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Scale-up of Caustic-Side Solvent Extraction Process for Removal of Cesium at Savannah River Site

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SUMMARY

In 2004, the Department of Energy (DOE) directed Westinghouse Savannah River Company (WSRC) to develop a Caustic-Side Solvent Extraction (CSSX) process at the Savannah River Site (SRS) capable of removing cesium from 1 million gallons a year of dissolved salt solution. This facility would provide interim processing for cesium containing salt solution until the Salt Waste Processing Facility (SWPF) comes on-line.

The DOE design inputs[1] were to utilize contactors similar in design to those to be used in the SWPF, assume class C waste with less than 0.5 Ci/gal Cs-137, achieve a Decontamination Factor (DF) greater than 12, include the ability to clean the contactors in place, and assume an operating life of three years. WSRC embarked on a design, test, and build program to achieve these criteria as described in the following text. All DOE design criteria have been met or exceeded by WSRC.

INITIAL PROCESS DEVELOPMENT

The initial development of the process was performed jointly at Argonne National Laboratory (ANL) and Oak Ridge National Laboratory (ORNL). Development of the solvent and the unique extractant was performed at ORNL and initial process flow sheet development and testing was performed at ANL. ANL used 2-cm centrifugal contactors for the initial testing[2], which was successful. An optimized solvent was subsequently developed by ORNL to decrease the probability of third phase formation and allow an increase in the extractant concentration[3].

Further flowsheet testing was performed at the Savannah River National Laboratory (SRNL) using real waste from the SRS tank farms. The testing utilized 2-cm contactors, 1 extraction contactors, 2 scrub contactors, 16 strip contactors, and 2 wash contactors. Using real waste containing Cs-137, the process was run in the SRNL Hot Cells. The results[4] demonstrated a DF >40,000 and a CF of 12-15.

CONTACTOR SCALE-UP TESTING

Maximum salt solution flow during the Hot Cell testing at SRNL was 43 ml/min. Maximum salt flow for the MCU is 8.5 gpm, a factor of ~750 times greater. The DOE directed WSRC to utilize technology similar to that being used for the Salt Waste Processing Facility (SWPF). The SWPF was using CINC (Costner Industries Nevada Corporation) centrifugal contactors. WSRC made an exhaustive search of centrifugal contactor manufacturers and CINC was the only viable option. The SRS MCU process flow rates were to support:

- Salt Solution 3.5 – 8.5 gpm
- Scrub 0.23 – 0.57 gpm
- Strip 0.23 – 0.57 gpm
- Wash 0.23 – 0.57 gpm
- Solvent 1.17 – 2.83 gpm

CINC contactors were chosen based on the manufacturer's recommendations for throughput. Ten-inch rotor contactors, model V-10, were selected for the salt solution bank (manufacturer specification of 30 gpm maximum flow) and five-inch rotors contactors, model V-05 (6 gpm maximum).

CINC V-10 Contactor with Motor

A vendor was chosen to design, build, and test the 18-stage contactor process. The first step in the process was to test both the V-05 and V-10 contactors individually. These tests were to determine optimum rotor speed and weir size to minimize solvent carryover and maximize throughput. The tests were to also allow observation of general hydraulic behavior and identify any areas for improvement.

To facilitate the individual contactor testing, a test stand capable of holding both contactors while providing a stable foundation to evaluate vibration was constructed. The test stand would also allow for cold feeds to be centrally located and easily accessible. The contactors mounted in the test stand are shown in Figure 1.

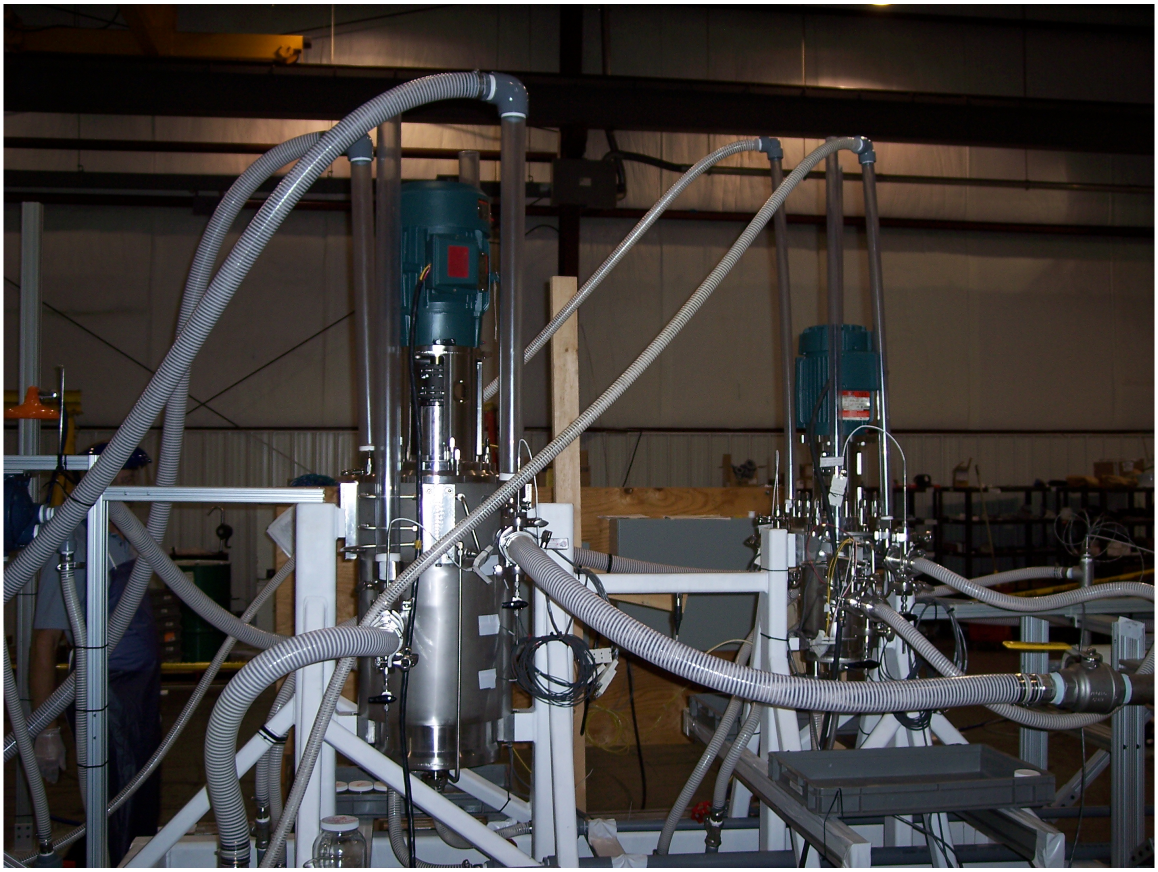


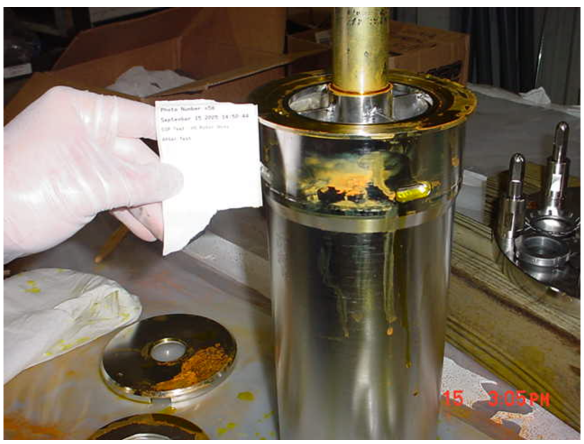
Figure 1

The most significant hydraulic issue that arose during Hot Cell testing with the 2-cm contactors was flow between stages. Due to the low flow rate, slope of the interstage lines was critical to obtain adequate flow. The inertial forces far exceed surface tension forces in five and ten inch rotor contactors, so obtaining adequate slope between contactors was not an issue.

The individual contactor testing involved selecting weir sizes and rotor speeds that provided the least organic carryover. Salt simulant that represented an average SRS feed and had been used for all previous CSSX was testing was utilized, along with actual solvent minus the extractant (hydraulically insignificant). Organic carryover >1000 ppm was obtained from both units, which was well above the design criteria. Further investigation revealed that the organic weir diameter in the V-05 was incorrectly specified. Using the corrected organic weir diameter a more appropriate heavy phase weir size was selected that reduced organic carryover.

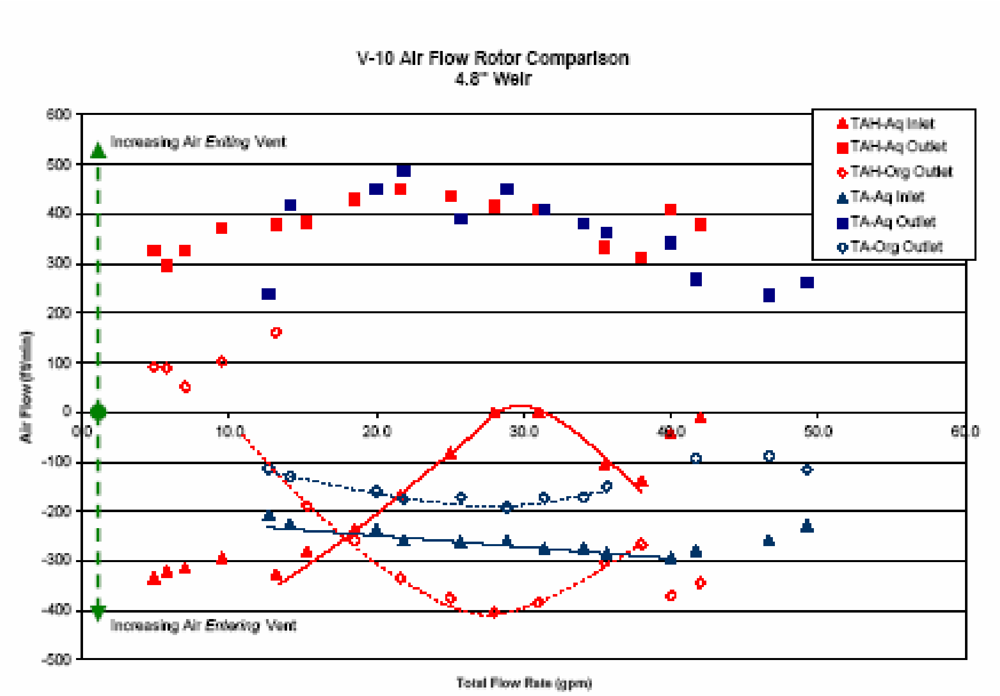
Hands-on maintenance for the contactors was a criteria due to the short three year operating life. CINC contactors have a Clean-in-Place (CIP) system as a standard feature; however, it was focused on cleaning the internal rotor and heavy phase underflow region. At SRS Cs-137 is the primary radionuclide of concern from a worker dose standpoint, so it was important to modify the contactors so all wetted areas within the contactor would be cleaned. The standard CINC CIP included perforations in the rotor shaft through which cleaning solution could be sprayed onto the rotor internal surface, as well as nozzles in the underflow region. SRS added spray ball nozzles in the annular region, 90 degrees apart, and two spray nozzles in each collector ring, 180 degrees apart. To test the system, all internal components were coated with yellow food dye and the system run. The CIP system was able to remove all food dye and considered to be acceptable for decontamination during hot operations to reduce dose rate.

Before Clean-in-Place



After Clean-in-Place

Both inlets and both outlets on CINC contactors are vented. During individual contactor testing anomalies in air flow through the vents was observed. The goal was to minimize air flow to the extent possible so that the bottom of the rotor would be "sealed" so no air flow could bypass the annular region and directly enter the rotor. This would allow the fluids in the annular region to mix more thoroughly and prevent surges in flow as the rotor bottom became covered then uncovered. The following figure show typical air flow through each of the inlet and outlet vents.



Typical Vent Air Flow

A comprehensive test plan was developed to explore the effect of a number of contactor variables and modify those that would accomplish the goal of efficient operation. The purpose of the testing was to:

(1) Identify differences between the standard CINC "TA" rotors (with bearings at the top and bottom of the rotor) and the new "TAH" or "hanging" rotor purchased for the MCU
(2) Increase the probability of successful mass transfer testing by investigating methods to increase the liquid height in the mixing zone (annulus)
(3) Recommend preferred contactor configurations for the V-05s and V-10s based on the testing.

The parameters to be investigated were:

- Rotor inlet size
- Bottom vane shape (curved vs. straight)
- Major vane diameter
- Vane-to-rotor gap height

A V-05 and V-10 were procured from the vendor that had removable bottom plates, adjustable rotor inlets, and windows on the housing that allowed the height of fluid in the annular region to be observed.

Varying the size of the rotor inlet indicated that this was not a viable parameter to change to increase the fluid height in the annulus. Increasing the rotor inlet diameter resulted in a step jump in fluid height. Several larger diameters led to essentially no increase, then the next step up filled the windowed region. The standard rotor allowed the contactor to operate in the fully pumping mode, then an increase in diameter put it into the partially pumping mode. This parameter was deemed to sensitive to consider for increasing fluid height.

The standard CINC design incorporates curved vanes on the bottom plate (Figure 2). All DOE design contactors have always had straight vanes. A series of tests was conducted using both style vanes to evaluate the effect on fluid height in the annulus.

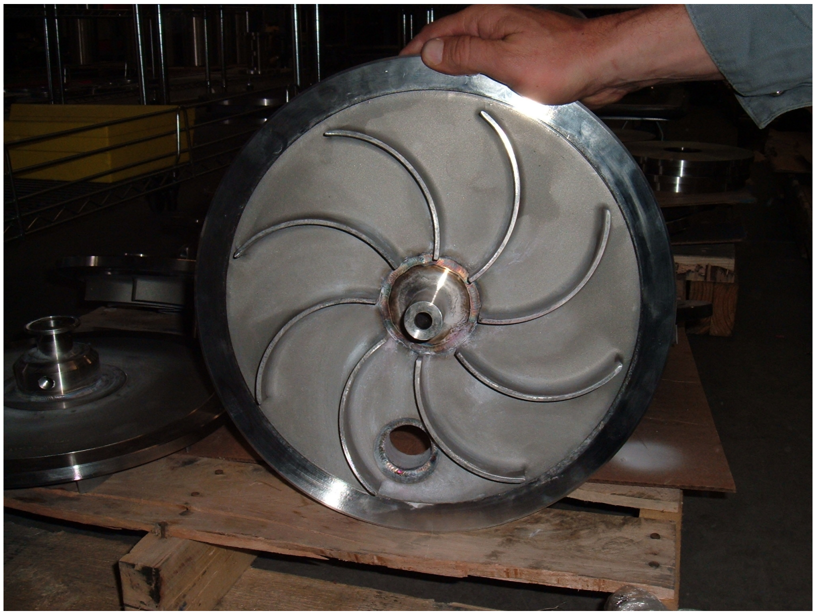


Figure 2 Curved Bottom Plate Vanes

Vane type and diameters were varied during testing, with only the curved V-05 vanes using slightly less diameter demonstrating any appreciable difference in annulus fluid height.

Varying the vane–to-rotor gap would reduce the pumping efficiency of the rotor and therefore raise the fluid level in the annulus. The effects were not as significant as those observed with changes in the vane diameter. Most of the increase in annulus height was seen only at the higher flow rates for the V-05. The V-10 showed only a small increase in annulus height over the MCU flow range.

Varying the V-05 rotor inlet had no appreciable affect on the annulus height. With curved vanes, increasing the gap and decreasing the major vane diameter resulted in increased annulus heights with the vane diameter having a slight advantage over the MCU flow range. Testing with straight vanes typically showed larger annulus heights than curved vanes, but the air flow was not reduced by the increasing liquid height. Therefore the major vane diameter on the V-05 contactors was reduced to with no change to any other parameters.

Varying the V-10 rotor inlet did not make a difference in annulus height, but unlike the V-05, the contactor eventually flooded. Decreasing the major diameter of the curved vanes resulted in an increase in annulus height up to the point the contactor was no longer able to pump. Testing with different straight vane diameters gave similar results, but the contactor lost pumping ability with a much smaller change that the curved vanes. Increasing the vane-to-rotor gap also increased the annulus level, but to a lesser extent than the vane diameter changes. None of the parameter changes showed any improvement in the air flow through the contactor. No changes were made to the V-10 contactors

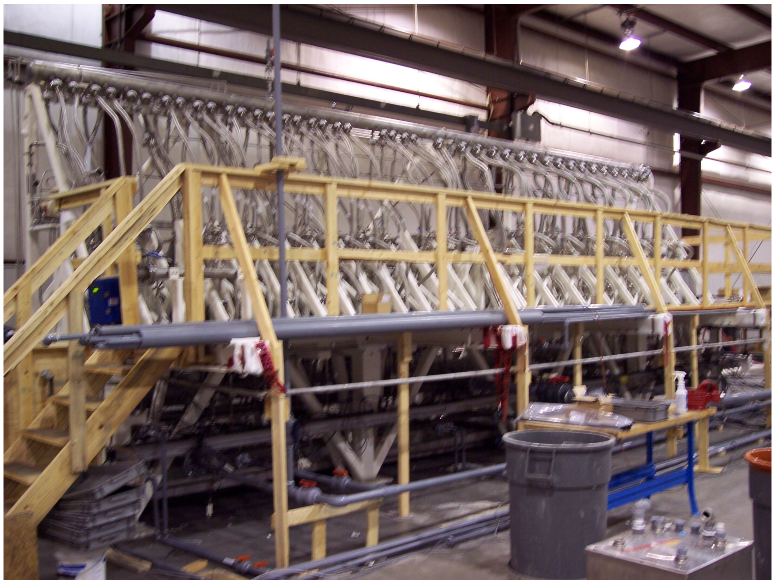
INDIVIDUAL CONTACTOR MASS TRANSFER TESTING

The extraction V-10 contactor was tested using salt simulant and actual solvent containing extractant and the strip V-05 contactor was tested using strip solution and solvent for removal and stripping of Cs-133. This testing was completed to assure that individual stage efficiency was high enough to allow the design DF and CF to be obtained.

The contactors were tested at the various design flow rates and various rpm. Efficiencies greater than 80% were obtained and validated the assumption that the CINC contactors would be able to meet the design criteria.

VENDOR FULL SCALE TESTING

When the specifications for the contactors had been finalized, testing of the assembled full bank of 18 contactors commenced at a vendor facility. A tank farm to supply feeds to the process was constructed and the full process was tested.


Contactor Assembly at Vendor Facility

The purpose of the vendor full scale test was to observe the hydraulics, demonstrate a decontamination factor (DF) ≥12, and demonstrate a concentration factor (CF) ≥12 when operating all seven V-10 contactors and all eleven V-05 contactors assembled as they will be in MCU. The operating conditions were selected based on the results of the Individual Contact test and Mass Transfer testing. This was the first test of the MCU flow sheet at full scale. Successfully meeting the objectives would conclude contactor testing until Cold Chemical Runs at SRS.

Testing conducted included:

- Mass Transfer Test (using “cold” Cs-133)
- Durability Test (ability to run continuously for 96 hours)
- Upset/Transient Conditions Testing
    - Transient Condition Testing involved stopping certain unit operations or feed streams in order to see how the contactors responded. The events tested were in order, as follows:
    - •Loss of Process Vessel Vent (PVV) system
    - •Loss of Temperature Control (tepid water) system
    - •Loss of solvent feed flow
    - •Loss of salt solution feed flow
    - •Loss of scrub feed flow
    - •Loss of strip feed flow
    - The hydraulic impacts of each of the events were observed, and samples for carryover (both aqueous and organic) were collected.
- Clean-In-Place Test

Integrated testing provided a vast amount of information pertinent to the operation of the contactors in MCU:

- Due to a control system programming error, the skid experienced an “uncontrolled shutdown.” A procedure was developed that successfully recovered the unit to an operable condition without having to drain the contactors.
- The MCU is capable of achieving a cesium Decontamination Factor at least an order of magnitude greater than the design specification.
- The MCU can meet and exceed the target concentration factor of 12 for cesium in the strip effluent, effectively minimizing the impact on DWPF cycle time due to boiling off excess water.
- Small variances in strip flow rate (2%) will have a noticeable impact on concentration factor. Accurate and precise control of process streams is needed to assure process requirements are met.
- Solvent carryover from the full bank of contactors is within the capability of the MCU coalescers and decanters to meet the output requirement of 50 ppm Isopar. Results are best at low to mid salt solution feed rates.
- Foaming occurs during flow rate changes; however, foam never reaches the vent header or causes process upsets. Foaming is less prevalent at low to mid salt solution feed rates. The operating scenario for the MCU should be to select a flow rate based on ARP process rates and avoid changes during operation.
- Due to pH changes, the Caustic Wash Tank must either be replenished at much shorter intervals than originally anticipated, or a feed-and-bleed system must be incorporated into the design.
- Operating parameters for the Caustic Wash contactors should be evaluated to minimize the aqueous carryover that results in a volume decrease in the Caustic Wash Tank.
- The CIP system appears to be effective in wetting all parts of the contactor rotor and housing. This should significantly reduce the radiation hazards associated with hands-on maintenance of the contactors.
- Process fluids are present in the “crud trap” of the bearing isolators/seals. The seals were

effective in protecting the bearings, but the material in the trap may pose a radiation hazard during maintenance. Further investigation demonstrated that the radiation rates will be manageable.

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