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UNIT OPERATIONS SECTION

QUARTERLY PROGRESS REPORT

OCTOBER–DECEMBER 1965

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
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U.S. ATOMIC ENERGY COMMISSION

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ORNL-3958

Contract No. W-7405-eng-26

UNIT OPERATIONS SECTION QUARTERLY PROGRESS REPORT

OCTOBER-DECEMBER 1965

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JULY 1966

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
Operated by
UNION CARBIDE CORPORATION
U. S. ATOMIC ENERGY COMMISSION

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ABSTRACTS

1. CHEMICAL ENGINEERING RESEARCH

1.1 Stacked-Clone Contactor

The stacked-clone solvent extraction contactor makes possible the multistage contacting of an organic extractant with a highly radioactive aqueous solution. A prominent advantage of the contactor is that it ensures a very short residence time for the organic phase. The operating capabilities of an 11-clone contactor are being ascertained by testing aqueous-organic systems having a wide range of physical properties. The flow capacities for an 0.08 M HNO_3 aqueous phase in contact with four different organic phases (mineral oil, 2-ethyl-1-hexanol, N-Paraffins-175, and Amsco) were found to be 3.9, 2.0, 3.8, and 3.8 liters/min, respectively, at 30°C and a flow-rate ratio (aqueous:organic) of 3.0. The batch-extraction rate constants for the extraction of benzoic acid from 0.08 M HNO_3 into mineral oil, N-Paraffins-175, and Amsco were 0.8, 25, and 3.1 min^{-1}, respectively, at a turbine speed of 400 rpm. The contactor continues to look promising for application to a wide range of chemical systems.

2. POWER-REACTOR FUEL PROCESSING

2.1 Shear-Leach Processing

During the head-end steps of shearing and leaching, which are a part of the reprocessing scheme for certain power-reactor fuels, the heat internally generated in the fuel by radioactive decay could conceivably lead to excessive temperatures or make necessary the provision of means for removing the heat from the fuel while it is in process and/or storage. The seriousness of this potential problem was investigated by calculating the temperature that would be reached in a typical power-reactor fuel at each step. The temperatures thus found were 75.5°F during receiving-canal storage, 191°F during mechanical disassembly, 548°F in the shear feed envelope, and 1586°F during storage

in a 10-in.-ID basket. We conclude that, in the operation of a batch dissolver, excessive pressure will not result from the heating produced by radioactive decay.

2.2 Graphite-Fuel Processing: Grind-Leach

The engineering problems of grinding, leaching, and solid-liquid separation are being examined experimentally to evaluate the grind-leach process as an alternative to the burn-leach process for those graphite-matrix fuel compacts that contain carbon-coated fuel microspheres. For a typical reactor the fuel kernels have a diameter of about 125 μ, necessitating grinding to -140 mesh or finer. Of two grinding devices tested, the double roll crusher was the more promising.

Grinding rates for an 8 x 2-in. roll-crusher were of the order of 100 g of Peach Bottom fuel per minute. The uranium loss during leaching of -140 mesh fuel was 0.35%, and the thorium loss was 0.67%. Permeability of the ground fuel was 6 to 25 gal $hr^{-1}ft^{-2}$ at a pressure gradient of 18 psi/ft. From these data we conclude that the process is technically feasible and that the principal need is to demonstrate that an improved roll-crusher can grind to -140 mesh without necessity for recycle.

3. THE ENGINEERING SEPARATION OF MACROMOLECULAR BIOLOGICAL SUBSTANCES

Engineering methods are being developed for separating and purifying large quantities of specific transfer ribonucleic acids (t-RNA's). We tested an extraction process in which the cell walls of E. coli are made selectively permeable with isoamyl alcohol instead of being ruptured with phenol. No t-RNA was obtained with this process. A batch of 632 g of mixed t-RNA was obtained by the phenol process. Our next step will be to devise means for separating the mixture.

4. SOL-GEL PROCESS DEVELOPMENT

4.1 Urania-Sol Preparation

We are investigating the preparation by the sol-gel technique of urania sols for application as urania or mixed-oxide reactor fuels.

The first four engineering-scale tests of a precipitation-dispersion process did not give satisfactory sols because of inadequate control of the individual steps. The tests are being continued in modified equipment.

5. THORIUM FUEL-CYCLE STUDIES

5.1 Microsphere-Preparation Development

We continued the development of equipment and procedures for converting sols into oxide microspheres suitable for fuel elements. The key process steps are formation of a sol drop of the correct size and gellation of the drop by extracting the water into an organic solvent. Rotary, shear, and electrostatic dispersers were improved for the large-scale dispersion of sol into uniform drops. The rotary dispersers gave calcined products of 88 to 575 μ mean diameter at flow rates of 4.8 to 20 ml/min, but the uniformity was variable. The electrostatic disperser gave good uniformity and mean product diameter of 240 to 310 μ for sol feed rates of 4.8 and 9.6 ml/min.

5.2 Microsphere Preparation Without Fluidization

The fluidized beds presently used for making microspheres by extracting the water require careful control; therefore, simpler procedures and equipment are being tested for remote operation. Long lengths of coiled tubing were tested to provide the residence time for gelling the sol drops. Satisfactory operation was possible for small drops only; low velocities at the tube walls caused larger drops to coalesce or settle. The coiled-tubing apparatus will not be tested further.

5.3 Coated-Particle Development Facility (CPDF) Microsphere Column

A microsphere-column system designed for processing 10 kg of ThO_2 per 24-hr day was installed and operated for studies of long-term

operation and for pilot-plant production of microspheres. The capacity is limited by the steam supply to the distillation apparatus and is about 25 ml of sol per min (1200 g of ThO_2 per hour for a 3 M ThO_2 sol). Both rotary and two-fluid-nozzle sol dispersers were used; 2 two-fluid nozzles will be used to prepare the 210- to 250- μ product needed for pyrolytic-carbon-coating studies.

6. TRANSURANIUM PROCESSING STUDIES

6.1 Pulse-Column Tests

The design and operation of process equipment for the Transuranium Facility are being tested with nonradioactive solutions. The pulse columns in the Tramex solvent-extraction rack operated for 80 hr at 50% of the design flow rates; flooding occurred at higher flow rates. The flooding may have been caused by partial plugging of the pulse-loss restrictors in the aqueous outlets of the column. The efficiency of the columns, measured with nickel chloride as the solute, was satisfactory for extraction and stripping.

6.2 Berkelium Extraction-Rack Tests

The equipment rack to be used in the Transuranium Facility for separating and recovering berkelium from californium and other actinides was tested for hydraulic stability and separation efficiency. With cerium and europium as standins for berkelium and californium, respectively, cerium extraction was greater than 99.6% complete after the third feed pass, and stripping was 99.7% complete after 5 liters of strip solution had been used. The europium decontamination factor was 1300 after 4 liters of scrub solution had been used.

7. WASTE PROCESSING

7.1 Rising-Level Glass-Making Pot Calcination

The feasibility of fixing radioactive wastes as glasses by *in situ* calcination in storage containers is being demonstrated with Purex

waste 1 (PW-1). In a previous test (R-91) the glass-making additives reacted chemically with the waste to produce a slurry that could not be satisfactorily handled. A 100-hr run was made in which 852 liters of PW-1 and 294 liters of additive were fed to the pot separately. The 0.525-in.-thick pot corroded nearly half way through near the bottom.

Previous Reports in this Series for Year 1965

January	ORNL-TM-1092
February	ORNL-TM-1094
March	ORNL-TM-1103
April-June	ORNL-3868
July-September	ORNL-3916

1. CHEMICAL ENGINEERING RESEARCH

1.1 Stacked-Clone Contactor

W. S. Groenier M. E. Whatley

The stacked-clone contactor is a high-speed solvent-extraction device developed for use with highly radioactive solutions. It consists of a cascade of axially aligned liquid cyclones, each operating with countercurrent flow of the two liquid phases. The countercurrent flow is produced by the induced underflow. An optimal component design and configuration were identified and tested exhaustively with a single chemical system. Subsequent studies were directed toward evaluating this 11-stage standard contactor with chemical systems exhibiting a range of liquid physical properties. Thus, such diverse organic substances as 100% TBP (tributyl phosphate) and hexone were tested to assist in determining an operating range for the contactor and the results reported.[1]

In tests made during the past quarter, the aqueous phase was 0.08 M HNO_3 and the organic phase was one of: mineral oil, 2-ethyl-1-hexanol, N-Paraffins-175,[2] and Amsco 125-82.[3] Flooding data for the standard 11-stage contactor show total liquid throughputs of 3.9, 2.0, 3.8, and 3.8 liters/min, respectively, for the above systems at a flow rate ratio (A/O) of 3 and a temperature of about 30°C. Further flooding data for these systems are presented in Fig. 1.1 and Table 1.1.

The lower flow capacity for 2-ethyl-1-hexanol is probably due to its lower interfacial tension of 13 dynes/cm, compared with 35 to 50 dynes/cm

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[1] M. E. Whatley *et al.*, *Unit Operations Section Quarterly Progress Report, July-September 1965*, ORNL-3916.

[2] A product of Union Carbide Corporation.

[3] A product of American Mineral Spirits Company.

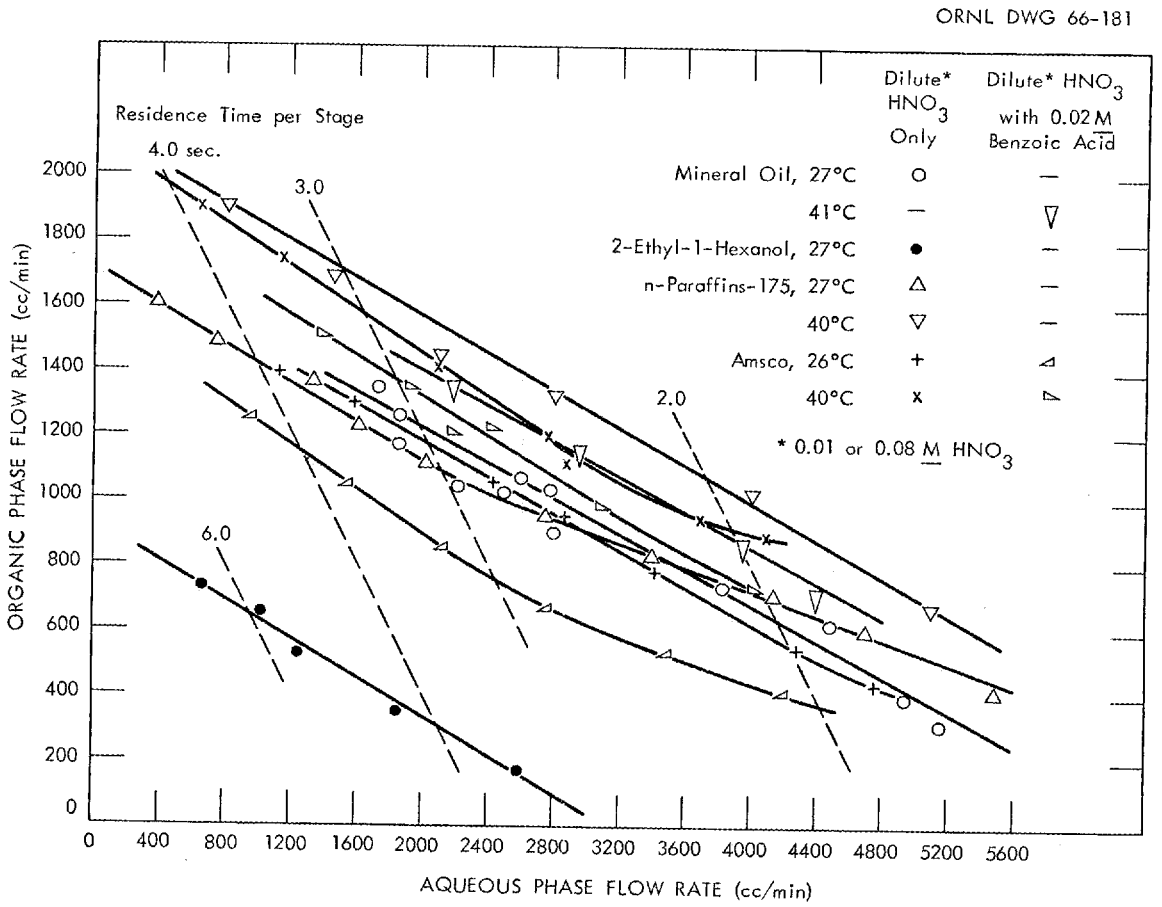

Fig. 1.1 Flooding Data.

Table 1.1 Flooding Data and Physical Properties of the Solutions

Aqueous phase	0.01 M HNO_3,		0.08 M HNO_3,	0.08 M HNO_3,		0.08 M HNO_3,			
Organic phase	Mineral Oil		2-ethyl-1-hexanol	N-Paraffins-175		Amsco			
Aqueous feed contains 0.02 M C_6H_5COOH	No	Yes	No	No	No	No	No	Yes	Yes
Density difference between phases, g/cc	0.121	0.124	0.171	0.254	0.258	0.248	0.254	0.248	0.254
Aqueous-phase density, g/cc	0.996	0.991	0.999	0.999	0.994	0.999	0.994	0.999	0.994
Organic-phase viscosity, centipoise	116.	57.	6.60	1.34	1.10	1.28	1.07	1.28	1.07
Aqueous-phase viscosity, centipoise	0.84	0.66	0.84	0.84	0.66	0.84	0.66	0.84	0.66
Interfacial tension, dynes/cm	52.7	51.9	13.4	35.7	35.4	45.2	44.3	34.5	35.1
Flow capacity at flooding, liters/min:									
A/O = 1/2	-	-	1.21	2.23	2.80	-	2.71	2.02	-
A/O = 1	2.77	-	1.43	2.64	3.32	2.78	3.16	2.35	2.92
A/O = 3	3.88	4.29	1.96	3.79	4.57	3.78	4.22	3.15	4.01
A/O = 10	5.16	-	2.55	5.63	6.10	5.06	-	4.56	-
Temperature, °C.	27	41	27	27	40	26	40	26	40

for the other systems (see Table 1.1). The dispersed-phase viscosity has little or no effect on flow capacity, as can be seen by comparing data for mineral oil (116 centipoises) with those for N-Paraffins-175 and Amsco (1.3 centipoises).

Solution residence time per physical stage ranged from 2 to 4 sec for these systems, depending upon the flow ratio chosen.

Benzoic acid was extracted from 0.08 M HNO_3 using seven stages. Stage efficiencies were only 30 to 40% for mineral oil and Amsco but were near 60% for N-Paraffins-175 - a mixture of normal hydrocarbons ranging from C_9 to C_{15} and having physical properties very near to those of Amsco. It is probable that the high viscosity of mineral oil is at least partially responsible for its low extraction efficiency. However, batch extraction-rate tests indicated lower rates for both mineral oil and Amsco than for N-Paraffins-175. Extraction efficiencies were computed in the usual manner,[4] using the CDC-1604 computer to determine the number of ideal extraction stages in a McCabe-Thiele type of calculation and adjusting a constant in the equilibrium relationship to equalize extraction efficiencies in each mode of operation. Thus, the reported efficiencies (Table 1.2) are for mass transfer into either the aqueous or the organic phase. The equilibrium data for these benzoic acid systems were fitted to parabolic expressions.

The batch-extraction rate studies were conducted at about 25°C in a baffled 6-in.-diam glass mixer with a 3-in.-diam, 6-bladed turbine. The turbine was located in the middle of the liquid, 3 in. from the bottom of the mixer. Extraction of benzoic acid from 0.08 M HNO_3 with N-Paraffins-175 was 99% complete in less than 10 sec at 400 or 700 rpm at an A/O of 1.0, and with either the aqueous phase or the organic phase continuous; this rate corresponds to a rate constant, ka, of greater than 25 min^{-1}. Extraction with Amsco at an A/O of 1.0 and the aqueous phase continuous indicated rate constants of 2.5 and 3.1 min^{-1},

---

[4] M. E. Whatley *et al.*, Unit Operations Section Monthly Progress Report, May 1963, ORNL-TM-613.

Table 1.2 Performance Data for the Standard Stacked-Clone Contactor

Run Series	Temperature (°C)	Adjusted Equilibrium Constant[a]	Average Flow Ratio (A/O)	Throughput at Flooding (liters/min)	Average Efficiency (%)	RMS Deviation from Average Efficiency	Performance Factor[b]	No. of Duplicated Runs
Mineral oil:								
BAMO- 1	31	0.0730	1.60	3.17	26.6	5.5	0.84	5
BAMO- 6	27	0.0765	1.60	3.17	31.7	7.4	1.00	5
BAMO-11	27	0.0728	1.42	3.05	39.3	5.4	1.20	5
BAMO-16	40	0.0651	1.70	3.58	38.2	7.3	1.37	5
N-Paraffins-175:								
DODEC- 1	27	0.1194	1.64	3.17	58.9	22.8	1.87	6
DODEC- 7	25	0.1263	2.20	3.47	80.3	17.0	2.78	6
DODEC-13	44	0.0919	2.06	4.08	56.6	9.4	2.31	6
DODEC-19	27	0.1388	2.04	3.39	56.0	9.5	1.90	6
Amsco:								
AMBA- 1	41	0.0838	2.04	3.90	31.5	2.4	1.23	6
AMBA- 7	26	0.0982	2.08	3.40	35.4	4.4	1.20	6
AMBA-13	41	0.0772	2.19	3.46	30.3	6.2	1.05	6

[a]Equilibrium line is parabolic, $y = ax^2$.

[b]Product of efficiency and throughput at flooding.

respectively, at 250 and 400 rpm. Values of ka for mineral oil were 0.8 and 1.9 min^{-1}, respectively, for aqueous-continuous dispersions at 400 and 700 rpm and an A/O of 1.0. These constants represent about 34 to 40% extraction after 10 sec for Amsco, and 12 to 27% for mineral oil.

2. POWER-REACTOR FUEL PROCESSING

2.1 Shear-Leach Processing

B. C. Finney

The development and evaluation of a batch shear-leach process for recovering fissile and fertile materials from spent nuclear power-reactor fuel assemblies was continued by computing temperatures attained by spent fuel in each of the various steps in the shear-leach process. The shear-leach process can be briefly described as consisting of five operations: (1) underwater storage prior to mechanical processing, (2) mechanical disassembly to remove massive end adaptors and inert hardware, (3) shearing to reduce the fuel to short sections in preparation for leaching, (4) storage of the sheared fuel pieces collected in a basket, and (5) batch leaching. All operations (except storage) are carried out in a concrete-shielded hot cell, necessitating remote maintenance and replacement of all processing equipment.

Spent fuel assemblies produce large amounts of high-energy radiation through radioactive decay of fission products. This radiation interacts with the fuel and its cladding, releasing energy as thermal decay heat. The amount of heat released by a given fuel depends primarily on three variables: burnup, specific power, and decay time (time out of the reactor).

The temperatures that metal-clad uranium oxide type fuels will attain during shear-leach processing as a result of the release of such decay heat were calculated; a rough draft of the calculations has been prepared. Decay heat generation rates were calculated over a burnup range of 8000 to 50,000 Mwd/ton of uranium, specific powers of 10 to 70 watts per gram of uranium, and cooling times of 180 to 1080 days. The computer code used was a modification of a code (PHØEBE), developed by E. D. Arnold of ORNL. In the calculations, a Yankee Atomic type sub-assembly (thirty-six 0.302-in.-OD x 120-in.-long UO_2-filled stainless-steel fuel rods on 0.422 in. centers) was selected to represent a typical power-reactor fuel. Maximum temperatures attained by the fuel

during reprocessing were calculated based on steady-state heat transfer conditions assuming that heat loss was by natural convection and radiation.

A Yankee subassembly was considered to be (1) suspended vertically in water, (2) suspended horizontally in air, (3) horizontally positioned in the shear feed envelope, (4) sheared and stored in an array of 16 baskets of 5, 7.75, and 10 in. inner diameter, and (5) leached in the sheared form in a basket in a batch dissolver. In addition to the temperature calculations just enumerated, the pressure rise in a batch dissolver (leacher) disengaging space caused by acid vaporization from quenching of a basket of hot fuel by cold acid was estimated. A computer code BIG DEAL was written in FORTRAN language and used to make all calculations.

A summary of results follows:

Maximum fuel temperatures during reprocessing will not cause melting of the fuel or clad. Acid may be added to the hot fuel in the dissolver as fast as mechanically feasible without fear of exceeding the maximum pressure specifications for the dissolver.

The lowest and highest fuel temperatures encountered for each operation of the shear-leach process over the range of burnups of 8000 to 50,000 Mwd/ton, specific powers of 10 to 17 w/g, and cooling times of 180 to 1080 days, are:

Operation	Lowest Temp. (°F)		Highest Temp. (°F)	
1. Receiving-canal storage	68.5		75.5	
2. Mechanical disassembly	123		191	
3. Shear-feed envelope	143		548	
4. Storage in baskets	Center	Skin	Center	Skin
5-in. ID	160	140	852	548
7-3/4-in. ID	196	155	1244	773
10-in. ID	232	171	1586	979

For the typical case of the Yankee subassembly irradiated to a burnup of 20,000 Mwd/ton, at a specific power of 23 w/g, and cooled 180 days, the temperatures are as follows:

Operation	Temp. (°F)	
1. Receiving-canal storage	71.5	
2. Mechanical disassembly	150	
3. Shear-feed envelope	350	
4. Storage in baskets	Center	Skin
5-in. ID	491	324
7-3/4-in. ID	720	453
10-in. ID	924	575

The highest temperature encountered (1586°F) is far below the melting point of both UO_2 (3947°F) and stainless steel (2642°F), indicating that the fuel and clad will not melt during reprocessing if the burnup, specific power, and cooling time are within the limits specified. The calculations indicate that a 180-day decay period should be sufficient. The calculated temperatures are based on heat loss by radiation and natural convection; somewhat lower temperatures could be attained during mechanical disassembly, in the shear feed envelope, or during storage by providing means for forced convective cooling. Of course for any given burnup and specific power, a lower temperature could be attained by extending the decay time but such an extension would require careful consideration of the expense involved in providing additional storage space for the irradiated fuel at either the reactor or reprocessing plant.

An acid flow rate greater than 1.16×10^5 lb/hr would be required to develop a pressure in excess of 1 psig in the dissolver when processing fuel with a burnup, specific power, and cooling time equal to or less than 50,000 Mwd/ton, 70 w/g,

and 180 days decay time, provided that the 6-in. off-gas line is fully open. This great an acid flow rate is impractical, and, consequently, it is concluded that a pressure buildup in the dissolver will not be a problem under normal operating conditions.

2.2 Graphite-Fuel Processing: Grind-Leach

B. A. Hannaford

Graphite-matrix fuels for high-temperature gas-cooled reactors (HTGR's) generally contain microspheres of fissile and fertile materials (as oxides or carbides) encased in an impermeable coating. The coating may consist of one or more layers of pyrolytic carbon or of alternate layers of pyrolytic carbon and silicon carbide. The coating protects the fuel microsphere from exposure to atmospheric moisture during fuel manufacture and helps prevent the release of fission products. HTGR fuels containing microspheres coated with pyrolytic carbon can be reprocessed by a burn-leach process[5] or by a grind-leach process, but only the grind-leach process appears to be potentially applicable to fuels that have a noncombustible coating for the microsphere. Current engineering-scale work on the grind-leach process is directed toward demonstrating (1) a reliable and rapid method for grinding HTGR fuel to -140 mesh, (2) satisfactory rates for separation of leachant and wash water from the leached graphite, and (3) low losses of uranium and thorium to the graphite residue.

2.2.1 Size Reduction: Crushing and Grinding

Quantitative recovery of uranium and thorium from coated-particle fuel requires that the coating of practically every particle be breached, to give access of the acid leachant to the carbide or oxide kernel.

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[5] E. L. Nicholson, L. M. Ferris, and J. T. Roberts, Burn-Leach Processes for Graphite-Base Reactor Fuels Containing Carbon-Coated Carbides or Oxide Particles, ORNL-TM-1096, (April 2, 1965).

Previous work[6] showed that a high-speed hammer mill was worn excessively when HTGR fuels containing SiC were ground to only 500 μ (35 mesh). Work with a small double-roll crusher showed encouraging results with Peach Bottom Fuel. A brief series of tests with ball-milling indicated that this method is undesirable on the basis of low grinding rate, batchwise operation, and no positive size classification of the product.

2.2.1.1 Crushing-Roll Grinding. -- In principle, outstanding advantages of a roll crusher are suggested for an application of this kind: (1) Separate equipment for size classification is unnecessary, since the minimum dimension of fuel discharged from the crusher is determined by the roll spacing on the final set of rolls. (2) The roll crusher is a once-through, continuous device. (3) Since it is a basically simple machine, remote lapping or replacement of the rolls, when necessary, should be possible.

HTGR fuel was satisfactorily prepared for feeding to the roll mill by a rough crushing in a conventional hammer mill having fixed hammers on a 15-in.-diam x 8-in.-long rotor and operating at 845 rpm. With the discharge bars set for 1/8-in.-wide slots, the mill produced a 75% yield of -6 mesh material in a single pass.

A small, commercial roll crusher having 8-in.-diam x 2-in.-wide rolls was used in preliminary work with graphite and Peach Bottom fuel to measure the nip angle and throughput rates. The position of one roll was adjustable (Fig. 2.1) in order to vary the roll spacing (nominally equivalent to the maximum product diameter, D_p). Both rolls were chain-driven at the same speed by a variable-speed DC compound motor.

The nip angle, A, as defined[7] in Fig. 2.1, was calculated to be about 12° for Peach Bottom fuel, based on crushing experiments at roll spacings of 0.045-in. and 0.003-in. The nip angle for ordinary rock is

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[6] M. E. Whatley et al., Unit Operations Section Quarterly Progress Report, April-June 1965, ORNL-3868.

[7] G. G. Brown et al., Unit Operations, pp 34-36, Wiley, New York, 1950.

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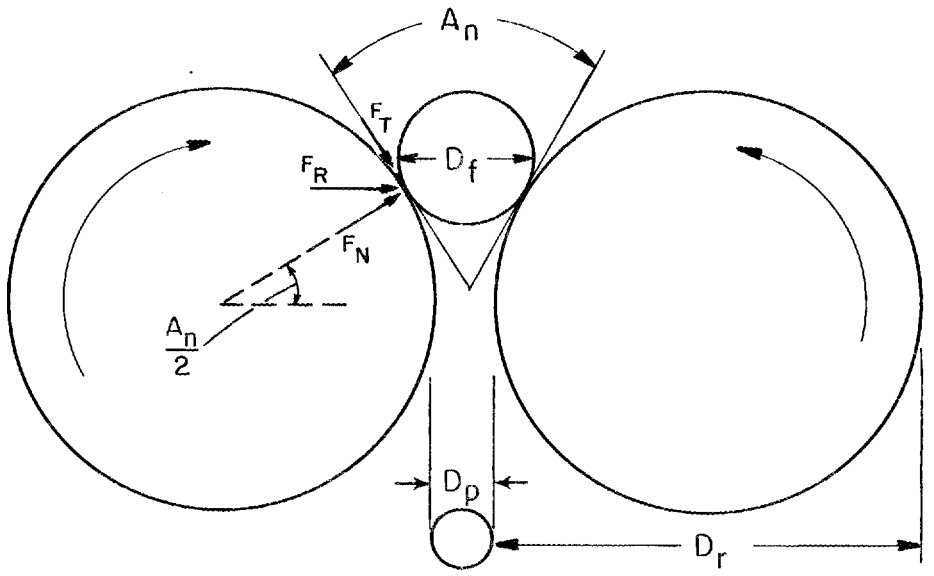

F_T = Tangential Force on Particle

F_N = Normal Force on Particle

F_R = Resultant of F_T and F_N

Fig. 2.1 Schematic Diagram of a Double-Roll Crusher. Angle of nip, A_n, is defined by $\cos A_n/2 = \frac{D_r + D_p}{D_r + D_f}$, for this critical condition in which F_R is horizontal.

about 32°. This comparatively low value for Peach Bottom fuel reflects the low coefficient of friction for graphite in contact with smooth steel. The commercial roll crusher used in the tests would not maintain a preset roll spacing under load, because of excessive play in the bearings of the crushing rolls and lack of rigidity in the machine frame. Therefore, the true nip angle may be even smaller than 12°. The determination of the correct value is of importance for predicting the size reduction ratio per pass.

Preliminary grinding results for Peach Bottom fuel are presented in Fig. 2.2. Although the final four passes were made at a nominal 0.002 in. (less than 50 μ) roll spacing, the fraction of product falling in the -325 mesh range (less than 44 μ) was small and was only 46% after the final pass. This was not unexpected, because of the lack of machine rigidity already described. Assuming that Peach Bottom compacts contain ThC_2-UC_2 microspheres of 125 to 200 μ diameter (random optical measurements showed beads as small as 175 μ), the minimum acceptable specification on the crushed product would be that it pass 140 mesh (105 μ). However, +140 mesh fraction remained close to 20% for the second and third passes, and actually increased with the fourth pass (Fig. 2.2). The explanation for this anomaly may be that, in addition to the comminutive effect on the fueled graphite, the roll mill has an agglomerative effect; that is, it compresses a great number of smaller particles (primarily from the -140 +325 mesh range) into relatively large flakes. These flakes were up to 3000 μ in diameter but were only 50 μ to 140 μ in thickness, as determined by optical measurements on several randomly selected flakes. Therefore the effectiveness of a roll crusher in fracturing coated particles may be much greater than is indicated by a sieve analysis of the product.

In addition to the shortcomings of the 8-in.-diam roll crusher in maintaining the preset roll spacing, the rolls were found to be slightly eccentric (about 0.003 in. on the diameter). This effect was partially offset by matching the high point on one roll with the low point on the other. Also, the clearance between the machine housing

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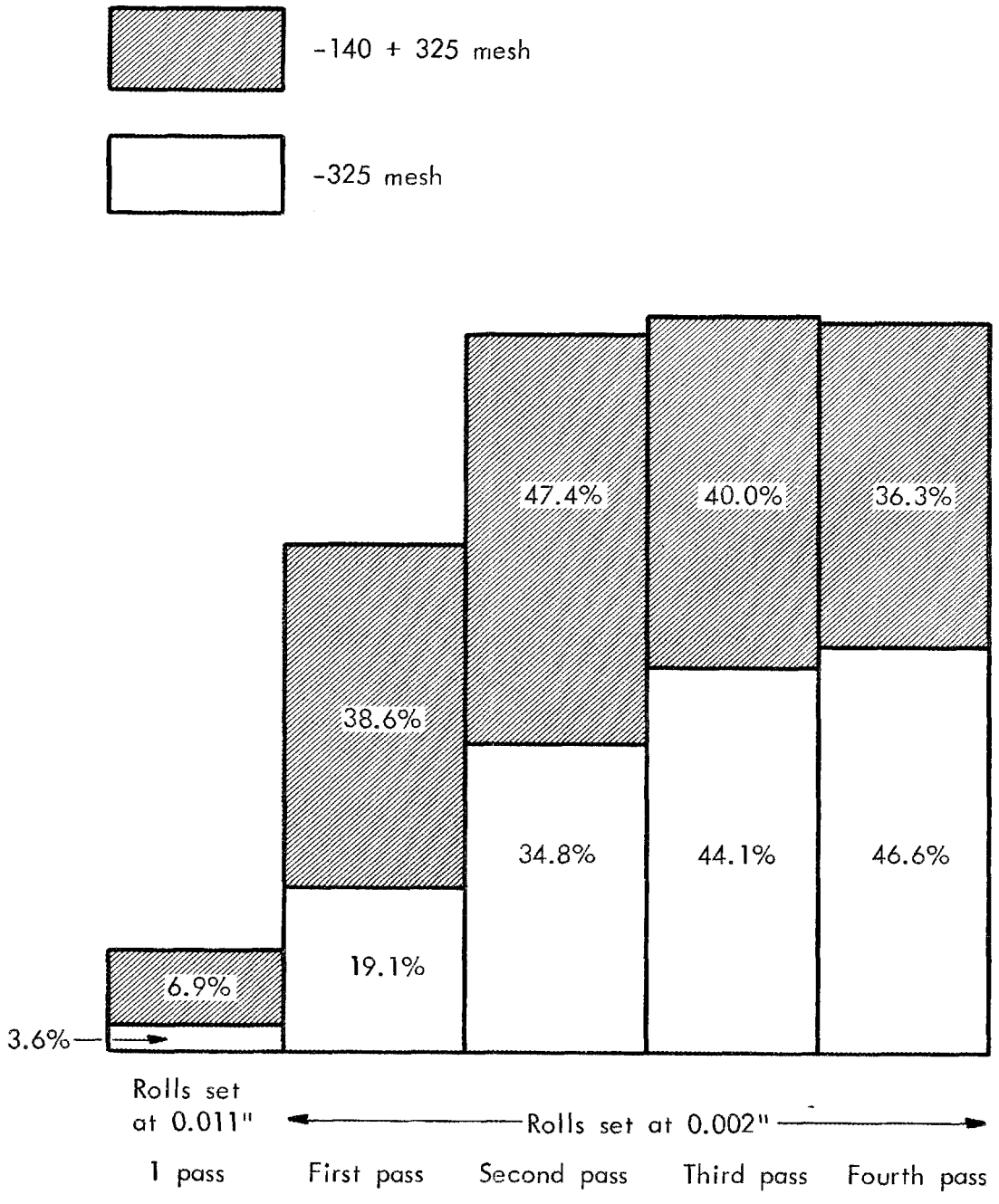

Fig. 2.2 Size Distribution of Peach Bottom Fuel Reduced by Successive Passes through an 8-in.-diam Double Roll Crusher. Preceded by roll-crushing the -4+6 mesh fraction from hammer-mill product at settings of 0.100 and 0.055-in. Roll speed, approximately 200 rpm; grinding rate 200 to 500 g/min.

and the ends of the rolls was excessive; this was partially corrected by fitting end plates of 3/8-in.-thick Teflon to ride on the cylindrical surface of the roll ends. These end plates confined the feed material to a path between the roll faces.

Machine throughput was measured at several values of roll spacing, roll speed, and nominal feed size but the data were inconclusive. A tentative specific conclusion was that, with the roll spacing set at the critical gap for a particular feed size (unfueled graphite), the throughput was proportional to roll speed up to about 200 rpm (7 ft/sec peripheral speed with the 8-in.-diam rolls). Increasing the speed from 200 rpm up to 300 rpm resulted in only a slight further increase in throughput. Typical grinding rates for unfueled graphite were 50 g/min through a nominal 0.003-in. gap, and 200 g/min for a 0.045-in. gap. If an improved roll crusher could reduce graphite to 100% -140 mesh in a single pass at the same rate per inch of roll length, an 8-in.-diam x 12-in.-long set of rolls could produce about 400 kg of ground fuel per day from 16-mesh feed.

Despite the deficiencies of this particular machine, it demonstrated that the application of a roll crusher to grinding of HTGR fuel is promising, and it indicated certain areas of machine design that should receive special attention. On the basis of information obtained with this machine, the General Engineering and Construction Division of ORNL began design of an improved roll crusher with the following features:

1. Roll dimensions, 12 in. diam x 4 in. long. The larger diameter should make possible reduction of material from -6 mesh to -12 mesh in one pass, and from -12 to -140 mesh in a second pass. Although the rolls could probably nip material as large as 2.5 mesh, the 15 x 8 hammer mill will be used, as before, to supply feed of -6 mesh. The roll will be made from AISI-01 steel hardened to about 60 Rockwell C.

2. The adjustable roll will be mounted in a pivoting yoke for setting the gap between rolls. Two heavy springs

between roll shafts will serve to preload the bearings and linkages which affect roll spacing.

3. The rolls will be geared for a differential roll speed of about 10% in order to promote a lapping action on the roll surfaces during normal operation. In-place lapping with fine abrasive will also be tested.

2.2.1.2 Ball-Mill Grinding. -- A ball mill was investigated[8] as an alternative device for fine grinding. Reactor-grade graphite was ground in a 4-in.-diam mill, using 1-in.-diam ceramic balls or 0.4-in.-diam steel balls, and the effect of grinding time and graphite inventory were investigated. Even at a low graphite inventory the +140 mesh fraction decreased from 90% to only about 30% in 30 min. The principal conclusion was that ball-milling is too slow to be of much interest, and no further work is planned.

2.2.2 Grind-Leach Run GL-2

HTGR fuel (General Atomic type "A" compacts for Peach Bottom) was ground to -50 mesh and leached with 13 M HNO_3--0.05 M F in bench-scale equipment. The loss to the washed residue was 2.1% of the thorium and 0.81% of the uranium. Most of the loss was caused by unfractured carbon-coated ThC_2-UC_2 beads in the -50+140 mesh fraction.

The fuel was first crushed in the 15 x 8 hammer mill, and then passed through stages of roll crushing, as outlined below:

Pass	Machine	Discharge Aperture	Feed Size	Product Size	Approx. Rate
1	Hammer mill, 15 x 8-in.	1/8 x 8-in. slots	1 in.	75% -6 mesh	-
2	Roll crusher, 8 x 2-in.	0.045 x 2 in.	100% -6 mesh	70% -16 mesh	130 g/min
3	Roll crusher, 8 x 2-in.	0.002 x 2 in.	70% -16 mesh	99% -35 mesh	70 g/min
4	Roll crusher, 8 x 2-in.	0.002 x 2 in.	99% -35 mesh	99% -50 mesh 48% -140 mesh	200 g/min

[8] H. O. Witte and L. M. Ferris, Ball-Milling of Graphite-Bed Reactor Fuel. ORNL-TM-1412 (February 3, 1966).

Microscopic examination of samples of the -50+100 mesh fraction showed no intact beads after the fourth pass. Therefore the entire -50 mesh portion from this final pass was used in experiment GL-2. The equipment and experimental procedure were the same as reported[9] for run GL-1, with one minor exception: 1 liter of leachant was put into the leacher before any of the ground fuel was added. Despite this precaution, the fuel formed a solid plug that required about an hour to disperse. Although the recirculation rate of leachant was maintained at about 120 ml/min (the lowest rate at which the charge of ground fuel was fully fluidized), about 35% of the charge was entrained out of the 3-in.-diam settling section of the leacher and was later recovered separately from the bulk of the leached residue. Permeability of the leached residue to downflow of water was about 23 gal hr^{-1} ft^{-2} for a 9-in. bed depth at a pressure drop of about 11 psi.

Leaching conditions and results are summarized in Fig. 2.3. Thorium and uranium analyses of all size fractions are shown in condensed form in Table 2.1. The uranium loss for all material smaller than 100 mesh was about 0.5%, and the -50+100 mesh fraction contained fuel beads with the still-intact carbon coatings amounting to about 3% of the total beads present in that size range before leaching. The reason for the thorium loss being higher than the uranium loss is not known.

The conclusions drawn from this experiment were: (1) Peach Bottom fuel should be ground to -140 mesh (105 μ) to ensure rupturing all particle coatings; (2) a means should be provided for continuous, controlled addition of ground fuel to the leacher; (3) future leaching equipment design should provide simple "well-mixed" contact between ground fuel and leachant with the objective of obtaining the maximum possible slurry concentration and solution loadings; (4) solid-liquid separation after leaching can probably best be done in a separate piece of equipment.

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[9] M. E. Whatley *et al.*, Unit Operation Section Quarterly Progress Report, July-September 1965, ORNL-3916.

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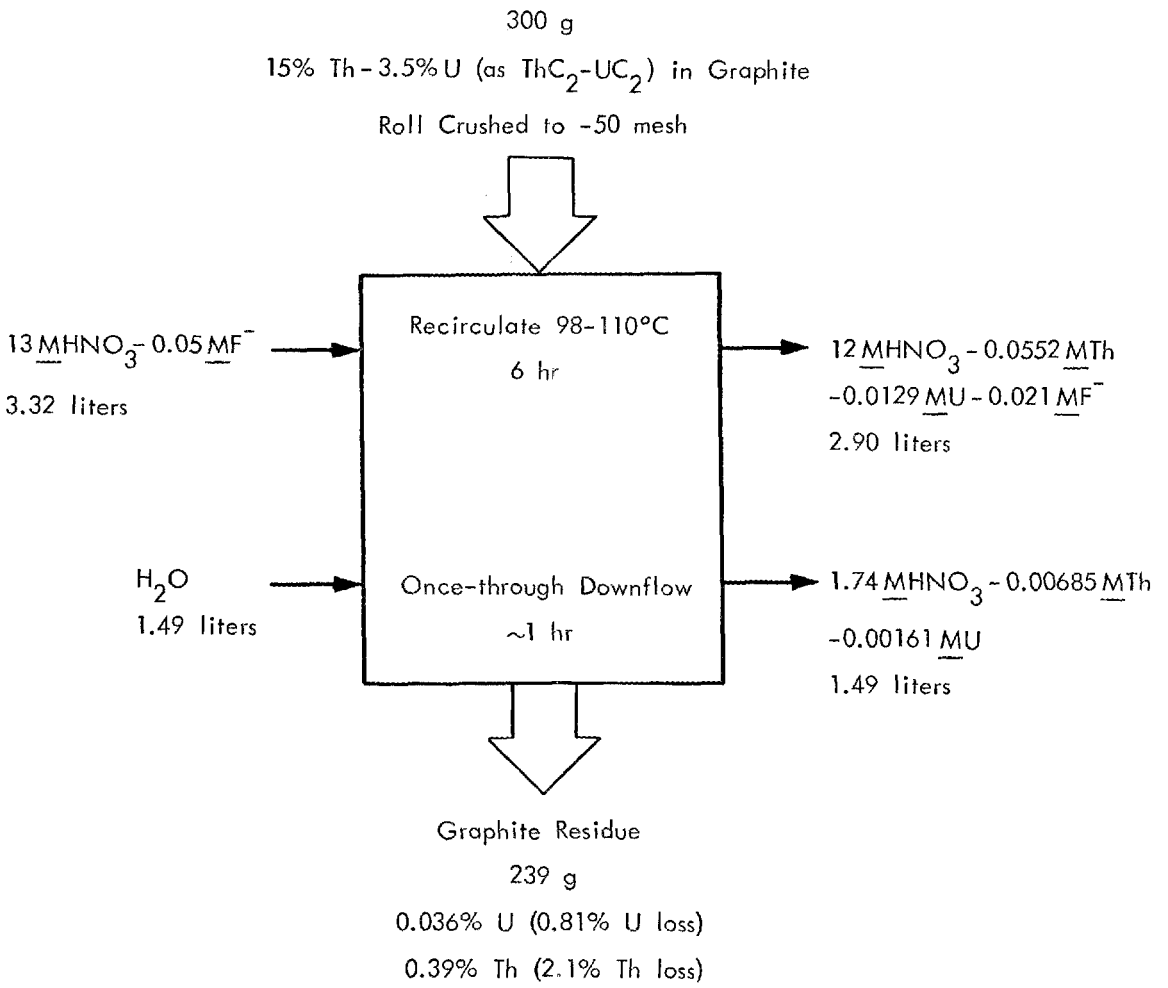

Fig. 2.3 Grind-Leach Processing of Peach Bottom Fuel (run GL-2). This fuel contains pyrolytic-carbon-coated microspheres of ThC_2-UC_2.

Table 2.1 Distribution of Thorium and Uranium Before and After Leaching, by Sieve Fractions (Run GL-2)

Mesh Size U.S. Series	Initial Inventory Total wt (g)	Initial Inventory Th (g)	Initial Inventory U (g)	Final Inventory Total wt (g)	Final Inventory Th conc (%)	Final Inventory Th wt (g)	Final Inventory U conc (%)	Final Inventory U wt (g)	Heavy Metal Loss to Residue Th (%)	Heavy Metal Loss to Residue U (%)
-50 +100	84	4.92	1.13	84	0.19	0.16	0.048	0.040	3.2	3.6
-100 +140	71	20.8	4.65	35	0.21	0.074	0.064	0.023	0.36	0.49
-140	145	19.4	4.71	120	0.59	0.70	0.019	0.023	3.6	0.48
Totals, g	300	45.1	10.5	239		0.93		0.086		

2.2.3 Grind-Leach Run GL-3

On the basis of conclusions reached from run GL-2, engineering-scale equipment was designed and assembled for leaching up to 5 kg of ground feed per batch. In the first experiment in the new equipment, run GL-3, losses (0.35% of the uranium and 0.67% of the thorium) to the leached and washed residue were lower. The improved recovery was largely due to finer grinding of the fuel (100% -140 mesh) preparatory to leaching. Also demonstrated were (1) a pneumatic method for controlled addition of ground fuel to the leacher, (2) higher solution loadings, (3) satisfactory suspension of a concentrated slurry of ground fuel and leachant during leaching, (4) rapid transfer of the slurry to a filtration column, and (5) reasonable rates for separation of leachate and washes from the graphite.

The engineering-scale batch leaching equipment, shown schematically in Fig. 2.4, was sized to process 10 liters of graphite-acid slurry containing as much as 5 kg of ground fuel. The -140 mesh roll-crusher product (Peach Bottom fuel) was air-lifted from the feeder by fluidizing the material only at the base of the feeder while injecting air into the vertical lift tube. The ground fuel flowed out of the upper feeder section into the leacher; the air for lifting plus the air purge to the upper section kept vapor from the boiling acid from penetrating into the addition line. The rate of addition of ground fuel was limited to about 75 g/min by the generation of foam from its initial reaction with the boiling 13 M HNO_3--0.05 M F^-. During the 7-hr leaching period, the ground fuel was suspended by boiling and by injecting air into the draft tube. Subsequent transfer of the slurry to the filter vessel was effected in 30 sec by applying a vacuum of about 20 in. of Hg to the filter vessel.

The flow rate of product solution and three water washes through the 6-in.-diam bed of ground fuel in the filtration vessel ranged from 320 ml/min to as low as 80 ml/min, for a total time of 48 min. Permeability data are summarized in Table 2.2. The slight decline in permeability at a pressure gradient of 13 psi/ft is probably caused primarily by

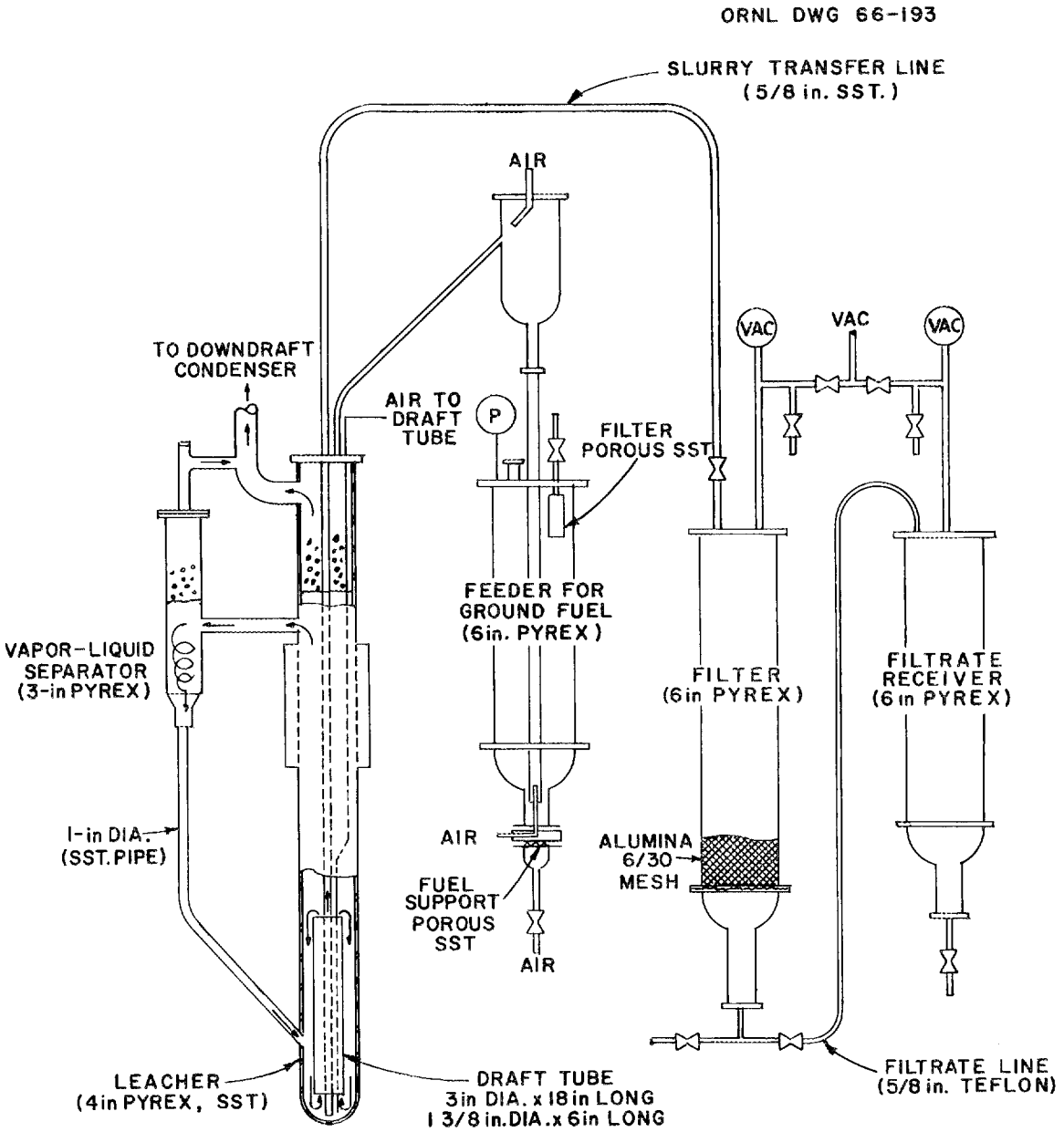

Fig. 2.4 Engineering-Scale Leaching Apparatus for Finely Ground HTGR Fuel.

Table 2.2 Permeability of Leached Graphite Residue to Leachate and Wash Solution

Equipment Diam (in.)	Equipment Area (ft^2)	Bed Height (ft)	Solution Description	Temp. (°C)	Permeability: Flux ($gal\ hr^{-1} ft^{-2}$)	Permeability: Pressure Gradient ($psi\ ft^{-1}$)	
6	0.2	0.67	Product solution[a]	~ 80	25	18	
			First wash[a]	~ 30	23	18	Water washing
			Second wash[a]	~ 25	6.2	18	
			Third wash[a]	~ 20	7.2	18	
1 1/2	0.012	0.88	Fourth wash[b]	90	21	13	A portion of the bed transferred to 1 1/2-in.-diam equipment for additional washing with 0.1 M HNO_3
			(b)	90	21	13	
			(b)	90	19	13	
			(b)	90	14	13	

[a]For composition and volume see Fig. 2.5.

[b]Each increment of washing = 160 ml ≈ one interstitial displacement.

compaction of the bed with time. However, the range of flows obtained at a pressure gradient of 18 psi/ft may also reflect the changing composition of the solutions. Extension of the permeability data to a wider range of pressure gradient will help define the optimum gradient.

Flowsheet conditions and leaching results are summarized in Fig. 2.5. Solution samples taken during leaching showed that the concentration of dissolved carbon reached a peak value of about 3.5 g/liter at 1.5 hr and slowly decreased to 1.15 g/liter at 7 hr. The composite solution would require a feed-adjustment step, which might include permanganate oxidation of the dissolved carbon, if its presence caused difficulties in solvent extraction.[10]

Loss of uranium and thorium to the residue under flowsheet conditions was 0.35% and 0.67%, respectively. However, additional washing of a portion of the residue using 0.1 M HNO_3 (Table 2.2) lowered the losses to about 0.2% and 0.5%, respectively. In terms of the original flowsheet, the additional washing would have increased the total mixed solution volume from 12.6 liter to 22.5 liter. Therefore the additional uranium and thorium recoveries were achieved at the cost of approximately doubling the volume of mixed solution requiring feed adjustment and of doubling the time required for washing.

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[10] L. M. Ferris, personal communication.

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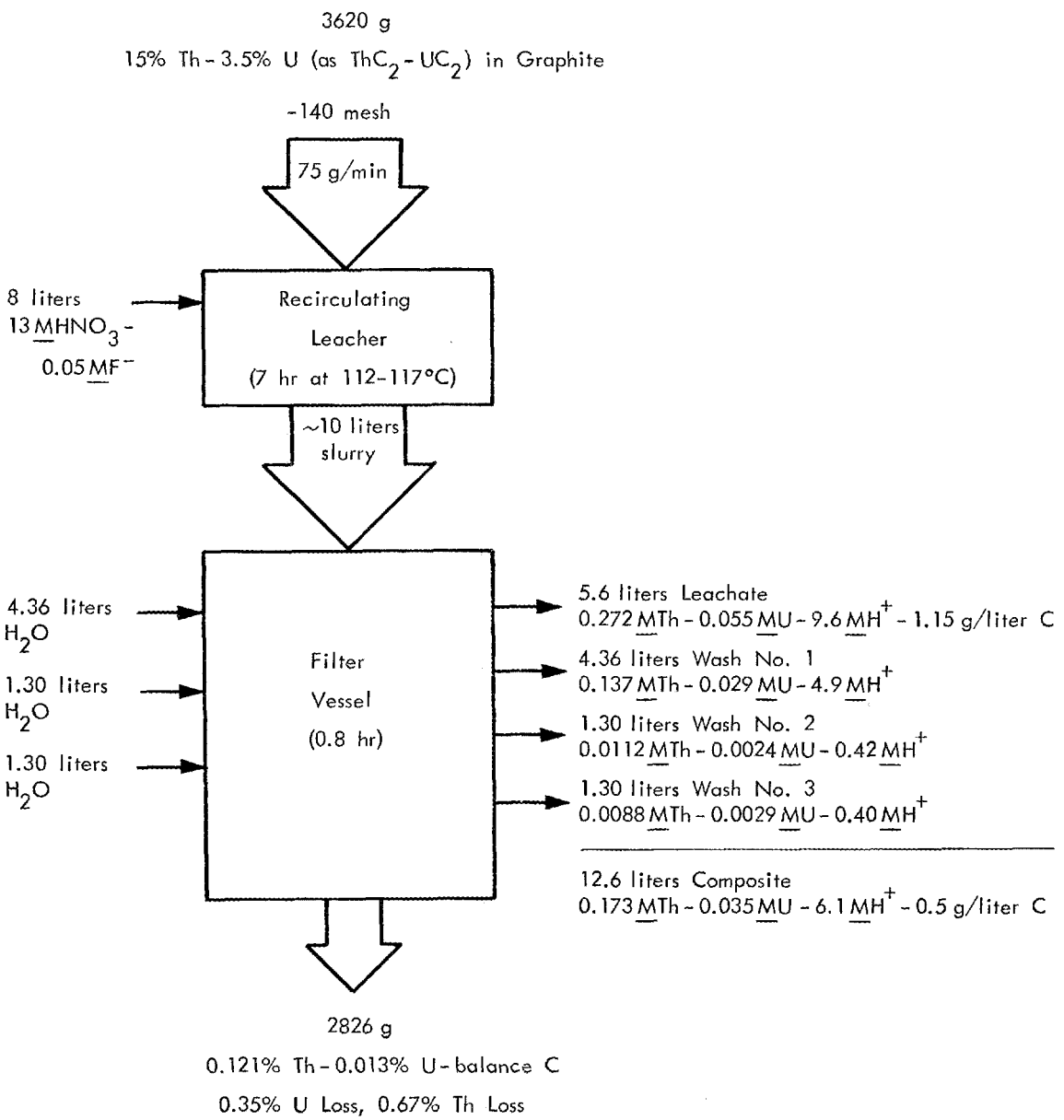

Fig. 2.5 Engineering Scale Grind-Leach Processing of Peach Bottom Fuel Ground to -140 Mesh (run GL-3).

3. THE ENGINEERING SEPARATION OF MACROMOLECULAR BIOLOGICAL SUBSTANCES

C. W. Hancher H. O. Weeren

The initial step in the program of separating kilogram quantities of purified mixed transfer ribonucleic acids (t-RNA) from E. coli has been partially completed. Approximately 300 kg of E. coli-w cells were processed to yield 632 g of mixed t-RNA. An amino acid acceptor activity analysis of the product showed that the activity is equivalent to commerical mixed t-RNA and to small batches of mixed t-RNA produced locally.

The initial processing steps were done in the previous quarter.[11] The E. coli-w cells were teated with phenol to rupture the walls. The phenol and cell residues were removed by centrifugation; the nucleic acids were precipitated with ethanol and separated by centrifugation. The RNA was separated from the bulk of the DNA (deoxyribonucleic acid) by fractional precipitation with isopropyl alcohol. The yield was 7.9 kg of crude product.

The next step of the process (purification of the RNA by adsorption on and subsequent elution from DEAE cellulose) removes whatever ribonuclease may have survived the phenol treatment and a great deal of other material besides. This operation can be most conveniently carried out as a series of batch treatments. The solids precipitated by isopropanol were dissolved in 0.05 M NaCl and slurried with DEAE cellulose. The solution was drained from the cellulose, and the cake was washed with 0.2 M NaCl. The RNA was then eluted with 0.6 M NaCl and precipitated with ethanol. The resulting solid was further purified by dissolution in NaCl solution, centrifuging to remove DEAE fines, precipitating the RNA with ethanol, and centrifuging to collect the precipitate. This precipitate was dried at 40°F under vacuum and weighed. A total of 632 g of dry solids was obtained from the 7.9 kg of solids treated with DEAE.

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[11] M. E. Whatley et al., Unit Operations Section Quarterly Progress Report, July-September 1965, ORNL-3916.

Samples of the purified dry solids were analyzed for optical activity, concentration of RNA and DNA by colorimetric analysis, and concentration of specific t-RNA. The analyses of the eight batches were much the same; a composite analysis of the whole 632 g is given in Table 3.1. A similar analysis of commerical t-RNA (strain B) is given for comparison.

The next step in the program is the further processing of the mixed t-RNA product to separate specific t-RNA from the mixture. Phenylalanine t-RNA is the first RNA for which a separation process has been developed and will consequently be the first to be separated in purified form. The separation process consists of a gradient elution from a diatomaceous earth column saturated with a quaternary ammonium compound. This has been highly successful on a small scale but must be scaled up manyfold to handle the large quantity of feed material now available.

A literature reference describes a process for the extraction of t-RNA from E. coli cells where phenol is not used to rupture the cell walls; isoamyl alcohol is used to make the cell walls selectively permeable. Since phenol is relatively expensive and somewhat inconvenient to use because of its effect on the skin, other processes look promising enough to warrant an intermediate-scale test. A batch of E. coli cells (strain B) was grown in the fermenter at Y-12, harvested, buffered, treated with isoamyl alcohol, and centrifuged. The solids were discarded, and the solution was treated with ethanol to precipitate the RNA. A total of 14.5 kg of E. coli cells were processed to yield 970 g of ethanol precipitate. A small batch was treated with DEAE cellulose and submitted for analysis of RNA activity. None was found. It is believed that nucleases that were extracted from the cells with the RNA destroyed the RNA before the processing had progressed far enough to remove these nucleases. In any event, this process does not look promising.

Table 3.1 Product Analysis

Amino Acid	μμ moles/ODU[a] charged	Commercial t-RNA (Strain B)
Alanine	16.3	14.4
Arginine	51.9	38.3
Aspartic Acid	24.5	23.0
Asparagine	32.4	33.0
Cysteine[b]		
Glutamic Acid[b]		
Glutamine[b]		
Glycine	15.9	9.1
Histidine	17.1	11.2
Isoleucine	13.4	10.0
Leucine	39.0	53.5
Lysine	47.0	36.8
Metheonine	62.0	60.0
Phenylalanine	37.4	35.5
Proline	4.0	6.3
Serine	30.9	36.7
Threonine[b]		
Tryptophane[b]		
Tyrosine	30.9	28.2
Valine	992.8	76.5

[a]ODU - optical density units, the greater the number the higher the activity.

[b]Counts per min too low to be meaningful.

ODU/mg at 206 - 12.0

% DNA 4.8

% RNA 76.3

4. SOL-GEL PROCESS DEVELOPMENT

P. A. Haas

The sol-gel process development studies have included laboratory investigations of sol preparation for most of the metal oxides that might be used in irradiated specimens or fuel elements. One purpose is to obtain the same advantages for these metal oxides as for thoria made by sol-gel processes. Another purpose is to be able to prepare mixed oxide products by preparing mixed sols or by mixing pure sols. Engineering-scale studies are being made for the urania, urania-carbon, and urania-thoria sol-gel processes. These studies are closely related to the thorium fuel-cycle studies covered in Section 5 of this report.

4.1 Urania Sol Preparation Studies

J. J. Perona

The process under development consists of (1) reducing uranyl nitrate in aqueous solution to uranous nitrate, (2) precipitating hydrous uranous oxide with 3 M NH_4OH--0.5 M N_2H_5OH solution, (3) filtering and washing with demineralized water, and (4) heating the washed filter cake to speed its conversion to a sol. The uranous nitrate solution, which is 0.5 M in U^{4+}, 2.5 M in NO_3, and 0.25 M in urea, is made 0.3 M in formic acid before the precipitation step. The chemical development of this process is described by McBride *et al.*[12]

4.1.1 Reduction of Uranyl to Uranous Nitrate in Aqueous Solution

A decrease in activity of the catalyst (0.5% platinum on alumina cylinders 1/8 in. in diameter and 1/8 in. long) was observed. Conversions greater than 99% for UO_2^{2+} to U^{4+} were previously reported[13] for a continuous tubular reactor containing about 1 kg of catalyst and

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[12] J. P. McBride, Preparation of UO_2 Microspheres by a Sol-Gel Technique, ORNL-3874, (also ORNL-TM-1110).

[13] M. E. Whatley *et al.*, Unit Operations Section Quarterly Progress Report, July-September 1965, ORNL-3916.

operating at 500 psig and 70°F with a flow rate of about 1 liter/hr. After about 15 liters of solution had been processed, the conversion dropped to an average of 84% for the next eight liters and to 81% for the next six.

The catalyst was regenerated by soaking it in 5 M HNO_3 and then washing it with water. After this, conversions greater than 99% were again obtained.

4.1.2 Precipitation and Washing

Four experiments were made, none of which resulted in a satisfactory sol. The first three filter cakes (Table 4.1) contained too little nitrate, probably because of excessive washing. The highest had a nitrate-to-uranium mole ratio of 0.059, compared with the desired value of 0.15 to 0.25. These filter cakes were sufficiently fluid to be removed from the filter by suction through 1/4-in. tubing, but they did not drain cleanly from the sides of the glass container. They appeared to form unstable sols, and within 24 hr they had coagulated. Nitric acid was added to adjust the nitrate-to-uranium ratio to 0.2, but the solids did not resuspend.

The last two filter cakes contained too much uranyl ion as a result of the deactivation of the catalyst in the reduction equipment. Sols made from them coagulated within 24 hr.

The filter was made from 12-in.-diam glass pipe 15 in. long with a porous stainless steel disk at the bottom end for the filter element (Fig. 4.1). Batches of sol containing up to 1.5 kg of uranium can be made with this filter. The uranium and neutralizing solutions are mixed before entering the filter by being pumped continuously through the inlet and bottom ports of a 0.5-in.-diam hydroclone. The slurry leaves through the top port and flows onto the filter plate, where a flow of argon up through the plate promotes further mixing and ensures an inert atmosphere.

Table 4.1 Urania Sol Preparation: Summary of Precipitation and Washing Experiments

Uranium solution: 0.5 M U^{4+}, 2.5 M NO_3^-, 0.25 M urea, 0.3 M HCOOH
Neutralizing solution: 3 M NH_4OH--0.5 M N_2H_5OH

Run Number	Volume ratio neutralizing solution / uranium solution	Final pH	Number of washes	Volume of H_2O per wash (liters)	Filter Cake Composition: NO_3/U^{4+} Mole ratio	Filter Cake Composition: U^{4+}/total U
F-1	0.70	~ 7[a]	10	2.5	0.030	0.868
F-2	0.72	~ 7[a]	10	3.25	0.059	0.847
F-3	0.637	7.23	8	3.5	0.035	0.758
F-4	0.71	6.7	5	2.25	0.205	0.728

[a] pH determined by use of pH indicator paper.

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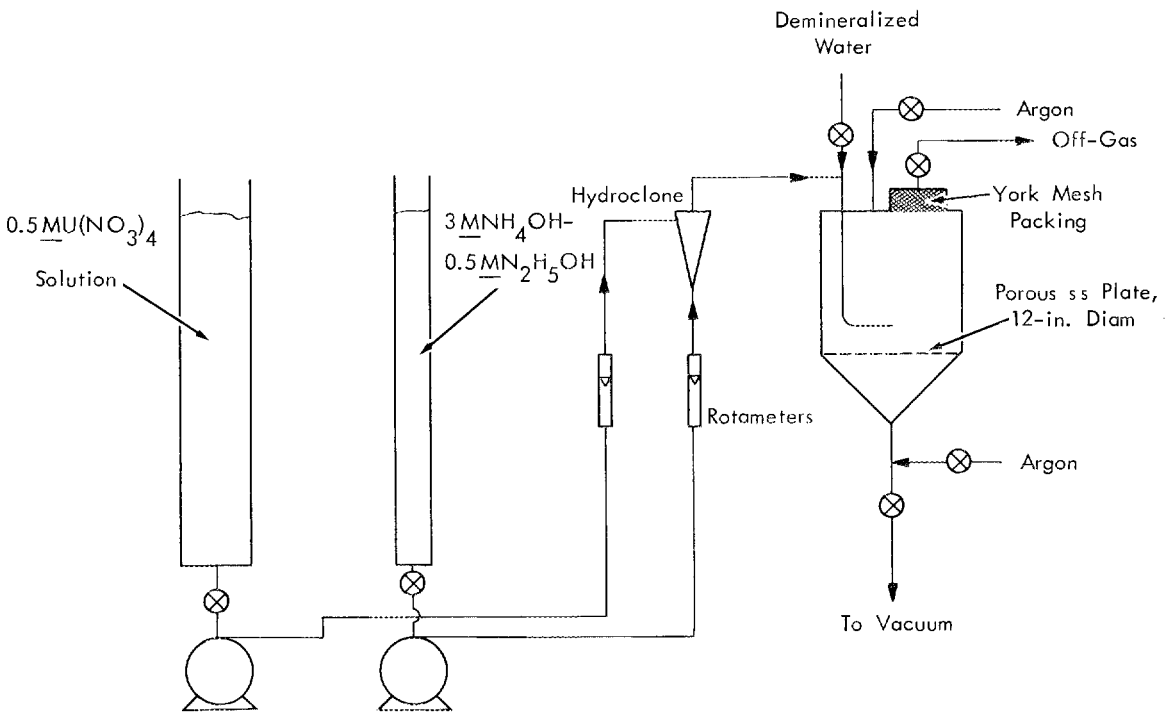

Fig. 4.1 Urania Sol Preparation. Precipitation and washing equipment.

For an 8-liter batch of $U(NO_3)_4$ solution, the neutralization was completed in about 30 min. The oxide precipitate must stand for 30 to 40 min while the pH of the filtrate drops before the final pH adjustment to 7 can be made. For example, the pH of the filtrate in F-4 was 6.0 after standing 15 min and 4.7 after 35 min.

A different procedure was used in F-3, in which the precipitation was done on the filter over a period of 2 hr 40 min, with mixing by the argon sparge. A smaller amount of neutralizing solution was used with this method, which probably shows that the mixing or diffusion time was not sufficient.

The settling rate of the precipitate was a strong function of pH. In run F-2, the mixing was stopped at a pH of 5, and the rate of increase of clear supernatant liquor was measured for 10 min. The rate was constant at 0.13 in./min, compared with a rate of about 1 in./min at a pH of 7.

The washed filter cakes were warmed to 40 to 50°C over a period of about four hours. The resulting material had the flow characteristics of a Bingham plastic, being quite fluid once some yield stress was exceeded.

5. THORIUM FUEL CYCLE STUDIES

P. A. Haas

We are developing the equipment and procedures necessary to adapt the thoria sol-gel process to remote operation in a shielded facility (TURF). A TURF cell mockup is used for experimental tests of remote maintenance procedures, manipulators, and services. We continued process development studies for preparation of thoria microspheres by dispersion of a sol into 2-ethylhexanol, extraction of water to give gel microspheres and drying and calcining to give high-density oxide microspheres. Development of high-capacity sol dispersers and additional tests of solvent recycle are necessary to demonstrate high process capacity and long-term operation. A microsphere column system designed for 10 kg/day of ThO_2 was placed in operation.

5.1 Microsphere Preparation Development

S. D. Clinton P. A. Haas

The process development studies for forming gelled oxide microspheres in 2-ethylhexanol (2EH) were continued in the 2-in. minimum diameter, tapered, glass column.[14] The capacity of the distillation system associated with this column is about 1 liter/min of 2EH, which limits the flow rate of sol droplets that can be gelled continuously to approximately 10 ml/min. Several different rotary dispersers were tested at sol flow rates up to 24 cc ml/min for short periods of time. The rotary and shear dispersers[15] are both considered to be promising methods for obtaining uniform sol dispersion on a large scale, particularly for producing thoria microspheres in the size range of 300 to 600 μ.

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[14] M. E. Whatley *et al.*, Unit Operations Section Quarterly Progress Report, July-September 1965, ORNL-3916.

[15] M. E. Whatley *et al.*, Unit Operations Section Monthly Progress Report, December 1964, ORNL-TM-1091, pp 24-35.

A multiple nozzle developed by D. M. Helton,[16] which controlled dispersion of sol by the application of electrical potentials, was tested in the microsphere column, and the resulting thoria size distributions compared favorably with those obtained from previous experiments with a two-fluid nozzle.

For a sol flow rate of 10 ml/min and a still temperature of 150°C, the steady-state water content of the 2EH in the column is about 1.5 wt %. At water concentrations between 1.5 and 2.0 wt % in 2EH, the probability of partially gelled microspheres clustering together seems to increase. The 2-in.-minimum-diameter, tapered, glass column has been operated continuously for 6 to 7 hr with no apparent problems at sol flow rates up to 10 ml/min. For higher sol flow rates the column operation has been limited to periods of less than 1 hr. The purpose of these short tests is to determine the performance of sol dispersion devices.

Five rotary dispersers (see Table 5.1) of principal types I and III[17] were tested in the tapered column. For the type-I dispersers, eight orifices are equally spaced in a horizontal plane on a cylindrical surface; for type III, the eight orifices are equally spaced on a circle in the base of a thin disk. The unpredictable wetting behavior between the aqueous sol and the stainless steel dispersers has a pronounced effect on the size distribution of droplets produced. Both types I and III gave product uniformity similar to that of the two-fluid nozzle, but the product from the rotary dispersers has not been consistently uniform.

The test conditions and sizes of the calcined product from the dispersion of a 3.0 M thoria sol by five rotary dispersers are shown in Table 5.2. The weight of each product sample was between 300 and 500 g

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[16] D. M. Helton, Dispersion of a Liquid Stream by an Electrical Potential: Applications to the Preparation of Thoria Microspheres, ORNL-TM-1395, January 17, 1966.

[17] M. E. Whatley *et al.*, Unit Operations Section Quarterly Progress Report, April-June 1965, ORNL-3868.

Table 5.1 Description of Rotary Dispersers

Each disperser has eight orifices located symmetrically on a specified pitch diameter

Code Letter	Type	Diameters (in.) Orifice	Pitch	Maximum	Other Features
H	III	0.016	1.50	1.75	Holes in base of 1/4-in.-high disk
I	III	0.010	1.50	1.75	Holes in base of shallow cone
J	I	0.010	1.50	1.50	Holes in sides of 1-in.-high disk
K	I	0.016	1.50	1.50	Holes in sides of 1-in.-high disk
FR	I	0.016	1.75	1.75	Holes in sides of 3/16-in.-high disk

Table 5.2 Rotary Disperser Product Size Distributions

Rotary Feeder Type	Rotary Feeder Code	Product Code	Sol Feed Rate (ml/min)	Tangential Velocity at the Orifice (ft/sec)	Mean Diameter d_{50} (μ)	$\left(\frac{d_{80} - d_{20}}{d_{50}}\right)100$ (%)
III	H	6X	9.6	0.8	525	18
III	I	6Y	9.6	0.8	480	36
		8E	9.6	1.9	250	34
		8A	9.6	2.2	240	46
		7G	9.6	3.1	120	41
		7F	9.6	3.5	88	41
I	J	7X	4.8	1.5	490	35
		7W	9.6	1.5	290	14
		7Y	9.6	1.5	320	30
		7Z	~ 20	1.5	300	25
I	K	6U	9.6	0.8	575	39
		6W	9.6	1.2	450	29
		7A	9.6	1.7	420	25
I	FR	7D	9.6	0.9	535	7
		7E	9.6	1.2	460	12

except for 6X and 7A, which were 850 and 1300 g, respectively. In general, the size-distribution results were similar to those reported previously for rotary dispersers,[18] but the size uniformity of products 7D and 7E was as good as the best obtained with any of the sol-dispersion devices. For the type-I dispersers, these results indicate that a shallow disk of the minimum thickness practical to contain the sol flow channels is desirable. The products 7F and 7G show that a rotary disperser can produce calcined microspheres in the size range of 100 μ with some loss in uniformity.

An electrostatic sol disperser (Fig. 5.1; dimensions shown on Drawing D-58139) was developed in Chemical Development Section A and tested in the Unit Operations microsphere column. This device has fifteen No. 24 hypodermic needles at a controlled positive potential with a copper ring at ground potential around each needle. The mode of dispersion varies with voltage in three distinct regions. At low voltages (up to about 500 v) the size of drops formed is independent of voltage. At intermediate voltage (from about 500 v to 1000 or 1200) a single stream of uniform drops is produced; the size of drops decreases as the voltage is increased. At voltages greater than the upper transition voltage a fan-like spray of nonuniform drops is produced. Over the range of intermediate voltages, the sol drop size is a function of voltage, sol flow rate, and sol concentration. With the electrostatic, multiple-nozzle disperser, the calcined product size distributions for a 3.0 M thoria sol were similar to the best obtained with a single two-fluid nozzle (Table 5.3). The products 7H, 7I, and 7J were formed during 1-hr tests, but product 7T was produced during a 6.5-hr continuous run. The operation of the electrostatic disperser for the extended run appeared to be satisfactory; although one of the 15 needles plugged about midway of the run, column operation and product size distribution were not significantly affected.

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[18] M. E. Whatley *et al.*, Unit Operations Section Quarterly Progress Report, April-June 1965, ORNL-3868.

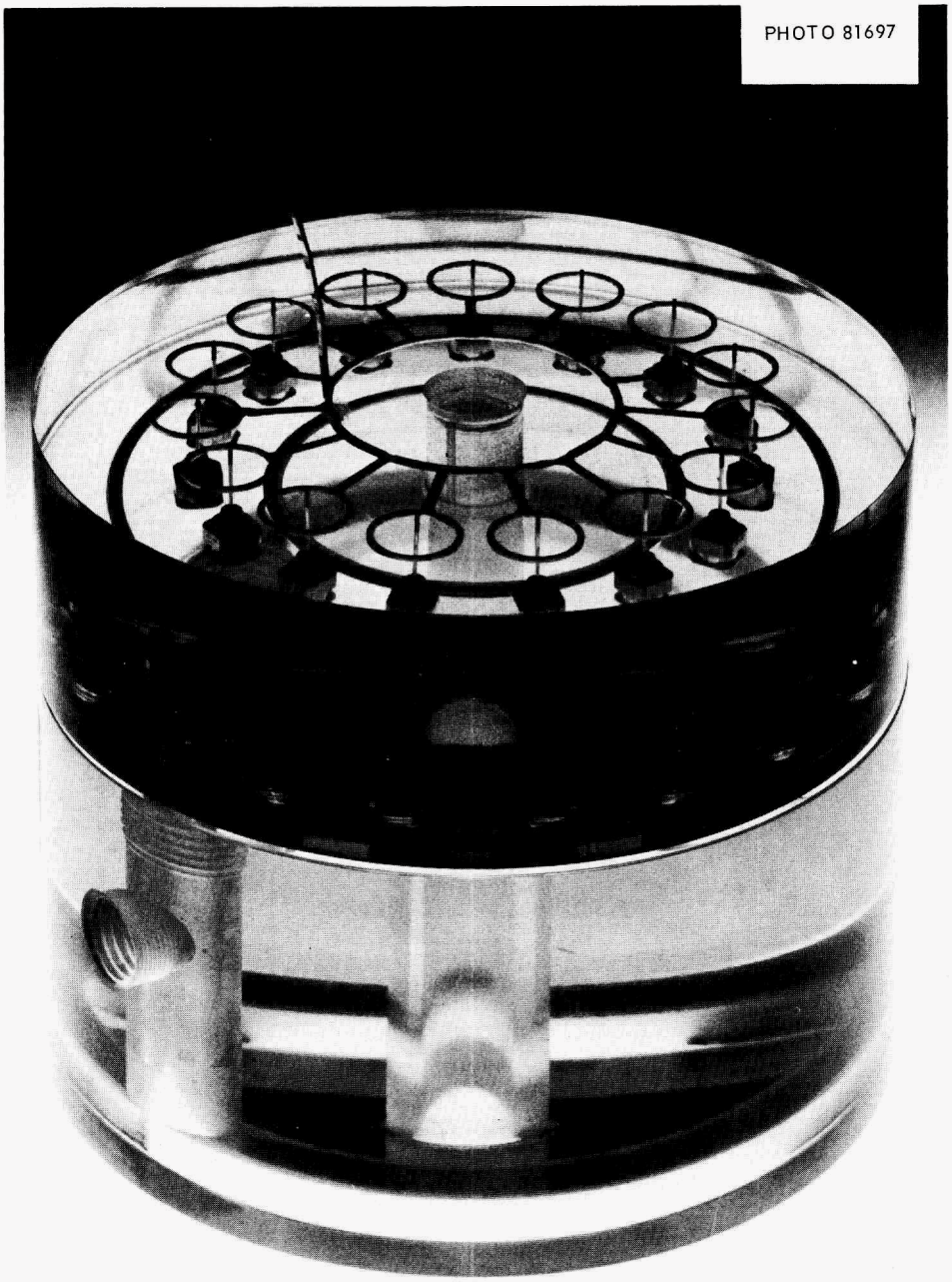

Fig. 5.1 Electrostatic Nozzle Disperser. Fifteen No. 24 hypodermic needles in parallel as one electrode, with a ring around each needle as the second electrode (details shown on DWG D-58139).

Table 5.3 Product Size Distributions from an Electrostatic Sol Disperser

Product Code	Product Weight (g)	Sol Feed Rate (ml/min)	Applied Potential (v)	Mean Diameter, d_{50} (μ)	$\left(\frac{d_{80} - d_{20}}{d_{50}}\right) 100$ (%)
7H	420	9.6	915	310	11
7I	210	4.8	915	240	10
7J	200	4.8	720	275	11
7T	2810	9.6	748	290	12

Several 100- to 200-g batches of urania microspheres were produced in the column to test the forming characteristics of different urania sol preparations. Future column runs will be made to study the gelation behavior of different urania and thoria-urania sols and to continue the testing of sol dispersion devices.

5.2 Microsphere Preparation Without Fluidization

P. A. Haas S. D. Clinton

Alternatives to the fluidized bed in a column are being considered for suspending sol drops during the extraction of water to cause gelation. Operation of a continuous fluidized-bed column requires uniform sol-drop sizes and an accurate matching of the fluidizing flows to the drop size. Also the accumulation of sol or gel spheres in a fluidized bed may increase the clustering or coalescence. The alternatives were considered with the expectation of avoiding one or both of these limitations.

Long lengths of 1/4-in.-ID polyvinylchloride tubing (100 ft for one set of tests and 200 ft for another) were used to provide the residence time necessary for gelation. Sol drops dispersed in 2-ethylhexanol (2EH)

were fed into this tubing from a two-fluid nozzle. The mixture discharged into a settler where the gel spheres accumulated; the wet 2EH overflowed to the distillation system for water removal. The variables were: tubing length, sol drop size, 2EH flow rate, and the dimensions and orientation of the coil of tubing.

A completely satisfactory set of conditions was not found. The principal difficulty was that the drops settled to and moved very slowly along the wall wherever the tubing was not vertical. For 2EH and 1/4-in.-diam tubing, the flow was laminar for all practical 2EH flow rates. Thus a 100- to 400-μ-diam particle at the tubing wall was in a layer of low average velocity. A second difficulty is that the tubing length required increases greatly with increasing product diameter, since both the time required for gelation and the average velocity needed to prevent accumulation of particles increases as the particle diameter increases. The tubing length required is the product of this average velocity and the gellation time. For these reasons, the testing of longer lengths of tubing and higher velocities appeared desirable. The sol feed pump and the 2EH pump limited the maximum pressure drop through the tubing to about 40 ft of 2EH. This pressure drop was adequate for all the flow rates of interest in the 100-ft length of tubing, but it limited the flow rates that could be tested in the 200-ft length.

The drops settled to the lower tubing surface in all nonvertical portions and were suspended only in vertical portions. The initial test was with the tubing coiled on an 18-in.-diam cylinder with the coil (cylinder) axis in the horizontal plane. The drops were suspended only for a few inches each side of each point which had a vertical tangent and were moving slowly along the tubing wall for about 80% of the tubing length. For this arrangement, the average sol or gel-sphere residence time was much longer than the average 2EH holdup time. Dried gel spheres of 100 to 300 μ diameter were discharged, but coalescence of sol drops at the feed end and beds of dried gel spheres near the outlet were both observed.

The remaining tests were in coils with 18-in. straight sections connected by turns on 3-in. radii. With the straight sections vertical and the coil axis horizontal, the settling was much less than that encountered in the 18-in. cylinder. With a 2EH flow rate of 340 cc/min (average 2EH residence time of 2.8 min), 175-μ gel spheres (120-μ calcined product) were satisfactorily prepared. The 100-ft length of tubing was not suitable for 210-μ gel spheres (150-μ calcined product) since all 2EH flow rates were either too low to keep the drops moving or too high to permit complete gelation (not enough residence time). For the 200-ft length, the maximum 2EH flow rate (as limited by the pumps) was that adequate for 210-μ gel spheres. When the straight sections were horizontal or slanted very slightly downward (coil axis vertical) with the 200-ft length, the accumulation and coalescence of wet sol drops near the entrance was worse than when the coil axis was in a horizontal plane. From these results, multiple vertical sections of enlarged cross section (columns) connected by short transfer connections of reduced cross section appear to be more attractive than simple coils. Use of a less viscous alcohol to permit turbulent flow in a coil of tubing would also be desirable.

5.3 Coated-Particle Development Facility (CPDF) Microsphere Column

C. C. Haws P. A. Haas

The microsphere column system in the Coated-Particle Development Facility (CPDF) was operated with thoria sols at rates up to 25 ml/min of thoria sol or 1200 g of ThO_2 per hr. The operation of the column itself has been excellent; the only significant difficulties were with accessories such as the dispersers and the product driers. The capacity of the system is limited by the package boiler system to about 25 ml of sol feed per minute. The capacity of the product driers appears to be about 10 kg/24 hr.

This system was operated with 3 M thoria sol feed using as dispersers a rotary disperser, a shear nozzle, and a pair of two-fluid nozzles. With a 1-7/8-in.-diam rotary disperser having twelve 0.010-in.-diam

orifices, 48% to 67% of the calcined ThO_2 microspheres were in the size range 250 to 297 μ at 220 to 270 rpm. An undesirable amount of fine particles was produced at 270 rpm, and smaller orifices would be preferred in order to make smaller microspheres. For test runs with 2 two-fluid nozzles at 4.9 ml of sol per minute to each nozzle, 79% of the calcined ThO_2 microspheres were 210 to 250 μ in diameter. The column will be operated with these nozzles to prepare 210- to 250-μ product needed for pyrolytic carbon coating studies.

6. TRANSURANIUM PROCESSING STUDIES

A. D. Ryon F. L. Daley

In support of the Transuranium facility, engineering studies are being made to form the basis for design and operation of the process equipment. This report presents the results of cold testing of two solvent extraction racks.

6.1 Pulse Column Tests

Final cold testing of the Tramex solvent extraction rack was completed. The rack consists of three pulse columns, pulsers, and receiver tanks. Each pulse column is 1.5 in. in diameter by 60 in. high and is packed with sieve plates (1/32-in. holes, 5% free area) spaced 1/4 in. apart. The columns were first operated at design flow rates (3:1:1:1 liters/hr of solvent, feed, scrub, and strip, respectively) with a feed whose physical properties at room temperature simulated the physical properties of Tramex process solutions at 50°C. During about 80 hr of operation the major difficulty was failure of Zircaloy-2 diaphragms in the pumps and pulsers. Then the columns were demonstrated to perform effectively with the solutions having compositions (other than radiation emitters) typical of those in the Tramex flowsheet. Again the main trouble was failure of diaphragms. These were replaced with alloy diaphragms (90% Ta--10% W), and no more failures occurred.

During the final period of testing, the columns were operated 24 hr a day for about 130 hr. The feed contained 100 ppm Zr, which can cause emulsions if it is hydrolyzed. The feed acidity was adjusted by evaporation to 120°C, which leaves just enough acid to prevent emulsions. The adjusted feed composition was: 10.2 M LiCl, 0.1 M $AlCl_3$, 0.2 M HCl, 0.1 M $SnCl_2$, 100 ppm Zr, and 2 vol % methanol. The scrub solution was 11 M LiCl and 0.03 M HCl. The solvent was 0.6 M Adogen (tertiary amine) in diethylbenzene; both were purified by contact with silica gel and activated alumina. The physical properties of the test solutions are shown in Table 6.1. A sample batch of Zr feed was mixed with solvent at an A/O ratio of 3 in a batch mixer at 25°C to determine

the extent of emulsion formation. The time required for phase separation ranged from 95 to 124 sec, and the amount of emulsion remaining after centrifugation was less than 0.005% of the feed.

Table 6.1 Physical Properties of Test Solutions

	Density (g/cc)	Viscosity (centipoises)
Tramex solutions at 50°C		
Solvent: 0.6 M Adogen in DEB	0.848	2.67
Feed: 10 M LiCl, 0.1 M $AlCl_3$, 0.2 M HCl	1.230	3.34
Scrub: 11 M LiCl, 0.03 M HCl	1.232	3.34
Strip: 8 M HCl	1.115	1.03
Simulated Tramex at 25°C		
Solvent: 0.3 M Alamine 304 in DEB	0.872	2.70
Feed : 1.5 M $Al(NO_3)_3$, 0.1 M $Ni(NO_3)_2$,	1.267	3.34
Scrub : 1 M HNO_3		
Strip: 0.16 M HNO_3, 0.1 M $Ni(NO_3)_2$	1.018	0.95

The columns were successfully operated for a while at design flow rates at 50°C, using the Zr feed, but after 22 hr it was necessary to reduce the flow rate to 60% of design to prevent flooding of the extraction and scrub columns. After 32 hr of operation at this rate, the flow rate had to be further reduced to 50% of design, at which rate the columns operated for 80 hr without further difficulty. No flooding of solvent out of the bottom of the columns was observed; the only sign of flooding was that the aqueous phase did not flow out fast enough even with no pressure applied to the pressure pot on the jackleg.

In an attempt to understand the loss of throughput experienced in the operation of the TRU pulse column, the same solutions were tested in the 3/4-in.-ID x 48-in.-high glass pulse-column assembly. A total of 23 liters of Zr feed was run through the extraction and scrub cycles. Some emulsion formed at the top of the extraction column and filled the

disengaging section. About two-thirds of the emulsion migrated down the extraction column and passed out with the raffinate stream (0.10 ml of emulsion per 100 ml). The remaining third overflowed as emulsion with the organic stream, passed through the scrub column, and overflowed to the strip column (0.05 ml of emulsion per 100 ml). The formation of emulsion and solids in no way interfered with column operations. In fact, the throughput (21:7:7 ml/min of organic, feed, and scrub) exceeded the throughput of previous tests (12:4:4 ml/min). The extraction column was operated organic-continuous for 8 hr and then returned to aqueous-continuous emulsion operation with no apparent effect. One possible explanation of reduced throughput in the TRU columns could be partial plugging of the pulse-loss restrictors on the aqueous outlets of the columns.

During the last 10 hr of operation, $NiCl_2$ (4 g Ni per liter) was added to the feed to permit mass transfer efficiency calculations. More than 99.9% of the nickel was extracted, thus the stage height in the extraction column was less than 10 in. This result agrees with previous data obtained in 3/4-in.-diam glass columns. Stripping of nickel with 8 M HCl was only 96.8% complete. Entrainment of aqueous phase in the waste solvent was 0.2%, and the aqueous phase was colorless, indicating low nickel concentration. At shutdown, the solvent holdup in the columns was measured and found to be as expected: 20% in the extraction and scrub columns and 10% in the strip column.

6.2 Berkelium Extraction Rack Tests

The equipment rack to be used in the Transuranium facility for the separation and recovery of berkelium from californium and other actinides was tested for hydraulic stability and separation efficiency. The major components of the extraction rack include a batch solvent spray column (1.4 in. in inner diameter, 57.5 in. long) containing two spray heads, a feed hold tank, a scrub hold tank, a product receiver tank, and a feed pump, all made of Zircaloy-2.

The product of the separation of americum and curium from berkelium and californium is collected and the aqueous solution is adjusted to 10 M HNO_3--0.1 M $KBrO_3$ (volume 1.5 liters, sp. gr. 1.30), in which the berkelium is oxidized to the extractable tetravalent state. Berkelium is extracted into 0.75 liter of 0.6 M 2-ethylhexylphosphoric acid in N-Paraffin-175 (sp. gr. 0.79), and the solvent is scrubbed with 2.5 liters of 10.0 M HNO_3--0.01 M $KBrO_3$ to remove the californium and other actinides. The berkelium is stripped from the solvent with 2.6 liter of 10 M HNO_3--1.0 M H_2O_2 strip solution, which reduces the berkelium back to the trivalent state. All tests were made at about 45°C.

The feed and scrub nozzles at the top of the column were found to be plugged. The scrub inlet was unplugged by backflushing, but the feed inlet remained plugged. Testing of the feed circuit was therefore done through the scrub inlet. It was possible to maintain a metered flow as low as 25 ml of 10 M HNO_3--1.0 M H_2O_2 per minute without gas binding of the feed pump. Aqueous flow rates in excess of 55 ml/min resulted in flooding of the column and dumping of about 25% of the solvent to the raffinate catch tank. The aqueous-phase holdup in the column at a flow rate of 50 ml/min was about 50%. The operation procedure[19] outlined by the TRU facility group was tested and the hydraulics of the system were verified except that, before the flushing operation, the interface should be raised in the column to prevent loss of solvent.

Two runs were made to test the chemical flowsheet using cerium and europium as standins for berkelium and californium, respectively. In the first run, extraction of the cerium was 92.9% complete after the second feed pass and 99.5% complete after the fourth. The decontamination factor (DF) from europium was only 120. Nearly 10% of the cerium remained in the solvent waste after stripping with 10 M HNO_3--0.1 M H_2O_2.

A second run was made using about three times as much scrub and strip, and the concentration of H_2O_2 was increased to 1 M in the strip to

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[19] O. O. Yarbro, ORNL, personal communication.

improve the stripping efficiency. Cerium extraction was 96.9% complete after the first pass and greater than 99.6% complete after the third. The concentration of europium in the third liter of aqueous scrub solution was only 0.001 g/liter. The DF of cerium from europium was about 1300 after contact with 4 liters of scrub solution. Cerium stripping was 98.1% complete after contact with 2 liters of strip solution and greater than 99.7% after 5 liters of strip solution. Table 6.2 contains the data.

Table 6.2 Berkelium Extraction Rack-Test Data

Operation	Run 1 Volume (liters)	Run 1 Concentration (g/liter) Cerium	Run 1 Concentration (g/liter) Europium	Run 2 Volume (liters)	Run 2 Concentration (g/liter) Cerium	Run 2 Concentration (g/liter) Europium
Feed	1.5	3.5	1.8	1.5	2.6	1.9
1st Extn. pass		0.14	1.2		0.08	1.4
2nd Extn. pass		0.25	1.4		0.02	1.4
3rd Extn. pass		0.042	1.3		< 0.01	1.4
4th Extn. pass		0.017	1.2			
1st Scrub	1.25	< 0.01	0.02	1.0	0.01	0.16
2nd Scrub	1.25	< 0.01	< 0.001	1.0	< 0.01	0.003
3rd Scrub				1.0	< 0.01	0.001
4th Scrub				1.0	< 0.01	< 0.001
5th Scrub				1.0	0.01	0.001
6th Scrub				1.0	< 0.01	< 0.001
7th Scrub				1.0	< 0.01	< 0.001
8th Scrub				1.0	< 0.01	< 0.001
9th Scrub				1.0	0.01	0.003
Total scrub	2.50			9.0		
1st Strip	3.0	1.0	0.006	1.0	3.30	0.003
2nd Strip				1.0	0.35	< 0.001
3rd Strip				1.0	0.03	< 0.0005
4th Strip				1.0	0.02	< 0.0005
5th Strip				1.0	< 0.01	< 0.0005
6th Strip				1.0	< 0.01	< 0.0005
7th Strip				1.0	< 0.01	< 0.0005
8th Strip				1.0	< 0.01	< 0.0005
9th Strip				1.0	< 0.01	< 0.0005
Total strip	3.0			9.0		
Waste organic	0.75	0.65	0.004	0.75	< 0.01	< 0.001

7. WASTE PROCESSING

C. W. Hancher J. C. Suddath

Waste-solidification test R-92 was a rising-level glass-making test with Purex waste 1 (PW-1). Test R-92 was made to determine the effectiveness of adding the Li_3PO_4 glass-making additive to the pot as a separate stream instead of premixing it with the waste feed solution as had been done in test R-91. When the glass additives were mixed with the feed in Test R-91, part of the chemicals precipitated and the resulting slurry could not be satisfactorily fed to the calciner pot.

7.1 Procedure and Material Balances

Purex waste 1 (PW-1) is a synthetic waste (Table 7.1) approximately 5 M in HNO_3, 1 M in $Fe(NO_3)_3$. It contains simulated fission products equivalent to a burnup of 20,000 Mwd/ton. Test R-92, which lasted for 100 hr, was done in a stainless-steel (304LC) pot, 12 in. in diameter and 6 ft high. The pot was heated in an induction furnace having a 15-in.-diam susceptor.

The glass-forming additives (Table 7.2) were added to the pot as a separate liquid stream. In the 100-hr test, the total amount of liquid feed was 852 liters of waste and 294 liters of additive; the average feed rates were therefore 8.5 liters/hr for waste and 3 liters/hr for additive. The condensate volume was the sum of the waste feed, the additive feed, feed-line washings (to remove plugs), and condenser washings (to remove entrained solids). The contributions to the condensate were (in liters):

Feed	852
Additive	294
Washings	260
Total	1406

The feed rate was controlled manually, starting at 15 to 20 liters/hr and then decreasing as the test progressed. After 68 hr of operation the pot became filled with a mixture of glass at the bottom, solid above

Table 7.1 Composition of Purex Waste 1

	Desired	Actual
	Chemical Constituents (M)	
H^+	5.0	4.9
Fe	0.93	0.93
Na	0.14	0.16
H_3PO_4	0.003	0.004
NO_3	7.8	8.0
Ni	0.005	-
Al	0.001	-
Cr	0.012	-
	Fission Products (M)	
Sr	0.015	-
Ba	0.019	-
Rare earths	0.118	-
Zr	0.065	-
MoO_3	0.065	-

Table 7.2 Composition of Glass-Making Additive Mixture

	Desired	Actual
	Concentration expressed as g-moles of additive per liter of original feed	
H_3PO_4	2.4	2.5
LiOH	1.37	1.1
NaOH	1.27	1.2
HNO_3	-	0.8

and on the walls, and a foam-liquid mixture in the center. After this condition was reached, the feed was stopped from time to time to prevent overfilling. The top thermocouples were used as level indicators. Inattention of the operators during the 44th and 45th hour allowed about 100 liters of feed to overflow from the pot to the condensing system.

The additive (Table 7.2) was made 0.1 M in sodium hypophosphite to decrease corrosion and was fed to the pot by means of a volume-ratio feed controller. For each volume of waste fed to the pot, 0.34 volume of additive solution was fed.

Both the feed line and the additive line plugged occasionally, although these lines were equipped with water purges intended to prevent plugging. When either of the feed lines plugged, the plug was removed with high-pressure water.

The additive liquid-level-probe line also plugged, resulting in a false indication of a high liquid level in the additive tank. The controller then caused the outlet valve on the additive feed tank to open fully, and the valve remained open until the plug was removed and the correct signal restored. This malfunction occurred about six or seven times during the test.

The material balance for the test (Table 7.3) was good except for iron (151%), which was high from pot corrosion, and phosphate, which was unexplainably low (60%). The semiglossy solid produced in the pot had the following composition of major constituents in wt %:

Fe	Na	PO_4	Li
22.1%	9.42%	18.2%	2.42%

The residual nitrate was 45 ppm.

The operating pot temperature was 900°C; the maximum temperature on the pot was 925 for not more than 30 min. After the feed was stopped, the pot was calcined for 9 hr at 900°C.

During the last half of the test, zone 1 was putting out additional heat to heat the overfilled pot. With zone 2 at its control temperature,

the large heat output from zone 1 caused zone 2 to overheat. Hence it was necessary to reduce the control temperature of zone 1 to about 800°C.

Test 7.3 Material Balance for Test R-92

	Gram-Moles of:			
	NO_3	Fe	Na	PO_4
Waste feed (852 liters)	6,816	792	136	3
Additive (294 liters)	646	-	981	1,990
Total in	7,462	792	1,117	1,993
Condensate (1404 liters)	8,564	252	449	744
Solids (236 kg)	-	938	968	454
Total out	8,564	1,190	1,417	1,198
Balance, %	115	151	127	60

7.2 Pot Corrosion

The pot was 12 in. in diameter by 97 in. long and was made of type 304L stainless steel about 500 mils thick. The thickness was measured at 30 places per heating zone by an ultrasonic thickness gauge. After the test the measurements indicated that the bottom zones of the pot were badly corroded (up to 239 mils; Table 7.4). The corrosion appears to be a function of the time that the area was covered with glass product: liquid, solid, or melt. The center thermocouple well was completely corroded away at the zone 5 and 6 levels.

Table 7.4 Wall Thickness of 12-in.-diam Type 304L Stainless-Steel Pot for Test R-92

Average wall thickness, in.; measured ultrasonically

Zone	Before Glass Test	After Glass Test
1	- 0.007 0.525 + 0.007	- 0.002 0.524 + 0.012
2	- 0.007 0.527 + 0.011	- 0.082 0.515 + 0.021
3	- 0.009 0.532 + 0.008	- 0.046 0.452 + 0.071
4	- 0.008 0.530 + 0.008	- 0.063 0.410 + 0.063
5	- 0.007 0.524 + 0.011	- 0.039 0.349 + 0.134
6	- 0.008 0.529 + 0.011	- 0.040 0.338 + 0.072

Maximum penetration - 0.239 in. in zone 6.

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