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CONTINUOUS DISSOLUTION OF URANIUM FUEL ELEMENTS IN A TOWER DISSOLVER

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CONTINUOUS DISSOLUTION OF URANIUM FUEL ELEMENTS
IN A TOWER DISSOLVER

By

T. F. Evans

Chemical Engineering Development
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CONTINUOUS DISSOLUTION OF URANIUM FUEL ELEMENTS
IN A TOWER DISSOLVER

I. INTRODUCTION

A continuous process for the dissolution of irradiated fuel elements is a potential means of solving criticality limitations in the dissolution of enriched uranium slugs and of achieving higher dissolver capacity in the Redox and Purex Plants. This report summarizes development studies aimed at determination of dissolution rates, effluent compositions, and effects of changes in operating conditions on mercury-catalyzed dissolution in a tower-type unit.

II. SUMMARY

Results obtained from a series of runs in a two-inch diameter continuous flooded tower dissolver showed dissolving rates in the range 0.3 to 1.8 tons of uranium per day per ton of heel for both aluminum jacketed and unjacketed slugs. The dissolving rate was found to depend principally on the nitrate concentration of the effluent solution (UNH + HNO₃ only), increasing from 0.4 tons/day/ton of heel at 5 M nitrate to 1.3 tons/day/ton of heel at 8 M nitrate concentration. Other variables, such as initial acid concentration, effluent acidity, feed entry point, presence or absence of slug jackets, etc., appeared to have little effect on dissolving rates other than as they affected the total nitrate concentration of the effluent.

III. EQUIPMENT AND PROCEDURE

The dissolver tower and auxiliary equipment are illustrated in Figure 1. The tower was a 2-inch Sch. 40 stainless steel pipe with four jackets along its length for heating or cooling. Slugs were added at the top of the tower through two gate valves which served as an air lock. Preheated feed acid from a weigh tank and reflux acid from the downdraft condenser normally entered the bottom of the column. The effluent overflowed at the 12-foot level except during runs D-13 and D-14 in which the effluent was removed from the bottom of the column (through a jack leg) and acid was fed at the 15-foot level. The off-gas system was maintained at a vacuum of about five inches of water by a steam jet.

Normally, at the start of a run the tower contained the heel from the previous run. After adjusting the off-gas vacuum and air addition rate, feed acid was run through the preheater to the dissolver at a rapid rate to fill the dissolver and then reduced to the desired flow rate. Steam to the preheater and tower jackets was adjusted to maintain a constant reflux rate as measured with an orifice-type flow meter. The hydrometer specific gravity of effluent samples was measured at about 30-minute intervals during the course of each run. Three to four hours was normally required to approach equilibrium and the run was continued until two or more duplicate samples were obtained. Slugs were added during the run to maintain a constant heel weight. Prior

to Run E-17 the heel was completely covered with solution during dissolution. Near the end of the run, samples of reflux acid and occasionally of solution from intermediate tower points were also taken in addition to effluent samples. The effluent flow rate was determined by sight-glass readings in the product tank except for Runs E-5, E-6, and E-12 to E-20 in which dilution water was added to the effluent stream just after it left the tower. An orifice-type flow meter in the product line did not perform satisfactorily because of intermittent plugging of the orifice. The weight of metal in the dissolver at the end of each run was determined by filling the dissolver with water and measuring the volume occupied by the metal.

IV. DISCUSSION OF RESULTS

The principal data from the present studies are tabulated in Table 1. With the exception of Run E-3, the measured product rates were between 80 percent and 100 percent of the acid feed rates. For the calculation of dissolving rates and acid efficiencies, an average product rate equal to 90 percent of the feed rate was assumed for each run. Dissolving rates were in the range of 1.1 to 5.0 pounds per hour (0.3 to 1.8 tons/day/ton of heel). They appear to depend principally on the total nitrate concentration in the effluent as shown in Figure 2. Only the nitrate concentration associated with the uranium and nitric acid were considered because of the absence of a reliable estimate of the aluminum nitrate concentration for most of the runs. A plot of dissolving rate versus effluent acidity rather than nitrate concentration shows much more scatter among the points. The effluent acidity appears to have had little effect on dissolving rate other than by virtue of the fact that dissolving to a low final acidity results in a lowered total nitrate concentration through loss of nitrogen oxides. As shown in Figure 2, deviations from the curve drawn do not appear to be associated with effluent acidity and the scatter of the points must be attributed to other factors. Acid feed rate, acid concentration, heel size, etc., were studied in the following ranges:

Acid feed rate, ml/min	22 to 100
Acid feed concentration, wt % HNO_3	40 and 60
Reflux rate, ml/min	50 to 120
Mercuric ion concentration, M	0.004 to 0.010
Heel weight, pounds	63 to 124
Tower position, degrees from vertical	0 and 20
Aluminum concentration, moles/mole of U	0 and 0.3
Acid entry point, feet above bottom of tower	0 and 15

These factors had no detectable influence on the dissolving rate other than as they influenced the effluent nitrate concentration.

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The variation of solution concentrations along the length of the tower for Run 13 are shown in Figure 3. The results, which are typical of those observed in other runs, indicate a significant gradient through the column in uranyl nitrate, nitric acid, and aluminum nitrate concentrations.

Results from the present study are compared with previous pilot plant results in Figure 4. Differences between the curves are probably attributable to several causes. The low dissolving rates in the batch dissolvings at high total nitrate concentrations are probably associated with the build up of catalytic agents in solution at the beginning of a cut and a relatively small area per unit weight of slugs. The high dissolving rates shown for continuous pot dissolutions are probably attributable to the presence of aluminum nitrate in the solution. The presence of aluminum increased the total nitrate concentration by 1.6 to 2.0 M. As shown in Figure 3 the aluminum was restricted to a small section of the tower dissolver rather than being distributed throughout the vessel as in the continuous pot dissolutions.

Acid efficiencies, as indicated in Table 1 by the moles of nitric acid consumed per mole of uranium dissolved, were in general lower than would be anticipated for an efficient downdraft condenser. Since the presence of variable amounts of aluminum in the tower effluent solution (depending on how long a period had elapsed since the last slug was charged) introduced major errors in the calculated acid recovery efficiency no attempt was made to study these data critically.

V. FUTURE WORK

The results reported above appear sufficiently promising to warrant construction of a larger dissolver. A five-inch diameter unit incorporating a more efficient acid recovery system and improved instrumentation is presently being designed.

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FIGURE 1
TWO-INCH CONTINUOUS
TOWER DISSOLVER

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No.	Ml/Min	Feet	Time	Temp	Rate	Pressure	Flow	Temp	Time	Rate
D-10	50	0	40	0.010	0.7	100	3.42	0	84	7.9-J 6.0
D-11	50	0	40	0.010	1.3	100	2.32	0	85	7.9-J 10.1
D-12	33	0	40	0.010	0.9	67	2.84	0	86	7.9-J 6.5
D-13	50	15	40	0.010	1.3	80	2.90	15	84	7.9-J 14.6
D-14	33	15	40	0.010	0.9	58	2.90	15	84	7.9-J 6.8
D-15	50	0	40	0.010	0.7	100	3.49	0	82	7.9-J 10.2
D-16	75	0	40	0.010	1.0	100	3.54	0	80	7.9-J 9.5
D-17	50	0	40	0.006	1.3	100	2.50	0	84	7.9-J 15.3
D-18	33	0	40	0.006	0.9	67	1.94	0	76	7.9-J 9.0
D-19	22	0	40	0.010	0.6	50	2.43	0	76	7.9-J 8.4
D-20	50	0	40	0.008	0.7	50	3.48	0	74	7.9-J 8.5
D-21	50	0	40	0.008	1.3	100	(10)	0	76	7.9-J 3.8
E-1	50	0	40	0.005	1.3	100	(10)	0	75	7.9-J 7.7
E-2	75	0	40	0.004	1.3	100	(10)	0	67	0 10.3
E-3	75	0	40	0.005	1.3	120	(10)	0	63	0 5.1
E-4	100	0	60	0.004	1.3	120	(10)	0	65	27.6 28.6
E-7	75	0	60	0.005	1.3	120	6.09	0	75	18.8 8.4
E-7a	75	0	60	0.005	1.3	60	9.96	0	75	0 5.0
E-12	50	0	60	0.005	1.3	120	4.94	0	64	12.4 4.8
E-13	35	0	60	0.005	1.3	80	(10)	0	68	22.3 14.8
E-14	75	0	60	0.005	1.3	120	6.48	0	63	15.9-J 25.4
E-15	50	0	60	0.005	1.3	105	5.34	0	67	23.8-J 14.2
E-16	35	0	60	0.005	1.3	80	4.96	0	74	15.9-J 16.1
E-17	50	0	60	0.005	1.3	70	6.80	0	F	103.2-J 16.2
E-18	35	0	60	0.005	1.3	95	(10)	0	F	7.9-J (10)
E-19	50	0	60	0.005	1.3	120	3.16	0	F	23.8-J 39.6
E-20	35	0	60	0.005	1.3	94	2.83	0	F	15.9-J 25.1

Notes:

1. Tower was inclined 20° from vertical during Runs E-1 to E-20.
2. Feed temperature = 110°C ± 5.
3. Air entered dissolver at 15 ft. level for all runs except D-21 in which it entered.
4. F = Tower filled above 15 ft. level. Head at 12 ft. level was approximately 124 lb.
5. J = Jacketed slugs.
6. 100 % efficiency = 2.0 moles HNO₃/mole U for bare slugs.
= 2.9 moles HNO₃/mole U for jacketed slugs.
7. D = Product diluted as it left the column so rate could not be measured.
8. By analysis.
9. Feet above bottom of tower.
10. No sample.

5.2	40	12	1.23	1.67	(10)	1.522	1.74	0.49	4.5
7.5	44	12	1.42	2.33	(10)	1.555	1.33	0.37	4.6
5.8	31	12	1.19	3.26	(10)	1.510	1.68	0.48	4.1
4.7	43	0	1.40	3.05	(10)	1.545	1.31	0.37	4.1
6.4	28	0	1.24	3.67	(10)	1.535	1.75	0.51	4.1
5.4	44	12							
3.9	62	12	1.10	4.20	(10)	1.500	2.30	0.69	4.2
6.2	49	12	1.05	3.35	(10)	1.481	1.49	0.43	5.2
7.2	27	12	1.38	1.86	(10)	1.557	1.29	0.41	5.0
7.2	23	12	1.74	1.51	(10)	1.641	1.08	0.34	4.2
5.2	46	12	1.13	2.87	(10)	1.500	1.60	0.52	5.2
2.6	41	12	0.83	3.10	(10)	1.460	1.18	0.37	6.9
6.4	48	12	0.93	3.73	(10)	1.464	1.32	0.42	5.4
4.6	73	12	0.87	6.14	(10)	1.422	1.85	0.66	3.0
2.9	42	12	0.86	5.18	(10)	1.420	1.82	0.70	4.2
5.9	92	12	1.33	6.20	(10)	1.652	3.69	1.36	6.2
4.5	61	12	2.34	4.00	(10)	1.857	4.96	1.59	4.5
1.8	64	12	1.84	5.45	(10)	1.734	3.90	1.25	4.9
5.0	D	12	3.05	1.74	7.96	2.070	4.31	1.62	4.2
4.9	D	12	3.50	1.07	7.84	2.154	3.47	1.23	3.8
5.7	D	12	2.19	3.84	8.65	1.787	4.65	1.77	4.8
5.5	D	12	2.39	2.64	8.33	1.882	3.38	1.21	4.9
5.6	D	12	2.53	1.94	8.44	2.024	2.50	0.81	4.9
6.2	D	12	1.85	1.56	8.85	1.840	2.62	0.51	7.0
6.8	D	12	2.55	1.49	8.91	2.023	2.52	0.49	5.1
9.5	D	12	2.88	1.03	7.88	2.051	4.07	0.79	4.7
8.2	D	12	2.98	0.91	7.24	2.009	2.95	0.57	4.5

. 0 ft. level.

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