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ABSTRACT

TRUEX mass transfer tests were performed using a single stage commercially available 12.5 cm centrifugal contactor and stable cerium (Ce) and europium (Eu). Test conditions included throughputs ranging from 2.5 to 15 LPM and rotor speeds of 1750 and 2250 rpm. Ce and Eu extraction forward distribution coefficients ranged from 13 to 19. The first and second stage strip back distributions were 0.1 to 1.4 and .001 to .004, respectively, throughout the dynamic test conditions studied. Visual carryover of aqueous entrainment in all organic phase samples was estimated at < 0.1% and organic carryover into all aqueous phase samples was about ten times less.

Mass transfer efficiencies of essentially 100% for both Ce and Eu in the extraction section were obtained over the entire range of test conditions. The first strip stage mass transfer efficiencies ranged from 75 to 93% trending higher with increasing throughput. Second stage mass transfer was greater than 99% in all cases. Increasing the rotor speed from 1750 to 2250 rpm had no significant effect on efficiency for all throughputs tested.

INTRODUCTION

Centrifugal contactors are being evaluated for the selective partitioning of actinide, lanthanide, and fission products from dissolved Light Water Reactor nuclear fuel solutions. Solvent extraction processes utilizing centrifugal contactors realize three important advantages over historically used equipment. First, the units are capable of processing plant scale throughputs with less in-process volume compared to pulse columns or mixer settlers. Secondly, the short residence times associated with contactor processing minimizes negative solvent effects such as hydrolysis and radiolysis when processing nuclear fuel solutions. Lastly, the shorter time to reach process equilibrium and the ability to tolerate in-process upsets and changes in flowrates and phase ratios makes centrifugal contactors quite attractive for more complex flowsheets. However, shorter residence times and differences in mixing dynamics can impact mass transfer rates. Therefore, the performance and efficiency of various sized contactors is being measured and their operating ranges are being defined for engineering-scale applications.

Commercialization of annular centrifugal contactor technology in the U.S. began about 15 years ago with the technology transfer of a patent from the Department of Energy's Idaho National Engineering Laboratory (1). Since that time, a number of design enhancements have been made and patented that led to a device better suited for a wide range of liquid-liquid processes. Multiple sizes were designed to provide total throughput ranging from 0.4 to 800 liters per minute (LPM) and interchangeable heavy phase weir rings were incorporated into the rotor to allow separation of a wide range of density pairs (2,3). A low-mix sleeve option was

added to aid in the separation of viscous and/or shear sensitive liquid mixtures (4). 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 to improve utility in both hands-on and remote applications (5).

In the past, various centrifugal contactor designs have been used at the Idaho National Laboratory (INL) to demonstrate novel solvent technologies for the selective partitioning of radioactive material from acidic media. The most recently designed, commercially available units have been evaluated at the INL for the past five years. Hydraulic and mass transfer test results for the CINC V-02 (5 cm) model were reported in September 2005 (6). The Clean in Place test results for a CINC V-05 (12.5 cm) model unit were reported in June 2006 (7), followed by the hydraulic and reliability test results for the 12.5 cm unit reported in September 2006 (8). Most recently the mass transfer testing of a single stage V-05 (12.5 cm) unit for the TRUEX process was completed.

EXPERIMENTAL

The experimental assembly consisted of a commercially available model V-05 CINC annular centrifugal contactor with a 12.5 cm (5 inch) diameter rotor fitted with a 2.600 inch heavy phase weir (9). A variable frequency drive was used to adjust rotor speed during testing. The V-05 contactor is designed with a rotor shaft that runs the entire length of the unit which provides clean-in-place capability and serves to stabilize the rotor. This added stability enhances reliability via minimized vibration and reduced bearing and seal wear when compared to top shaft only rotor designs. A detailed listing of the test equipment used for the mass transfer testing is found in Table 1.

Table 1. Detailed description of the mass transfer test equipment.

Contactor: model CINC V-05TA CIP 12.5 cm rotor (O.D.) fitted with 2.600 inch weir	
Material of construction	316L stainless steel
Inlet and outlet ports	Male 1” NPT, tangential
Motor	Baldor, inverter duty, 2 hp, 3450 rpm, 230/460 V XP Class 1 Group D
Speed controller	Allen Bradley Powerflex 4, cat. # 22A-V1P5N104
Rotor speed	Fully adjustable 0-5400 rpm
Configuration	Single stage units, interconnectable
Size	Footprint 16” X 16”, height 42” with motor
Weight	140 lbs (63.6 kg) with motor
Rated Capacity	0-19 LPM, (5 gpm)
Feed pumps (2)	Micropump series 5000/56C model GL H25 FFSE
Capacity	Variable 2-27 LPM
Material of construction	316 Stainless steel
Drive motors (2)	LEESON Elec. Co. model C6T34F41D 1 hp 208-230 V 3 phase
Speed controllers (2)	LEESON Speed Master micro series model 174915
Tubing 1” supply 1.25” discharge	Gates Mustang 45 HW type K polyethylene

Mass transfer testing of the contactor was limited to a non-radioactive area. Therefore, the Transuranic Extraction (TRUEX) process solvent was chosen for the extraction of stable cerium and europium. The TRUEX solvent was prepared by combining octyl(phenyl)-N,N-diisobutyl-carbamoylmethylphosphine oxide (CMPO) and tributyl phosphate (TBP) to a final concentration of 0.2 M CMPO + 1.4 M TBP in n-dodecane. CMPO was acquired from Eichrom Technologies, Inc. (Darien, IL). The reagent grade TBP and n-dodecane were received from Aldrich Chemical Co. (Milwaukee, WI). Following the adjustment, the solvent was washed three times with 0.25 M Na_2CO_3 at an O/A of 4 using fresh wash solution each time. The washed solvent was then reacidified with 0.1 M HNO_3 at an O/A of 3. The solvent was then pre-equilibrated twice with 3.2 M HNO_3 at an O/A of 1.

The feed for the extraction section was 3.2 HNO_3 with stable cerium and europium added at 1.0 and 0.25 g/L, respectively. The extraction section tests were then performed by pumping the TRUEX solvent and aqueous feed into the contactor at an O/A ratio of 0.5 using the test parameters listed in Table 2. The system was allowed to reach equilibrium before both aqueous and organic samples were collected and all extraction, scrub and strip tests were run at ambient temperature (19-21^{o}C). Samples were visually examined for phase carryover at each test condition. No noticeable phase carryover was observed in the samples obtained for all test parameters for the extraction section testing.

Table 2. Mass transfer testing parameters for the extraction section.

Total throughput (LPM)	Rotor speed (rpm)	Aqueous flowrate (LPM)	Organic flowrate (LPM)
3	1750	2	1
3	2250	2	1
5	1750	3.4	1.7
5	2250	3.4	1.7
10	1750	6.6	3.4
10	2250	6.6	3.4
15	1750	10	5
15	2250	10	5

The scrub section testing consisted solely of a single pass of the loaded organic solvent contacted with 0.1 M HNO_3 acid at an O/A of 1, processing a total throughput of 10 LPM at a rotor speed of 2250 rpm. The scrub reduces the acid in the solvent so the lanthanides can be stripped. During the scrub testing, collected samples were not submitted for analysis but were used only to observe phase carryover of either phase during processing. No evidence of other phase carryover was observed in the scrub section.

The strip section consisted of contacting the scrubbed organic with a strip solution at an O/A of 1.3. The strip solution consisted of a 1.5 M lactic acid and 0.05 M diethylenetriaminepentaacetic acid (DTPA) in H_2O solution with a measured pH of 3. The strip section test conditions are shown in Table 3. Again, samples were examined for other phase carryover observations at each test condition. No measurable phase carryover was observed in any samples taken during the first strip section testing.

Table 3. Mass transfer testing parameters for the strip sections.

Total throughput (LPM)	Rotor speed (rpm)	Aqueous flowrate (LPM)	Organic flowrate (LPM)
2.5	1750	1.1	1.4
2.5	2250	1.1	1.4
5.7	1750	2.5	3.2
5.7	2250	2.5	3.2
8.5	1750	3.7	4.8
8.5	2250	3.7	4.8
13	1750	5.7	7.3
13	2250	5.7	7.3

RESULTS AND DISCUSSION

ICP-MS sample results were used to calculate distribution coefficients of Ce and Eu for single stage contacts for each test condition in the extraction and strip sections. Forward distribution coefficients for Ce and Eu in the extraction stage ranged from 13 to 19 at all process flow rates and rotor speeds tested. The higher rotor speeds combined with higher throughput levels tested had somewhat higher distribution coefficients. First strip stage distribution values differed when comparing Ce to Eu. Distributions for Ce ranged from 1.4 to 0.5 while those for Eu were measured at 0.8 to 0.1. In every set, Eu stripping was at least twice the magnitude of that for Ce. Overall, the strip distributions indicated better stripping with higher rotor speed and throughput. In addition, higher efficiency stripping was expected in the first strip stage based on modeling. However, measured acid concentrations of the scrubbed organic entering the strip section were slightly higher than those predicted. The single scrub employed in this test may have been insufficient for efficient stripping in the first stage. For this reason, a second stage strip study was conducted.

The TRUEX solvent was washed with Na_2CO_3 solution after the first strip study and recycled for another bulk extraction, scrub, and first strip process to prepare it for the second stage strip study. Fresh Ce and Eu feed was prepared for the second TRUEX solvent loading and fresh scrub and strip solutions were used. The extraction, scrub, and first strip processes were repeated, but with only several process verification samples, and the once stripped organic was collected for the second strip stage testing. This first strip, done to just provide organic feed for the second strip, was all processed at 8.5 LPM and 2250 rpm. In the second strip stage test, the same test parameters as used for the original first stage strip were repeated and each set sampled.

Distribution coefficients for the second strip stage were measured from .003 to .001 for Ce and .004 to .002 for Eu for all test parameters. No trends were observed due to flow rate or rotor speed as very efficient second stage stripping occurred.

Mass transfer efficiencies for extraction and strip section test parameters were calculated using the ICP-MS sample results taken from each test parameter and re-equilibrated samples. The mass transfer equation used is of the form:

$$\eta = \frac{(X - X_{in})}{(X_{eq} - X_{in})} * 100$$

Where X is the metal concentration of the effluent, X_{in} is the inlet metal concentration, and X_{eq} is the metal concentration of the effluent following re-equilibration. Efficiency can be calculated based on aqueous or organic phase concentrations. Aqueous phase concentrations were used for all extraction calculations and organic phase concentrations for strip calculations since these phases contained the Ce and Eu.

The mass transfer efficiencies for the V-05 were calculated for the extraction, first strip stage and second strip stage. For all parameters studied, the forward extraction efficiency was essentially 100%. The first strip stage efficiencies ranged from 75 to 93% with the least effective stripping at the lowest flow rate of 2.5 LPM. However, the second stage strip mass transfer efficiency was also essentially 100%, even for the lowest flow rate studied.

CONCLUSIONS

Two mass transfer tests were performed using a single stage commercially available 12.5 cm centrifugal contactor. The TRUEX portion of the uranium extraction (UREX) +1a flowsheet format was processed to measure the distribution coefficient and mass transfer efficiencies of stable Ce and Eu. Mass transfer efficiency measurements of essentially 100% for both Ce and Eu in the extraction section indicated the 12.5 cm contactor is very capable of efficiently extracting Ce and Eu from a simulated dissolved fuel solution with the TRUEX solvent at tested throughputs and rotor speeds. The first strip contact mass transfer efficiencies were lower than those determined for the extraction section. Efficiencies determined for Ce ranged from 75 to 91% while the Eu efficiencies ranged from 76 to 93 % each increasing with increasing throughput. The rotor speeds studied had no measurable impact on efficiency for all throughputs.

As a result of the lower than expected efficiency measurements in the first strip contact, a second mass transfer test incorporating a second strip contact was performed. The mass transfer efficiencies for all second stage strip test conditions were > 99 % for both Ce and Eu. The higher than expected acidity of the scrubbed organic significantly impacted the strip efficiency in the first stage.

Overall, visual and measurable carryover levels of aqueous entrainment in all organic phase samples were less than or equal to 0.1%. However, organic carryover into all aqueous phase samples was estimated to be at least an order of magnitude lower.

The high single stage mass transfer efficiencies indicate the 12.5 cm centrifugal contactor is a viable equipment option for the TRUEX portion of the UREX +1A solvent extraction flowsheet. In addition, very good phase disengagement using centrifugal contactors is common and should lead to process savings via reduced extractant loss in plant applications.

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