Impact of the Small Column Ion Exchange Process on the Defense Waste Processing Facility - 12112

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ABSTRACT

The Savannah River Site (SRS) is investigating the deployment of a parallel technology to the Salt Waste Processing Facility (SWPF, presently under construction) to accelerate high activity salt waste processing. The proposed technology combines large waste tank strikes of monosodium titanate (MST) to sorb strontium and actinides with two ion exchange columns packed with crystalline silicotitanate (CST) resin to sorb cesium. The new process was designated Small Column Ion Exchange (SCIX), since the ion exchange columns were sized to fit within a waste storage tank riser. Loaded resins are to be combined with high activity sludge waste and fed to the Defense Waste Processing Facility (DWPF) for incorporation into the current glass waste form. Decontaminated salt solution produced by SCIX will be fed to the SRS Saltstone Facility for on-site immobilization as a grout waste form. Determining the potential impact of SCIX resins on DWPF processing was the basis for this study. Accelerated salt waste treatment is projected to produce a significant savings in the overall life cycle cost of waste treatment at SRS.

INTRODUCTION

The projected completion of salt waste processing at the SRS is several years later than the projected completion of sludge waste processing. Radionuclides recovered from salt waste, however, were to be combined with sludge waste and vitrified in the DWPF. Accelerating salt waste processing would bring the two completion dates into the same time frame and would eliminate the possibility of having to combine salt waste radionuclides with glass former chemicals for vitrification. SCIX as currently envisioned has the potential to process salt waste at about half the rate of the SWPF and to significantly accelerate salt waste processing.

Implementation of SCIX introduces CST resin into SRS waste processing. A research program was conducted at the Savannah River National Laboratory (SRNL) to identify potential processing issues from the simultaneous operation of SWPF and SCIX with the on-going sludge waste treatment within DWPF. Both salt treatment processes produce loaded MST for disposal in DWPF, though the two feed routes into DWPF are different. SWPF uses a solvent extraction system to remove cesium. The cesium-rich stream fed to DWPF has few other solids, and the main DWPF impacts are from small concentrations of volatile organics and some residual acid that tends to promote catalytic hydrogen generation (both issues relate to potential off-gas flammability concerns within DWPF).

The production volumes of MST and CST from parallel salt processing operations have the potential to displace 20-40% of the sludge waste oxides in the DWPF flowsheet for a fixed number of waste glass canisters per year. This is an overall favorable shift, since sludge waste processing was projected to finish a number of years earlier than salt waste processing, but the DWPF would need to continue operating to vitrify the radionuclides separated during salt waste processing. Suitable glass waste forms would require more components than are currently

found in DWPF glass frit and salt processing solids, i.e. glass former chemicals would be needed.

This paper focuses on the projected impacts of the SCIX-SWPF combination on the DWPF Chemical Processing Cell (CPC, the chemical pretreatment upstream of the waste melter). This paper gives a high level summary of results presented in an SRNL technical report [1]. Studies of the impact of MST and CST on the glass properties and DWPF melter performance were also conducted by SRNL [2,3]. Overall, the impact of MST and CST was projected to be relatively benign to DWPF CPC operations. The impact of the two resins on slurry rheology, foaminess, and processing chemistry were all evaluated during a series of simulant-based DWPF flowsheet simulations at lab-scale.

METHOD

Experimental Description

Testing was accomplished in a 4-L lab-scale CPC pilot plant previously developed to support other DWPF programs. The equipment simulates the 30 m^3 Sludge Receipt and Adjustment Tank (SRAT) and Slurry Mix Evaporator (SME) that are the main CPC processing vessels as well as critical portions of the chemical addition system and off-gas system. The trimmed SRAT receipt volume was about 2.9 L of simulated waste slurry. Volumetrically scaled DWPF design basis operating conditions were used. The chemical processing strategy used was as follows:

- The caustic SRAT starting slurry was brought to boiling under reflux for a minimum of two hours (when CST or MST was present).
- A rheology sample was obtained after cooling to 93 °C.
- Nitric acid, equivalent to about 15% of the demand, was added at 7.6 L/min (~100 min).
- Formic acid, equivalent to about 85% of the demand, was added at 7.6 L/min (~4 h).
- Process samples were obtained to monitor SRAT chemistry.
- The SRAT slurry was taken to boiling.
- Condensate was removed from the SRAT to concentrate solids (~3 h).
- Once the target water mass was removed, the SRAT slurry was refluxed for 12 hours.
- Process samples were obtained periodically.
- The end of the 12 hour reflux period defined the end of the SRAT cycle.
- End of SRAT samples were obtained for chemical analysis and rheology.
- A SME cycle was performed on the remaining SRAT product slurry.
- Two frit-water-formic acid additions were made targeting a 40% waste oxide loading in the ultimate glass waste form.
- The SME slurry was concentrated following each frit slurry addition (~3 h boiling for each concentration).
- The final SME total solids target was 50 wt% (undissolved plus dissolved solids).
- SME product samples were obtained for chemical analysis and rheology.

Minimum SRAT acid requirements (nitric plus formic) were calculated from a previously developed equation in terms of the chemical composition of the feed slurry. Actual processing was generally at 100-150% of the minimum total acid to simulate typical DWPF practice. The actual total SRAT acid was partitioned between formic and nitric acids using the current REDOX equation (REDuction-OXidation). This equation predicts the distribution of oxidation states of iron in the DWPF glass product from the chemical composition of the SME product. The target

was 0.2 Fe(II)/Total Fe. About 85% of the total acid added during the SRAT cycle was formic acid by moles.

A slurry sample was taken following caustic boiling to check for adverse impacts on slurry rheology which might occur if MST or CST were actively forming alumino silicate species. Ten to fifteen samples were taken during the SRAT cycle to monitor major chemical reactions. Reaction progress is also monitored *in situ* by a pH probe. Major reactions include acid-base neutralization, conversion of soluble and insoluble carbonates to CO_2, formic acid reduction of HgO, homogeneous and catalytic destruction of nitrite ion, formic acid reduction of MnO_2 to Mn(II), dissolution of $CaCO_3$ and $Mg(OH)_2$ (or equivalent insoluble forms), catalytic conversion of formic acid/formate ion to H_2 plus CO_2, and catalytic conversion of nitrate ion to ammonium ion plus CO_2. Separation of elemental mercury from the slurry was tracked by sampling plus weighing the elemental mercury recovered in condensates (mercury both steam strips to the off-gas and settles on the SRAT/SME vessel bottom).

Samples of the SME slurry were obtained after frit addition and concentration. Samples of SME slurry were checked for composition. The flow curve rheology of the SME products was measured. Samples of SME slurry were also held for two months to evaluate the impact of MST and CST on the yield strength of settled solids. This measurement used a four paddle vane attached to the rheometer to deform the settled solid layer. Additional SME product samples were vitrified in sealed crucibles and measured for REDOX. Condensate samples were obtained from the SRAT and SME cycles.

Analytical Methods

The DWPF monitors the SRAT and SME off-gas composition for hydrogen every minute during processing in order to avoid forming a flammable atmosphere inside the equipment. The simulation off-gas was sampled and analyzed every 4.5 minutes by a two column, micro gas chromatograph (GC). Compositions were converted to molar flowrates using a known flow rate of helium used as an internal standard (He-H_2 separation requires a slower scan method than DWPF). Flow rates for hydrogen, oxygen, nitrogen, CO_2, and N_2O were obtained directly by taking a concentration ratio with helium times the helium flowrate. N_2O forms during nitrite destruction and also in parallel with ammonium ion formation. A flow rate for NO_2/N_2O_4 was estimated from the loss of oxygen that occurs in the reaction sequence: $2NO + O_2 \rightarrow 2NO_2 \leftrightarrow N_2O_4$. Typically 70-90% of the NO produced during nitrite destruction is converted to NO_2/N_2O_4. Small amounts of NO were detected directly on the GC simultaneously with the loss of O_2. The NO peak on the GC, however, was not calibrated. Historical area data for the GC permits estimates of NO concentration to be made to confirm its relatively minor status.

Process simulation samples (both slurries and supernatant liquids) were analyzed for major elements using inductively coupled plasma-atomic emission spectroscopy. Slurry samples were analyzed for soluble anions using 100-fold weighted dilutions with water followed by filtration as well as for ammonium ion using ion chromatography. Samples were analyzed for wt.% total and dissolved solids in tared crucibles dried at 110 °C to constant weight. Slurry and supernatant liquid densities were measured on a commercial instrument. Rheological data were measured using a Haake RheoStress rheometer (RS600).

PROCESS RESULTS

DWPF currently receives MST with a small amount of entrained sludge solids directly into the CPC Sludge Receipt and Adjustment Tank (SRAT) in parallel with ~23 m^3 (6,000 gallons) of fresh sludge waste slurry. The MST comes from a pair of coupled small-scale facilities called ARP-MCU, (Actinide Removal Process-Modular CSSX Unit), which resembles the SWPF in design concept (MST strike coupled with a cesium solvent extraction (CSSX) process). SWPF is designed to replace ARP-MCU in a few years, and to feed MST to DWPF in the same manner.

In contrast, the design basis for SCIX is to transfer MST to the SRS Tank Farm vessel used for DWPF sludge batch preparation, blending, and washing (~4,000 m^3 or ~10^6 gallons). Once sludge preparation is complete, the sludge-MST blend will be transferred to the Tank Farm vessel dedicated to feeding the DWPF (~4,000 m^3). The design basis for the loaded CST is to sluice it from the ion exchange column as needed (roughly every 7-14 days), grind it, and then transfer it to the above DWPF feed holding tank. There are expected to be about 15-30 ground CST resin transfers during the duration of a normal DWPF sludge batch. The Tank Farm vessels being integrated with SCIX have heels remaining in them after inter-vessel transfers. These heels will begin to load with MST and CST over the course of the first few sludge batch preparations following SCIX start-up.

Material balance projections were performed to estimate the impact of heel resins on the feed to DWPF in combination with the fresh additions of CST to the sludge batch holding tank and SWPF MST additions directly to the DWPF SRAT. (SCIX MST is fully integrated into the sludge batch before it becomes feed for DWPF.) The bounding case evolution of the fractions of sludge, CST, and MST on an oxide basis over a single sludge batch lasting 111 SRAT batches is shown in Figure 1.
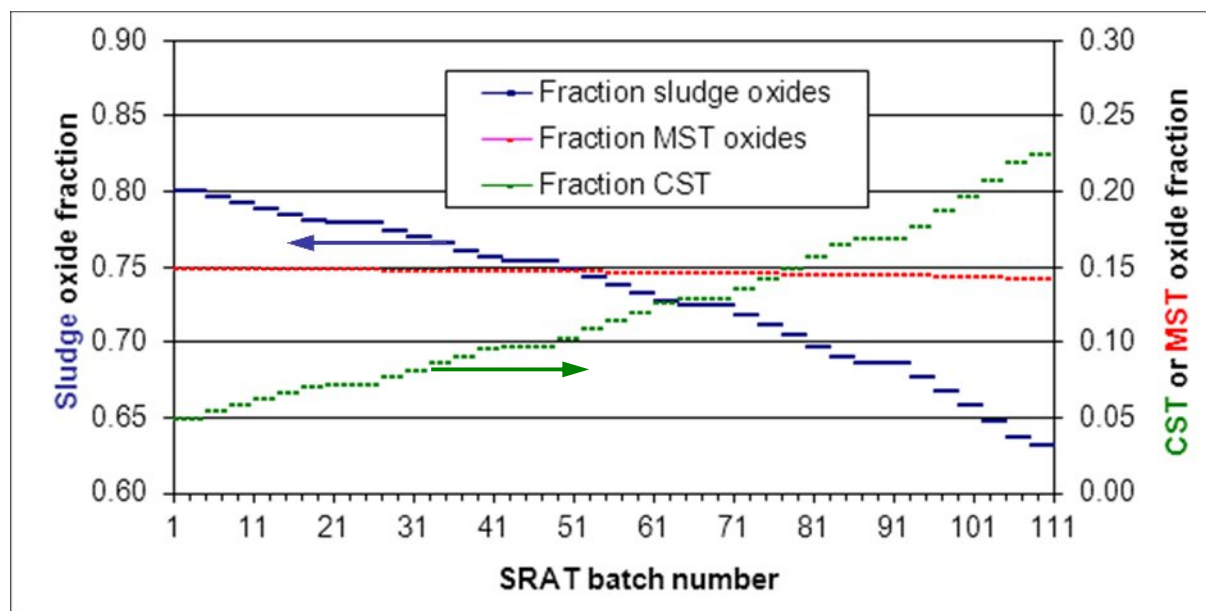

Figure 1. Shift in oxide fractions during a typical future sludge batch

Sludge waste oxides are about 80% initially plus 15% MST oxides, along with 5% CST oxides originating from the heel of the prior sludge batch. Regular periodic additions of CST to the sludge batch feed tank slowly build up the fraction of CST relative to sludge and MST seen in

the DWPF SRAT. The MST fraction remains relatively constant because of the regular SWPF transfers to the SRAT, which are ~80% of the total MST. Only the 20% contribution of MST in the DWPF feed tank is being diluted by CST. So what is roughly 11% MST from SWPF plus 4% in the sludge slurry feed (15%) drops to 11% from SWPF plus 3% in the sludge slurry feed (14%) over the 111 loadings of the SRAT vessel with feed (~23 m^3 per batch) in this example.

The projected overall impact of SCIX plus SWPF is to reduce the sludge waste oxides in the DWPF feed from the high 90's today into the 64-80% range several sludge batches after both salt processes are operational. Since the throughput of total oxides in DWPF is fairly constant, the time to process sludge oxides could be extended by 1.2-1.4 times. The exact extension will depend on the volumes of resin required during processing. Simultaneously, SCIX plus SWPF would reduce the total time for salt waste processing. The implication is that the remaining SRS sludge waste would last closer to the expected end of salt waste processing than current projections without SCIX. This would minimize the need to use glass former chemicals to vitrify the radioactive components recovered from salt waste in the absence of sludge, and could reduce the overall number of glass canisters produced during SRS tank clean-up and closure.

The above material balance results were used to define a bounding simulant (in terms of MST and CST content). A conservative sludge composition was selected from projected future DWPF sludge batches. A matching, non-radioactive, simulant slurry was prepared by co-precipitating nitrate salts, washing, and adding soluble sodium salts to produce a washed supernate composition. The simulant and baseline process simulations were also used to support a study on the impact of steam reforming the contents in Tank 48 at SRS [4].

One portion of the conservative simulant was trimmed to the bounding MST and CST content, while a second portion was trimmed to 50% of the bounding MST and CST content. The remainder was left untrimmed. The three simulants (0%, 50%, and 100% salt processing impacted) were used to perform seven lab-scale simulations of the DWPF CPC, Table I.

Table I. Description of process simulations.

Run ID-acid level	Description
SB0-lo	Baseline sludge plus 0% MST, CST at low acid
SB0-hi	Baseline sludge plus 0% MST, CST at high acid
SB50-lo	Sludge plus 50% MST, CST at low acid
SB100-lo	Sludge plus 100% MST, CST at low acid
SB100-hi	Sludge plus 100% MST, CST at high acid
SB100-hi-Ir	Sludge plus 100% MST, CST (Irradiated) at high acid
SB100-hi-EF	Sludge plus 100% MST, CST at high acid with Extended Flowsheet

The focus of the three low acid runs was the impact of MST and CST on the determination of the minimum acid requirement for processing, while the focus for the high acid runs was on the impact of excess acid on catalytic hydrogen generation from formic acid. Excess acid also impacts slurry rheology, so the tests provided a reasonable range of rheological behaviors.

CST and MST were conservatively irradiated in one high acid test to determine if irradiation could activate the titanium to a catalytically active form for hydrogen generation (SB100-hi-Ir). An extended flowsheet confirmation simulation was performed to check the impacts of slower MST additions and SWPF strip effluent additions to subsequent processing (SB100-hi-EF). In this run, a dilute MST slurry was fed to the SRAT sludge at boiling to simulate how the MST

from SWPF will likely be added to the SRAT. The SRAT sludge simulant slurry at the start of dilute MST addition contained the balance of the MST (from SCIX) and the CST.

Process Chemistry Results

The impact of MST and CST on the required acid for nitrite destruction is summarized in Table II.

Table II. Description of low acid process simulation outcomes.

Run ID	Description
SB0-lo	~30% nitrite destroyed after acid, 100% by end of SRAT
SB50-lo	~45% nitrite destroyed after acid, 100% by end of SRAT
SB100-lo	~30% nitrite destroyed after acid, 100% by end of SRAT

The calculation of percent nitrite destroyed after acid addition is subject to propagation of uncertainties in slurry mass and nitrite concentration that combine with the uncertainty in the feed composition of nitrite to make the values obtained only approximate. All three runs were ultimately successful (nitrite was not detected in the three SRAT products). No impact of MST, which has a base structure, on predicted acid consumption requirements was detected.

Low acid testing did not show any appreciable impact of MST and CST on the solubility of sludge species during processing. Typically some of the insoluble calcium, magnesium, manganese, nickel, and zinc will dissolve during SRAT cycle acid addition. Generally similar behavior was seen in the three tests. Marker species for MST and CST dissolution (titanium, niobium) were not detectable in the aqueous phase. Sodium data were ambiguous, but suggested that some sodium may have been leached out of the MST and CST in addition to the ~1M sodium in the aqueous phase of the blended starting materials. Sodium leaching from the resins is not expected to create any special processing concerns for DWPF.

The maximum hydrogen generation rates seen in the SRAT and SME cycles are summarized in Table III. None of the tests with CST and MST approached the catalytic hydrogen generation rates seen in the test with no CST and MST.

Table III. Description of high acid process simulation outcomes.

Run ID	Description
SB0-hi	Produced up to 0.40 kg/h hydrogen in SRAT; 0.34 in SME
SB100-hi	Produced up to 0.12 kg/h hydrogen in SRAT; 0.05 in SME
SB100-hi-Ir	Produced up to 0.07 kg/h hydrogen in SRAT; 0.11 in SME
SB100-hi-EF	Produced up to 0.13 kg/h hydrogen in SRAT; 0.03 in SME

The test with irradiated CST and MST seemed to lag behind the other two high acid runs in terms of catalytic activity. As a consequence, relatively less excess acid was destroyed in the SRAT cycle and more was available in the SME cycle for conversion to hydrogen toward the end of the simulation. The time evolution of the hydrogen generation rate in the SRAT cycle is shown in Figure 2. Results are presented on a DWPF-scale basis. The DWPF design basis limits are 0.295 kg H_2/h in the SRAT cycle and 0.10 kg H_2/h in the SME cycle.
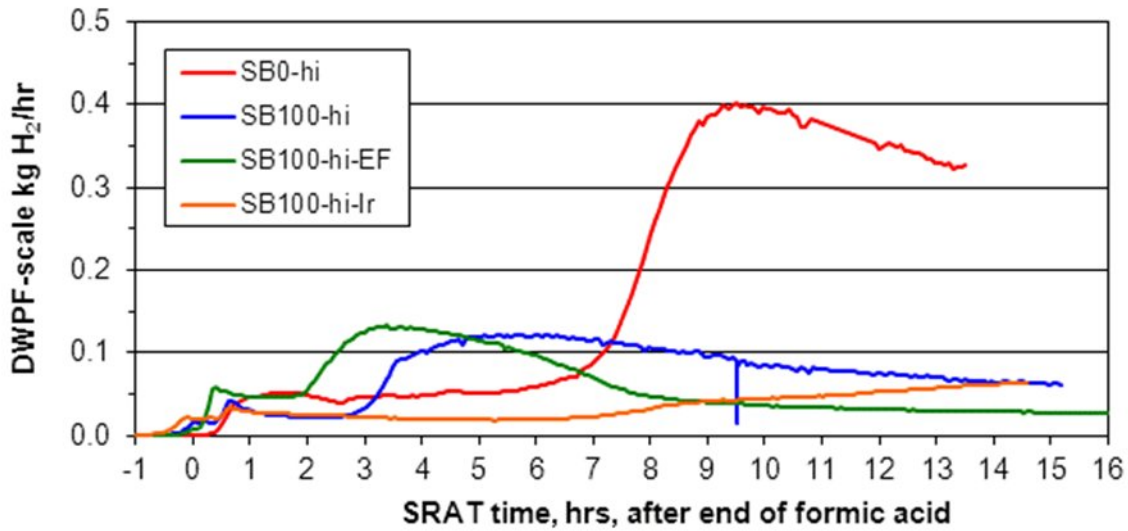
Figure 2. Comparison of hydrogen generation rates in the SRAT cycle

The baseline run exceeded the DWPF SRAT design basis limit indicating that the high acid tests were slightly aggressive. The continuation of hydrogen generation into the SME cycle (with frit-water-formic acid slurry additions) is shown in Figure 3.
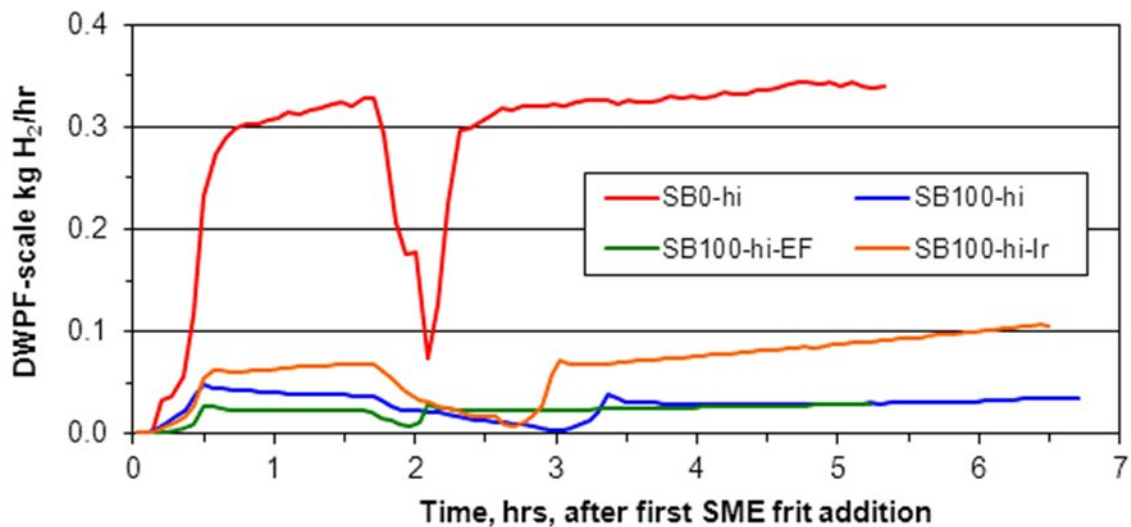

Figure 3. Comparison of hydrogen generation rates in the SME cycle

The baseline run continued to produce hydrogen at levels above the DWPF design basis for the SME cycle. The irradiated CST and MST test eventually approached the design basis limit. This is not seen as a negative outcome for SCIX, however, since the rates were less than a third of the CST and MST-free baseline.

Rheological data results

Flow curves for the slurries with and without MST and CST were obtained before, during, and after the SRAT/SME simulations. Vane yield strength measurements were made on settled SME product solids as well. The results in the tables that follow are averages of duplicate measurements. In the case of the vane measurements, the results are averages of single measurements on duplicate samples. No unusual variations were seen in the two individual results for a given sample (or sample pair).

Bingham plastic constants for the three starting simulants (0%, 50%, and 100% of bounding CST and MST oxides) are given in Table IV along with flow curve results for mildly diluted slurry samples taken from the SRAT vessel following two hours of boiling (before acid addition). A small de-ionized water dilution occurs when the SRAT is charged with feed slurry and minor trim chemicals.

Table IV. Impact of caustic boiling on simulants containing CST and MST

Slurry Description	Yield Stress Pa	Plastic Viscosity cP	Wt. % Insoluble Solids	Slurry Dilution
0% simulant	2.3	8.1	11.2	None
50% simulant	1.5	6.2	13.6	None
SB50-lo	0.8	4.8	12.5	8%
100% simulant	1.0	5.1	12.9	None
SB100-lo	0.4	3.8	12.1	6%
SB100-hi	0.4	3.8	12.1	6%
SB100-hi-EF	0.6	4.3	12.1	6%
SB100-hi-Ir	0.4	3.8	12.1	6%

No evidence was found to indicate that MST and/or CST were contributing to sodium alumino silicate formation which was expected to negatively impact the rheological properties of the unprocessed slurry. The drop in yield stress observed in the SRAT vessel samples after caustic boiling is larger than expected from dilution alone, and likely indicates some thermal aging of the sludge simulant particles.

Rheological properties for SRAT product slurries are compared in Table V. The slurries differ in total and insoluble solids contents. The SRAT slurry was targeted to an endpoint that produced the same number of equivalent glass waste canisters. Since the CST and MST are close to an oxide waste form, less mass of these two species is required than of sludge waste to produce the same mass of waste oxides in glass.

Table V. Comparison of SRAT products with/without CST and MST

Run ID	Yield Stress Pa	Plastic Viscosity cP	Wt % Insoluble Solids
SB0-lo	14.7	13.6	16.8
SB50-lo	5.7	5.9	12.6
SB100-lo	5.7	11.6	13.2
SB0-hi	1.6	8.5	15.5
SB100-hi	0.3	4.9	12.0
SB100-hi-EF	0.7	5.9	12.7
SB100-hi-Ir	0.3	4.0	10.5

An indirect benefit of maintaining waste oxide throughput was a reduction in the yield stress of SRAT product slurries containing CST and MST compared to the baseline runs. This shift could be exploited by reducing the number of SRAT cycles needed for a given canister rate (nominally 325 canisters per year at 40% waste loading) while running at correspondingly higher solids concentrations.

SME product slurries were aged for eight weeks at room temperature without mixing. After measurements were made on the settled solids, the slurry was regenerated with mixing and fresh flow curves were measured, Table VI. High acid slurries were generally less viscous than low acid slurries except for the baseline simulant tests without MST and CST.

Table VI. Comparison of aged SME products with/without CST and MST

Run ID	Yield Stress Pa	Plastic Viscosity cP	Wt % Insoluble Solids
SB0-lo	17	37	39.7
SB50-lo	22	37	39.5
SB100-lo	22	36	40.4
SB0-hi	27	34	38.8
SB100-hi	10	29	40.2
SB100-hi-EF	14	36	42.1
SB100-hi-Ir	11	29	39.2

The yield strength of the settled solids was measured by vane rheometry for the eight week old samples of settled SME product, and the results are given in Table VII.

Table VII. Comparison of settled SME products yield strength

Run ID	Vane Yield Strength, Pa
SB0-lo	77
SB50-lo	66
SB100-lo	61
SB0-hi	77
SB100-hi	56
SB100-hi-EF	70
SB100-hi-Ir	117

Generally, the settled solids containing MST and CST were similar to lower in yield strength compared to the corresponding baseline simulant SME product. One exception was the test with irradiated CST and MST. The yield strength result was not prohibitively different from the baseline slurry, but was more than double that of the corresponding run with unirradiated CST and MST (117 vs 56 Pa).

DISCUSSION

Process simulations were executed with few signs of unusual behavior. Steam stripping of mercury occurred in a manner consistent with prior process simulations. Foaminess of the slurry during gas generation and boiling was well controlled by the current antifoam agent. Samples taken shortly after formic acid addition indicated a reasonably high fraction of MnO_2 had been reduced to Mn(II). Mn(II) causes fewer off-gas issues in the melter. Catalytic formation of ammonium ion and ammonia vapor from nitrate ion was present at very low levels and was not of concern.

One observation was made on the high acid runs with MST and CST. There was an increase in iron solubility during processing. The percentage of iron dissolved was not large, but it was considerably more iron than is normally seen in the aqueous phase. The iron discolored the aqueous phase, Figure 4.
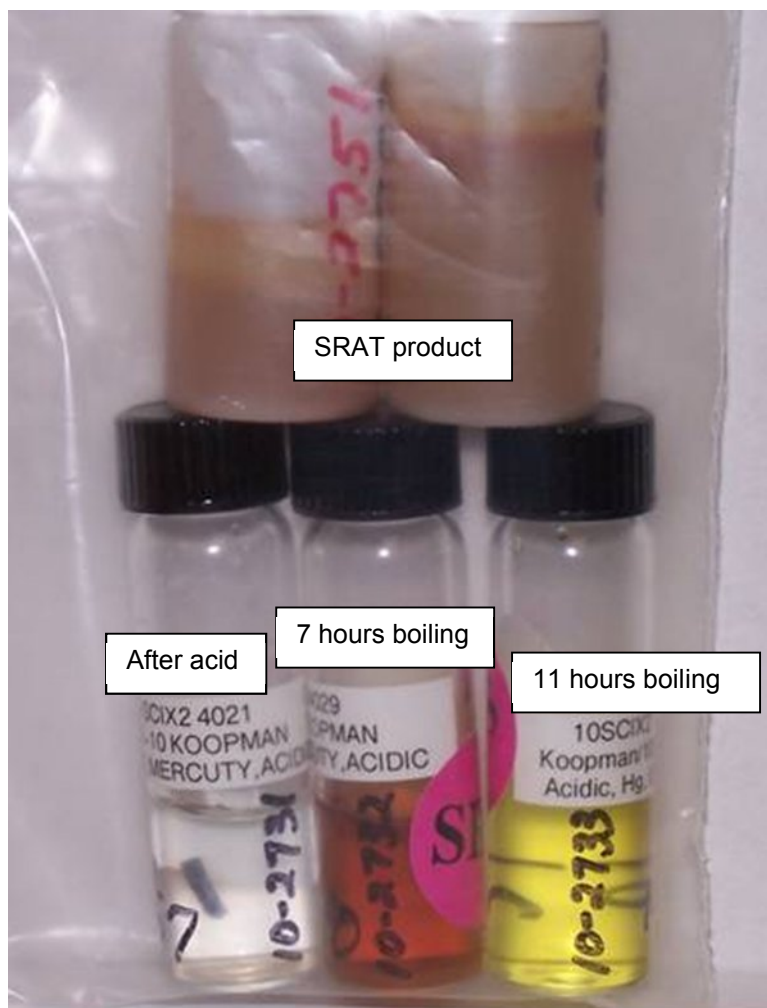
Figure 4. Impact of dissolved iron on the aqueous phase

The aqueous phase was nearly water white from a sample that was taken, centrifuged, and decanted immediately after the completion of formic acid addition. About seven hours later, the centrifuged and decanted aqueous phase was a rusty brown color, but after eleven hours of boiling the color had turned to yellow. When SRAT product slurry samples were being prepared for analytical work following the run (at room temperature for over a day), the aqueous phase separated from the slurry began to precipitate out fresh brown solids (top samples in Figure 4). If the bounding MST and CST tests saw the maximum iron dissolution that could be expected during actual processing, then it is probably not an issue going forward with the current DWPF coupled sludge processing flowsheet (nitric acid-formic acid flowsheet). Alternative flowsheets are under consideration, and limiting the dissolution of one of the neutron poisons in the criticality analysis for settled waste solids is already under study.

CONCLUSIONS

Lab-scale simulations of the DWPF CPC were performed to evaluate the impacts of bounding fractions of MST and ground CST resins on the physical and chemical behavior of the waste slurry as it processed through the DWPF to the melter feed tank. No negative impacts to slurry rheology, chemical reaction behavior including catalytic hydrogen generation, and foaming tendencies were found.

The main findings of the study were that these materials could be handled at the projected volumes (up to 40% of the waste oxide content in the final glass waste form) and that sludge waste processing would consequently be slowed down as salt waste processing was accelerated.

Primary findings were that the resins reduced slurry viscosity, did not contribute to foam stabilization, had no effect on the equation for setting the stoichiometric acid requirement, and reduced the potential for producing excessive catalytic hydrogen from the formic acid added as a reductant during processing. Collectively, the findings indicate that a potential increase in CPC throughput could be achieved in kg of waste oxides processed per year relative to current production rates that depends on the actual quantities of MST and CST in the CPC waste feed.

Compositional data for other off-gas species were generally more similar across the tests than the results for hydrogen. One exception was SME cycle CO_2 generation which was fairly similar to the results obtained for hydrogen in terms of relative differences. SME cycle CO_2 occurs when catalytic reactions are essentially the only source of both CO_2 and H_2, so this similarity was not unexpected.

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