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August 6, 1984

M E M O R A N D U M

TO: H. D. HARMON

FROM: J. C. CHESNA/M. E. HODGES

MASS TRANSFER IN 12-CM CENTRIFUGAL CONTACTORS

INTRODUCTION

Oak Ridge National Laboratory (ORNL) requested SRL to design, build, and test two 8-stage centrifugal contactor units (8-packs). These 8-packs will be used in the ORNL Integrated Equipment Test Facility. These contactors are of an advanced design, which evolved from the original SRP contactors in use in F Area.

The SRL test program consisted of two phases. In the first, the hydraulic operation of the contactors was determined using 3N nitric acid and 30% tributyl phosphate in n-dodecane. The results of this study were reported in DPST-84-202, Hydraulic Study of 12-CM Centrifugal Contactors.[1] The second phase was to determine the mass transfer efficiency of an eight-stage array of centrifugal contactors. Since these contactors were designed for the first cycle of the conceptual Hot Experimental Facility (HEF), all the experiments were run at the HEF flowsheet operating conditions.

This memorandum describes the results of the mass transfer studies of an 8-stage unit operating in both extraction and stripping modes.

SUMMARY

One eight-stage unit (8-pack) of centrifugal contactors was tested in both extraction and stripping modes. Efficiencies approaching 100% were obtained in both modes. The contactors were operated successfully at a wide range of combined flow rates, including the HEF conditions.

DISCUSSION

Contactor Description

These contactors are of an advanced design (see Figure 1), which evolved from the original SRP design of the 1960's. The original contactors are currently in use as the F-Area 1-A bank. In this original design the two process streams, aqueous and organic, are fed to the base of the contactor where the two streams are intimately mixed with a paddle. This mixed solution then passes into the upper part of the housing where it enters the rotor. The two phases are separated by centrifugal force in the rotor.

With the advanced contactors, the separate phases enter the side of the contactor housing near its upper end. The phases are then mixed in the annulus between the housing and the rotor by Couette flow, which induces Taylor vortices. The mixed solution enters the rotor through a port in the base and is separated by centrifugal force. The aqueous phase, being heavier, is thrown to the wall of the rotor. The organic phase, being the lighter, remains in the rotor center. The aqueous and organic phases exit the rotor via a system of weirs. The location of the organic-aqueous interface is controlled by means of air pressure which is supplied to the aqueous overflow weir. Varying the pressure changes the radial position of the aqueous-organic interface. The interface is adjusted so there is minimal entrainment of either phase.

Extraction Efficiency

The Murphree stage efficiency relation is used to express the extraction performance of the 8-stage array.[2] The Murphree efficiency relations measure the approach of one of the effluent

streams to equilibrium with the other at its actual final concentration. Thus, the Murphree extract stage efficiency is

$$E_{ME} = \frac{Y_2 - Y_1}{Y_2^* - Y_1}$$

For the raffinate

$$E_{MR} = \frac{X_2 - X_1}{X_1 - X_2^*}$$

where

Y_1 = inlet extract concentration

Y_2 = outlet extract concentration

Y_2^* = theoretical equilibrium outlet concentration

X_1 = inlet raffinate concentration

X_2 = outlet raffinate concentration

X_2^* = theoretical equilibrium outlet concentration

The raffinate is that stream which originally contained the uranium. The extract is that stream which extracted the uranium. So, either the aqueous or organic phase could be the extract, or raffinate, depending upon the mode of contactor operation. An efficiency of each stage of the 8-pack were not determined, since sample ports were not located between each stage. Thus, the efficiencies calculated are for the whole bank. The theoretical equilibrium outlet concentrations were determined using the computer code SEPHIS-MOD4.[3]

Experimental

The first phase of mass transfer testing was with the 8-pack in the extraction mode. This condition entails uranyl nitrate in the aqueous phase being fed to the unit, where the uranium is transferred to the organic phase. A summary of run conditions is shown in Table 1.

The second phase of testing was with the contactors in a stripping mode, to transfer or "back-extract" the uranium into the aqueous phase. The conditions for this mode are different from those for the extraction run, and are shown in Table 2.

Two types of experiments were run in each mode, so that the approach to steady state and the efficiency of mass transfer at steady state, could be studied.

A typical experiment in the extraction mode took place as follows. The contactor motors were allowed to warm up at the test speed (1750 or 2400 RPM). The organic flow was started at a predetermined flow rate. The weir air pressure was set to a value calculated to be near the center of the operating envelope (see Figure 2).[1] The aqueous flow was then started and adjusted so the O/A ratio was 1.4, as specified in the HEF flowsheet. Samples of both outlet streams were taken at short, timed intervals for up to 2 hours after the initial startup. The data collected from repetition of this sequence at varying throughputs is summarized in Table 3.

The same experimental procedure was used for testing in the stripping mode. A summary of the data is shown in Table 4.

RESULTS

The 8-pack operated successfully in both extracting and stripping modes under the conditions of the HEF flowsheet. The mass transfer efficiency in both modes of operation approached 100%, at a wide range of O/A ratios and total throughputs, including the HEF conditions. This information is detailed in Tables 5 and 6 for the extraction mode and Table 7 for the stripping mode.

Sample ports were located in both outlets, as well as in the interstage piping between stages 4 and 5. This is shown in Figure 4. Uranium analyses were run on samples taken from the middle of the bank in all runs. The results are listed in Tables 8 and 9 for the extraction mode and the stripping mode. As can be seen from the tables, effectively all the mass transfer occurred in the first four stages of contactors. The feed concentration of 126.9 g U/L was reduced to the detectable limit concentration, which is on the order of 1 part per million (ppm). This attests to the high efficiency of the advanced centrifugal contactors.

The initial run allowed data to be collected for the contactor's approach to steady state. This initial run was the first introduction of uranium into the system. Thus, the change in uranium concentration in the organic outlet with time was plotted and fit to the following equation (Figure 3):

$$U_o = U_s \left(1-e^{-t/\tau}\right)$$

where

U_o = uranium outlet concentration, g/L

U_s = steady state uranium concentration, 88.7 g/L in the extract stream

τ = time constant of 8.3 min

t = time, min

This equation is of the form of a first-order lag system response to a step change in input. So, at 8.3 minutes, τ, the bank of 8 contactors is 63.2% toward reaching equilibrium, and at 33 minutes, the bank is 98% of steady-state operation.

The steady-state condition being discussed here is attained when the outlet concentrations of the bank do not change with time. The data points for the approach to steady state shown in Figure 3 are listed in Table 10. The low value obtained for the sample at t = 28 minutes is most likely due to experimental error in the analysis. Due to time constraints only one approach to steady state run at each concentration could be made.

A number of runs, as outlined in Table 6, used a feed of 51.74 g U/L. The conditions for these runs ranged from total throughputs of 1.92 to 5 gpm and O/A's of 0.29 to 0.56. Again, excellent extraction performance was exhibited, with efficiencies for the bank in excess of 99.9%.

PROGRAM

The mass transfer studies were the final phase of a series of tests requested by Oak Ridge National Laboratory (ORNL) as part of the Consolidated Fuel Reprocessing Program (CFRP). SRL coordinated the design, fabrication, and testing of the two 8-stage arrays of centrifugal contactors. The 8-packs were shipped to ORNL in June of 1983.

Additional studies are in progress on centrifugal contactors varying in diameter from 5.5 to 25 cm.

REFERENCES

1. J. C. Chesna and M. E. Hodges, "Hydraulic Study of 12-CM Centrifugal Contactors," DPST-84-202, April 1984.

2. Treybal, R. E., "Liquid Extraction," McGraw Hill, 2nd Ed., 1963.

3. Mitchell, A. D., "Sephis-MOD4: A User's Manual to a Revised Model of the Purex Solvent Extraction System," ORNL-5471, 1979.

TABLE 1

Run Conditions for Extraction Mode

Aqueous Phase	126.9 g U/L in 3.5M HNO_3
Organic Phase	30% TBP in n-DD
O/A	1.4, 0.56, 0.36, 0.29
Rotor Speed, rpm	1750, 1200, 2400
Throughput, gpm	1.9 - 5.0

TABLE 2

Run Conditions for Stripping Mode

Aqueous Phase	0.05M HNO_3
Organic Phase	38.7 g U/L in 30% TBP and 70% n-DD
O/A	0.8
Rotor Speed, rpm	1750
Throughput, gpm	2 - 6

TABLE 3

Summary of Data for Extraction Mode

Run No.	Rotor, rpm	Weir Pressure, in. H_2O	Total Flow, gpm	O/A Ratio	Temp, °C
1	1750	30	1.92	1.4	21.1
2	1750	30	5.02	1.4	21.1
3	1750	30	3.0	1.4	21.1
4	1750	30	1.92	0.56	30.3
5	1750	30	5.0	0.56	30.3
6	1200	12	2.46	0.56	30.8
7	1200	12	1.92	1.4	21.1
8	2400	60	2.46	0.56	30.8
9	2400	60	3.33	0.36	30.8
10	2400	60	3.88	0.29	30.8

TABLE 4

Summary of Data for Stripping Mode

Run No.	Rotor, rpm	Weir Pressure, in. H_2O	Total Flow, gpm	O/A Ratio	Temp, °C
1	1750	40	3.0	0.8	25.0
2	1750	30	5.0	0.8	25.0
3	1750	30	2.0	0.8	23.3
4	1750	30	6.0	0.8	23.3
5	1750	40	5.5	0.8	37.2
6	1750	40	6.0	0.8	37.2

TABLE 5

Efficiencies for the Extraction Mode Runs with Aqueous Feed of 126.9 g U/L

Run No.	Total Flow, gpm	O/A Ratio	Temp, °C	Uranium Concentration				
				Aqueous Outlet, g/L	Organic Outlet, g/L	Equilibrium Outlet* Aqueous, g/L	Equilibrium Outlet* Organic, g/L	% Efficiency**
1	1.92	1.4	21.1	4.3×10^{-5}	88.5	6.6×10^{-12}	88.6	99.9
2	5.02	1.4	21.1	2.7×10^{-5}	92.2	6.6×10^{-12}	88.6	99.9
3	3.0	1.4	21.1	2.9×10^{-5}	82.9	6.6×10^{-12}	88.6	99.9
7	1.92	1.4	21.1	2.4×10^{-5}	74.5	6.6×10^{-12}	88.6	99.9

* Determined by computer code SEPHIS-MOD4.

** Aqueous Basis.

TABLE 6

Efficiencies for the Extraction Mode Runs with Aqueous Feed of 51.7 g U/L

Run No.	Total Flow, gpm	O/A Ratio	Temp, °C	Uranium Concentration				% Efficiency**
				Aqueous Outlet, g/L	Organic Outlet, g/L	Equilibrium Outlet* Aqueous, g/L	Equilibrium Outlet* Organic, g/L	
4	1.92	0.56	30.3	3.2×10^{-4}	75.2	1.27×10^{-7}	90.4	99.9
5	5.0	0.56	30.3	1.8×10^{-4}	78.9	1.33×10^{-7}	90.6	99.9
6	2.46	0.56	30.8	2.7×10^{-5}	73.9	1.4×10^{-7}	90.9	99.9
8	2.46	0.56	30.8	4.4×10^{-3}	73.7	1.4×10^{-7}	90.9	99.9
9	3.33	0.36	30.8	2.7×10^{-4}	85.2	4.2×10^{-2}	100.8	99.6
10	3.88	0.29	30.8	13.5	93.9	12.2	100.7	96.9

* Determined by computer code SEPHIS-MOD4.

** Aqueous Basis.

TABLE 7

Efficiencies for the Stripping Mode Runs with Organic Feed of 38.7 g U/L

Run No.	Total Flow, gpm	O/A Ratio	Temp, °C	Uranium Concentration				
				Aqueous Outlet, g/L	Organic Outlet, g/L	Equilibrium Outlet* Aqueous, g/L	Equilibrium Outlet* Organic, g/L	% Efficiency**
1	3.0	0.8	25.0	33.3	0.65	30.4	3.6×10^{-4}	98.3
2	5.0	0.8	25.0	33.4	0.50	30.4	3.6×10^{-4}	98.7
3	2.0	0.8	23.3	30.9	0.28	30.4	3.6×10^{-4}	99.3
4	6.0	0.8	23.3	32.1	0.28	30.4	3.6×10^{-4}	98.3
5	5.5	0.8	37.2	30.5	0.39	30.4	3.6×10^{-4}	97.5
6	6.0	0.8	37.2	33.0	0.66	30.4	3.6×10^{-4}	98.3

* Determined by computer code SEPHIS-MOD4.

** ORGANIC BASIS.

TABLE 8

Summary of All Data Collected in the Extraction Mode

Run	Time	Aq-out Uranium ppm	Aq-out Acid M	Aq-out SpG	Aq-aid Uranium ppm	Aq-aid Acid M	Aq-aid SpG	Org-out Uranium g/l	Org-out Acid M	Org-out SpG	Org-aid Uranium ppm	Org-aid Acid M	Org-aid SpG
1	8	0.02	2.29	1.0699	0.04	2.58	1.0771	57.57	0.36	0.8878	0.17	0.54	0.8250
	19	0.02	3.36	1.1023	0.25	3.83	1.1156	79.12	0.27	0.9264	0.27	0.78	0.8298
	28	0.03	3.36	1.1094	0.08	3.80	1.1200	76.25	0.27	0.9234	0.31	0.73	0.8299
	58	0.04	3.49	1.1105	0.23	3.87	1.1210	88.50	0.22	0.9304	0.35	0.74	0.8299
2	4	0.22	3.48	1.1087	0.25	3.85	1.1174	85.80	0.25	0.9274	1.51	0.73	0.8297
	34	0.10	3.33	1.1081	0.17	3.92	1.1188	85.53	0.21	0.9342	0.50	0.71	0.8301
	64	0.27	3.47	1.1084	0.27	3.91	1.1193	92.21	0.22	0.9330	0.35	0.75	0.8303
3	5	0.21		1.1085	0.42	3.73	1.1193	87.11	0.21	0.9360	0.32	0.84	0.8299
	19.2	0.29		1.1173	0.18		1.1067	82.95		0.9287	0.23		0.8291
4	5	0.14	2.95	1.0838	2.71	2.89	1.0920	81.76	0.18	0.9296	7.71	0.64	0.8245
	10	0.71	2.90	1.0816	4.42	2.92	1.0894	78.11	0.20	0.9255	19.98	0.50	0.8243
	15	0.71	2.92	1.0825	3.45	2.94	1.0903	74.79	0.22	0.9212	31.19	0.52	0.8245
	30	0.35	2.91	1.0829	9.69	2.96	1.0901	78.28	0.21	0.9244	29.51	0.53	0.8246
	60	0.32	2.98	1.0827		2.95	1.0001	75.18	0.23	0.9211	23.35	0.51	0.8247
5	0.5	0.17	2.91	1.0826	5.09	2.86	1.0888	69.90	0.26	0.9146	0.89	0.50	0.8249
	7.5	0.18	2.90	1.0833	7.30		0.9944	76.81	0.21	0.9240	19.38	0.45	0.8245
	16.5	0.16	2.88	1.0834	0.20		0.9922	83.19	0.19	0.9281	24.08	0.47	0.8245
	49.5	0.23	2.89	1.0834	11.20		1.0860	78.78	0.20	0.9225	25.70	0.47	0.8241
	79.5	0.16	2.86	1.0826	0.04		0.9932	79.01	0.20	0.9219	28.78	0.50	0.8243
6	5	0.23	2.76	1.0885	58.12	3.00	1.0881	77.21	0.21	0.9187	20.00	0.61	0.8296
	10	0.22	2.80	1.0889	4.63	2.92	1.0880	82.33	0.20	0.9206	9.22	0.63	0.8289
	15	0.32	2.84	1.0884	9.67	2.98	1.0878	77.60	0.19	0.9247	17.67	0.62	0.8313
	30	0.24	3.18	1.0879	9.85	2.85	1.0880	78.33	0.21	0.9240	20.36	0.62	0.8307
	60	0.27	2.91	1.0871	8.48	2.93	1.0878	73.89	0.20	0.9191	19.07	0.62	0.8300
7	28	0.02	2.99	1.1000	0.21	3.36	1.1245	74.47	0.30	0.9478	0.28	0.46	0.8463
8	5	0.36	2.85	1.0855	4.64	2.92	1.0863	77.44	0.20	0.9189	29.55	0.68	0.8284
	10	0.29	2.83	1.0857	7.22	1.97	1.0863	77.21	2.04	0.9225	21.64	0.64	0.8277
	15	0.25	2.88	0.1858	6.48	2.86	1.0865	72.72	0.20	0.9217	22.18	0.62	0.8279
	30	0.29	2.86	1.0860	4.74	2.94	1.0864	73.83	0.21	0.9191	17.46	0.63	0.8277
	60	0.25	2.93	1.0843	4.34	2.96	1.0867	73.67	2.03	0.9197	37.99	0.64	0.8277
9	25	0.27	2.94	1.0851	327.17	2.90	1.0869	85.25	0.21	0.9361	0.59	0.62	0.8279
10	28	13460	2.79	1.0969	650.04	2.96	1.0872	93.86	0.20	0.9432	91040.00	0.12	0.9449

	Uranium g/l	Acid M	SpG	TBP %
Aqueous Feed	126.86	3.54	1.2588	
Organic Feed	2.5E-04	0.49	0.8251	29.13

TABLE 9

Summary of All Data Collected in the Stripping Mode

Run	Time	Aq-out Uranium g/l	Aq-out Acid M	Aq-out SpG	Aq-mid Uranium g/l	Aq-mid Acid M	Aq-mid SpG	Org-out Uranium ppm	Org-out Acid M	Org-out SpG	Org-mid Uranium g/l	Org-mid Acid M	Org-mid SpG
1	10	33.28	0.30	1.0647	20.55	0.05	1.0336	0.77	0.002	0.8079	30.00	0.006	0.8749
	62	33.28	0.30	1.0666	7.34	0.05	1.0223	0.65	0.002	0.8080	11.12	0.005	0.8350
2	16	34.45	0.30	1.0668	9.99	0.05	1.0196	0.55	0.002	0.8073	15.56	0.006	0.8382
	60	33.37	0.33	1.0650	NR	NR	NR	0.50	0.002	0.8073	11.91	0.005	1.0496
3	15	29.55	0.36	1.0459	15.90	0.07	1.0181	311.65	0.003	0.8089	21.12	0.006	0.8380
	34	30.99	0.36	1.0486	13.65	0.07	1.015	275.76	0.003	0.8083	16.08	0.005	0.8334
	69	30.83	0.36	1.0484	9.73	0.07	1.0093	285.81	0.002	0.8089	12.32	0.004	0.8264
	109	30.88	0.36	1.0487	8.65	0.06	1.0078	280.07	0.002	0.8083	11.31	0.004	0.8243
4	5	34.00	0.33	1.0514	NR	0.05	NR	274.33	0.002	0.8089	18.71	0.005	0.8346
	10	29.67	0.35	1.0473	NR	NR	1.016	257.10	0.002	0.8086	8.50	0.004	0.8193
	31	29.33	0.36	1.0463	NR	NR	NR	283.85	0.002	0.8087	7.75	0.004	0.8876
	59	29.87	0.36	1.0488	NR	NR	NR	359.22	0.002	0.8080	12.97	0.005	0.8247
	119	32.10	0.36	1.0510	NR	NR	NR	448.41	0.002	0.8081	17.66	0.005	0.8317
5	10	30.49	0.33	1.0492	NR	NR	NR	384.10	0.002	0.8070	4.53	0.004	0.8184
	19	33.01	0.53	1.0555	NR	NR	NR	663.00	0.002	0.8072	28.51	0.004	0.8436
	34	30.49	0.32	1.0454	NR	NR	NR	389.00	0.002	0.8074	1.43	0.005	0.8076
6	7	34.00	0.37	1.0512	NR	NR	NR	949.00	0.003	0.8086	8.34	NR	0.8172
	31	34.18	0.37	1.0507	NR	NR	NR	359.00	0.002	0.8069	3.69	0.003	0.8102

	Uranium g/l	Acid M	SpG	TBP %
AQUEOUS FEED	0.00	0.05	1.0016	
ORGANIC FEED	31.60	0.30	0.8774	31.60

TABLE 10

Organic Outlet Uranium Concentration vs. Time for the Approach to Steady State Determination

Time, min.	Uranium Concentration
0	0
8	57.6
19	79.1
28	76.2
58	88.5

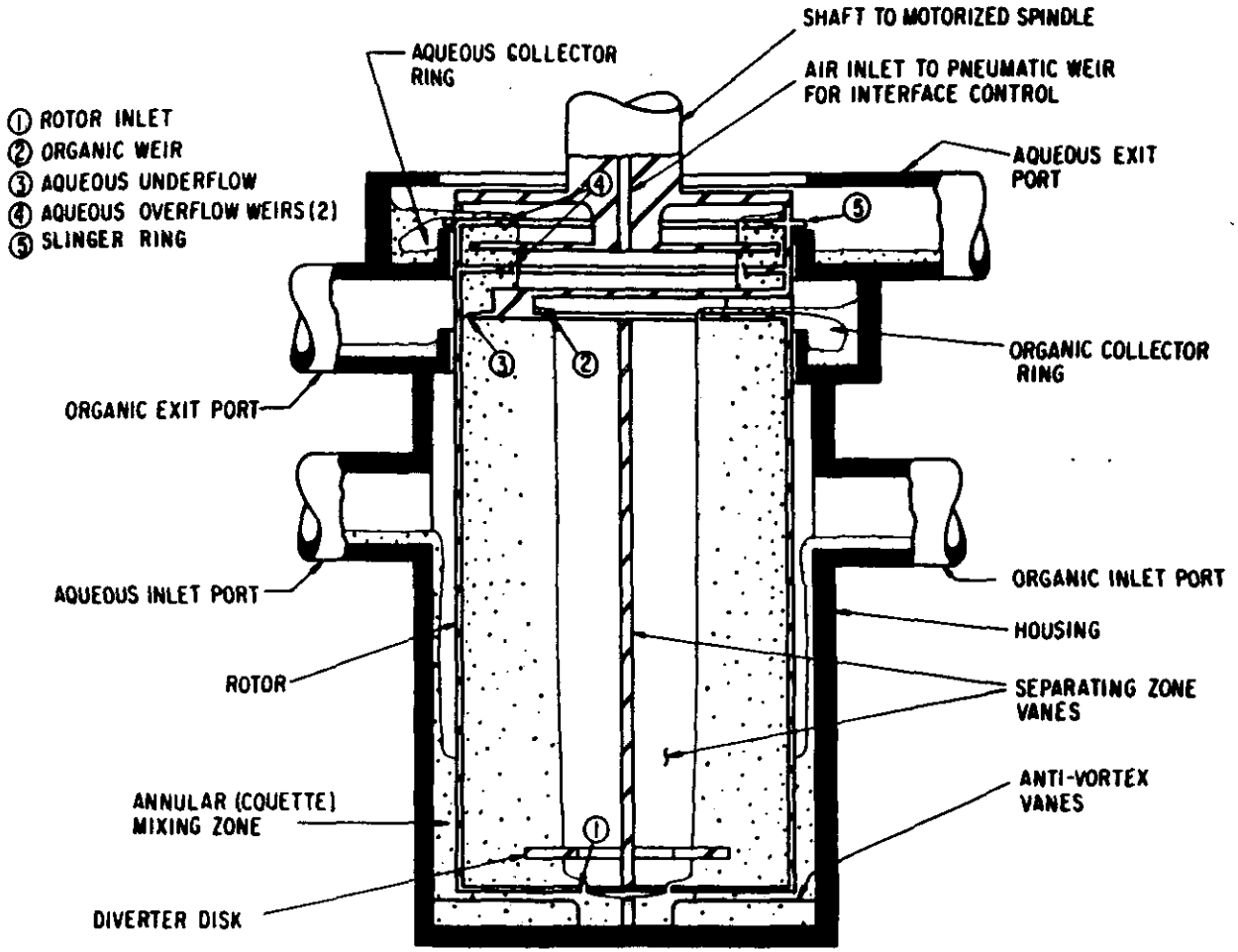

FIGURE 1. Schematic of the Advanced Centrifugal Contactor with Air Controlled Aqueous Weir

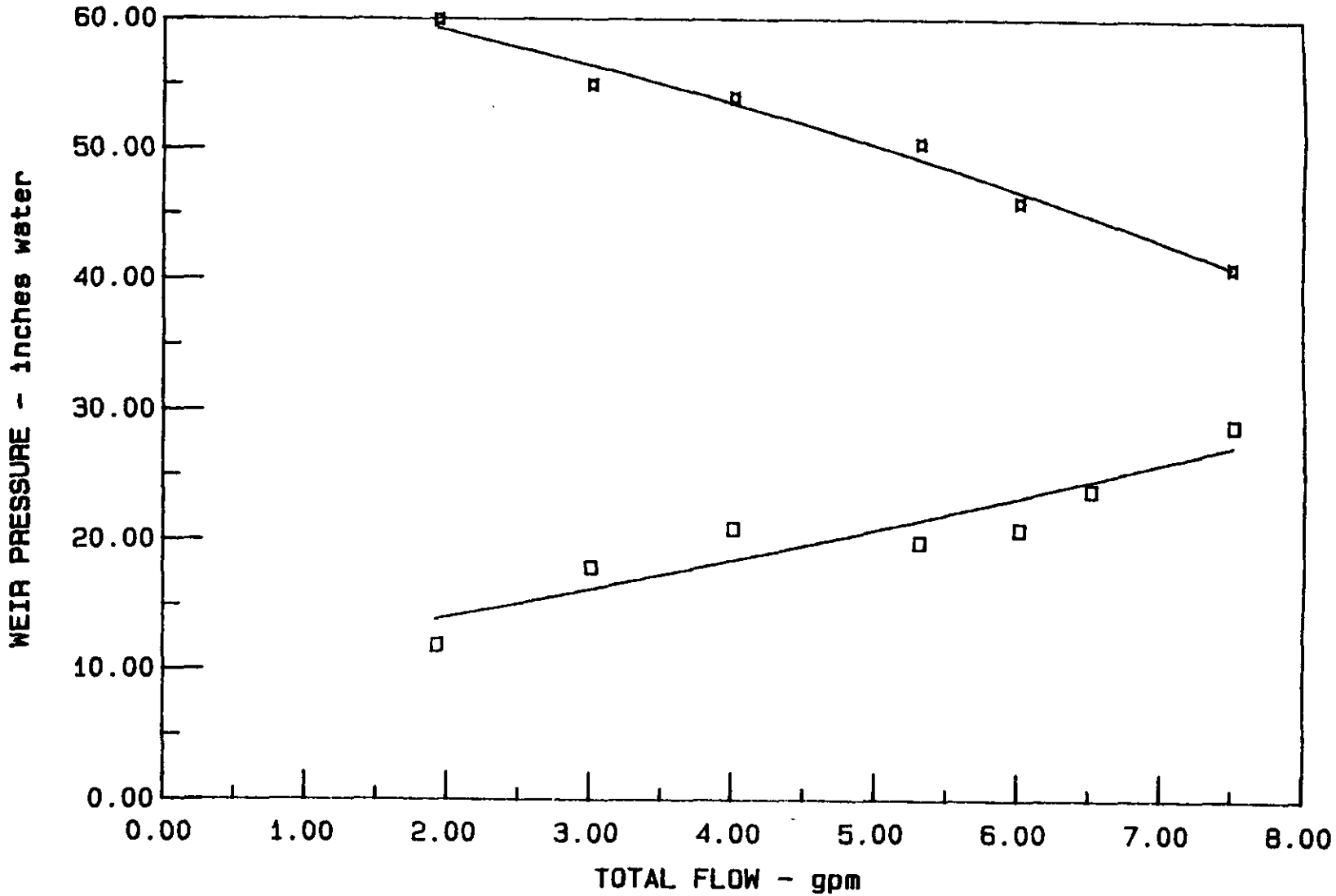

FIGURE 2. Operating Envelope for 8-Pack 8-I at 1750 rpm and O/A = 1.4

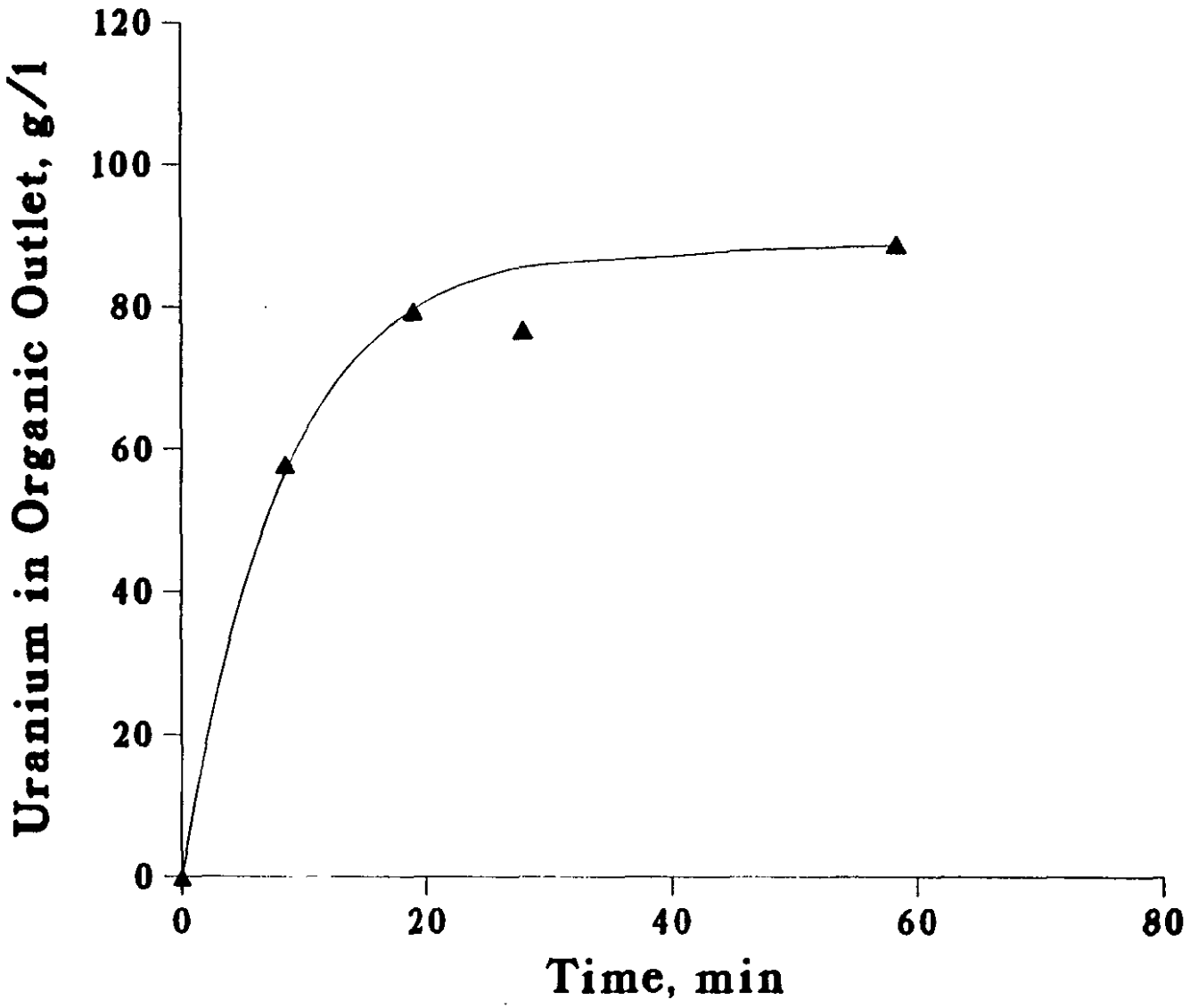

FIGURE 3. Approach to Steady State in an 8-Stage Array of Centrifugal Contactors

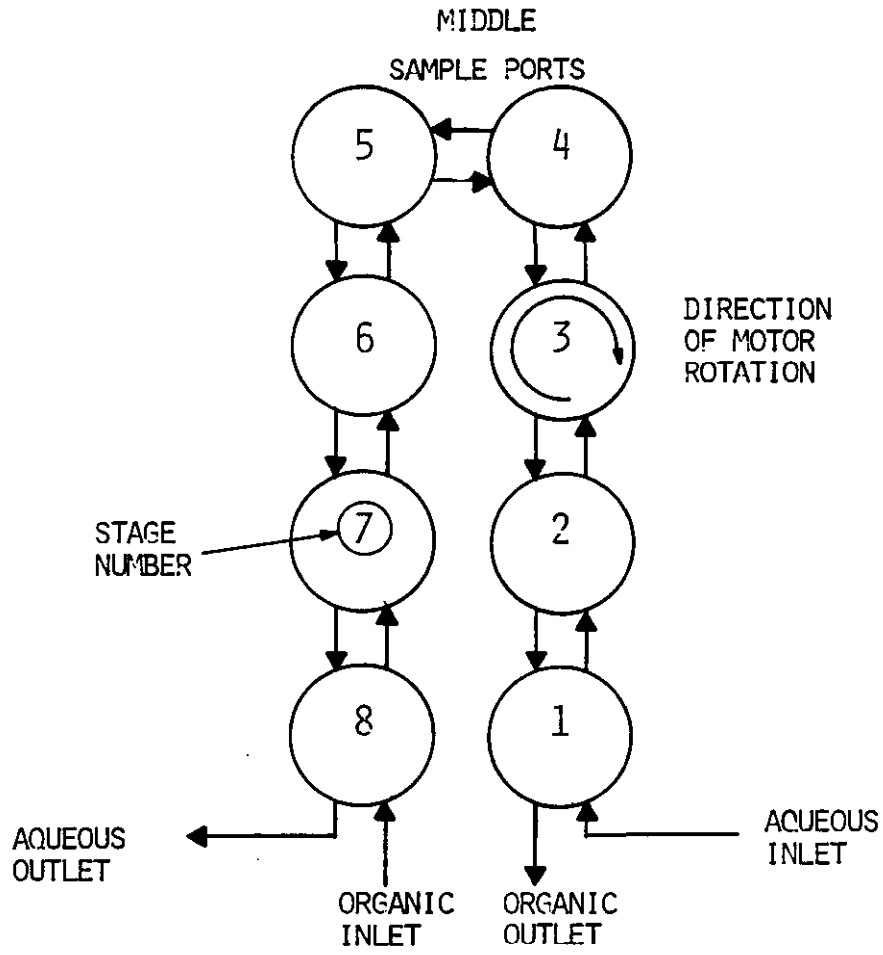

FIGURE 4. Top View of an 8-Pack of Centrifugal Contactors