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Development and Performance of a New Annular Centrifugal Contactor for Semi-Industrial Scale

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Abstract: Centrifugal contactors have several advantages such as low hold-up volume, short residence time, low solvent degradation, small space requirements, and short start-up time. Therefore, centrifugal contactors are favored for the reprocessing of spent fuel. Institute of Nuclear and New Energy Technology, Tsinghua University, China (INET) has been developing annular centrifugal contactors since the late 1970s. Recently, a new annular centrifugal contactor for semi-industrial scale, which the inside diameter of its rotor is 70 mm, has been developed to be used in nuclear industry. A modular design and an overflow structure design are adopted in the contactor. The hydraulic performance and the mass-transfer efficiency have been studied in a single-stage contactor and in a three-stage cascade. The maximum total flow of the single stage contactor can reach 290 L/h at suitable operation conditions for H_2O-kerosene system. The extraction stage efficiency of the single-stage contactor is greater than 95% at suitable operating conditions for extracting Nd^{3+} with 30% TRPO-kerosene solution from $Nd(NO_3)_3$-1 mol/L HNO_3 solution and from the simulated high-level liquid waste (HLLW). The hydraulic performance and the mass-transfer efficiency of the three-stage cascade are also excellent. Even though one stage or two nonadjacent stages ceased to work, the cascade can continue to run.

Keywords: Annular centrifugal contactor, semi-industrial scale, hydraulic performance, mass-transfer efficiency

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INTRODUCTION

Centrifugal contactors are efficient extraction equipments in extraction processes. Compared with conventional contactors such as mixer-settlers and pulsed columns, centrifugal contactors offer the following advantages, which are particularly beneficial to the nuclear fuel reprocessing (1–3):

- Low liquid inventories;
- Short residence time and therefore less solvent degradation;
- Excellent phase separation;
- High mass-transfer efficiency;
- Greater safety with respect to nuclear criticality;
- Compact and short therefore low capital costs;
- Without destroying the steady-state when shut down;
- Rapid start-up, shut-down, and wash out of the process liquors.

The primary centrifugal contactor, which was the paddle type, had been successfully developed and operated for many years at Savannah River Plant (SRL). In the late 1960s, the paddle type centrifugal contactor was modified to the annular type at Argonne National Laboratory (ANL). The ANL centrifugal contactor was reliable and easy to operate and maintain. In particular, it was used to hot test the TRUEX process in ANL (4–6). In the late 1970s, INET has developed its own annular centrifugal contactors. A series of INET centrifugal contactors have been developed with the rotor diameter from 10 mm to 230 mm (7–8). 10-mm INET annular centrifugal contactors were successfully used in the hot tests of the TRPO process in the hot cell of Institute for Transuranium Element (ITU) of EU in Karlsrule, Germany and in INET, respectively (9–12). The TRPO process has been developed in China for removing actinides from Chinese HLLW since 1980s, which the 30% trialkyl phosphine oxide (TRPO)-kerosene is been used as the extractant in the process.

Recently, a new 70-mm annular centrifugal contactor for semi-industrial scale has been developed for the semi-industrial scale test of the TRPO process in the future in INET. In order to meet the requirement of the process test, the design goal of the 70-mm annular centrifugal contactor is as follows:

- The remote maintenance is easy and rapid;
- The cascade can continue to operate when one stage or nonadjacent stages are ceased to work;
- The expected total flow (F) of two phases can reach 100 L/h;
- The range of flow ratio (aqueous/organic) is from 5/1 to 1/5;
- The rotor speed can be varied from 2000 r/min to 2800 r/min;
- The mass-transfer efficiency can reach 95%.

In this paper, both design and operation of the 70-mm annular centrifugal contactor are described. Experimental results on the hydraulic performance and the mass-transfer efficiency of both a single-stage contactor and a three-stage cascade are presented.

THE 70-mm ANNULAR CENTRIFUGAL CONTACTOR

Principle of Operation

The 70-mm annular centrifugal contactor made of stainless steel is shown in Fig. 1. Its main structures are shown in Fig. 2.

The inside diameter of its rotor is 70 mm, and the hold-up volume is about 350 mL.

Two immiscible liquids are fed from the opposite sides into the annular mixing zone between the spinning rotor and the stationary housing, and are mixed by skin friction as they flow down the annular space. Four radial vanes in the bottom of the housing inhibit the rotation of the mixture and direct it through the inlet in the bottom and into the inside of the rotor. Here the emulsion breaks rapidly under the high centrifugal force. The separated phases flow separately through the heavy phase weir and the light phase weir of the rotor into their collector rings in the housing. Then each liquid leaves its collector ring through a tangential exit, and flows into an adjacent stage, respectively. The extraction cascade is formed by linking the exit to its corresponding inlet of the neighbor contactor in the opposite direction.

NEW DESIGN CHARACTERISTICS

Modular Design

In order to make the contactor remote maintenance easy and reliable, the mating joint of every part of the contactor need not use screws or pins. The

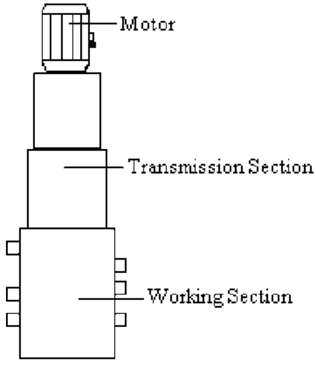

Figure 1. General view of the 70-mm annular centrifugal contactor.

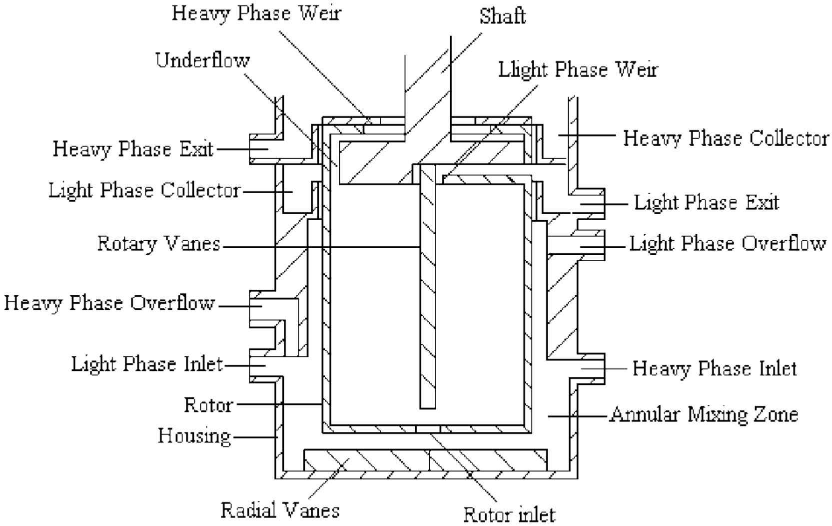

Figure 2. Cutaway view of the 70-mm annular centrifugal contactor.

recent 10-mm INET annular centrifugal contactor was divided into two parts (10). One was a housing part, and the other was a rotor part. Two parts were mounted separately, and jointed by cylindrical join with clearance fit, and without screws and nuts. In order to prevent the rotor part from rotating when the rotor speed was high, two stop pins were used between two parts. H. Takeda et al. reported the design to apply the magnet coupling in the centrifugal contactor (13). The application of the magnet coupling to drive the rotor meant the motor was replaced independently of the rotor. This feature provided an easier mode of remote maintenance and decreased the space requirement on the up side when replacing the motor. However, screws or pins were adopted for the mating joint of every part of the contactor.

The 70-mm annular centrifugal contactor is made up of three modules that are the motor module, the rotor module, and the housing module. Modules are installed in each other by simply inserting one on the other one, and they are fitted by mating cone without screws and nuts or pins. The contactor is assembled and disassembled fast by simply moving modules up and down. The magnetic coupling is also used in the contactor to provide the torque to rotate the rotor without loss of any speed. All these designs make the contactor remote maintenance easy and reliable.

Overflow Structure Design

For the use of the previous centrifugal contactors in a multistage system or cascade, if a stage of the cascade became inoperative such as caused by the failure of a drive motor, then the entire cascade must be shut down. To settle this problem, Jubin et al. invented a way to install horizontally oriented overflow ports below the organic collector rings and above the feed points with these overflow ports serially coupling the centrifugal contactors in the cascade (14). The overflow ports provided for the overflow of both aqueous and organic phases from the inoperative stage into the mixing volume in the operating adjacent stages. However, the overflow of liquid through the overflow ports consisted of both aqueous and organic phases, so the overflow ports were not useful when the inoperative stage was an end stage in the cascade. J. A. Jenkins et al. also adopted this overflow designs in the centrifugal contactors (2).

Two kinds of overflow line are designed in the housing module of the 70-mm annular centrifugal contactor (as shown in Fig. 2). One is the light phase overflow, and the other is the heavy phase overflow. The light phase overflow is below the organic collector ring and above the heavy phase overflow. The heavy overflow is below the light phase overflow and above the feed inlets. If a contactor ceases to work due to a motor failure or due to other abnormal accidents, the heavy phase and the light phase can separate by gravity in the housing of the stopped contactor and overflow in opposite directions through their own overflow line into adjacent stages, respectively. In this way, the overflows are also useful when the stopped stage is an end stage in the cascade. The use of the overflow structure design provides a satisfactory solution to the failure or inoperativeness of one stage or nonadjacent stages of a multistage centrifugal contactor arrangement or cascade, in other words, when a multistage cascade is operated, even though one stage or non-adjacent stages cease to work, the cascade can continue to operate.

EXPERIMENTAL

Hydraulic Tests

The hydraulic performance of the single-stage centrifugal contactor and the cascade with three-stage centrifugal contactors was tested with H_2O-kerosene system. The experimental flowsheets of the single-stage centrifugal contactor and the three-stage cascade are shown in Figs. 3 and 4 respectively. The nominal total flow of two phases at different flow ratio was defined by end stream entrainment below 0.5%. Each phase liquid was pumped through a rotameter to its inlet pipes of the contactor, and effluent streams were returned to proper tanks. To determine the amount of the opposite phase entrained, effluents were collected in a graduate; and after two phases separated, the entrainment was determined by volume measurement. The rotor speed of the contactor was adjusted by frequency modulation.

Overflow tests of the contactor were carried out with the three-stage cascade, which included four scenarios as follow: stages 2 and 3 worked

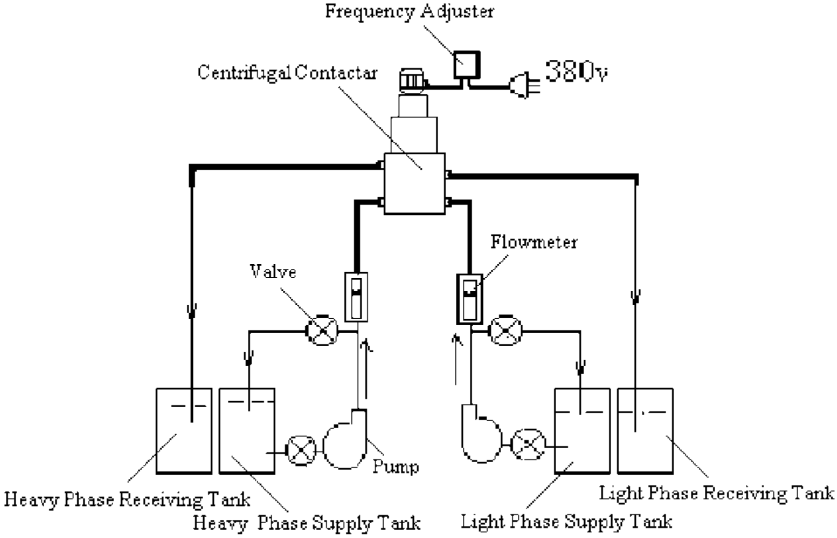

Figure 3. Flowsheet of the single-stage contactor test.

but stage 1 stopped; stages 1 and 3 worked but stage 2 stopped; stages 1 and 2 worked but stage 3 stopped; stages 1 and 3 stopped but stage 2 worked.

Mass Transfer Tests

Experimental systems of mass-transfer tests were the same as that of hydraulic tests (see Figs. 3 and 4). The extraction stage efficiency of the contactor was determined by extracting Nd^{3+} with 30% TRPO-kerosene solution from $Nd(NO_3)_3$-1 mol/L HNO_3 solution and from the simulated HLLW. The main composition of the simulated HLLW is shown in Table 1. The concentration of Nd^{3+} in the aqueous phase was analyzed with ICP-AES (Inductive Coupled Plasma) and the concentration of HNO_3 in the aqueous phase was determined by titration with the standard NaOH solution after adding $K_2C_2O_4$ as the masking agent. The concentration of Nd^{3+} in the organic phase was determined by firstly stripping with 5.5 mol/L HNO_3 three times

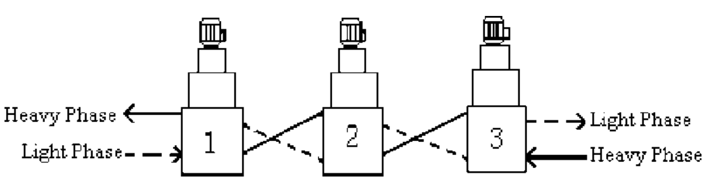

Figure 4. Three-stage contactor cascade.

Table 1. The main composition of the simulated HLLW

Element	Na	Al	Fe	Ni	Nd	Cs	Cr	Mo
Concentration (g/L)	51.2	15.9	17.4	8.2	0.5	1.8	2.0	0.8

and then by analyzing the stripping solution with ICP-AES. The extraction stage efficiency was calculated by Murphree equation as follows (15):

$$E_A = \frac{(C_{A,in} - C_{A,out})}{(C_{A,in} - C_{A,eq})} \times 100\% \quad \text{(for the aqueous phase)}$$

$$E_O = \frac{(C_{O,out} - C_{O,in})}{(C_{O,eq} - C_{O,in})} \times 100\% \quad \text{(for the organic phase)}$$

where E_A and E_O are the extraction stage efficiency of the aqueous phase and the organic phase, respectively (%), $C_{A,in}$ and $C_{A,out}$ are the inlet and the outlet concentrations of Nd^{3+} in the aqueous phase, respectively (mg/L), $C_{O,in}$ and $C_{O,out}$ are the inlet and the outlet concentrations of Nd^{3+} in the organic phase, respectively (mg/L), $C_{A,eq}$ and $C_{O,eq}$ are the equilibrium concentrations of Nd^{3+} in the aqueous phase and in the organic phase, respectively (mg/L).

The mass transfer of the three-stage cascade were also tested with $Nd(NO_3)_3$ by using 30% TRPO-kerosene solution as the organic phase, and $Nd(NO_3)_3$-1 mol/L HNO_3 as the aqueous phase. Two kinds of tests were carried out. One was that all of three stages worked, the other was that only two stages worked and another stage stopped. The extraction rate was described as follows (13):

$$\rho = \frac{(C_{O,out} - C_{O,in})V_O}{C_{A,in}V_A} \times 100\%$$

where ρ is the extraction rate (%), V_O and V_A are the volume of the aqueous phase and the organic phase, respectively (L), the definition of $C_{O,in}$, $C_{O,out}$ and $C_{A,in}$ is the same as above.

RESULTS AND DISCUSSION

Hydraulic Performance of the Single-Stage Contactor

The hydraulic performance of the single-stage 70-mm centrifugal contactor is presented in Figs. 5 and 6. For the 70-mm annular centrifugal contactor, the nominal total flow (F) of two phases is a function of the flow ratio (aqueous/organic), the rotor speed, and the diameter of the heavy weir. At a definite rotor speed the F changes with the flow ratio and has a maximum. When the rotor speed is 2940 r/min, the nominal total flow can reach

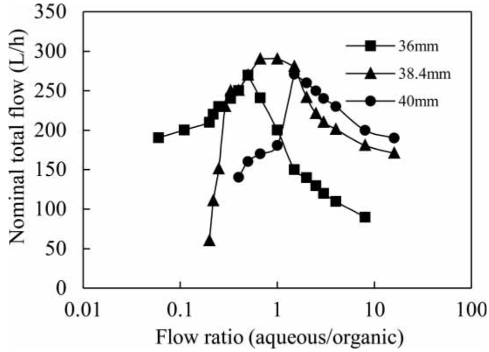

Figure 5. Variation of the nominal total flow with the flow ratio for various diameter of the heavy weir. The rotor speed: 2940 r/min.

290 L/h, and there is different operating range with the different diameter of the heavy weir. The heavy weir of 38.4 mm (diameter) is the best one among three heavy weirs, which are the heavy weir of 36.0 mm, the heavy weir of 38.4 mm and the heavy weir of 40.0 mm. This is because the contactor with the heavy weir of 38.4 mm can cover the expected operational range of the conditions, namely, the expected total flow (F) of two phases can reach 100 L/h when the range of flow ratio (aqueous/organic) is from 5/1 to 1/5 and the rotor speed is varied from 2000 r/min to 2800 r/min, but the contactor with one of two heavy weirs cannot cover the expected operational range of the conditions.

Hydraulic Performance of the Three-Stage Cascade

The hydraulic performance of the three-stage cascade is presented in Fig. 7. When the diameter of the heavy weir is 38.0 mm and the rotor speed is

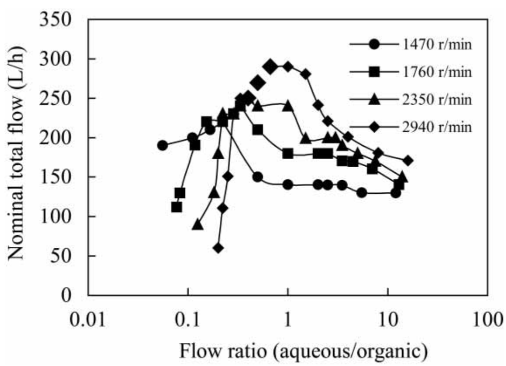

Figure 6. Variation of the nominal total flow with the flow ratio for various rotor speeds. Diameter of the heavy weir: 38.4 mm.

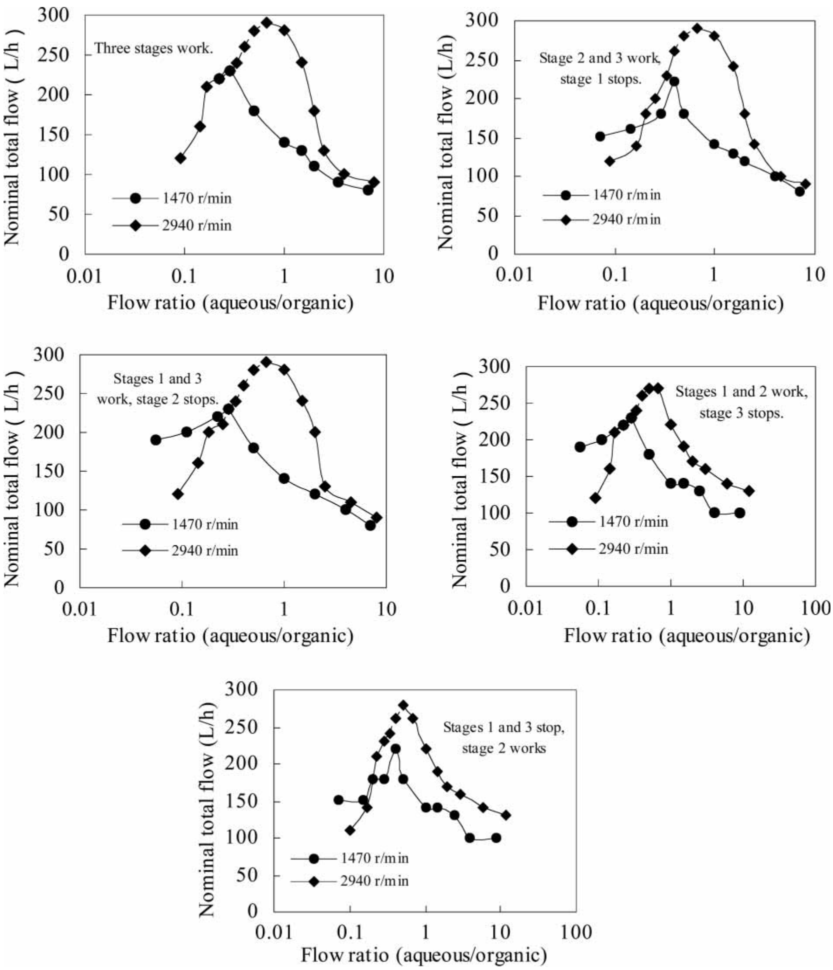

Figure 7. Variation of the nominal total flow with the flow ratio for various rotor speeds. Diameter of heavy weirs: 38.0 mm.

2940 r/min, the maximum nominal total flow of the three-stage cascade can reach 280 L/h, and the expected total flow of two phases of the cascade can reach 100 L/h when the range of flow ratio (aqueous/organic) is from 5/1 to 1/5. Moreover, even though one stage or two nonadjacent stages cease to work the cascade can continue to operate. The hydraulic performance of the cascades is different with the difference of the stopped stage. The hydraulic performance of the three-stage cascade can also satisfy the design goal. For the cascade with the different stopped stage or stages, the operating range and the nominal total flow in the same operating conditions are different, and variation of the nominal total flow of two phases with the flow ratio

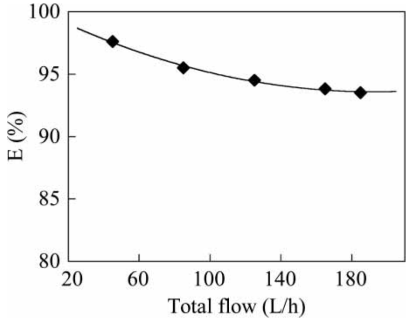

Figure 8. Variation of the efficiency (E) with the total flow. Flow ratio (aqueous/organic): 1/1; rotor speed: 2650 r/min.

(aqueous/organic) and the rotor speed is also different. Therefore the hydraulic performance of the cascade with the different stopped stage or stages may be determined by the working stages (or stage) and the stopped stage (or stages).

Mass Transfer Efficiency of the Single-Stage Contactor

The extraction stage efficiency (E) of the single-stage contactor for aqueous phase is presented in Figs. 8 and 9 for extracting Nd^{3+} with 30% TRPO-kerosene solution from $Nd(NO_3)_3$-1 mol/L HNO_3 solution, and in Figs. 10 and 11 for extracting Nd^{3+} with 30% TRPO-kerosene solution from the simulated HLLW. It can be seen that the E is greater than 95% at suitable operating conditions, namely, when the rotor speed is no less than 2650 r/min, the total flow of two phases is no more than 100 L/h, and the flow ratio (aqueous/organic) is 1/1 or 2/1, moreover, these operating conditions accord with the design requirements. When the total flow of two

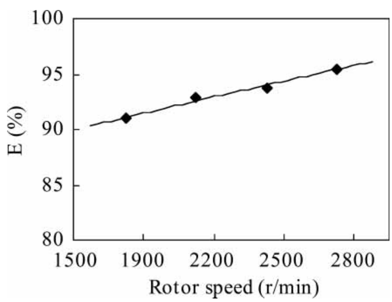

Figure 9. Variation of the efficiency (E) with the rotor speed. Flow ratio (aqueous/organic): 1/1; total flow: 80 L/h.

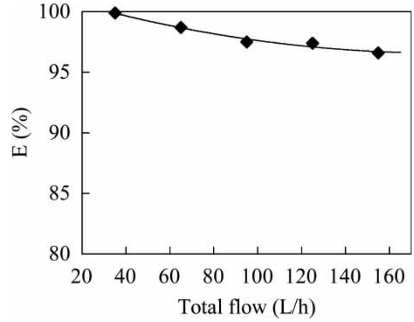

Figure 10. Variation of the efficiency (E) with the total flow. Flow ratio (aqueous/organic): 2/1; Rotor speed: 2650 r/min.

phases is increased at the given rotor speed and the flow ratio, the E decreases, because the mixing time of the two phases in the annular mixing zone is shorter when the total flow is increased. However, when the rotor speed of the contactor is increased at the given total flow of the two phases and a flow ratio (aqueous/organic), the E increases, because the mixing of the two phases is more sufficient when the rotor speed is increased. In addition, from the results shown in Figs. 8, 9, 10 and 11, the extraction stage efficiency using the simulated HLLW (Figs. 10 and 11) is slightly higher than that using just Nd^{3+} (Figs. 8 and 9). This is mainly because of the different flow ratio (aqueous/organic), namely, the former flow ratio is 2/1, while the latter flow ratio is 1/1. This result is the same as the previous study on extraction of nitric acid with 30% TRPO-kerosene solution (16).

Mass-Transfer Efficiency of the Three-Stage Cascade

The extraction rate (ρ) of the three-stage cascade is presented in Table 2 for extracting Nd^{3+} with 30% TRPO-kerosene solution from

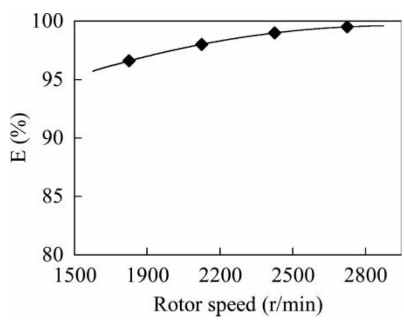

Figure 11. Variation of the efficiency (E) with the rotor speed. Flow ratio (aqueous/organic): 2/1; total flow: 60 L/h.

Table 2. Results of the three-stage cascade test for mass transfer

N	ω (r/min)	Q_A (L/h)	Q_O (L/h)	$C_{A,in}$ (mg/L)	$C_{O,in}$ (mg/L)	$C_{A,out}$ (mg/L)	$C_{O,out}$ (mg/L)	ρ (%)
3	1800	61.2	30	740.16	64.8	49.97	1468.8	93.0
3	2100	61.2	30	740.16	64.8	35.34	1480.3	93.75
3	2400	61.2	41.4	740.16	64.8	29.66	1094.4	94.1
2	1800	61.2	30	740.16	64.8	119.95	1324.8	83.4
2	2100	61.2	30	740.16	64.8	85.45	1385.2	87.4
2	2400	61.2	41.4	740.16	64.8	54.72	1072.8	92.1

Where N is number of working stages in the cascade test, ω is the rotor speed (r/min), Q_A and Q_O are the flow rate of the aqueous phase and the organic phase, respectively (L/h), $C_{A,in}$ and $C_{A,out}$ are the inlet and the outlet Nd^{3+}concentrations in the aqueous phase, respectively (mg/L), $C_{O,in}$ and $C_{O,out}$ are the inlet and the outlet Nd^{3+}concentrations in the organic phase, respectively (mg/L), ρ is the extraction rate (%).

$Nd(NO_3)_3$-1 mol/L HNO_3 solution. It is shown that at the given total flow of two phases and the given flow ratio, the ρ increases with the increase of the rotor speed, because the mixing of the two phases is more sufficient when the rotor speed is increased. When one stage of the cascade ceases to work, ρ is less than that of all three stages working at the same operational conditions, because the stopped stage has no mixing of the two phases, but phase separation. It is also shown that although one stage ceases to work, the cascade can continue to operate.

CONCLUSION

The new 70-mm annular centrifugal contactor for semi-industrial scale has been proved by a series of tests to have good hydraulic performance and mass-transfer efficiency, and satisfy the expected design goal. The modular design makes the contactor easy to be disassembled and assembled fast by simply moving modules up and down. With the overflow structure, even though one stage or nonadjacent stages in the multistage cascade cease to work, the cascade can continue to operate. All results show that the new 70-mm annular centrifugal contactor can be used in the semi-industrial scale test of the TRPO process in the future in INET, and has good prospect for nuclear industry use.

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