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Centrifugal Contactor Operations for UREX Process Flowsheet

Chemical Science and Engineering Division

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CENTRIFUGAL CONTACTOR OPERATIONS FOR UREX PROCESS FLOWSHEET

1 OVERVIEW

The UREX process separates uranium, technetium, and a fraction of the iodine from the other components of the irradiated fuel in nitric acid solution. The time and equipment requirements were evaluated for treatment of 260-L batches of a solution containing 130 g-U/L using commercial annular centrifugal contactors from CINC Industries. An earlier evaluation was done assuming 150 L of feed solution per batch.

Two different contactor versions from CINC were evaluated, the V-2 and V-5 units. The V-2 has a 2-in. (5-cm) rotor with a reported maximum total organic-plus-aqueous throughput of 1.9 L/min. We evaluated this version for a throughput of 1 L/min. The V-5 has a 5-in. (~12.5-cm) rotor with a reported maximum throughput of 19 L/min. We evaluated this unit for throughputs of 10 and 15 L/min. The lower-than-maximum throughputs selected for this evaluation are based on the Argonne National Laboratory (Argonne) ROTOR model, which calculates the operational envelope for a given process O:A (organic-flow-rate/aqueous-flow-rate) ratio based on contactor parameters (weir dimensions, rotor diameter, etc.), the physical properties of the two immiscible fluids, and the allowable other-phase-carryover. Because the UREX process designed for this application has O:A ratios ranging from 0.8 to 5.6, adjustments to the weirs in different sections of the contactor bank can broaden the operational envelope for satisfactory separations, but throughput will remain short of the equipment maximum due to the physical characteristics of the two phases.

2 CALCULATIONS

The reference UREX flowsheet consists of four sections with a total of 30 stages. Four additional solvent-wash stages may be required, but they would be used only periodically and are not included in these calculations. Based on processing a single batch, Table 1 shows the process and cumulative time requirements for a V-2 contactor at 1 L/min and for a V-5 contactor operating at maximum throughputs of 10 and 15 L/min. As can be seen, the startup and shutdown times are comparable for all three cases. Once the hot feed is exhausted, cold feed is introduced to initiate the uranium clean-out; all other flows are maintained. When the hot feed ends, this cleanout moves essentially all the uranium in the extraction section through the contactor and out into the uranium strip product. Nearly all of the residual uranium remaining after processing will be collected in the initial pass, but a three-residence time flush through the entire system is assumed in order to recover residual uranium due to other-phase-carryover. If the process is run continually, it may be feasible to overlay the startup and clean-out so that the contactor bank is maintained in operational mode except for periodic maintenance.

As Table 1 shows, the fuel processing time for the V-2 will be a factor of 10 to 15 times longer than for the V-5, and the overall processing time is larger by a factor of 3 to 6. The required process times can be reduced by adding multiple lines of the V-2, although hold-up and footprint begin to approach those of the V-5. Table 2 provides some design parameters for the two different contactor systems from CINC Industries. The 30-stage case assumes that the inter-stage lines required for multiple units in series will double the lateral dimension of the contactor based on the mount supplied with the individual units by CINC Industries and the orientation shown on the CINC Web site (http://www.cincind.com), where the interconnect tubing is aligned with the contactor stages. The Argonne design (as seen on the 2- and 4-cm rotor units currently onsite) places the inter-stage lines perpendicular to the stages, which allows the stages to be situated much closer together, reducing the lateral dimension but increasing the forward and back dimensions in a multi-stage bank. The perpendicular arrangement also allows easier access to the lines if both the front and back of the bank can be accessed. It may be possible to align the CINC units in this manner, but further study is needed; the Argonne arrangement could drop the length of the contactor bank by almost half. Figure 1 illustrates the different inter-stage line arrangements and how they impact footprint. A single V-5 bank will require approximately 10 m^2 of floor space for the arrangement with inline inter-stage lines (0.4 m × 25 m), while the V-2 would require approximately 3.2 m^2 (0.25 m × 15 m) of floor space. The contactor can be segregated at various points to break the straight line into sections. For example, there is a natural break between the second scrub and strip section, where the contactor could be divided into an 18-stage and a 12-stage unit. It can be separated at other points, but a pump may be required to move the aqueous phase from one section to the next. By breaking up the sections in this way, the two segments can be arranged in separate rows, or even tiered. Three V-2 multistage-contactor lines would require approximately the same floor space as a single V-5, although the need for access ready may increase the space needed beyond that for the V-5.

Along with processing time, the major factor driving the contactor size selection is the hold-up and the effluent volumes. Table 3 provides the in-process hold-up for the three contactor

TABLE 1 Processing Time Requirements for V-2 and V-5 Contactor Banks

	Run Time (min)			Cumulative Run Time (min)		
Process Step	V-2 (1 L/min)	V-5 (10 L/min)	V-5 (15 L/min)	V-2 (1 L/min)	V-5 (10 L/min)	V-5 (15 L/min)
Cold startup of all feeds	31	37	25	31	37	25
Initial hot feed to activity in raffinate	4	4	3	35	41	28
Initial hot feed to uranium in product	22	23	16	57	64	44
Process 260 L of hot feed	972	97	65	1029	162	109
Initial hot feed to exit last uranium in product	994	121	81	1051	185	125
Non-mandatory residual U rinse	44	47	32	1077	213	144
Incidental residual AHA rinse[a]	54	56	37	1131	269	182

[a] The AHA rinse may be considered if AHA removal from the contactor is required for extended stoppage.

TABLE 2 Dimensional CINC Specification for V-2 and V-5 Contactors[a]

	V-2		V-5	
Characteristic	1-Stage	30 Stages[b]	1-Stage	30 Stages[b]
Footprint	0.5 m^2	3.2 m^2	1.7 m^2	9.9 m^2
Height	0.6 m	0.6 m	1.0 m	1.0 m
Weight	12.8 kg	384 kg	68.2 kg	2046 kg
Maximum throughput	1.9 L/min		19.0 L/min	
Rotor speed	2000–6000 rpm		800–4000 rpm	
Hold-up	0.2L		3.3L	

[a] From http://www.cincind.com.

[b] 30-stage case includes an estimate of added space required for inter-stage lines.

cases examined here. As can be seen, hold-up in a given section is 10 to 15 times larger for the V-5 than it is for the V-2 contactor. Based on the ROTOR code prediction, upon completion of a batch the V-2 will contain approximately 2.9 L of organic phase and 5.7 L of aqueous phase; the V-5 will contain approximately 39 L of organic and 64 to 73 L of aqueous phase. The composition of the phases will vary significantly across any section during operation, so the U rinse initiated after all of the fuel feed has been introduced would use a cold feed to force all of the uranium to the strip product prior to shut down. It may be possible to have the uranium rinse serve as the startup for the subsequent batch of fuel, or at least reduce the duration of the cold startup if it is suitably effective.

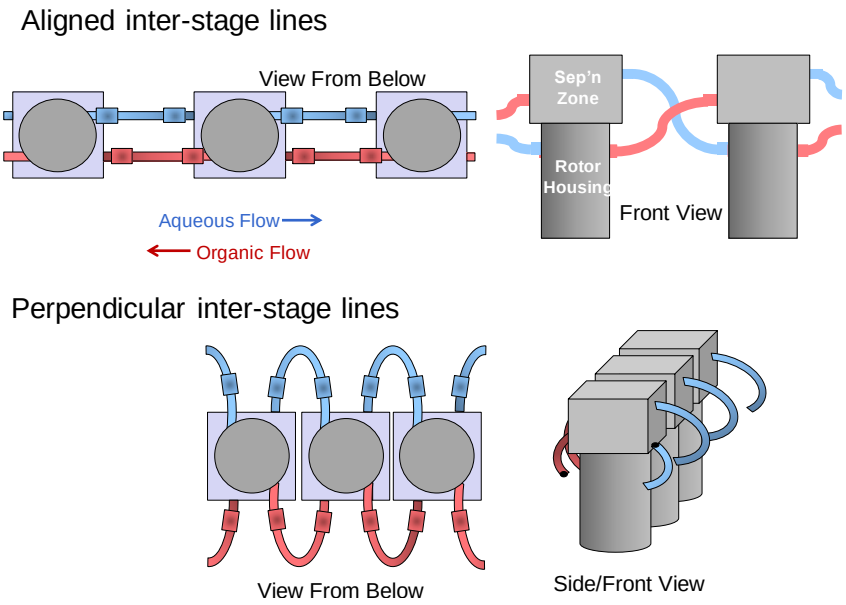

FIGURE 1 Illustration of Contactor Inter-stage Line Orientation

TABLE 3 Hold-up per Section for V-2 and V-5 Contactors Based on UREX Flowsheet

	V-2 (1 L/min)		V-5 (10 L/min)		V-5 (15 L/min)	
Hold-up (L)	Organic	Aqueous	Organic	Aqueous	Organic	Aqueous
Extraction	0.7	1.4	8.8	15.3	8.7	17.5
Scrub 1	1.1	1.2	17.6	12.1	18.5	11.4
Scrub 2	0.1	0.1	2.0	1.3	2.2	1.2
Strip	0.8	2.2	10.0	25.1	9.5	29.8
Overall	2.9	5.7	39.1	64.0	39.0	72.8

The individual stages can be equipped with drain valves so that they can be emptied and the contents collected separately and kept isolated once a run is complete. This may be useful for material accountancy and recovering from processing upsets. Draining may also reduce rinse volumes. However, valves are potential sources of leaks during operations and will require periodic maintenance, particularly if organic seats and seals are used.

Required input volumes for both the cold and irradiated fuel feeds were calculated based on the UREX process times shown in Table 1 and the volumetric feed flow rates shown in Table 4. These input volumes are shown in Table 5. The solvent value assumes that three times the hold-up is circulated during processing; however, this value will depend on the solvent wash schedule and the tankage allotment in the hot cell facility. Because it degrades in acid on standing, AHA will have to be added as a solid to scrub 1 just prior to processing, and thus the scrub 1 feed tank will require a mixing function. There are several options for acid management.

TABLE 4 Process Flow Rates for V-2 and V-5 Contactor Banks

Input Flow Rate (L/min)	V-2 (1 L/min)	V-5 (10 L/min)	V-5 (15 L/min)
Cold and Irradiated-Target Feed	0.27	2.7	4.0
Solvent	0.45	4.5	6.8
Scrub 1	0.013	0.13	0.20
Scrub 2	0.08	0.8	1.2
Strip	0.55	5.5	8.2
Raffinate	0.36	3.6	5.4
Product	0.55	5.5	8.2

TABLE 5 Process Feed Volumes Required for V-2 and V-5 Based on UREX Flowsheet

Process Feed (L required)	V-2 (1 L/min)	V-5 (10 L/min)	V-5 (15 L/min)
Irradiated Fuel Feed	260	260	260
Startup/Rinse Feed	26	286	293
Solvent[a]	9	117	117
Scrub 1	14	24	24
Scrub 2	84	145	147
Strip	574	991	1003

[a] Solvent in recycle mode assumed to require ~3 times hold-up.

All of the UREX feeds contain nitric acid and thus may be generated periodically from a single concentrated acid source, or procured pre-diluted, as determined by cost, tankage, plumbing, and other factors. The actual facility tankage will depend on the treatment schedule and the available tankage for cold chemical storage, but large bulk storage coupled with smaller process feed tanks would seem optimal for hot cell operations.

Table 6 lists the total volume of effluent generated per processing step for the three operational cases. As is readily evident, the greatest benefit arising from use of the small contactor comes from the reduced start-up and rinse volumes due to the smaller contactor hold-up, as given in Table 3. After introducing all of the irradiated fuel, a substantial fraction of the uranium remains in the contactor, so a cold feed must be introduced to clean out the uranium. This cold feed would likely be 1-M nitric acid, although a solution of un-irradiated uranyl nitrate is also a possibility. If multiple lines of the V-2 are used, hold-up and U-product volumes begin to approach those calculated here for the V-5. The effluent volume numbers suggest that there is substantial benefit to larger contactors if the processes are run continuously (if material accountancy can be managed) because the startup and rinse cycles may be shortened or perhaps eliminated. Batch operation seems to favor smaller units.

TABLE 6 Process Effluent Volumes Generated for V-2 and V-5 Based on UREX Flowsheet

Process Volumes (L)	V-2 (1 L/min)	V-5 (10 L/min)	V-5 (15 L/min)
Raffinate generated during cold startup	11	133	137
Product generated during cold startup	17	202	207
Raffinate generated processing fuel	351	351	351
U Product from fuel treatment	533	533	533
Raffinate generated during U rinse	16	169	173
Dilute product generate during U rinse	24	256	263

3 CONCLUSIONS

The analysis shown here is a first attempt at elucidating the contactor requirements. The tradeoff between the two different contactor sizes is very clear. The V-2 requires a substantially longer processing time, up to 18 hr for a single bank. The V-5 reduces the processing time by a factor of four to six, but the volumes of waste generated go up by a factor of 3 to 4. The quantities of cold chemicals needed simply to start up and shut down the larger unit are substantial for the V-5, and may be problematic if such operations occur frequently, although this is much less costly than waste disposal. The total waste generated for both cases may be reduced by optimizing contactor operations such as reducing rinses and startup times; this would improve V-5 performance in particular. The V-2 footprint is a third the size of that of the V-5. However, because the contactor stages are normally arranged side by side, the relative width ratio of 5:3 would likely have a greater impact on facility design than would the areal footprint.

The long process times required for the smaller V-2 unit can be reduced by employing multiple lines of contactors. Multiple banks increase facility flexibility by reducing the downtime associated with equipment failure and enabling simultaneous treatment of multiple irradiated fuel batches. However, the advantages of the V-2 over the V-5 in terms of waste volumes and footprint are reduced significantly with multiple lines. In addition, more banks require additional pumps, feed lines, product and raffinate tanks, and all of the additional material tracking and handling associated with the added equipment complexity. Ultimately, the contactor selection will depend on the integration of the UREX process with the other facility operations, but in isolation the tradeoffs are clear.

4 FURTHER WORK

Clearly, a systematic study needs to be done regarding the relative benefits of the longer process times necessary for the smaller units versus the larger liquid volumes required for the larger units. Given the process volumes and times associated with UREX, the mode in which the facility is operated will have a substantial impact. If operation in a semi-continuous mode is viable; if fuel solutions are processed in series, the contactor cleanup steps can be simplified by overlapping or combining the startup and rinse steps, resulting in a potentially substantial reduction in waste volumes.

The stage composition profile in the contactor will deviate significantly from the steady-state profile at startup and rinse because of the change in uranium concentration in the feed. This is particularly relevant for the V-5 or if multiple V-2 lines are employed, because the fuel processing times are similar in duration to the startup and rinse cycles. The product collected will begin to become relatively dilute in uranium upon introduction of cold feed, but the strip should become more effective, which would tend to mitigate this dilution initially. The dynamic AMUSE will predict the dynamic behavior of the system at startup and rinse. This will provide a better estimate of the uranium hold-up in the contactor bank over time, particularly for short processing times, and will therefore improve accountancy.

In the revised flowsheet studied here, the number of stages used for the second UREX scrub section was reduced by assuming that the concentration of AHA does not need to essentially zero because processing the uranyl nitrate product solution to uranium trioxide will destroy AHA and its hydrolysis products.

The UREX flowsheet assumes ideal behavior. Although the extraction sheets are generally robust, a sensitivity analysis should be done to determine where the designed flowsheet may break down. Typically, this would involve varying a number of process parameters, such as flow rate and feed concentrations, and calculating the system's response using the AMUSE code. Regions of unstable performance can be identified, as can cases where flowsheet parameters are particularly sensitive to process conditions. The dynamic AMUSE code would be particularly useful in providing information on the dynamic response, such as the time required for recovery from a process upset and the composition profile of the products as a function of startup conditions.

In terms of equipment, there is substantial room for optimization of contactor design. Because of the limited options, there is a significant change in process volumes when going from the V-2 to the V-5. It seems that the optimal scale may be somewhere between these two designs. It should be noted that contactors do have significant turndown ratios, so the V-5 throughput can be reduced by a factor of 10 with relatively small reductions in performance. So, if required, processing time can be extended to accommodate other processing. However, the hold-up volumes will not change substantially, so startup and rinse times will be similar to those listed here. An intermediate size unit can be designed at Argonne, but at substantial cost in terms of design time and manufacturing setup. Argonne-designed contactors are also developed for a

specific process, while commercial units are more generic, with substantial rework done to meet user-specific requirements.

One example of a simple modification mentioned above was the inter-stage line orientation. Other factors come into play, particularly for engineering larger contactors. As contactors become larger, vibration becomes more significant, so mounts must be designed to mitigate the effects of vibration, particularly if a large number of units are mounted in tandem, as will be necessary for UREX. Any modifications to the contactor support structure should undergo a vibration analysis.

The weirs within the rotors can be optimized to specific separations using the ROTOR model. The CINC units are equipped with multiple weirs, from which the user selects that most suited to the process of interest. Appropriate weir dimensions can improve throughput or stage efficiency by expanding the operational envelope in the appropriate direction. Another adjustable design element is the vanes layout at the base of the housing. CINC units are shipped with curved vanes that improve separation but reduce mixing. Other vane configurations can be installed. An analysis of vane orientation and height can be done for the specific system of interest, particularly as the different sections of the bank operate at very different O:A ratios. Similarly, the motors on the CINC units have variable-speed motors; inappropriate selection of the motor speed will impact performance. Motor replacement costs can also be reduced if variable-speed motors are replaced with single-speed motors, if these are determined to be sufficient for the desired separation. In sum, there is a number of design parameters associated with the operation of annular centrifugal contactors that should be determined in order to obtain optimal performance in solvent extraction.

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