,500 MT of NaOH (3740 MT of Na) that is required. This savings is identical for both cases since the caustic requirement is constant.

Table 17. Cost savings basis for SRS

	Baseline @ 4.6 M-Na	Baseline @ 6.8 M-Na	Case 1	Case 2
LAW immobilization & disposal (m 3-final)	369,000	520,000	478,000	356,000
LAW immobilization & disposal (1998 \$M)	992	671	616	458
Caustic Recycle & evaporation capital (\$M)	n/a	n/a	39.8	52
Caustic Recycle & evaporation operating (\$M)	n/a	n/a	143.2	191.7
Caustic Recycle & evaporation D&D (\$M)	n/a	n/a	8.9	11.7
Caustic Recycle & evaporation subtotal (\$M)	n/a	n/a	191.9	255.4
Caustic cost (\$M)	2	2	0	0
Total Cost (\$M)	994	n/a	808	713
Caustic Recycle savings (\$M)	0	n/a	186	281

DISCUSSION AND CONCLUSIONS

The conclusions described in the Summary section are that the potential cost savings at Hanford range from \$194M to \$215M for Case-1, and \$293M to \$324M for Case-2. The potential cost savings at the SRS is \$186M for Case-1, and \$281M for Case-2. It is important to note that a significant portion of the caustic recycle cost savings at SRS, is due to the addition of an evaporator which concentrates the LAW feed from 4.6 M-Na to 6.8 M-Na.

While significant debate continues over the basis used for estimating UST remediation costs, it has not been within the scope of this study to ascertain all uncertainties related to the existing cost basis. This would be a monumental effort considering the scope of the remediation effort, and consequently the authors have chose to use engineering judgement in assessing the cost savings uncertainty. Based upon the single sensitivity comparison for Hanford, and assuming the SRS cost uncertainties are less than Hanford due to greater maturity, it appears the cost savings uncertainty for caustic recycle is less than \$100M at Hanford, and probably tens of millions at SRS. This implies that with reasonable certainty, the use of caustic recycle at Hanford or SRS could yield cost savings of hundreds of millions of dollars.

Before detailed design and cost estimates can be performed, the following issues require resolution. (1) Purity and concentration requirements for onsite customers must be determined, (2) How to best deliver recovered caustic to onsite customers must be determined, (3) Cell performance and service life of the electrochemical reactors must be determined, and (4) options for excess sodium disposition for Case 2 must be identified.

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