Gas Generation and Bubble Formation Model for Crystalline Silicotitanate Ion Exchange Columns

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R0197663

by

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DOE Contract No. **DE-AC09-96SR18500**

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WSRC-TR-2000-00177, Rev. 0

Keywords: Waste processing, ion
exchange, modeling, gas
generation, bubble formation

Retention: Permanent

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FOR CRYSTALLINE SILICOTITANATE ION EXCHANGE COLUMNS**

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Publication Date: June 16, 2000

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SUMMARY

The authors developed a transient model to describe the process of gas generation due to radiolysis and bubble formation in crystalline silicotitanate (CST) ion exchange (IX) columns using the Aspen Custom Modeler (ACM) software package. The model calculates gas concentrations and onset of bubble formation for large CST IX columns. The calculations include cesium loading as a function of time, gas generation as a function of cesium loading, and bubble formation as a function of gas solubility.

Using this model, we made a number of predictions to examine the effect of feed stream composition, air saturation of feed stream, temperature, and feed stream flow rate on gas generation and bubble formation. Additionally, the model calculated radiolytic gas generation and bubble formation during the three-column CST IX operation to process the average waste feed. The results indicate the necessity to install gas disengagement devices between Columns 2 and 3 to mitigate the formation of gas bubbles in Column 3.

INTRODUCTION

The Salt Disposition Systems Engineering Team identified non-elutable ion exchange as one of the preferred alternatives to replace the In-Tank Precipitation process used to decontaminate soluble radioactive waste at the Savannah River Site (SRS) (Dimenna et al., 1999). The CST sorbent in the engineered form (UOP IONSIV® IE-911) serves as the ion exchange medium in this process option. Previous testing with simulated and radioactive waste solutions demonstrated the feasibility of the process (Walker et al., 1998; Walker, 1999 [b]; Walker et al., 1999 [a]). Tests previously performed in simulated waste under irradiation conditions similar to process operations also showed the formation and growth of bubbles. These findings raised questions regarding potential effects of gas bubbles on IONSIV® IE-911 performance. A performance tests plan (Walker et al., 1999 [b]) addresses these operational issues and additional areas. As part of this plan, we developed a computer model to calculate gas concentrations and onset of bubble formation for large CST IX columns as a function of location in the column and cesium loading. The model results together with experimental data will estimate the magnitude and effects of gas generation during CST IX process operations. This report summarizes the model development and predictions.

MODEL DEVELOPMENT

Assumptions

A prior report discusses the assumptions involved in the porous particle diffusion model for predicting the loading of cesium on the CST fixed bed (Hang and Dimenna, 2000). We added the following assumptions to predict the radiolytic gas generation and bubble formation during the CST IX operations.

1. Gas volume negligible compared to the solution volume.
2. Constant volumetric flow rate.
3. Homogeneous nucleation as the only mechanism to form bubbles.
4. Bubble formation mechanism in the mobile phase applicable to the pore phase.
5. Bubble velocity equals interstitial velocity of the mobile phase.

Material Balance

The porous particle diffusion model provides the material balance equations for cesium in both the mobile phase and the pore phase (Hang and Dimenna, 2000). The following equations account for the gas concentrations:

$$V \frac{dC_{i,out}}{dt} = C_{i,in}F_{in} - C_{i,out}F_{out} + R_i \quad (1)$$

$$R_i = R_{i,g} + R_{i,b} + R_{i,d} \quad (2)$$

The model tracks three gases (i.e., H₂, N₂, and O₂) in two phases (i.e., dissolved in the solution, and in bubbles). We account for two sources of gas: (1) that present in the feed as dissolved gas from air (i.e., N₂ and O₂), and (2) that radiolytically generated in the column (i.e., H₂ and O₂). The model requires specification of the concentration of dissolved gas in the feed stream at the start of the calculation.

Gas Generation

Hydrogen and oxygen form radiolytically in the waste stream. Hydrogen forms from the breakdown of water. Oxygen forms by breakdown of water and decomposition of sodium nitrate. The following reactions depict the stoichiometry to form hydrogen and oxygen.

Hydrogen

Oxygen

If $G_{H_2} < 2G_{O_2}$:

If $G_{H_2} = 2G_{O_2}$:

If $G_{H_2} > 2G_{O_2}$:

Hydrogen and oxygen generation rates are calculated from experimentally measured G values (i.e., the radiolytic yields for the gases) and dose rate. The G value varies for each gas and for the feed stream composition. The G values typically vary between 0.001 and 1 molecules/100 eV. Appendix A gives the G values for a column packed with UOP IONSIV® IE-911.

Assuming an adiabatic column, the equation used to calculate the generation rate of gas i follows.

$$R_{i,g} = 5.07548 \times 10^{-3} G_i C_{Cs} \quad (9)$$

C_{Cs} is the total cesium concentration loaded on the CST bed. The constant 5.07548×10^{-3} derives from the following combination of factors.

$$5.07548 \times 10^{-3} \frac{\text{mol}_i}{\text{mol}_{Cs} h} = \frac{\text{molecules}}{100 \text{ eV}} \times \frac{4.95 \text{ Watts}}{10^3 \text{ Ci}_{137Cs}} \times \frac{1 \text{ Ci}_{137Cs}}{0.01153 \text{ g}_{137Cs}} \times \frac{137 \text{ g}_{137Cs}}{\text{mol}_{137Cs}} \times$$

$$\frac{0.23124 \text{ mol}_{137Cs}}{\text{mol}_{Cs}} \times \frac{1 \frac{\text{eV}}{\text{s}}}{1.602 \text{ E} - 19 \text{ Watts}} \times \frac{1 \text{ mol}_i}{6.022 \text{ E} + 23 \text{ molecules}} \times \frac{3600 \text{ s}}{h}$$

All of these factors represent physical constants except for the 0.23124 ratio of ^{137}Cs to the total cesium in SRS waste (Dimenna et al., 1999). The combination of factors calculates the number of curies of ^{137}Cs in the waste from the total cesium concentration (radioactive and non-radioactive); then converts this value to the decay energy (in eV). The model assumes that all

the decay energy from ^{137}Cs and its daughter, Ba-137m, except the neutrino from the beta decay of ^{137}Cs , is absorbed within the column at the location of decaying nucleus. Deposition of gamma ray energy away from the cesium loading zone is not included in the model.

Bubble Formation/Dissolution

According to the homogeneous nucleation theory, bubbles form when the difference between the total dissolved gas pressure (P^*) and the local pressure in the surrounding liquid (P) exceeds the surface pressure (Kwak and Kim, 1998; Li and Yortsos, 1995).

$$P^* - P \geq \frac{2\gamma}{r_b} \quad (10)$$

Due to the lack of surface tension (γ) data for the SRS waste and the difficulty determining bubble radius r_b , we used the following criterion for the bubble formation.

$$P^* - P \geq 0 \quad (11)$$

Equation (11) predicts an earlier onset of bubbles, hence provides more conservative results than those obtained from Equation (10).

Based on Equation (11), bubbles form when the sum of gas partial pressures are equal to or larger than the local pressure of the liquid phase. If bubbles already exist, increasing the local pressure to a value larger than the sum of gas partial pressures will cause bubbles to dissolve into the liquid phase. Appendix B summarizes the system pressure profiles in CST IX columns calculated for different feed streams. We approximate the total rate of bubble formation or dissolution (R) by the following equation.

$$R = k(P^* - P) \quad (12)$$

$$(P^* - P) \geq 0: \quad R_b = R \quad (\text{Bubble formation})$$

$$(P^* - P) < 0: \quad R_d = R \quad (\text{Bubble dissolution})$$

To obtain the bubble formation rate for each dissolved gas, one multiplies the total formation rate by the ratio of component dissolved gas concentration to the total dissolved gas concentration. Similarly, one determines the dissolution rate for each gas in the bubbles by multiplying the total dissolution rate by the component mole fraction in the bubble gas phase.

Assuming linear phase equilibrium, one calculates the sum of the gas partial pressures P^* by applying Henry's law.

$$P^* = \sum \frac{C_i}{S_i} \quad (13)$$

Appendix A lists solubilities, S_i , of hydrogen, nitrogen and oxygen for different waste types. We

proposed the following empirical equation for the rate constant k to provide value changes with an S-shaped curve.

$$k = k_0 e^{\frac{-10^{-3}}{|P^* - P|}} \quad (14)$$

We used a value of 1.5 for k_0 . The formulation of k provides fast bubble formation and dissolution rates so that supersaturation of the liquid phase is negligible.

Aspen Custom Modeler Software

We developed the gas generation/bubble formation model using the Aspen Custom Modeler (ACM) software package marketed by Aspen Technology. ACM serves as the new generation of SpeedUp™ used for the previous IX column model (Hang and Dimenna, 2000). Like SpeedUp™, ACM provides the architecture to build complex transient models and equation solvers that prove exceptionally robust and very well suited to solving a system of differential and algebraic equations. In addition, ACM includes a much-improved user-friendly graphical user interface to facilitate model development. The ACM supports the personal computer platform running Microsoft Windows 95, 98, and NT.

Model verification

We tested the model against a series of simple cases. Appendix C contains the results of these test cases.

CALCULATIONS OF FLOWSHEET CASES

We performed a number of calculations for the design IX columns (i.e., 16' length; and 5' diameter) to determine the effects of different process parameters on radiolytic gas generation and bubble formation. These calculations included the following.

1. Effect of gas dissolved in feed stream
2. Effect of feed stream composition
3. Effect of errors in G values
4. Effect of higher temperature
5. Effect of flow rate
6. Three-Column operation

Except in Case 6, we performed all test cases for the first column only. We stopped the calculations after reaching 99.9% breakthrough in Cases 1 to 5. In Case 6, we terminated the simulation when cesium mass concentration at the outlet of column 2 reached 1.3×10^{-6} mg/ml. Table 1 lists the conditions of the six cases. Appendix A provides the chemical compositions of the three waste feeds used in these cases. The following sections discuss the results.

Table 1: Conditions of the Flowsheet Cases

Case	Effect To Be Studied	Conditions
1	Gas dissolved in the feed stream	Average waste with and without air saturation of feed stream @ 21 gpm and 25 °C
2	Feed stream composition	High-OH waste with and without air saturation of feed stream @ 21 gpm and 25 °C
2a.		
2b.		High-NO ₃ waste with and without air saturation of feed stream @ 21 gpm and 25 °C
3	Errors in G values	Average waste with air saturation of feed stream @ 21-gpm-and-25-°C-and 20% increase in G values
4	Higher temperatures	High-NO ₃ waste with air saturation of feed stream @ 21 gpm and 35 °C
5	Flow rates	Average waste with and without air saturation of feed stream @ 10 gpm and 25 °C
6	Three-column operation	Average waste with air saturation of feed stream @ 21 gpm and 25 °C

Case 1 - Effect of Gas Dissolved in Feed Stream

Using average waste composition and the design flow rate of 21 gpm, we performed calculations with and without air saturation of the feed stream at 25 °C. Figures 1 to 5 show the results. Figure 1 shows the CST bed pressure profile. Figures 2 and 4 display the profiles of pressure difference (P^*-P) at different times. Figures 3 and 5 give the concentration profiles of total dissolved gas. As expected, the pressure difference and dissolved gas concentration increase due to air saturation in the feed stream. No bubble formation resulted due to negative (P^*-P). However, no bubble formation resulted because the gas solubility (due to increasing local pressure down the column) increased faster than the gas concentration.

Figure 1: Column Pressure Profile (Case 1 – Average waste)

Figure 2: Pressure Difference Profile (Case 1 - Average waste without air saturation)

Figure 3: Dissolved Gas Concentration Profile (Case 1 - Average waste without air saturation)

Figure 4: Pressure Difference Profile (Case 1 – Average waste with air saturation)

Figure 5: Dissolved Gas Concentration Profile (Case 1 – Average waste with air saturation)

Case 2 - Effect of Feed Stream Composition

This case examined high-OH and high-NO₃ waste compositions. Similar to Case 1, calculations assumed a flow of 21 gpm with and without air saturation of the feed stream at 25 °C. Changes in waste compositions cause changes in density and viscosity which result in different system pressure profiles (See Appendix B). Furthermore, gas generation rates and solubility depend on feed composition. Compared to average waste, gas generation is 23% slower in high-OH waste and 15% faster in high-NO₃ waste. Figures 6 to 10 show the column pressure, the profiles of pressure difference ($P^* - P$) and dissolved gas concentration at different times for high-OH waste while Figures 11 to 15 show results for high- NO₃ waste. Again, no bubble formed under these conditions.

Figure 6: Column Pressure Profile (Case 2 - High-OH waste)

Figure 7: Pressure Difference Profile (Case 2 – High-OH waste without air saturation)

Figure 8: Dissolved Gas Concentration Profile (Case 2 – High-OH waste without air saturation)

Figure 9: Pressure Difference Profile (Case 2 – High-OH waste with air saturation)

Figure 10: Dissolved Gas Concentration Profile (Case 2 – High-OH waste with air saturation)

Figure 11: Column Pressure Profile (Case 2 – High-NO₃ waste)

Figure 12: Pressure Difference Profile (Case 2 – High-NO₃ waste without air saturation)

Figure 13: Dissolved Gas Concentration Profile (Case 2 – High-NO₃ waste without air saturation)

Figure 14: Pressure Difference Profile (Case 2 – High-NO₃ waste with air saturation)

Figure 15: Dissolved Gas Concentration Profile (Case 2 – High- NO_3 waste with air saturation)

Case 3 - Effect of Errors in G values

To evaluate the impact of the uncertainty in G values on gas generation, we increased the measured values by 20%. Using larger G-values, we repeated Case 1 only with the feed stream saturated with air. With increasing G-values, more gases form. Hence, compared to Case 1, the total dissolved gas concentration should increase as shown by Figure 17. Compared to Figure 4, however, Figure 16 clearly indicates that the 20% increase in G-values did not result in a positive value of (P^*-P) as required to form bubbles.

Figure 16: Pressure Difference Profile (Case 3 – Average waste with air saturation and 20% increase in G-values)

Figure 17: Dissolved Gas Concentration Profile (Case 3 – Average waste with air saturation and 20% increase in G-values)

Case 4 – Effect of Higher Temperature

This case determined the effect of higher temperature on bubble formation. We assumed high- NO_3 waste with air saturation in the feed stream at 35 °C. Gas solubility, density and viscosity of the feed all decrease with increasing temperatures (See Appendix A), hence resulting in lesser dissolution of gases in the liquid phase. Additionally, a faster breakthrough should occur because less cesium will load on the CST bed at higher temperatures. Figure 18 displays the column pressure profile. Figures 19 and 20 show the calculated pressure difference and dissolved gas concentration profiles, respectively. Compared with the results at 25 °C displayed in Figures 14 and 15, the model correctly predicted lower dissolved gas concentration and closer-to-zero pressure difference. No bubble formation occurred at this condition.

Figure 18: Column Pressure Profile (Case 4 – High- NO_3 waste at 35 °C)

Figure 19: Pressure Difference Profile (Case 4 – High-NO₃ waste with air saturation at 35 °C)

Figure 20: Dissolved Gas Concentration Profile (Case 4 – High-NO₃ waste with air saturation at 35 °C)

Case 5 – Effect of Flow Rate

In this case, we decreased the feed flow rate by ~50% to 10 gpm. A slower flow rate results in a lower system pressure profile in column 1 as shown in Figure 21 (Also see Appendix B) and a much larger breakthrough time (~7500 hours compared to ~4000 hours in Case 1). With increasing time more gases form and a lower system pressure would favor bubble formation as displayed in Figures 22 and 23. Figure 24 shows that at about 4000 hours bubbles begin to form in the middle of Column 1.

Figure 21: Column Pressure Profile (Case 5- Average waste at 10 gpm)

Figure 22: Pressure Difference Profile (Case 5 – Average waste with air saturation at 10 gpm)

Figure 23: Dissolved Gas Concentration Profile (Case 5 – Average waste with air saturation at 10 gpm)

Figure 24: Bubble Gas Concentration Profile (Case 5 – Average waste with air saturation at 10gpm)

Case 6 - Three-Column Operation

In this case, we tested the ability of the model to compute the SRS design base case of the three-column CST IX operation. Figures 25, 26 and 27 show the predicted pressure difference profile, the dissolved gas concentration profile and the bubble gas concentration profile, respectively, for Column 1; Figures 28, 29 and 30 show the predicted results for Column 2; and Figures 31, 32 and 33 display the results for Column 3. The model predicted no bubble formation in Column 1. In Column 2, bubbles started forming at 3500 hours, then dissolved into the liquid phase due to increasing system pressure. In the model, to avoid negative concentration values, we set the dissolution rate to zero when the bubble gas concentration becomes less than 1×10^{-6} mol/L. Therefore, in Figure 30, the dissolution of bubble gases results in a final concentration value between 1×10^{-6} mol/L and 5×10^{-7} mol/L. At time less than 2500 hours, no bubbles formed in Column 3. Starting from 2500 hours, due to the large pressure drop between Column 2 and Column 3, increasing total gas pressure P^* caused bubbles to form after the waste stream leaves Column 2 and before it enters Column 3. In Column 3, at any given time less than 2500 hours, the dissolved gas concentration remained unchanged with respect to location. From time 2500 hours on, the dissolution of bubble gases caused the dissolved gas concentration to increase and the bubble gas concentration to decrease.

Figure 25: Pressure Difference Profile in Column 1 (Case 6 – Three-column operation)

Figure 26: Dissolved Gas Concentration Profile in Column 1 (Case 6 – Three-column operation)

Figure 27: Bubble Gas Concentration Profile in Column 1 (Case 6 – Three-column operation)

Figure 28: Pressure Difference Profile in Column 2 (Case 6 – Three-column operation)

Figure 29: Dissolved Gas Concentration Profile in Column 2 (Case 6 – Three-column operation)

Figure 30: Bubble Gas Concentration Profile in Column 2 (Case 6 – Three-column operation)

Figure 31: Pressure Difference Profile in Column 3 (Case 6 – Three-column operation)

Figure 32: Dissolved Gas Concentration Profile in Column 3 (Case 6 – Three-column operation)

Figure 33: Bubble Gas Concentration Profile in Column 3 (Case 6 – Three-column operation)

CONCLUSIONS

Using Aspen Technology's Aspen Custom Modeler software package, we developed a computer model to predict radiolytic gas generation and bubble formation in a non-elutable ion exchange column. Calculations examined six flowsheet test cases to provide the following results.

- The model predicted no bubble formation in Column 1 for all waste types at 21 gpm.
- At 10 gpm, bubbles formed in Column 1 for the average waste feed.
- In the three-column operation to process the SRS average waste at 21 gpm, bubbles formed in Column 2 near the end of the cycle. Bubbles also formed between Column 2 and Column 3 due to a large pressure drop between the two columns.

The model results indicate the necessity to install gas disengagement devices between Columns 2 and 3 to possibly eliminate gas bubble in Column 3.

RECOMMENDATIONS FOR FUTURE WORK

- Conduct experiments to obtain surface tension data for SRS waste solutions.
- Conduct tests to determine bubble radius.
- Conduct experiments to evaluate the impact of heterogeneous bubble formation inside particle pores on cesium loading.
- Investigate methods that more realistically approximate the spatial deposition of gamma ray energy in the columns.
- Experimentally confirm the calculated gas solubilities in SRS waste streams.

NOMENCLATURE

C	Concentration, mol/l
F	Volumetric flow rate, l/h
g	Acceleration of gravity, cm/s ²
G	G value, molecules/100eV
k	Rate constant
L	Column length, ft
P	Liquid pressure, atm
P*	Total pressure of bubble gases, atm
R	Rate, mol/h
r	Bubble radius
S	Solubility @ 1 atm, mol/l
t	Time, h
u	Superficial velocity, cm/min
V	Volume of the liquid phase, l
z	Position in axial direction, ft
γ	Surface tension
μ	Viscosity, cP
ρ	Density, g/ml
Δ	Difference

Subscript

i	Gas component (i = H ₂ , N ₂ , or O ₂)
b	Bubble formation
d	Bubble dissolution
in	Inlet
out	Outlet

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APPENDIX A – PHYSICAL PROPERTY DATA

Densities

Temperature (°C)	Density (g/mL)		
	<i>Average</i>	<i>High-NO₃</i>	<i>High-OH</i>
25	1.255	1.266	1.233
30	1.251	1.263	1.230
35	1.248	1.259	1.226

Viscosities

Temperature (°C)	Viscosity (cP)		
	<i>Average</i>	<i>High-NO₃</i>	<i>High-OH</i>
25	2.8	2.7	3.1
30	2.3	2.4	2.7
35	2.1	2.1	2.4

Gas Solubilities

The solubilities of hydrogen, nitrogen, and oxygen in simulated SRS waste were calculated using the Schumpe model (a modification to the Sechenov Equation) as described by J. D. Norton and L. R. Pederson (Norton and Pederson, 1995).

$$\log (c_{G,O}/c_G) = \Sigma(h_{ion} + h_{gas})c_i$$

where $c_{G,O}$ = solubility of a particular gas in pure water
 c_G = solubility of a particular gas in waste solution
 c_i = concentration of ion i (molar)
 h_{ion} = ion-specific parameter
 h_{gas} = gas-specific parameter

Table A1 lists the compositions of the three simulated waste solutions (Walker, 1999 [a]). Table A2 gives the values of the ion-specific (h_{ion}) and gas specific (h_{gas}) parameters obtained from Hermann, Dewes, and Schumpe (Hermann et al., 1995). The parameter values measured at 25°C were assumed to be constant over the temperature range 25 to 35 °C used in the calculations. Three minor components (oxalate, silicate and molybdate ions) normally added to simulated waste solutions were not included in the calculations because no parameter values were available. Sulfate ion was increased to compensate for the loss of the three minor anions. Minor cations (Cs^+ and K^+) were not included.

Table A1: Composition of Simulated Waste Solutions (‡)

Ion	Concentration (molar)		
	<i>Average</i>	<i>High-NO₃</i>	<i>High-OH</i>
Na ⁺	5.6	5.6	5.6
OH ⁻	1.91	1.17	3.046
AlO ₂ ⁻	0.31	0.31	0.27
NO ₃ ⁻	2.14	2.86	1.10
NO ₂ ⁻	0.52	0.37	0.74
CO ₃ ²⁻	0.16	0.16	0.17
SO ₄ ²⁻	0.156	0.22	0.03
PO ₄ ³⁻	0.010	0.010	0.008
Cl ⁻	0.025	0.040	0.010
F ⁻	0.033	0.050	0.010

(‡) Walker, 1999 [a]

Table A2: Values of Ion-specific (h_i) and Gas-specific (h_G) Parameters (*)

Ion	h_i	Gas	H_G
Na ⁺	0.1079	H ₂	-0.0176
OH ⁻	0.0918	N ₂	0.0002
AlO ₂ ⁻	0.1 (**)	O ₂	0.0000
NO ₃ ⁻	0.0136		
NO ₂ ⁻	0.0726		
CO ₃ ²⁻	0.1558		
SO ₄ ²⁻	0.1164		
PO ₄ ³⁻	0.2243		
Cl ⁻	0.0381		
F ⁻	0.1058		

(*) Hermann et al., 1995

(**) Estimated

Temperature-dependent equations for the gas solubilities (in mole fraction, X) in pure water follow (Norton and Pederson, 1995).

$$\ln X_{\text{hydrogen}} = -125.939 + 5528.45/T + 16.8893 \ln(T)$$

$$\ln X_{\text{nitrogen}} = -181.587 + 8632.13/T + 24.7981 \ln(T)$$

$$\ln X_{\text{oxygen}} = -3.717984 - 5596.17/T + 1049668/T^2$$

Note that the equation for $\ln X_{\text{nitrogen}}$ given in Norton and Pederson Reference is in error. The correct equation was obtained from the original source (Battino, 1982). For each equation for $\ln X$ (in mole fraction), a corresponding equation for $\ln K_H$ (mole/kg water-atm) is obtained by

subtracting the number of moles of water in 1 kg (ln55.5). The latter expressions yield values for $c_{G,O}$ in the calculations.

The calculated gas solubility values at 1 atm of gas pressure at 25, 30, and 35 °C in each of the simulated waste solutions follow.

Oxygen

Temperature (°C)	Solubility (mmoles/l)		
	<i>Average</i>	<i>High-NO₃</i>	<i>High-OH</i>
25	0.151	0.173	0.236
30	0.140	0.160	0.219
35	0.131	0.149	0.204

Nitrogen

Temperature (°C)	Solubility (mmoles/l)		
	<i>Average</i>	<i>High-NO₃</i>	<i>High-OH</i>
25	0.0772	0.0883	0.121
30	0.0724	0.0827	0.113
35	0.0684	0.0782	0.107

Hydrogen

Temperature (°C)	Solubility (mmoles/l)		
	<i>Average</i>	<i>High-NO₃</i>	<i>High-OH</i>
25	0.144	0.164	0.226
30	0.140	0.160	0.221
35	0.138	0.157	0.216

G-Values

The following G values assume a column packed with UOP IONSIV® IE-911.

Waste Type	G (molecules / 100 eV)	
	<i>Hydrogen</i>	<i>Oxygen</i>
Average, 5.6 M Na ⁺	0.040	0.052
High-OH, 5.6 M Na ⁺	0.058	0.0124
High-NO ₃ , 5.6 M Na ⁺	0.028	0.078

Note that these G values account for the distribution of deposited energy between the salt solution and the IE-911 particles.

Air Saturation of Feed

The solubility values given above assume 1 atm of pure gas over the waste solution. Since air contains 21% oxygen and 79% nitrogen by volume, gas compositions in an average waste feed saturated with air at 25 °C follow.

	Oxygen	Nitrogen
Solubility (mmoles/l)	0.151	0.0772
Concentration (under air) (mmoles/l)	$(0.151) \times 21\% = 0.0317$	$(0.0772) \times 79\% = 0.0610$
(mg/l)	1.0144	1.708

The following table lists the compositions of dissolved oxygen and nitrogen in the three waste feeds saturated with air at 25 °C.

Component	Concentration (molar)		
	<i>Average</i>	<i>High-NO₃</i>	<i>High-OH</i>
O ₂	3.1710E-5	3.6330E-5 3.1290E-5 (@ 35 °C)	4.9560E-5
N ₂	6.0988E-5	6.9757E-5 6.1778E-5 (@ 35 °C)	9.5590E-5

Feed Compositions

The following chemical compositions were used for the three waste feeds without air saturation at 25 °C. For feeds saturated with air, we added oxygen and nitrogen compositions listed above to the feeds.

Component	Concentration (molar)		
	<i>Average</i>	<i>High-NO₃</i>	<i>High-OH</i>
H ₂ O	4.940000E+01	4.837925E+01	4.829090E+01
Misc. Na ⁺ Salts	1.321370E-01	1.442400E-01	7.037000E-02
NaNO ₃	1.955000E+00	2.840000E+00	1.100000E+00
NaNO ₂	4.730000E-01	3.700000E-01	7.400000E-01
NaOH	2.218863E+00	1.165760E+00	3.019630E+00
NaAlO ₂	2.770000E-01	3.200000E-01	2.700000E-01
CsOH	1.370000E-04	1.400000E-04	3.700000E-04
KOH	1.400000E-02	4.100000E-03	3.000000E-02
Na ₂ CO ₃	1.410000E-01	1.600000E-01	1.700000E-01
Na ₂ SO ₄	1.310000E-01	2.200000E-01	3.000000E-02

APPENDIX B - SYSTEM PRESSURE CALCULATIONS

Figure B1 schematically represents the CST IX. Based on the pre-conceptual design, the columns measure 20 feet long with 16 feet of bed length. In Figure B1, subscripts indicate column number. Point A shows the highest point before the column, assumed as 6 feet above the top of the column. Point B, 4 feet below the top of the column, represents the upper surface of the CST. Point C represents a point a short distance below the surface of the CST. Point D represents the bottom of the CST, 20 feet from the top of the column, and before the Johnson screen. Point E immediately follows the Johnson screen at the low point between columns, 20 feet below the top of the column. Point F represents the recycle blend tank before the pump. Point G shows the high point before the product hold tank. Point H depicts the product hold tank.

Figure B1: Schematic Flow Diagram of the CST IX Process

The design orients the long axis of the IX column along the vertical. Liquid enters the top of the column. Two pressure effects on the column pressure exist: (1) static head due to the height of liquid mass (increasing from top to bottom), and (2) dynamic head due to flow through the packed bed (decreasing from top to bottom). If we divide the column into a number of volume elements, the system pressure in element n comes from the following equation.

$$P_n = P_{n-1} + (\Delta P_S - \Delta P_D) \Delta L$$

with

$$\Delta P_S = \rho g$$

Based on tall column data from ORNL testing (Welch et al., 1999), Pike (Pike, 2000) estimated ΔP_D as 0.01414 psi/cm for the average waste feed at 21 gpm. The following expression provides a generalized form of ΔP_D adjusted for viscosity and flow rate of the feed stream.

$$\Delta P_D = 0.01414 \left(\frac{u}{4.36} \right) \left(\frac{\mu}{2.8} \right)$$

In the following sections, we calculate system pressure profiles for the cases studied in this report.

Average Waste @ 21 gpm and 25 °C

Position	Pressure (psig)	Position	Pressure (psig)	Position	Pressure (psig)
A1	25.34	A2	16.89	A3	8.45
B1	30.77	B2	22.32	B3	13.88
C1	29.72	C2	21.27	C3	12.83
D1	31.51	D2	23.06	D3	14.62
E1	31.01	E2	22.56	E3	14.12

High-OH Waste @ 21 gpm and 25 °C

Position	Pressure (psig)	Position	Pressure (psig)	Position	Pressure (psig)
A1	27.55	A2	18.37	A3	9.18
B1	32.89	B2	23.70	B3	14.52
C1	31.84	C2	22.65	C3	13.47
D1	32.74	D2	23.55	D3	14.37
E1	32.24	E2	23.05	E3	13.87

High-NO₃ Waste @ 21 gpm and 25 °C

Position	Pressure (psig)	Position	Pressure (psig)	Position	Pressure (psig)
A1	24.60	A2	16.40	A3	8.20
B1	30.08	B2	21.88	B3	13.68
C1	29.03	C2	20.83	C3	12.63
D1	31.14	D2	22.94	D3	14.74
E1	30.64	E2	22.44	E3	14.24

High-NO₃ Waste @ 21 gpm and 35 °C

Position	Pressure (psig)	Position	Pressure (psig)	Position	Pressure (psig)
A1	20.17	A2	13.44	A3	6.72

B1	25.61	B2	18.89	B3	12.17
C1	24.56	C2	17.84	C3	11.12
D1	28.11	D2	21.38	D3	14.66
E1	27.61	E2	20.88	E3	14.16

Average Waste @ 10 gpm and 25 °C

Position	Pressure (psig)	Position	Pressure (psig)	Position	Pressure (psig)
A1	14.46	A2	9.64	A3	4.82
B1	19.89	B2	15.07	B3	10.25
C1	18.84	C2	14.02	C3	9.20
D1	24.26	D2	19.44	D3	14.62
E1	23.76	E2	18.94	E3	14.12

APPENDIX C – MODEL VERIFICATION

We established a number of simple test cases to verify the model. In these verification tests, we estimated the pressure drop per unit length ΔP_D in the equation to calculate the system pressure (See Appendix B) by the following correlation.

$$\Delta P_D = -1.51169E-3 + 0.01674f$$

ΔP_D has units of psi/cm with f as the flow rate per unit area ($\text{gal min}^{-1} \text{ft}^{-2}$). The correlation derives from ORNL test data for small columns (Walker, Jr., et al., 1998).

Verification Test #1

Base case:

Average salt composition with $[\text{Cs}] = 0.14$ mmolar

Temperature = 25°C

$G(\text{H}_2) = 0.04$

$G(\text{O}_2) = 0.052$

No dissolved gas in feed stream

Run column until effluent $[\text{Cs}]$ exceeds 99.9% of the feed value. Determine total H_2 and O_2 generated using the following two approaches.

- Cs loading and gas generation rate:

$$\text{\$H2_Moles1} * 1000 = 5.07548E-3 * \text{H2_G} * (\text{Solute_Mass} * 453.59 / \text{Solute_MW});$$

$$\text{\$O2_Moles1} * 1000 = 5.07548E-3 * \text{O2_G} * (\text{Solute_Mass} * 453.59 / \text{Solute_MW});$$

- Gas concentration at column inlet and outlet

$$\text{\$H2_Moles2} * 1000 = \text{X_H2_Out} * \text{mF_Out} - \text{X_H2_In} * \text{mF_In};$$

$$\text{\$O2_Moles2} * 1000 = \text{X_O2_Out} * \text{mF_Out} - \text{X_O2_In} * \text{mF_In};$$

Results: $\text{H2_Moles1} = 1.129648 \text{ kmol}$

$\text{H2_Moles2} = 1.129413 \text{ kmol}$

$\text{O2_Moles1} = 1.468542 \text{ kmol}$

$\text{O2_Moles2} = 1.468237 \text{ kmol}$

The results indicate that the model consistently predict the total amount of gases generated. Figures C1, C2, C3, C4 and C5 show the profiles for cesium concentration, column pressure, pressure difference, dissolved gas concentration and bubble gas concentration, respectively.

Figure C1: Cesium Concentration Profile (Verification Test #1)

Figure C2: Column Pressure Profile (Verification Test #1)

Figure C3: Pressure Difference Profile (Verification Test #1)

Figure C4: Dissolved Gas Concentration Profile (Verification Test #1)

Figure C5: Bubble Gas Concentration Profile (Verification Test #1)

Verification Test #2

Examine the effect of 1/10 decrease in bubble formation rate constant. With smaller bubble formation rate constant, fewer gas bubble will form and higher dissolved gas concentration would result. Figures C6 and C7 correctly reflect these trends.

Figure C6: Dissolved Gas Concentration Profile (Verification Test #2)

Figure C7: Bubble Gas Concentration Profile (Verification Test #2)

Verification Test #3

This test examined the effect of increase in G values for hydrogen and oxygen. We repeated Test #1 with double G values. The model predicted earlier bubble formation as shown in Figure C8 and the total amounts of hydrogen and nitrogen generated also doubled.

Results: $H_2_Moles1 = 2.262836 \text{ kmol}$
 $H_2_Moles2 = 2.262366 \text{ kmol}$

$O_2_Moles1 = 2.941687 \text{ kmol}$
 $O_2_Moles2 = 2.941076 \text{ kmol}$

Figure C8: Bubble Gas Concentration Profile (Verification Test #3)

Verification Test #4

We repeated Test #1 with G values equal to zero. The model correctly predicted no gases (both soluble and bubbles) present in the column.

Verification Test #5

We repeated Test #4 with feed stream saturated with air. As shown below, the model correctly predicted effluent stream with the same amount of gas as influent

Influent:

C N2,dissolved = 6.1774524E-5
C N2,bubble = 0
C O2,dissolved = 3.2118815E-5
C O2,bubble = 0

Effluent:

C N2,dissolved = 6.17745239E-5
C N2,bubble = 0
C O2,dissolved = 3.21188146E-5
C O2,bubble = 0

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