Multi-Day Test of the Caustic-Side Solvent Extraction Flowsheet for Cesium Removal from a Simulated SRS Tank Waste

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ABSTRACT

To demonstrate that the caustic-side solvent extraction (CSSX) process could remove cesium from Savannah River Site (SRS) high-level waste over long periods of time, an improved minicontactor (2-cm centrifugal contactor) was needed that could be operated for several days. In particular, the contactor temperature had to be controlled and contactor hydraulic performance needed to be improved. Because the process was to be continuous, provisions were made for a three-shift operation. With the improvements made and the operators trained, the CSSX process was run in a 33-stage minicontactor over a period of three days to remove cesium from an average SRS simulant for the waste feed. The two key process goals were achieved: (1) the cesium was removed from the waste with decontamination factors greater than 40,000 and (2) the recovered cesium was concentrated by a factor of 15 in dilute nitric acid. These goals were maintained for 71 h as 1.4 L of solvent was recycled 42 times while processing 180 L of SRS simulant. The average decontamination factor for cesium was 157,000 and the average concentration factor was 14.9. The process had to be shut down twice for minor problems, which were fixed and testing resumed. This multi-day demonstration confirmed that the CSSX process could be used to help decontaminate the millions of gallons of SRS waste now stored in underground tanks.

I. INTRODUCTION

Savannah River Site (SRS) has 34 million gallons of high-level waste in 48 tanks that need to be decontaminated [LEVENSON-2000]. As a part of this process, the cesium will be removed

1

from waste containing both supernatant liquid and dissolved salt cake, then vitrified for disposal. After the cesium is removed, the decontaminated solution will be immobilized in low-level grout.

Work performed at Oak Ridge National Laboratory (ORNL) resulted in the development of a new extractant that is very specific for removing cesium from alkaline solutions such as DOE tank wastes at Savannah River, SC, and Hanford, WA [BONNESEN-1998]. The new extractant is a calixarene-crown ether, calix[4]arene-bis(*tert*-octylbenzo-crown-6), designated BOBCalixC6. In the tests reported here, BOBCalixC6 is one component in a four-component solvent. The other three components are (1) a modifier, 1-(2,2,3,3-tetrafluoropropoxy)-3-(4-*sec*-butylphenoxy)-2-propanol, also called Cs-7SB, which is an alkyl aryl polyether that keeps the extractant dissolved in the solvent and increases the ability to extract cesium in the extraction section; (2) a suppressant, trioctylamine (TOA), which suppresses effects from organic impurities to ensure that the cesium can be back-extracted from the solvent in the strip section; and (3) a diluent, Isopar$^®$L, which is a mixture of branched hydrocarbons. The specific solvent composition for the tests reported here is 0.01 $\underline{M}$ BOBCalixC6, 0.50 $\underline{M}$ Cs-7SB, and 0.001 $\underline{M}$ TOA in Isopar$^®$L and is designated the "CSSX solvent." After the work reported here was completed, the composition of the CSSX solvent was changed to 0.007 $\underline{M}$ BOBCalixC6, 0.75 $\underline{M}$ Cs-7SB, and 0.003 $\underline{M}$ TOA in Isopar$^®$L [KLATT-2002]. In FY1998, an earlier version of the CSSX solvent with a different modifier and no suppressant was used for solvent extraction tests carried out in centrifugal contactors at Argonne National Laboratory (ANL). These tests first demonstrated that cesium could be extracted from caustic simulants representative of the high-level waste at SRS using plant-type equipment [LEONARD-1999, -2001C]. After additional work at ORNL in FY1999 [BONNESEN-2000] to improve the solvent and at ANL in FY2000 [LEONARD-2001A] to improve stage efficiency in the 2-cm centrifugal contactor, the CSSX flowsheet required for removing cesium from high-level waste at SRS was demonstrated with a waste simulant [LEONARD-2000]. In these proof-of-concept tests, the CSSX process was demonstrated both for once-through solvent operation and for solvent recycling up to four times in three hours. The improved CSSX process achieved both key process goals: (1) the cesium was removed from the waste with decontamination factors greater than 40,000 and (2) the recovered cesium was concentrated by a factor of 15 in dilute nitric acid.

The annular centrifugal contactor used in testing the CSSX flowsheet was a design that had been developed at ANL in the early 1970s for carrying out solvent extraction operations required in the nuclear industry [BERNSTEIN-1973]. It improved on the earlier design developed at SRS in the 1960s in which a paddle mixer below the rotor mixed the two immiscible phases [WEBSTER-1969]. A schematic of the annular centrifugal contactor, given in Fig. 1, shows where the more-dense phase (the aqueous phase for the CSSX process) and the less-dense

phase (the organic phase for the CSSX process) enter and exit the unit. Detailed discussion of contactor operation can be found elsewhere [BERNSTEIN-1973, LEONARD-1980]. Many contactor stages are combined to run the CSSX flowsheet. Seventeen stages make up the extraction and scrub sections where cesium is recovered from the waste feed stream. Fifteen stages make up the strip section where cesium is recovered from the solvent. The final stage is the wash section where degradation products are removed from the solvent. In the contactor used for the test discussed here, the rotor has a 2-cm diameter (hence, the name "2-cm centrifugal contactor"). Its design has been changed over the years to improve operability and increase stage efficiency [LEONARD-1997, -2001A]. In this work, two new ways were found to improve hydraulic performance of the contactor; one is specific to the CSSX process using the 2-cm contactor and the other is general for all contactors. In addition, two ways were evaluated for controlling the temperature in the contactor stages– thermoelectric devices and water-cooled chilling bars.
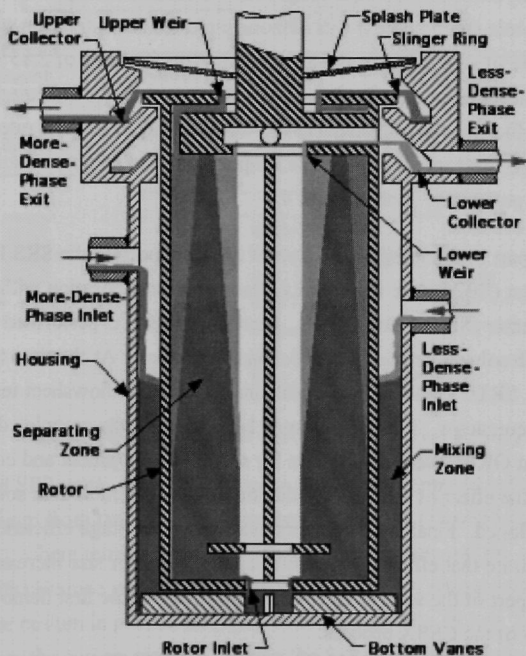
Fig. 1. Schematic of Operating Contactor Stage

The test reported here extends the above-mentioned proof-of-concept work by carrying out a multi-day test of the CSSX flowsheet. The flowsheet for the multi-day test, given in Fig. 2, shows the nominal flow rates for the test. This flowsheet is different from the previous flowsheet [LEONARD-2000] in that a solvent-wash stage has been added, stage 33. The solvent-wash stage is located next to the extraction section, stages 1-15, so that the organic strip effluent (EP) can be sampled directly to determine the effectiveness of the strip section in removing cesium from the solvent. As with the earlier test, there were two scrub stages, stages 16 and 17, and fifteen strip stages, stages 18-32. In this test, designated "CS27," 1.4 L of CSSX solvent was used to process 180 L of SRS simulant. The solvent was recycled a total of 42 times. The initial test plan was to operate at the nominal maximum throughput of the 2-cm contactor, which is 40 mL/min (both phases) and occurs in the extraction section, so that the SRS simulant feed rate would have been 28.7 mL/min and the aqueous strip feed rate would have been 1.90 mL/min. At these rates, it would take 105 h (4.4 d) to process the 180 L of simulant. This initial plan gave rise to the name "5-day test." In subsequent hydraulic tests of the 2-cm contactor, a maximum throughput of 60 mL/min (both phases) was demonstrated for the extraction section, allowing an SRS simulant feed rate of 43.0 mL/min and an aqueous strip feed rate of 2.85 mL/min. The higher feed rate for the aqueous strip feed was used, since experience indicated that higher flow rates improve stage efficiency [LEONARD-1999, -2001B]. At these flow rates, it takes only 71 h (2.94 d) to process the 180 L of simulant while keeping the total number of solvent cycles, the desired experimental parameter, unchanged at 42.

This work is part of the integrated scope of work supporting the SRS High-Level Waste Salt Processing Project (SPP). The work was performed in collaboration with the Savannah River Technology Center (SRTC) and ORNL. Personnel at SRTC performed tests with real waste, including batch solvent extraction and solvent irradiation. At the time the work reported here was being done, SRTC personnel were planning for a CSSX flowsheet test with real waste in the 2-cm centrifugal contactor. This test has now been successfully completed [CAMPBELL-2001A]. Personnel at ORNL were responsible for solvent development and commercialization. They also evaluated the effect of heat and irradiation on the solvent so that solvent cleanup methods can be developed. Finally, ORNL personnel measured stage efficiency in a larger (5-cm) contactor to demonstrate that efficiency increases as the contactor size increases. The multi-day test at ANL is a key part of the solvent extraction effort, as it is the first demonstration of the long-term operability of the CSSX process.

Caustic-Side Tank
Waste Feed
(Avg SRS Sim)
(DF)
Flow = 43.0 mL/min

Scrub Feed
0.05 $\underline{M}$ HNO$_3$
(DS)
Flow = 2.82 mL/min

Wash Feed
0.01 $\underline{M}$ NaOH
(FF)
Flow = 2.85 mL/min

Strip Feed
0.001 $\underline{M}$ HNO$_3$
(EF)
Flow = 2.85 mL/min

FX

| 1 Stage | 15 Stages | 2 Stages | 15 Stages |

Wash (33)   Extraction (1-15)   Scrub (16-17)   Strip (18-32)

Wash Effluent
(Only NaOH)
(FW)
Flow = 2.85 mL/min

Aqueous Raffinate
(All components except Cs)
(DW)
Flow = 45.82 mL/min

Strip Effluent
(Only CsNO$_3$)
(EW)
Flow = 2.85 mL/min

CSSX Solvent
0.01 $\underline{M}$ BOBCalixC6
0.50 $\underline{M}$ Cs-7SB
0.001 $\underline{M}$ TOA
Isopar®L (rest)
(FX, EP)
Flow = 14.12 mL/min

EP

Fig. 2. CSSX Flowsheet for Multi-Day Test (CS27)

In the overall SPP work, the CSSX process was being compared with two alternatives that also remove cesium from tank waste. These processes were (1) small tank tetraphenylborate precipitation (STTP), where tetraphenylborate is used to precipitate out the cesium in small tanks, and (2) crystalline silicotitanate non-elutable ion exchange (CST), where crystalline silicotitanate is used to remove the cesium in packed beds [LEVENSON-2000]. In July 2001, the CSSX process was chosen as the cesium removal step for the Salt Waste Processing Facility (SWPF) designed to treat the SRS tank waste.

II. EQUIPMENT AND MATERIALS

This solvent extraction test was performed in a 33-stage, 2-cm annular centrifugal contactor located in an ANL glovebox. The contactors, which are manufactured in banks of four stages, were built at ANL (see ANL print number CMT-E1265 entitled "2-cm Contactor" and dated 1/6/94). The 33 contactor stages were modified to improve stage efficiency to 88 ± 4%, as discussed elsewhere [LEONARD-2000, -2001A]. Because the distribution ratio for cesium is very sensitive to temperature, increasing with decreasing temperature, the extraction stages must be kept between 20 and 32°C. If they get too hot, they will no longer be able to maintain the desired decontamination factor. To control the extraction section temperature at 26 ± 2°C, a water-cooled chiller bar was used. The water, which was at 7.5°C, was recirculated from a cooling bath located outside the glovebox. Since higher temperatures improve stripping, no chiller bar was needed for the strip section. The block temperatures there reached 35 ± 3°C during the test.

A "four-component simulant" was used for hydraulic testing in single-stage and multistage 2-cm contactors, preliminary CSSX flowsheet tests, and start-up of the multi-day CSSX flowsheet test. This simulant approximates the composition of the SRS tank waste solution. It was prepared by dissolving 127.48 g of NaOH, 105.04 g of $Al(NO_3)_3 \cdot 9H_2O$, 34.5 g of $NaNO_2$, and 99.85 g of $NaNO_3$ in water for each liter of solution. It has an estimated density of 1226 g/L at 22°C [WALKER-1998]. As this simulant is prepared, NaOH reacts with $Al(NO_3)_3$ as follows:

$$Al(NO_3)_3 + 4\ NaOH \rightarrow NaAlO_2 + 3\ NaNO_3 + 2\ H_2O \qquad (1)$$
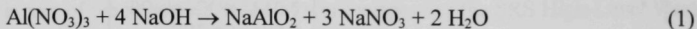
Thus, the final ionic composition of the four-component simulant is 4.86 $\underline{M}$ Na^+, 2.07 $\underline{M}$ OH^-, 2.01 $\underline{M}$ NO_3^-, 0.5 $\underline{M}$ NO_2^-, and 0.28 $\underline{M}$ AlO_2^-.

For the multi-day CSSX flowsheet test, highly alkaline simulant for tank supernate waste was prepared using a method supplied by SRS personnel [PETERSON-2000A]. It has the composition given in Table 1 and is designated the "average SRS simulant" or "SRS simulant." Based on its composition, its estimated density is 1258 g/L at 22°C [WALKER-1998]. Its measured density for the multi-day test was 1250.8 ± 2.6 g/L at ambient temperature. When it was prepared for the proof-of-concept tests, the SRS simulant had a measured density of 1254.9 ± 0.9 g/L at 20.4 ± 0.1°C. Before the start of the multi-day test, the SRS simulant was spiked with 0.5 mCi/L of ^{137}Cs. The scrub feed was 0.05 $\underline{M}$ HNO_3, the strip feed was 0.001 $\underline{M}$ HNO_3, and the solvent wash feed was 0.01 $\underline{M}$ NaOH. All four aqueous feeds were prepared at ANL. The CSSX solvent, whose composition was 0.01 $\underline{M}$ BOBCalixC6, 0.50 $\underline{M}$ Cs-7SB, and 0.001 $\underline{M}$ TOA in Isopar®L, was prepared at ORNL and shipped to ANL.

Table 1. Composition of Average SRS Simulant

Component	Conc., mol/L	Component	Conc., mg/L
Na^+	5.6	Copper	1.44
K^+	0.015	Chromium	75
Cs^+	0.00014[a]	Ruthenium	0.82
OH^-	2.06	Palladium	0.41
NO_3^-	2.03	Rhodium	0.21
NO_2^-	0.50	Iron	1.44
AlO_2^-	0.28	Zinc	8
CO_3^{2-}	0.15	Tin	2.4
SO_4^{2-}	0.14	Mercury	0.05
Cl^-	0.024	Lead	2.1
F^-	0.028	Silver	0.01
PO_4^{3-}	0.007	Tri-n-butyl phosphate (TBP)	0.5
$C_2O_4^{2-}$	0.008	Di-n-butyl phosphate (DBP)	25
SiO_3^{2-}	0.03	Mono-n-butyl phosphate (MBP)	25
MoO_4^{2-}	0.000078	n-Butanol	2
NH_3	0.001	Formate	1500
		Tri-methylamine (TMA)	10

[a] This is the total Cs concentration in the average SRS tank waste. It is the Cs concentration used here with the addition of ^{137}Cs at a tracer level of 0.5 mCi/L. The ^{137}Cs concentration in the average SRS tank waste is 22.6% of the total Cs concentration.

III. TEMPERATURE CONTROL

Since the distribution ratio for cesium is a strong function of temperature, good temperature control is required for successful CSSX process operation [BONNESEN-2000]. In general, the extraction section should be kept cooler than the strip section. During the proof-of-concept tests for the CSSX flowsheet, the original temperature management plan (operate at 25°C) had to be revised (operate at 18°C, cool the waste (DF) feed using an ice bath, and start the motors in the extraction section last) to achieve a successful demonstration [LEONARD-2000, -2001A]. In the first test, it was found that the aqueous (DF) raffinate from the extraction section had to have a temperature of 32.5 ± 0.5°C or less to achieve a cesium decontamination factor greater than 40,000. With the revised temperature management plan, the temperature of DF raffinate leveled out at 30.9 ± 0.5°C after two hours and stayed there for the final hour of the test. As a result, the cesium decontamination factor was greater than 40,000. While the revised temperature management plan worked, the temperature of the DF raffinate rose and leveled out

only 1.4°C below the maximum allowable temperature. Thus, better temperature control was needed for a test that would last several days.

As a first step in getting better temperature control, the sources and amount of the heat were determined. This work was reported elsewhere [ARAFAT-2001]. Most of the heat was found to be coming from the motors spinning the contactor rotors. These motors, which operate at 3600 rpm, are AC induction motors, model 710 from the Bodine Electric Company, Chicago, IL. They have an output power of 0.83 W (1/900 hp), about twice the 0.3 to 0.4 W required per stage to run the rotors [ARAFAT-2001]. Since the input power is 12 W (0.102 A at 115 V), most of the power is dissipated as heat. Much of this heat travels by conduction from the motor to the motor mount plate to the contactor housing. Some travels down the rotor shaft to the rotor body. The rest is dissipated directly to the air. For a conservative design, the cooling system must be able to remove 12 W of heat per stage, 180 W of heat for the 15 extraction stages. Since higher temperatures give better stripping and the scrub and wash sections are not temperature sensitive, there was no attempt to cool these sections.

To improve temperature control for a multi-day test, two methods were evaluated for heat removal: thermoelectric cooler (TEC) modules and chiller bars. The chosen method would be applied to the 15 extraction stages. It was determined that either device would work. The advantages of the TEC modules are that they allow individual temperature control for each stage and would make direct contact with the contactor housing. The disadvantages of the TEC modules are (1) each stage would have two pairs of wires, with each pair requiring a device to control voltage and current; (2) the heat sink and fan for each stage would take up significant room; (3) the modules, while sealed, could be compromised if liquid got on them; and (4) the modules would be hard to remove, as they are glued to the contactor body. The advantages of the chiller bars are (1) only one line into and out of the glovebox would be required and those penetrations were already in place, (2) the chiller bars and connecting lines would not be affected by the chemicals in the glovebox, (3) the chiller bars could be placed out of the way of other operations, and (4) they could be removed easily, if needed. The disadvantages of the chiller bars are (1) all stages would be cooled to the same temperature and (2) a spacer would be required to conduct the heat from the contactor to the chiller bar.

Chiller bars were chosen, since they were easier to install in the glovebox, easier to remove, and less likely to be damaged by other glovebox operations. Cooling all of the extraction stages to the same temperature was not considered to be a problem. A spacer, a half-moon piece as shown in Fig. 3 (crescent-shaped heat-transfer block), was tested and found to transfer enough heat from the contactor to the chiller bar for control of stage temperature. The half-moon spacer

was 7/8 in. (22.2 mm) high by 1 in. (25.4 mm) wide. Its outer surface was flat to match the chiller bar. Its inner surface was curved to match the lower contactor housing. The minimum distance between these two surfaces was 1/4 in. (6.4 mm). The chiller bars and half-moon pieces were held on by a clamping system that was out of the way. To improve heat transfer between the contactor and the chiller bars, a heat-transfer paste, Thermon® T-85 (Thermon Manufacturing company, San Marcos, TX), was placed on two surfaces of each half-moon piece, the one that touched the chiller bar and the one that touched the lower contactor housing. The chiller bars were cooled by water flowing in series from bar to bar. The exiting chiller water was recirculated, passing through a rapid-cool, constant-temperature circulating bath, model U-12101–40 (Cole-Parmer, Vernon Hills, IL), with a temperature range from -45 to 200°C, a cooling water flow rate of 11 to 24 L/min with no pressure head, a bath volume of 13 L, and a cooling capacity of 900 W at 20°C. This system worked very well.
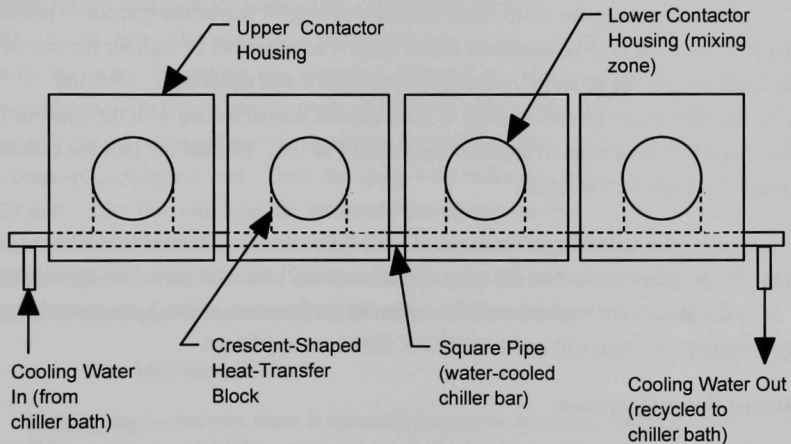
Fig. 3. Schematic Showing Top View of Setup for Four-Stage Chiller Bar

Further details on the heat-up of the contactor stage by the rotor motor, setup and operation of the TEC module, and the setup and operation of the chiller bars are given in Appendix A.

IV. HYDRAULIC PERFORMANCE

Some hydraulic problems were observed when the 2-cm contactor was used to carry out the three proof-of-concept tests [LEONARD-2001A]. While these problems were not severe enough to ruin the proof-of-concept demonstration of the CSSX process, a more hydraulically

stable system was desired for the multi-day test. Some problems observed during these three tests were as follows: (1) The liquid level in some interstage lines was high, with 50 to 90% of the line being filled. (2) In subsequent tests for the extraction section, different interstage lines would exhibit these high liquid levels; sometimes the liquid level in the interstage lines would change during the test. (3) Aqueous droplets were seen flowing with the organic phase in the organic interstage lines of the extraction section. (4) Significant liquid appeared in the standpipe of one extraction stage even though the interstage lines were not completely filled. (5) The liquid levels in the organic interstage lines were typically greater than those for the aqueous interstage lines. After the first test, CS23, a motor/rotor assembly was replaced at stage 27 in the strip section. This change solved the problem of high liquid level in both the aqueous and organic interstage lines of the strip section for the other two tests. During the third test, CS25, a motor/rotor assembly was replaced at stage 4 in the extraction section to eliminate the high liquid level in the standpipe. At times, the liquid level was so high that liquid flowed over the top of the standpipe into the glovebox. Replacing the motor/rotor assembly at stage 4 solved the immediate problem, but the liquid level in the organic interstage line to stage 4 continued to be high for the rest of the test. The liquid level in the aqueous interstage line to stage 4 was never high. After the motor/rotor assembly was replaced at stage 4, some solvent started exiting with the aqueous DW raffinate at stage 1. The amount of solvent varied from 1 to 9%. This did not hurt the cesium decontamination factor for test CS25.

To improve the hydraulic performance of the 2-cm contactor, various ideas were considered. These ideas focused on (1) rotor operation and (2) the inlet ports into the contactor stages. After the ideas were implemented, the hydraulic performance of the 2-cm contactor was tested by operating a 4-stage unit under the CSSX flowsheet conditions.

1. Improved Rotor Operation

To improve the hydraulic performance of rotor operation, three items were considered: (1) improving the maximum throughput for a single-stage rotor, (2) minimizing the total indicating runout (TIR) of the rotor, and (3) maintaining the rotational speed of the rotor.

a. Increased Maximum Throughput

As a general rule of thumb, multistage banks of rotors should be operated at no more than 50 to 67% of their maximum throughput for single-stage operation. The maximum throughput is defined as the point where, as the flow rate is increased, the other-phase carryover in at least one effluent stream becomes greater than 1%. By keeping flow rates at no more than 50 to 67% of their maximum throughput for single-stage operation, one prevents the occurrence of a positive

feedback loop. Such a loop can occur when the flow rate fluctuates above its steady-state value while operating near the maximum throughput for single-stage operation. The increased flow causes some other-phase carryover to occur. This carryover returns to the stage from which it just came. It is immediately recycled back to the current stage, giving a further increase in total throughput for the current stage, and so generates even more other-phase carryover. This process creates a positive feedback loop that will not recover even when the initial flow fluctuation disappears. By keeping stage throughput (both phases) at no more than 50 to 67% of maximum throughput, flow fluctuations in multistage operation are able to dissipate without upsetting contactor hydraulics.

For the extraction section of the 2-cm contactor used to carry out the CSSX flowsheet, this general rule of thumb was not followed in earlier tests. All flow rates were increased proportionally so that the aqueous flow rate in the strip section would be above 2 mL/min. The maximum total throughput for single-stage operation at the CSSX extraction section conditions was 82 ± 10 mL/min. The actual total throughput was 60 mL/min for multistage operation. When the maximum throughput was exceeded, the mode of failure was aqueous phase in the organic effluent (A in O). When the mode of failure is of this type, that is, the more-dense phase exiting with the less-dense phase, the maximum throughput can be increased by opening up the more-dense-phase (upper) weir. Thus, the upper-weir radius of the rotor was increased from 6.12 to 6.24 mm. After this was done, the maximum throughput increased to 102 ± 10 mL/min. As before, the mode of failure for the modified rotor was A in O when the maximum throughput was exceeded. Since the modified rotor met the criteria set by the rule of thumb for multistage operation of centrifugal contactors, all of the rotors were modified in this way.

b. Minimizing TIR

In designing contactors, there is currently no way to determine the required TIR for the rotor in advance. In general, it should be small. However, if the TIR specification is set too tight, machining costs become higher than necessary. The TIR represents the combined effect of (1) the rotor body being out of round, (2) the rotor body being not quite concentric with the rotor shaft, (3) the rotor shaft being not quite concentric with the motor shaft, (4) the motor shaft being not quite concentric with the motor bearings, and (5) the motor shaft being bent. The method used for measuring TIR is given in Appendix C. If the TIR is high, it can generate a pressure pulse that would move around the annulus as the rotor spins and create a pressure on rotor inlets. This pressure could cause the liquid flowing between stages to backup in the interstage lines. Initially, allowable TIR values had been as great as 0.012 in. (0.305 mm). Since the annular gap is 0.062 in. (1.59 mm), the TIR was 19% of the gap width. To improve contactor performance, this

specification was tightened to 0.010 in. (0.254 mm). Our experience indicates that this specification is adequate. If a 2-cm contactor were built with tangential inlet ports, as discussed next, it should be possible to return to the original limit for the TIR. For larger contactor rotors, some increase in the maximum allowable TIR may be possible. This is an area where careful attention is needed.

c. Maintaining Rotor Speed

For proper contactor operation, the rotor should be rotating at its design speed. This is especially important when the unit is designed to be partially pumping, which is the case here [LEONARD-2001A]. Initially the motor was checked to see that it rotated in the right direction (clockwise when looking down on the contactor from above) at the proper speed (3600 rpm). When this test was done, the time it took for the motor to spindown was noted. Then, after each step in the assembly of the motor/rotor system, the spindown time was checked to see if it had changed. This allowed us to determine that the spinning parts were not rubbing on the motor mount plate, the vanes below the rotor, the plastic inserts, or the wire ropes. If the spinning portion of the motor/rotor assembly were rubbing against any of these objects, the rotor speed could be reduced. Before the multi-day test, each stage was tested to be sure that the rotor was turning freely and that its speed and direction of rotation were appropriate.

2. Improved Inlet Ports

For the 2-cm contactor used here, the inlet port is perpendicular to the annular mixing zone. In addition, with the partially pumping rotor and the high flow rate in the extraction section, the liquid level in the annular mixing zone is up to the level of the inlet ports. As a result, the swirling annular liquid can put pressure on the liquid coming in from the interstage line and cause it to back up. This degrades the hydraulic performance of the contactor. Subtle differences in machining and welding around the inlet ports as well as a high TIR can contribute to this pressure on the incoming liquid. Note that, if the liquid level is not up to the inlet ports, their orientation is not important. Also, if liquid is being pumped to the inlet port, its orientation is not important. To improve the operation of the inlet ports, three design options were considered: (1) tangential inlet port, (2) inlet port between tangential and perpendicular, and (3) modified perpendicular inlet port.

a. Tangential Inlet Port

The best design option would be to have the inlet port tangential to the annular mixing zone so that the incoming liquid joins with the swirling liquid in the annular mixing zone, both liquids moving in the same direction. For this design, annular liquid actually helps draw the

incoming liquid into the mixing zone. Based on the work here, a tangential inlet port is recommended anytime a new contactor is built. For an existing unit, this change can be very hard to make. As that was the case here, the tangential inlet port was not used.

b. Inlet Port between Tangential and Perpendicular

A second design option is to place the inlet port between the tangential and perpendicular orientations. For the 2-cm contactor, which has a 1.000-in. (25.4-mm) diameter for the outer wall of the annular mixing zone and a 0.875-in. (22.2-mm) diameter for the inner wall (the outer surface of the rotor), the aqueous phase inlet was displaced 0.156 in. (4.0 mm) from the perpendicular towards the tangential. This displacement is probably the reason that the liquid levels in the aqueous interstage lines for the proof-of-concept tests were almost never a problem, even when the organic interstage lines were very full [LEONARD-2001A].

c. Modified Perpendicular Inlet Port

The third design option is to improve the existing perpendicular inlet ports. This is done by modifying the inlet opening so that the flowing interstage liquid is protected from the swirling annular liquid as the interstage liquid enters the mixing zone. For the existing 2-cm contactor, a protective space was created by using an insert. It extended from the inlet port halfway into the annular mixing zone, that is, 0.8 mm into the 1.6-mm width of the mixing zone. As shown in Fig. 4, the insert was oriented so that it shielded the entering liquid from the swirling liquid in the annulus. This improvement can be applied to all existing contactors. When the inlet ports are tangential, the need for an insert should be eliminated. When the inlet port is between the tangential and perpendicular orientations, the need for an insert is minimized. In the tests reported here, inserts were used in the aqueous inlet ports as well as the organic inlet ports.
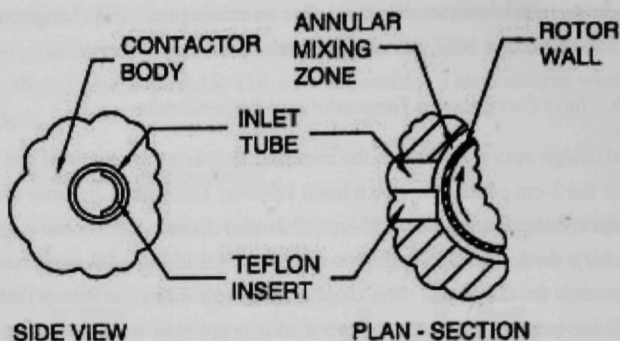
Fig. 4. Position and Orientation of an Insert in the Inlet Tube That Goes to the Annular Mixing Zone

3. Multistage Operation

After all of these improvements were implemented in the 2-cm contactor, hydraulic performance was tested by operating a 4-stage unit under the CSSX extraction section conditions shown in Fig. 2. These tests showed that the hydraulic performance had been improved. As given in Table 2, the liquid level in the organic interstage lines, which had been 40 to 100%, dropped to 5%. At the same time, the aqueous phase seen flowing in the organic interstage lines disappeared. Finally, the liquid level in the aqueous interstage lines, which had been good (10%), became even better (5%). As shown in Table 3, these improvements eliminated the significant carryover of aqueous phase in the organic effluent. With these improvements, the hydraulic performance was satisfactory for a multi-day test.

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Table 2. Hydraulic Performance of Interstage Lines in a Four-Stage 2-cm Contactor under Extraction Section Conditions for the CSSX Flowsheet

Conditions	Side where Interstage Line Was Located	Rotor Series	Interstage Line	Amount of Liquid in Interstage Line, %	Other Phase Flowing through Interstage Line (Yes or No)
Original[a,b]	O	X	1 to 2	50	Yes
	O	X	2 to 3	40	Yes
	O	X	3 to 4	40	Yes
	O	B	1 to 2	50	No
	O	B	2 to 3	40	Yes (~30%)
	O	B	3 to 4	100	Yes (~25%)
	A	X	4 to 3	10	No
	A	X	3 to 2	10	No
	A	X	2 to 1	10	No
	A	B	4 to 3	10	No
	A	B	3 to 2	10	No
	A	B	2 to 1	10	No
Improved[b,c]	O	X	1 to 2	5	No
	O	X	2 to 3	5	No
	O	X	3 to 4	5	No
	A	X	4 to 3	5	No
	A	X	3 to 2	5	No
	A	X	2 to 1	5	No

[a] The X series rotors, X1 - X4, and the B series rotors, B1 - B4, have upper-weir radii of 6.12 mm. Tests were done at 60 mL/min total flow, O/A flow ratio of 0.3, and the same contactor configuration used in the CSSX proof-of-concept tests [LEONARD-2001A].

[b] The aqueous feed was the four-component simulant.

[c] In these tests, the X series rotors had upper-weir radii of 6.24 mm. Tests were done at 60 mL/min total flow, O/A flow ratio of 0.3, and the improved contactor configuration. The improvements included the larger upper weirs, good spindown times, TIR values of 0.010 in. (0.254 mm) or less, and plastic inserts for the inlet ports. Wire ropes were not used in the interstage lines.

Table 3. Hydraulic Performance of Effluents in a Four-Stage 2-cm Contactor under Extraction Section Conditions for the CSSX Flowsheet

Conditions	Effluent	Rotor Series	Amount of Other-Phase Carryover, %
Original[a,b]	O	X	40
	O	B	40
	A	X	0
	A	B	0
Improved[b,c]	O	X	0
	A	X	0

[a] The X series rotors, X1-X4, and the B series rotors, B1-B4, have upper-weir radii of 6.12 mm. Tests were done at 60 mL/min total flow, O/A flow ratio of 0.3, and the same contactor configuration used in the CSSX proof-of-concept tests [LEONARD-2001A].
[b] The aqueous feed was the four-component simulant.
[c] In these tests, the X series rotors had upper-weir radii of 6.24 mm. Tests were done at 60 mL/min total flow, O/A flow ratio of 0.3, and the improved contactor configuration. The improvements included the larger upper weirs, good spindown times, TIR values of 0.010 in. (0.254 mm) or less, and plastic inserts for the inlet ports.

Further details on the hydraulic performance of the 2-cm contactor stage are given in Appendix B.

V. PREPARATIONS AND OPERATIONS DURING THE MULTI-DAY TEST

Preparations for the multi-day test (CS27) included working with the contactor housings, rotor bodies, and feed tanks; training personnel; and an operational readiness review. The contactor housings and rotor bodies were cleaned before the test. The upper weir of each rotor was enlarged after appropriate hydraulic performance tests. After this change, each rotor was tested hydraulically to see that its operation was satisfactory. As the motor/rotor assemblies were mounted, they were tested to be sure that rotor speed and direction of rotation were correct; the TIR was within specifications; and rotor rotation was not hindered by the contactor housing, the wire rope, or the insert. An additional contactor stage, stage 33, was added to provide for the solvent wash section. The cooling system for the extraction section was built and installed. The hot feed drum for test CS27 was located in a limited access area outside the glovebox to minimize radiation exposure to personnel during the test. Two low-level waste (raffinate) drums were located in a limited access area outside the glovebox to minimize the number of bag-outs required during the test. Enough of each feed solution was prepared to allow 1.4 L of the solvent to be recycled 42 times. Because of the extended operation of the contactor, extensive personnel training was given to the operators on all three shifts. This training included hands-on operation of the contactor by each shift. Before test CS27 was started, our preparations were examined by an operational readiness review team selected by SRS [CAMPBELL-2001B, HINDS-2001].

Operations during the multi-day test included the process operation itself, process upsets, shift operation, sampling and analysis, and flow-rate measurements. Process operations were designed to minimize individual and collective radiation dosages. Log sheets were developed to record the pertinent data and observations. A typical procedure was used to start up the contactor. During the test, two process upsets occurred. First, a pump failed and had to be replaced. Second, the rotor in the simulant feed stage, stage 15, became partially plugged and was cleaned out. A special log sheet was developed to facilitate the transmission of information between shifts. Samples were taken and analyzed on a timely basis during the test to identify any process problems while the test was still in progress. During the test, flow rates were measured on a regular basis and corrections were made as needed.

Further details about the preparations for and work during the multi-day test are given in Appendix C.

VI. RESULTS

The CSSX process was tested in a 33-stage 2-cm centrifugal contactor using the flowsheet shown in Fig. 2. In the extraction section, the solvent extracts cesium from the aqueous SRS simulant as the two phases flow countercurrent to each other. That is, as the solvent flows from stage 1 to 15 (see Fig. 2), its Cs concentration increases; as the SRS simulant flows in the opposite direction, from stage 15 to 1, its Cs concentration decreases. The effectiveness of the extraction section is defined by the decontamination factor, which is the concentration of cesium in the aqueous SRS simulant (DF) entering stage 15 divided by that in the aqueous raffinate (DW) exiting stage 1. As shown in Fig. 5, the average decontamination factor for the test CS27, 157,000 ± 44,000, exceeded the process goal of 40,000 by a factor of 4.
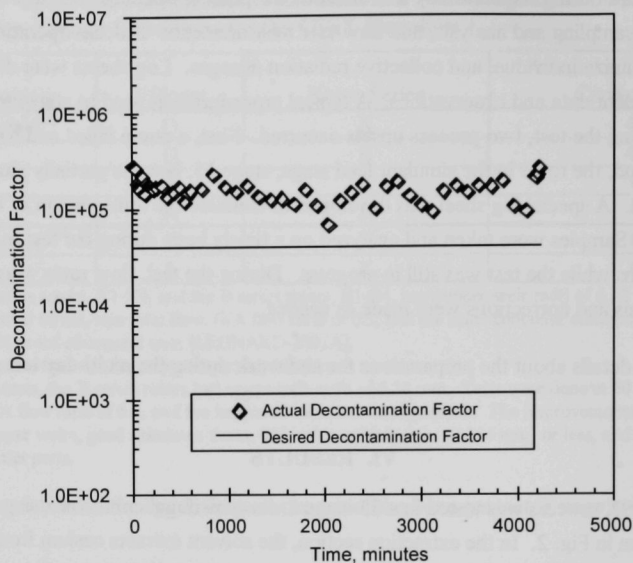
Fig. 5. Changes in the Decontamination Factor vs. Time for Test CS27

The solvent leaving the extraction section at stage 15 is loaded with essentially all of the Cs that entered with the SRS simulant waste feed (DF). The solvent enters the scrub section, stages 16 and 17 in Fig. 2, where weakly extracted impurities are removed along with any entrained aqueous phase. In addition, the entrained alkaline aqueous phase is neutralized and made slightly acidic. The scrubbed solvent then enters the strip section at stage 18. In the strip section, the Cs is recovered from the solvent and comes out in the aqueous strip effluent (EW). Since the flow of the aqueous strip feed (EF) is 1/15 of the aqueous waste feed (DF) flow and since essentially all of the Cs entering in the DF feed goes out in the aqueous strip effluent (EW), the Cs is concentrated by a factor of 15 in the EW effluent. This [EW]/[DF] ratio, called the concentration factor, is plotted in Fig. 6 as a function of time for test CS27. The average value of the concentration factor is 14.9 ± 1.0, which meets the second process goal. At 1043 min into test CS27, the waste (DF) feed rate was determined to be low and was increased by 10%. Before this change, the average concentration factor was 13.8 ± 0.4; afterwards, 15.6 ± 0.6. These results show clearly how the concentration factor depends on the waste (DF) and strip (EF) feed rates and is controlled by them.

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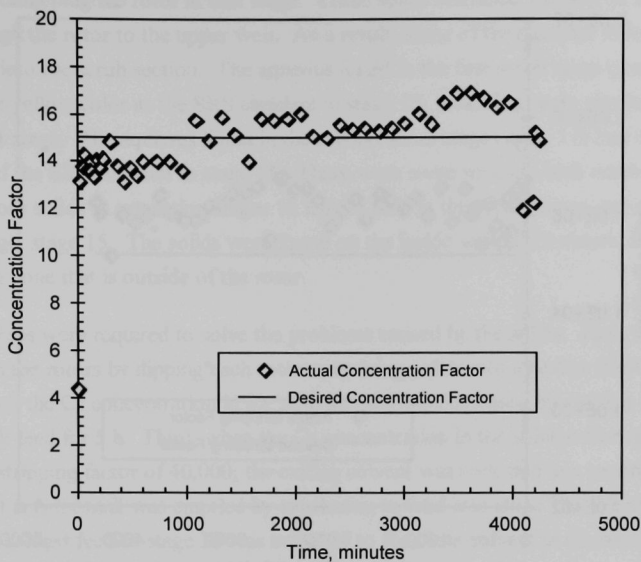
Fig. 6. Changes in the Concentration Factor vs. Time for Test CS27

In the strip section, the solvent flows countercurrent to the aqueous strip solution of 0.001 $\underline{M}$ HNO$_3$. This solution strips the cesium from the solvent. That is, as the solvent flows from stage 18 to 32 in Fig. 2, its Cs concentration decreases; as the aqueous strip solution flows in the opposite direction, from stage 32 to 18, its Cs concentration increases. The effectiveness of the strip section is defined by the stripping factor, which is the Cs concentration in the aqueous SRS simulant (DF) entering stage 15 divided by that in the organic effluent (EP) exiting stage 32. As shown in Fig. 7, the average stripping factor for the test CS27, 125,000 ± 48,000, exceeded the process goal of 40,000 by a factor of 3. The required stripping factor is the same as the decontamination factor so that the solvent can be recycled.
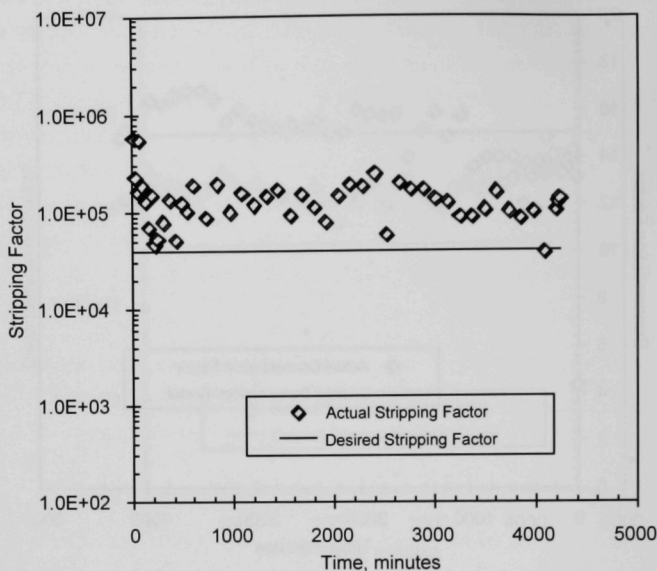
Fig. 7. Changes in the Stripping Factor vs. Time for Test CS27

One of the feed pumps had to be replaced during test CS27. This event occurred at 956 min (15.9 h) into the test. The test was stopped for about one hour to replace the DF transfer pump. This pump transferred the feed from the DF feed drum outside the glovebox into the DF feed tank inside the glovebox. After the pump was replaced, a new feed line was installed from the pump into the DF feed drum, and the test was restarted. The location of the pump is given in more detail in Appendix C. As Figs. 5-7 show, this shutdown and restart had no effect on the key process parameters.

Solids became a problem late in the test. The solids were not formed in the contactor. Rather, they were precipitated from the SRS simulant over time. In future tests, the feed will be filtered just before it enters the contactor. This solids problem was noticed at 4080 min (68.0 h) into the test. At that time, the EW effluent was found to be slightly alkaline, with a pH of 9. The Cs concentration in the solvent coming out in the EP effluent was unchanged. The DS feed rate was doubled in an effort to reduce the pH of the EW effluent, but it did not work. About one hour later, a solvent (EP) effluent sample had a Cs concentration about half that of the DF feed. At this point, 4160 min (69.3 h) into the test, the test was stopped for a second time. Sufficient solids were found in the rotor at stage 15 (where the DF feed enters the contactor extraction

section) to partially plug the rotor at that stage. These solids restricted the flow of the SRS simulant through the rotor to the upper weir. As a result, some of the simulant followed the organic phase into the scrub section. The aqueous liquid in the first scrub stage (stage 16) had about the same yellow color as the SRS simulant in stage 15, indicating more simulant than scrub solution in that stage. The aqueous liquid in the second scrub stage (stage 17) had about half the yellow color of the SRS simulant in stage 15. There were some solids in both scrub stages. There were more solids in extraction stages 13 and 14 than in the scrub stages, but not as much as in the feed stage, stage 15. The solids were found on the inside wall of the rotors, but not in the annular mixing zone that is outside of the rotor.

Four steps were required to solve the problems caused by the solids. First, the solids were dissolved from the rotors by dipping each rotor containing solids into a beaker filled with 2 $\underline{M}$ HNO_3. Second, the Cs concentration in the strip section was decreased by running the process without the DF feed for 5 h. Third, when the Cs concentration in the solvent from the strip section had a stripping factor of 40,000, the exiting solvent was collected in a second solvent tank. The first solvent tank was emptied by continuing to feed it to stage 33. When this solvent was gone, the solvent feed to stage 33 was switched to the clean solvent in the second solvent tank. Finally, when the Cs concentration in the DW effluent was sufficiently low so that the decontamination factor was 40,000, the test was resumed. The remaining feed was processed with appropriate values for the decontamination and concentration factors. The test was over at 4233 min (70.6 h) when all of the 180 L of SRS simulant waste had been processed.

Thus, test CS27 met both process goals; that is, a decontamination factor >40,000 and a concentration factor of 15 were maintained, while recycling the solvent 42 times over a test time of 71 h. No evidence of degradation in solvent performance was observed based on the relatively constant values for the decontamination and stripping factors over the entire 71 h of the test. Subsequent analysis of the solvent at ORNL using high-performance gel permeation chromatography and reverse-phase high-performance liquid chromatography also showed no qualitative differences between the pristine solvent and the solvent left after the multi-day test, that is, no degradation products were observed [MASKARINEC-2002]. Results from test CS27 were used to guide and evaluate the test with real waste at SRTC [CAMPBELL-2001A].

While the ORNL evaluation of the solvent left after the multi-day test showed no qualitative changes, it did show some quantitative changes [MASKARINEC-2002]. There was a 10% loss of TOA. Most of this loss could be attributed to the formation of dioctylamine. While there was a trace of dioctylamine in the pristine solvent, its concentration was six times higher in the solvent left after the multi-day test. In addition, there was a 20% increase in the concentration

of BOBCalixC6 and a 32% increase in the concentration of Cs-7SB. These increases suggest that there was a 20 to 32% loss of diluent during the multi-day test. A gas chromatographic analysis of the pristine and used solvents showed the Isopar®L diluent to be unchanged in its composition during the test, either qualitatively or quantitatively [MASKARINEC-2002]. This diluent loss was confirmed by measurements of the solvent density at ANL. The pristine solvent had a density of 822 g/L at 27°C. A composite solvent volume consisting of six EP effluent samples taken near the end of the multi-day test (from 3600 to 4200 min) had a density of 857 g/L at 20°C.

A second ORNL evaluation of the solvent left after the multi-day test showed some quantitative changes in the distribution ratio for cesium [SLOOP-2001]. F. V. Sloop did an extraction-scrub-strip (ESS) test at 25°C. The one batch extraction was done at an O/A ratio of 0.33 using the solvent and the SRS simulant. The one batch scrub was done at an O/A ratio of 5 using the solvent from the batch extraction and 0.05 $\underline{M}$ HNO$_3$. The three batch strips were done at an O/A ratio of 5 using the solvent from the batch scrub and 0.001 $\underline{M}$ HNO$_3$. Average results for the pristine solvent are compared with used solvent from test CS27 in Table 4. The increase in the cesium distribution ratio for the used solvent is from 31 to 69%. Again, these increases in D_{Cs} suggest that there was a loss of diluent during the multi-day test.

Table 4. Cesium Distribution Ratios for Pristine and Used CSSX Solvent[a]

	Extraction	Scrub	Strip #1	Strip #2	Strip #3
D_{Cs} (pristine solvent)	16.9	1.59	0.148	0.089	0.067
D_{Cs} (used solvent)	22.1	2.45	0.250	0.139	0.113
D_{Cs} (pristine)/ D_{Cs} (used)	1.31	1.54	1.69	1.56	1.69

[a]Data from ORNL, see [SLOOP-2001].

Additional results from the multi-day test are given in Appendix D. This includes flow rates, temperatures, pH values for the EW and FW effluents, interstage line appearance, the cesium concentrations in the effluent streams, and the cesium concentrations in both phases for each stage at the end of the test. The cesium concentration profile is compared with a computer model to determine stage efficiency. The calculations give an average stage efficiency of 89% for the extraction section and 83% for the strip section. These mass transfer efficiencies for multistage operation are similar to the 89 to 92% seen in earlier CSSX tests [LEONARD-2001A].

VII. DISCUSSION

The approach of the CSSX process to steady state can be followed by looking at the data for the concentration factor. The rate at which this factor approaches steady state is controlled by the rate at which the DF feed brings cesium into the contactor. The concentration factor was 4.4 at 5 min (0.08 h) into the test and 13.1 at 25 min (0.42 h). At 4160 min (69.3 h) into the test, when the contactor was being restarted after all of the Cs had been flushed out, the concentration factor reached 12.3 at 18 min (0.30 h) into the restart. At 38 min (0.63 h), the concentration factor was 15.2. Thus, the CSSX process run in centrifugal contactors reaches steady state in 25 to 30 min (0.42 to 0.50 h).

When calculating the average concentration factor and the average stripping factor in the Results section above, some data points were omitted; however, no data points were omitted when calculating the average decontamination factor. For the average concentration factor, the two low data points that occurred during the two start-up periods were omitted. Also omitted was the low concentration factor at 4080 min, which was probably caused by the solids accumulation in stage 15. These three concentration factors are included in Fig. 6. For the average stripping factor, the low stripping factor at 4080 min was omitted for the same reason. The data points for the first hour were also omitted from the average stripping factor, as they were all higher than the average. During start-up, the stripping factor will be greater than the average value, so this indicates the stripping section took more than one hour to reach steady state. This is expected, because, after one hour, there were only 1.4 residence times with respect to the aqueous phase in the strip section. This residence time was calculated using the aqueous strip flow rate of 2.85 mL/min, the aqueous-phase volume of 8 mL/stage in the strip section, and the 15 strip stages. While these stripping factors were omitted from the average value, they are included in Fig. 7.

In the Results section above, it was noted that solids were a problem. These solids, which entered with the DF feed at stage 15, had precipitated from the SRS simulant in the 10 to 12 days between the time when the simulant was filtered and the test started. Since the DF inlet line went to the bottom of the DF feed drum, solids would have been picked up around the feed point at time zero. When the transfer pump for the DF feed drum to the glovebox failed at 956 min, a new feed pickup line was put in the drum and would have picked up solids at a second point on the bottom. The addition of 22 L of DF feed to the 13 L of DF feed left in the DF feed drum was started at 3459 min and continued for 120 min. This would have stirred up more solids and suspended them in the DF feed in the drum. Thus, there were three inadvertent transfers of solids into the DF feed going to the contactor. After the test, solids were found in the DF feed tank in

the glovebox and in the line from the DF feed drum to the DF transfer pump. Because of this inadvertent entrainment of solids, enough particles eventually reached the contactor to cause the aqueous underflow in the rotor at stage 15 to become partially blocked. This forced some of the aqueous phase (SRS simulant) over the organic weir of the rotor along with the solvent into stage 16, the first scrub stage. This highly alkaline solution in the scrub section overwhelmed the dilute nitric acid and eventually made the first strip stage, stage 18, alkaline as well. The excess nitrate entering the strip section kept the cesium from being stripped from the solvent. Thus, solvent with a high Cs concentration was able to move through the strip section and gave a high Cs concentration in the organic strip (EP) effluent. This was the reason the run was stopped. At this point, the solids were discovered in the contactor. As stated above, the solids were dissolved out of the rotors, the excess cesium was cleared from the strip and extraction sections, and normal process operations were resumed. This test demonstrates the importance of (1) keeping solids out of the contactor; (2) monitoring the pH in the scrub section or the aqueous strip (EW) effluent or both, which allows early detection if solids force the aqueous phase out of the waste (DF) feed stage along with the organic phase; and (3) having a plan for responding to solids in the DF feed.

An elemental analysis of the solids showed the presence of sodium, aluminum, and sulfur. X-ray diffraction (XRD) analysis indicated the solids were crystalline and contained many components. The main components identified were $NaNO_3$ (~75%), sodium aluminosilicate (also known as sodium montmorillonite, a clay, at ~13%), Na_2SO_4 (~4 to 8%), and $Al_2(SO_4)_3 \cdot 5H_2O$ (~4%). Other possible components were NaCl and $NaAl_3(SO_4)_2(OH)_6$ (also known as natroalunite or alunite). In the sodium-containing components, K may sometimes replace the Na. Several weeks after the test, the residual liquid in the DF feed tank was filtered and these solids were also analyzed using XRD. They were found to have the same major components identified from the solids in the rotor as well as traces of two additional ones, $Na_9(AlO_2)_9(SiO_2)_{15} \cdot 27H_2O$ and $Na[AlO_4Al_{12}(OH)_{24}(H_2O)_{12}](SO_4)_2 \cdot 10H_2O$. These new solids may have precipitated during the four weeks that the residual feed solution sat in the DF feed tank after the 5-day test. Thus, it appears that the solids in the contactor rotors formed before the waste (DF) feed reached the contactor.

The increases in extractant and modifier concentrations and the increases in the cesium distribution ratios for the final solvent indicate a loss of diluent, which should increase the solvent density. A simple density model was developed to predict the increase in extractant and modifier concentrations from an increase in solvent density. The model assumes there is no interaction between solvent components that would affect their density. In this model, BOBCalixC6 has a molecular weight of 1149.53 g/mol and a density of 1054 g/L at 25°C; Cs-7SB has a molecular

weight of 338.35 g/mol and a density of 1173.5 g/L at 25°C; TOA has a molecular weight of 353.69 g/mol and a density of 809 g/L at 25°C; and Isopar®L has a density of 759.7 g/L at 25°C. Finally, the effect of temperature on density for all components is -0.77 (g/L)/°C in the neighborhood of 25°C. With this model, the density of fresh CSSX solvent at 25°C is calculated to be 823 ± 2 g/L. For a CSSX solvent density of 857 g/L at 20°C near the end of test CS27 but prior to the solids problem, the concentration factor is 1.492. This is higher than the concentration factor of 1.20 to 1.32 observed by [MASKARINEC-2002] analyzing the final solvent. In recovering from the process upset caused by the solids, some solvent was lost. To complete the test, some fresh solvent was added. Thus, as was seen experimentally, the solvent at the end of the test should be less dense than the solvent collected shortly before. If the extraction and modifier concentrations increased 20%, the solvent density would be 835 g/L at 25°C; if 32%, then 843 g/L at 25°C.

All of the factors considered when improving hydraulic performance in the 2-cm contactor are important. As most TIR values were already within specifications and the motors were mostly turning at 3600 rpm, these factors did not have much effect on the further improvement of hydraulic performance. The rotor change (increase in the diameter of the upper weir) to increase the maximum throughput in the extraction section of the CSSX process had some improvement on hydraulic performance. It reduced the possibility that positive feedback would occur when the aqueous flow rate fluctuated to higher flow rates. The most important change was the improved inlet to the mixing zone from the interstage lines, that is, the use of an insert in the perpendicular inlet ports. This change to the multistage 2-cm contactor gave the long-term stable operation needed for the multi-day test. When building 2-cm contactors and larger units in the future, the interstage inlet to the mixing zone should be tangential to the spinning rotor so that the incoming liquid is drawn into the mixing zone by the rotor.

In the test done here, the solvent was recycled every 1.65 h. In a process plant with 3785 L (1000 gal) of solvent and a solvent flow rate of 25.0 L/min (6.6 gpm), the solvent will be recycled every 2.53 h. Thus, the 42 solvent cycles after 2.94 d of operation in the minicontactor test reported here are equivalent to the solvent recycle for 4.49 d of plant operation.

VIII. SUMMARY AND CONCLUSIONS

Two methods for temperature control in the extraction section of the CSSX flowsheet were considered, thermoelectric cooler modules and chiller bars. It was determined that either device would work. Chiller bars were chosen because they were easier to install in the glovebox, easier to remove, and less likely to be damaged by other glovebox operations. The bars were cooled in series by water recirculating from a chiller bath.

Hydraulic performance of the 2-cm contactor was improved in four ways. The first improvement involved opening the upper-weir diameter of each rotor so that it would allow a higher throughput in the extraction section. This prevented the occurrence of a positive feedback loop where small amounts of aqueous phase start flowing with the organic phase in the organic interstage. While the first improvement was specific to the 2-cm contactor rotor being used, it is based on the general rule that no rotor should be operated at more than 50 to 67% of its maximum throughput in multistage operation. If this more general rule is followed, positive feedback loops will not occur.

The second way in which the hydraulic performance was improved was to tighten the specifications on the total indicating runout at the bottom of the rotor. The operation of the 2-cm contactor was good for rotor TIR values up to 10 mils (0.254 mm). Initially, allowable TIR values had been 12 mils (0.305 mm). If the 2-cm contactor housing were built with inlet ports tangential to the rotor in the mixing-zone cavity, as discussed next, this greater limit for the TIR value might be acceptable. For larger contactor rotors, some increase in the maximum allowable TIR may be possible. This is an area where careful attention will be needed as larger contactors are built and used.

The third way in which the hydraulic performance was improved was by ensuring that the spinning portion of the motor/rotor assembly was not rubbing against any object that might slow it down. This was done by first measuring the time that it takes for the motor itself to spindown. The spindown time was then checked after each step in the assembly of the motor/rotor system to see if it had changed. This allowed us to determine that the spinning parts were not rubbing on the motor mount plate, the vanes below the rotor, the plastic inserts, or the wire ropes.

The fourth way in which the hydraulic performance was improved was to change the opening where the flowing interstage liquid enters the mixing zone so that it is protected. This allows interstage liquid to enter without being hindered by the swirling liquid already in the mixing zone. For the 2-cm contactor used here, the inlet tube is perpendicular to the annular mixing zone. To improve this setup, a protective space was created by the use of an insert that extended from the inlet port halfway into the annular mixing zone. The insert was oriented so that it shielded the entering liquid from the swirling liquid already in the annulus. This improvement is applicable to all contactors. In new contactors, inlet ports should be tangential to the annulus so that the swirling liquid in the mixing zone will carry the incoming liquid into the annular mixing zone. A tangential inlet port should eliminate the need for an insert.

Operator selection and training were very important for the three-shift operation. On each shift there was a need for people who (1) understood the overall process, (2) knew what data had to be taken, (3) understood the equipment, (4) could fix the equipment, and (5) could measure the cesium concentrations. Classroom training gave every operator some knowledge in each area. This was supplemented by laboratory training in which the CSSX flowsheet was run cold, that is, with no radioactive cesium. The laboratory training was found to be very important. It could not have been replaced with more classroom training. The most important skill was being able to shut down and restart the CSSX process. With this skill, the team was prepared to face any emergency.

With the new temperature control system, the changes to improve hydraulic performance, and the operators trained, the CSSX process to remove cesium from SRS high-level waste was tested for three days in a 33-stage 2-cm centrifugal contactor. This multi-day demonstration of CSSX flowsheet was done using an average SRS simulant for the waste feed. The two key process goals were achieved and maintained: (1) the cesium was removed from the waste with decontamination factors greater than 40,000 and (2) the recovered cesium was concentrated by a factor of 15 in dilute nitric acid. In the multi-day test, 1.4 L of solvent was recycled a total of 42 times while processing 180 L of SRS simulant. The cesium had an average decontamination factor of 157,000 and an average concentration factor of 14.9. In these tests, the average stage efficiency was 89% for the extraction section and 83% for the strip section, which were very close to the mass transfer efficiencies seen in earlier CSSX tests [LEONARD-2001A].

During the multi-day CSSX test, the contactor had to be shut down twice. Both times it was restarted easily. The first time, a feed pump had to be replaced. This did not cause any change in the process, which was restarted at essentially steady-state conditions. The second time, with 1 h and 20 min remaining until the end of the test, solids from the SRS simulant partially plugged the rotor where the simulant was being fed. This forced some of the alkaline aqueous phase into the scrub section and made it alkaline. This, in turn, increased the nitrate concentration in the first strip stage and prevented the strip section from removing the cesium from the solvent. After the solids were dissolved out of the contactor rotors and the high cesium concentration was cleared from the strip section, the solvent recycle tank, and the extraction section, the test was restarted and continued until the SRS simulant was processed. In the three samples taken after recovering from the solids problem, the system was found to have the same high decontamination factor as before. This quick recovery from emergency shutdowns gives a clear indication of the robustness of the CSSX process when run in a centrifugal contactor.

This multi-day test showed the ability of the solvent to be used for at least 42 solvent recycles, the ability to recover from process upsets, and the importance of keeping solids out of the contactor. After 42 solvent cycles, there was no indication that the solvent had lost any of its ability to remove and concentrate cesium. Based on the success of this test, SRTC did a two-day test with real waste from the SRS tanks, which also met the CSSX process goals [CAMPBELL-2001A].

ACKNOWLEDGMENTS

APPENDIX A

FURTHER INFORMATION ON TEMPERATURE CONTROL

Further information on temperature control for the 2-cm contactor is given here on (1) the heat-up of a contactor stage by the rotor motor, (2) setup and operation of the TEC module, and (3) the setup and operation of the chiller bars.

1. Heat-up of Contactor Stage

An analysis of the heat-up in a contactor stage indicates that most of the heat comes from the rotor motor [ARAFAT-2001]. A simple test was done to evaluate the effect of the rotor motor on stage temperature in the 2-cm contactor used for testing the CSSX flowsheet. The motor of one contactor stage was turned on with no liquid flowing into the stage. Using calibrated digital thermometers that were accurate to ±1.0°C, the temperature was measured at four locations on the external surfaces of the contactor stage as a function of time. During this test, the average ambient room temperature was 25°C. Time zero was when the motor was turned on. The results, given in Table A-1, show how the temperature increases with time.

Table A-1. Temperature Measurements on Contactor Housing

Motor Operation Time, min	Temperature of Rotor Motor,[a] °C	Temperature of Motor Mount Plate,[b] °C	Temperature of Contactor (Front),[c] °C	Temperature of Contactor (Side),[d] °C
0	25.1	25	24.9	24.9
18	29.6	27.3	26.2	25.7
41	32.4	29.6	28.5	28.0
65	32.9	29.4	30.5	28.3
110	34.4	30.3	31.5	28.7
190	33.4	31.0	31.8	30.5
230	34.2	32.0	30.8	29.5

[a] The digital thermometer probe was placed at the back of the motor. Since the motor was point down, this was the top of the motor. The probe was located in the space between the back of the motor and the attached junction box.
[b] The motor mount plate is a 0.25-in thick plate that is between the motor and the contactor block. The digital thermometer probe was placed on one corner of the top surface of this plate.
[c] The upper stainless-steel block of the contactor housing is 3 in wide by 3 in deep with a height of 2.5 in. It contains the collector rings, the inlet and outlet ports, and the upper region of the annular mixing zone. The digital thermometer probe was placed in the middle of the front face.
[d] The digital thermometer probe was placed in the middle of the side face of the upper stainless-steel block of the contactor housing.

Two other tests, which were run in multistage 2-cm contactors for the same or longer times, resulted in even higher temperatures. In one test, a 4-stage 2-cm contactor was operated

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without any liquid. After 16 h, the temperature on the front face of the upper contactor body was found to be 39.5°C. In the other test, a 32-stage 2-cm contactor test (CS23) [LEONARD-2001A], the DW raffinate from the extraction section had reached 36.9°C after operating the CSSX flowsheet for several hours in a glovebox with a temperature of 25°C. No attempt was made to cool either the contactor stages or the feed. The motors had been on for 3.5 h at the time of the temperature reading. Liquid had been flowing through the 15 extraction stages for the last 3.3 h. The actual CSSX flowsheet test had been the last 1.8 h. Thus, in a laboratory at 25°C without any cooling of the feed or the 2-cm contactor, the contactor housing and the liquids inside can reach temperatures between 36 and 40°C after several hours of operation. These temperatures, which are higher than those for single-stage operation shown in Table A-1, suggest that the contactor temperature continues to rise slowly even after three to four hours of operation.

Based on these results, it was decided that, for the multi-day test, each contactor stage in the extraction section should be cooled. The cooling device should be able to remove more heat from each stage than the rotor motor can generate. The devices considered were TEC modules and chiller bars.

2. Thermoelectric Cooler Modules

Because of their widespread use in cooling central processing units of personal computers, TEC modules are widely available at reasonable prices in a variety of shapes and cooling capacities. The TEC modules use the Peltier effect. This effect gives rise to a temperature change when an electrical current passes through the junction of two dissimilar metals or semiconductor materials. The rate at which heat is pumped is proportional to the electrical current flow through the junction. Using this principle, a TEC module consists of a number of tiny semiconductor couples connected electronically in series and sandwiched between two ceramic plates. When connected to a DC power source, the flow of electrical current causes heat to move from one side of the TEC device to the other. This creates hot and cold faces on the TEC device. By controlling the direction of the flow of electrical current to the TEC device, it can be made to either cool or heat a piece of equipment. In our case, since we want to cool the extraction section, the cold face contacts the contactor housing while the hot face contacts a heat sink that has a fan on the other side. The fan blows air over the heat sink helping to move the heat to the surrounding air.

A commercially available TEC module, heat sink, and fan were used in this study. They are shown in Fig. A-1 in the upper left, lower left, and lower right corners, respectively. A ruler with 1-mm markings is shown along the top. The TEC module is catalog no. 6301/071/030AS from Ferrotec, Nashua, NH. It is square with 22-mm sides and a thickness of 3.6 mm. The

square face of the module can be placed on the upper portion of the upper contactor housing. To protect its internals from moisture, the module is sealed with an aerospace-grade epoxy. At its maximum rating of 3 A at 9.8 V, the module will provide 16 W of cooling. To improve heat transfer between the module and the contactor, a Ferrotec thermal compound, a zinc oxide paste, was used. Its catalog no. is 111-01ZS-FE. The heat sink is the OptiPin Heat Sink, catalog no. 371924B00032, from Arrow Electronics, Itasca, IL. It is square with 35-mm sides and a thickness of 14 mm. The face that touches the TEC module has a two-sided pre-applied thermal tape. The DC fan is catalog no. 412F from Resource Electronics, Chicago, IL. It is square with 40-mm sides and a thickness of 10 mm. The rated airflow for the fan is 15 cfm. While the fan is normally bolted to the housing, a strap (a cotton string) was used to hold the fan in place over the heat sink and the TEC module. This strap also held the entire assembly against the contactor housing.
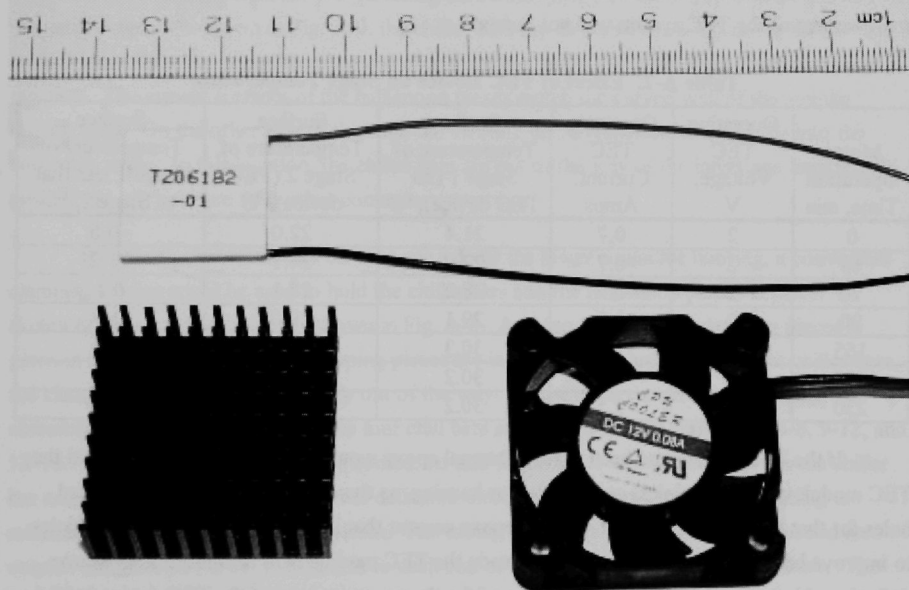
Fig. A-1. TEC Module, Heat Sink, and Fan Used in TEC Device Test

This TEC device was tested as follows. The motors of two adjacent 2-cm contactor stages were started. One stage had a TEC device attached to it, while the other one did not. A digital thermometer was attached to the side of the upper contactor housing opposite the TEC

device. A second digital thermometer was attached to the same point on the other stage with no TEC device. These two surface temperatures were measured and compared as a function of time, starting at time zero when the motors were turned on. At time zero, a DC voltage was applied across the TEC device and the heat-sink fan was started. The results, given in Table A-2, show that the TEC device does very little cooling at 25% of its rated voltage and current. At 50% of its rated voltage and current, the surface temperature of the cooled stage was 2.5°C cooler than the non-cooled stage. Because of the limits of our power supply, the test was stopped here. However, with a better power supply, even further temperature differences would have been seen. The TEC device should be able to provide the cooling needed, as it can remove more heat than the rotor motor generates. Also, slightly larger TEC devices with even greater cooling capacities are available. In addition, better contact between the TEC device and the contactor housing, for example, using thermal epoxy, will enhance heat transfer and provide better cooling. Since, as discussed in Section III of this report, the chiller-bar concept was rated superior in overall design, improvement of the TEC system was not pursued.

Table A-2. Effect of TEC Device on Stage Temperature

Motor Operation Time, min	Operating TEC Voltage, V	Operating TEC Current, Amps	Surface Temperature of Stage 1 (no TEC device), °C	Surface Temperature of Stage 2 (TEC device), °C	Surface Temperature of Stage 1 less that of Stage 2, °C
0	2	0.7	21.4	22.0	−0.6
18	2	0.7	22.9	24.1	−1.2
70	2	0.7	28.2	28.1	0.1
90	2	0.7	29.1	29.0	0.1
155	4	1.5	30.3	28.3	2.0
200	4	1.5	30.2	27.8	2.4
230	4	1.5	30.2	27.7	2.5

If the TEC system had been used, a thermal epoxy would have been required to hold the TEC module onto the stainless-steel contactor housing, as there were no drilled and threaded holes for that purpose. A thermal epoxy is epoxy cement that is loaded with aluminum particles to improve heat transfer. This would have made the TEC module hard to replace if it became defective. Note that the combined thickness of the three components of the TEC device is 27.6 mm. Since it is mounted on the upper contactor housing, it would be in the way and easy to damage.

While the TEC system was not chosen here, it does have features that would make it very attractive in systems that are properly designed from the ground up. First, the TEC module is very responsive. When holding the TEC module between two bare fingers and applying 2 V

across the wires to the module, it quickly becomes both too hot and too cold to hold. Second, the TEC system can be changed from cooling to heating by reversing the flow of current to the TEC module. Third, no liquid flow is required for cooling or heating. Finally, a different amount of cooling or heating can be applied to each module or stage.

3. Chiller Bar

The chiller bars used to cool the extraction section were attached to the contactor housing as shown in Fig. 3. This design is a modification of one developed at SRTC under the leadership of R. A. Peterson [PETERSON-2000B]. In the SRTC design, the chiller bars and half-moon pieces were placed on both sides of each stage in the extraction section. Thus, the design used here has about half the cooling capacity of the SRTC design. The chiller bar is a 3/4-in. square pipe with a 1/32-in. wall. Each chiller bar is 12-in. long so that it can cover 4 contactor stages. At each end of the bar, there are 3/8-in. tubing barbs with pipe threads that screw into the side of the square pipe. As shown in Fig. A-2, the chiller bars slip in below the 3-in. square blocks (2.5 in. high) that form the upper housings and above the box beam that holds each contactor stage in position. The curved surfaces of the half-moon pieces match the curved wall of the annular mixing zone. On the other side of the wall, the swirling liquid enhances heat transfer into the contactor stage. In this location, the chiller bars are out of the way of the interstage lines and, in general, do not interfere with other contactor operations.

By putting the chiller bars on only one side of the lower contactor housing, a convenient clamping system could be used to hold the chiller bars and the half-moon pieces in place. A sketch of the clamping system is shown in Fig. A-3. A photo showing the clamping pieces is given in Fig. A-4. Because the clamping pieces are in the recessed area opposite the chiller bars, the clamp system is also conveniently out of the way. At the same time the whole chiller bar assembly can be removed easily. The four chill bars are located next to stages 1-4, 5-8, 9-12, and 13-16. Since stage 16 was not to be cooled, no half-moon piece was placed between the chiller bar and stage 16. The cooling water is delivered through fairly large flexible PVC tubing to minimize the pressure drop in the system. The tubing has an inside diameter of 3/8 in. and an outside diameter of 1/2 in. It connects to each end of the square pipe via the tubing barb. The PVC tubing was held onto the tubing barb with a stainless-steel hose clamp. To prevent tight turns of the PVC tubing, the connection order for the chiller bars was as follows: The water was pumped from the circulating bath through the glovebox wall to the chiller bar beside stages 1 to 4, stages 8 to 5, stages 9 to 12, and stages 16 to 12. From stage 12, the cooling water returned through the glovebox wall to the circulating bath.
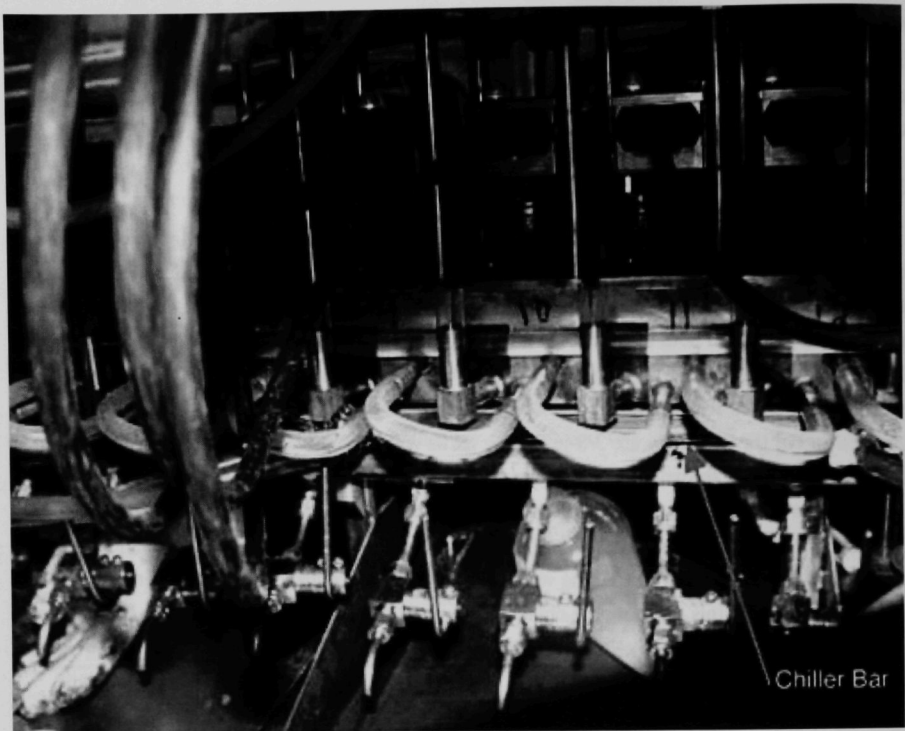
Fig. A-2. Location of Chiller Bars on 2-cm Contactor
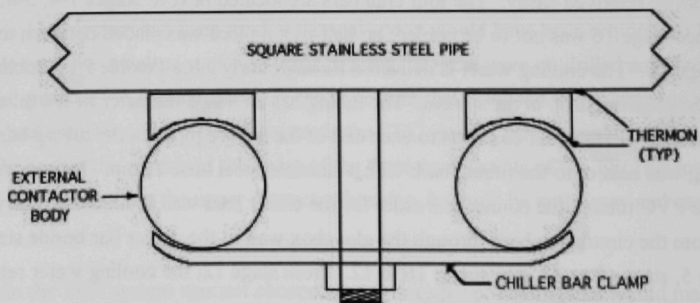
Fig. A-3. Schematic Showing Chiller Bars Clamped onto the Lower 2-cm Contactor Housing

Chiller Bar Clamp

Fig. A-4. Clamps for Chiller Bars

The chiller-bar cooling system was tested as follows. One chiller bar was set up on a 4-stage 2-cm contactor. The chiller water flow was set at 15 L/min and the chiller water bath was set at a fixed temperature. Next, the rotor motors were turned on and process water was pumped through four contactor stages at 53 mL/min. At this flow rate the contactor water has the same total heat capacity as the waste/solvent mixture in the extraction section of CSSX test CS27. This is because the solvent has half the heat capacity of the aqueous waste. The system was allowed to run until the recycled process water flowing through the contactor had reached a steady-state temperature. In this way, it was hoped to model the operation of many contactor stages for conditions where the temperature would not increase from chiller bar to chiller bar. For a room temperature of 21.6 ± 0.3°C, the results in Table A-3 show that as the chiller-bath temperature goes from 5 to 15°C, the steady-state temperature of the process liquid flowing through the contactor goes from 22 to 26°C. As a rough rule of thumb, one can say that the temperature of the process liquid flowing through the contactor will decrease 2°C for every 5°C decrease in chiller-bath water temperature. If more cooling is necessary, ethylene glycol can be

added to the cooling water and the chiller bath can be set at temperatures below 0°C. This was not required for the tests reported here.

Table A-3. Effect of Chiller Bath Temperature on the Process Liquid Temperature in the 2-cm Contactor

Water Temperature in Chiller Bath, °C	Inlet Process Water Temperature to Contactor at Steady-State Conditions, °C	Outlet Process Water Temperature from Contactor at Steady-State Conditions, °C	Room Temperature, °C
5	21.9	22.0	21.8
10	23.1	23.5	21.3
15	25.7	26.1	21.9

Based on these results, it was decided to operate the chiller system using cooling water at a temperature of 7.5°C during test CS27. If necessary, the temperature could be changed. This was not needed. Because the chiller water flows into and out of the glovebox during the test, the chiller water lines inside the glovebox were not opened. This keeps the chiller water from becoming contaminated with radioactive materials.

APPENDIX B

FURTHER INFORMATION ON HYDRAULIC PERFORMANCE

Further information on the hydraulic performance of the 2-cm contactor is given here for (1) one-phase flow tests, (2) two-phase flow tests for the extraction and strip sections, (3) hydraulic evaluation of the solvent wash section, and (4) the effect of insert orientation. The one-phase flow test results were evaluated using a model that takes into account the height of the liquid over each weir based on flow rate and liquid density [DAVIS-1961]. The two-phase flow test results were evaluated using the same model combined with the thickness of the dispersion band in the rotor as given by the dispersion number [LEONARD-1981]. To allow the establishment of steady-state conditions, the flows were maintained for at least three residence times before samples were taken. Both models give an apparent radius for the more-dense-phase weir that can be compared with the measured (actual) radius for the more-dense-phase weir.

1. One-Phase Flow Tests

To modify the 2-cm contactor rotors so that the maximum throughput is increased, each more-dense-phase weir (upper weir) was opened using electrical discharge machining (EDM). Before machining, the upper-weir radius was 6.12 mm; after machining, 6.24 mm. After each weir was machined, its diameter was measured to see that it met the new specification to within ± 0.03 mm. Then, the effect of this change on hydraulic performance was determined using one-phase flow tests to measure the zero-point flow rate for the rotor. The one-phase zero-point flow rate is defined as the point where, as the flow is increased, liquid starts to come out the less-dense-phase exit. This flow rate is called the "zero-point flow rate" or the "zero point." For these tests, water is used for the liquid phase and flow rates are measured both above and below the zero point. Typical data, plotted in Fig. B-1, are for a 2-cm contactor with an upper-weir radius of 6.24 mm. The data are interpolated to determine the zero-point flow rate (95.2 mL/min). Then the one-phase flow model is used to calculate the apparent radius for the more-dense-phase weir (6.16 mm) based on this zero-point flow rate. Finally, the apparent radius is compared with the actual radius and found to be slightly less.
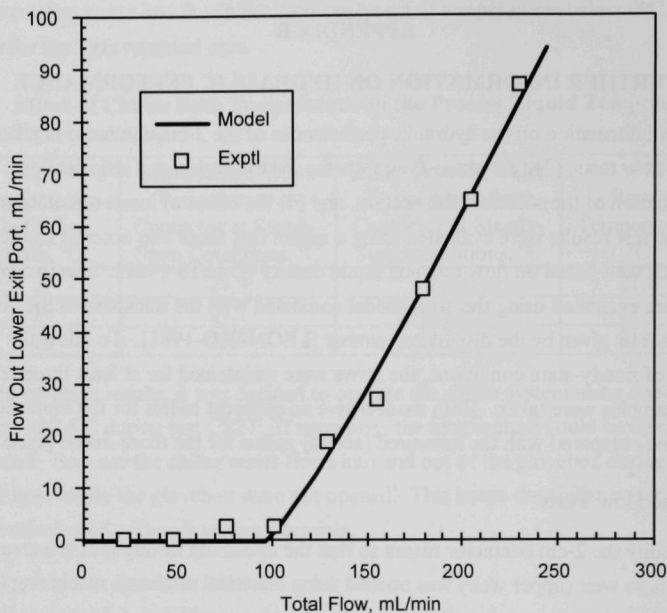
Fig. B-1. Typical Zero-Point Data

For the two-phase hydraulic performance tests in the 1- and 4-stage 2-cm contactor, rotors B-1 through B-4 and X-1 through X-4 were used. Before the two-phase tests, the zero point of each rotor was determined. After the upper-weir radii of the X rotors were enlarged from 6.12 to 6.24 mm, the zero point was measured again. The results, given in Table B-1, show that, with one exception, the apparent radii are slightly less than the actual radii. The results in Table B-1 are summarized in Table B-2 along with the results from three other sets of 2-cm rotors. In all cases, the apparent radius for the upper weir is less than the measured radius. This difference indicates that the model only approximates the actual flow conditions. For the rotors whose radii are 6.12 mm, the weirs were machined and then welded in place. The apparent radii for these weirs are 0.14 ± 0.04 mm less than the actual radius. When these machined radii are enlarged to 6.24 mm using EDM, the difference between the measured and apparent radii is less, 0.07 ± 0.03 mm. Thus, the EDM process seems to make the measured weir performance closer to that predicted by the model.

Table B-1. Results of Zero-Point Tests for Several 2-cm Rotors

Rotor ID	Measured Upper-Weir Radius, mm	Apparent Upper-Weir Radius, mm	Zero-Point Flow Rate, mL/min	Liquid Volume in Mixing Zone, mL	Liquid Volume Inside the Rotor, mL
1B	6.12	5.97	37.4	4.7	5.5
2B	6.12	5.99	45.2	5.8	4.8
3B	6.12	6.07	68.7	4.4	5.6
4B	6.12	6.06	64.8	4.4	5.6
Average		**6.02**	**54.0**	**4.8**	**5.4**
1X	6.12	5.93	25.7	5.0	6.0
2X	6.12	6.05	60.9	6.1	6.1
3X	6.12	5.96	33.4	5.0	6.0
4X	6.12	5.92	22.0	5.0	6.6
Average		**5.97**	**35.5**	**5.3**	**6.2**
1X	6.24	6.16	95.2	4.0	4.5
2X	6.24	6.19	102.5	3.9	7.1
3X	6.24	6.24	106.1	5.4	6.2
4X	6.24	6.22	113.2	5.5	6.6
Average	**6.24**	**6.20**	**104.3**	**4.7**	**6.1**

Table B-2. Summary of Zero-Point Results for 2-cm Rotors

Rotor Set	Rotors Tested	Measured Upper-Weir Radius, mm	Apparent Upper-Weir Radius, mm	Zero-Point Flow Rate, mL/min	Liquid Volume in Mixing Zone, mL	Liquid Volume inside the Rotor, mL	Notes
1	4	6.12	5.97 ± 0.06	36 ± 18	5.3 ± 0.5	6.2 ± 0.3	X rotors, built before 2000
2	4	6.12	6.02 ± 0.05	54 ± 15	4.8 ± 0.7	5.4 ± 0.4	B rotors, built before 2000
3	36	6.12	5.95 ± 0.04	33 ± 11	5.7 ± 0.2	4.5 ± 0.8	Built 2000
4	4	6.24	6.20 ± 0.03	104 ± 7	4.7 ± 0.9	6.1 ± 1.1	Rotor set 1 after upper-weir radius enlarged
5	37	6.24	6.15 ± 0.05	91 ± 14	4.7 ± 1.2	5.0 ± 0.4	Built before 2000
6	40	6.24	6.15 ± 0.03	91 ± 8	5.9 ± 0.6	4.4 ± 1.1	Built 2000/2001

After each zero-point test was made, the water flowing to the rotor was stopped while the rotor itself was kept running. After water had stopped flowing out of the contactor stage, the

liquid in the mixing zone was drained and its volume measured. Then the rotor was turned off, the liquid inside the rotor drained, and its volume measured. These two volumes, liquid in the mixing zone and liquid inside the rotor, respectively, are also given in Tables B-1 and B-2. They show that, under no-flow conditions, the liquid volume in each stage of a 2-cm contactor is 10.4 ± 0.1 mL.

2. Two-Phase Flow Tests for the Extraction and Strip Sections

The upper weir was opened to increase the maximum throughput of the 2-cm contactor for the extraction section of the CSSX process. Before machining, the upper-weir radius was 6.12 mm; after machining, it was 6.24 mm. In previous 2-cm contactor work at ANL, such as the proof-of-concept tests [LEONARD-2000], the upper-weir radius had been 6.12 mm. In this section, results are reported on how performance was improved by increasing the upper-weir radius to 6.24 mm. Most of the tests focused on the extraction section, since this section had the maximum throughput. The strip section was evaluated to be sure that it had not been adversely affected by the modification. The scrub section was not checked, as it is like the strip section, only at a slightly higher acid concentration. The solvent wash section, which is new, was evaluated separately. Tests of the solvent wash stage are discussed in Section 3.

a. Single-Stage Operation

To evaluate the hydraulic performance of the CSSX solvent in the various sections of the process, two-phase single-stage tests were carried out in the 2-cm contactor. In these tests, the flow rate of each phase was increased while maintaining the desired O/A flow ratio. When hydraulic performance became unsatisfactory, the increases were stopped. As we have defined hydraulic performance, contactor operation goes from satisfactory to unsatisfactory when the other-phase carryover in at least one of the effluent streams goes from less than 1% of the other phase entrained in it to more than 1%. At each flow rate, the tests of hydraulic performance were done as follows. First, the flow of aqueous phase is started through the empty but spinning rotor. After the aqueous phase is coming out of the exit port so that operation is initially aqueous continuous, the organic flow is started. The amount of organic in the aqueous effluent (O in A) and the amount of aqueous in the organic effluent (A in O) are measured and the appearance of each effluent is noted. Then the aqueous flow is stopped, but the rotor is kept spinning and the organic phase continues flowing. The contactor stage is operated for 3 to 5 residence times in this way so that only organic phase is in the mixing zone and this zone is organic continuous. The aqueous phase trapped in the rotor prevents the organic phase from coming out of the aqueous exit port. Finally, the aqueous flow is restarted with the organic phase as the continuous phase. Again, the amounts of O in A and O in A are measured and the appearance of each effluent noted.

If only one phase can be the continuous phase at the flow condition, then the results will be about the same no matter which phase was the initial continuous phase. By testing in this way and finding that the final hydraulic performance is the same no matter which phase is the initial continuous phase, one can conclude that the process section should be resistant to process upsets.

Single-stage tests under extraction section conditions were done using the 2-cm contactor. Test results before the upper weirs of the rotors were enlarged are given in Table B-3. Test results after the upper weirs of the rotors were enlarged are given in Table B-4. Only rotor X1 was subjected to the single-stage tests both before and after rotor enlargement. While tests were done at O/A flow ratios of both 0.2 and 1.0, the tests at 0.2 are most representative of the CSSX extraction section conditions where the O/A flow ratio is 0.3. The total flow rate (both phases) is highest in the extraction section, 60 mL/min. This is close to the maximum throughput. Before the upper weir was enlarged, the maximum throughput was 82 ± 10 mL/min. This can be seen from Table B-3 if, when the organic phase is the initial continuous phase, the 2% O in A at a total throughput of 72 mL/min is ignored. After the upper weir was enlarged, the maximum throughput was 102 ± 10 mL/min. This can be seen from Table B-4 if, when the organic phase is the initial continuous phase, the 2% O in A at a total throughput of 72 mL/min is ignored. The other three X rotors, X2-X4, had maximum throughputs of 62 mL/min or less before they were modified. After these three rotors were modified, they were used in the 4-stage hydraulic performance tests.

Single-stage tests under strip section conditions were done using the 2-cm contactor after the rotors were modified. The results, given in Table B-5, are for the one rotor tested, X1. The tests were done at an O/A flow ratio of 5, which is used in the strip section. Since the total flow rate in the strip section is 17.0 mL/min, testing was stopped when the total flow rate reached 70 mL/min, even though hydraulic performance was still satisfactory. Thus, for the CSSX strip section, the maximum flow rate is >70 mL/min. These results indicate that enlarging the upper weir will not cause hydraulic performance problems in the strip section.

Table B-3. Hydraulic Performance of a Single-Stage 2-cm Contactor with Upper-Weir Radius of 6.12 mm Using CSSX Solvent and Four-Component Simulant

Rotor	O/A Flow Ratio	Total Flow Rate, mL/min	Initial Continuous Phase	O in A Carryover, %	A in O Carryover, %	Appearance of A Phase[a]	Appearance of O Phase[a]
X1	0.2	72	A	0	<1	Cldy	Clr
"	0.2	72	O	0	2	Cldy	Cldy
"	1	72	A	0	<1	V cldy	Clr
"	1	72	O	0	0	Cldy	
"	0.2	92	A	0	9.4	Cldy	Cldy
"	0.2	92	O	0	9.9	Cldy	Cldy
"	1	92	A	0	16.8	Cldy	Cldy
"	1	92	O	0	1.5	Cldy	Clr
"	0.2	112	A	0	14.3	Cldy	Cldy
"	0.2	112	O	0	14.4	Cldy	Cldy
"	1	112	A	0	5.3	Cldy	Cldy
"	1	112	O	0	5.4	Cldy	Cldy
X2	0.2	52	A	0	0	V cldy	Cldy
"	0.2	52	O	0	0	V cldy	Cldy
"	1	52	A	0	4.6	Cldy	Cldy
"	1	52	O	0	0	Cldy	Cldy
"	0.2	72	A	0	4	Cldy	Cldy
"	0.2	72	O	0	4.4	V cldy	Cldy
"	1	72	A	0	2.6	Cldy	V cldy
"	1	72	O	0	0	Cldy	Cldy
X3	0.2	52	A	0	3.7	Cldy	Cldy
"	0.2	52	O	0	9.7	Cldy	Cldy
"	1	52	A	0	4.1	Cldy	Cldy
"	1	52	O	0	2.9	Cldy	Cldy
"	0.2	72	A	0	4.9	Cldy	Cldy
"	0.2	72	O	0	6.7	Cldy	Cldy
"	1	72	A	0	11.3	Cldy	V cldy
"	1	72	O	0	1.5	Cldy	V cldy
X4	0.2	52	A	0	9.1	Cldy	Cldy
"	0.2	52	O	0	13.8	Cldy	Cldy
"	1	52	A	0	7	Sl cldy	Cldy
"	1	52	O	0	2	Cldy	Cldy
"	0.2	72	A	0	7.5	Cldy	Cldy
"	0.2	72	O	0	6	Cldy	Cldy
"	1	72	A	0	14.8	Cldy	Cldy
"	1	72	O	0	2.1	Cldy	Cldy

[a] Appearance Code: cldy = cloudy, clr = clear, sl = slightly, and v = very.

Table B-4. Hydraulic Performance of a Single-Stage 2-cm Contactor with Upper-Weir Radius of 6.24 mm Using CSSX Solvent and Four-Component Simulant

Rotor	O/A Flow Ratio	Total Flow Rate, mL/min	Initial Continuous Phase	O in A Carryover, %	A in O Carryover, %	Appearance of A Phase[a]	Appearance of O Phase[a]
X1	0.2	72	A	0	0	Cldy	Sl cldy
"	0.2	72	O	0	2.1	Cldy	Cldy
"	1	72	A	0	4.3	Cldy	Cldy
"	1	72	O	0	0	Cldy	Cldy
"	0.2	92	A	0	<1	Cldy	Cldy
"	0.2	92	O	0	<1	Cldy	Cldy
"	1	92	A	0	9.1	Clr	Cldy
"	1	92	O	0	1.2	Hazy	Cldy
"	0.2	112	A	0	4.7	V cldy	Cldy
"	0.2	112	O	0	4.9	V cldy	Cldy
"	1	112	A	0	8.6	Cldy	V cldy
"	1	112	O	0	1.5	Cldy	Cldy

[a] Appearance Code: cldy = cloudy, clr = clear, sl = slightly, and v = very.

Table B-5. Hydraulic Performance of Single-Stage 2-cm Contactor with Upper-Weir Radius of 6.24 mm Using CSSX Solvent and 0.001 $\underline{M}$ HNO_3

Rotor	O/A Flow Ratio	Total Flow Rate, mL/min	Initial Continuous Phase	O in A Carryover, %	A in O Carryover, %	Appearance of A Phase[a]	Appearance of O Phase[a]
X1	5	50	A	<1	<1	Sl cldy	Cldy
"	5	50	O	<1	<1	Sl cldy	Cldy
"	5	60	A	0	0	Crys clr	Cldy
"	5	60	O	0	<1	Hazy	Cldy
"	5	70	A	<1	<1	Sl cldy	Cldy
"	5	70	O	<1	<1	Clr	Cldy

[a] Appearance Code: cldy = cloudy, clr = clear, crys = crystal, hazy = hazy, and sl = slightly.

Typically, when there is a difference in the cloudiness of the two effluents, the cloudier phase is the continuous phase. With an O/A flow ratio of 5 for the strip section tests, the continuous phase was expected to be the organic phase. That this was the case seems to be confirmed in that it was also the cloudier phase. Such observations were harder to make for the extraction section tests as almost everything came out cloudy. However, when the unit starts out aqueous continuous with an O/A of 0.2, the aqueous phase is expected to be continuous and the aqueous effluent is expected to be cloudier when there is a difference between the two effluents.

There was only one case (see line 1 in Table B-4) where this was not the case. What is not clear is whether the cloudiness is due to very finely dispersed other-phase carryover or to very fine air bubbles trapped in the continuous phase.

b. Four-Stage Operation

The results of the two-phase hydraulic performance tests done in the 4-stage 2-cm contactor are given in Tables 2 and 3. In these tests, the four-component simulant flow rate was 46 mL/min with an O/A of 0.3 so that the conditions in the extraction section for the CSSX flowsheet shown in Fig. 2 were duplicated. Using the B rotors, the test was repeated at a lower simulant flow rate, 30 mL/min. The test operators still observed backup in the organic-side interstage lines, other-phase carryover in the organic interstage lines, and aqueous phase in the organic effluent. When the aqueous flow rate was reduced further to 25 mL/min with an organic flow rate of 14 mL/min, the test operators continued to observe other-phase carryover in organic interstage lines and aqueous phase in the organic effluent.

After the contactor was improved, its hydraulic performance, given in Tables 2 and 3, shows that the extraction section was now working well. Other tests were made under strip section conditions, 0.001 $\underline{M}$ HNO_3 and an O/A of 5. These tests also showed good operation even though backup in the organic-side interstage lines was higher, up to 50% in some interstage lines. This increased liquid backup probably reflects on the position of the plastic insert. If the insert is not positioned correctly, that is, if the insert does not extend into the mixing zone sufficiently (0.8 mm), the backup in the interstage line can be increased to 50 or 60%.

Hydraulic performance was also tested in a 4-stage 2-cm contactor for three possible problems: (1) high rotor runout, (2) low rotor speed, and (3) non-spinning rotor. The high runout was caused by the motor shaft being slightly bent. The low-speed rotor was a motor manufacturing defect. The motor of the non-spinning rotor had been turned off. In these tests, the aqueous feed was pumped into stage 4. When it came out of stage 1, the organic phase was started pumping into stage 1. For each problem, the observed hydraulic behavior is given in Table B-6. In each case, the hydraulic problems were severe with liquid backup in interstage lines, especially on the organic side, and high other-phase carryover in both the interstage lines and the effluents. The corrective action for each problem is also given in Table B-6. These tests were done without the plastic inserts in the inlets to each stage. If these inserts had been in place, the effect of high rotor runout would probably have been less.

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Table B-6. Hydraulic Behavior Caused by Various Rotor Problems

Aqueous Phase	O/A	Rotor Problem	Observed Hydraulic Behavior and Problems	Corrective Action	Hydraulic Behavior after Corrective Action
0.001 $\underline{M}$ HNO$_3$	5	High rotor runout in stage 2 [a]	25-50% backup in A-side interstage line, 50-100% backup in O-side interstage line, high carryover in both sides interstage lines, 8% A carryover in O effluent and 13% O in A effluent	Replace defective motor	5-10% backup in A-side interstage line, 40-50% backup in O-side interstage line, low carryover in O-sides interstage lines, 5% carryover in O-effluent
Four-component simulant	0.3	High rotor runout in stage 3 [a]	40-95% backup in O-side interstage line, high carryover in O-side interstage lines, 19% A carryover in O effluent and 3% O in A effluent	Replace defective motor	50% backup in O-side interstage line, Less carryover in O-side interstage lines, 6% carryover in O-effluent
0.001 $\underline{M}$ HNO$_3$	5	Low motor speed in stage 2 [b]	40-50% backup in A-side interstage lines, 50-95% backup in O-side interstage line, some emulsion-like liquid in O-side interstage lines, 1% A carryover in O effluent and 85% O in A effluent	Replace defective motor	(See 0.001 $\underline{M}$ HNO$_3$ results above)
0.001 $\underline{M}$ HNO$_3$	5	Rotor stopped in stage 2	45% backup in A-side interstage lines, plenty of emulsion-like liquid in O-side interstage lines, no organic effluent coming out of the unit, 91% O carryover in A effluent	Turn on motor	(See 0.001 $\underline{M}$ HNO$_3$ results above)

[a] Rotor runout was 0.028 in. (0.71 mm).
[b] Rotor speed was 1800 rpm instead of the manufacturer-specified 3600 rpm.

3. Solvent Wash Section

Earlier CSSX tests had been run without stage 33, the solvent wash stage shown in Fig. 2. As a result, no data was available on the operation of such a stage. To get the required data, batch tests were done to determine the dispersion number (also called the Leonard number). The dispersion number results provide an indication of hydraulic performance over a range of aqueous-phase concentrations and O/A ratios [LEONARD-1995]. Then, one-stage tests in a 2-cm contactor give actual hydraulic performance for planned operating conditions. While the composition of the solvent wash was specified to be 0.01 $\underline{M}$ NaOH, we were told by Bruce Moyer at ORNL, that either higher or lower NaOH concentrations might be required. Because of

this, the range of NaOH concentrations investigated using the dispersion number was varied from 0 (pure water) to 0.5 $\underline{M}$.

a. Batch Tests

To evaluate the general ability of a two-phase dispersion to separate quickly and yield a good solvent wash section, batch tests that yield the dispersion number were applied to the CSSX solvent for the range of possible aqueous wash feeds from 0 (pure water) to 0.5 $\underline{M}$ NaOH. The procedure for the dispersion number test is described in LEONARD-1995. The tests were done at room temperature (22°C) in a 100-mL graduated cylinder. The dispersion numbers, N(Di), for the various aqueous phases and O/A ratios are given in Table B-7. The performance ratings for N(Di) in this table are based on the definitions summarized in Table B-8. The results suggest that contactor operation will be good except when the NaOH concentration is very low, 0.01 $\underline{M}$ or less, and the O/A ratio is low, which would give aqueous-continuous operation. Since the O/A ratio will be high in the solvent wash section, the hydraulic performance should be satisfactory. If a process upset allows a low O/A ratio to occur in the solvent wash stage, an emulsion could form that would take a long time to coalesce.

Table B-7. Gravity-Settling Dispersion Numbers for Various Aqueous Phases with CSSX Solvent at 22°C

Aqueous Phase	O/A	Avg. N(Di)	Performance Rating	Dispersed Phase	Appearance of Aqueous Phase[a]	Appearance of Organic Phase[a]
Water	0.2	1.8E-04	P	O	V cldy	Crys clr
"	1	1.1E-03	VG	A	Clr	V cldy
"	5	1.4E-03	VG	A	Crys clr	V cldy
0.001 $\underline{M}$ NaOH	0.2	1.9E-06	VP	O	V cldy	Clr
"	1	1.0E-03	VG	A	Clr	Cldy
"	5	1.0E-03	VG	A	Clr	V cldy
0.01 $\underline{M}$ NaOH	0.2	1.8E-05	VP	O	V cldy	Clr
"	1	1.1E-03	VG	A	Clr	V cldy
"	5	1.2E-03	VG	A	Crys clr	V cldy
0.1 $\underline{M}$ NaOH	0.2	5.9E-04	G	O	V cldy	Hazy
"	1	9.6E-04	VG	A	Crys clr	V cldy
"	5	1.1E-03	VG	A	Crys clr	V cldy
0.5 $\underline{M}$ NaOH	0.2	1.7E-03	E	O	V cldy	Cldy
"	1	1.0E-03	VG	A	Crys clr	V cldy
"	5	1.2E-03	VG	A	Crys clr	V cldy

[a] Appearance Code: cldy = cloudy, clr = clear, crys = crystal, hazy = hazy, and v = very.

Table B-8. Criteria for Performance Rating Using the Dispersion Number

N(Di) Value	Performance Rating
N(Di) > 16 E-4	Excellent (E)
8 E-4< N(Di) < 16 E-4	Very good (VG)
4 E-4< N(Di) < 8 E-4	Good (G)
2 E-4< N(Di) < 4 E-4	Fair (F)
0.2 E-4< N(Di) < 2 E-4	Poor (P)
N(Di) < 0.2 E-4	Very poor (VP)

b. Hydraulic Performance

While the dispersion number test quickly test indicates how a liquid-liquid pair will perform in solvent extraction equipment, some of the performance is specific to the equipment. In addition, process upsets and starting from an aqueous-continuous condition, which cannot be avoided in the centrifugal contactor, have the potential to cause problems based on the results from the dispersion number test. For both of these reasons, it is important to do additional hydraulic performance tests in the type of equipment that will be used in the process. Thus, single-stage 2-phase tests were carried out in a 2-cm contactor over a range of appropriate conditions. The aqueous solutions were 0.001, 0.01, and 0.1 M NaOH, the O/A flow ratios were 1 and 5, and the highest flow rate (both phases) was 28 mL/min, which was 65% higher than the planned flow rate of 17 mL/min in the solvent wash section. The results, given in Table B-9, show that hydraulic performance is satisfactory over the range of expected conditions. In addition, they show that starting up from initially aqueous-continuous conditions is not a problem. These results indicate that operating the solvent wash stage at an O/A flow ratio of 5 using 0.01 M NaOH as the solvent wash (FF) feed will not present any hydraulic problems.

Table B-9. Single-Stage Hydraulic Performance of 2-cm Contactor for the CSSX Solvent with Dilute NaOH Solutions

Aqueous Solution	O/A Flow Ratio	Total Flow Rate, mL/min	Initial Cont. Phase	O in A Carryover, %	A in O Carryover, %	Appearance of Aqueous Phase[a]	Appearance of Organic Phase[a]
0.001 M NaOH	5	17	A	0	0	Hazy	Clr
"	5	17	O	<1	0	Hazy	Cldy
"	1	28	A	0	0	Crys clr	Crys clr
"	1	28	O	<1	0	Clr	V cldy
0.01 M NaOH	5	17	A	0	0	Hazy	Clr
"	5	17	O	0	0	Clr	Crys clr
"	1	28	A	0	0	Hazy	Clr
"	1	28	O	0	0	Hazy	Cldy
0.1 M NaOH	5	17	A	0	0	Clr	Clr
"	5	17	O	0	0	Cldy	Clr
"	1	28	A	0	0	Clr	Hazy
"	1	28	O	0	0	Clr	Cldy

[a] Appearance Code: cldy = cloudy, clr = clear, crys = crystal, hazy = hazy, and v = very.

4. Effect of Insert Orientation

As reported in the main body of this report, the use of Teflon™ inserts in the perpendicular inlet ports improved hydraulic performance. These inserts were cut from thin-walled Teflon tubing consisting of a copolymer of tetrafluoroethylene and hexafluoropropylene (FEP). A special gauge was used to position the inserts in the mixing zone so that they do not interfere with the spinning rotor. As shown in Fig. B-2, the insert should be oriented so that it shields the liquid entering from the interstage line from the swirling turbulent liquid in the annular mixing zone. To determine the effect of insert orientation, four different orientations were tested. Without any insert, the liquid level in the interstage line was 50 to 60%. When insert orientation was in position a or c in Fig. B-2, the liquid level was even higher. When insert orientation was in position of b or d shown in Fig. B-2, the liquid level was lower. For the tests, position b was chosen. While position d would have been preferred, it was close to the worst position, position a. In practice, because the inserts were hard to place in the contactors in the glovebox, the orientation of an insert was considered satisfactory if it was between positions b and d.
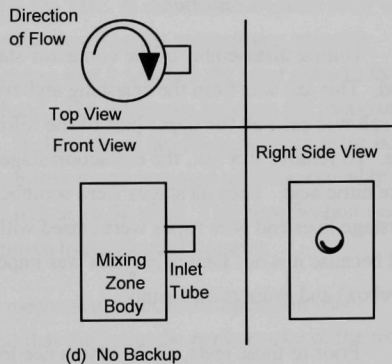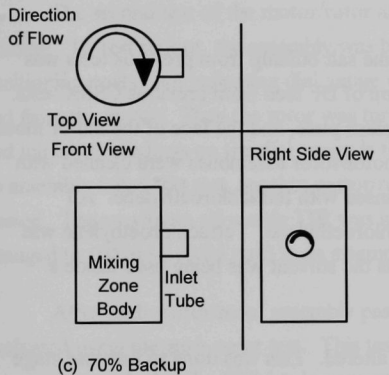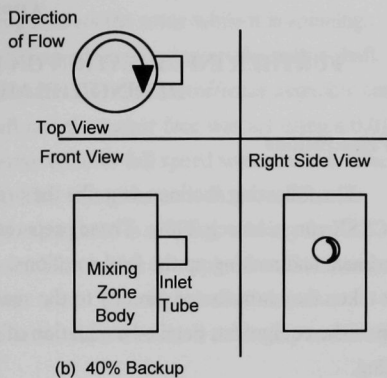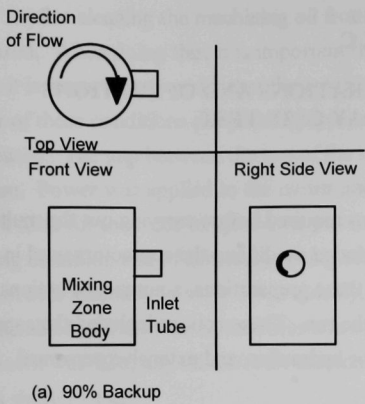
Fig. B-2. Effect of Insert Orientation on Liquid Backup in the Organic Interstage Line

APPENDIX C

FURTHER INFORMATION ON PREPARATIONS AND OPERATIONS
DURING THE MULTI-DAY CSSX TEST

1. Preparations

The following sections describe the preparations required before carrying out the multi-day CSSX run in March 2001. These preparations included modifying the contactors used in this experiment and making up the feed solutions. During these preparations, a number of actions were taken that contributed directly to the success of the run. These actions included the careful setup of the equipment, detailed evaluation of contactor hydraulics, and extensive personnel training.

a. Contactor

During disassembly of the contactor stages, some salt buildup from previous tests was noted. This salt was from the splashing and evaporation of DF feed from previous CSSX tests. The salt was seen on the upper part of the rotor, the splash plate, and the face of the motor mount plate. To remove this salt, the extraction stages and motor/rotor assemblies were cleaned with dilute nitric acid. Then all stages were scrubbed and rinsed with tetrachloroethylene. All interstage lines and wire ropes were rinsed with tetrachloroethylene. Tetrachloroethylene was used because it is not flammable (this was important, as the solvent was being used inside a glovebox) and evaporates cleanly.

Prior to these tests, all the rotors needed to be altered. This was done to increase stage efficiency and throughput. The first modification was to increase the rotor inlet diameter from 7.92 mm (0.312 in.) to 10.72 mm (0.422 in.) for any rotor that had not been modified previously (see LEONARD-2001A). This increases the liquid level in the mixing zone of the contactor stages, thus increasing residence time there. The second modification was to increase the upper-weir radius of each rotor from 6.12 to 6.24 mm to allow a higher throughput. The upper-weir opening was enlarged using electrical discharge machining (EDM). Even though the rotors were being modified in a "hot" shop, they still needed to be decontaminated before being machined. The rotors were decontaminated by soaking them in 8 $\underline{M}$ nitric acid and then rinsing thoroughly in water. The rotors were decontaminated down to a few hundred to a few thousand dpm of beta/gamma activity by direct reading. During the machining, cutting oil was used. To remove the residual oil from the rotors prior to reinstalling them in the contactor stages, the rotors were ultrasonically washed in a Citranox™ degreasing solution and then thoroughly rinsed in water.

After cleaning the machining oil from the rotors, the motor/rotor assemblies were prepared. When doing this, it is important that nothing hinders the rotor while it is spinning. Also, it is important to verify that the rotor is not wobbling while spinning on the motor shaft. If either of these conditions are present, hydraulic performance for that motor/rotor assembly can be decreased. The gap between the top of the rotor shaft and the motor face was set using a 0.015-in shim. Power was applied to the motor and then removed after full speed was attained. The time it took for the rotor to spindown was measured. This test gives an indication of motor bearing condition and of something being off-center so that there is rubbing, causing the motor/rotor assembly to slow down quickly. Any assemblies with a low spindown time were disassembled and another motor/rotor combination was tried. The target cutoff time for spindown was 12 s; any motor/rotor assembly with a lower time was fixed unless the motor alone had a shorter time.

The second test of the motor/rotor assembly is the TIR. It shows how true the rotor is running. To test runout, the assembly was bolted upside down on the motor/rotor assembly positioning posts. An indicating dial gauge was placed so that it touched the rotor close to the far end from the motor. Then the rotor was turned by hand. The difference between the maximum and minimum readings on the dial gauge is the TIR. It shows how far off-center each rotor is. If an assembly failed this test, another motor/rotor combination was prepared until the assembly passed. The maximum allowable TIR was initially 0.012 in. (0.30 mm). This specification was changed to 0.010 in. (0.25 mm) in an attempt to improve hydraulic performance.

After each motor/rotor assembly passed the mechanical tests, rotor operation was evaluated using the zero-point test. This test verified that the hydraulic performance of the rotor was as expected after the EDM enlargement of the upper weir. To do this test, the motor/rotor assembly was placed into a contactor stage and water was pumped through the stage. The flow rate of water was increased until water exited the less-dense-phase exit port. The zero-point is the flow rate at which the water first exits from the less-dense-phase exit port. Using the zero-point flow rate, the apparent weir diameter of the rotor can be calculated. The apparent weir diameter was compared with the actual weir diameter to determine if the rotor was acceptable. In these tests, the apparent diameter was less than the actual diameter by $1.7 \pm 1.1\%$. This difference is attributed to the limitations of the model.

Inserts were added to the interstage line inlets prior to placing the motor/rotor assemblies into the contactor stages. When the liquid level in the annular mixing zone is so high that it reaches the inlet port, the liquid can be forced to back up into the interstage line between contactor stages. Since the inlet tube is perpendicular to the mixing-zone wall, the incoming

liquid can, in some cases, back up all the way to the exit port of the previous stage. Such a backup decreases the hydraulic performance of the contactor and can stop the experiment. To eliminate most of this liquid backup in the interstage lines, a small piece of tubing is inserted into the tube on the contactor housing that goes from the interstage line to the mixing zone. The insert deflects the liquid spinning in the annular mixing zone away from the inlet, creating a space where the interstage liquid can enter the stage. The inserts must be properly positioned to be effective. The inserts are pushed into the housing inlet tube until they are about 1/32 in. from the rotor wall. A schematic giving the proper insert orientation relative to the spinning rotor is shown in Fig. 4 (see the main body of this report).

To obtain the correct spacing for the insert, a jig was fabricated that could be inserted into the contactor body. The jig had a diameter that was 1/16 in. (1.6 mm) larger than the diameter of the rotor (1/32 in. (0.8 mm) larger than the radius). In this way, the gap between the insert and the rotor was kept at 1/32 in. (0.8 mm). While the interstage lines were fabricated using perfluoroalkoxy (PFA) Teflon tubing with an outside diameter of 1/2 in. (12.7 mm) and an inside diameter of 3/8 in. (9.5 mm), the inserts are fabricated using FEP tubing with an outside diameter of 3/8 in. (9.5 mm) and an inside diameter of 5/16 in. (7.9 mm). The FEP tubing was cut into lengths, then a longitudinal section of the tubing covering an arc of about 165° was cut out to make the insert. The insert length was initially 4 in. (102 mm) long. It appeared that, at this length, the insert interacted with the wire rope. Thus, most inserts were made shorter, about 3 5/8 in. (92 mm) long. The length was chosen so that only 3/8 in. (9.5 mm) of the insert stuck out of the stainless-steel inlet tube that is part of the contactor housing. With this much of the insert sticking out of the tube, the insert can be rotated to the proper orientation. After fabrication, the inserts were pushed into the housing inlet ports until they touched the spacing jig. The jig was removed and the ends of the wire ropes and the inserts were inspected using a mirror. Then the motor/rotor assembly was replaced. To verify that the inserts and wire ropes did not interfere with the rotation of the rotor, an *in situ* spindown test was completed.

The *in situ* spindown tests are done as follows:
- The motor/rotor assembly is inserted into the stage.
- The motor is turned on.
- The rotor is allowed to reach full operating speed.
- The motor is switched off at the same time a stopwatch is started.
- After 10 to 20 s, the still spinning motor/rotor assembly is lifted out of the stage.
- The rotor is observed until it stops spinning (typically an additional 10 to 20 s) and the stopwatch is stopped.
- The spindown time for the motor/rotor assembly is noted.

The first *in situ* test is done before the inserts are added. It is a baseline test and is compared with the results obtained when the motor/rotor assembly was put together. These two spindown times should be the same if the rotor is not rubbing against the contactor housing. This test was the reference point for future tests and was completed with neither the insert nor the interstage line in place. Once the baseline spindown was measured, the inserts were placed into the interstage lines using the spacing jig. The spindown time was measured again. If it was significantly less than the baseline, the inserts were removed and repositioned. This process was repeated until the spindown time with the inserts in place was approximately equal to the baseline spindown time. Next, the interstage lines were attached. Each interstage line contains a wire rope that extends from the collector ring to the annular mixing zone of the contactor. A second spacing jig was made so that the interstage lines can be positioned correctly on the contactor body. The wire ropes reduce slug flow between stages by acting as wicks that draw liquid out of the collector ring into the interstage lines. Slug flow, which occurs only at low throughputs, is thought to be one of the major reasons for the decreased stage efficiency seen in earlier multistage 2-cm tests. Once the interstage lines were installed, the spindown time was measured again. If the spindown time with the interstage lines in place was significantly less than the baseline, the interstage lines were removed and repositioned until the spindown time was close to the baseline.

For the multi-day test, one additional stage (stage 33) was added as a solvent wash stage next to stage 1 (adjacent to the extraction section). The position of stage 33 is shown in Fig. 2. The purpose of the solvent wash stage is to strip impurities and keep anionic species from building up in the solvent. The stage has a special design so that it could be disassembled into pieces that could be brought into the glovebox through the port with an 8-in. diameter. This is the only port that was available for moving materials into the glovebox. Once inside, the stage was reassembled.

When the motors are running, the entire contactor block heats up by conduction of heat from the motor. The Cs distribution ratio for the CSSX solvent is very sensitive to temperature, decreasing with increasing temperature. To keep this ratio high in the extraction section, a set of four chiller bars was added to that section (stages 1-15). This heat exchanger consisted of four sections of square pipe, each one was about 12-in. (305-mm) long, which was enough to cool four contactor stages. Metal blocks that were crescent-shaped on one side were used to conduct the heat from the contactor body to the chiller bar. The square pipes and crescent-shaped blocks were made of stainless steel to minimize corrosion from the acidic and caustic solutions. These crescent blocks were clamped to the outside of the contactor by the square pipe at the level of the annular mixing zone. Thermon® T-85, a thermally conductive paste, was applied to the contact surfaces of the crescent block to improve heat transfer. A sketch of this heat-transfer assembly is

shown in Fig. A-3. Water from a recirculating chiller bath outside the glovebox was pumped through the square pipe to keep it cool. During the multi-day test, the recirculating chiller bath was set at 7.5°C, which kept the contactor bodies around 26°C. To see that the chiller bath was working, an adhesive liquid-crystal thermometer was placed on either end of the chiller bar.

Several temperatures were measured during the test to verify that the heat exchanger was cooling the extraction section. The housing temperature for the stages in the extraction section was measured either by a digital thermometer taped to the outside of the stage or by an adhesive liquid-crystal thermometer. Temperatures for most of the other contactor housings were measured as well. Of the 33 stages used for test CS27, 13 stages had digital thermometers taped to them and 17 had liquid-crystal thermometers. The liquid-crystal adhesive thermometers were used to avoid the clutter of each stage having a digital thermometer taped on. The digital thermometers were easier to read and more accurate. The liquid-crystal thermometers are not recommended. Each time an effluent sample was taken, its temperature was measured, in addition to the housing temperatures.

b. Rotor

Previous operation and hydraulic testing of the 2-cm centrifugal contactors had shown the need to verify the correct direction of rotor spin and its speed [LEONARD-1999, -2001A]. During placement of each motor/rotor assembly, the rotor speed and direction of rotation were checked. This was done before it was verified that the rotor was not rubbing on the side or bottom of the contactor housing, the plastic insert, or the wire rope installed in each interstage line.

c. External Feed and Raffinate Tanks

The radioactive feed and raffinate tanks were placed outside the glovebox for three reasons, all of which worked to minimize radiation exposure to operating personnel and observers. First, radiation control and protection was simplified and improved by having the tank drums in a restricted access storage area. Second, operation during the test was simplified, since it was no longer necessary to bag waste (DF) feed into or DW raffinate out of the glovebox in bottles. Third, it simplified preparation of the radioactive feed in that the addition of the ^{137}Cs spike was done once to the 55-gal drum instead of adding radiotracer to several carboys or bottles full of simulant.

During the multi-day test, radioactive feed was pumped into a surge tank inside the glovebox using a jack pump. This pump was located outside of the glovebox, as shown in Fig. C-1, and potential leak points around the fittings and the pump head were wrapped in a plastic bag.

The pump with the bag was seated in a tray to contain any leaks. The transfer line into the glovebox consisted of a plastic line inside a stainless-steel line. The plastic transfer line had a valve on the discharge side of the pump to stop flow in case of an emergency.
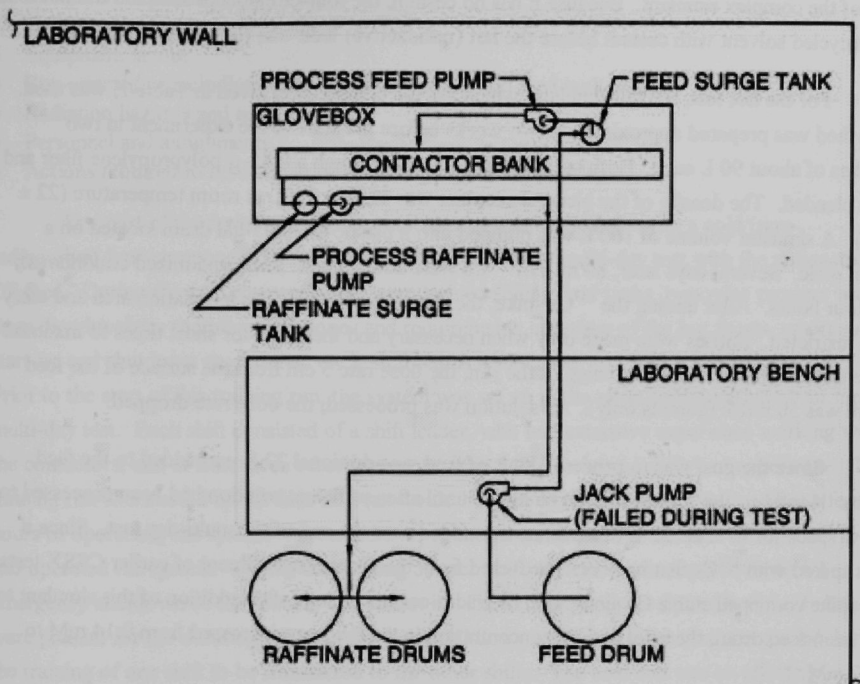
Fig. C-1. Schematic Layout of Feed and Raffinate System for Multi-Day Test

In a similar fashion, the DW raffinate in the glovebox was pumped to an external collection drum. The DW raffinate exiting the contactors was collected in a surge tank and pumped out of the glovebox using the same kind of double line used for the DF feed. The continuous plastic line discharged directly into a 55-gal drum with a plastic liner. Since the radioactive feed was being diluted by about 5% with the scrub solution and the wash solution was also being pumped out from this surge tank, the waste liquid would not fit into a single 55-gal drum. Thus, it was necessary to change the raffinate drum during the experiment. This was done when about 150 L had been collected in the first drum.

d. Feed

The four-component simulant was used for hydraulic performance tests, the training run (CS26), and the start-up hot test (CS27). This simulant has a density and alkalinity close to that of the complex simulant. Because it has no cesium, the four-component simulant did not load the recycled solvent with cesium before the hot (radioactive) feed was started for test CS27.

For the hot test, the full simulant, which has the composition given in Table 1, was used. This feed was prepared approximately two weeks before the start of the experiment in two batches of about 90 L each. Both batches were filtered through a 0.45-μ polypropylene filter and then blended. The density of the blended simulant was 1251 ± 3 g/L at room temperature ($22 \pm 3°C$). A simulant volume of 160 L was pumped into a plastic-lined 55-gal drum located on a drum scale. Several days later, 80 mCi of ^{137}Cs were added to the drum and mixed continuously for four hours. After adding the ^{137}Cs spike, the drum was roped off as a radiation area and entry was restricted. Entries were made only when necessary and then only for short times to minimize dose to personnel. At the beginning of the run, the dose rate 5 cm from the surface of the feed drum was 50 mR/h (gamma only). As solution was processed, the dose rate dropped.

Since the goal was to process 180 L of feed, an additional 22 L was added to the feed drum. However, the 22 L could not be added until after sufficient solution had been processed to allow space for it. This 22 L of simulant was added near the end of the multi-day test. Since it was spiked with ^{137}Cs that had been recovered from the Cs product effluent of earlier CSSX tests, the spike contained stable Cs along with the radio-cesium. Thus, after addition of this simulant to the main feed drum, the total cesium concentration in the feed was increased from 0.14 m$\underline{M}$ to 0.17 m$\underline{M}$.

Aqueous solutions were also prepared for scrub, strip, and solvent wash feeds. The test required 20 L each of 0.01 $\underline{M}$ sodium hydroxide, 0.05 $\underline{M}$ nitric acid, and 0.001 $\underline{M}$ nitric acid. These solutions were prepared and titrated at ANL. The 0.01 $\underline{M}$ sodium hydroxide was used for the wash (FF) feed, the 0.05 $\underline{M}$ nitric acid was used for the scrub (DS) feed, and the 0.001 $\underline{M}$ nitric acid was used for the strip (EF) feed.

e. Personnel Training

Because of the length of the run and the need to operate 24 h per day, 16 operators and two health physics people were needed for this test. Since several of the operators were not familiar with centrifugal contactors or the CSSX process and few of us had experience with a multi-shift operation, a training session was conducted to provide everyone with the needed background. In this training, the following items were covered:

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- Flowsheet
- Solution and solvent compositions
- Process goals
- Operation of centrifugal contactors
- Results from previous CSSX tests
- Contactor hydraulic performance
- Equipment layout
- Run operations including sampling, flow checks, and feed tank refilling
- Radiation hazards and requirements
- Personnel and assignments
- Actions required to resolve various potential operating problems

As a part of the training, a short contactor run was carried out with a cold (non-radioactive) four-component DF feed prior to the start of the multi-day test with the radioactive DF feed. During the cold run, personnel were trained (filling feed tanks, removing product bottles from the glovebox, sampling techniques and requirements, a review of the log sheets, emergency start-up and shutdown procedures, etc.), shift teams established, and shift turnover practiced. Prior to the start of this training run, the system was set up in the same configuration as for the multi-day test. Each shift consisted of a shift leader, who had extensive experience working with the contactors, and at least three other operators, who had a variety of skills and experience. The training run was started by the first shift (as would be done for the multi-day test). After a few hours of operation, the system was turned over to the next shift, and so on until all three shifts had operated the system. During the training of each shift, the crew was required to do an emergency shutdown of the process and restart it. The shift supervisors for each of the 3 shifts were present for the duration of the training run. This allowed knowledge that was gained during the training of one shift to be transferred to the other shifts. The cold run was an effective training tool. It allowed inexperienced operators to operate the system before being asked to run the experiment. It also verified that the shift-change procedure was effective. As the first training run was rather rough, procedures were revised and a second training run carried out. This one went very smoothly. Overall, approximately 5 d were spent conducting training for the 2-cm centrifugal contactors.

In addition to training personnel, the cold test provided (1) a check of the tubing connections, pump settings, and general setup of the system; (2) additional cleaning of the contactors, flushing out any residual radioactive isotopes and other materials that might still be left from previous tests; and (3) a multistage test of the hydraulic operation of the modified contactors. It was during the second training run that the problem with the backup of solution in the interstage lines was identified. By developing the inserts for the inlet tubes to each stage, this

problem was solved. A third cold run was done with the inserts in place to verify that they had solved this problem.

f. Operational Readiness Review

Before the multi-day test was started, an operational readiness review was carried out by people from SRS and ORNL. The review team consisted of Robert Hinds (Chair) and Seth Campbell from SRS and Leon Klatt from ORNL. The review team did a final check of the equipment installation, the training, the quality assurance plan, the implementation of safety and radiation procedures, and the method for data collection and documentation.

2. Operations during Multi-day Test

This section describes operations during the multi-day (nominal 5-d) test.

a. Process Operation

The cesium in the waste (DF) feed was extracted by the solvent. The cesium was subsequently stripped from the solvent and concentrated in the aqueous (EF) effluent. This solution was collected inside the glovebox in 2-L Nalgene® bottles. To reduce the activity level inside the glovebox, these bottles were bagged out of the glovebox as soon as they were full and stored in a shielded area next to the DF feed drum. Dose rates on a full EW bottle averaged 180 mR/h at contact.

Operations were carried out to minimize the individual and collective radiation dose. Because of the presence of the cesium effluent in the glovebox and the high radiation levels that it generated, operations on the aqueous side of the glovebox were controlled to limit operator exposure. This included wearing lead-lined aprons during activities associated with that side of the glovebox and limiting the time spent in this area. Activities that were impacted included examining interstage lines, reading the stage temperatures, and sampling the EW effluent. To further control radiation doses, the operator started the EW sample collection and then moved away until the 5-min collection period ended. Additional radiation dose was received during the preparation of samples for scintillation counting. This was especially important during the start of the test when samples were being collected on a frequent basis, every 15 to 20 min. As the samples built up in the preparation hood, the operator preparing these samples would start to receive a significant dose. By removing the samples from the hood to a shielded area in timely fashion, the dose to the operator was reduced. The overall dose was significantly lower that the maximum amount anticipated for the test.

Log sheets were developed to record data and observations during the run. These log sheets included pump calibrations and flow checks, interstage line appearance, stage and effluent temperatures, feed and effluent tank volumes, and the master run schedule with the sample times.

The start-up procedure used was typical for centrifugal contactors when the aqueous phase is the more-dense phase. First, the stages were filled with the appropriate aqueous feed solutions, then the organic feed flow was started. The detailed procedure was as follows: Before any liquid was fed to the contactor, the contactor motors were turned on and the chiller was started. The first pump to be switched on was the strip (EF) feed, as it took the longest to fill the strip section. The scrub (DS) feed was started second. The cold waste (DF) feed, which was the four-component simulant, was started third. The solvent wash (FF) feed was started fourth. Once the aqueous strip (EW) effluent exited at stage 18, its pH was checked to verify that it was acidic. When the solvent (FW) wash exited stage 33, its pH was checked to verify that it was basic. Once all of the aqueous effluents were flowing and the flow rates and appropriate pH values had been checked, the solvent was started. The solvent flowed into stage 33, the solvent wash stage, and then through stages 1-32. Once the organic (EP) effluent was detected, the system was checked for hydraulic stability by looking for changes in liquid heights in the interstage lines. After verifying that the system was stable, the DF feed was switched from the cold (non-radioactive) four-component simulant to the radioactive waste simulant.

b. Process Upsets

Two upsets to the CSSX process occurred during the multi-day test. Recovery from the process upsets is discussed here. The ramifications of these upsets to the experiment itself are discussed main body of this report. The first process upset was due to the failure of the jack pump for the DF feed. This pump was located close to the DF feed drum on a laboratory bench, as shown in Fig. C-1. The pump failed 16 h into the run. This failure was not noticed until after the DF surge tank had run dry. After the surge tank ran dry, only scrub feed was flowing into the extraction section. Thus, the scrub feed was slowly diluting the liquid in the extraction section and appeared to have completely flushed the waste feed out of the first two extraction stages. Once the failed jack pump was discovered, the entire system was shut down: that is, all of the remaining pumps and the contactor motors were stopped. The jack pump was replaced with a spare and DF feed was pumped into the surge tank. The system was restarted by turning on the rotors and then, approximately 30 s later, turning on all the feed pumps.

Inspection of the failed jack pump, which was a positive-displacement rotary piston pump, showed that the piston had seized inside the pump housing. The pump motor was not affected. It

is not known why the piston seized in the pump housing. The replacement pump operated normally for the remainder of the multi-day test.

The second process upset was the partial plugging of the rotor in stage 15 with white solids. This was observed to happen in the following way. After 68 h of operation, a pH check of the aqueous strip (EW) effluent showed that it was slightly basic. The scrub (DS) feed rate was increased by about a factor of two in an effort to make the EW effluent acidic again. By checking the Cs concentration in the organic (EP) effluent from the strip section, it was verified that the Cs was still being stripped out. After an hour with the high DS feed rate, the EW effluent was still basic. When the EP effluent was checked again, its Cs concentration was found to be very high. At that point, the test was stopped by turning off all of the feed pumps and rotor motors. The stages were inspected by draining the stage liquids and pulling up the motor/rotor assemblies. The stage liquid was added back to the stage before the motor/rotor assembly was re-inserted. When stage 15 was inspected, a thick layer of white solids was found on the inside wall of the rotor, as shown in Fig. C-2. Lesser amounts of solids were found in nearby stages, especially in the extraction section. The source of the white solids and how they caused the high pH in the EW effluent are discussed in the main body of this report and in LEONARD-2001B. A simple experiment showed that the white solids were soluble in 2 $\underline{M}$ HNO$_3$. Each of the rotor/motor assembles from the extraction and scrub sections was removed and the rotor was soaked in 2 $\underline{M}$ HNO$_3$ until all of the white precipitate was dissolved. Each rotor went through two water washes to remove any residual nitric acid before being re-installed.

Start-up from this upset was more difficult because the strip section and the organic feed tank had become contaminated with [137]Cs. First, the strip section was decontaminated by restarting all contactor sections. The rotors were started first. Then, 30 s later, the solvent, strip, scrub, and wash feeds were started. The caustic DF feed was not started. This allowed the organic phase and the aqueous strip (EF) feed to flow in a countercurrent fashion through the strip section and flush out the Cs contamination. Samples of organic (EP) effluent were taken and analyzed for Cs. Eventually the EP effluent was "clean," that is, it contained very little [137]Cs. This indicated that the strip section had been decontaminated. However, the organic phase in the solvent feed tank was still contaminated. Since the EP effluent from the strip section was now clean, it was collected in a separate tank. Solvent was fed from the original feed tank until it was gone. Then the process was switched to full solvent recycle using the new tank containing the decontaminated solvent. The solvent was allowed to run until the raffinate samples contained very little [137]Cs. This indicated that both the extraction and the strip section had been cleaned up. Running the contactor to clean up the strip and extraction sections, took about 9 h. At this point,

the radioactive DF feed was restarted and normal operation was resumed. Normal operation was experienced for the remaining hour of the run.

Fig. C-2. White Solids Plugging the Rotor in Stage 15

c. Shift Operation

Because of the need to run 24 hours per day for three to five days, operators were assigned to 8-h shifts. Since four operators were needed for each shift, a total of 12 operators were required. As health physics coverage was required around the clock, two health physicists, who each worked 12-h shifts, were used. The same two health-physics technicians were assigned for the duration of the test. This worked very well as they both were familiar with our experiment and the radiation levels associated with various aspects of the test. In addition, there were two observers, Seth Campbell from SRS and Leon Klatt from ORNL. They worked the first two 8-h shifts. Shift assignments were made prior to the start of the run, and a shift-change checklist was

developed to aid in the transfer of information from shift to shift. One operator was assigned as the experimental lead for each shift. This person was in charge of the run and was responsible for making decisions during the test. Other duties were assigned to the other operators as well as the experimental lead. One operator was assigned duties as the note taker and timekeeper, responsibilities that included filling out checklists and writing down general observations. That person also had to make sure that someone was recording data (temperatures and interstage line appearance), providing times during sampling, and watching feed-tank volumes. Two operators were assigned the duty of sampling effluents and watching contactor operation. One collected the DW and FW effluent samples; the other, the EP and EW samples. Because of the dose associated with the EW sample (the concentrated cesium effluent), sampling of this stream was shared between all four operators. The fourth operator was in charge of preparing samples for scintillation counting, loading the samples into the counters, and collecting results from counted samples. These results were important, as they indicated how well the system was operating during the run. In practice, when an operator could not do a task for which he or she was responsible, one of the other operators filled in.

d. Sampling and Analysis

When the DF feed switched from cold (non-radioactive) to hot (radioactive), the run clock was started at t = 0. During the first hour, samples were collected at 5, 20, 40, and 60 min. For the next three hours, samples were collected every half hour. Then, for the following six hours, samples were collected hourly. After that, samples were collected every two hours.

Results were needed quickly to identify process upsets and determine if changes were needed. Qualitative real-time results were available from several sources. First, hydraulic upsets were detected by looking at the interstage lines. Large changes in the flow rate of any effluent during sampling would also have indicated a process upset. Radioactive dose rate was qualitatively used to verify that the ^{137}Cs was coming out in the aqueous strip effluent. The EW effluent samples were counted first to verify that the ^{137}Cs was being concentrated. Finally, the DW and EP samples were counted, initially for a short time, to verify cesium was being removed from both of these effluents.

Samples were counted for ^{137}Cs using liquid scintillation. The protocol for counting the beta decay of ^{137}Cs was to use a window of 2 to 400 keV. Three different liquid scintillation counters were used to analyze the process samples. A standard sample was counted on each of the three counters so that all results had the same basis. Samples were prepared for liquid scintillation counting by placing aliquots of the sample into a liquid scintillation cocktail. Slightly different sample preparations were used for the various samples. Organic samples were added to

a vial containing 10 mL of Ultima Gold™ scintillation cocktail. For the aqueous samples from the DW and FW streams, a 2 mL volume of sample was added to the cocktail vial with no other additions. This sample volume is high enough to ensure a good emulsion of the sample with the cocktail. For the lower volume samples used in the EW and DF samples, 200 µL of water were added to improve the emulsion of the sample with the cocktail. For preparation of stage samples taken at the end of the test, a matrix of sample volumes (established by trial and error) was used to ensure proper counting rates. Again, for small aqueous sample volumes, a small volume of water was added to the vial to improve the emulsion.

e. Feed Flow Rate Measurements

To measure the flow rate of the wash (FF), scrub (DS), and strip (EF) feeds, the three feed bottles were placed on digital balances as described in LEONARD-2001A. The flow rates were measured by noting the mass difference over time. In this way, an accurate flow rate measurement could be made in 5 min. The feed bottles were not changed as they were emptied. Instead, they were filled with the appropriate solution when necessary and the corresponding mass changes noted.

APPENDIX D

ADDITIONAL RESULTS

Further results from the multi-day CSSX flowsheet test (CS27) are given here. These results include measurements made before the test as well as measurements and observations made during the test and measurements made after the test. These details supplement the test results given in the body of this report.

1. Measurements before the Test

Before the multi-day CSSX flowsheet test (CS27) was carried out, batch-equilibrium D_{Cs} values were measured using the solutions and the volume ratios specified for the tests. The D_{Cs} results, given in Table D-1, agree fairly well with earlier data for the same alkaline-waste simulant, scrub feed, and strip feed [LEONARD-2001A]. The no-load D_{Cs} values in the extraction and scrub sections and the D_{Cs} values at low Cs concentrations in the strip section were calculated as given in [LEONARD-2001D].

Table D-1. D_{Cs} Values from Batch-Equilibrium Measurements before Test CS27

Section	Batch Test Number	O/A Volume Ratio	D_{Cs} at 25°C	Notes
Extraction	1	0.31	15.86	a
Extraction	2	0.31	14.67	a
Scrub	1	5.0	1.069	b
Scrub	2	5.0	1.102	b
Strip	1	5.0	0.1856	c
Strip	2	5.0	0.1397	c
Strip	3	5.0	0.099	c
Strip	4	5.0	0.082	c

^a Using these data, the no-load D_{Cs} value for the extraction section is 16.1 ± 0.6.
^b Using these data, the no-load D_{Cs} value for the scrub section is 1.14 ± 0.02.
^c Based on these data, the D_{Cs} value at low Cs concentrations in the strip section is 0.078 ± 0.005.

2. Measurements and Observations during the Test

To put the various measurements and observations in their proper time orientation, the key test times are given in Table D-2, starting with time zero. The test was continued until all SRS simulant was gone.

Table D-2. Key Times during Test CS27

Time date h:min	Total Elapsed Time min	Total Test Time min	Total Test Time d:h:min	Notes
3/12/01 14:15	0	0	0:00:00	Start of test CS27 (DF feed with 0.5 mCi/L of ^{137}Cs is started; see Notebook 1671-174).
3/12/01 14:20	5	5	0:00:05	Take first EW sample (measure of approach to steady state; see Notebook 1800-34).
3/12/01 14:40	25	25	0:00:25	Take second EW sample (measure of approach to steady state; see Notebook 1800-34).
3/13/01 6:11	956	956	0:15:56	Stop to replace DF-T pump (Notebook 1800-14).
3/13/01 7:11	1016	956	0:15:56	Start after replace DF-T pump (start at 7:10 am, allow 1 min to reach steady state; see Notebook 1800-15).
3/13/01 8:38	1103	1043	0:17:23	Increase the DF flow rate setting by 10% as it is running low by this much (Notebook 1800-15).
3/15/01 0:54	3519	3459	2:09:39	Start to add last 22 L of DF feed to 13 L already in DF drum (takes 2 h; see Notebook 1800-17).
3/15/01 11:15	4140	4080	2:20:00	Last samples taken before solids cause the test to be stopped (Notebook 1800-45).
3/15/01 12:35	4220	4160	2:21:20	Stop test to discover cause of high pH in EW and loss of stripping in EP (Notebook 1800-20).
3/16/01 4:22	5167	4160	2:21:20	Start test after recover from high pH in EW and loss of stripping in EP (Notebook 1800-29).
3/16/01 4:40	5185	4178	2:21:38	First EW sample taken 18 min after restart flowsheet conditions (measure of approach to steady state; see Notebook 1800-45).
3/16/01 5:00	5205	4198	2:21:58	Second EW sample taken 38 min after restart flowsheet conditions (measure of approach to steady state; see Notebook 1800-46).
3/16/01 5:25	5230	4223	2:22:23	Third EW sample taken 63 min after restart flowsheet conditions (Notebook 1800-46).
3/16/01 5:35	5240	4233	2:22:33	End of test CS27 (Notebook 1800-29).

During flowsheet test CS27, effluent flow rates were measured by taking timed effluent samples. For flow rates above 10 mL/min, the collection time was 1 min; for lower flow rates, 5 min. In addition, the effluent appearance was checked, any other-phase carryover was noted, the effluent temperature was measured, and a sample of the liquid was taken for later determination of its Cs concentration. Using pH paper, the pH was measured for the aqueous effluent from the strip and wash sections. These measurements and observations are summarized in Table D-3 for the DW and EW effluents and Table D-4 for the FW and EP effluents. The overall DW flow rate was 44.9 ± 3.2 mL/min. This flow rate breaks down to 41.7 ± 1.0 mL/min before 1043 min into the test (when the DF pump setting was increased) and 47.0 ± 2.3 mL/min after that time. The DS flow rate, measured using an electronic balance to weigh the DS feed tank (see LEONARD-2001A), was 2.94 ± 0.08 mL/min. The EW flow rate was 2.84 ± 0.32 mL/min. The FW flow rate was 3.00 ± 0.16 mL/min. The EP flow rate was 14.0 ± 1.1 mL/min.

Table D-3. Aqueous (DW) Raffinate and Aqueous Strip (EW) Effluents for Test CS27

Sample Number	Time, min	DW Flow Rate, mL/min	DW Temp, °C	DW Appearance[a]	DW Other-Phase Carryover, %	EW Flow Rate, mL/min	EW Temp, °C	EW Appearance[a]	EW Other-Phase Carryover, %	pH for EW
1	5	43.2	25.8	Clr, yel	<0.5	3.30	28.8	Sl cldy	<1.0	4
2	20	43.0	26.2	Hazy, yel	<0.5	2.80	29.4	Sl cldy	<1.0	4
3	40	42.3	26.9	Hazy, yel	<0.5	2.50	29.8	Sl cldy	<1.0	3
4	60	43.0	26.4	Hazy	<0.5	2.90	29.8	Sl cldy	<1.0	4
5	90	43.0	26.1	Hazy	<0.5	3.10	30.0	Sl cldy	<1.0	3
6	120	41.0	26.0	Clr, yel	<0.5	2.80	30.1	Sl cldy	<1.0	2.75
7	150	40.2	26.6	Clr	<0.5	2.96	30.0	Hazy	<1.0	4
8	180	40.0	26.4	Clr	<0.5	2.70	30.0	Hazy	<1.0	3.5
9	210	42.0	26.0	Clr	<0.5	2.90	29.7	Hazy	<1.0	3.5
10	240	43.0	26.3	Clr	<0.5	2.85	29.7	Hazy	<1.0	4
11	300	41.0	26.0	Clr	<0.5	2.86	30.0	Hazy	<1.0	3.5
12	360	41.0	26.3	Clr, yel	<0.5	2.80	30.0	Hazy	<1.0	3.5
13	420	41.0	26.0	Clr	<0.5	2.64	29.4	Hazy	<1.0	4
14	480	41.5	26.0	Clr	<0.5	3.10	30.0	Hazy	<1.0	3.5
15	540	41.0	26.1	Clr	<0.5	3.20	30.2	Hazy	<1.0	3.5
16	600	40.5	25.5	Clr	<0.5	2.80	29.2	Hazy	<1.0	3
17	720	42.0	25.4	Clr, yel	<0.5	2.60	29.6	Hazy	-	3
18	840	42.0	25.7	Clr, yel	<0.5	3.00	29.6	Hazy	<1.0	3
19	960	42.0	23.8	Clr, yel	<0.5	2.30	27.2	Clr	<1.0	3
20	1080	45.0	25.6	Clr, yel	<0.5	2.68	28.5	Hazy	<1.0	4.5
21	1200	45.0	25.6	Hazy, yel	<0.5	3.00	29.1	Hazy	<1.0	3
22	1320	45.7	26.0	Clr, yel	<0.5	3.20	29.3	Clr, col	<1.0	3
23	1440	47.0	25.9	Clr, yel	<0.5	2.30	29.0	Hazy, col	<1.0	3
24	1560	46.0	26.2	Clr	<0.5	3.04	30.1	Hazy	<1.0	3.5
25	1680	45.5	26.0	Clr	<0.5	2.90	29.8	Hazy	<1.0	4
26	1800	45.0	25.9	Clr	<0.5	2.90	34.0	Hazy	<1.0	3.5
27	1920	48.5	26.1	Clr	<0.5	2.70	29.4	Hazy	<1.0	3.5
28	2040	45.0	25.9	Clr, yel	<0.5	3.40	25.9	Hazy	<1.0	3.5
29	2160	45.0	25.8	Clr, yel	<0.5	2.60	29.6	Hazy	<1.0	3
30	2280	48.0	25.5	Clr, yel	<0.5	2.30	29.8	Cldy	<1.0	4
31	2400	45.0	23.2	Clr	<0.5	2.60	30.1	Hazy	-	3
32	2520	46.2	25.0	Clr, yel	<0.5	2.74	29.7	Hazy	<1.0	3
33	2640	45.0	25.2	Hazy	<0.5	3.10	29.9	Sl Hazy	<1.0	3.5
34	2760	46.8	25.1	Hazy, yel	<0.5	3.24	30.0	Hazy	<1.0	3
35	2880	45.0	25.5	Clr, yel	<0.5	2.00	29.0	Hazy, col	<1.0	3
36	3000	47.0	25.6	Clr	<0.5	2.80	30.1	Hazy	<1.0	3.5
37	3120	45.5	25.5	Clr	<0.5	3.26	30.0	Hazy	<1.0	3.5
38	3240	46.0	25.4	Clr	<0.5	3.40	30.3	Hazy	<1.0	4
39	3360	50.0	25.9	Clr	<0.5	2.90	30.3	Hazy	<1.0	3.5
40	3480	45.0	25.7	Clr, yel	<0.5	3.00	30.3	Hazy	<1.0	3
41	3600	47.0	25.4	Clr, yel	<0.5	2.40	29.9	Hazy	<1.0	3
42	3720	48.5	25.6	Clr, yel	<0.5	2.40	29.6	Cldy	<1.0	3
43	3840	53.0	25.7	Clr, yel	<0.5	2.80	30.0	Cldy	<1.0	3
44	3960	48.0	25.6	Clr, yel	<0.5	2.88	30.1	Hazy	<1.0	3
45	4080	47.0	25.3	Sl cldy	<0.5	2.60	30.1	Hazy	<1.0	3
46	4178	52.0	25.6	Clr, yel	<0.5	2.60	29.4	Clr	8	3
47	4198	50.0	25.7	Clr, yel	<0.5	3.00	28.7	Hazy	7	4
48	4223	51.0	25.4	Clr, yel	<0.5	3.50	28.1	Hazy	23	5

[a] Appearance Code: cldy= cloudy, clr= clear, col= colorless, crys= crystal, hazy= hazy, sl= slight, v= very, and yel= yellow

Table D-4. Aqueous (FW) Wash and Organic Strip (EP) Effluents for Test CS27

Sample Number	Time, min	FW Flow Rate, mL/min	FW Temp, °C	FW Appearance[a]	FW Other-Phase Carryover, %	pH for FW	EP Flow Rate, mL/min	EP Temp, °C	EP Appearance[a]	EP Other-Phase Carryover, %
1	5	3.00	24.8	V clr	<1.0	12+	14.0	30.3	Crys clr	<0.01
2	20	2.88	25.3	V clr	<1.0	12+	15.5	29.6	Crys clr	<0.01
3	40	2.84	25.0	V clr, col	<1.0	12+	14.4	30.1	Crys clr	<0.01
4	60	3.06	25.2	V clr, col	<1.0	12+	15.0	30.0	Crys clr	0.2
5	90	2.66	25.3	Clr	<1.0	12+	15.0	29.0	Clr, col	<0.01
6	120	3.00	25.1	V clr	<1.0	12+	14.0	27.9	Crys clr	0.06
7	150	2.80	24.8	Clr	<1.0	12+	13.0	29.8	Clr	0.06
8	180	2.90	25.2	Clr	<1.0	12+	13.5	29.8	Clr	0.2
9	210	2.80	25.1	Clr	<1.0	12+	14.0	29.5	Clr	0.08
10	240	3.04	25.2	Clr	<1.0	12+	13.8	29.8	Clr	0.7
11	300	2.90	25.1	Clr	<1.0	12+	14.8	29.6	Clr	0.7
12	360	3.00	25.5	Clr	<1.0	11	15.0	30.1	Clr	0.7
13	420	3.00	25.0	Clr	<1.0	12+	14.0	29.6	Clr	1.4
14	480	2.80	27.0	Clr	<1.0	12+	14.5	30.1	Clr	<0.01
15	540	3.06	24.6	Clr	<1.0	11	14.0	30.1	Clr	0.7
16	600	2.70	24.8	Clr	<1.0	12	14.5	29.6	Clr	<0.01
17	720	2.90	24.0	Clr	-	12	13.0	29.1	Clr	<0.01
18	840	3.00	24.5	Clr, col	<1.0	11	14.0	29.0	Clr	3.6
19	960	2.90	23.4	-	<1.0	12	13.0	27.1	Clr	3.8
20	1080	2.72	25.5	Crys clr	<1.0	11	13.0	28.6	V clr, col	<0.01
21	1200	3.06	24.7	Clr	<1.0	12+	13.5	28.1	V clr, col	0.06
22	1320	3.00	25.9	Crys clr, col	<1.0	11	14.0	28.5	Crys clr	<0.01
23	1440	3.04	25.2	Clr	<1.0	12+	14.2	28.8	V clr, col	<0.01
24	1560	3.30	25.0	Clr	<1.0	12+	9.0	29.8	Clr	2.2
25	1680	2.80	24.9	Clr	<1.0	12+	13.8	30.1	Clr	0.7
26	1800	2.80	26.7	Clr	<1.0	12+	14.0	30.2	Clr	1.4
27	1920	2.96	25.0	Clr	<1.0	12+	14.8	28.9	Clr	0.3
28	2040	2.80	24.6	Clr	<1.0	12	15.5	28.6	Clr	0.6
29	2160	2.90	24.0	Clr	<1.0	11	15.0	30.0	Clr, col	<0.01
30	2280	3.20	25.6	Clr	<1.0	12	15.5	29.6	Clr	<0.01
31	2400	3.00	24.7	Clr	-	12	14.0	29.2	Clr	0.1
32	2520	3.10	25.0	Clr	<1.0	12	14.0	29.4	Clr	<0.01
33	2640	3.08	24.9	Clr	<1.0	12	14.5	29.4	Clr	0.01
34	2760	3.00	25.6	Clr	<1.0	12+	14.4	29.9	Clr	0.03
35	2880	3.00	25.0	Crys clr, col	<1.0	11	13.0	28.3	Crys clr	0.05
36	3000	3.10	25.3	Clr	<1.0	12+	14.0	29.1	Clr	1.4
37	3120	3.20	26.8	Clr	<1.0	12+	15.0	29.4	Clr	3.3
38	3240	2.90	24.9	Clr	<1.0	12+	13.0	28.2	Clr	1.5
39	3360	3.00	24.7	Clr	<1.0	12+	14.0	29.8	Clr	0.4
40	3480	3.00	25.8	Clr	<1.0	12	13.0	29.6	Clr	<0.01
41	3600	3.40	25.0	Cldy	6	11	12.0	29.4	Clr	<0.01
42	3720	3.20	25.7	Cldy	9	11	16.0	29.8	Clr	<0.01
43	3840	3.20	25.5	Cldy	9	11	15.0	29.5	Clr	7
44	3960	3.26	25.6	Clr	<1.0	12	12.0	30.5	Clr	8
45	4080	3.00	24.3	Clr	<1.0	12	14.2	29.5	Clr	<1.0
46	4178	3.20	25.1	Clr	<1.0	12	14.0	27.2	Clr	<1.0
47	4198	3.20	25.2	Clr	<1.0	12	14.0	30.1	Clr	<1.0
48	4223	3.20	25.4	Clr, yel	<1.0	12	14.0	26.8	Clr	<1.0

[a] Appearance Code: cldy= cloudy, clr= clear, col= colorless, crys= crystal, hazy= hazy, sl= slight, v= very, and yel= yellow

Other conditions measured during flowsheet test CS27 were the temperature of selected contactor stages and the level of the liquid flowing through selected interstage lines. The temperatures of selected contactor stages, which were measured using digital thermometers taped to the contactor block, are given in Table D-5 over the period of the test. They show that all contactor temperatures were higher than the surrounding (glovebox) temperature. The stages cooled by the chiller bar, stages 1-15, had the lowest block temperatures ranging from 24.5 to 26.6°C. The stages away from the chiller bar and with no external feeds, stages 20-30, had the highest block temperatures, ranging from 35.1 to 38.2°C. The level of the liquid flowing through selected interstage lines was estimated by the operators. The results, summarized in Table D-6, include only those lines that were 50% full or more at some point during the multi-day test. All interstage lines looked very good, having a liquid level of 20% or less some of the time. The highest liquid levels on the organic side were seen in the interstage line going from stage 9 to 10. The highest liquid levels on the aqueous side were seen in the interstage line going from stage 30 to 29. Four other organic interstage lines (5 to 6, 6 to 7, 13 to 14, and 22 to 23) were generally low, but at some point exceeded 65%. No interstage line was greater than 50% until we stopped to replace the DF-T pump. When the contactor was restarted after stopping to clean out the solids, only two interstage lines exceeded a liquid level of 50%, 22 to 23 on the organic side and 30 to 29 on the aqueous side.

Table D-5. Block Temperatures for Selected Contactor Stages during Test CS27

Total Test Time, min		Stage 1	Stage 3	Stage 8	Stage 13	Stage 15	Stage 16	Stage 17	Stage 18	Stage 20	Stage 25	Stage 30	Stage 32	Stage 33	Notes
							Block Temperature, °C								
125	22.2	26.6	26.3	25.4	26.4	24.8	31.6	32.4	34.6	35.1	37.2	37.8	34.0	29.2	
566	22.1	26.3	26.5	25.4	26.2	24.6	31.7	32.6	34.8	36.3	37.4	37.9	33.0	29.0	
876	21.9	26.1	26.1	25.1	26.1	24.6	31.6	32.4	34.6	36.3	37.3	37.7	33.8	28.5	a
1110	22.1	26.1	26.2	25.2	25.9	24.5	31.4	32.2	34.1	35.4	36.6	36.8	33.3	28.1	b
1420	22.1	26.5	26.6	25.4	26.1	24.6	31.7	32.3	34.0	36.2	37.6	37.9	33.7	28.8	
1834	22.2	26.1	26.2	25.3	26.0	24.5	31.5	32.6	34.8	35.6	36.8	37.7	33.8	28.6	
2250	22.3	26.3	26.2	26.0	26.1	24.6	31.8	32.9	35.0	37.4	36.9	38.0	33.8	29.0	
2667	22.5	26.4	26.3	25.4	26.2	24.8	31.6	32.8	34.8	37.6	36.8	37.9	33.9	28.9	
3150	22.5	26.5	26.4	25.5	26.3	24.8	31.1	32.5	34.8	37.7	37.2	38.1	33.9	29.1	
3690	22.3	26.5	26.4	25.7	26.4	24.9	30.7	32.6	34.9	37.4	37.3	37.4	34.0	29.1	
4050	22.5	26.5	26.4	25.6	26.3	25.2	30.3	32.5	34.8	38.2	36.9	37.5	33.9	29.3	
Avg:	22.2	26.4	26.3	25.5	26.2	24.7	31.4	32.5	34.7	36.7	37.1	37.7	33.7	28.9	
Std Dev:	0.2	0.2	0.1	0.2	0.2	0.2	0.5	0.2	0.3	1.1	0.3	0.4	0.3	0.4	

a) Before first stop
b) After first sto|

Table D-6. Liquid Levels for Selected Interstage Lines during Test CS27

Total Test Time, mir	A(30 to 29)	A(25 to 24)	O(6 to 7)	O(9 to 10)	O(11 to 12)	O(13 to 14)	O(23 to 24)	O(26 to 27)	O(27 to 28)	Notes
74	20	50	30	30	50	20	50	50	30	
182	10	30	10	10	20	10	20	10	15	
253	10	20	15	10	30	10	30	10	15	
315	25	25	25	25	30	20	30	20	30	
421	5	40	15	20	30	10	30	10	20	
540	10	30	20	20	40	10	20	10	15	
659	10	40	30	30	45	30	45	10	45	
780	20	30	30	40	45	30	35	30	40	
855	20	35	30	30	40	30	40	10	20	
903	30	40	35	30	40	30	40	25	30	
1090	60	35	30	60	40	20	40	40	30	a
1140	60	40	20	65	40	20	50	50	30	
1275	60	40	20	65	40	20	30	50	30	
1380	65	30	30	65	40	20	40	45	30	
1450	65	40	25	65	45	20	40	45	40	
1560	65	10	20	80	45	10	30	30	20	
1620	70	40	20	90	45	15	40	60	20	
1740	70	30	20	85	20	10	45	50	30	
1800	70	45	20	80	45	15	45	50	20	
1860	80	40	25	80	50	15	50	60	40	
1920	70	50	30	90	50	30	40	60	40	
2063	60	40	25	60	45	15	45	45	35	
2160	60	30	25	70	45	30	50	50	50	
2287	60	40	20	70	40	30	50	40	30	
2340	50	30	20	65	40	40	50	50	40	
2461	60	40	20	70	50	40	50	50	40	
2584	60	40	20	70	50	40	40	50	40	
2650	60	40	20	70	50	40	40	50	40	
2780	60	40	20	80	50	50	40	50	40	
2890	60	40	20	88	55	50	45	55	40	
2950	60	15	30	100	50	50	50	50	40	
3060	60	40	30	100	60	45	45	50	40	
3180	60	40	30	100	60	50	50	55	45	
3240	60	45	30	100	60	55	45	60	40	
3360	60	40	35	100	65	60	45	55	40	
3480	60	40	30	100	60	50	50	50	40	
3540	60	40	40	90	50	50	40	50	45	
3600	60	30	25	10	50	50	50	50	40	
3718	60	40	30	20	50	50	50	50	40	
3840	50	30	20	10	60	50	50	50	40	
3973	60	40	20	10	60	60	40	60	50	
4020	60	40	30	10	60	80	40	50	40	
4120	70	40	85	10	60	30	50	50	40	b
4188	70	30	45	40	40	30	30	40	40	c

a) First look at interstage lines after stopping to replacing pump DF-

b) Other high liquid levels are O(4 to 5), 60%; O(5 to 6), 90%

c) First look after remove solids. Other high liquid level is O(22 to 23), 70%

3. Measurements after the Test

After test CS27, the Cs concentration was measured for all available effluent samples. The results are listed in Table D-7 along with the DF concentration that is the basis value and the calculated decontamination, concentration, and stripping factors. The average Cs concentration

for the DW raffinate is 1.00E-09 ± 0.33E-09 $\underline{M}$; for the EW effluent, 2.11E-03 ± 0.33E-03 $\underline{M}$; for the FW effluent, 7.0E-10 ± 8.2E-10 $\underline{M}$; and for the EP effluent, 1.32E-09 ± 0.76E-09 $\underline{M}$.

Table D-7. Effluent Concentrations during Test CS27

Sample Number	Time, min	Cs Conc in Aqueous (DW) Raffinate, M	Cs Conc in Aqueous Strip (EW) Effluent, M	Cs Conc in Aqueous Wash (FW) Effluent, M	Cs Conc in Organic Strip (EP) Effluent, M	Cs Conc in Aqueous (DF) Feed, M	Decon Factor, [DF]/[DW]	Conc Factor, [EW]/ [DF]	Stripping Factor, [DF]/[EP]
1	5	5.04E-10	6.12E-04	4.7E-10	2.39E-10	1.40E-04	277,673	4.4	586,448
2	20	5.14E-10	1.84E-03	3.3E-10	6.10E-10	1.40E-04	272,195	13.1	229,337
3	40	7.66E-10	1.93E-03	2.3E-10	8.70E-10	1.40E-04	182,740	13.8	160,986
4	60	8.61E-10	2.00E-03	2.3E-10	2.53E-10	1.40E-04	162,675	14.3	553,389
5	90	6.27E-10	1.90E-03	5.8E-11	7.51E-10	1.40E-04	223,431	13.6	186,366
6	120	9.58E-10	1.90E-03	8.8E-11	1.08E-09	1.40E-04	146,192	13.6	129,240
7	150	7.77E-10	1.87E-03	2.4E-10	1.96E-09	1.40E-04	180,204	13.3	71,430
8	180	7.95E-10	1.97E-03	2.2E-10	9.23E-10	1.40E-04	176,177	14.1	151,634
9	210	8.66E-10	1.93E-03	3.2E-10	2.96E-09	1.40E-04	161,722	13.8	47,353
10	240	8.31E-10	1.98E-03	4.8E-10	2.65E-09	1.40E-04	168,383	14.1	52,922
11	300	8.10E-10	2.08E-03	1.5E-09	1.74E-09	1.40E-04	172,866	14.9	80,517
12	360	1.04E-09	1.93E-03	2.5E-10	1.02E-09	1.40E-04	135,264	13.8	136,800
13	420	9.18E-10	1.84E-03	3.6E-10	2.69E-09	1.40E-04	152,452	13.2	52,114
14	480	8.24E-10	1.93E-03	1.3E-10	1.11E-09	1.40E-04	169,808	13.8	126,160
15	540	1.17E-09	1.88E-03	4.0E-10	1.37E-09	1.40E-04	119,389	13.4	101,966
16	600	9.09E-10	1.97E-03	N/A	7.41E-10	1.40E-04	153,940	14.0	188,817
17	720	8.67E-10	1.96E-03	N/A	1.60E-09	1.40E-04	161,485	14.0	87,349
18	840	6.19E-10	1.97E-03	3.6E-10	7.23E-10	1.40E-04	226,179	14.0	193,517
19	960	8.35E-10	1.91E-03	3.7E-10	1.38E-09	1.40E-04	167,616	13.6	101,325
20	1080	9.25E-10	2.20E-03	4.7E-10	8.87E-10	1.40E-04	151,302	15.7	157,821
21	1200	7.92E-10	2.08E-03	5.4E-09	1.18E-09	1.40E-04	176,742	14.9	118,407
22	1320	1.04E-09	2.22E-03	1.1E-10	9.76E-10	1.40E-04	134,358	15.9	143,505
23	1440	1.11E-09	2.12E-03	1.3E-09	8.32E-10	1.40E-04	126,143	15.2	168,370
24	1560	1.10E-09	1.96E-03	7.4E-10	1.50E-09	1.40E-04	127,307	14.0	93,334
25	1680	1.26E-09	2.22E-03	4.6E-10	9.22E-10	1.40E-04	111,434	15.8	151,867
26	1800	9.01E-10	2.21E-03	4.3E-10	1.26E-09	1.40E-04	155,349	15.8	110,948
27	1920	1.32E-09	2.21E-03	2.5E-10	1.78E-09	1.40E-04	106,079	15.8	78,512
28	2040	2.09E-09	2.24E-03	4.9E-10	9.55E-10	1.40E-04	66,955	16.0	146,585
29	2160	1.18E-09	2.11E-03	3.4E-10	7.26E-10	1.40E-04	118,492	15.1	192,855
30	2280	9.77E-10	2.10E-03	1.5E-09	7.58E-10	1.40E-04	143,344	15.0	184,717
31	2400	8.04E-10	2.18E-03	6.2E-10	5.72E-10	1.40E-04	174,093	15.6	244,785
32	2520	1.43E-09	2.15E-03	6.0E-10	2.39E-09	1.40E-04	98,111	15.3	58,695
33	2640	8.26E-10	2.16E-03	9.4E-10	7.12E-10	1.40E-04	169,417	15.4	196,554
34	2760	7.30E-10	2.15E-03	1.0E-09	8.09E-10	1.40E-04	191,790	15.3	173,022
35	2880	1.05E-09	2.16E-03	5.2E-10	8.09E-10	1.40E-04	133,223	15.4	173,022
36	3000	1.31E-09	2.19E-03	3.5E-10	1.05E-09	1.40E-04	106,875	15.6	132,929
37	3120	1.53E-09	2.25E-03	3.5E-10	1.11E-09	1.40E-04	91,655	16.0	125,808
38	3240	9.43E-10	2.18E-03	5.4E-10	1.56E-09	1.40E-04	148,404	15.6	89,605
39	3360	8.20E-10	2.31E-03	9.5E-10	1.59E-09	1.40E-04	170,793	16.5	88,065
40	3480	9.88E-10	2.39E-03	8.6E-10	1.35E-09	1.50E-04	151,836	15.9	111,313
41	3600	9.99E-10	2.62E-03	9.0E-10	9.73E-10	1.60E-04	160,149	16.4	164,443
42	3720	1.12E-09	2.83E-03	1.1E-10	1.68E-09	1.70E-04	151,181	16.7	101,038
43	3840	9.92E-10	2.77E-03	2.5E-10	1.97E-09	1.70E-04	171,347	16.3	86,431
44	3960	1.81E-09	2.80E-03	1.2E-09	1.73E-09	1.70E-04	93,715	16.5	98,266
45	4080	2.04E-09	2.03E-03	8.1E-10	4.49E-09	1.70E-04	83,165	11.9	37,903
46	4178	9.13E-10	2.09E-03	1.9E-09	1.63E-09	1.70E-04	186,219	12.3	104,234
47	4198	8.51E-10	2.59E-03	7.1E-10	1.37E-09	1.70E-04	199,678	15.2	124,526
48	4223	7.33E-10	2.53E-03	9.8E-10	1.25E-09	1.70E-04	232,077	14.9	136,401

An overall Cs material balance for test CS27 was calculated using the flow rates and Cs concentrations for the incoming and outgoing process streams. The results, summarized in Table D-8, show that 98.6% of the Cs was recovered.

Table D-8. Overall Material Balance for Test CS27

Stream Identity	Stream In or Out	Average Stream Flow Rate, mL/min	Average Stream Cs Conc, $\underline{M}$	Cs Flow, m$\underline{M}$/min
DF	In	41.99	1.45E-04	0.006087907
DS	In	2.94	0	0.000000000
EF	In	2.84	0	0.000000000
FF	In	3.00	0	0.000000000
FX	In	13.99	7.00E-10	0.000000010
			Total Cs Flow In, m$\underline{M}$/min: 0.006087917	
DW	Out	44.93	1.00E-09	0.000000045
EW	Out	2.84	2.11E-03	0.006004309
EP	Out	13.99	1.32E-09	0.000000019
FW	Out	3.00	7.00E-10	0.000000002
			Total Cs Flow Out, m$\underline{M}$/min: 0.006004375	
			Amount of Cs Recovered: 98.6%	

Shortly after test CS27 was completed, the aqueous and organic liquids in each stage were drained and collected in 60 mL polyethylene bottles. The stage samples were later equilibrated by shaking for 15-20 s in the collection bottle. Immediately after equilibration, the sample temperature was measured. Then the two phases were separated and each phase was analyzed for its cesium concentration. From these concentrations, the apparent D_{Cs} value can be calculated for each stage. These concentrations, temperatures, and D_{Cs} values are given in Table D-9. Some of the concentrations and D_{Cs} values in the scrub, strip, and wash sections are higher than expected based on the values seen in Table D-1. This is attributed either to residual high concentrations of cesium in the stage drain lines or to residual high concentrations of either acid or base in the drain lines. These high concentrations could have been left by either the upset conditions caused by the solids in the contactor or the subsequent procedure used to recover from this upset. To get an idea of how well the stage samples represent the actual stage conditions, the cesium concentration in the each effluent stream is also listed. This comparison shows that the sample from stage 32 was definitely contaminated with cesium, probably from the stage drain line. The high Cs concentration for the organic phase from stage 33, but not the aqueous phase, suggests that the organic phase was contaminated after the stage sample was equilibrated and the two phases separated.

Table D-9. Cesium Concentration in Equilibrated Stage Samples

Stage Number	Cs Conc. (Aqueous) after Stage Sample Equilibrated, M	Cs Conc. (Organic) after Stage Sample Equilibrated, M	Equil. Temp, °C	D_{Cs}	Cs Conc. (Aqueous) in Last Effluent Sample, M	Cs Conc. (Organic) in Last Effluent Sample, M
1	5.32E-10	7.16E-09	23.3	13.5	7.33E-10	
2	1.25E-09	3.10E-08	22.4	24.7		
3	8.37E-10	1.03E-08	22.5	12.3		
4	9.50E-10	1.46E-08	22.9	15.3		
5	7.64E-10	9.86E-09	22.5	12.9		
6	9.86E-10	1.26E-08	23.0	12.8		
7	2.41E-09	2.73E-08	22.4	11.3		
8	3.22E-09	6.68E-08	22.5	20.7		
9	6.51E-09	1.69E-07	22.7	26.0		
10	2.74E-08	7.14E-07	22.3	26.0		
11	1.03E-07	2.49E-06	22.6	24.2		
12	3.51E-07	9.16E-06	22.8	26.1		
13	1.39E-06	3.72E-05	22.6	26.7		
14	4.53E-06	1.19E-04	22.8	26.2		
15	2.02E-05	4.90E-04	22.9	24.2		
16	5.51E-05	7.30E-04	21.6	13.2		
17	4.79E-05	5.95E-04	22.8	12.4		
18	1.19E-03	5.66E-04	22.0	0.48	2.53E-03	
19	1.03E-03	3.72E-04	22.1	0.36		
20	6.11E-04	1.71E-04	22.4	0.28		
21	3.34E-04	8.70E-05	22.1	0.26		
22	1.09E-04	2.68E-05	21.6	0.25		
23	7.32E-05	1.49E-05	21.8	0.20		
24	4.37E-05	8.06E-06	21.7	0.184		
25	4.01E-05	6.48E-06	21.7	0.162		
26	2.52E-05	4.05E-06	21.5	0.160		
27	5.03E-07	2.53E-07	21.7	0.50		
28	8.85E-07	1.07E-07	22.0	0.121		
29	2.80E-05	3.27E-06	21.6	0.117		
30	4.54E-07	4.49E-08	22.6	0.099		
31	6.40E-08	6.50E-09	22.4	0.102		
32	2.92E-05	2.27E-06	22.3	0.078		1.25E-09
33	9.65E-10	1.63E-07	23.0	169	9.81E-10	

The concentration of Cs in both phases of each stage at the end of test CS27 can be calculated using the SASSE workbook [LEONARD-1994]. The procedure for doing this for the CSSX process at a given temperature is given in LEONARD-2001D. The effect of temperature on the D_{Cs} values for the CSSX solvent was added by using information from BONNESEN-2000. The enthalpy (H) for this cesium extraction reaction is obtained from the slope of each curve in Fig. 5 of BONNESEN-2000. It is 42.8 kJ/mol for the extraction section, 61.8 kJ/mol for the scrub section, and 62.5 kJ/mol for the strip section. This means that, going from 20 to 30°C,

D_{Cs} will drop by 1.79x in the extraction section, 2.31x in the scrub section, and 2.33x in the strip section. The equation for D_{Cs} that gives D_1 at T_1 knowing D_0 at T_0 is

$$D\_1 = D\_0 \bullet \exp[(H/R) \bullet \{(1/T\_1) - (1/T\_0)\}] \qquad (D\text{-}1)$$

where T is the absolute temperature in K, H is as given above in kJ/mol, and R is 0.0083144 kJ/(mol•K). Using the temperature data above, the liquids flowing through the stages were estimated to be 25.7°C in stages 1-13, 24.6°C in stage 14, 24.7°C in stage 15, 27.5°C in stages 16-17, 32.1°C in stage 18, 34.6°C in stage 19, 37.2°C in stages 20-30, 33.2°C in stage 31, and 29.3°C in stage 32. Stage 33 was omitted from the model. In the SASSE calculations, the effect of diluent loss is included by increasing the no-load D_{Cs} values for the extraction and scrub sections and the D_{Cs} value at low concentrations in the strip section (see Table D-1) by 31%. Using the SASSE model with solvent recycle and the stage-to-stage concentration profile for Cs given in Table D-9, the best fit of the model to the data was found to be for a stage efficiency of 89% in the extraction section and 83% in the strip section. The model results are plotted along with the measured Cs concentrations in Fig. D-1. Except for high Cs concentrations at some stages in the strip section, probably caused by contamination in the stage drain lines, there is reasonable agreement between the data and the model.

For aqueous strip (EW) effluent, the concentrations of Na, K, and Al were measured using inductively coupled plasma/atomic emission spectrometry (ICP/AES). These are the major components in the EW effluent along with the cesium. The results, given in Table D-10 along with the corresponding Cs concentrations, show that cesium was the major cation in the EW effluent throughout test CS27. The total Cs concentration near the end of the test increased 20% because the ^{137}Cs spike for the final 22 L of DF feed also had some non-radioactive cesium. This spike was obtained from EW effluents of earlier CSSX flowsheet tests by evaporating the water and dissolving the residual solids. The mole fraction of Na relative to Cs in the EW effluent was 0.23 ± 0.10; of K relative to Cs, 0.13 ± 0.12; of Al relative to Cs, 0.003 ± 0.001. Relative to its concentration in the DF feed, the Na concentration in the EW effluent was 13,000 times lower; the K concentration, 98 (20 to 226) times lower; the Al concentration, 55,000 times lower. This shows that, while the distribution ratio for K is much less than 1.0, it is large enough to allow some K to be extracted. If more scrub stages were used, the concentration of K in the EW effluent would be reduced even further.
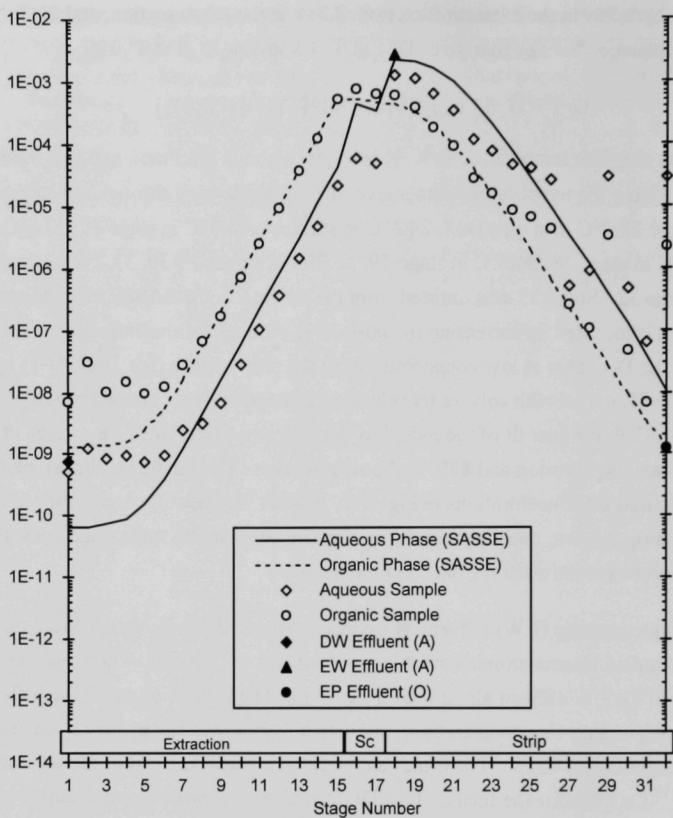
Fig. D-1. Cs Concentration Profile at the End of Test CS27

Table D-10. Concentration of Major Cations in the Aqueous Strip Effluent for Test CS27

Time into Test CS27, min	Component	Concentration in Extraction (DF) Feed, mM	Concentration in Aqueous (EW) Strip Effluent, mM	Amount that Component in DF Extracted into EW Effluent Relative to Cs, %
90 ± 30	Cs	0.14	2.1	Not applicable
	Na	5,600	0.61	0.0007
	Al	280	0.006	0.0001
	K	15	0.23	0.10
1560 ± 120	Cs	0.14	2.1	Not applicable
	Na	5,600	0.39	0.0005
	Al	280	<0.004	0.0001
	K	15	0.18	0.08
3900 ± 180	Cs	0.17	2.6	Not applicable
	Na	5,600	0.83	0.0010
	Al	280	0.004	0.0001
	K	15	0.77	0.34
4200 ± 23	Cs	0.17	2.6	Not applicable
	Na	5,600	0.26	0.0003
	Al	280	0.011	0.0003
	K	15	0.07	0.03

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