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A BENCH-SCALE NATURAL-RECIRCULATION DISSOLVER

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ATOMIC ENERGY DIVISION

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A BENCH-SCALE NATURAL-RECIRCULATION DISSOLVER

by

E. E. Erickson

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Atomic Energy Division

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A BENCH-SCALE NATURAL-RECIRCULATION DISSOLVER

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E. E. Erickson

A B S T R A C T

A natural-recirculation dissolver closely approaching a stirred-tank reactor in behavior has been developed, and its feasibility has been demonstrated in the mercury-catalyzed dissolution of aluminum in nitric acid. It was designed to utilize the heat of reaction and evolution of gaseous reaction products for recirculation and mixing. The dissolvent was 5-molar nitric acid, containing mercury in concentrations from 10^{-4} to 10^{-3} molar as a catalyst. Dissolution rates for 1/7 scale, simulated MTR fuel elements of 2-S aluminum ranged from 0.25 to 4.0 mg/(cm²)(min), depending on the catalyst concentration in the dissolvent and the dissolvent flow rate. For comparable catalyst and acid feed concentrations, dissolution rates, based on unit net superficial velocities, were nearly 25 times those for dissolution of randomly-packed flat plates in a pilot plant, continuous, flooded dissolver.

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TABLE OF CONTENTS

	<u>Page</u>
I. SUMMARY	9
II. INTRODUCTION	11
III. BENCH-SCALE DISSOLVER	12
A. Equipment	12
1. Fuel Element	12
2. Dissolver	13
3. Auxiliaries	14
B. Method of Operation	15
IV. LABORATORY TESTS	17
A. Experimental Program	17
B. Data and Results	17
C. Discussion	19
V. ACKNOWLEDGEMENT	22
VI. REFERENCES	23

TABLE

DATA FROM STEADY-STATE DISSOLUTIONS	18
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LIST OF FIGURES

Figure 1	Aluminum Fuel Element for Laboratory Dissolver.	13
Figure 2	Schematic of Concentric-Tube Natural-Recirculation Dissolver (Laboratory Model).	13
Figure 3	Dissolution Rates in Recirculating Dissolver at Several Catalyst Concentrations.	20

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A BENCH-SCALE NATURAL-RECIRCULATION DISSOLVER

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I. SUMMARY

A semi-continuous recirculating dissolver, closely approaching a stirred-tank reactor in behavior, was built and successfully operated over a wide range of dissolvent flow rates for the mercury-catalyzed dissolution of aluminum in nitric acid. At similar catalyst and acid-feed concentrations, dissolution rates for 2-S aluminum parallel-plate elements were nearly 25 times as great as rates estimated from pilot plant data (1) for randomly-packed flat plates of the same material in a continuous flooded dissolver. The comparison was made in the linear operating range, and was based on unit superficial velocity of dissolvent.

In the recirculating dissolver, the evolution of gaseous reaction products and steam generated by the heat of reaction caused rapid circulation and mixing of the liquid phase. The flow was upward through a central dissolver tube, the liquid overflowing into an annulus and returning to the bottom of the tube. Fresh acid feed (containing catalyst) was injected at the bottom of the dissolver, where it mixed with the recirculating liquid. Product overflow through a jack-leg maintained a constant liquid holdup in the system. A reflux condenser returned condensate to the dissolver, and non-condensables were taken to an off-gas system maintained at about 2 inches-of-water vacuum. Feed and discharge were continuous over most of the dissolution period for the single-element batch.

The dissolver was fabricated of Pyrex pipe with stainless steel end-plates and connecting tubing. The central dissolver tube was 3-inch Pyrex pipe, 10 inches long, and the outer tube was 4-inch Pyrex pipe. Acid feed was heated to near its boiling point, and the dissolver was heated electrically to compensate for heat loss at low dissolution rates.

The fuel elements were fabricated from 16-gage 2-S aluminum sheet. Average retention times (dissolver holdup/feed rate) ranged from 0.13 to 2.1 hours. The dissolvent was 5-molar nitric acid with mercury concentrations ranging from 1×10^{-4} to 1×10^{-3} molar.

Dissolution rates varied from about 0.25 to 4.0 mg/(cm²)(min). The dissolution rates for 5×10^{-4} and 1×10^{-3} molar catalyst were identical and, at these catalyst concentrations, varied inversely in proportion to the retention time (or directly with the feed rate) for the range of flow rates covered. At lower catalyst concentrations, dissolution rates were identical only at high retention times, dropping off from linearity as the time decreased or when the flow rate was too high to allow efficient acid utilization.

II. INTRODUCTION

A dissolver was needed to fit the concept of an on-site processing plant to serve a single nuclear reactor, a so-called "miniature plant". The plant was to be designed to process one fuel element per day from an MTR-type reactor. The requirements for the dissolution of the fuel elements were set by the following considerations:

1. Limitations in the time allowable for development of technology and instrumentation demanded that present technology be utilized as far as possible.
2. Space limitations dictated a continuous or semi-continuous process in which the dissolver size would be established by the size of the fuel element rather than by the volume of a batch of dissolvent.
3. The desirability of continuous measurement of product concentration, either directly or indirectly, meant that sampling of the dissolver or of the dissolver product, with little time lag, was essential.
4. For effective control, there should be no rapid fluctuations in concentration of the dissolver contents or effluent.

The above requirements appeared to be met best by a dissolver operating on the semi-continuous stirred-tank principle. The semi-continuous feature, the continuous flow of dissolvent throughout most of the dissolution period for one fuel element, would permit the size of the dissolver to be determined primarily by the dimensions of the element. Also, the complications of feeding elements to an operating dissolver, and the requirement of additional space for adding an element while part of another remained in the dissolver, could be avoided. Stirred-tank behavior, or rapid and complete mixing, would provide a product of the same composition as the dissolver contents, with composition changes occurring only gradually. It thus would be possible to sample a product solution that represented the actual composition in the dissolver. Instrumentation would present no problems not associated with either batch or true continuous operation.

A study was undertaken to develop an efficient dissolver which would meet the requirements for the miniature plant and have general application in the processing of nuclear fuels, and to evaluate a small-scale model of the dissolver in the laboratory.

III. THE BENCH-SCALE DISSOLVER

There are several means of obtaining mixing in a chemical reactor. Probably the simplest and most practicable means in a radioactive and corrosive atmosphere for most fuel element dissolutions is the utilization of the evolved gases to obtain rapid circulation.

A vertical continuous dissolver packed with fuel elements, in which acid is fed to the bottom and product is taken off at the top, called a continuous flooded dissolver, theoretically gives highly efficient tubular flow ⁽²⁾. In practice, however, gaseous reaction products and steam generated by the heat of reaction create violent surging and mixing ⁽¹⁾. The liquid holdup in the column accounts for only a small part of the column void space, and contact between metal and acid appears to be quite sporadic. The mixing is so great that very little concentration gradient is found over the length of the column; the behavior more closely approaches that of a stirred tank. By providing means for the liquid carried upward through the dissolver by the gas evolution to return to the bottom for recirculation, the surging can be avoided, the holdup increased, and the contact improved.

Two means for recirculation were considered and given preliminary laboratory evaluation: an external leg, and an annular space around the dissolver tube. Although both could be considered satisfactory, the annular-space method gave better heat utilization and smoother operation. It was thus the method chosen for further development.

A. Equipment

1. Fuel Element

The size of the mock element for the dissolution study was selected to fit available standard Pyrex pipe. Based on the surface area and weight, features which control the overall dissolution rate and time, a scale of approximately 1/7 of an MTR element proved suitable for a 3-inch dissolver tube. The ratio of length to width and to thickness of the element was held roughly the same for the model as for a full-sized element, but the number of plates was reduced. Because considerable laboratory and pilot plant dissolution data were available for comparison, 2-S aluminum was selected for the mock element. The design of this element is shown in Figure 1.

MATERIAL: 16 GAUGE 2-S ALUMINUM SHEET
 SPACERS BETWEEN PLATES: 2- 3/4" DIA - 16 GA. AL. WASHERS
 TIE BOLTS: 1/4" STAINLESS STEEL BOLTS & NUTS

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Figure 1

Aluminum Fuel Element for Laboratory Dissolver

2. Dissolver

The bench-scale dissolver contained about 2.4 liters of solution during operation. It was constructed of standard Corning Double-Tough Pyrex pipe with stainless steel end-plates and interconnecting tubing. Pertinent details of the dissolver are shown in Figure 2.

Figure 2

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Schematic of Concentric-Tube Natural-Recirculation Dissolver (Laboratory Model)

The inner - or dissolver - tube was a 10-inch length of standard 3-inch Corning Pyrex pipe having a uniform outside diameter over its entire length. It rested on a stainless steel distributor plate 3-3/8 inches in diameter and 1/2 inch thick. The distributor plate was attached through a feed funnel and 3/8-inch feed tube to the bottom end-plate of the outer tube of the dissolver. The distributor plate allowed the acid feed from the feed tube and funnel to enter the dissolver tube through two concentric rings of 1/16-inch holes in the plate (through 38 holes). In the outer part of the distributor plate, outside of the feed funnel, 150 holes in four concentric rings allowed entry of the recirculating solution from the annulus. The general arrangement and the flow through the plate are indicated in Figure 2. It might appear that this arrangement would channel the incoming acid feed and cause dissolution in the center of the element to be more rapid than elsewhere. No significant evidence of such action was found in partially dissolved elements. Dissolutions with other distributor plates indicated that the design details of the dissolver-tube support and the distributor have little influence on the dissolver performance. A conical retaining screen of perforated stainless steel sheet at the top of the dissolver tube prevented the carryover of solids into the recirculation loop or product overflow.

The outer tube was a section of 4-inch Pyrex pipe 24 inches long, attached by Corning flanges to the end-plates. Teflon gaskets were used. A water-cooled reflux condenser was connected to the top of the dissolver by a 6-inch length of 1-inch Schedule-40 stainless steel pipe. The condensing coil consisted of about 23 feet of 1/4-inch stainless steel tubing housed in the annulus between a section of 3-inch Pyrex pipe and a 1-inch Pyrex tube. The tube simulated a charging chute. The condenser was vented to an exhaust system under a vacuum of about 2 inches of water.

3. Auxiliaries

Dissolvent was fed to the dissolver by a multiple-head Milton Roy Mini-Pump. For most tests, two heads were used in parallel for a maximum feed rate of about 165 ml/min; in one test, however, four heads were connected for a 300 ml/min rate. Electrical heating tapes wrapped around the feed tube and the lower part of the dissolver were used to heat the feed to near its boiling point and to heat the dissolver to compensate for heat loss at low dissolution rates.

Product was discharged through a jack-leg overflow; the height of the jack-leg controlled the liquid holdup in the dissolver or, in effect, the submergence for the vapor-lift action. The height could be varied, but an overflow at the same level as the top of the dissolver tube was satisfactory and was used for all runs.

Temperature measurements, one in the dissolver tube, one in the vapor space, and one in the cooling water, were made with iron-constantan thermocouples in stainless steel thermowells and recorded on a multirange Brown "Electronic" recorder.

There were no means of direct, continuous measurement of aluminum concentrations readily available. Since specific gravity could be measured with available equipment, it was used for monitoring the product composition. The sampling system for this measurement consisted of: (1) an air lift to carry solution from the bottom of the annular space of the dissolver to a disengaging pot; (2) a cooling coil, through which solution flowed downward by gravity from the pot, to provide a constant-temperature sample; (3) a vented sample chamber of 1-inch Pyrex pipe, 12 inches long, arranged for an upward flow of sample; and (4) an overflow line to return the sample stream to the dissolver and product line. Two 1/4-inch stainless steel tubes were inserted in the sample chamber, at a 10-inch separation in elevation, as specific gravity probes. They were supplied with filtered air from a constant pressure (20 psig) source through metering bubble chambers. The signal was transmitted pneumatically to a Minneapolis Honeywell density recorder by a Taylor differential-pressure transmitter calibrated for a specific-gravity range of 1.0 - 1.5. While specific gravity was not as sensitive a measurement of product concentration as was desired, it proved to be a satisfactory indicator of the attainment of steady state and of significant changes in process conditions.

B. Method of Operation

There are several options for operating procedures with a recirculating dissolver. With an appropriate type and shape of fuel, continuous charging of fuel gives a continuous process; or with a unit of adequate size, batch operation is possible. Semi-continuous operation also is feasible. For the latter, one charge of fuel constitutes a batch, but dissolvent feed and discharge are continuous over the greater part of the dissolution cycle. The semi-continuous process was applicable for dissolution of MTR-type fuel, and operation was simplified because a parallel-plate element has a nearly constant area exposed over most of the dissolution period. The mode of operation found most suitable for this particular element is outlined below. The general procedure, with minor modifications, would be applicable for a full-scale MTR fuel dissolving unit, and could be adapted to automatic control.

(1) Charging: The laboratory model was charged by removing the bottom end-plate, distributor plate, and dissolver tube, placing the element in the dissolver tube, and reassembling the dissolver. Obviously, such techniques could be considered only in the case of a small test unit with non-radioactive materials.

(2) Startup: Dissolvent feed to the dissolver at the run conditions could be started with the dissolver containing only the element, and steady state would be reached shortly after the dissolver was filled. For more rapid attainment of steady state, however, the dissolver was first filled with product from a previous run and heated. When the dissolver contents were near boiling or the reaction was well underway, addition of heated dissolvent feed at the rate predetermined for the run was begun. Steady state generally was approached closely in less than one-half hour. The preheating of the feed was continued throughout the run period, but heating of the dissolver was continued only at low reaction rates where it was needed to compensate for heat loss.

(3) Steady state period: A leveling off of the specific gravity to a constant value indicated the attainment of steady state. Acid feed was continued at the preselected rate until a decrease in specific gravity, usually accompanied by a drop in dissolver temperature, indicated a decrease in dissolution rate.

(4) Heel dissolution: While several methods were tried for completing the dissolution of the plate fragments near the end of the cycle, only that most readily followed with the dissolver instrumentation is described. Following indication of a decrease in dissolution rate, the dissolver was heated; when the specific gravity dropped again, the catalyst concentration in the feed was increased by a factor of five to ten. On further decrease in specific gravity, the acid feed was shut off and heat to the dissolver was continued to hold the contents at the boiling point. If the specific gravity then increased, the feed was started again and continued until a further decrease was noted. Heating was continued for about 1/2 hour after no further change in gravity was noted.

(5) Shutdown: The dissolver was drained, flushed with water, and the bottom end-plate removed for recharging. For an installation in which a drain line is impractical, the dissolver could be emptied readily by an air lift.

Where dissolution-rate data under steady state conditions were to be obtained, the dissolver was operated at apparent steady state (stage 3 above) until several samples had been taken at 10- to 15-minute intervals. Approximately 15 ml, or 0.6 per cent of the dissolver contents, was withdrawn during sampling to flush the sample line and provide a sample. The acid-feed rate was then changed to the next desired value, the dissolver again allowed to reach apparent steady state, and sampling repeated. Runs could be made at as many as three flow rates before an element began to disintegrate and the surface area to change appreciably.

IV. LABORATORY TESTS

A. Experimental Program

The experimental program was intended to test briefly the concept of the dissolver, the operating procedure, and the means of control. Thus, the program was primarily a qualitative study, with a limited quantitative evaluation in the range of flow rates and catalyst concentrations of direct interest to full-scale dissolution of one MTR fuel element per day. The general procedure for the experimental runs was described in the previous section. All runs were made at the boiling point of the solution.

The feed was 5M HNO_3 containing 1×10^{-4} , 2×10^{-4} , 5×10^{-4} , or 1×10^{-3} M $\text{Hg}(\text{NO}_3)_2$. Feed solutions were prepared to 5.00 ± 0.05 M acid. Catalyst was prepared by dissolving a weighed quantity of mercury in nitric acid.

From visual observation, the time for complete mixing in the dissolver was estimated to be about 5 seconds. The mixing time was determined by injecting a dichromate solution into the top of the dissolver and noting the time required for the maximum color intensity to reach the product sampler. The observed time was corrected for the time delay in the sample line.

The dissolution rates were based on the aluminum concentration in the product and on the 2750 cm^2 surface area calculated from the dimensions of the element. Precision and accuracy of aluminum and acid analyses, at the 95 per cent confidence level, were as follows:

Acid precision	= ± 0.03 N
accuracy	= zero bias

Aluminum precision	= ± 0.41 g/l
accuracy	= slight negative bias (about 0.01 g/l)

Most of the results were obtained from data taken where steady state was reasonably established, i.e., where no further trend in the aluminum concentration was noted.

B. Data and Results

The data and calculated dissolution rates are tabulated on page 18.

The concentrations are averages of two or more samples. Since the primary objective of the experimental program was a demonstration of the operability of the dissolver, no attempt was made to analyze the data statistically.

DATA FROM STEADY STATE DISSOLUTIONS

Run	Feed Conditions				Product		Dissolution Rate	Stoichiometry	
	Hg ⁺⁺ Conc. M	HNO ₃ Conc. M	Feed Rate Liters/Hr.	Retention Time, Hrs.	Al ³⁺ , M	H ⁺ , M	mg/(cm ²)(min)	Moles Acid Consumed	Mole Al Dissolved
1	2 x 10 ⁻⁴	5.04	3.6	0.67	1.36	0.34	0.802		3.64
2		5.01	1.2	2.00	1.35	0.37	.265		3.44
3			2.4	1.00	1.38	0.42	.543		3.34
4			6.0	0.40	0.52	3.20	.513		3.47
5			4.8	0.50	1.09	1.37	.852		3.35
6			7.2	0.33	0.90	2.13	1.060		3.21
7			6.0	0.40	1.01	1.42	0.992		3.52
8	1 x 10 ⁻⁴	5.02	1.2	2.00	1.32	0.68	0.260		3.35
9			3.6	0.67	0.55	3.08	.322		3.56
10			4.8	0.50	0.51	3.24	.403		3.47
11		5.03	2.4	1.00	0.86	1.65	.337		3.95
12			3.6	0.67	0.59	2.64	.347		4.06
13			1.2	2.00	1.22	0.28	.240		3.90
14			2.4	1.00	0.83	1.98	.327		3.66
15			3.6	0.67	0.48	3.24	.282		3.75
16	5 x 10 ⁻⁴	5.01	1.2	2.00	1.30	0.18	0.255		3.72
17			2.4	1.00	1.37	0.28	.542		3.45
18			3.6	0.67	1.32	0.35	.777		3.56
19			4.8	.50	1.29	0.50	1.010		3.51
20		5.006	4.8	.50	1.34	0.33	1.050		3.50
21		5.035	6.0	.40	1.34	0.34	1.315		3.51
22			7.2	.33	1.30	0.55	1.527		3.46
23			8.4	.29	1.31	0.49	1.799		3.42
24	1 x 10 ⁻³	5.00	1.8	1.33	1.38	0.15	0.450		3.52
25			2.1	1.13	1.38	0.19	.475		3.50
26			2.4	1.00	1.33	0.14	.522		3.66
27			3.0	0.80	1.31	0.23	.643		3.65
28			3.6	0.67	1.31	0.26	.770		3.62
29			1.5	1.60	1.31	0.21	.322		3.67
30			1.08	2.22	1.36	0.19	.240		3.53
31			3.6	0.67	1.34	0.22	.790		3.55
32			4.8	.50	1.32	0.34	1.037		3.53
33		5.03	18.	.13	1.34	0.46	3.956		3.40

In Figure 3, the results are plotted as dissolution rate in $\text{mg}/(\text{cm}^2)(\text{min})$ as a function of the average retention time, θ , in hours. The dissolver holdup was essentially constant, so the retention time was inversely proportional to the flow rate.

C. Discussion

At the two higher catalyst concentrations, 1×10^{-3} and 5×10^{-4} M, the dissolution rate is inversely proportional to the average retention time, or directly proportional to the flow rate, over the range of flow rates covered. In this range of operation, for a given catalyst concentration, the dissolution rate was established by the retention time or flow rate only, the product concentration being essentially constant. At the lower catalyst concentrations, the linearity did not hold over the entire range of flow rates. Rather, the product concentration was controlled by the acid-feed rate. Apparently, at the higher catalyst concentrations, flow rates great enough to show the non-linearity were not reached.

The dissolver operated without violent surges or rapid changes in behavior or in product composition. The contact between metal and dissolvent appeared, by visual observation, to be excellent. Control of the dissolution cycle could be obtained readily by regulation of flow rate, within the range of flow rates used, as long as the catalyst concentration was 5×10^{-4} M or higher. The tapering off of dissolution rates at lower catalyst concentrations as flow rates increased was expected; the reaction rates were not high enough for good acid utilization in the retention time allowed.

The data obtained from the recirculating dissolver were compared, as far as possible, with data obtained by Boeglin and Buckham ⁽¹⁾ in a 2-inch continuous, flooded dissolver. Although a point-by-point comparison could not be made, since a higher acid concentration, higher flow rates, and different catalyst concentrations were used in the flooded dissolver, an idea of relative effectiveness of the two dissolvers can be obtained. The flooded dissolver gave a nearly linear relationship between dissolution rate and dissolvent feed rate in the lower range of feed rates. In the recirculating dissolver, the same was true for the lower feed rates and for all feed rates at higher catalyst concentrations. Therefore, dissolution rates for the linear regions can be compared on the basis of rate per unit dissolvent flow rate, if the flow rate is expressed as superficial velocity. In both cases, the material dissolved was 2-S aluminum. Various shapes of material were dissolved in the flooded dissolver, but the most pertinent for comparison was a flat plate, $5 \times 0.625 \times 0.0625$ inches. The plates were dumped into the operating column as random packing.

Boeglin and Buckham ⁽¹⁾ estimated the average values for surface areas per foot of column, so that a value for dissolution rate per unit area could be calculated. The average area was 3470 cm^2 per foot of column height for the flat plates, and the average bed height was 9 feet.

Figure 3

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Dissolution Rates in Recirculating Dissolver
at Several Catalyst Concentrations

The flooded dissolver, with 5.2 - 5.6 M HNO_3 and 7.5×10^{-4} M catalyst feed, gave a dissolution rate estimated to be 4.17×10^{-4} mg/(cm²)(min) per unit flow rate at 652 cm/hr superficial velocity. One cm/hr superficial velocity was taken as unit flow. For 3.75×10^{-4} M catalyst and 687 cm/hr flow, the dissolution rate per unit flow was the same. In the recirculating dissolver, with 5.0 M HNO_3 and 1×10^{-3} M catalyst, the parallel-plate elements dissolved at 1×10^{-2} mg/(cm²)(min) per unit flow rate at up to 395 cm/hr net flow based on the dissolver tube cross section. The same rate was obtained with 5×10^{-4} M catalyst at lower flow rates. The rate was linear to the extent of the data taken, and extrapolation to nearly the above flow rate seems reasonable. On this basis of comparison, the recirculating dissolver gives almost 25 times the dissolution rate of the flooded dissolver. The fact that much higher flow rates were used for the flooded column may make such a comparison subject to question, quantitatively. Also, packing of the flat plates in the column may reduce somewhat the area of metal exposed to the dissolvent. The rate difference is great enough, however, to indicate that a definite improvement in effectiveness is possible.

A comparison of the two dissolver types at maximum flow rates is still favorable to the recirculating dissolver. In the flooded column, 2687 cm/hr flow gave 1.29 mg/(cm²)(min) dissolution for the plates, and 2485 cm/hr flow gave 1.80 mg/(cm²)(min) for packed round rods. The recirculating dissolver, at 18 l/hr or 395 cm/hr flow, gave 3.95 mg/(cm²)(min) or about two-to-three times the dissolution rate at less than 1/6 the flow rate. It should be noted also that the acid requirement for the recirculating dissolver, at the higher feed rates, was about 3.5 to 3.6 moles per mole of aluminum dissolved. This compares quite favorably with the average value of 3.88 for the flooded dissolver (1).

The improved dissolution rate comes, apparently, from better contact efficiency. Total and continuous immersion of fuel in dissolvent solution results from the high rate of recirculation through the dissolver, and more than makes up for the theoretical disadvantage of dissolving at product composition. Dissolution in the flooded column, on the other hand, is accompanied by violent surging and considerable longitudinal mixing. The mixing prevents an approach to desirable tubular-flow behavior, and the surging causes a low holdup and resulting low contact efficiency.

The recirculating dissolver is well suited to applications where flexibility and smoothness of operation are required, and offers the advantage of increased dissolution rate over the flooded column dissolver for aluminum elements. Since recirculation can be obtained by introducing air or heat to the dissolver if the gas evolution and heat from the reaction are inadequate, application need not be limited to highly exothermic reactions. The recirculating dissolver, therefore, appears to be adaptable to a wide variety of processes in which highly efficient contact is desired.

V. ACKNOWLEDGEMENT

The experimental work reported herein was carried out by E. L. Dayley, J. B. Echo, and M. E. Jacobson. Feed solutions were prepared and samples were analyzed by the CPP Analytical Laboratory.

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