Sodium Recycle Economics for Waste Treatment Plant Operations – 8341

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ABSTRACT

Sodium recycle at the Hanford Waste Treatment Plant (WTP) would reduce the number of glass canisters produced, and has the potential to significantly reduce the cost to the U.S. Department of Energy (DOE) of treating the tank wastes by hundreds of millions of dollars. The sodium, added in the form of sodium hydroxide, was originally added to minimize corrosion of carbon-steel storage tanks from acidic reprocessing wastes. In the baseline Hanford treatment process, sodium hydroxide is required to leach gibbsite and boehmite from the high level waste (HLW) sludge. In turn, this reduces the amount of HLW glass produced. Currently, a significant amount of additional sodium hydroxide will be added to the process to maintain aluminate solubility at ambient temperatures during ion exchange of cesium. The vitrification of radioactive waste is limited by sodium content, and this additional sodium mass will increase low-activity waste-glass mass.

An electrochemical salt-splitting process, based on sodium-ion selective ceramic membranes, is being developed to recover and recycle sodium hydroxide from high-salt radioactive tank wastes in DOE's complex. The ceramic membranes are from a family of materials known as sodium (Na)—super-ionic conductors (NaSICON)—and the diffusion of sodium ions (Na+) is allowed, while blocking other positively charged ions.

A cost/benefit evaluation was based on a strategy that involves a separate caustic-recycle facility based on the NaSICON technology, which would be located adjacent to the WTP facility. A Monte Carlo approach was taken, and several thousand scenarios were analyzed to determine likely economic results. The cost/benefit evaluation indicates that 10,000–50,000 metric tons (MT) of sodium could be recycled, and would allow for the reduction of glass production by 60,000–300,000 MT. The cost of the facility construction and operation was scaled to the low-activity waste (LAW) vitrification facility, showing cost would be roughly $150 million to $400 million for construction and $10 million to $40 million per year for operations. Depending on the level of aluminate supersaturation allowed in the storage tanks in the LAW Pretreatment Facility, these values indicate a return on investment of up to 25% to 60%.

INTRODUCTION

Sodium is one of the most common components of the Hanford tank wastes, and is a major contributor to the waste-oxide loading in the low-activity waste (LAW) glass. In addition to the large amounts of sodium already in the wastes, the current waste-treatment approach necessitates the addition of supplementary sodium (primarily as NaOH) while pretreating the tank wastes, which would potentially increase the volume of LAW glass. Since the tank wastes already contain significant amounts of sodium, the potential benefit exists for a caustic-recycle process that would separate sodium hydroxide for recycle at the Hanford Site. This potentiality would reduce the volume of LAW glass and minimize the need to purchase new NaOH.

The current pretreatment flowsheet indicates that approximately 6500 metric tons (MT) of Na will be added to the tank waste, primarily for removing Al from the high-level waste (HLW) sludge [1]. An assessment [2] of the pretreatment flowsheet, equilibrium chemistry, and laboratory results indicates that the quantity of Na required for sludge leaching will increase by 6000–12,000 MT to dissolve sufficient Al from the tank-waste sludge material. This is to maintain the number of HLW canisters produced at 9400 canisters, as defined in the Office of River Protection (ORP) System Plan [3]. The additional Na will significantly increase the volume of LAW glass and extend the processing time of the Waste Treatment and Immobilization Plant (WTP). Future estimates on sodium requirements for caustic leaching are expected to significantly exceed the 12,000-MT value, and approach 40,000 MT of total sodium addition for leaching [4].

Electrochemical salt-splitting technologies for caustic recycle were investigated in the 1990s for application to the treatment of tank wastes at the Hanford, Savannah River, and Idaho National Engineering Laboratory sites. These investigations, which were primarily funded by the EM-50 Efficient Separations and Processing Program, included testing of commercially available, organic-based, ion-exchange membranes (i.e., Nafion) and ceramic-based, sodium-selective membranes (NaSICON) developed by Ceramatec, Inc. Both membrane types were tested with simulants at the pilot scale and with actual radioactive-waste samples at the bench scale. The Nafion membranes were found to have a lower current efficiency than the ceramic membranes. The Nafion membranes also transported radioactive cesium at a higher rate than the sodium, resulting in a contaminated caustic product.

The likely increase in caustic demand for pretreating tank wastes has resulted in renewed interest in the caustic-recycle methods. Ceramatec, Inc., has continued to develop the NaSICON membranes for caustic recycle. As part of this development effort, Ceramatec has engaged staff at the Pacific Northwest National Laboratory (PNNL) to assist with the application of this technology for caustic recycling at Hanford. This paper addresses the economic issues involved in the deployment of such a facility at Hanford.

SCOPE AND OBJECTIVE

This paper contains a review of the potential cost benefits of NaSICON ceramic membranes for the separation of sodium from Hanford tank waste. The primary application is for caustic recycle to the WTP pretreatment-leaching operation. The report includes identification of the benefits and costs for a caustic-recycle facility, and Monte Carlo results obtained from a model of these costs and benefits. The use of existing cost information has been limited to those sources that are publicly available. This paper is intended to be an initial evaluation of the economic feasibility of a caustic-recycle facility based on NaSICON technology.

Overall Process Description

Sodium is recovered via the electrochemical process with a Ceramatec membrane as shown in **Error! Reference source not found.**. Anode, cathode, and overall reactions from this process are shown in the equations below. In this process, sodium ions are selectively transported across a ceramic membrane, driven by an applied electrical potential.

Anode: $2H_2O \rightarrow O_2 + 4H^+ + 4e^-$ (Eq. 2)

$H^+ + OH^- \rightarrow H_2O$ (Eq. 3)

Cathode: $4H_2O + 4e^- \rightarrow 2H_2 + 4OH^-$ (Eq. 4)

Overall: $2H_2O \rightarrow 2H_2 + O_2$ (Eq. 5)

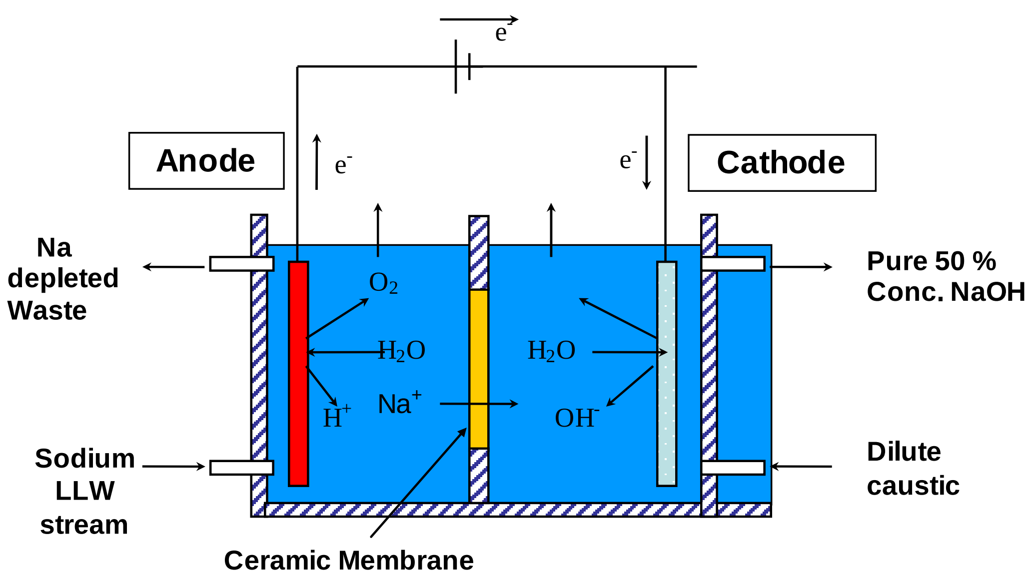

Fig. 1. Schematic of a Two-Compartment Electrochemical Process Using the NaSICON.

METHODS

The cost benefit for caustic recycling is presumed to consist of four major contributions: 1) the cost savings realized by not producing additional immobilized low-activity waste (ILAW) glass, 2) caustic-recycle capital investment, 3) caustic-recycle operating and maintenance costs, and 4) research and technology (R&T) costs needed to deploy the technology. In estimating costs for each of these components, several parameters are used as inputs. Due to the uncertainty in assuming a singular value for each of these parameters, a range of possible values is assumed as shown in Table I. A Monte Carlo simulation is then performed in which the range of these parameters is exercised, and the resulting range of cost benefits is determined. The remainder of this section is dedicated to discussing the range of parameters used for the major contributors to the cost.

Table I. Range of Major Parameters Used in the Monte Carlo Model

Estimated Costs	Low	Nominal	High
Electrical $/kWh	0.05	0.10	0.15
Labor Rates ($/hr)	47	52	57
Fixed Capital Exponent	0.33	0.5	0.67
Membrane Replacement Frequency	Twice Per Year	Once Per Year	Once Every Two Years
Membrane Cost ($/m^2)	12K	16K	18K
Research & Technology ($)	10M	20M	30M
Plant Availability (%)	50	60	70
Na Removal (MT)	10	30K	50K
Na_2O ILAW Limitation (%)	20	23	26
ILAW Cost Saving ($/MT)	12.8K	15.9K	19.1K
Operating Life	27 years	27 years	27 years

Sodium Removal

For every mole of sodium entering the cathode cell, a mole of water is consumed, and a mole of hydroxide ions and one-half mole of diatomic hydrogen are produced. For every mole of sodium transported from the pretreated LAW across the membrane, the anode reaction results in the consumption of a mole of hydroxide ions, and the production of one-half mole of water and one-quarter mole of diatomic oxygen. Consequently, the pH of the pretreated LAW stream will drop as the reaction proceeds.

The drop in pretreated LAW pH can be significant enough to precipitate dissolved species in the LAW. This study limits the amount of sodium transported across the membrane to an amount that will not result in aluminate saturation to a series of assumed temperatures. One limitation of any caustic-recovery stream would be to avoid fouling the caustic-recovery cell from the precipitation of aluminum. Therefore, to understand how much caustic can be removed from a process stream, understanding the equilibrium conditions between aluminate and hydroxide is necessary. The solubility model used for this estimate is a simple conservative model [5] that does not include the effects of other ions that increase the solubility of the aluminate. Some degree of supersaturation is present, based on the model. This was allowed on the basis of actual laboratory tests, the conservative nature of the solubility model, and the very slow precipitation kinetics.

ILAW Glass Model

Hamel and coworkers [6] have recently presented a revised U.S. Department of Energy (DOE) glass model. The oxide loading in the ILAW glass has a maximum sodium oxide loading of 23% by mass. The maximum SO_3 loading in this model corresponds to 1.2% by mass.

ILAW Cost Savings

Curtis [7] provided an economic assessment of Hanford's waste-treatment options. The focus of this thesis is the trade-off of pretreatment partitioning technologies against the total mass of HLW and low-level waste (LLW) produced during the lifecycle of each flowsheet. These costs are summarized in Table II, with a total cost of $93,400/MT Na ILAW.

DOE/ORP-2007-03 provides an economic assessment of Hanford waste-treatment options [9]. One of the options, case 2, provides operating cost estimates for a second LAW vitrification facility. These costs are estimated at $114M/yr for a facility with a 1220 MT Na/yr throughput capacity. As shown in Table II this translates to a cost of $93,400/ MT Na. After converting this value to a MT of glass basis, a Monte Carlo distribution assumption for ILAW cost savings is shown in Table III.

Table II. Estimation of ILAW Immobilization Costs, adapted from Curtis [7] in 2008 Dollars, Assuming 3% Annual Rate of Inflation

Case 2 Supplemental LAW Vitrification Operating Costs $M/yr	**Case 2 Supplemental LAW Vitrification Capacity** MT Na/yr	**Case 2 Supplemental LAW Vitrification Operating Costs** $/MT Na
$114	1220	$93,400

Table III. Monte Carlo Distribution Assumptions for ILAW-Cost Savings

Description	Distribution Type	Low	Median	High
ILAW Cost Savings ($/MT ILAW), ΔC_{ILAW}	Triangular	$ 11,100/MT ILAW	$13,800/MT ILAW	$16,600/MT ILAW

Estimated Capital Costs

Capital costs were estimated from Equation 1.

$$C_{CR} = \mathrm{C}_{LAW} \cdot \left(\frac{A_{CR}}{A_{LAW}} \right)^{n_{CR}} \qquad \text{(Eq. 1)}$$

where A_{CR} = area of the caustic-recycle facility (ft^2)
A_{LAW} = area of the LAW vitrification facility (260,000 ft^2)
n_{CR} = scaling factor exponent (–)
C_{CR} = estimated capital costs of the caustic-recycle facility ($)
C_{LAW} = capital costs of the LAW vitrification facility ($).

For the Monte Carlo simulations used in this cost-benefit analysis, the range of values for the scaling factor and the facility-size ratio are presented in Table I. The low value of one-third for the scaling-factor exponent was selected because it was used as a basis for estimating vitrification costs in a report by the National Research Council [8]. The median value of one-half was selected based on the use of this value to scale nuclear power plants [9]. The high value of two-thirds was selected because of the use of a scaling factor of 0.7 for Hanford treatment and immobilization facilities in the Curtis thesis [7].

Estimated Facility Size

The caustic-recycle facility size and resulting capital cost are significant factors in the overall costs. The following section describes the process and the facility housing the process equipment. This description is the basis for the facility size in the model.

Facility Description

The overall process system provides for recirculation loops of LAW and NaOH that pass through multiple electrochemical modules in parallel. Each module contains many membranes and cells. The flow loops are supported by buffer tanks to receive and transfer material to the WTP-pretreatment facility. The system would reside in a contact-maintainable facility, although most reactor maintenance would be performed in a separate area for as-low-as-reasonably-achievable (ALARA) reasons and to maximize operating efficiency. Replacement electrochemical modules would be available for rapid change-out, while the module requiring maintenance would be moved with a crane to a separate area. All the tanks and piping would be fabricated with stainless steel to reduce fire loading, while significant parts of the cells and pumping systems may be polyethylene.

The facility contains process and building-ventilation systems to mitigate environmental releases and for worker protection. The power supplies for the modules are large and would be placed in a room alongside the main processing room as seen in the layout in Fig. 2. The LAW vitrification facility has similar radionuclide content and dose rates, and is the reason it was used for scaling cost for the sodium-recycle facility.

The building is proposed to be a canyon-type facility with an overhead crane. The central canyon would be enclosed by a concrete wall with electrical cabinets, controls, and cooling-water services supplying the electrochemical system from rooms on the opposite side of the canyon walls. The building will consist of an electrochemical-cell area, approximately 60 feet by 35 feet. Twenty electrochemical-cell modules will be contained in this area, using a ratio of 100 ft^2 per module. The remainder of the facility will consist of feed and product tanks, piping, and pumps. The center section, where the tanks will be located, will be approximately 80 ft long and 35 ft wide. The total facility footprint will be approximately 70 feet by 120 feet. The sum of 6420 ft^2 is reached by subtracting out the area for the electrochemical modules.

The system layout is based on the following functional requirements and assumptions:

- process system shall be able to produce 1600 metric tons/year of NaOH from Hanford LAW waste with a 60% operating efficiency
- electrochemical cells shall transfer 23,000 MT of sodium from the waste over 27 years
- the Hanford LAW waste has been aluminum leached and ion exchanged
- facility design life shall be for greater than 27 years
- process system shall be capable of operating for 12 hours without transfers to or from WTP
- system shall produce a 50 wt% NaOH solution at a rate of 8.0 liters/minute
- system components shall be designed to allow for remote replacement or removal (crane and impact wrench) for all equipment with life expectancy of less than one year, and contact maintained for other equipment
- contamination from the process shall be confined and controlled within the processing area
- recycling availability of 23,000 MT of Na.

One end of the processing area will be used for an airlock, which will allow module change-out and maintenance. The airlock will include a containment area to service the electrochemical modules and will have a connection to the Feed Receipt Tank for flushing the modules. The airlock will also function as a service area for the LAW receipt tank. Next to the airlock will be a truck bay for bringing in equipment. This will also allow for the eventual loading out of waste from failed-process equipment. The truck bay will be serviced by an overhead crane. The other end of the building, outside the process area, will contain cooling-water heat exchangers, cooling-water pumps, process-ventilation filters, ventilation blowers, and other miscellaneous building services. The building layout is shown in Fig. 2.

RESULTS

The results discussed in this section include the return on investment (ROI), the cost savings, the capital costs, the operations and maintenance (O&M) costs, and the sale and production costs. A sensitivity analysis on the Monte Carlo results is also presented.

Return on Investment

Monte Carlo simulations were performed using the model described above. A decision matrix was established in which the amount of sodium added for leaching purposes was varied between 10,000 MT and 50,000 MT in 10,000-MT increments. The amount of sodium recovered was fluctuated to correspond with an aluminate-saturation temperature between 25ºC and 100ºC in 15ºC increments. The simulation results in 30 scenarios to map out the feasible operating region for a caustic recycle facility.

In each scenario, 1000 realizations were performed. The probability distribution for ROI was calculated for each of these scenarios and the results shown in Fig. 3. The 10% line represents the set of points in which 10% of the realizations are below a particular ROI at a given amount of sodium recycled. Likewise, the 50% and 90% lines represent the points at which 50% and 90% of the realizations are below a particular ROI. Interestingly, the breakeven point for each of these cases is about 5000 MT sodium recycled.

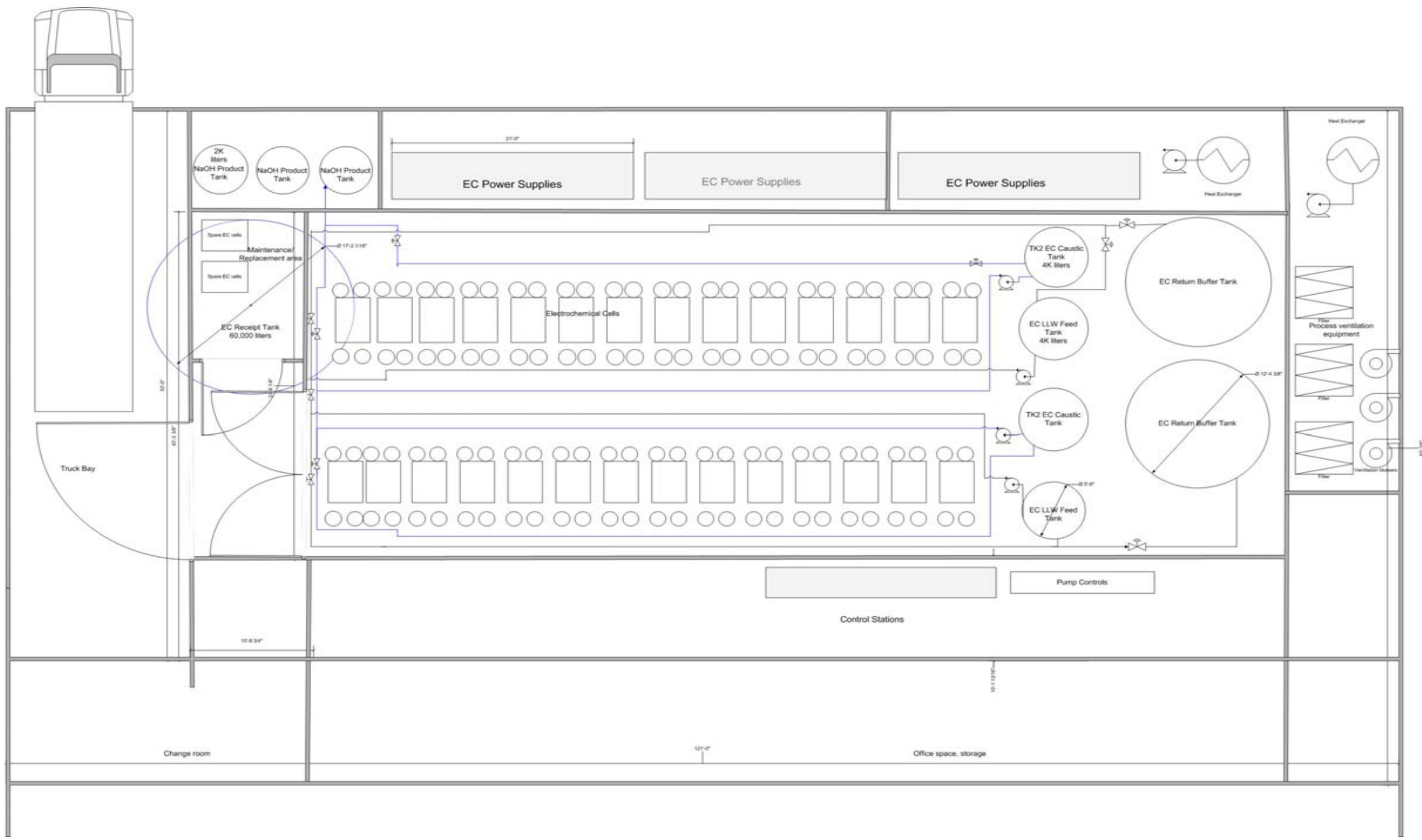

Fig. 2. Preliminary Na Recycle Plant Layout.

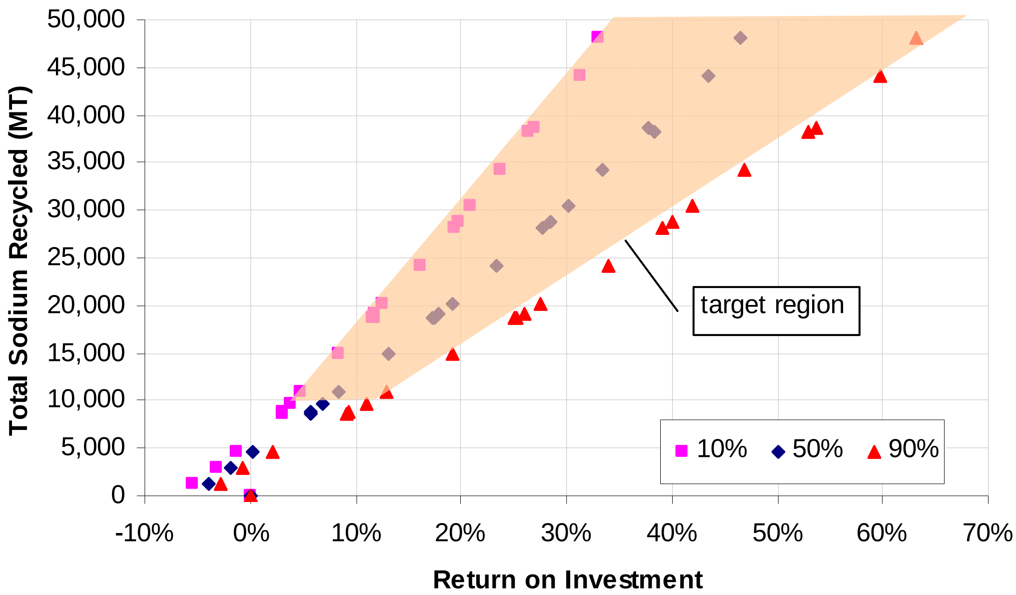

Fig. 3. ROI for Various Amounts of Caustic Recycled.

A threshold region for minimal plant economic feasibility is 3%–12% ROI, centered on approximately 7.5%. This corresponds to approximately 10,000 MT sodium recycled for each case.

Approximately 30,000 MT to 40,000 MT of sodium must be added during caustic-leaching operations to reach the aluminate saturation level at 25ºC. The pretreated LAW would then be processed through the cesium ion-exchange process under less-than-saturated conditions. The pretreated LAW—with cesium removed—would then be used as a feed to the caustic-recycle facility. Recovering any sodium from this point will supersaturate the solution at 25ºC. For instance, if 30,000 MT of sodium is added for leaching and 20,000 MT of the sodium is recovered in the caustic-recycle facility, the solution would be saturated with aluminate at 70ºC and be supersaturated at 25ºC.

Sensitivity Analysis

Fig. 4 shows a sensitivity analysis on the Monte Carlo results in the form of a tornado chart. The figure indicates that to obtain a more accurate forecast on ROI, the following major questions must be answered:

1. How much sodium will be added for caustic leaching?
2. What level of supersaturation can be tolerated?
3. What is the cost savings from preventing additional ILAW-glass production?
4. What are the total capital costs for the caustic-recycle facility?

5. What are the operating and maintenance costs for the caustic-recycle facility?

An improved cost-benefit analysis for caustic-recycle facilities should focus on answering the questions in the order presented.

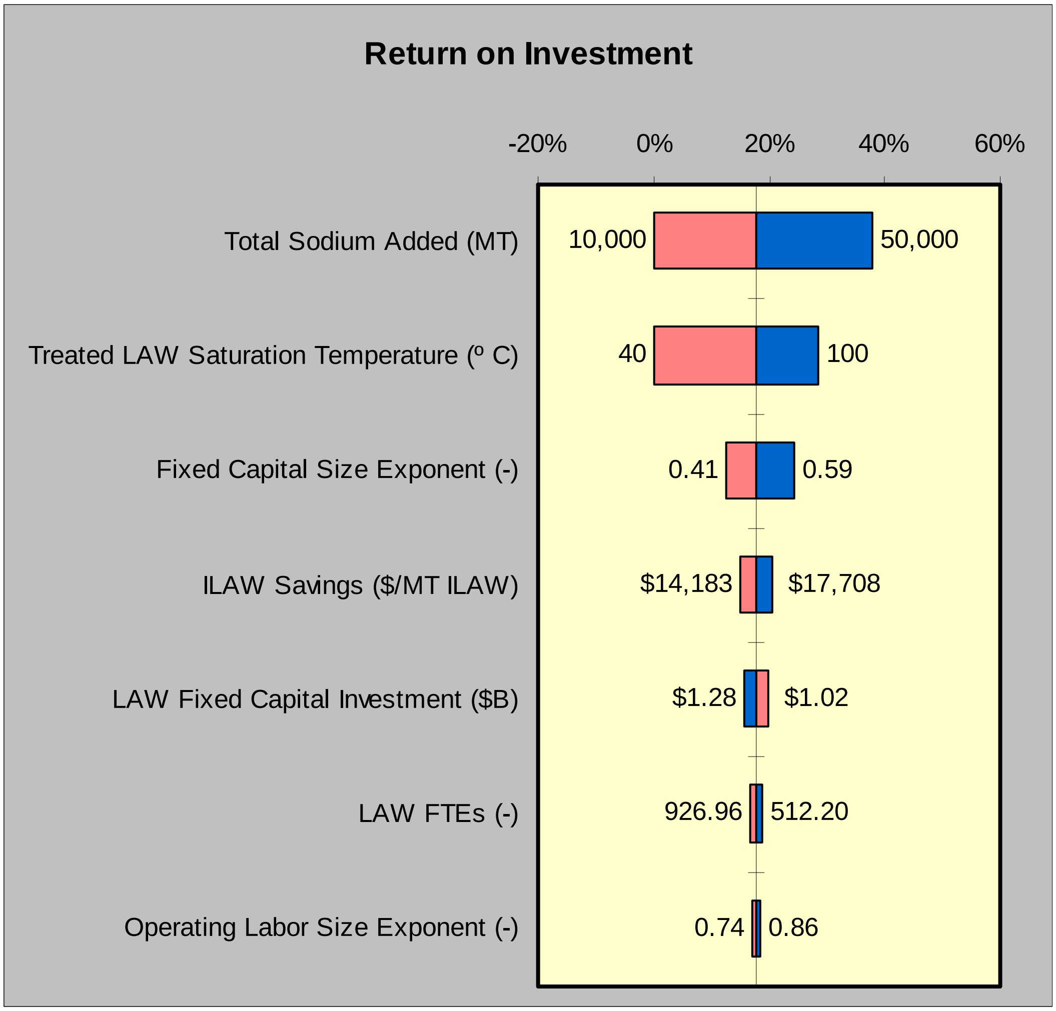

Fig. 4. Tornado Chart Illustrating the Sensitivity of the Model Parameters on ROI.

CONCLUSIONS

The major conclusions from the Monte Carlo model results discussed in this report are summarized below:

- A feasible region for minimal plant economics (e.g., 10% ROI) corresponds with approximately 28,000 MT sodium recycled for the 10% case, 23,000 MT sodium recycled for the 50% case, and 17,000 MT sodium recycled for the 90% case.

- Recycling 17,000 MT to 28,000 MT of sodium would require 25,000 MT to 50,000 MT of sodium addition. This results in aluminate-saturation ratios (i.e., ratio of aluminate concentration in sample over aluminate concentration at saturation) between 1.5 and 5. Bench-scale tests with actual waste have been conducted at a saturation ratio of 8.

- A minimum of 20,000 MT of sodium must be added for caustic leaching to achieve a reasonable ROI. In this case, the resulting LAW sodium-recycled product will have a saturation ratio of approximately 5. If 40,000 MT of sodium is added for caustic leaching, a reasonable ROI is achieved at a saturation-ratio range of 1.5 to 3.

- Recycling 20,000 MT of sodium results in a cost savings in ILAW glass of $700 million to $1.2 billion. If 30,000 MT of sodium is recycled, $1.2 billion to $2.2 billion would likely be realized. Total saving minus capital cost is hundreds of millions.

- Recycling 20,000 MT of sodium results in an estimated range of total capital cost for the caustic-recycle facility to be $310 million to $520 million. If 30,000 MT of sodium is recycled, $320 million to $550 million would likely be realized.

- If 20,000 MT of sodium is recycled, a specific production cost is estimated to be in the range of $30/kg to $40/kg.

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