# Filtration of Envelope C Waste Simulant Treated by the Sr/TRU Precipitation Process

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# Filtration of Envelope C Waste Simulant Treated by the Sr/TRU Precipitation Process

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Test Specification: TSP-W375-01-00003 Test Plan: CHG-TP-41500-019 Test Exceptions: None R&T Focus Area: Pretreatment Test Scoping Statement(s): B-36

Battelle—Pacific Northwest Division Richland, Washington 99352

#### **COMPLETENESS OF TESTING**

This report describes the results of work and testing specified by Test Specification TSP-W375-01-00003 and Test Plan CHG-TP-41500-019. The work and any associated testing followed the quality assurance requirements outlined in the Test Specification/Plan. The descriptions provided in this test report are an accurate account of both the conduct of the work and the data collected. Test plan results are reported. Also reported are any unusual or anomalous occurrences that are different from expected results. The test results and this report have been reviewed and verified.

**Approved:** 

Gordon H. Beeman, Manager WTP R&T Support Project Date

Research and Technology Manager

Date

# Summary

The River Protection Project-Waste Treatment Plant baseline for pretreating Envelope C low-activity waste (LAW) at Hanford includes a filtration step for removing entrained solids and the Sr/TRU precipitate before the waste is vitrified. The purpose of the work discussed in this report was to conduct crossflow filtration tests with a treated simulant representing the "active" (actual) waste blend tested by Hallen et al. (2002). This waste blend is a mixture of Tank AN-102 samples and C-104 sludge pretreatment solutions (supernatant, wash, caustic leach, and rinse solutions), and is referred to as the "AN-102/C-104 waste blend." These simulant tests were performed in accordance with Test Specification TSP-W375-01-00003 (Reynolds 2001) and Test Plan CHG-TP-41500-019 (Hallen 2001), as part of Scoping Statement B-36 (BNI 2002).

In the active waste testing with the treated AN-102/C-104 waste blend (Hallen et al. 2002), there was insufficient slurry volume to conduct the complete test matrix of filtration conditions and any dewatering or washing tests in the cells unit filter (CUF). A simulant was developed that represented the chemical constituents of the waste blend. The simulant was treated by adding the necessary reagents for Sr/TRU precipitation under the same conditions as the actual waste blend prior to the filtration test. The target concentrations of both strontium and permanganate were 0.02M (based on the final volume after addition of both reagents). No additional hydroxide was specified for this test, since the free hydroxide (0.33M) was shown to be adequate for Sr/TRU removal in previous testing. [Note: The results do not reflect the WTP baseline and that the flux results are not applicable for design verification or flowsheet validation.]

The objective of the "inactive" testing was to verify that the treated simulant adequately mimicked filtration behavior of the treated actual waste blend by directly comparing filtration performance data for the two treated materials. Once the simulant was shown to be representative of the actual treated waste blend, additional simulant tests, not possible with the active waste due to insufficient slurry volume, were conducted in order to demonstrate that the simulant could be successfully filtered. The average targeted design flux was  $0.02 \text{ gpm/ft}^2$ 

During the simulant testing, the filterability of a 0.8 wt% undissolved solids slurry (low solids concentration) was measured with a 0.1-µm, sintered metal, high purity Mott filter using a single-element, crossflow filtration system (i.e., the CUF). The slurry was filtered using a matrix of thirteen 1-hour conditions of varying transmembrane pressure (TMP) (20 to 60 psid) and axial velocity (7 to 15 ft/s), with the permeate being recirculated. The system was backpulsed between each condition. The testing mimicked the filtration testing of the active waste.

The average permeate  $flux^{(a)}$  was 0.037 and 0.034 gpm/ft<sup>2</sup> for the active and simulant tests, respectively. However, a statistical comparison of the two empirical models indicated that, although the average flux is similar, the two models are statistically different. Because of the similar flux results obtained when running the matrix, and the known chemical similarities resulting from the precipitation process, the

<sup>(</sup>a) Average matrix conditions were 40 psid TMP and 11 ft/s axial velocity.

simulant used is believed to mimic the filtration properties of the active waste, even if there are some differences in response to changes in the independent variables. Consequently, the more extensive simulant testing described below (dewatering and solids washing) is considered representative of what would be expected from the active waste if an adequate waste volume had been available.

Additional testing of the simulant was conducted with an industrial grade Mott filter,<sup>(a)</sup> with the same dimensions as the high purity filter. The test matrix described above was repeated with a 0.8 wt% undissolved solids slurry (low solids concentration). These tests produced a permeate flux that ranged from 0.018 to 0.044 gpm/ft<sup>2</sup>. This flux was almost exclusively dependent on TMP. Following the matrix testing, the slurry was tested for 3 hours at a single condition without backpulsing.

The slurry was concentrated (dewatered) to 2.7 wt% undissolved solids (high solids concentration). Permeate flux of the slurry during the dewatering decreased linearly with the log of the undissolved solids concentration. The test matrix was repeated with the high solids concentration (2.7 wt%) slurry and produced a permeate flux that ranged from 0.015 to 0.031 gpm/ft<sup>2</sup>. This flux was primarily dependent on TMP, but also was dependent on time and axial velocity. After the matrix testing was completed, the slurry was tested for 3 hours at a single condition without backpulsing.

Once testing of these two matrices was completed, the material was washed four times in the CUF with equal volume batches (1.25 liters) of 0.01M NaOH and dewatered between washes back to the original slurry volume. For each wash, approximately 50% dilution of the resulting permeate occurred. The permeate flux during washing was shown to vary inversely proportional to permeate viscosity.

Battelle—Pacific Northwest Division (PNWD) implemented the RPP-WTP quality requirements in a quality assurance project plan (QAPjP) as approved by the RPP-WTP quality assurance (QA) organization. Active testing was performed in May 2001 in accordance with PNWD's quality assurance project plan, CHG-QAPjP, Rev. 0, which invoked PNWD's Standards Based Management System (SBMS), compliant with DOE Order 414.1A Quality Assurance and 10 CFR 830, Energy/Nuclear Safety Management, Subpart A -- Quality Assurance Requirements. Due to a change in the contract QA requirements in September 2001, the inactive testing, conducted in September 2001 and January 2001, was conducted in accordance with PNWD's quality assurance project plan, RPP-WTP-QAPjP, Rev. 0, which invoked NQA-1-1989 Part I, Basic and Supplementary Requirements, and NQA-2a-1990, Subpart 2.7. These quality requirements were implemented through PNWD's Waste Treatment Plant Support Project Quality Assurance Requirements and Description (WTPSP) Manual. The quality of the data gathered during the earlier experiments was not impacted by the change in requirements.

PNWD addressed verification activities by conducting an Independent Technical Review of the final data report in accordance with procedure QA-RPP-WTP-604. This review verified that the reported results were traceable, that inferences and conclusions were soundly based, and the reported work satisfied the Test Plan objectives. The review procedure is part of PNWD's WTPSP Manual. There were no exceptions to the test plan.

<sup>(</sup>a) The industrial grade filter is preferred by the Contractor. A comparison of the 0.1-μm industrial grade and high purity (also known as the 9-log reduction media) filters indicated no significant difference in the permeate flux with the simulant tested (Hallen et al. 2002).

Issues:

- Time between reagent addition for Sr/TRU precipitation and filtration may impact the permeate flux time dependency. Although more data are necessary to confirm the cause, we observed that the simulant filtered 11 hours after reagent addition demonstrated less time dependency than the simulant filtered 6 hours after reagent addition. Data from Hallen et al. (2002) and Lilga et al. (2002) support this observation, as the soluble strontium concentration decreases by ~50% between 4 and 24 hours after reagent addition. These data suggest a benefit of waiting, as much as 24 hours, after reagent addition before filtration.
- 2. Based on extrapolation of dewatering data, the permeate flux will be below the BNI targeted flux<sup>(a)</sup> of 0.02 gpm/ft<sup>2</sup> at undissolved solids concentration greater than 4.6 wt% (at 40 psid TMP and 11 ft/s axial velocity).

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<sup>(</sup>a) The targeted flux in the test specification is  $0.065 \text{ gpm/ft}^2$ . The target was subsequently changed by BNI to  $0.02 \text{ gpm/ft}^2$  and documented in Stiver (2002).

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# 1.0 Introduction

Flowsheets developed for the River Protection Project-Waste Treatment Plant (RPP-WTP) specify crossflow filtration for initial dewatering of the waste and to separate wash solutions from the solids. In crossflow filtration, the majority of the filter cake is swept away by the fluid flowing across it. This filtration method is especially beneficial when there are very fine particles and when system simplicity is required. Traditional dead-end filtration has a declining filtration rate caused by the growth of a filter cake on the surface of the filter medium.

The purpose of the work discussed in this report was to conduct crossflow filtration tests with a treated simulant representing the "active" (actual) Envelope C<sup>(a)</sup> waste blend tested by Hallen et al. (2002b,c). This waste blend is a mixture of Tank AN-102 samples and C-104 sludge pretreatment solutions (supernatant, wash, caustic leach, and rinse solutions), and is referred to as the "AN-102/C-104 waste blend." The "inactive" supernatant simulant used here was derived from targeting analytical concentrations measured from Tank AN-102 samples (Hay et al. 2000, Urie et al. 2002b) and C-104 sludge pretreatment solutions (Brooks et al. 2000). No attempt was made to simulate the entrained solids present in the actual AN-102 samples used for preparing the active waste blend. Before filtration testing, the simulant was treated with similar Sr/TRU precipitation conditions as the actual waste blend. Similar Sr/TRU precipitates were expected for both the simulant and actual waste blend, since the reagent and conditions were similar.

In the active waste testing reported in Hallen et al. (2002c), the volume of slurry (1.35 wt% undissolved solids) was insufficient to conduct dewatering or washing tests in the cells unit filter (CUF). The scope of the testing described in this report was to match the conditions tested with the active waste to verify that the treated simulant adequately mimicked the active slurry filtration by directly comparing filtration data. Because 6.5 liters of simulant were available, additional testing beyond the original scope of this study was performed in order to obtain as much filtration data as possible. These additional tests, presented in Sections 3.2.2 and 3.2.3, included dewatering the simulant to 2.7 wt% undissolved solids and conducting tests with the concentrated slurry.

The test matrix used for this study consisted of thirteen 1-hour conditions of varying transmembrane pressure (TMP) (20 to 60 psid) and axial velocity (7 to 15 ft/s), with the permeate being recirculated. The system was backpulsed between each condition. Because of the limited volume in the active filtration tests, not all matrix conditions could be obtained. The active and simulant filtration comparison was conducted with data from the test matrix at low solids concentration ( $\sim$ 1 wt%) using a 0.1-µm, sintered metal, high purity Mott filter (2-ft-long, single element).

The additional simulant testing was conducted with a Mott 0.1-µm, sintered metal, industrial grade filter. The 13-hour matrix was repeated with 0.8 wt% undissolved solids slurry. Following the matrix testing, the slurry was tested for 3 hours at a single condition without backpulsing. The slurry was concentrated to 2.7 wt% solids (high solids concentration), and the test matrix and extended run at a single condition

<sup>(</sup>a) Envelope designations are explained in DOE-ORP (2000).

without backpulsing was repeated. Once testing of these two matrices was completed, the material was washed four times in the CUF, with permeate removed by filtration.

The simulant tests were conducted using a "cold" CUF located in the Simulant Development Laboratory in the Hanford 300 Area. This filtration system is identical to the "hot" CUF, located in the High Level Radiochemistry Facility hot cells (in the Radiochemistry Processing Laboratory, 300 Area), that was used for the active tests.

The testing was performed in accordance with Test Specification TSP-W375-01-00003 and Test Plan CHG-TP-41500-019, as part of Scoping Statement B-36. There were no test exceptions.<sup>(a)</sup> Section 2.0 of this report describes the test conditions and experimental procedures; Section 3.0 discusses the results; and Section 4.0 provides conclusions based on the results. The appendices contain the simulant recipe and particle size distribution data.

<sup>(</sup>a) Based on a targeted flux of 0.02 gpm/ft<sup>2</sup>. The targeted flux in the test specification is 0.065 gpm/ft<sup>2</sup>; however, the target was subsequently changed by BNI to 0.02 gpm/ft<sup>2</sup> and documented in Stiver (2002).

# 2.0 Test Conditions and Approach

This section describes the equipment, test conditions, simulant, and empirical modeling used for the testing. Additional details on the active waste testing are provided in Hallen et al. (2002c).

### 2.1 Testing Apparatus

The CUF process flow diagram is shown in Figure 2.1. The slurry feed is introduced into the CUF through the 4-liter slurry reservoir. An Oberdorfer progressive cavity pump (Model 101B), powered by a Gast air motor (Model 4AM-NRV-92), pumps the slurry from the slurry reservoir through the magnetic flow meter (Bailey Fischer & Porter, series 10D1476 K-MAG) and the Mott filter element (sintered metal, 2-ft active length, 1/2-inch OD and 3/8-inch ID).

Axial velocity and TMP are controlled by the pump speed (which is controlled by the pressure of the air supplied to the air motor) and the discharge throttle valve (V4) position. Permeate that passes through the filter can be sent to the backpulse chamber, reconstituted with the slurry in the slurry reservoir, or removed. The permeate flow rate is measured by means of a graduated glass-flow monitor that is fill-and-drain operated. Higher permeate flow rates can be monitored with an in-line rotometer. Slurry samples are taken directly from the slurry reservoir with a 10-mL pipette. Permeate samples are taken at the three-way valve upstream from the slurry reservoir. This is also the point at which permeate is removed for the dewatering step. Filter backpulsing is conducted by partially filling the backpulse chamber with permeate, pressurizing the backpulse chamber with air, and forcing the permeate in the chamber back through the filter. An impeller in the reservoir tank agitates the slurry, while a contact probe determines the slurry height. Welded connections and VCO fittings are used on the slurry side instead of Swagelok fittings to reduce areas where solids could hold up in the CUF.

During the tests, the slurry temperature was maintained at  $25 \pm 5$ °C by pumping cooling water through the heat exchanger just downstream of the magnetic flow meter. The slurry temperature was measured by a thermocouple installed in the slurry reservoir and controlled by a proportional-integral-derivative (PID) temperature controller that was part of the chiller.

### 2.2 Filter Elements Used in Testing

The filter elements<sup>(a)</sup> used in this work are 2 ft long, in a tube-in-shell design, with the 3/8-inch ID sintered metal tube mounted inside a 3/4-inch stainless steel pipe with threaded and compression fittings. The high purity (HP) filter was used because it is the same filter type tested in Hallen et al. (2002c). The industrial grade (IG) filter (used for the additional simulant testing described in Section 3.3) was included

<sup>(</sup>a) Both the "industrial grade" and "high purity" (also known as 9-log reduction media) filters are liquid service filters with a 0.1-µm rating.



Figure 2.1. Flow Diagram of the Crossflow Filtration Process

2.2

because it is preferred by the Contractor. Mott reports slight variations between the two filters, although both have a similar porosity ( $\sim$ 30% open pores). The industrial grade filter is reported to have a larger average pore size (with fewer total pores) and more mechanical strength when compared to the high purity filter. A comparison of the two filters using clean water, a 0.35M SrCO<sub>3</sub> slurry, and the AN-102/C-104 simulant is provided in Hallen et al. (2002c). The two filters were shown to give similar results.

### 2.3 Overview of Tests

Table 2.1 provides an overview of the testing. The active tests were conducted with HP #1, while the simulant (inactive) tests were conducted with HP #2 and IG #2. As part of the simulant testing,  $SrCO_3$  slurry tests were conducted in addition to clean water flux (CWF) to help determine filter resistance. The test conditions for the CWF and the  $SrCO_3$  slurry tests are listed in Table 2.2.

Active Tests—HP #1	Inactive Tests—HP #2	Inactive Tests—IG #2 <sup>(a)</sup>
Testing Date: May 2001	Testing Date: Sep. 2001	Testing Date: Jan. 2002
CWF, 1 hour	CWF, 1 hour	CWF, 1 hour
5 wt% kaolin clay slurry, 6 hours	0.35M SrCO <sub>3</sub> slurry, 1 hour	0.35M SrCO <sub>3</sub> slurry, 1 hour
CWF, 1 hour	CWF, 1 hour	CWF, 1 hour
AN-102/C-104 active test matrix,	AN-102/C-104 inactive test matrix,	AN-102/C-104 inactive test matrix
12 hours (precipitation reagents	13 hours (precipitation reagents	(0.8 wt%), 13 hours (precipitation
added 6 hours before filtration	added 6 hours before filtration	reagents added 11 hours before
testing. <sup>(b)</sup> )	testing.)	filtration testing. <sup>(c)</sup> )
CWF, 1 hour	Low Volume Run, 2 hours	Extended run, 2 hours
	CWF, 1 hour	Dewater, 4 hours
	0.35M SrCO <sub>3</sub> Slurry, 1 hour	AN-102/C-104 inactive test matrix
		(2.7 wt%), 11 hours
	CWF, 1 hour	Extended run, 2 hours
		Four wash/dewater, 1 hour
		CWF, 1 hour
		0.35M SrCO <sub>3</sub> slurry, 1 hour
		CWF, 1 hour

Table 2.1. Ove	erview of 7	Fests Cond	ucted with	Each Filter
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(a) IG #1 was used for active testing with AZ-101. This nomenclature is consistent with that presented in Hallen et al. (2002c).

(b) Reagents necessary for the Sr/TRU precipitation are provided in Appendix A.

(c) The time between Sr/TRU reagent addition and start of filtration testing for the 0.8 wt% slurry was 11 hours. During the dewatering step, the simulant used in the September 2001 testing was added to the CUF to reach 2.7 wt% solids.

Test	TMP (psid)	Velocity (ft/)	Comments
Clean Water Flux with 0.01M NaOH	10, 20, 30	11	Hold each condition for 20 minutes; backpulse between conditions
0.35M SrCO <sub>3</sub> Slurry	10, 20, 30	11	Hold each condition for 20 minutes; backpulse between conditions

 Table 2.2.
 Summary of Conditions During System Verification Testing

The matrix of filtration conditions for the active and inactive testing with the AN-102/C-104 Sr/TRU slurry is shown graphically in Figure 2.2. The matrix includes 13 combinations of TMP and crossflow (axial) velocity each run for an hour and in the order listed. The first condition (center point) was run for 3 hours with a backpulse after each hour before proceeding to the next condition. The center point was then repeated in the middle and at the end of testing to assess the effect of filter fouling with time over the course of testing. The system was backpulsed once between each condition. Average flux data reported from the matrix testing were calculated by averaging all data taken between 10 and 60 minutes. The flux results from this testing are presented in Section 3.0.

Test conditions during extended runs and dewatering were 40 psig TMP and 11 ft/s axial velocity. There was no backpulsing during the either the extended runs or the dewatering. Results from the low-volume inactive testing with HP #2 are discussed in Hallen et al. (2002c).



Figure 2.2. Crossflow Filtration Test Matrix for Sr/TRU Slurry Filtration (Conditions 1 through 13)

#### 2.4 Description of the AN-102/C-104 Waste Blend Simulant

The AN-102/C-104 waste blend simulant was based on the composition of a specific waste blend prepared by mixing Tank AN-102 waste and Tank C-104 sludge pretreatment solutions (supernatant, wash, caustic leach, and rinse solutions). The volume of each solution used to make the waste blend was specified in Test Specification TSP-W375-00-00007 (Johnson 2000) and was, by volume, approximately 4 parts AN-102 (containing 2 wt% entrained solids), 6 parts C-104 supernatant, 2 parts C-104 wash, 2 parts C-104 caustic leach, and 1 part C-104 rinse. The resulting waste blend was approximately 3M sodium (Urie et al. 2002a) and was evaporated to approximately 5.5M sodium (Lumetta et al. 2002) before Sr/TRU precipitation. The composition of the simulant representing the AN-102/C-104 waste blend was approximated by mathematically combining the specified volumes of each component using the composition data from the characterization of the AN-102 waste samples (Hay et al. 2000, Urie et al. 2002b) and the various C-104 sludge pretreatments solutions (Brooks et al. 2000). The free hydroxide concentration in the as-received AN-102 waste was in question based on three different values reported by Hay et al. (2000) and the values reported in the Hanford TWINS database and by Urie et al. (2002b). Urie et al. reported a concentration of 0.25M free hydroxide in the as-received waste, which is consistent with earlier AN-102 Sr/TRU removal tests (Hallen et al. 2002a) and was used for the waste blend calculations. The sodium concentration was adjusted mathematically to 5.5M, and the resulting composition were used to formulate the simulant for the inactive tests (see Appendix A for recipe).

An initial 2-liter batch of simulant was prepared August 23, 2001, to determine if the recipe provided a suitable simulant for precipitation and filtration testing. The original recipe was analyzed by inductively coupled plasma-atomic emission spectroscopy (ICP-AES). The simulant was found to be a good representation of the waste blend, except for chromium, which was well below the target concentration of 130  $\mu$ g/mL. This was a result of using Cr (III) acetate in the recipe instead of Cr (VI). In the final recipe, sodium chromate was used to obtain target levels of soluble chromium. The free hydroxide was also found by titration to be approximately 0.1M below the target of 0.3M. The simulant was corrected by adding an additional 0.1M NaOH. The recipe was then repeated on September 23, 2001, to prepare a 5-liter batch of simulant. The resulting simulant had a density of 1.28 g/mL and a viscosity<sup>(a)</sup> of 5.4 cP at 22°C.

The simulant batches were sampled after at least 1 week of aging, and the composition remained unchanged. However, 4 months after the first batch of simulant was prepared, it showed clear signs of dark brown/black solids on the sides of the storage container. Chemical analysis revealed approximately 90% of the manganese and 40% of the iron had precipitated from solution. The second batch was sampled at the same time and showed very little, if any, manganese and iron removal. The second batch was stored in a large translucent container under the counter, whereas the first batch was stored on the counter top in a small transparent bottle. The general observation has been that the solids form on the sides of the bottles, and the precipitation is likely a result of light-initiated reactions of the organic-metal complexes (Lilga et al. 2002). It is recommended that the Envelope C simulants be stored in air-tight containers protected from the light to prevent oxidation/precipitation of the soluble metal complexes.

<sup>(</sup>a) The kinematic viscosity was determined by flow of liquid through a tube, thus no shear stress and shear rate information is available.

Prior to filtration tests, the simulant was treated by adding the reagents necessary for the precipitation of Sr/TRU from Envelope C wastes. The target concentrations of both strontium and permanganate were 0.02M (based on the final volume after addition of both reagents). No additional hydroxide was specified for this test, since the free hydroxide (0.33M) was shown to be adequate for Sr/TRU removal in previous testing (Hallen et al. 2002c). Reagents added for precipitation are provided in Appendix A. The same reagent addition and conditions were used to treat the simulant and the active waste. The resulting precipitated solids in the simulant comprised approximately 0.8 wt% undissolved solids, which is less than the 1.4 wt% observed in the active waste (Hallen et al. 2002c). However, there was no attempt to simulate the entrained solids from the active waste, which was likely the difference between the two values. The rationale for not including the entrained solids is that 1) the exact composition and physical properties of the entrained solids. The basis for preparing the simulant solids was to use identical Sr/TRU precipitation reagent addition, as the Sr/TRU precipitate process is the major process step that determines the filterability of Envelope C wastes.

The simulant prepared on August 23, 2001 was precipitated on September 3, 2001, 6 hours prior to filtration testing of the high purity filter. Samples were removed during the precipitation reaction to allow comparison to the actual waste tests. The change in simulant composition as reagents were added was similar to the actual waste tests. Strontium was initially at 2  $\mu$ g/mL and increased to 20  $\mu$ g/mL on Sr(NO<sub>3</sub>)<sub>2</sub> addition, similar to actual waste tests. On permanganate addition, iron removal was around 90%, but manganese actually increased. In contrast, in actual waste tests the manganese decreased on permanganate addition. During filtration tests, the permeate was observed to have an orange color, characteristic of the Mn(IV) gluconate complex observed in reaction mechanism experiments (Lilga et al. 2002). Permeate samples were collected for chemical analyses. On setting, when exposed to light, these samples were observed losing all of the orange color. Additional samples were removed and stored in the dark. The samples in the dark retained the color for hours/days, whereas the sample exposed to light became colorless in minutes. Both the light-exposed and unexposed samples were submitted for ICP-AES analysis, and contained high levels of manganese, 20 µg/mL. For comparison, the manganese concentration from the verification tests using actual waste blend samples was 2 µg/mL (Hallen et al. 2002b).

Sr/TRU precipitation of 1.5 liters of the large batch of simulant (prepared on September 3, 2001) was performed on September 5, 2001. The permeate from this batch of treated simulant was again orange in color, light sensitive, and contained higher-than-expected levels of manganese,  $20 \mu g/mL$ . The higher-than-expected manganese level was consistent for both of the treated batches of simulant, and most likely a result of the gluconate levels used in simulant recipe.

Approximately 4 months later (January 7, 2002), 3 liters of the second large batch of AN-102/ C-104 simulant (initially prepared on September 3, 2001) was precipitated in preparation for additional filtration testing with the industrial grade filter. The Sr/TRU precipitation was conducted approximately 11 hours prior to the initial filtration testing. Samples taken during this testing showed about 50% manganese removal on permanganate addition, more consistent with the actual waste tests. The conclusion is that the initial simulant recipe contained high levels of organic complexants, which resulted in high soluble manganese. On aging of the simulant, some organic likely degraded to a level that better represents the actual waste blend.

The simulant prepared during the September 2001 testing (total of 3 liters) was combined and saved for additional feed during the dewatering tests. Consequently, the dewatering, the high solids test matrix, and the solids washing conducted in the January 2002 filtration testing was performed with a blend of treated simulants prepared in September 2001 and January 2002.

Lilga et al. (2002) used the remaining AN-102/C-104 waste blend simulant, 0.5 liter of the first batch and 1 liter of the second batch, for experiments to assess the reaction mechanisms of the Sr/TRU removal process.

## 2.5 Empirical Modeling

Empirical models, described in Section 3.0, evaluating the influence of time, TMP, and axial velocity on permeate flux were analyzed using a JMP statistical software package (version 4.0.4, SAS Institute, Inc.). The models use linear regressions of the three variables to predict permeate flux. Linear regression was used because non-linear regression generally resulted in little to no improvement of the models' predictive capabilities. The leverage plots in Section 3.0 have confidence curves that indicate whether the test is significant at the 5% level by showing a confidence region for the line of fit. If the confidence region between the curves contains the horizontal line, then the effect is not significant. If the curves cross the line, the effect is statistically significant. The values P are statistical p-values, which can range from zero to 1, with small values indicating the statistical significance of the associated factor. For example, Figure 3.2a shows the significance of the time factor with the very small p-value 0.0003 and the associated confidence curves crossing the dotted line. In contrast, the 0.1305 p-value in Figure 3.3a indicates that velocity is not significant at the 5% level (since 0.1305 > 0.05), and the curves contain the dotted line and do not cross it.

### 3.0 Results and Discussion

All the flux data presented in this section have been corrected to 25°C using the following formula to correct for viscosity and surface tension changes:

Flux<sub>25C</sub> = Flux<sub>T</sub>e<sup>2500(
$$\frac{1}{273+T} - \frac{1}{298}$$
) (3.1)</sup>

where  $Flux_{25C}$  is the corrected permeate flux, and T is the temperature (in °C) at the flux measurement (Flux<sub>T</sub>).

### 3.1 Active and Simulant Filtration Tests with High Purity Filter

This section compares the results from filtration tests using the Sr/TRU-treated sample of active (actual) AN-102/C-104 waste blend with the results from the simulant (inactive) testing. As shown in Figure 3.1, the flux rates are very similar, especially when the first 2 hours of testing are excluded. The average permeate flux was 0.037 and 0.034 gpm/ft<sup>2</sup> for the active and simulant tests, respectively.<sup>(a)</sup>

As can be seen in Figure 3.1, the active test was more time dependent. For the active testing, the initial flux started quite high and dropped quickly over the three conditions. For the simulant testing, the flux results started at more of an average level and actually increased slightly over the initial three conditions.



Figure 3.1. Comparison of the AN-102/C-104 Active and Simulant Tests

<sup>(</sup>a) As previously noted, the solids loading of the active and simulant waste tested was 1.35 wt% and 0.8 wt% undissolved solids, respectively. Using the empirical relationship established during dewatering (refer to Section 3.2, Figure 3.15), the estimated permeate flux for the simulant at a solids concentration equal to that of the active slurry (1.35 wt%) is 0.029 gpm/ft<sup>2</sup>. However, as noted in Section 2.4, the basis for simulant solids preparation was identical Sr/TRU precipitation reagent addition rather than solids concentration.

The cause is thought to be, at least partially, differences in initial filter conditioning. Eliminating these differences is the main reason the center point was held constant for the first 3 hours of testing. Another contributing cause is thought to be the different slurry volumes tested. The active waste was tested with approximately 40% less volume than the simulant test (0.9 liter and 1.5 liter for the active and simulant wastes, respectively). On a unit volume basis, the active waste was exposed to more pump shear, and, consequently, more time dependency should be expected.

The average flux data as a function of filtration conditions were analyzed using the JMP statistical software package (SAS Institute, Inc.) to determine the dependency of the flux on time, axial velocity, and TMP. Using linear regression and the three variables, the simulant slurry empirical model predicts 84% of the data variation (Rsquare=0.84). Of this predictive capability, 56% of the model is influenced by time, 39% by TMP, and 5% by axial velocity. The equation for permeate flux as predicted by the simulant model is:

Flux =  $2.29 \times 10^{-2} - 8.29 \times 10^{-4} \times 10^$ 

With Flux in gpm/ft<sup>2</sup>, Time in hours, Velocity in ft/s, and TMP in psid.

The active waste empirical model predicts 95% of the data variation (Rsquare = 0.95). Of this predictive capability, 89% of the model is influenced by time, 7% by axial velocity, and 4% by pressure. The equation for permeate flux as predicted by the active waste model is:

Flux =  $2.04 \times 10^{-2} - 2.06 \times 10^{-3} \times 10^{-3} \times 10^{-3} \times 10^{-3} \times 10^{-3} \times 10^{-4} \times 10^$ 

With Flux in gpm/ft<sup>2</sup>, Time in hours, Velocity in ft/s, and TMP in psid.

A visual comparison of the empirical models from the simulant and active tests is shown in Figures 3.2 through 3.4. The leverage residuals are the flux variations that remain after applying all the model parameters, except the parameter represented on the y-axis. The leverage plots are shown with confidence curves that indicate whether the test is significant at the 5% level by showing a confidence region for the line of fit. If the confidence region between the curves contains the horizontal line, then the effect is not significant. (For example, Figure 3.3a indicates that axial velocity is not a statistically significant parameter of Equation 3.2, as the confidence region contains the horizontal line.) If the curves cross the line, the effect is significant.

For both models, run order was the most important parameter. For the inactive tests, the next most important parameter was pressure, with axial velocity of marginal statistical significance. For the active tests, the axial velocity was the next most important parameter, with TMP of marginal statistical significance.

Velocity is statistically significant for the active model but not for the simulant model, while the reverse is true for TMP. This may be an artifact of the different solids loading between the active and simulant testing. It is well documented that velocity dependence increases with increased undissolved solids concentration.<sup>(a)</sup>

<sup>(</sup>a) This was observed in the current testing also, as the 2.7 wt% simulant had more velocity dependence than the 0.8 wt% simulant; see Section 3.3.



**Figure 3.2**. Comparison of Time Leverage Plot for AN-102/C-104 Sr/TRU Precipitation Slurries: (a) 0.8 wt% simulant, and (b) 1.3 wt% active



Figure 3.3. Comparison of Axial Velocity Leverage Plot for AN-102/C-104 Sr/TRU Precipitation Slurries: (a) 0.8 wt% simulant, and (b) 1.3 wt% active



**Figure 3.4**. Comparison of Transmembrane Pressure Leverage Plot for AN-102/C-104 Sr/TRU Precipitation Slurries: (a) 0.8 wt% simulant, and (b) 1.3 wt% active

To further compare the simulant data with the active data, the active model was used to establish performance criteria. These criteria consist of predictions that would be expected to result from the factor combinations in the active model. Then the simulant observed values can be compared to the active model.<sup>(a)</sup>

In Figure 3.5, the middle line is a regression through the square symbols, which represent observed flux values generated by the model for the active test. The two outer irregular lines are the 95% prediction bands for individual flux values. The simulant results (triangles) start out within the confidence bands, but fall out to the low side at higher predicted flux.

A more direct statistical comparison of the simulant and active group models can be performed. In Figure 3.6, the observed flux values for the simulant and active tests (groups) are plotted against the average of the prediction values. The dashed line in the center fits the combined data from both groups. The line highest on the right is the regression line through the square symbols representing the observed active flux values, while the line lowest on the right is the regression line through the triangle symbols representing the observed simulant flux values. If the two models were identical, the two symbol types would be more intermixed, and both regression lines would then lie on top of the dashed line.



Figure 3.5. Comparison of the Active Waste Model to the Simulant Data

<sup>(</sup>a) For both simulant and active studies, the initial filtration conditions were repeated for 3 consecutive hours before filtration conditions were varied (refer to Figure 2.2), because some of the greatest changes in flux occur in the first few hours. Such changes are of minor interest in determining the expected flux in the longer term or when comparing filters, as differences in the prior condition of the filter can bias the results. For this reason, and since this initial behavior difference has substantial impact on the resulting models, the first two conditions in each case are omitted in the comparison discussed in the remainder of this section.



Figure 3.6. Direct Comparison Between the Active and Simulant Empirical Models

A formal statistical test for equal regression lines leads to the conclusion that the two lines, and thus the two underlying models, are indeed "statistically different" from each other. The statistical test used is based on an F-distribution that compares the residual (error) sums of squares for a single regression line model (only two parameters, the slope and intercept; this is represented in Figure 3.6 by the center dashed line) to the necessarily reduced residual sums of squares when two separate regression lines are fit (four parameters, two slopes and two intercepts, represented by the other two lines in the figure). The magnitude of the reduction in residual sum-of-squares is evaluated relative to adding the two parameters. For these data, the associated F-distribution p-value, which can again range from zero to 1, with small values now indicating statistical differences, is 0.0018.

However, since both the simulant and active models fit their respective data well with minimal resulting error, even a small difference between the two models will make them "statistically significant." That is likely the case here as the underlying Rsquare values for simulant and active wastes are, respectively, 0.84 and 0.95. Thus, as stated, since these models fit their respective data sets so strongly, the rather minimal difference between the two lines becomes statistically significant. With more unexplained error for the two models, and corresponding smaller Rsquare values, the magnitude of difference observed between the two lines would not be significant. The practical impact of the differences should be considered as well as their statistical significance.

#### 3.2 Simulant Filtration Tests with Industrial Grade Filter

The purpose of the simulant filtration described in Section 3.1 was to mimic the active tests in order to make a direct comparison. The simulant results were judged to be sufficiently representative of the active waste to merit further testing. This section describes simulant testing without the slurry volume constraints of the active testing. The 13-point test matrix (shown in Figure 2.2) was run with the 0.8 wt% insoluble solids slurry. Following the test matrix, the slurry was tested for 3 hours at a single condition

without backpulsing. The slurry was then concentrated to 2.7 wt% solids, and the test matrix and extended run were repeated. Once these tests were completed, the material was washed four times in the CUF by batch additions of 1.25 liters of 0.01M NaOH, and the permeate removed by filtration.

#### 3.2.1 Low Solids (0.8 wt% Solids) Matrix

A graph of the permeate flux as a function of time for conditions 1, 2, 3, 8, and 13 (the center points of the matrix) is shown in Figure 3.7. The benefits of backpulsing in terms of increased flux are difficult to discern, as the flux immediately following backpulsing is approximately the same an hour later. The flux decreases with run order (i.e., condition), but to a much lesser extent than observed in the active testing. Lilga et al. (2002) demonstrated a 50% reduction in soluble strontium between 4 and 24 hours after precipitation reagent addition. It is speculated that at least part of the reason this slurry was less time dependent than the slurry in the September simulant testing is the additional 5 hours between reagent addition and start of filtration, suggesting a benefit of waiting as much as 24 hours after Sr/TRU precipitation reagent addition before filtration.

The variation in flux can be modeled empirically using only linear relationships with an Rsquare of 0.99; that is, roughly 99% of the variation in permeate flux is captured by the model. Of this predictive capability, approximately 97% of the model is influenced by TMP, 2% by time, and less than 1% by axial velocity. Axial velocity is of marginal statistical significance. See Figures 3.8 through 3.10 for a graphical presentation of the model.

The model is:

Flux = 
$$1.02 \times 10^{-2} - 2.30 \times 10^{-4} \times 10^$$



With Flux in gpm/ft<sup>2</sup>, TMP in psid, Velocity in ft/s, and Time in hours.

**Figure 3.7**. Permeate Flux as a Function of Time for the 0.8 wt% Solids Slurry at Nominally 11 ft/s Axial Velocity and 40 psid TMP



Figure 3.8. Effect of Transmembrane Pressure on Permeate Flux



Figure 3.9. Effect of Time on Permeate Flux



Figure 3.10. Effect of Axial Velocity on Permeate Flux

The flux is principally dependent on the TMP, which is typical of low solids slurries. In contrast, the axial velocity shows almost no influence on the flux. The time dependence is also less than observed for the simulant test with the high purity filter.

#### 3.2.2 High Solids (2.7 wt% Solids) Matrix

After the 3-hour extended run, with the low solids slurry, all available simulant slurry was dewatered to 2.7 wt% and the test matrix was repeated. The dewatering was stopped at 2.7 wt% undissolved solids due to the volume of simulant available. The permeate flux as a function of time for conditions 1, 6, and 11 (the center points of the matrix) is shown in Figure 3.11.<sup>(a)</sup> The permeate flux values are generally lower than the 0.8 wt% slurry because of the higher concentration of undissolved solids.

The variation in flux can be modeled empirically using only linear relationships with an Rsquare of 0.88; that is, roughly 88% of the variation in permeate flux is captured by the model. Of this predictive capability, 61% of the model is influenced by TMP, 26% by time, and 13% by axial velocity. As was the case for the low solids slurry, axial velocity is of marginal statistical significance. See Figures 3.12 through 3.14 for a graphical presentation of the model.

The model is:

$$Flux = 7.25 \times 10^{-3} - 6.61 \times 10^{-4} \times Time + 7.22 \times 10^{-4} \times Velocity + 3.23 \times 10^{-4} \times TMP$$
(3.5)



With Flux in gpm/ft<sup>2</sup>, TMP in psid, Velocity in ft/s, and Time in hours.

**Figure 3.11**. Permeate Flux as a Function of Time for the 2.7 wt% Solids Slurry at Nominally 11 ft/s Axial Velocity and 40 psid TMP

<sup>(</sup>a) Since the filter had already been conditioned during the 0.8 wt% testing, the first two matrix conditions in Figure 2.2 were omitted.



Figure 3.12. Effect of Pressure on Permeate Flux



Figure 3.13. Effect of Time on Permeate Flux



Figure 3.14. Effect of Axial Velocity on Permeate Flux

Comparing Equations 3.4 and 3.5 indicates that the 2.7 wt% slurry filter flux exhibited greater time dependency than the 0.8 wt% slurry (both tests were conducted in January 2002). Recall, however, that addition of simulant from the September 2001 testing was necessary to reach 2.7 wt% undissolved solids during dewatering. It appears that simulant age and time since reagent addition for Sr/TRU precipitation and the start of filtration may impact the flux time dependency.

#### 3.2.3 Extended Runs and Dewatering

Following the tests with the low and high solids matrices, the slurry was tested for 3 hours at 11 ft/s axial velocity and 40 psid TMP, without backpulsing. The results are shown in Figure 3.15. After the first 60 to 80 minutes, the slurry flux generally stopped decreasing and held within a range. The average permeate flux (excluding the first 60 minutes) was 0.029 and 0.020 gpm/ft<sup>2</sup> for the low and high solids slurries, respectively.

After the first extended run, the slurry was dewatered at 11 ft/s axial velocity and 40 psid TMP. The dewatering brought the slurry from 0.8 to 2.7 wt% undissolved solids. The system was backpulsed prior to dewatering, but not during. Figure 3.16 displays the instantaneous dewatering flux measured as a function of the log of the undissolved solids concentration, Cs. The dewatering results compare well with the first 60 minutes of the extended run. The predicted flux at 0.8 wt% and 2.7 wt% undissolved solids is 0.033 gpm/ft<sup>2</sup> and 0.024 gpm/ft<sup>2</sup>, respectively, which matches well with results in Figure 3.15. Extrapolation of the dewatering data in Figure 3.16 indicates that at undissolved solids concentrations greater than 4.6 wt% (at 40 psid TMP and 11 ft/s axial velocity) the permeate flux will be below the targeted design flux of 0.02 gpm/ft<sup>2</sup>. (The targeted design flux is an average, not a minimum.) Although extrapolations should be treated cautiously, such relationships (i.e., the flux decreasing linearly with the log of the undissolved solids concentration) have held over fairly broad ranges of wt% undissolved solids. For example, this general relationship held at all solids concentrations tested during the initial dewatering



Figure 3.15. Extended Run Without Backpulsing at Nominally 11 ft/s Axial Velocity and 40 psid TMP



Figure 3.16. Effect of Solids Concentration on Permeate Flux at Nominally 11 ft/s Axial Velocity and 40 psid TMP

of AZ-101 (between 7.6 and 17.9 wt% undissolved solids), during the dewatering of the first wash (between 14.5 and 20.2 wt% undissolved solids), and during the second wash (15.0 and 26.6 wt% undissolved solids) (Geeting et al. 2002).

After the second extended run, the slurry was batch washed four times with a 1.25-liter/batch of inhibited water (0.01M NaOH), and dewatered at 11 ft/s axial velocity and 40 psid TMP back to the original volume. The 1.25-liter/batch was approximately an equal volume wash, as the starting volume of slurry was 1.4 liters. Because all dewaterings were conducted at the same axial velocity and TMP without backpulsing, the results are directly comparable with the first dewatering data at a similar solids loading.

Figure 3.17 shows the average flux during the initial dewatering and during dewatering of each of the washes. The flux increases as the washes progress. The increase in flux is attributed to the decreased viscosity of the fluid, as shown in Figure 3.18. The viscosity of the original (prewash), wash 1, and



Figure 3.17. Average Permeate Flux Before and During Dewatering at Nominally 11 ft/s Axial Velocity and 40 psid TMP



Figure 3.18. Effect of Permeate Viscosity on Average Permeate Flux at Nominally 11 ft/s Axial Velocity and 40 psid TMP

wash 4 supernatant was measured from permeate samples to be 5.40 cP (at 22.0°C), 1.68 cP (at 21.6°C), and 1.04 cP (at 21.7°C), respectively. Not unexpectedly, the data indicate that the permeate flux is proportional to (permeate viscosity)<sup>-1</sup>. The linear fits shown have a forced zero intercept.

After simulant testing was completed, the CUF was drained and rinsed thoroughly with inhibited water (0.01M NaOH), and the flux was measured when running clean (inhibited) water and a SrCO<sub>3</sub> slurry (Section 3.3).

### 3.3 Flux Measured with Clean Water and Standard SrCO<sub>3</sub> Slurry

Flux was measured using clean water (0.01M NaOH in pre-filtered deionized water) and a 0.35M SrCO<sub>3</sub> slurry (recipe provided in Hallen et al. 2002c). The CWF discussed in this section was measured immediately before and immediately after the AN-102/C-104 Sr/TRU simulant or active slurry (as appropriate). The SrCO<sub>3</sub> slurry flux was measured immediately before and immediately after the CWF test.

The filter flux from the SrCO<sub>3</sub> slurry is less sensitive to the cleanliness of the CUF. For that reason the WTP project has begun to use the 0.35M SrCO<sub>3</sub> slurry comparison rather than CWF data to qualitatively assess the permeability of the filter before and after testing.

The SrCO<sub>3</sub> slurry results are provided in Figure 3.19. The initial SrCO<sub>3</sub> flux (before testing simulant) is approximately the same for the HP and IG filters shown, although there is greater deviation at 30 psid TMP. After the simulant testing, the HP #2 has a higher flux than the IG filter, indicating less fouling. [Recall, however, from Table 2.1, that HP #2 filter was run with the AN-102/C-104 waste blend simulant for approximately 15 hours, while IG #2 was run for approximately 33 hours. Thus, the increased fouling observed for the IG #2 is expected.]

Although more sensitive to the cleanliness of the system, the CWF data are shown in Figure 3.20 for completeness. A comparison of the final CWF from HP #2 and IG #2 indicates that IG #2 had a greater hydraulic resistance after simulant testing, which is consistent with the  $SrCO_3$  data from Figure 3.19. The



Figure 3.19. SrCO<sub>3</sub> Slurry Results Before and After Simulant Testing



Figure 3.20. Clean Water Flux Results Before and After Testing AN-102/C-104 Sr/TRU Slurry (Simulant or Active Waste, as Appropriate)

final CWF from the HP filters (used in the active and simulant testing) is virtually the same, and suggests that greater time dependency observed in the active AN-102/C-104 testing was not due to filter fouling. This result is consistent with the analysis presented in Hallen et al. (2002c), which states that the changes in the slurry caused the large time dependence.

The initial CWF from HP #2 is much higher than the initial flux from either HP #1 or IG #2. This may be indicative of a well-cleaned system. The CWF from the IG #2 filter both before and after testing was lower than that for any other filters tested, which contradicts the results from the SrCO<sub>3</sub> slurry testing and prior comparisons of the CWF from the HP and IG filters (Hallen et al. 2002c). This type of data led to the use of the standard SrCO<sub>3</sub> slurry for assessing filter permeability.

### 3.4 Particle Size Data

The particle size data were determined using a Microtrac X-100 particle analyzer. Particle size data were obtained at two flow velocities and after sonication. All particle size distributions measured are provided in Appendix B. Table 3.1 summarizes the samples analyzed and the process step in which the samples were gathered. The particle size distribution data measured were from simulant slurry samples taken during testing with the IG #2. For comparison purposes, Table 3.1 also includes particle size data from the active testing and reported in Hallen et al. (2002c).

Sample Designation	Process Step
1.4	Sample taken during low-solids slurry matrix, after approximately 2.5 hours of filtration testing.
1.8.1	Sample taken during high-solids slurry matrix, after approximately 20 hours of filtration testing.
1.9B	Sample taken during high solids extended run, after approximately 30 hours of filtration testing.
WS-1.10	Sample taken after solids wash, after approximately 31 hours of filtration testing.
Active (washed solids)	Sample taken after wash, after approximately 13 hours of filtration testing reported in Hallen et al. (2002c).

As seen in Figure 3.21, there is little change in particle size between each process step. The mean particle size shows a minor decrease after sample 1.8.1. No replicate samples were analyzed, and the variations in sample means may be a result of variability between samples. The simulant samples show a lack of trend, which is consistent with the reduced time dependency of the permeate flux when compared with the active waste.



Figure 3.21. Number Mean Particle Size With and Without Sonication

The most directly comparable sample between the simulant and active testing is the washed solids samples [samples designated WS-1.10 and Active (washed solids)]. Without sonication, the Active (washed solids) sample has a number mean over twice that of the simulant. With sonication, the number mean of the sample is approximately 60% greater than that of the stimulant, which may help explain why the filter flux of the active waste was approximately equal to that of the simulant, even though the active waste had a higher solids concentration (1.35 wt% undissolved solids vs. 0.8 wt% undissolved solids).

# 4.0 Conclusions

The following conclusions are based on the results of the testing discussed in this report:

- The average simulant flux matches the average active waste flux (both using the HP filter) within 10%, although statistical analysis of empirical models generated from testing of each indicate the models are statistically different. A comparison of the empirical models indicates that for both models time is the most important parameter, although the simulant model showed less time dependence than the active waste model. The cause is thought to be at least partially a result of differences in initial filter conditioning. Another contributing cause is thought to be the different slurry volumes tested. The active waste was tested with approximately 40% less volume than the inactive simulant test. As a result, the active waste was exposed to more pump shear, and, consequently, more time dependency should be expected.
- Because of the similar flux results obtained when running the matrix, and the known chemical similarities resulting from the precipitation process, the simulant used is believed to adequately mimic the filtration properties of the active waste, even if there are some differences in response to changes in the independent variables. Consequently, the simulant tests at high solids loading, and during dewatering and washing are considered representative of what to expect with the active waste.
- Simulant testing with the IG filter showed significantly less time dependency than was observed using the HP filter. The 0.8 wt% slurry flux was almost exclusively dependent on TMP. The 2.7 wt% slurry was also primarily dependent on TMP, but also was dependent on time and axial velocity.
- Simulant age and time since reagent addition for Sr/TRU precipitation may impact the permeate flux time dependency. Although more data are necessary to confirm the cause, we observed that the simulant filtered 11 hours after reagent addition demonstrated less time dependency than the simulant filtered 6 hours after reagent addition.
- Extrapolation of dewatering data suggests that undissolved solids concentrations greater than 4.6 wt% solids will have a flux below the targeted design flux of 0.02 gpm/ft<sup>2</sup> at 11 ft/s axial velocity and 40 psid TMP. Higher axial velocities and TMP will likely allow operation at higher solids concentrations while maintaining the flux at or above 0.02 gpm/ft<sup>2</sup>. It should be noted that the design flux of 0.02 gpm/ft<sup>2</sup> is an average over the concentration cycle. It is not a minimum.
- The flux decreased linearly with the log of the undissolved solids concentration.
- The permeate flux was shown to vary inversely with the permeate viscosity.
- During simulant testing the industrial grade 0.1-µm Mott filter fouled more than the high purity 0.1-µm Mott filter. This result is consistent with the length each filter was tested, as the industrial grade filter was used for approximately twice as long as the high purity filter.

- Particle size distribution measurements of the simulant tested in January 2002 indicate that there is not a significant change in particle size during testing, which is consistent with the lesser time dependency exhibited by the simulant compared with the active waste.
- The high purity and industrial grade filter flux from a standard SrCO<sub>3</sub> slurry were very similar, which is consistent with the analysis in Hallen et al. (2002c) that concludes there is no significant difference in the filtration characteristics between the two filters.

## 5.0 References

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Appendix A

Simulant Recipe

# Appendix A

# Simulant Recipe

Compound	Formula Weight	Grams Needed for 2-L Batch	g/L		
Organics—Prepare in 5-L stainless steel beaker.					
Water		400			
trisodium citrate	294.1	7.811	3.906		
glycolic acid	76.04	11.111	5.555		
NaFormate	68.01	16.481	8.241		
HEDTA	278.26	5.985	2.993		
Na <sub>2</sub> EDTA . 2H <sub>2</sub> O	372.2	9.891	4.945		
Iminodiacetic Acid	133.1	3.269	1.635		
Nitritotriacetic Acid	191.1	0.764	0.382		
NaGluconate	218.1	2.180	1.090		
Cations—Prepare in separate 1	-L stainless steel	beaker, then add to n	nain vessel.		
Water		400	200		
$Al(NO_3)_3 \cdot 9H_2O$	375.13	282.8	141.4		
H <sub>3</sub> BO <sub>3</sub>	61.84	0.248	0.124		
$Bi(NO_3)_3 \cdot 5H_2O$	485.07	0.008	0.004		
$Ca(NO_3)_2 \cdot 4H_2O$	236.15	2.4348	1.217		
$Cd(Cl)_2 \cdot 2.5H_2O$	228.3	0.1056	0.528		
$Ce(NO_3)_3 \cdot 6H_2O$	434.25	0.0773	0.0387		
$Co(NO_3)_2 \cdot 6H_2O$	291.03	0.0179	0.00895		
$Na_2CrO_4 \cdot 4H_2O^{(a)}$	234.0	1.197	0.598		
$Cu(NO_3)_2 \cdot 6H_2O$	295.55	0.1048	0.0524		
$Eu(NO_3)_3 \cdot 6H_2O$	446.07	0.0734	0.0367		
$Fe(NO_3)_3 \cdot 9H_2O$	404	0.2311	0.116		
KNO3	100.1	5.1485	2.574		
$La(NO_3)_3 \cdot 4H_2O$	379	0.0312	0.0156		
LiOH · H <sub>2</sub> O	41.96	0.138	0.069		
$Mg(NO_3)_2 \cdot 6H_2O$	256.41	0.0022	0.0011		
$MnCl_2 \cdot 4H_2O$	197.9	0.0536	0.0268		
$Na_2MoO_4 \cdot 2H_2O$	241.95	0.1317	0.0659		

### Table A.1. AN-102/C-104 Supernatant Simulant Composition

Compound	Formula Weight	Grams Needed for 2-L Batch	g/L	
$Nd(NO_3)_3 \cdot 5.5H_2O$	429	0.057	0.0285	
$Ni(NO_3)_2 \cdot 6H_2O$	290.81	1.8724	0.936	
$Pb(NO_3)_2$	331.23	0.2489	0.1245	
$SnCl_4 \cdot 5H_2O$	390.58	0.1678	0.0839	
Sr(NO <sub>3</sub> ) <sub>2</sub>	211.63	0.0059	0.00295	
$Zn(NO_3)_2 \cdot 6H_2O$	297.49	0.0522	0.0261	
$Zr(NO_3)_4 \cdot 5H_2O$	249.23	0.0398	0.0199	
Anions—Prepare in separate 1-	L stainless steel l	beaker, then add to m	ain vessel.	
Water		400		
NaOH <sup>(b)</sup>	40	168.71	0.084355	
NaCl	58.44	6.3874	0.0031937	
$Na_2HPO_4 \cdot 12H_2O$	380.12	20.3973	0.01019865	
$Na_2SO_4$	142.04	17.3985	0.00869925	
Na <sub>2</sub> oxalate	134	5.1342	0.0025671	
NaF	41.99	4.0758	0.0020379	
Add to main vessel				
$Na_2CO_3 \cdot 1H_2O$	159.91	255.84	0.12792	
NaNO <sub>2</sub>	69	117.79	0.058895	
NaNO <sub>3</sub>	84.99	89.55	0.044775	
<ul> <li>(a) Chromium(III) acetate was used in the original recipe, but was replaced with Na<sub>2</sub>CrO<sub>4</sub>.</li> <li>4H<sub>2</sub>O because it did not give soluble Cr near the target levels.</li> <li>(b) NoOH in original recipe was 160.71 grams, which by titration gave free hydroxide as</li> </ul>				
0.22M. An additional 8 grams of NaOH was added to increase the free hydroxide to the 0.3M target.				

 Table A.1. (contd)

**Comments**: Simulant prepared in order listed above. Simulant stirred at  $35^{\circ}C \pm 5$  during preparation and overnight after all reagents were added. Dilute final volume to 2 liters with DI water. Less than 0.1 wt% of insoluble solids was observed. Simulant composition remained constant for 2 weeks after initial preparation. However after 4 months of storage, dark solids had formed on the sides of the storage bottle and chemical analysis by ICP-AES revealed approximately 90% of the manganese and 40% of the iron had precipitated.

#### **Physical Properties of the supernatant simulant:**

Density=1.277 g/mL

Viscosity=5.397 cP

#### Sr/TRU Precipitation

Conduct precipitation within 24 hours of planned filtration tests.

#### Sr/TRU precipitation:

- 1. Dissolve 6.6134 g of Sr(NO<sub>3</sub>)<sub>2</sub> (MW 211.63) in 30 g of water.
- 2. Dissolve 4.9981 g of NaMnO<sub>4</sub> (MW 159.94) in 30 g of water.
- 3. Place 1.5 L of simulant in stirred vessel.
- 4. Add Sr solution to simulant over 5 minutes, and then stir for 30 minutes.
- 5. Add Mn solution to simulant over 5 minutes, and then stir for 30 minutes.
- 6. Digest precipitate for at least 4 hours at room temperature.

Appendix B

Particle Size Distribution Data



B.1



6 00- 3/M/02



**B.3** 



Ì Adn 3/14/02 e  $\leq$ 

B.4



NAN 3/19/02



San 3/19/02 9 5 1



Du 3/19/02 



Dul 3/19/02 D





577 3/1n/or



JAAn 3/11/02

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