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The Testing of Commercially Available Engineering- and Plant-Scale Annular Centrifugal Contactors for the Processing of Spent Nuclear Fuel

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THE TESTING OF COMMERCIALLY AVAILABLE ENGINEERING AND PLANT SCALE ANNULAR CENTRIFUGAL CONTACTORS FOR THE PROCESSING OF SPENT NUCLEAR FUEL

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1. Introduction

Annular centrifugal contactors (ACC s) are being studied for rapid and efficient processing in numerous nuclear spent fuel partitioning applications by liquid-liquid solvent extraction technologies. They have been studied and utilized in limited applications for over forty years. Webster et al. at Savannah River provided the basic design and Bernstein et al. at Argonne, added the annular mixing zone [1,2]. Leonard and co-workers, also at Argonne, developed many practical concepts and improved upon the design and utility of these devices [3,4].

Commercially available, mass produced, contactors are now sold with significant numbers of units being utilized in a wide range of separations in both the industrial and government sectors. Separation, washing, and extraction operations all benefit from the elegant design of this relatively new tool. Processing advantages of this technology include; low in-process volume per stage, rapid mixing and separation in a single unit, connection-in-series for multi-stage use, and a wide operating range of input flow rates and phase ratios without adjustment. Therefore, the centrifugal contactor is the equipment of choice for contemporary solvent extraction based separations. Evaluation of these mass-produced units in support of the various nuclear fuel cycle and radioactive waste management goals is thus ongoing at the Idaho National Laboratory (INL) and other locations.

Commercialization of annular centrifugal contactor technology in the U. S. began eleven years ago with the technology transfer of a patent from the Department of Energy's Idaho National Engineering and Laboratory [5]. Since that time, a number of design enhancements have been made and patented that led to a device better suited to a wide range of liquid-liquid processes. Multiple sizes were designed to provide total throughput ranging from 0.1 to 200 gallons per minute and interchangeable heavy phase weir rings were incorporated into the rotor to allow separation of a wide range of density pairs [6,7]. A low mixing sleeve was added to aid in direct separations of viscous and shear sensitive liquid mixtures [8]. Clean in place (CIP) capability was achieved by adding a hollow central shaft with spray nozzles to the rotor design to enhance use as a clarifier and improve utility in hands-on and remote applications [9].

The primary objective of this work was to test and evaluate several sizes and features of commercially available centrifugal contactors for use with currently proposed Advanced Fuel Cycle Initiative (AFCI) flowsheets. It builds upon the Argonne evaluation, by R. A. Leonard et al, of this same 5 cm contactor for removal of cesium from high-level waste at the Savannah River Site (SRS) [10]. In that work, the commercially designed 5 cm unit compared favorable with the performance obtained in past studies using both 2 and 4 cm diameter ANL centrifugal contactors. The Argonne study resolved the questions raised by an earlier study on a modified 5 cm rotor at Oak Ridge National Laboratory (ORNL) [11]. Further efforts obtained in a later ORNL study confirmed the ANL results when good performance was observed using a conventional rotor in the 5 cm centrifugal contactor [12].

Numerous successful partitioning tests using Argonne 2 cm contactors on actual radioactive feed streams have been conducted at several of the National Laboratories. Small contactors are convenient to use for "hot" work to minimize radioactive waste and provide multi-stage data in a limited space such as conventional hot cells. However, the steady-state flow characteristics of these small devices requires careful leveling of all units, provides little tolerance for solids, and exhibits flooding due to intermittent flow in connections and phase inversions in the mixing annulus [13]. All of these flow related issues can easily lead to loss of stage efficiency in multistage "hot" testing of 2 cm units. Therefore, contactor testing on sizes of units having rotor diameters of 4 cm or larger has been recommended to ensure accurate input to pilot or production plant design [13,14].

Additional performance enhancements such as unit by unit heavy phase weir diameter optimization for changing process steps, phase ratios, and densities, low mixing contactors for terminal stage separations and solvent washing, and use of the CIP equipped units for remote cleaning and as clarifiers are among the aspects for study at the INL. This report details the hydraulic and initial mass transfer tests of the commercial 5 cm contactors.

2. Equipment Description

All testing was performed on commercially available 5 cm diameter rotor ACC s, Model V-02, purchased from CINC Processing Equipment [15]. A cutaway diagram of an ACC is shown in Figure 1. The units were also equipped with optional clear polymer housings to aid in observing mixing, flooding, and discharge of separated liquid phases. However, these housings are not compatible with many of the extractants used for AFCI separations so their use in actual flowsheet testing is somewhat limited. Figure 2 shows the contactors configured with stainless steel and clear polymer housings including a close-up of the clear housing in operation. The 5 cm units are individually driven by 240 volt three phase motors each with variable frequency drives that plug into standard 120 volt outlets. Salient features of the CINC contactors, as purchased, are given in Table 1.

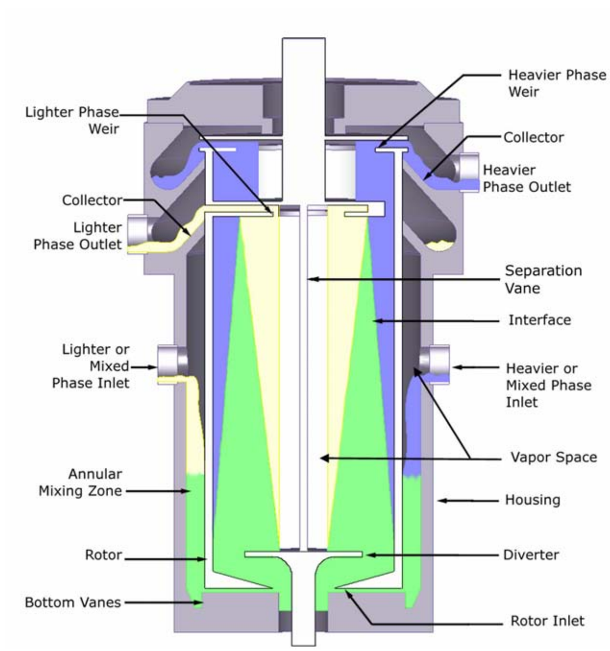

Figure 1. Annular Centrifugal Contactor

Table 1. Description of the CINC 5 cm centrifugal contactors

Size	5 cm rotor (O.D.)
Material of construction	316L stainless steel
Inlet and outlet ports	Male 3/8" NPT, tangential
Motor	Dietz, 0.125 hp, 3410 rpm 230/460 v. XP group C, D
Speed controllers	Allen Bradley Powerflex 4, cat. # 22A-V1P5N104
R[illegible]	[illegible]00 rpm
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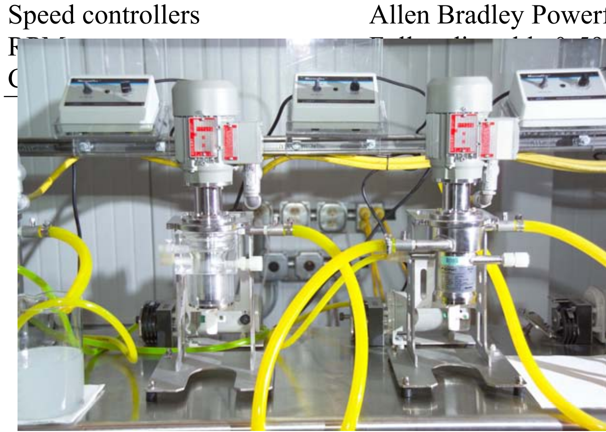

Figure 2. CINC 5 cm contactors equipped with polymer and stainless steel housings

3. Results and Discussion

3.1 Hydraulic testing with lamp oil and water

Two-phase hydraulic testing was initially accomplished using tap water and commercial grade of blue dyed lamp oil. Lamp oil is a blend of normal paraffin hydrocarbons, from C_{14} to C_{16}. The color aids in the observing of mixing in the contactor annulus and also in seeing carryover in separated samples. Its physical and chemical properties, provided in Table 2, make it convenient and safe to use for basic hydraulic contactor performance studies.

Table 2. Lamp Oil properties

Specific Gravity (H_2O = 1): 0.773@ 16^0 C/16^0 C
Viscosity: 2.5-2.7 cSt @ 40^0 C
Vapor Pressure (mm Hg): 0.05 @ 20^0 C
Flashpoint: 121^0 C
Solubility in water: negligible
Odor: very mild hydrocarbon

Clearly, it appeared that any of the three tested heavy phase weirs would be useful over a wide range of ratios and throughputs for the lamp oil/ water couple. The middle weir, 0.975", was chosen for the next series of tests as it provided a good balance of O/A ratios within the rotor volume. The effect of rotor speed on separation efficiency at increasing flowrates was studied for O/A ratios of 0.5 and 2. The results of those tests are summarized in Table 3. Slight carryover was defined as the presence of a few organic droplets on the surface of a 2 L aqueous sample or a droplet equaling less than 0.1% aqueous in the organic sample seen after centrifuging a 30 mL organic outlet sample for several minutes. Carryover was defined as any quantity above slight but was not further quantified in these tests.

3.2 Hydraulic testing with TRUEX process

The TRUEX portion of the UREX process consists of extraction, scrub and strip sections. Prior to hydraulic testing of each section, the TRUEX solvent was pre-equilibrated in the 5 cm ACC with the respective solutions used as the process feed to the various sections. All pre-equilibrations were performed at an O/A = 0.5 at 3500 RPM for an equivalent of three solvent turnovers. The following Tables 4 and 5 provide hydraulic testing results on solutions that were used in actual mass transfer tests. All samples taken for phase separation quality were centrifuged offline in a general-purpose unit at 4000 RPM for 3 minutes. Unless otherwise quantified, carryover was noted as very slight if barely visible and slight if more visible but less than measurable from comparison with visual standards having a minimum carryover of 0.1%.

Table 3. Contactor tests with lamp oil/ tap water vs. rotor speed, O/A ratio (weir=0.975")

O/A	RPM	Total Flow	Carryover	O/A	RPM	Total flow	Carryover
0.5	2000	1.0 lpm	No	2.0	2000	1.0 lpm	No
0.5	2000	1.5 lpm	No	2.0	2000	1.5 lpm	No
0.5	2000	2.0 lpm	No	2.0	2000	2.0 lpm	No
0.5	2000	2.5 lpm	Slight (A in O)	2.0	2000	2.5 lpm	Slight (A in O)
0.5	2000	3.0 lpm	Yes (A in O)	2.0	2000	3.0 lpm	Yes (A in O)
0.5	2500	2 lpm	No	2.0	2500	2 lpm	No
0.5	2500	2.5 lpm	Slight (A in O)	2.0	2500	2.5 lpm	No
0.5	2500	3.0 lpm	Yes (A in O)	2.0	2500	3.0 lpm	No
0.5	3000	2 lpm	No	2.0	3000	2 lpm	No
0.5	3000	2.5 lpm	No	2.0	3000	2.5 lpm	Slight (A in O)
0.5	3000	3.0 lpm	Slight (A in O)	2.0	3000	3.0 lpm	Slight (A in O)
0.5	3500	2 lpm	No	2.0	3500	1.5 lpm	No
0.5	3500	2.5 lpm	Slight (A in O)	2.0	3500	2 lpm	Slight (A in O)
0.5	3500	3.0 lpm	Yes (A in O)	2.0	3500	2.5 lpm	Slight (A in O)
0.5	4000	2 lpm	No	2.0	3500	3.0 lpm	Slight (A in O)
0.5	4000	2.5 lpm	Slight (A in O)				
0.5	4000	3.0 lpm	Yes (A in O)				

Table 4. TRUEX hydraulic extraction section test using 2.5 M HNO_3 (weir = 0.976”)

RPM	Total flow	O/A	comments
2500	1 lpm	0.5	Very slight O in A
3000	1 lpm	0.5	Very slight O in A
3500	1 lpm	0.5	Very slight O in A
4000	1 lpm	0.5	Slight O in A
4500	1 lpm	0.5	No carryover
2500	1.5 lpm	0.5	Slight O in A
3000	1.5 lpm	0.5	Slight O in A
3500	1.5 lpm	0.5	Slight O in A
4000	1.5 lpm	0.5	Slight O in A
4500	1.5 lpm	0.5	Very slight O in A
2500	2.0 lpm	0.5	A in O at 5%
3000	2.0 lpm	0.5	A in O at 5%
3500	2.0 lpm	0.5	A in O at 2%
4000	2.0 lpm	0.5	A in O at 1%
4500	2.0 lpm	0.5	A in O at 1%

Table 5. TRUEX hydraulic strip test using Lactic acid and DTPA (weir = 0.975”)

RPM	Total Flow	O/A	Comments
3000	1 lpm	1.2	Very slight O in A, A in O < 0.2%
3500	1 lpm	1.2	Very slight O in A, A in O < 0.1%
4000	1 lpm	1.2	Aq good, A in O <0.5%
2500	0.6 lpm	1.3	Very slight O in A, A in O 0.2%
3000	0.6 lpm	1.3	Aq good, A in O 0.2%
3500	0.6 lpm	1.3	Aq good, A in O 0.2%
4000	0.6 lpm	1.3	Aq good, A in O 0.5%

3.3 Mass transfer testing with TRUEX process

The results of these tests provided the parameters required for the TRUEX mass transfer testing. To prepare, the lactic acid stripped TRUEX extractant was conditioned for mass transfer studies by first washing, three times, with a solution of 0.25 M sodium carbonate at an O/A of 4 and then thrice equilibrated with 0.1 M nitric acid at an O/A of 0.5. These solvent treatments were made using the centrifugal contactor at 3500 RPM in three discreet passes. The third contact with the TRUEX solvent immediately emulsified the two phases however that was reversed by washing the emulsion with 0.1 M nitric acid.

Subsequent tests of virgin TRUEX solvent under identical conditions resulted in a very cloudy organic when contacted thrice with sodium carbonate but no emulsion formed. Another washing test with virgin solvent using a low mix sleeve to reduce shear in the mixing annulus gave better results. The low mix sleeve, a cylinder that shields the input liquids from the spinning rotor is shown in Figure 3. The carbonate washed solvent remained clear even after five washes. Following three washes with 0.1 M nitric acid, this solvent was processed again using a standard high mix bottom plate. No emulsion formed but the solvent was visibly cloudier after carbonate washing at the higher shear. Application of the low mix option in solvent extraction flowsheets as a final section disengaging stage and for carbonate washing will be evaluated further in the future.

A total of 1.4 L of TRUEX solvent was prepared, purity checked via ^{241}Am extraction [16], carbonate washed as described above, and subsequently pre-equilibrated with nitric acid prior to use as organic feed to the extraction stage. These solutions, at a total flow of 1 LPM, were fed into the contactor operating at 3000 RPM. Contacting the solvent three times with fresh acid on a single organic pass basis provided pre-equilibration. The aqueous feed was a nitric acid solution containing stable cerium and europium. Temperatures of the discharged phases were 23^{0}C during the first pass and 22^{0}C during the second pass. Extraction section tests at total flowrate of 1 and 1.5 Lpm were then completed (Table 6). Samples of the discharged phases were collected while at operating equilibrium, each after one minute of elapsed time from start up or parameter change. After each operating parameter change, equilibrium was reestablished by waiting one minute before sampling again. Stage efficiencies in the extraction section were all in the mid to high 90 percentage range, independent of flow rate or rotor speed.

The high stage efficiencies in the strip section were comparable to those obtained in the extraction section without dependence on flow or rotor speed (Table 6). No other phase carryover was observed in any of the samples taken during the TRUEX mass transfer study.

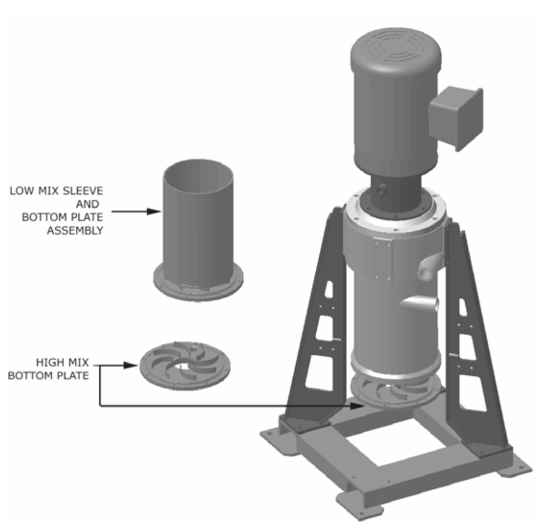

Figure 3. High and low mixing options

Table 6. Mass transfer efficiency results

Total Flowrate	RPM	Extraction Efficiency Ce	Extraction Efficiency Eu	Total Flowrate	RPM	Strip Efficiency Ce	Strip Efficiency Eu
1.0 lpm	3000	95.9	95.1	0.77 lpm	3000	95.8	96.2
1.0 lpm	4000	98.5	98.4	0.77 lpm	4000	94.9	97.2
1.42 lpm	4000	99.2	99.0	0.59 lpm	4000	96.1	98.1
1.42 lpm	3000	94.4	93.8	0.59 lpm	3000	98.9	---

7. Conclusion

Hydraulic testing of annular centrifugal contactors for use in pilot and production processes provides valuable data and guidance for plant and process design. This preliminary testing of 5 cm commercial contactors demonstrated the differences obtained when evaluating such operating hydraulics as maximum throughput for two different organic solvent systems (lamp oil/water and TRUEX/ aqueous).

Mass transfer efficiency for a single stage was greater than 93% in all cases and did not change measurably over a throughput range of 50-75% of maximum for the TRUEX process. Likewise, rotor speed did not affect performance, either in terms of mass transfer or phase separation. Selection of the optimum rotor speed for a given unit size and throughput should be governed by the lowest rpm that provides good phase disengagement and mass transfer, a function of mixing. Tests conducted to date would thus choose 3000 RPM for either 1 or 1.5 Lpm total flow in the TRUEX extraction section.

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