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Computational Analysis of the SRS Phase III Salt Disposition Alternatives

by

R. A. Dimenna

Westinghouse Savannah River Company
Savannah River Site
Aiken, South Carolina 29808

M. V. Gregory

T. Hang

P. K. Paul

F. G. Smith III

G. A. Taylor

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R. A. Dimenna, M. V. Gregory, T. Hang, P. K. Paul, F. G. Smith, III, G. A. Taylor
Westinghouse Savannah River Company
Savannah River Technology Center
Aiken, SC 29808
Phone: 803-725-8203
Fax: 803-725-8829
Email: richard.dimenna@srs.gov

ABSTRACT

In late 1997, the In-Tank Precipitation (ITP) facility was shut down and an evaluation of alternative methods to process the liquid high level waste stored in the Savannah River Site High Level Waste storage tanks was begun. The objective was to determine whether another process might avoid the operational difficulties encountered with ITP for a lower cost than modifying the existing ITP plant. The analysis described in this report was part of a structured approach to evaluating proposed alternatives on a common basis to identify the best one. Results from the computational analysis were a key part of the input used to select a primary and a secondary salt disposition alternative.

This paper describes the process by which the computational needs were identified, addressed, and accomplished with a limited staff under stringent schedule constraints.

INTRODUCTION

In late 1997, the In Tank Precipitation facility was shut down and a process of evaluating alternative methods to process the liquid high level waste stored in the Savannah River Site High Level Waste storage tanks was begun. The original SRS solicitation for salt disposition alternatives resulted in more than a hundred suggestions. In Phase I, the list of candidate processes was pared to eighteen. These were subjected to a more detailed quantitative analysis (Phase II) addressing technical feasibility, perceived risk, and estimated cost. A key part of the Phase II analysis was the use of a flowsheet model for each alternative to determine the rate at which the liquid waste, or salt solution, could be processed and converted into a stable form for permanent storage. The result was a reduction from eighteen alternatives to four. Phase III was a still more detailed analysis of the four selected alternatives (Dimenna *et al.* 1999). The analytical models used for the Phase III analysis were modifications of those used for the Phase II analysis to include more physical detail in the models themselves and a better representation of the feed streams for the various options. The result was an improved assessment of the performance of each option under the most limiting conditions each was likely to experience during the lifetime of the plant.

The focus of this paper is the process by which the large amount of work for the Phase II and Phase III analysis was accomplished with limited staff in a short amount of time. The project originated as a ten-week effort during which a list of approximately eighteen candidate processes would be identified by a qualitative description of each. The analytical effort would then define each process quantitatively, assess it for performance potential, and evaluate it against all of the others on the list on a common basis. Part of the quantitative assessment of each option included incorporating applicable results from an ongoing experimental program into the models being developed with SPEEDUP™, a flexible, modular, dynamic equation solving package. With only ten weeks to evaluate all of the processes and a base case representing the existing ITP process, it was critically important that the objectives of the analysis be carefully delineated and that the analytical work be strictly limited to address only those objectives.

The Phase III effort was to perform a more detailed analysis of the four processes selected as a result of the Phase II work. It was similar to the Phase II analysis in that the models employed to evaluate the four options came from those used for the Phase II work, but they were modified to address more details. Phase III was not part of the ten-week effort, but it followed directly from the Phase II work and it used the same approach to defining the objectives and carefully controlling the scope of the analysis.

The discussion below begins with the approach to defining the problem. It then moves to assigning resources to that problem, and concludes with a brief description of the four processes selected for further analysis. Some of the computational results that were used to support the final decision making will be presented as an example of the computer model output.

OVERALL ANALYSIS APPROACH

The fundamental approach used to complete the ten-week analytical task in the shortest time possible can be reduced to three essential steps:

1. Define the objectives of the analytical effort.
2. Define the method by which the objectives will be accomplished.
3. Keep all work focused on the task objectives.

These three steps can be described as common sense, but without the focus they demand, analyses can become excessively detailed, take too long to complete, and fail to satisfy the basic needs of the customer. Therefore, it helps to use them as a means to assess all modeling options and requests for added detail, and to limit the analyses to only those phenomena which support the decision making process.

Define the objectives

A team approach was used to address all aspects of the analytical tasks including problem definition. A "Definition Team" was

made up of engineering personnel familiar with the various processes being proposed. Their initial goal was to describe the criteria against which each of the candidate processes would be evaluated, and thereby help define the objectives of the computer analysis. Because the ultimate goal of the processes being evaluated was to immobilize radioactive waste, the objectives of the analysis were to supply information such as the number of glass canisters produced per year, the waste loading in each canister, the properties of the glass, and the rate at which the liquid waste in the High Level Waste storage tanks was being removed. Other objectives included flow rates and composition of associated waste streams that would have to meet regulatory requirements. A second team of engineers, the "Modeling Team," reviewed each of the objectives as it was proposed to ensure that it could be accomplished with enough accuracy to be meaningful in the time available.

Define the method

Defining the method to accomplish the analytical objectives required the close cooperation of both the Definition and Modeling teams. The first step was to define the level of analytical detail needed to satisfy the evaluation requirements for each process. For instance, cycle times in the DWPF were known to be affected by the amount of water introduced in the input streams, and cycle time affected the glass production rate, one of the key objectives of the analysis. A pressure and temperature equilibrium calculation could be used to estimate the water evaporation rate, but it would have been more time consuming than a simple mass balance and would have required more development time to ensure that it was computationally stable. The teams agreed that the DWPF model could use an empirical boil off rate based on previous DWPF operations as a reasonable approximation of system performance and still obtain a good estimate of cycle time.

With modeling assumptions tailored to the accuracy needs of the review group, the Definition and Modeling teams developed process flowsheets for each of the eighteen options and the base case. Part of the flowsheet development included the governing equations to be used to write computational models of the processes. The goal was to define equations which approximated a proposed process simply enough to be calculated quickly, but accurately enough to evaluate the option.

The Definition team also defined which parameters were required to evaluate the various options. The Modeling team formulated the output to match those requirements. A constant focus on the objectives kept the list of output parameters to the minimum needed to assess the calculations and evaluate the options. As the modeling task progressed, the Definition team changed from defining the processes to evaluating the calculations. After several iterations between the teams, the final results for each option were assimilated and cast into a common form for input to the design panel.

The number of candidate processes evaluated in Phase II was too great for all to be used here as examples, so the processes described in this paper will be those evaluated in Phase III. The discussions which follow are applicable to all of the options studied, but where examples or descriptions are needed, these four will be used.

1. Small Tank TPB Precipitation
2. Caustic Side Solvent Extraction
3. CST Non-Elutable Ion Exchange
4. Direct Disposal in Grout

Keep the work focused

Several general modeling assumptions and approximations were made to simplify the models, ensure that components of each model were interchangeable, and ensure that the necessary output information would be calculated. They included:

1. Identifying required output. Model development was aimed at providing the required information needed to evaluate specific salt alternative processing options. The models were simplified to include only those processes that affected the output parameters. To accomplish this goal, the Modeling Team required that the Definition Team specify the required parameters and an acceptable degree of approximation. Once a set of output parameters was defined, a standard output format was agreed upon and all results were reported in this format.

2. Use of a common vector structure. The salt vector consisted of 51 compounds. The same structure was used for all of the salt alternative models, even in situations where most of the constituents were not present. The DWPF models also used a standard vector structure, but the DWPF model had to accommodate both sludge and salt inputs. A 27-component sludge vector was used for the average sludge composition for a 20-year period.

The 51-component salt vector from the salt models had to be modified to mesh with a 45-component salt vector used in the DWPF model. To accomplish this, a special module was written as an interface between the salt models and the standard DWPF models. The translator module accepted a 61-component salt stream which included the materials introduced by the various salt processing options, as well as the standard 51 components comprising the common salt vector. Each of the 61 components was broken into compounds included in the 45-component DWPF salt vector such that, with only a few exceptions, the mass of each element was conserved. Because the glass forming components were already carried in oxide form and no chemical reactions were modeled in the DWPF module, this approach was an acceptable approximation to a DWPF mass balance.

3. Defining a standard input set. A standard input set was used to evaluate all the options in Phase II. It was based on a 20-year average for both the sludge and salt input streams and formed a common basis for all of the cases evaluated. The actual sludge stream was modified to accommodate the actinide removal process with monosodium titanate (MST) prior to processing the salt stream in any of the options. In Phase II, the MST stream was added to the sludge vector to ensure that the actinides were included in the DWPF calculation without complicating the DWPF model. In the Phase III analysis, the MST stream was separated from the sludge stream and input to the DWPF calculation explicitly.

The sludge stream assumptions were the same for Phases II and III, but the salt stream assumptions changed. Rather than use a 20-

year average for the salt stream, the blending operation as the various tanks are emptied was modeled and a time dependent salt input vector was determined. The options evaluated in Phase III were then driven with the limiting salt vector conditions for each process.

Model Structure

The computational models evolved from two models that had been developed previously, the High Level Waste Integrated Flowsheet Model (IFM) (Gregory *et al.* 1995) and a detailed DWPF model developed by Smith (1998). The IFM was used as a template for the salt processing models, in particular the precipitation option and the salt blending. The modularity of the SPEEDUP™ (Aspen Technology 1993) environment made the task of removing the existing ITP model and replacing it with a modified precipitation model straightforward. Control logic built into the external data interface routines within the IFM SPEEDUP™ model was generally applicable to the revised option of small tanks precipitation.

The models were fundamentally mass balances. The energy equation was not modeled explicitly, although some of the DWPF modules incorporated a fixed evaporation rate to estimate cycle times affected by excess water inventory. Feedback loops were removed from the entire model to prevent potential convergence problems from impacting computational performance. SPEEDUP™ can calculate flowsheets with feedback loops, but we felt the models would be faster to develop and more robust with the feedback loops removed. This mathematical simplification introduced a physical assumption that the feedback loops would not significantly alter the input flow rates or compositions, an acceptable approximation given the simplicity of the model.

An additional simplification was to run the salt alternative models and the associated DWPF calculations separately. The reason was to improve computational performance, as well as to hasten the development of the models. It also avoided the difficulties of coupling the models. Because feedback loops had already been removed, this was not an additional physical approximation.

A sketch of the overall model structure is shown in Figure 1. The modeling differences for the various salt alternatives were in the salt processing option and the specific DWPF modules which were needed to process the salt option effluent.

There were several salt side models and two basic DWPF side models, depending on the needs of a given option. The general salt processing options assessed during Phase II included precipitation, ion exchange, solvent extraction, fractional crystallization, and direct injection to DWPF, which was actually just a DWPF model, and direct disposal in Saltstone. A filter model to remove actinides from the salt stream was used for several of the options. This step took place before the salt stream was introduced to the salt processing module. Most of the salt processing options required that the effluent stream carrying cesium be sent to DWPF for vitrification. Two different DWPF models were developed to account for the different nature of the incoming salt streams. They were:

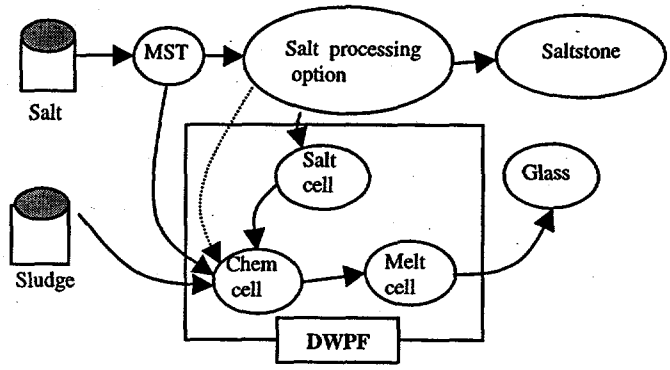

Figure 1. General Model Layout

1. A precipitation model that included the salt cell for treatment of organic precipitate.
2. A direct injection model that introduced the salt stream directly into the SRAT in the chemical processing cell.

Special Process Models

Several special process models were developed as components for most of the salt processing options. A brief description of each follows.

Blending model To provide a general capability to generate time-dependent salt blend compositions, a dynamic SPEEDUP™ model called TFARM was created. Its input was an initial state for each waste storage tank, specified for supernate, salt cake, and sludge regions as appropriate, all extracted from the standard HLW tank farm database. A blending schedule derived from ProdMod (Gregory and Paul 1995) System Plan-like analyses was also required. The dynamic unit operation models in TFARM were all variants of the same basic equation set applied to each tank being represented: ordinary differential equations which track mass, mass fractions (for 34 constituents), and volume changes. Distinct sets of equations were maintained for the supernatant liquid (supernate), salt cake, and sludge regions within each tank as needed.

Alpha Sorption Tank The model feeds salt solution in 60 kgal batches into the Alpha Sorption Tank. MST is added to the tank to complex uranium, strontium and plutonium salts in the solution. Each AST batch is held for 24 hours to simulate the adsorption process. Following the hold time, the AST batch is continuously passed through a 100% efficient filtration step. Output from the filter is separated into filtrate and solids slurry streams. The filtrate is collected in the Salt Solution Holding Tank and from there fed continuously to the grout (Saltstone) plant. The solids are added as an input stream to the SRAT in the DWPF model with the volume prorated over each SRAT cycle.

Defense Waste Processing Facility A DWPF model similar to that used in Phase II was used for each of the four Phase III options. With the exception of the small tank precipitation option, the treated waste streams were fed directly to the SRAT in the Chem Cell. These streams included the salt stream from the

salt processing alternative as well as an additional stream carrying actinides removed from the salt stream prior to cesium removal processing. The precipitation option included a model of the salt cell in the DWPF model. Principal outputs from the DWPF model were the glass canister production rate, the cycle time of the SRAT and SME processes, workoff rates for the sludge and salt inventories, and recycle water volume.

The DWPF model uses a version of the PCCS calculation (Smith 1996) to determine the minimum amount of frit that can be added to each SME batch to make acceptable glass that meets all property constraints, including liquidus temperature, melt viscosity and glass durability (ΔG). Durability was cast into the form of limits on the glass free energy.

ProdMod An integrated computational tool called ProdMod (Gregory and Paul 1995) was developed to simulate the waste processing behavior of the entire HLW complex at SRS for its life cycle operation. The existing ITP was modeled as the salt processing facility in ProdMod. ProdMod is a simplified integrated pseudo-dynamic simulation code based on SPEEDUP™ which uses algebraic equations only. The dynamic nature of the plant processes is modeled in linear constructs in which time dependence is implicit. Figure 2 shows the processes and facilities along with the main flow streams modeled. ProdMod provides HLW facility process and flow stream information for the entire range of simulation.

ProdMod was modified to reflect each new salt disposition option and used to evaluate that option for the entire life cycle to clean out all the waste tanks.

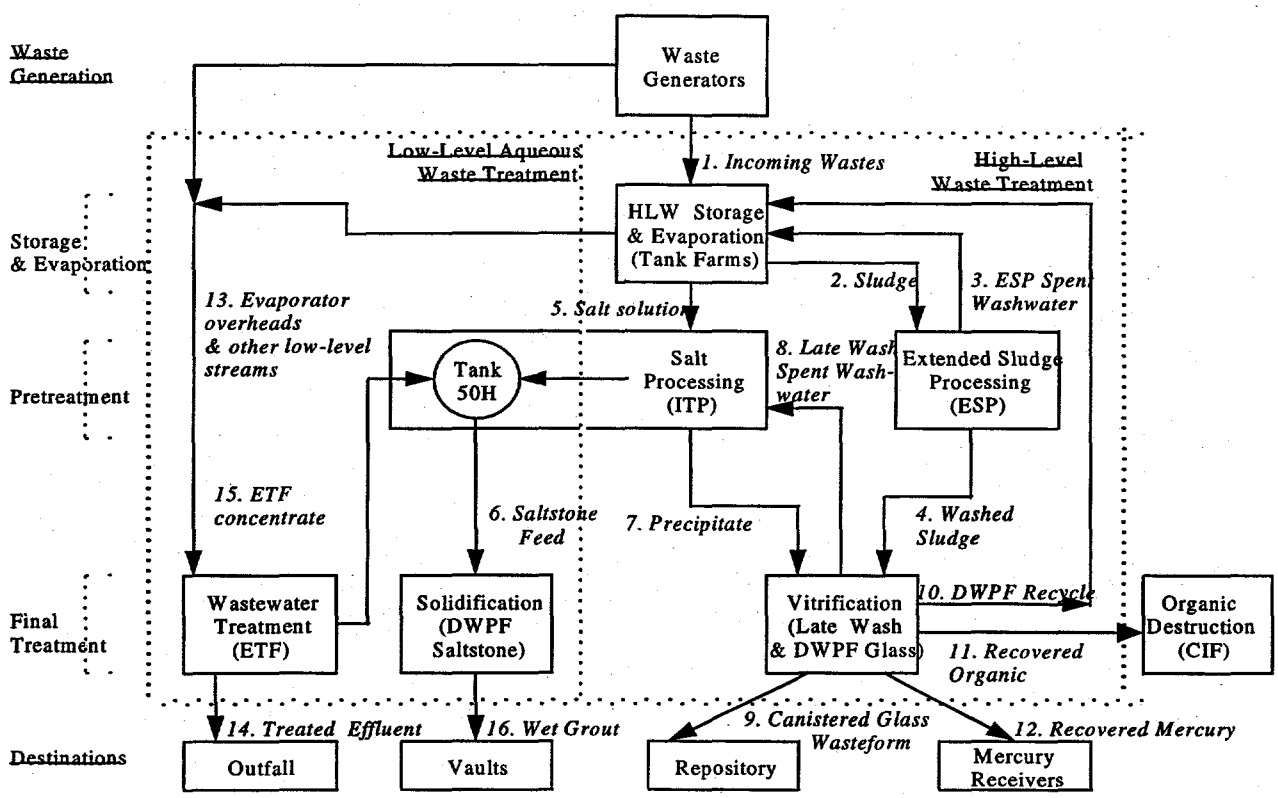

Figure 2. SRS HLW Process Streams Modeled in ProdMod

SALT PROCESSING ALTERNATIVES

Small tank TPB precipitation

The SPEEDUP™ representation of the Small Tank TPB Precipitation alternative was a comprehensive dynamic model of the overall process. Precipitation kinetics, filtration, solubility, evaporation, decomposition, absorption, and dissolution were treated in full dynamic detail as part of the mass balance with over 34 distinct chemical species. The basic building block of the models was the original HLWIFM ITP model, with modifications to suit the new small tank design. The main extension to the original model was the addition of the dynamic equations to calculate benzene generation due to soluble NaTPB decomposition in the presence of copper or palladium catalyst.

The following table lists summary results from the DWPF calculation for the Small Tank TPB Precipitation process.

Can production: .....308.817 per year
Glass properties:
Durability: ..... -7.271
Viscosity: 47.424
Liquidus: 1004.035
Effective Percent_Ti: 0.317
Recycle volume: ..... 1328.252 kgal/yr
Organic waste: ..... 59.557 kgal/yr
NO2: ..... 61.709 klbm/yr

In addition to this overall summary, a process summary showing the results of the DWPF calculation for each option were provided. This allowed the Definition team to compare the overall model results with input concentrations and flowrates to ensure that at least an approximate mass balance had been obtained. A process summary for one of the small tank runs is shown below.

PROCESS SUMMARY

	Item	Mass (klb/yr)	Vol (Kgal/yr)
57	PRFT to PR	4255.237	494.393
60	Copper nitrate	270.034	31.645
61	Formic to PR	205.728	20.726
68	Benzene to CIF	431.513	59.557
81	Sludge to SRAT	2835.358	310.253
82	MST to SRAT	5.154	0.621
83	Formic to SRAT	97.819	9.855
84	Nitric acid to SRAT	124.621	12.483
85	NaNO2 to RCT	222.188	20.963
86	Caustic to RCT	1149.015	101.532
87	Recycle H2O to TnkFrm	11178.643	1328.252
92	Melter feed	2848.909	250.942
96	Formic to SME	61.591	6.205
97	Process frit to SME	1149.261	93.074
97A	Decon frit to SME	1216.374	142.716
98	SRAT to SME	2568.937	274.281
99	PRBT (Salt) to SRAT	5489.802	647.468
100	Glass	1191.355	55.653

Caustic Side Solvent Extraction

The SPEEDUP™ model for the solvent extraction process was completed and benchmarked against data. Calculations with the SPEEDUP™ model for the TRUEX process using the parameters reported in a paper by Leonard and Regalbuto (1994) compared well to literature values reported in the referenced paper using the SASSE model.

The alpha decontamination and grout production parts of the model were the same as those used with the other options. The solvent extraction process adds a continuous counter-current solvent extraction operation to remove cesium from the salt solution in the SSHT prior to grouting. The solvent extraction model consists of three units: an extraction unit where cesium is removed from the salt solution into an organic solvent, a scrubber where 0.5 M NaOH is added to the aqueous phase that then enters the extraction unit, and a stripper where 0.01 M nitric acid is used to strip the cesium from the organic phase back into the aqueous phase. Parameters used for the model include the number of stages in each unit and the cesium distribution coefficients. The model assumes that cesium is the only component transferred between the different phases in the solvent extraction process.

The following table lists summary results from the DWPF calculation for the Solvent Extraction process.

Can production: .....426.479 per year
Glass properties:
Durability: .....-7.642
Viscosity: 33.889
Liquidus: 999.837
Effective Percent_Ti: 2.724
Recycle volume: ..... 4057.098 kgal/yr
NO2: ..... 51.372 klbm/yr

CST Non-Elutable Ion Exchange

The SPEEDUP™ model for the CST Non-Elutable Ion Exchange column describes the ion exchange loading of Cs onto the granular CST fixed bed as an adsorption process. The computational method applicable to an unsteady operation of a packed column provided by Bird et al. (1960) is used. For simplicity, the method involves a number of assumptions:

1. The solution contains only two components, i.e. a solvent and a single solute.
2. No axial dispersion* or back-mixing of the fluid. The solution volumetric flow rate through the column is constant.
3. Solute concentration is uniform over any cross section.
4. Equilibrium is established between the solute concentration adsorbed on the solid and the local solute concentration in the solution.
5. Solute concentration in the solution is small, so the operation may be considered isothermal.
6. The solid bed only adsorbs the solute from the solution. Hence, no competing adsorption occurs.
7. Resistance of the solid to mass transfer is negligible.

The CST IX process is operated as follows. (Beck 1998) A three-column carousel arrangement is assumed. This arrangement includes a lead column followed by a guard column to assure sufficient decontamination. The third column is maintained standby loaded with fresh resin. A cycle is completed when either of the following criteria is satisfied: (1) Molar concentration ratio C/C_o for Cs at the exit of the lead column reaches 0.9, (2) the Cs concentration in the liquid at the exit of the guard column reaches 1.3 micrograms Cs/L (or 20 nCi/g). At the end of the cycle, the loaded lead column is rotated out of service, the guard column becomes the lead column, and the standby column becomes the guard column. The resin is slur-

* Neglecting axial dispersion proved to be a significant assumption in model development work completed after this project was done. The SPEEDUP column model was enhanced to include axial dispersion in an effort to understand differences between our results and those of outside contractors. When this term was included, the SPEEDUP model agreed closely with independent column calculations.

ried from the loaded column for transfer to DWPF, fresh resin is added, and this column then goes to standby.

Two cases were run to determine the cycle time. In both, the results showed that Cs was totally removed within the first two columns. All cycles were completed because of the first requirement (i.e. C/C_o for Cs at the exit of the lead column = 0.9) being satisfied. Due to the fresh resin loaded in the lead column at the start of the operation, the cycle time was longer in the first cycle, then approached a stable value in subsequent cycles. The cycle time was about 960 hours for Case 1, and about 650 hours for Case 2.

The DWPF model was modified to accommodate the CST which was unique to this option. Rather than modify the composition vector for all applications, a position with a known concentration of zero was used for the significant CST constituent which had to be tracked. Results for the CST runs were similar in format to the previous cases already shown, so they are not shown here.

Direct Disposal in Grout

A one-dimensional thermal saltstone model was developed and enhanced by M. A. Shadday (personal communication) to determine the effect of Cs-137 activity on the temperature response of the grout. Results indicated that for anticipated cesium loading in the grout, the radioactive energy source term was a small but noticeable addition to the energy of hydration released during the curing process. Calculations showed acceptable temperatures at pour rates high enough to satisfy estimated operational requirements. DWPF and Saltstone calculations were also made for this case to show material balances around the processes, but they did not need to be coupled to the thermal analysis.

SUMMARY

Adherence to a well defined list of objectives and a strict assessment of all model details against that list allowed us to complete the entire process of defining and evaluating nineteen different process options in only fourteen weeks. The selection of SPEEDUP™ as the principal analytical tool allowed development of fairly simple models such as mass balances in the initial phase of the project, yet provided for later addition of more complexity with no penalty in either model or development efficiency. Additional relationships such as energy balances, additional chemical reactions, or more chemical species could be included with no rework of existing models. Some model additions to be considered for the design phase of the project are:

1. Couple the facility models computationally so that an integrated plant response can be evaluated.
2. Identify additional chemical species that must be tracked within the model.
3. Develop energy balances for those facilities where temperature dependence is important.
4. Develop a parallel computational structure to allow additional detail to be included without degrading the calculational performance of the model.

NOMENCLATURE

C, C_0	Concentration, initial concentration
CST	Crystalline Silicotitanate
DWPF	Defense Waste Processing Facility
HLW	High Level Waste
HLWIFM	High Level Waste Integrated Flowsheet Model
IFM	Integrated Flowsheet Model
ITP	In-Tank Precipitation
MST	Monosodium Titanate
NaTPB	Sodium tetraphenylborate
PCCS	Process Composition and Control System
SRAT	Slurry Receipt and Adjustment Tank
SRS	Savannah River Site
SRTC	Savannah River Technology Center
SSHT	Salt Solution Holding Tank
TPB	Tetraphenylborate

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