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UNIT OPERATIONS SECTION

QUARTERLY PROGRESS REPORT

JULY-SEPTEMBER 1965

M. E. Whatley
P. A. Haas
R. W. Horton
A. D. Ryon
J. C. Suddath
C. D. Watson

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
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ORNL-3916

Contract No. W-7405-eng-26

UNIT OPERATIONS SECTION QUARTERLY PROGRESS REPORT

July-September 1965

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MARCH 1966

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ABSTRACT

Chemical Engineering Research. - The flow capacity of the stacked-clone contactor is much greater when hexone is used instead of Amsco, while substitution of mineral oil, which is very viscous, has little effect on capacity. The average stage efficiency for uranium transfer in the hexone-aluminum nitrate system was 69%.

Processing Studies for the Molten-Salt Reactor. - One of the problems in close-coupled processing of molten-salt reactor fuels is the separation of volatile chromium fluoride from the UF_6, both of which derive from fluorination of the molten salt. Chromium fluoride can be sorbed on NaF at 400°C with a uranium loss of less than 0.01%. The capacity of low-surface-area NaF pellets is 20 g of chromium per 100 g NaF.

Processing of Power-Reactor Fuels. - Mechanical shearing of unirradiated Consolidated Edison and Elk River fuel assemblies was successfully demonstrated. On-site consultation about mechanical head-end equipment was provided the Nuclear Fuel Services processing plant. Initial testing of $HF-O_2$ decladding of stainless steel fuel in a fluidized bed has been unsuccessful because of caking of the aluminum oxide fluidizing medium. Vapor-liquid equilibrium data for UF_6-NbF_5 are being measured to aid in design of distillation equipment for their separation.

Thorium Fuel-Cycle Studies. - Thoria microsphere preparation and continuous calcination of thoria gels were studied. Both rotary and shear-nozzle dispersers produced satisfactory microspheres in the continuous drying column. Nineteen kg of thoria microspheres were prepared during an extended run. Microspheres smaller than 80 μ in diameter were easily made in a mixer-settler. A vertical tube furnace was demonstrated for continuous calcination of thoria-urania gel. Operating procedures were developed to avoid bridging and consequent breakage of the tube during calcination.

Sol-Gel Process Development. - The application of the sol-gel process to oxides other than thoria is being investigated. Natural, depleted, and enriched UO_2 microspheres were prepared in the continuous drying column. Several methods of preparing U^{4+} sols were tested.

Separation of Biological Macromolecular Substances. - The laboratory process for extracting mixed t-RNA from E. coli cells was modified and scaled up to the production of 1 kg. An outline of the purification procedure is presented. Continuous operation of the fermenter was demonstrated for 80 hr and was stopped after 300 kg of cells had been produced.

Radiation Resistance of Protective Coatings (Paints). - Continued screening of coatings has discovered three that can tolerate up to 8×10^9 r of gamma radiation when exposed in deionized water. This is 60% more than for any coating previously tested. The retention of radionuclides after exposure to conditions simulating release of radioactivity from a reactor excursion has been measured. The vinyl coatings show less retention than other generic types. In an attempt to standarize decontamination tests of protective coatings, a new method was tested. It is more reproducible and causes less damage to the coatings than the old method.

Previous Reports in this Series for Year 1965

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

1. CHEMICAL ENGINEERING RESEARCH

1.1 Stacked-Clone Contactor

W. S. Groenier A. D. Ryon M. E. Whatley

The Standard Mark X stacked-clone contactor is an 11-clone, stainless-steel, solvent-extraction apparatus featuring countercurrent liquid-liquid contact and a short residence time for the solvent. It consists of seven extraction stages and four polishing stages to reduce organic entrainment in the raffinate. The contactor has potential for use with highly radioactive systems, and its low liquid holdup and rapid response characteristics make it the ideal solvent extraction contactor for operations of short duration.

The contactor is being tested with a variety of chemical systems to evaluate its performance with liquids having a range of physical properties. A detailed description of the contactor, its development, and its performance with several chemical systems may be found elsewhere.[1,2]

The most recent testing has been with the system hexone--uranyl nitrate--acid-deficient aluminum nitrate. This flowsheet is characterized by a rather high aqueous-phase viscosity (2-5 centipoises), which would tend to decrease flow capacity, in conjunction with a solvent (hexone) known to have excellent flow-capacity characteristics. Previous work showed that systems having aqueous-phase viscosities as high as 1.2 to 1.3 centipoises [TBP-Amsco-$Al(NO_3)_3$ or Na_2CO_3 systems] with adequate interfacial tension and density difference between phases exhibit flow capacities of only 1.4 to 1.8 liters/min (A/O = 3), whereas flow capacities as high as 3.6 liters/min (A/O = 3) are observed where the aqueous-phase viscosity is near 0.7 centipoise (TBP-Amsco-$NaNO_3$ systems). Surprisingly, flow capacities

---

[1] M. E. Whatley and W. M. Woods, Jr., The Performance of an Advanced Stacked-Clone Contactor: A High Performance Solvent Extraction Machine with Potential for Application to Very Highly Radioactive Solutions, ORNL-3533 (April, 1964).

[2] Unit Operations Section Quarterly Progress Report, April-June 1965, ORNL-3868.

exceeding 7 liters/min (A/O = 3) were commonplace with hexone-acetic acid systems, where the physical properties are not very different from those for TBP-Amsco-$NaNO_3$. (Solvent residence times for the hexone-acetic acid systems were as low as 2 sec per theoretical stage.)

In the present work, the observed flow capacities for hexone--1.7 and 1.0 M $Al(NO_3)_3$, 0.2 M acid deficient, were 2.8 and 5.9 liters/min (A/O = 3), respectively. Although the density differences between phases were somewhat higher (0.474 and 0.356 g/cc, respectively) than for the systems described above, these flow capacities are amazingly high when considering the aqueous-phase viscosities and the past history of the contactor operating with aqueous solutions having viscosities greater than 1 centipoise. The data shown in Fig. 1.1, (largely taken from a previous report)[3] show quite vividly that the hexone flowsheets consistently exhibit higher flow capacities than flowsheets using an Amsco diluent, in spite of the high aqueous-phase viscosities when using acid deficient aluminum nitrate.

Flooding data were also obtained for a system exhibiting an extremely high solvent-phase viscosity; namely, mineral oil--0.01 M HNO_3. The flow capacity (A/O = 3) for this flowsheet was 3.9 and 4.3 liters/min at 28 and 40°C, respectively, and fits rather well with the flooding data correlation for flowsheets using an Amsco diluent (Fig. 1.1). This high throughput for a system exhibiting a solvent-phase viscosity of 116 centipoises (28°C) bears out the assumption that dispersed-phase viscosity has little if any effect on flow capacity. The high interfacial tension for this system (more than 50 dynes/cm) contributes to the observed throughput.

Complete flooding data for the present flowsheets are shown in Fig. 1.2 and Table 1.1. Physical-property data for the solutions tested are also given in Table 1.1. The range of physical properties for all the solutions tested so far with the stacked-clone contactor is as follows: density

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[3] Unit Operations Section Quarterly Progress Report, April-June 1965, ORNL-3868.

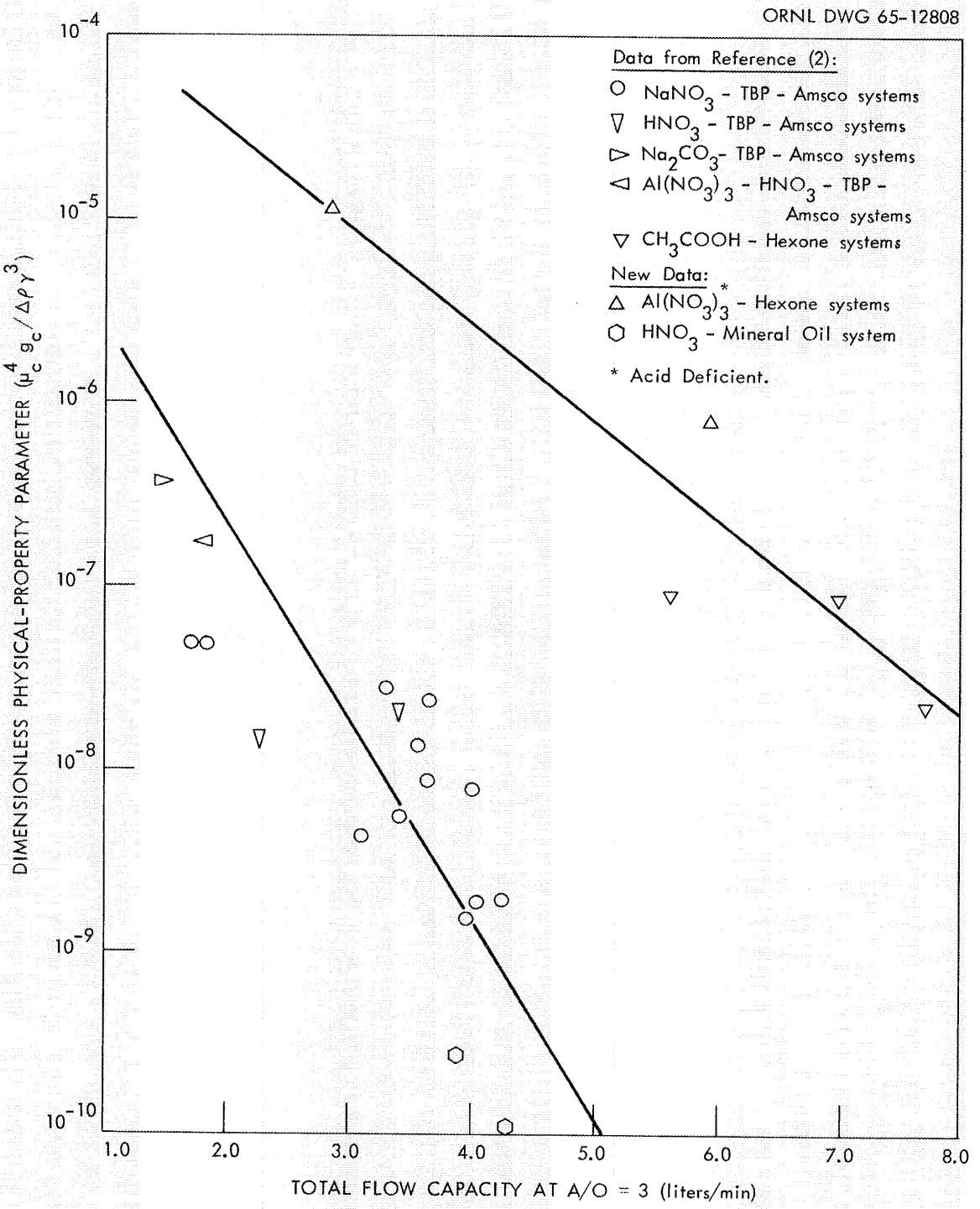

Fig. 1.1 Flooding Data Correlation for the Stacked-Clone Contactor.

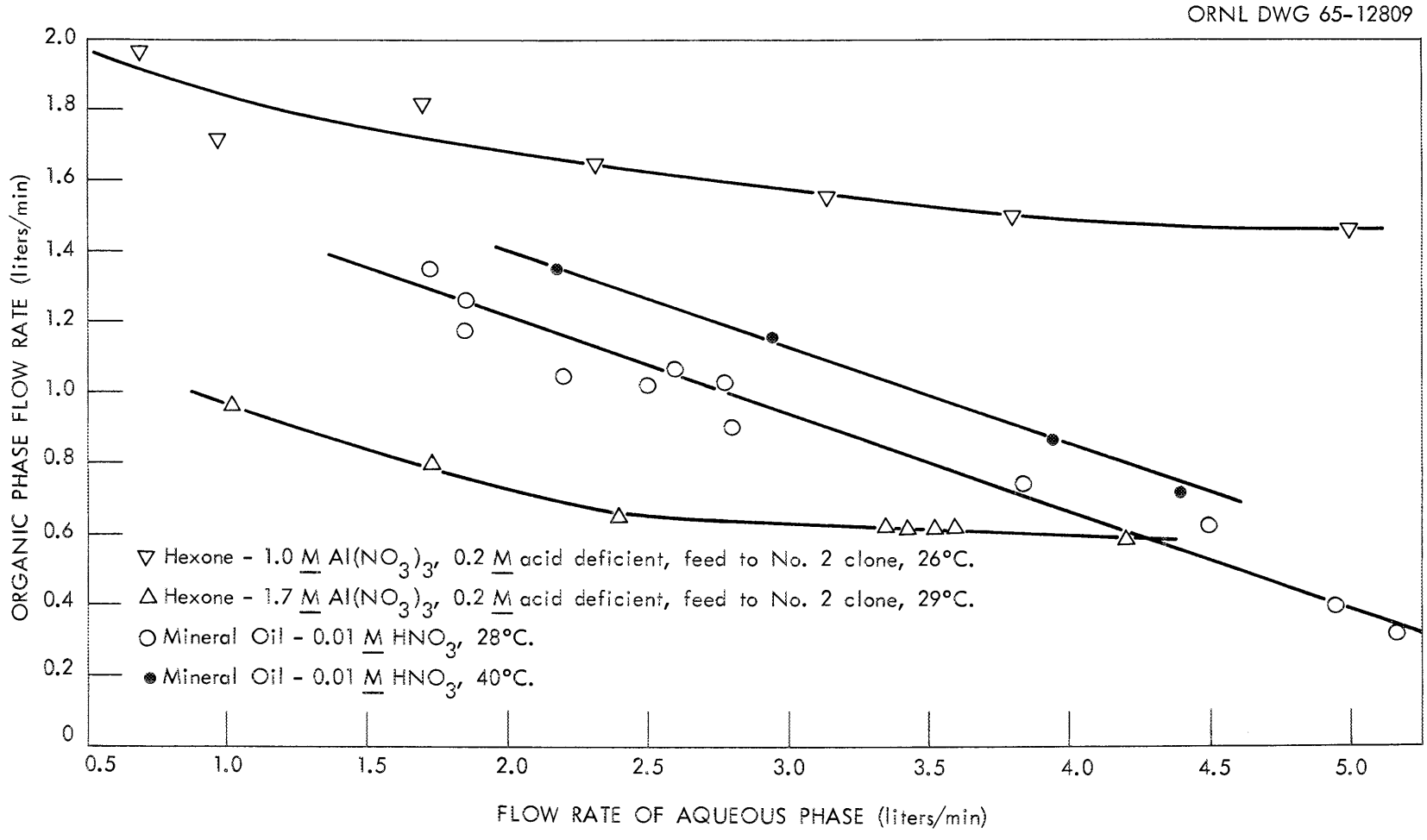

Fig. 1.2 Flooding Data.

Table 1.1 Flooding Data and Physical Properties of the Solutions

Aqueous phase	1.0 M $Al(NO_3)_3$[a]	1.7 M $Al(NO_3)_3$[a]	0.01 M HNO_3	0.01 M HNO_3
Organic phase	Hexone	Hexone	Mineral Oil	Mineral Oil
Density difference between phases, g/cc	0.356	0.474	0.121	0.124
Aqueous-phase density, g/cc	1.155	1.273	0.996	0.991
Organic-phase viscosity, centipoise	0.56	0.56	116.	57.
Aqueous-phase viscosity, centipoise	1.88	4.17	0.84	0.66
Interfacial tension, dynes/cm	7.6	8.1	52.7	51.9
Flow capacity at flooding, cc/min:				
A/O = 1/2	2790	-	-	-
A/O = 1	3430	1950	2770	-
A/O = 3	5880	2830	3880	4290
A/O = 10	-	-	5160	-
Temperature, °C	26	29	28	40

[a] 0.2 M acid deficient, aqueous feed to No. 2 clone.

difference between phases, 0.121 to 0.474 g/cc; aqueous-phase viscosity, 0.66 to 4.17 centipoises; organic-phase viscosity, 0.53 to 116 centipoises; interfacial tension, 6.7 to 52.7 dynes/cm.

Performance data for the stacked-clone contactor for the mass transfer of uranyl nitrate in a hexone--1.7 M $Al(NO_3)_3$--0.2 M acid deficient system are presented in Table 1.2. The uranium distribution coefficient for this flowsheet was dependent upon the sixth power of the aluminum concentration, a fact necessitating extremely precise measurements of aluminum concentration. Slight variations in aluminum nitrate content between aqueous feed solutions in extraction and stripping modes prevented the use of the usual approach to equalize efficiencies in each mode, where the slope of the equilibrium relation is adjusted slightly to effect the equalizing of efficiencies. Rather, adjustments were made by using different values of the slope in each mode, maintaining the proper difference between the slopes (distribution coefficients) as dictated by results of aluminum determinations. This procedure also minimized the efficiency dependence on flow ratio.

Although 36 runs were made in duplicate with this flowsheet, only 17 were useful in establishing a good average mass-transfer efficiency. In other runs, either the acid deficiency and/or the flow ratio was not proper, or material balances were poor, causing McCabe-Thiele-type calculations to give meaningless results. In some cases only extraction runs were made, so no stripping data were available to allow adjusted efficiencies to be obtained.

For the 17 useful duplicated runs, an average stage efficiency of 69% was obtained at a nominal flow ratio (A/O) of 11 and at flows approximately 60% of the flow capacity at flooding.

Table 1.2 Performance Data for the Stacked-Clone Contactor

Aqueous phase: 1.7 M $Al(NO_3)_3$, 0.2 M acid deficient
Organic phase: Hexone
Distributing species: $UO_2(NO_3)_2$

Run Series	Temperature (°C)	Adjusted Distribution Coefficient[a]	Average Flow Ratio (A/O)	Throughput at Flooding (cc/min)	Average Efficiency with Adjusted Distribution Coefficient (%)	RMS Deviation from Average Efficiency	Performance Factor[b]	No. of Duplicated Runs
Hexur - 6	26	12.65 E 10.68 S	10.55	5430	69.2	6.0	3.76	4
Hexur - 14	29	9.28 E 8.13 S	10.47	5440	83.5	10.0	4.54	5
Hexur - 29	28	10.38	10.14	5370	67.9	7.7	3.65	5
Hexur - 34	28	11.16	11.53	5650	46.6	2.4	2.63	3
					69.0 (Average)			

[a] Slope of the straight equilibrium line obtained by plotting organic vs aqueous equilibrium analyses, adjusted to force equal efficiencies in stripping and extraction modes. Different values were used for each mode where the $Al(NO_3)_3$ content varied.

[b] Product of average efficiency and throughput at flooding.

2. PROCESSING STUDIES FOR THE MOLTEN-SALT REACTOR

2.1 Chromium Fluoride Trapping

L. E. McNeese

At the conclusion of tests on the Molten-Salt Reactor Experiment, uranium will be recovered from the fuel salt as UF_6 by sparging the salt with F_2. Fluorides of chromium will be present in the fuel salt as a result of corrosion of the reactor piping and of the equipment used for hydrofluorinating or fluorinating the salt. A potential problem associated with recovery of the uranium is the presence of volatile fluorides of chromium (CrF_4 and CrF_5) in the fluorinator off-gas; these fluorides can not only contaminate the UF_6 product but can also render equipment inoperative by depositing in lines, valves, etc. Work is in progress which will permit the design of a trapping system for removing these fluorides from the off-gas, which will also contain UF_6 and F_2. A description of equipment and initial tests for evaluation of NaF as a trapping material has been reported previously.[4] It was concluded from these tests that pelleted NaF having a surface area 0.074 m^2/g and a void fraction of 0.277 was superior to pelleted NaF having a surface area of 0.45 for removing chromium fluorides from a gas stream containing F_2 at 400°C.

Six additional tests on the low-surface-area NaF for determining its capacity for chromium fluoride and for determining the extent of interaction between UF_6 and the chromium-sodium fluoride complex have been carried out and are discussed in the remainder of this section.

2.1.1 Conclusions and Future Work

Based on the results to date, it can be concluded that:

1. Fixed beds of NaF at 400°C are effective in removing fluorides of chromium from a gas stream which also contains UF_6 and F_2.

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[4] Unit Operations Section Quarterly Progress Report, April-June 1965, ORNL-3868.

2. Pelleted NaF having a surface area of 0.074 m^2/gm and a void fraction of 0.277 is superior to material having a surface area of 1 m^2/gm and a void fraction of 0.45, and has an effective capacity of about 20 g of chromium per 100 g of NaF.

3. Uranium losses of less than 0.01% are achievable when working with a gas stream that contains 0.4 mole of chromium fluoride per mole of UF_6 in F_2.

Future work will be directed toward determining conditions that minimize the loss of uranium to the 400°C NaF beds.

2.1.2 Procedure

In each of the runs, a salt charge of 3 kg of 38 mole % ZrF_4--62 mole % NaF containing 0.2 to 5.0 wt % chromium as CrF_3 was sparged with a F_2 flow of 0.9 slm at 650°C. During the last two runs, the sparge gas consisted of 0.9 slm of F_2 and 0.1 slm of UF_6. The fluorinator off-gas passed into NaF beds operated at 400°C for removal of chromium fluoride and thence to NaF beds operated at 100°C for removal of UF_6. The gases then passed into soda-lime beds at 25°C for removal of F_2.

Samples of the molten salt were taken after each run by inserting a cold 3/8-in.-diam nickel rod into the molten salt and withdrawing a sample. After each run, the NaF beds, which had been operated at 400°C, were divided into sections and sampled for analysis.

2.1.3 Results

Experimental conditions, quantities of chromium added to the salt before runs and removed during runs, and chromium content of the salt after the runs (by analysis) are shown in Table 2.1. During the runs, a total of 350 g of chromium was volatilized from the salt and collected on the 400°C NaF beds. The chromium content of the salt (chromium in solution) is not known since the rate at which metallic chromium dissolves in the salt is unknown.

Table 2.1 Experimental Conditions for Runs on Chromium Fluoride Removal

Run	F_2 Rate (slm)	UF_6 Rate (slm)	Sparge Time (hr)	Cr Addition Before Run	Cr Removed During Run (g)	Cr Content of Salt After Run (wt %)
CR-7	0.9	0	5	62.6 g as CrF_3	57.5	0.37
CR-8	0.9	0	6	104 g as CrF_3	30.4	0.218
CR-9	0.9	0	6	100 g as Cr	22.3	5.1
CR-10	0.9	0	9	None	43.1	-
CR-13	0.9	0.1	14	222 g as CrF_3	116.8	4.55
CR-16	0.9	0.1	15	None	80.0	2.90

The distribution of chromium on the NaF beds for runs CR-7 through CR-10 are shown in Fig. 2.1. The chromium loadings at the entrance to the bed range from about 11.5 to 17.5 g of chromium per 100 g of NaF.

The distribution of chromium and uranium on the NaF beds during runs CR-13 and CR-16 are shown in Figs. 2.2 and 2.3. Inlet chromium concentrations of 30 and 18 g of chromium per 100 g of NaF, respectively, were observed. For run CR-13, the uranium concentration was about 10% that of the chromium concentration and represented a uranium retention of 1% of the uranium passing through the bed. During run CR-16, a much lower uranium content, relative to chromium, was observed, and only 0.0075% of the uranium passing through the bed was retained. It is felt that the higher uranium retention during run CR-13 was caused by the presence of H_2O, introduced with the CrF_3. No CrF_3 addition was made before CR-16. The lack of a F_2 concentration sufficient to prevent decomposition of UF_6 on the NaF at 400°C during run CR-13 may also have contributed.

2.2 Containment Facility for Beryllium Fluoride Experiments

R. S. Lowrie

2.2.1 Pressure-Drop Measurements Across Soda-Lime Beds

The hoods being designed to house the beryllium molten-salt equipment will use a fixed-bed soda-lime trap to remove fluoride and other acidic gases from the exhaust air to protect the absolute filters, which are made of fibrous glass. The available space will limit the cross-sectional area of the traps, and, as a result, the maximum air velocities will be in the range 7.5 to 15 ft/sec. The fans commonly used in this service will limit the pressure drop across the bed to 6-8 in of water. Several tests were run with air at ambient temperature to determine the pressure drop across beds of commercial-grade soda-lime (-4 to +8 mesh, with a bulk density of 0.73 g/cc).

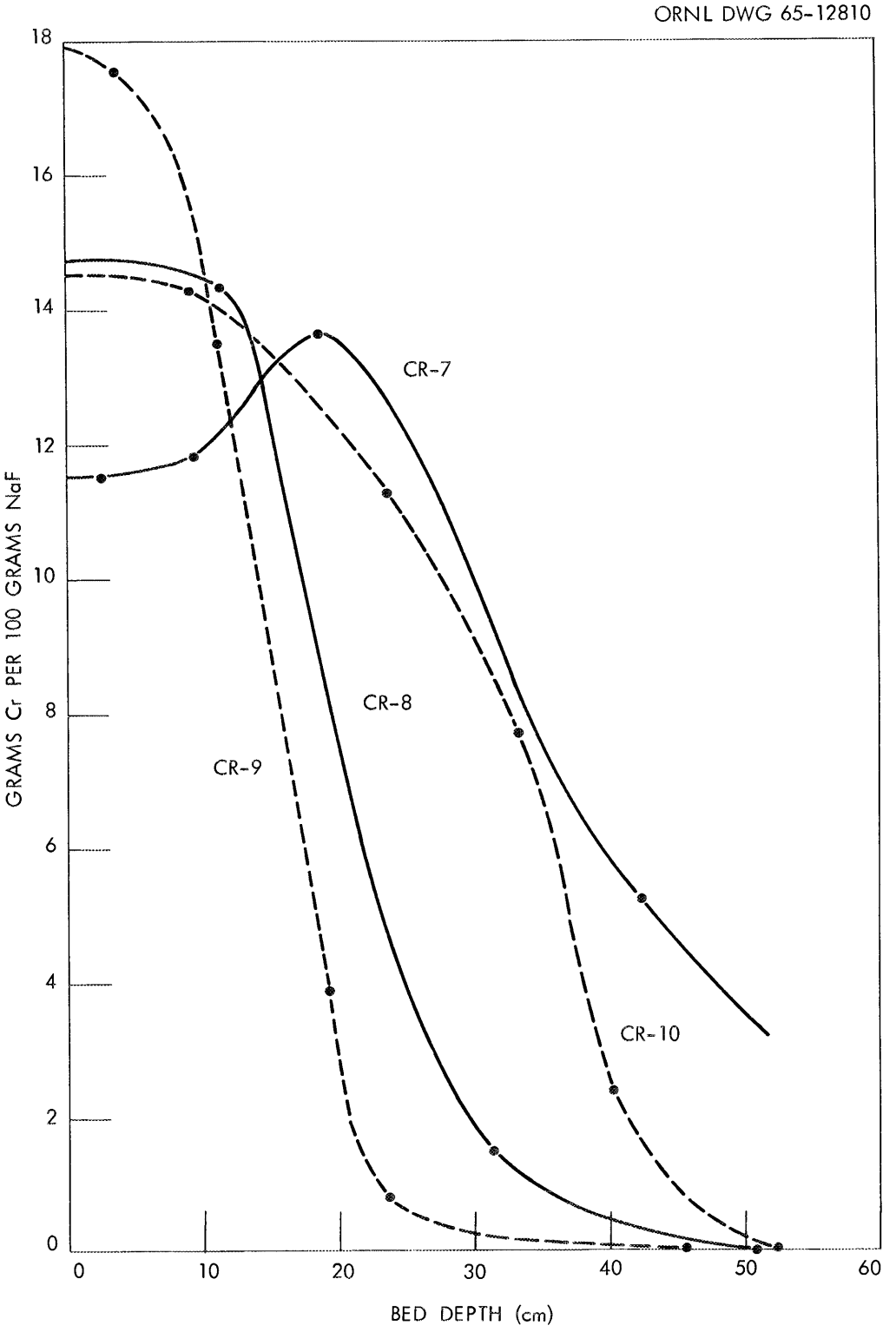

Fig. 2.1 Distribution of Chromium on 400°C NaF Beds for Chromium Fluoride Removal Runs CR-7 through CR-10.

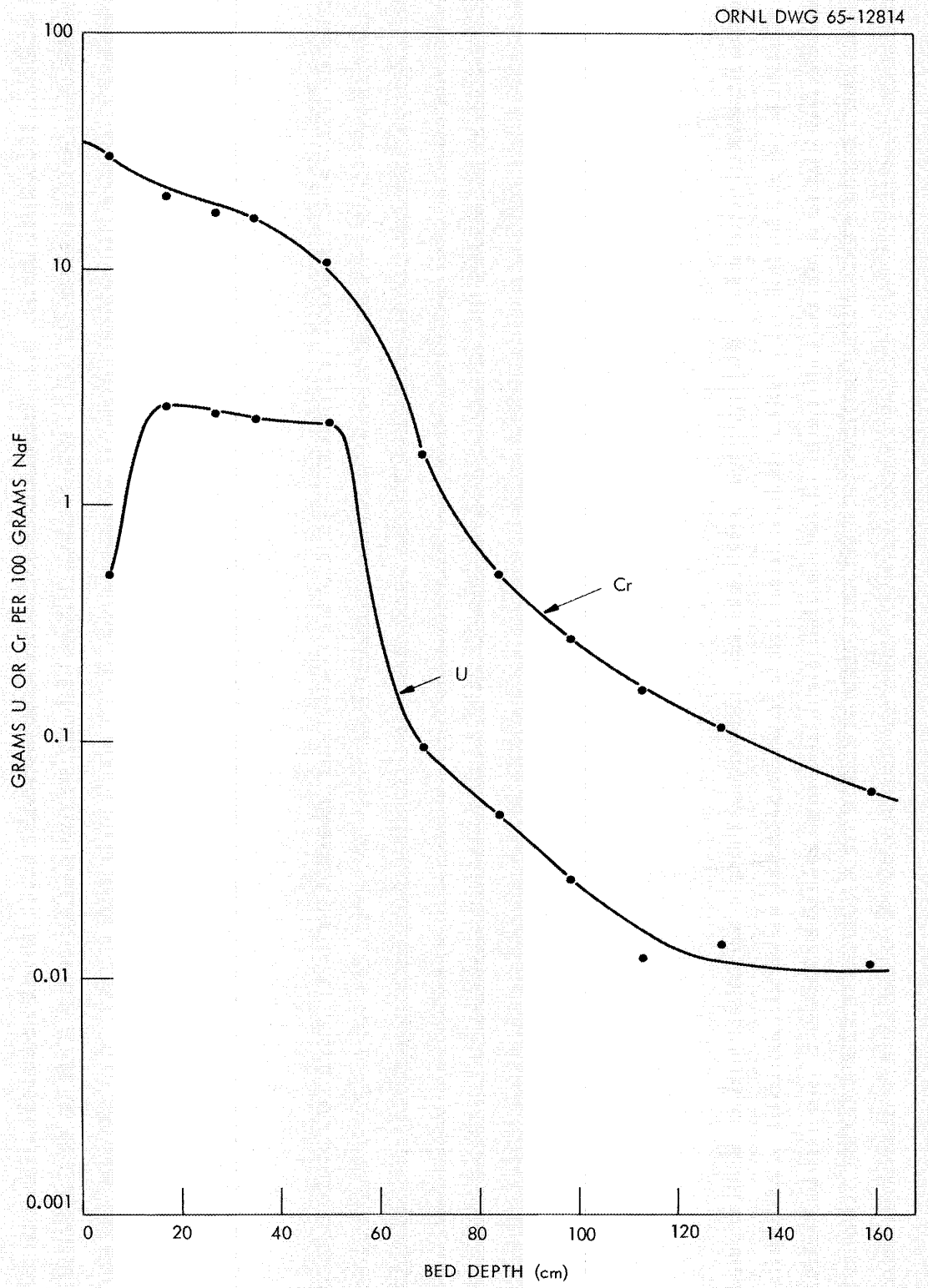

Fig. 2.2 Distribution of Chromium and Uranium on 400°C NaF Beds for Chromium Fluoride Removal Run CR-13.

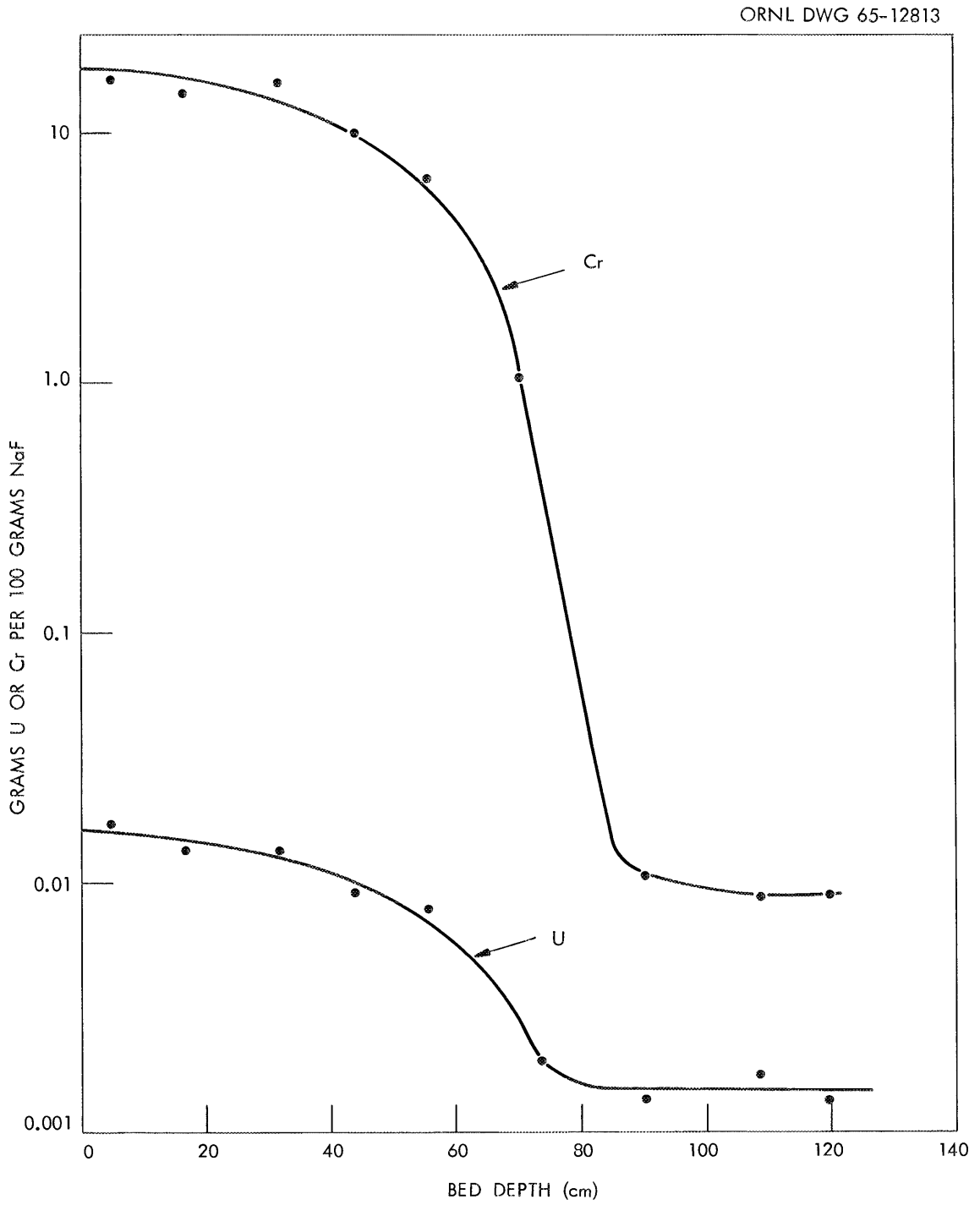

Fig. 2.3 Distribution of Chromium and Uranium on 400°C NaF Beds for Chromium Fluoride Removal Run CR-16.

The results of tests with the exhaust gas flowing downward through the bed are shown in Fig. 2.4. Acceptable pressure drops at design velocity could be obtained only with beds less than an inch deep.

Upward-flow tests with the bed unconstrained resulted in the bed's fluidizing at gas velocities above 2.5 ft/sec. Upflow tests with the bed constrained by a 10-mesh screen gave the same pressure drop as downflow.

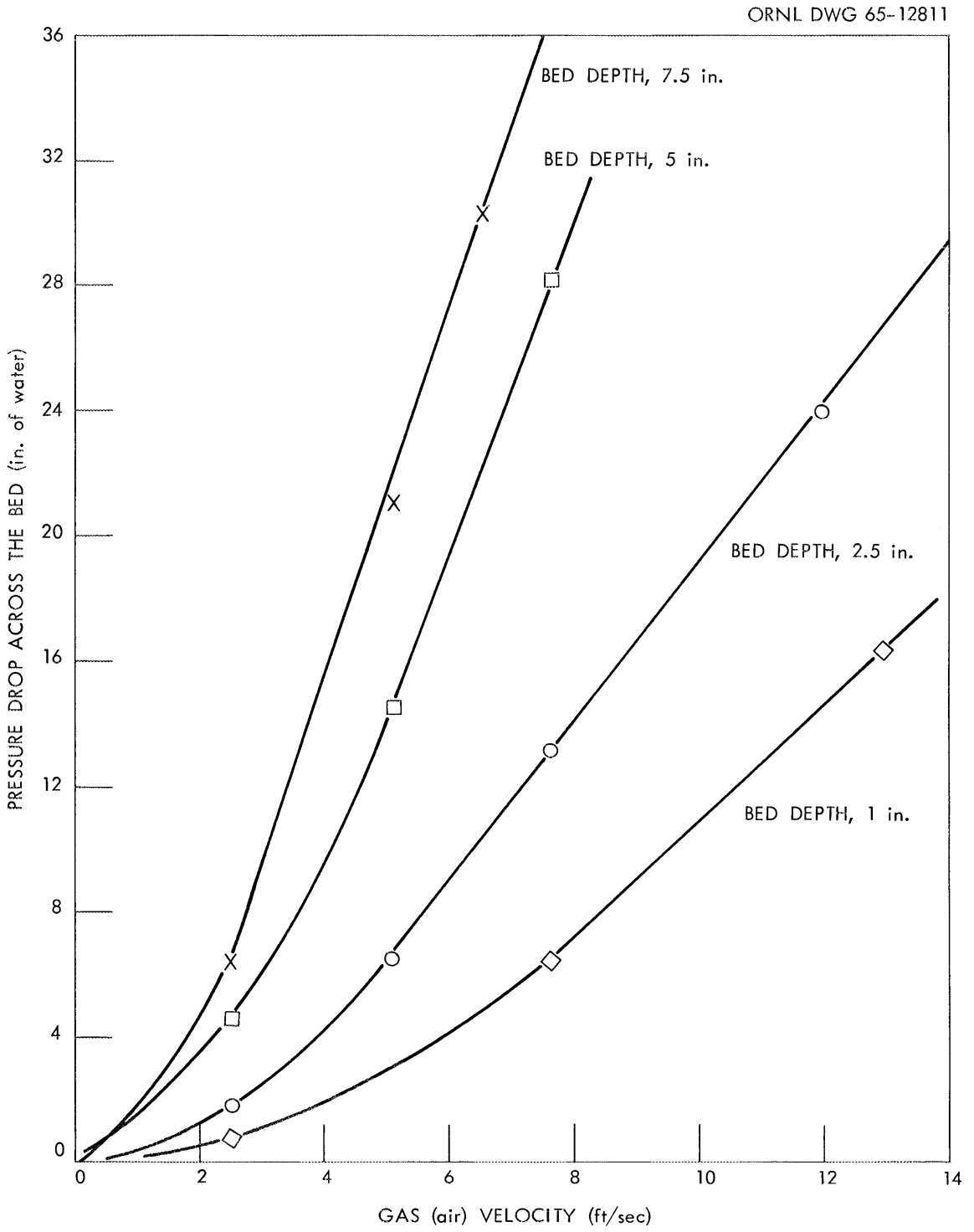

Fig. 2.4 Pressure Drop Across Soda-Lime Bed of Varying Thickness as a Function of Air Velocity.

3. PROCESSING OF POWER-REACTOR FUELS

3.1 Shear-Leach

B. C. Finney C. D. Watson

The development and evaluation program of a batch shear-leach process for recovering fissile and fertile core material from spent power-reactor fuel assemblies while leaving the sheared metallic cladding intact was continued. This program, which has been active since 1959, is being concluded; consequently, there are no new major areas of interest under investigation.

Unirradiated prototype assemblies (Consolidated Edison and Elk River) were successfully sheared, and on-site consultation was provided Nuclear Fuel Services, West Valley, New York, at their request.

To determine the feasibility of shearing complete Consolidated Edison Core B type fuel assemblies, several short sections (15 in. to 36 in. long) were sheared with all the fuel rods in place. In preparation for the tests, the 2 1/2-in.-diam cylinder supplying the holding force for the outer gag was replaced with a 4-in.-diam cylinder, and the internal hydraulic pressure was increased from about 900 psig to 1900 psig, giving greater than a fivefold increase in force on the gag. In one test, a 3-ft-long section consisting of four stainless-steel leaf-spring grid sections and 231 porcelain-filled stainless-steel tubes was sheared. At one point, the fuel jammed in the fixed blade and could not be fed properly. Tubes pulled out of the grid section, and the terminal pieces consisted of a piece of sheath 4 to 5 in. wide, and sections of tubing 2 1/2 to 6 in. long.

In general, it was shown that a dull shear blade and excessive clearance (20 mils) between the moving and fixed blade produced a poor product and incomplete cutting of the sheath (Fig. 3.1). Replacing the moving blade and decreasing the clearance to 3 to 5 mils gave an excellent product (Fig. 3.2) and a good terminal cut (Fig. 3.3). In contrast to the life of the moving blade, the fixed blade of the shear is not easily dulled and has not been replaced during several years of use. It was concluded

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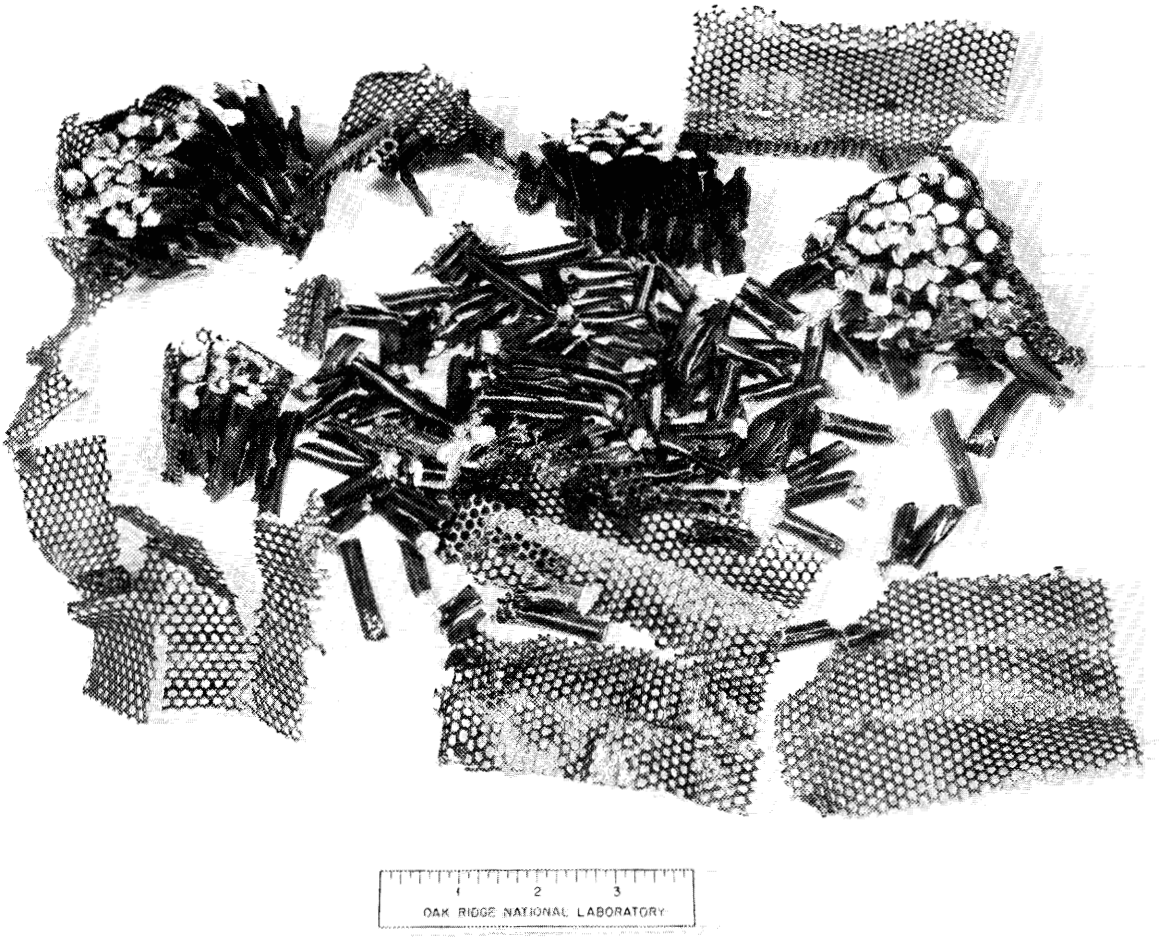

Fig. 3.1 Sheared Consolidated Edison Core B Fuel.

Note: Clearance between fixed and moving blade 20 mils.
Blade: Stepped shape dulled by previous use.

Fig. 3.2 Sheared Consolidated Edison Core B Fuel.

Note: Clearance between fixed and moving blade 3-5 mils.
Blade: Stepped shape in new condition.

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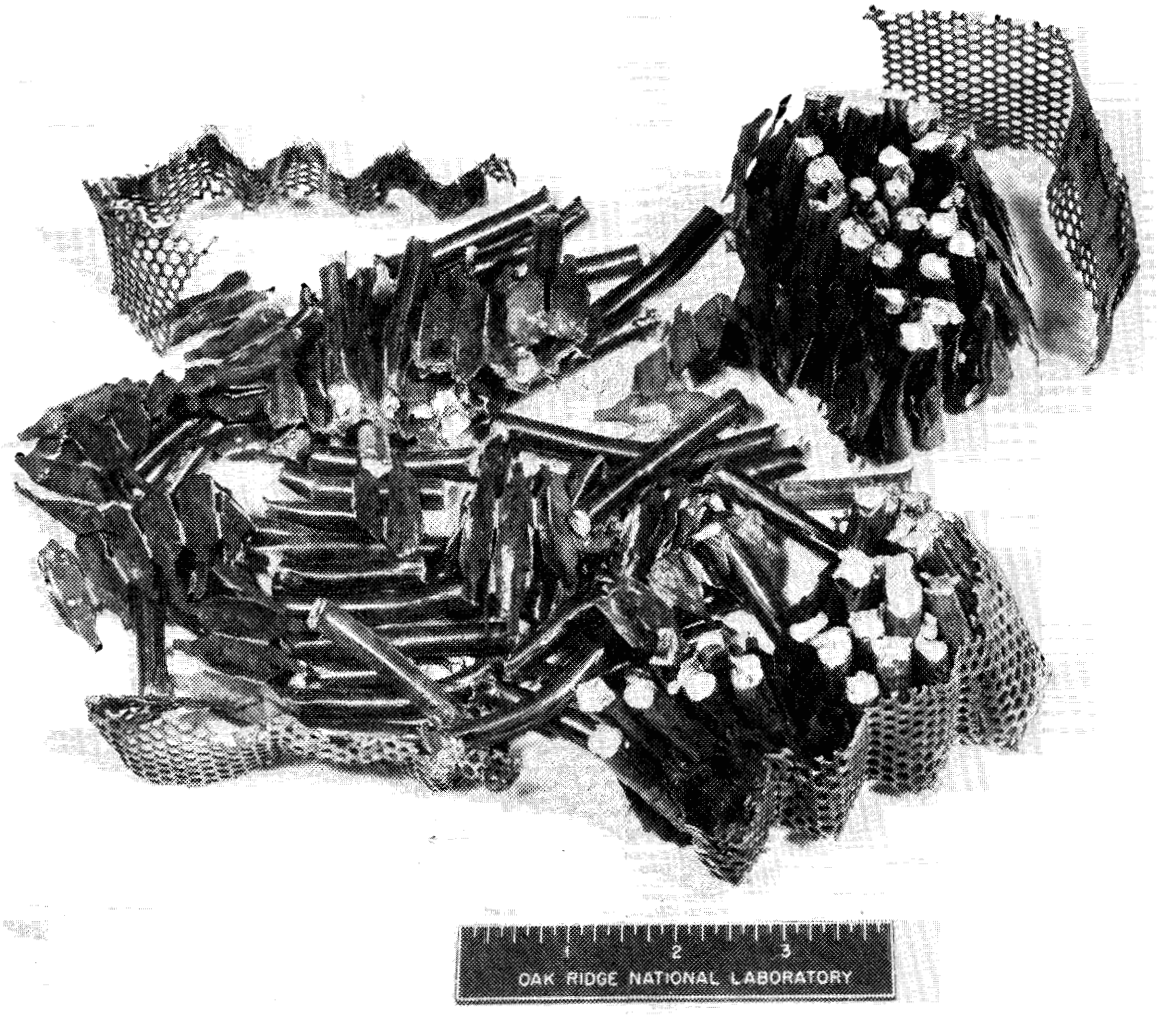

Fig. 3.3 Terminal Cut on Consolidated Edison Core B Fuel.

Note: Clearance between fixed and moving blade 3-5 mils.
Blade: Stepped shape in new condition.

that intact Consolidated Edison Core B fuel can be sheared satisfactorily if (a) the blade is reasonably sharp, (b) if blade clearance is 3 to 5 mils, and (c) if some "chunking" of fuel at the grids can be tolerated, along with some terminal pieces up to 3-in. long.

An intact porcelain-filled Elk River Core II fuel assembly was successfully sheared into 1 in. sections (Fig. 3.4). The assembly consisted of 25 stainless-steel tubes. They were 0.452 in. in outer diameter, had a 20-mil-thick wall, and were 62 in. long, filled with porcelain pellets. Two leaf-spring grid sections on 21-in. centers were used to separate the tubes. An ORNL-Mark I feed envelope (3.75 x 3.75 in.) was used to confine the assembly during shearing. After the last grid section was sheared, the tubes were loose (about 20-in. long), with a resultant 61.6 vol % voidage in the envelope and nearly the same in the outer gag. Even though there appeared to be excessive voidage, the loose tubes were confined by the outer gag and fuel-mechanism pusher head, and it was noted that the tubes were randomly clustered and that the inner gag was partially effective. All terminal pieces of tubing were 2-in. long except that two were 2 1/2-in., one was 3-in., and one was 4-in. long.

Consultation pertaining to the head end portion of the plant, primarily the Process Mechanical Cell, was made at the Nuclear Fuel Services, Inc., Power Reactor Fuel Reprocessing Plant located at West Valley, New York for the period June 13 to July 23, 1965. The plant is now essentially 99% complete and the final phase of construction involves mechanical check out of equipment to establish operability and remote maintenance procedures. NFS is now receiving spent nuclear fuel assemblies from Yankee Atomic and Dresden power reactors.

3.2 Fluidized-Bed Volatility-Process Development

R. W. Horton

The fluidized-bed volatility process is being developed as an extension of the fluoride-volatility process, which has been successfully demonstrated for the recovery of enriched uranium. The fluidized-bed process is expected to demonstrate the recovery of uranium and plutonium from low

PHOTO 81201

Fig. 3.4 Sheared Elk River Fuel. Long pieces in foreground are from terminal cut; pieces of grid-type spacers are on the right.

Note: Clearance between fixed and moving blade 3-5 mils.
Blade: Stepped in new condition.

enrichment uranium oxide fuel, with a minimum of preliminary processing and a very low volume of liquid waste. The process development originated at Argonne National Laboratory, is now being pursued at ANL, BNL, ORGDP, and ORNL, and will culminate in a pilot plant at ORNL for demonstration with irradiated fuel assemblies.

The Unit Operations section is involved in studies of process instrumentation, development of operating methods, and engineering evaluation of equipment needed for various process steps. To facilitate these studies, an engineering-scale process facility has been installed. Additional studies being made for basic data are done in auxiliary equipment as needed. The total scope of the program includes decladding, fluorination, and several phases of product separation. All the work is done with unirradiated UO_2 containing no plutonium.

3.2.1 Decladding of Stainless Steel Fuel Elements with $HF-O_2$ (R. S. Lowrie)

The fluidized-bed reactor test facility in Building 3503 has been completed and placed in operation. The reactor is made of 2-in. sched-40 nickel pipe and includes off-gas filters, heating jackets, cooling coils, and temperature and pressure-measuring devices. It can be supplied with metered flows of HF, F_2, N_2, and O_2. The off-gas system includes an HF condenser (operated at -50°F) with a liquid receiver and neutralizer, a reactor to eliminate F_2 by contacting it with SO_2 to form SO_2F_2, and the necessary scrubbers and filters to satsify safety and containment requirements.

This reactor is now being used to devise and test monitoring systems for controlling remote operations and to correlate operating variables and information read-out in order to provide a remote-operation program for fluidized-bed volatility-process flowsheets.

Three experiments, primarily to test equipment operation, have been made by using the $HF-O_2$ reference flowsheet for decladding stainless-steel-clad fuel elements. Sections of Consolidated Edison power-reactor fuel elements containing UO_2 pellets in a stainless steel tube (0.3 in. in outer diameter, 11-mil wall, type 304 stainless steel) were used in all tests. The fluidized bed was -60 mesh tabular T-61 commercial-grade Alcoa alumina.

A typical screen analysis is shown in Table 3.1. Experimental conditions for these tests are shown in Table 3.2.

Operation during the first run (FBVE-2) was uneventful. The pressure difference across the bed remained constant at about 1.0 psig. The pressure drop across the filter reached a maximum of 0.1 psig, well below the 0.8 psig at which blowback was scheduled. The fuel assembly rested on a bed of nickel shot about 4-in. from the bottom of the reactor, and the temperature of the bottom part of the fuel assembly probably did not exceed 300°C. A radiograph showed extensive attack at the top and along one edge of the fuel assembly. When the bottom of the reactor was removed, the unreacted fuel was found to be embedded in a cake of alumina that had a large blow hole in it next to the edge of the fuel assembly, which had reacted with the $HF-O_2$ mixture. Since the caked alumina was not mixed with any of the finely divided reaction products, it apparently had caked before the reaction started, perhaps during preheating.

The fuel assembly for the second run (FBVE-3) rested on a nickel grate 9 in. above the bottom of the reactor to ensure that the entire assembly was at the reactor temperature. No temperature or pressure transients occurred during the run (bed ΔP, 3.0 psig; filter ΔP, 0.12 psig, max.), and filter blowback was not required. A radiograph of the vessel indicated extensive attack on all the side rods of the fuel array, with some attack on the bottom of the center rod. Examination of the material again indicated only partial fluidization because the alumina below the grate contained no fine reaction products.

The third run (FBVE-4) was made with a 12-in.-long fuel-rod section cut from the corner of the Consolidated Edison fuel assembly (Fig. 3.5). After 2 hr of $HF-O_2$ exposure, the bed started to cake. Vibrating the reactor did not help, and the reactor plugged completely in the next hour. Pressure drop across the bed increased from 3.0 psig to more than 10 before shutdown. Radiographs showed extensive attack on the bottom and along one edge of the fuel assembly. Bed material that had caked around the unreacted sides of the fuel assembly contained no reaction products, indicating that it had caked early in the run. Apparently caking started

Table 3.1 Sieve Analysis of -60-mesh Alcoa T-61 Alumina

Size of Particles in Fractions U. S. Sieve Series		(μ)	Weight Fraction (%)
- 60	+ 70	-250, +210	0.6
- 70	+ 80	-210, +177	1.6
- 80	+100	-177, +149	2.2
-100	+120	-149, +125	4.9
-120	+140	-125, +105	6.9
-140	+170	-105, + 88	8.7
-170	+200	- 88, + 74	7.8
-200		- 74	67.2

Table 3.2 Experimental Conditions for HF-O_2 Decladding of Stainless Steel Fuel[a]

Run	FBVE-2	FBVE-3	FBVE-4
Fuel element length, in.	6	6	12
Contained UO_2, g	523	525	996
Stainless Steel, g	76	90	302
Total wt of fuel, g	599	615	1298
Alumina charge, kg	2	3.2	2.6
HF oxidation time, hr	4	4	3
Temperature, °C	550	550	600
Superficial gas velocity, fps	1.25	1.25	1.25

[a]Nine fuel-rod sections were arranged in a 3 x 3 square-pitch array for all runs.

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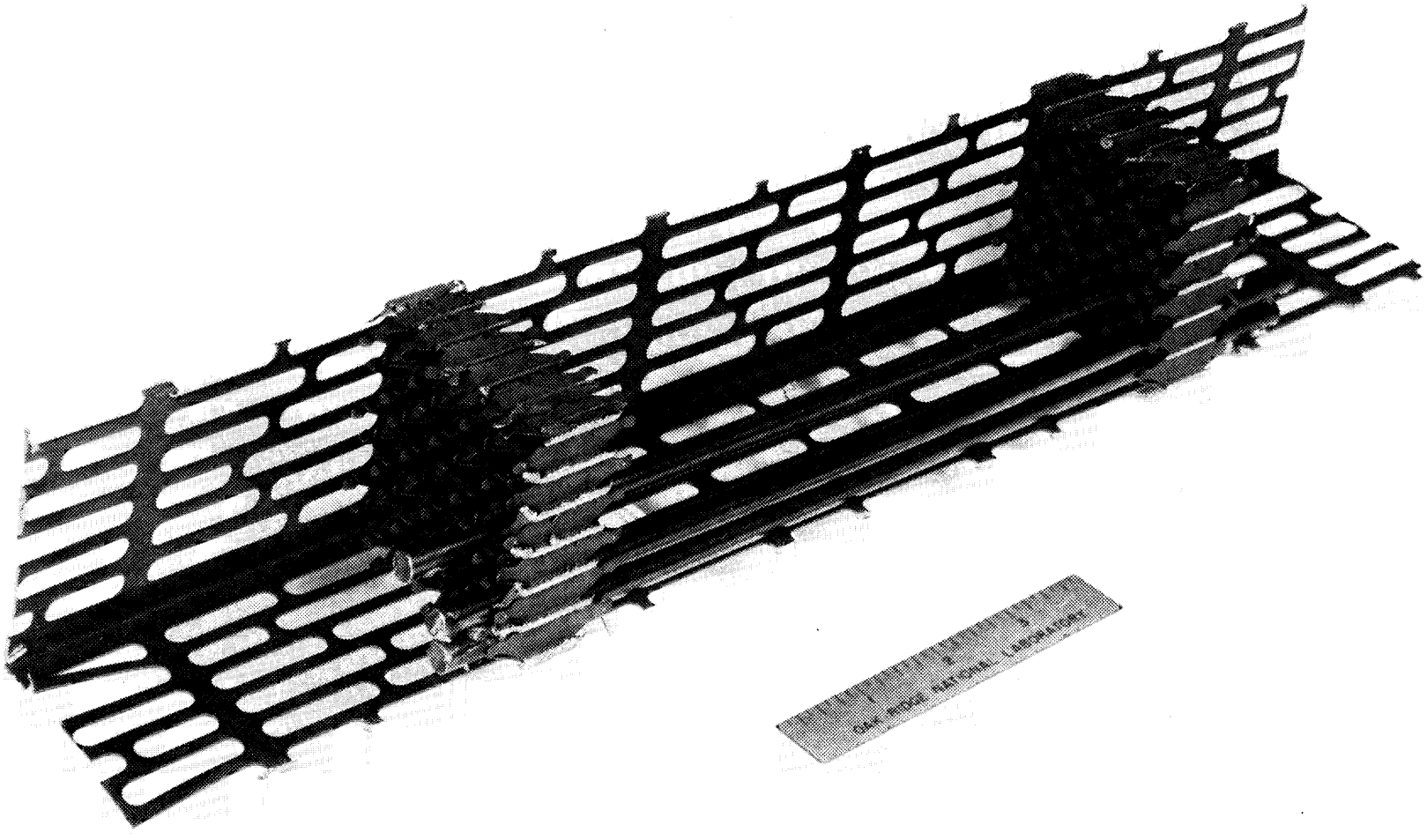

Fig. 3.5 Sectioned Consolidated Edison Element.

soon after the HF-O_2 was admitted, forcing the gas to pass through a blow hole, which in turn was plugged by reaction products as the run progressed. About 40% of the fuel assembly was oxidized.

Further tests are planned with both Alcoa tabular T-61 and Norton RR-grade alumina in an attempt to find out why the bed is caking.

The high efficiency of the sintered nickel filters (E porosity) in retaining uranium was indicated by analysis of the filter cake, which showed a uranium content of 4 to 15% in various samples, contrasted with a uranium concentration of less than 5 ppm in the HF condensed from the reactor off-gas stream.

A side stream from the decladding-process off-gas was sent through a gas-chromatograph column (20 ft of 1/2-in.-OD nickel tubing packed with KEL-F molding powder coated with 20 wt % No. 10 KEL-F oil) to measure HF and O_2 concentrations. No data was obtained because of mechanical failure of the automatic sample valve. The valve has been repaired, and the chromatograph will be evaluated as a monitoring device.

3.2.2 Vapor-Liquid Equilibria Studies of the UF_6-NbF_5 System (W. W. Pitt)

In fluoride-volatility processes, one proposed method for separating UF_6 from other volatile fluorides is fractional distillation. Knowledge of the vapor-liquid equilibria of mixtures of UF_6 and the other volatile fluorides is desirable to design the distillation equipment. The vapor-liquid equilibria of the UF_6 and NbF_5 system have been obtained.

Procedure and Results. - The isothermal dependence of total pressure on the liquid composition of UF_6-NbF_5 mixtures at 150°C was determined with the apparatus previously described.[5] A positive deviation from Raoult's law was found; but, assuming that Dalton's law of additive partial pressure held, the data were fit with the Scatchard-Hamer equation[6] (Fig. 3.6). With this equation, the activity coefficients of UF_6 and NbF_5 were determined for the entire liquid composition range (Fig. 3.7).

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[5] Unit Operations Section Monthly Progress Report, February 1964, ORNL-TM-836.

[6] G. Scatchard and W. J. Hamer, Jr., J. Amer. Chem. Soc. 57, 1805 (1935).

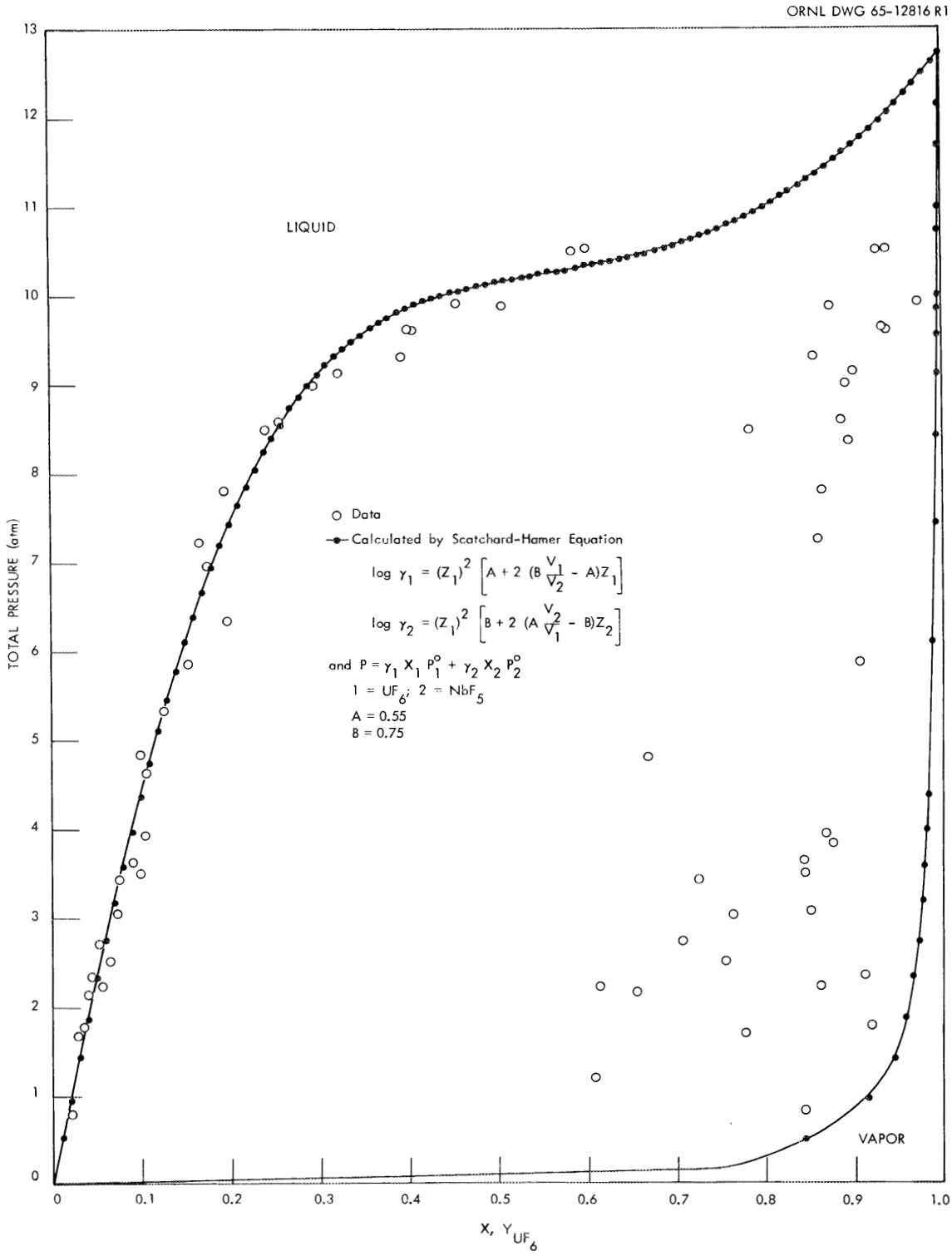

Fig. 3.6 Isothermal Pressure Composition Diagram for UF_6-NbF_5 System at 150°C.

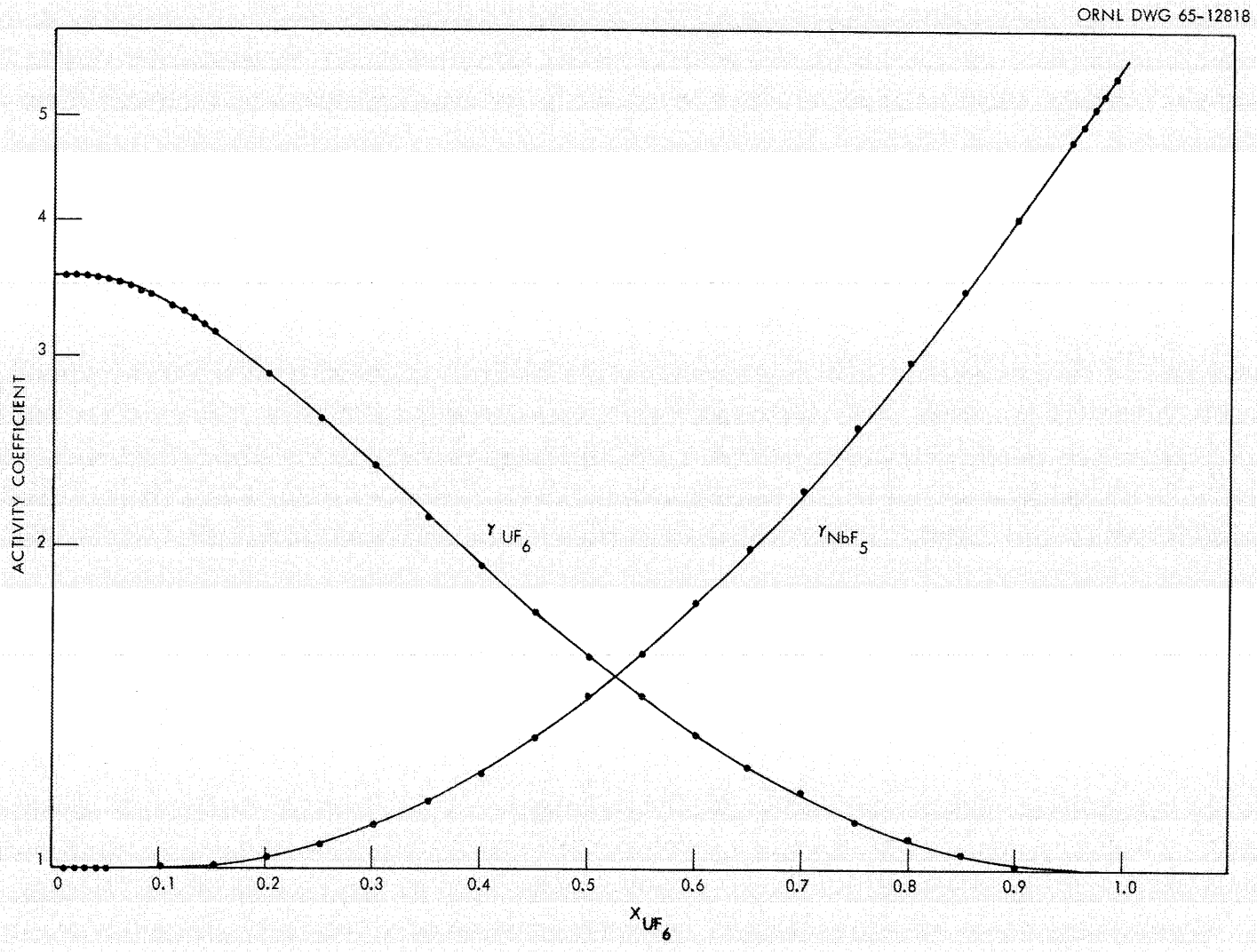

Fig. 3.7 Isothermal Dependence of the Activity Coefficients of UF_6 and NbF_5 on Liquid Composition at 150°C. Ideal vapor phase assumed.

Several methods for obtaining the composition of the vapor in equilibrium with the liquid under isothermal condition were attempted, but all were unsatisfactory. The vapor composition data, also shown on Fig. 3.6, were so scattered they were useless in determining activity coefficients, thus the indirect P-x data were used.

An Othmer-type equilibrium still[7] was built and then operated isobarically at 4500 mm Hg. The bubble and dew-point curves were determined for the entire liquid composition range (Fig. 3.8).

Method for Verifying Activity Coefficients. - Recognizing that the assumptions of an ideal vapor phase and the validity of Dalton's law are questionable at the pressures experienced during these studies (6 atm in the isobaric still and up to 12.8 atm in the isothermal still), we are trying to correct the calculated activity coefficients for the nonideality of the vapor phase. This will be done by an empirical method based on the theorem of corresponding states. To do this, the critical constants of UF_6 and NbF_5 are required. Those for UF_6 are available from the literature, but none for NbF_5 are available. These are being determined by measuring the pressure-temperature relationships (isochores) of several quantities of NbF_5 in a constant-volume bomb. By analyzing the shape of these P-vs-T curves, it is possible to estimate the critical pressure and critical temperature reasonably well, and to also obtain a fair estimate of critical density. This is possible, since a loading of material less than the critical loading (for a critical loading, the weight of material charged divided by the volume of the bomb equals critical density) gives an isochore which breaks away (downward) from the vapor-pressure curve at a temperature below the critical temperature, and loading greater than critical loading gives an isochore that breaks away (upward) from the vapor pressure curve at a temperature below the critical temperature. The isochore for a critical loading follows the vapor-pressure curve up to the critical point, at which point it becomes a straight line. This method should give the critical pressure within 5%, and the critical temperature within 1%.

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[7] D. F. Othmer and T. R. Morley, Ind. Eng. Chem. 38, 751 (1946).

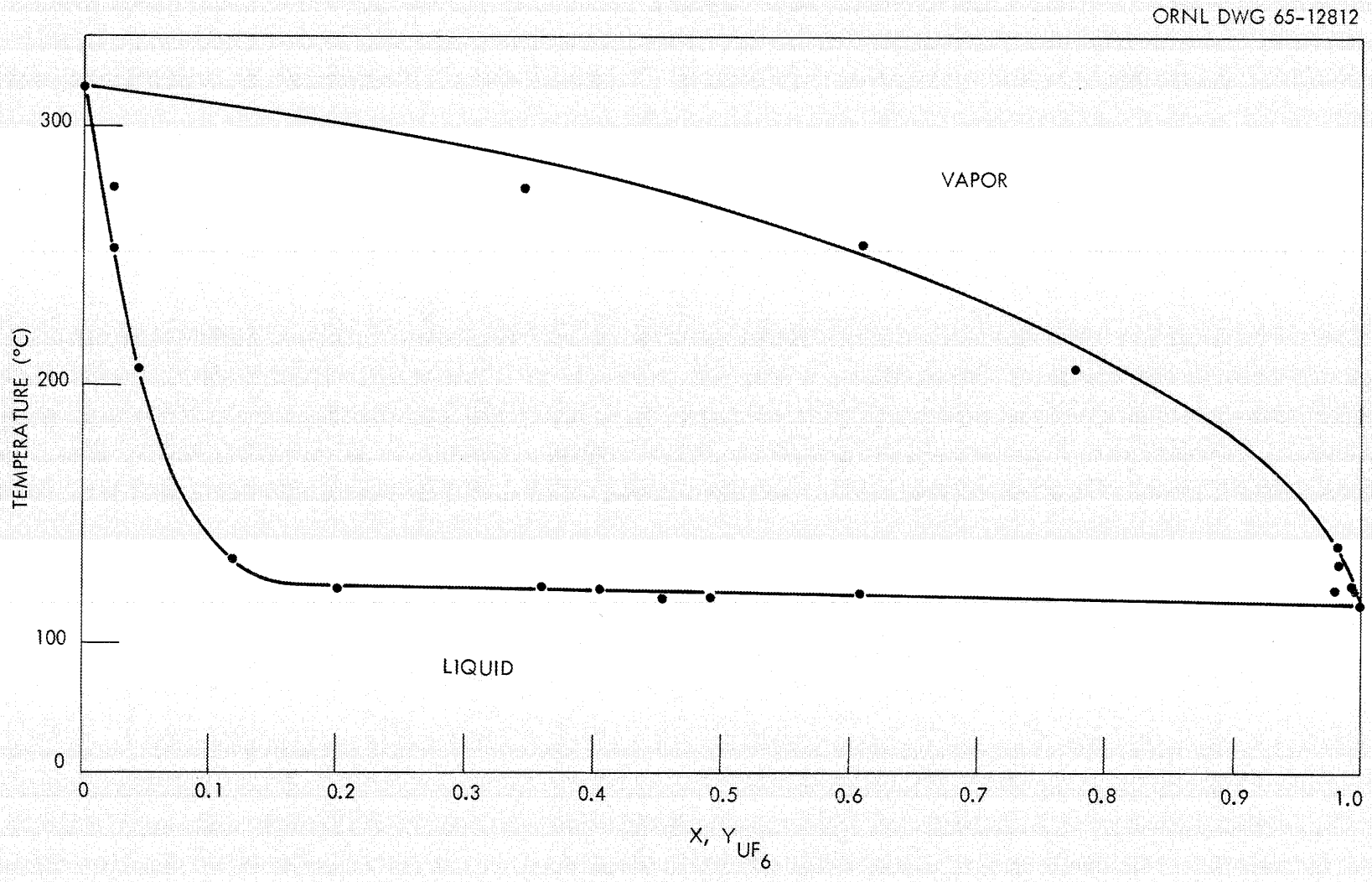

Fig. 3.8 Isobaric Temperature Composition Diagram for UF_6-NbF_5 System at 4500 mm Hg.

4. THORIUM FUEL-CYCLE STUDIES

P. A. Haas

We are developing the equipment and procedures necessary to adapt sol-gel processes to a completely remote operation, as required for the Thorium-Uranium Recycle Facility (TURF). A TURF cell-mockup installed in Building 4501 is used for testing remote maintenance procedures, manipulators, and services. Experimental testing of a vertical tube calciner (inner diameter, 3 in.) was completed, and a run is in progress to prepare product for other development studies.

We continued process-development studies for preparing thoria microspheres by dispersing a sol into 2-ethylhexanol, extracting the water to give gel microspheres, and drying and calcining to produce high-density oxide microspheres. The present emphasis is on studies of large-capacity sol dispersers and solvent recycle for the column-type system to permit high capacity and long-term operation. Pre-operational testing of a column system designed for an output of 10 kg of ThO_2 per day was completed; minor alterations were completed, and the system is ready for operation with thoria sol. A continuous, agitated-vessel type of microsphere-preparation system was tested and appears promising for preparing microspheres ranging in diameter from 1 to 100 μ.

4.1 Microsphere-Preparation Studies

S. D. Clinton P. A. Haas

The process development studies for dispersing oxide sols into droplets of uniform size, and then forming gelled microspheres in 2-ethylhexanol were continued in the tapered glass column of 2-in. minimum diameter. At present, the most promising method for obtaining uniformly dispersed sols on a large scale is to shear the sol stream emerging from an orifice by maintaining a velocity gradient in the organic solvent. The column was operated on a semicontinuous basis for 103 hr, with a sol feed rate of 285 cc/hr. The overall performance of the column and distillation systems was satisfactory. More downtime was accumulated than during the previous 100-hr run, mainly because thoria fines plugged the filter in the

recirculation system. During the last 25 hr of operation, the loss of surfactant from the 2-ethylhexanol caused some problems -- coalescence of droplets and sticking of partially gelled particles to the column walls.

4.1.1 Development of Sol Dispersers

The sol-dispersers now being evaluated can be grouped as follows: two-fluid nozzle, rotary disperser, and shear disperser (Fig. 4.1). The two-fluid nozzle continues to give excellent size distribution; however, at present, this nozzle is limited to sol flow rates of 1 to 5 cc/min. The different types of rotary dispersers[8] continue to show promise and have been operated for short times at sol flow rates up to 25 cc/min. The distillation system associated with the present column limits continuous operation to a sol flow rate of about 10 cc/min. This limitation will not exist in the column of 3 in. minimum diameter, now nearly ready for operation in Building 4508.

The shear disperser was made as a possible improvement over the rotary disperser. Both disperser types are designed to shear the sol stream emerging from an orifice by maintaining a velocity gradient in the 2-ethylhexanol at the orifice. With the shear disperser, the orifices remain stationary, and the organic is pumped by the orifices through a confined flow channel. A shear disperser with eight 16-mil-diam holes equally spaced around a 1/4-in.-OD stainless steel tube centered within a 3/8-in.-ID glass tube produced size distributions that were at least favorable, compared with the output of the rotary disperser (Table 4.1). At sol flow rates of 4.8 and 9.6 cc/min, the droplets appeared to cling to the stainless steel tube in a continuous stream for a distance of 0.5 to 1 in. immediately below the orifices. At a flow rate of 15 cc/min, the eight sol streams were jetting out into the center of the annular organic flow path. As the sol streams bent downward, the breakup appeared to be very similar to that obtained with the two-fluid nozzle. At a sol flow rate of 20 cc/min, and with an organic-drive-fluid flow rate of

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[8] Unit Operations Section Quarterly Progress Report, April-June 1965, ORNL-3868.

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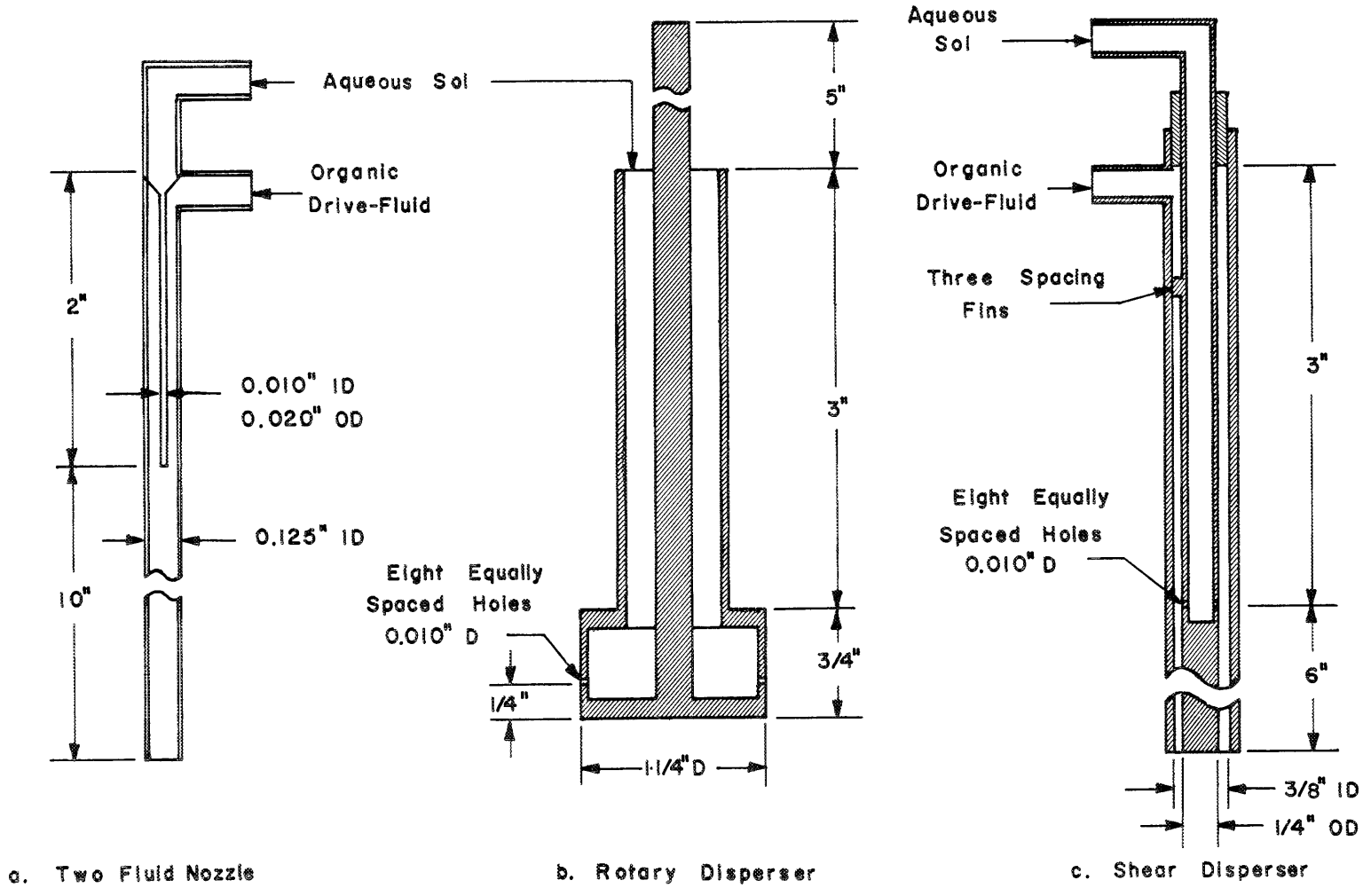

Fig. 4.1 Sol-Dispersion Devices.

Table 4.1 Size of Microspheres from a Shear-Nozzle Disperser

Sol Feed: A 3.4 M thoria sol which forms drop diameters of 2.2 times the diameters of the theoretically dense thoria product

Flow Rate of Organic Drive Fluid (cc/min)	Flow Rate of Sol (cc/min)	Sample Size (g)	d_{50} (μ)	$100 \frac{d_{80} - d_{20}}{d_{50}}$
220	4.8	390	310	15
220	9.6	380	380	16
220	~ 15	460	425	15
345	~ 20	245	465	17

of 220 cc/min, the sol streams impinged on the 3/8-in.-ID glass wall and appeared to break up into several different droplet sizes. By increasing the organic flow rate to 345 cc/min, the streams were bent downward before impinging on the wall.

Three different orifice diameters have been used with the shear disperser: 6, 10, and 16 mils; however, initial investigations indicated that this variable may have little effect on the droplet size produced. For a given sol flow rate, the orifice size determines the momentum of the sol stream, which may be an important factor in controlling dispersion. The shear disperser may be very inefficient at producing microspheres less than 250 μ in diameter. Early results show an increasing spread in the size distribution as the organic flow rate is increased for a fixed sol flow rate.

One problem with shear dispersers and rotary dispersers is the unpredictable wetting characteristic of the stainless steel. When the sol does wet the stainless steel in either the shear or rotary dispersers, the resulting distribution of droplet sizes can be very broad and highly unpredictable. This problem may possibly be eliminated by coating the stainless steel with either polyethylene or Teflon.

The operation of multiple two-fluid nozzles in parallel was tested as a possible scaleup procedure. A 5.4-hr run was made in the tapered glass column in which a definite separation of the sol droplets and gelled spheres was obtained. Two syringe pumps were used to feed two of the two-fluid nozzles at a total sol rate of 2.4 cc/min. The calcined product weighed 540 g, with 92 wt % in the size range 250 to 297 μ. The factor $100\ (d_{80} - d_{20})/d_{50}$, used previously for expressing the uniformity of microsphere size distributions, was less than 8. The flow conditions of this run were repeated in a 1.5-hr run in which a single syringe pump was used to feed both nozzles, and the calcined product (150 g) contained 86 wt % in the 250- to 297-μ size range. The possibility of operating two-fluid nozzles in parallel was tested further by using a single syringe pump to feed four two-fluid nozzles at a total sol flow rate of 4.8 cc/min. After running for about 7 hr, the run was interrupted due

to the formation of nonuniform droplets, which was caused by the variation in sol flow rate to each nozzle. At times, the sol flow to one or another of the four nozzles would stop completely. The uniformity decreased as shown by the value of 100 $(d_{80} - d_{20})/d_{50}$; less than 8 for the short test, 11 for normal long-term operation, and 19 for the four nozzles in parallel (Table 4.2).

A semicontinuous 100-hr run was made to demonstrate longer operation of several dispersers in addition to the primary objectives. The run was started with the four two-fluid nozzles in parallel. It was continued with a shear disperser that had eight 10-mil-diam holes at two different organic drive-fluid flow rates and two of the two-fluid nozzles with a sol syringe pump for each nozzle. About 17,700 g of thoria was calcined and screened, and 75 wt % of the product was in the size range 250 to 350 μ. An analysis of the product size distributions during the 100-hr run showed that these four tests resulted in products having mean diameters ranging from 270 to 310 μ.

4.1.2 Effects of Long-Term Operation

During the semicontinuous 100-hr run, 24.4 liters of a 3.0 M thoria sol (19.3 kg of thoria) was fed to the tapered column, and 19.3 liters of water was removed by the distillation system. In addition to the 17.7 kg of calcined microspheres, about 1 kg of gel spheres could be accounted for as samples. The remaining 0.6 kg probably collected on the replaceable Micro-Klean filters in the recirculating system. The particle crushing strengths of the calcined microspheres in the size range of 250 to 297 μ varied from 1050 to 1250 g, and from 1200 to 1650 g in the 297 to 354 μ range. As in the previous 100-hr run, there was no correlation of crushing strength with operating time of the 2-ethylhexanol. The BET surface area of the calcined spheres (250 to 354 μ) varied from 0.003 to 0.005 m^2/g.

Sol was fed to the column at the rate of 285 cc/hr for 88.3 hr during the 103 hr. The organic fluidizing flows were shut off ten times. Five of the shutdowns were caused by oxide fines plugging the Micro-Klean filter in the circulating system. None of these shutdowns occurred while

Table 4.2 Sizes of Microspheres Produced During Nominal 100-hr Run

Sol Feed: A 3.0 M thoria sol

Disperser	Total Sol Flow Rate (cc/min)	Sample Size (g)	Mean Diameter, d_{50} (μ)	$100 \frac{d_{80} - d_{20}}{d_{50}}$
2 two-fluid nozzles, separate pumps	4.8	4800	270	11
4 two-fluid nozzles, single pump	4.8	1450	280	19
Shear nozzle, 220 cc of organic per min	4.8	6975	310	13
Shear nozzle, 470 cc of organic per min	4.8	2700	280	25

using two of the two-fluid nozzles with separate sol supplies. Apparently, the other sol-dispersion techniques used during the run generated more fines than the normally used two-fluid nozzle. Three of the ten shutdowns were made in order to change the sol disperser. The remaining two shutdowns occurred toward the end of the run and were due to coalescence of the sol droplets and sticking of partially gelled particles to the column wall.

To prevent the sol droplets from coalescing, the original 2-ethylhexanol contained 0.5 vol % Ethomeen S/15 (surfactant by Armour). During the last 25 hr of the run, two additions of Ethomeen S/15 were made to the organic in order to stop particle clustering and sticking. Each of these additions increased the Ethomeen S/15 concentration in the organic by about a 0.1 vol %. The analyses on the amine and nitrate concentrations in the 2-ethylhexanol (Fig. 4.2) were similar to those obtained in the previous 100-hr run[9]. For the first 8 hr of the run, only the distillation system was in operation to see how the 150°C temperature in the still affected the organic. During this time with no sol feed to the column, the amine concentration decreased with a 37-hr half-life, and the concentration of nitrate in the organic remained low (less than 0.0005 M). After sol had been fed to the organic for about 10 hr, the amine began to disappear somewhat faster (half-life of about 19 hr). The nitrate concentration increased to a maximum of 0.0017 M at about the same time the disappearance rate of the amine increased. For 45 hr, the nitrate concentration decreased with an approximate half-life of 33 hr and then seemed to remain constant at 0.00065 M. Although the data are somewhat scattered, the additions of Ethomeen S/15 can be detected during the latter part of the run. The distillation system was operated for 16 hr after stopping the sol feed. During this time, the nitrate concentration decreased by a factor of 1.3, and the amine decreased by a factor of 2.7. According to the analytical results of the two 100-hr runs, the Ethomeen S/15 seems to be a weaker base than Amine O, as shown by the decreased concentration of nitrate in the organic.[9] The results from the two 100-hr

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[9] Unit Operations Section Quarterly Progress Report, April-June 1965, ORNL-3868.

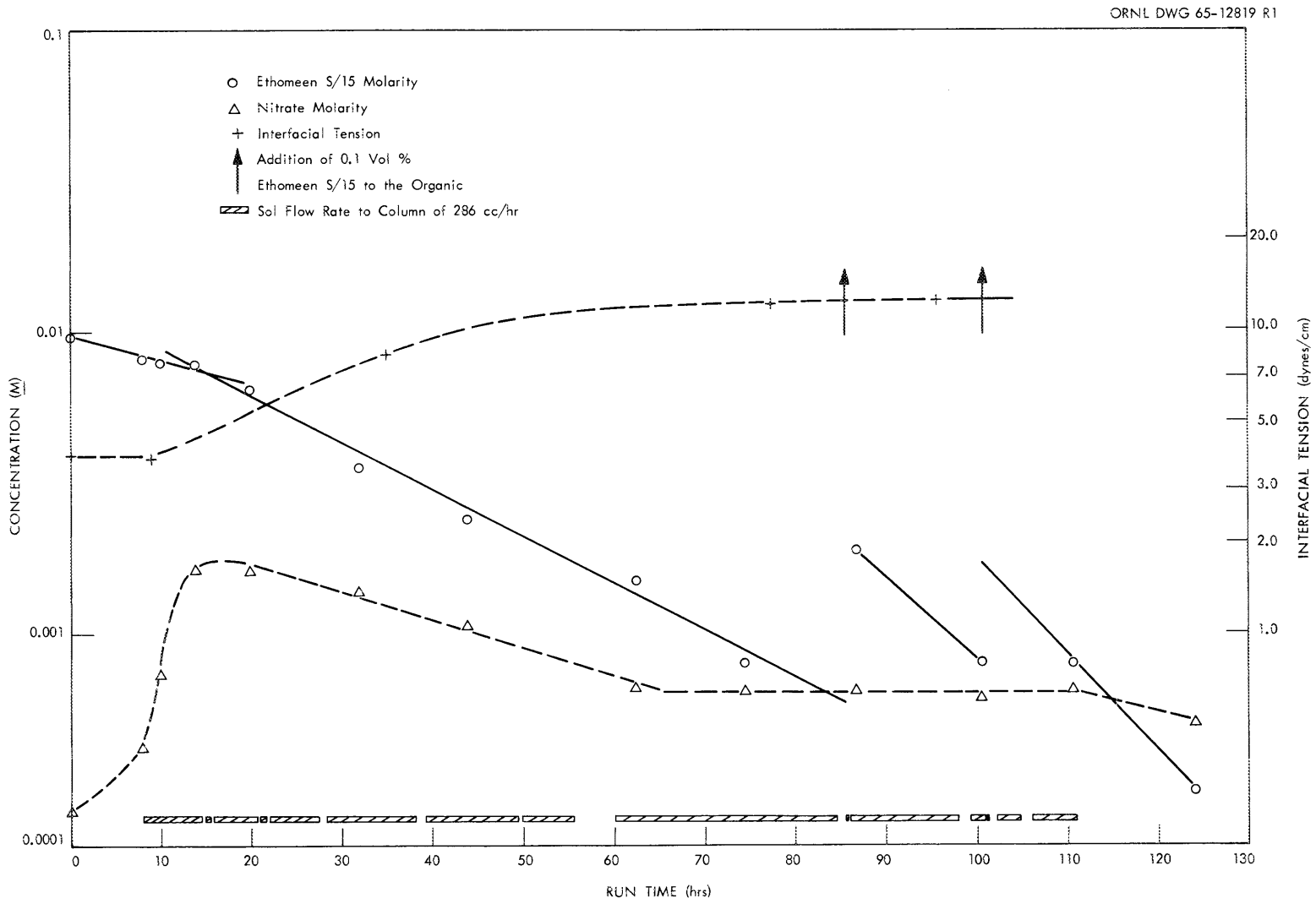

Fig. 4.2 Concentration of Nitrate and Ethomeen S/15 in 2-Ethylhexanol Interfacial Tension vs Run Time.

runs are still not conclusive; however, the amount of nitrate extracted from the aqueous sol into the organic solvent does seem to depend on the concentration and type of amine being used as a surfactant. The disappearance of both the amine and nitrate from the organic appears to be a first-order adsorption or decomposition reaction. The interfacial tension between the organic and aqueous sol increased from 4 to 12 dynes/cm and remained constant during the last half of the run (Fig. 4.2).

Future runs will be designed to continue the study of long-term recycle effects on different surfactants in 2-ethylhexanol and to test different sol-dispersion methods at increased sol flow rates. The results from the last 100-hr run suggest the feasibility of continuously adding Ethomeen S/15 to the organic solvent to maintain its concentration in the range 0.001 to 0.002 M.

4.1.3 Preparation of Microspheres in a Column Without Fluidization

The extraction of water from thoria sol drops (to cause gelation) was tested by allowing the droplets to fall through 6 ft of 2-ethylhexanol in the column without fluidization. The settling velocity is proportional to the square of the drop diameter, and the surface/volume ratio for a sphere is proportional to the diameter. Therefore, the column length required to give adequate drying should be approximately proportional to the cube of the diameter of the drop. An experiment was performed with a 6-ft column using 3 M Th sol, controlling the sol droplet size by the operating conditions of a two-fluid nozzle. The results were as follows:

Observed Gel Sphere Diameter (μ)	Amount of Drying
100-200	Very well dried
170-190	Adequately dried
250	More than 50% of adequately dried spheres, but large fraction of useless product from inadequate drying

This type of column operation does not require careful flow control nor uniformity of droplet size, which are necessary for fluidization in the column. Also, the coalescence, deposition, and clustering problems are much less severe. During operation with fluidization, the deposition is most severe where the sol drops are fluidized near the top of the taper, and the clustering is most severe where the gel spheres are fluidized near the minimum diameter of the column. Neither type of accumulation occurs for a free-fall type of operation.

4.1.4 Preparation of Microspheres by Dispersion and Suspension Through Mechanical Agitation

Sol-gel microspheres of relatively nonuniform diameter and less than 80-μ mean diameter (calcined product) can be easily and efficiently prepared by using mechanical agitation to disperse and suspend the droplets in 2-ethylhexanol during extraction of water. This method of preparing microspheres has two important advantages: Suspension by agitation requires neither the careful control of fluidization conditions nor the uniformity of sol drop size, which are necessary for fluidization in the column; also, scaleup of a process based on mechanical agitation is much simpler than scaleup of the column and the dispersers which give uniform sol drops. The original interest in microspheres was principally in products of uniform diameter (mean diameters of 100 to 500 μ). Therefore, most of our development studies were with fluidization of the sol drops in tapered columns during extraction of water. However, the preparation of microspheres in mixer-settler systems was demonstrated, and the results illustrate the advantages and limitations of such a system.

Continuous operation with dispersion and suspension by agitation was demonstrated by using two mixers and two settlers (Fig. 4.3). The mixers were 4-liter beakers altered to provide a low inlet for solvent, a high outlet, and four 3/4-in.-deep indentations as baffles. Dry 2-ethylhexanol was fed in tangent to the wall at the bottom of the first mixer. Sol was fed into the first mixer above the agitator, which also served as a disperser. The agitator consisted simply of four 1/2 x 1/2-in. paddles. The 2-ethylhexanol containing suspended sol drops and gel spheres overflowed near the top, through the second mixer by a similar path, and to the

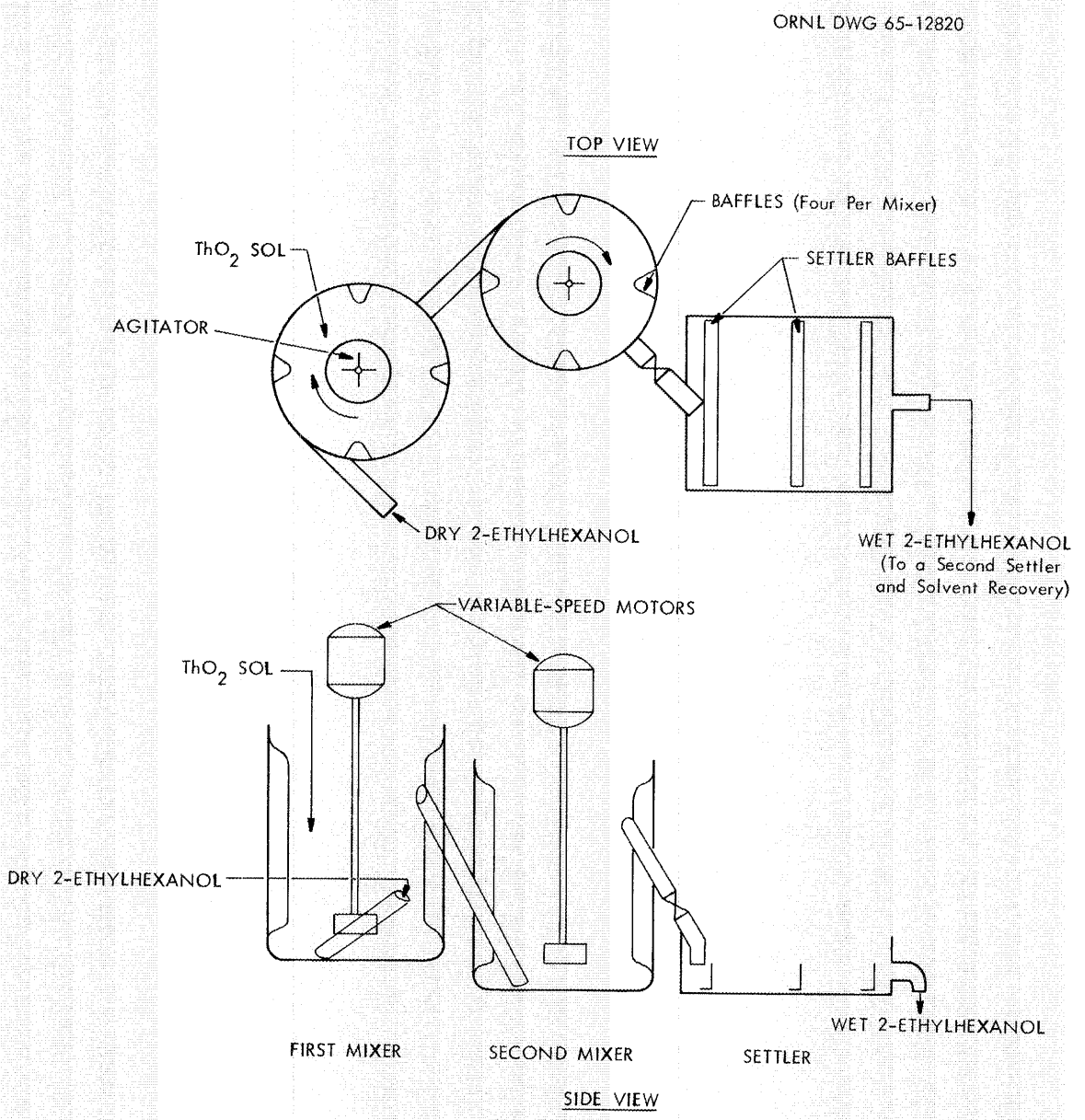

Fig. 4.3 Mixer-Settler System for Preparing Microspheres.

settler. Over 99 wt % of the gel spheres settled out in the first settler. The flow ratio of 2-ethylhexanol to sol was about 100. The variables were the speed of the first agitator and the feed rate of the sol. At the end of a period of continuous operation, the 2-ethylhexanol was decanted from the first settler. The gelled microspheres were dried by placing the settler pan in an oven at about 120°C.

The size distributions of the products show that product diameters increase as the mixer speed is decreased or as the flow rates are increased (Fig. 4.4 and Table 4.3). The purpose of these studies was to determine the practicality of making microspheres larger than 44 μ in diameter. Previous studies had shown that vigorous agitation could be used to make products that were nearly all -325 mesh (less than 44 μ in diameter). The product of 90-μ mean diameter at 575 rpm appeared to be the largest spheres that could be made with 2-ethylhexanol in these mixers. At lower agitator speeds, suspension of the drops was incomplete, and large masses of coalesced or clustered drops soon covered the bottom of the mixer. When a similar mixer was operated with CCl_4-isopropyl alcohol as the gelling agent, mean diameters as large as 200 μ were possible.[10] This product was dispersed and suspended at an agitator speed of 330 rpm; the high density of CCl_4 permits the suspension of larger drops by less agitation.

The uniformity of the diameters is poor compared with that for microspheres prepared in the tapered-column. This is shown by values of $(d_{80} - d_{20})/d_{50}$ of 50 to 120% (Table 4.3), compared with less than 20% for normal operation of the column system. When the suspension is less uniform (less agitation for a given system), the product from a mixer-settler system is less uniform. A more uniform product might be possible if the mixer were designed to only suspend drops fed to it from a separate disperser.

The design of a practical mixer-settler system for preparing microspheres should allow for the following factors:

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[10] Unit Operations Section Monthly Progress Report, December 1964, ORNL-TM-1091.

ORNL DWG 65-12815 R1

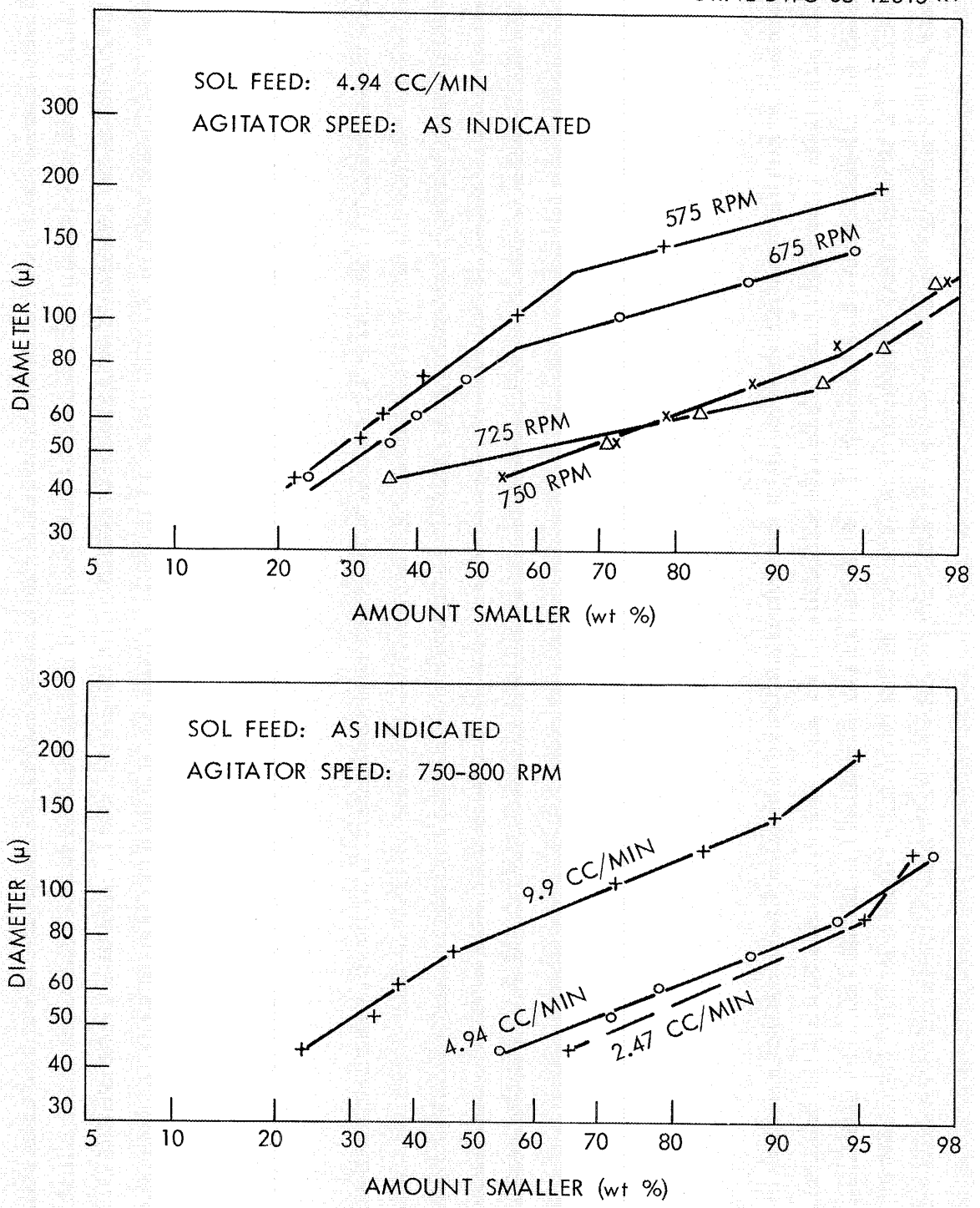

Fig. 4.4 Size Distribution for Calcined Microspheres from Mixer-Settler System.

Table 4.3 Size Distributions for Calcined Microspheres from Mixer-Settler Tests

2-ethylhexanol/sol volume ratio: 100
Sol concentration: 3.0 M ThO_2

Feed Rate of Sol (cc/min)	Agitator Speed (rpm)	Product Size Distributions	
		$d_{50}(\mu)$	$\frac{d_{80} - d_{20}}{d_{50}}(100)$
2.0	625	64	70
2.47	680	60	70
	750	36	80
4.94	575	90	120
	675	75	100
	725	48	50
	750	42	80
9.9	800	78	100

1. The holdup time in each stage may be short, but two or more stages are necessary to prevent excessive discharge of undried spheres. Extra stages are much more effective than increasing the volume of a single stage. The holdup for the experimental tests were about 3 liters per stage, or 3 to 13 min average holdup time per mixer.

2. Sharp corners must be avoided because sol drops accumulate and coalesce in such corners. For the tests discussed here, the mixers were baffled beakers. The baffles were shaped into the beaker walls by heating until they softened and then indenting the wall from the outside. Baffles that did not extend to the bottom of the beaker required less agitation to maintain suspension.

3. In deciding on the degree of agitation required, both the dispersion and suspension operations should be considered. The preparation of fine products requires intense localized agitation for dispersion, but the suspension of the fines is relatively easy. To prepare larger-diameter products, the agitation must provide effective suspension throughout the whole mixer while avoiding any intense local agitation, which would further disperse drops of the preferred size.

Tests of mixer-settler system for preparing microspheres indicate the following:

1. The maximum practical mean product diameter is about 80 μ when 2-ethylhexanol is the organic liquid. Sol drops that would give larger microspheres are dispersed into smaller drops by the agitation necessary to suspend them. Products with a larger mean diameter could be prepared in organic liquids of higher density and/or viscosity.

2. The mixer-settler system would be a simple and efficient system if a relatively nonuniform product is satisfactory and if the limit on the maximum mean diameter is acceptable.

3. The uniformity of the product improves and the mixer-settler system becomes more attractive for smaller mean diameters. A large-scale system for a product less than 50 μ in diameter could be easily designed.

4.2 VERTICAL TUBE CALCINING FURNACE FOR THE TURF

C. C. Haws, Jr.

Testing of a prototype vertical furnace[11] for continuous calcination of ThO_2-UO_3 gel in the TURF nears successful completion. In the tests, reliable mechanical operation was repeatedly obtained; procedures and equipment necessary to prevent and, or, detect bridging of the product in the tube were developed; expected product quality standards were met; and instrumentation adequate for remote operation was proved. A sustained operating test is now in progress and will represent the final acceptance test of this equipment for remote operation.

Two problems were expected to prove troublesome in the vertical tube calciner. The first of these was bridging of the product in the tube during calcination, while the second was the breaking of the slender, (3-in. ID x 54-in. long) alumina bore tube by thermal and mechanical shock. Bridging was encountered. Tube breakage also has given trouble but has occurred usually as a result of attempts at breaking bridged product loose from the tube. Thus the solution of the tube-breakage problem lay primarily in preventing bridging and secondarily in developing procedures for breaking bridges.

Bridging was found to be time-temperature-motion dependent. Thus, when the furnace bed was kept moving down the tube at the design rate and temperature (1150 to 1200°C), no bridge formed. The bed could be stopped for 4 hr at design temperature without forming a bridge; but, stopping for 12 hr caused bridging. However, the furnace could be "parked" at 900 to 1000°C indefinitely without bed movement, and no bridging occurred. Operation of the furnace within these limitations is practical and reliable.

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[11] Unit Operations Section Monthly Progress Report, October 1964, ORNL-TM-1069.

Tube breakage was found to be the result of thermal shock. When a bridge is formed, material below the bridge continues to pass down the tube, leaving a void below the bridge. This void is in the hottest region of the furnace. When the bridge is broken, cold material above the bridge falls into the hot portion of the tube and shatters it. A void estimated at about 2 in. deep was sufficient to cause breakage of a tube. Allowing the furnace to cool to room temperature before attempting to break a bridge solved this problem successfully.

More than 150 hr of service was obtained from the last bore tube, and this was under conditions far more severe than those expected in the final application. This tube was thermally cycled repeatedly and was used for the entire series of tests designed to find ways of preventing or breaking bridges. The tubes should have indefinite life in a routine operation; however, means of replacing the tube (or furnace) upon breakage must be provided in the facility.

The quality of the product from the vertical tube calciner is equal to that obtained in static-bed calciners. Particle densities (by mercury pycometry at 500 psig) of 10.0 were routinely obtained, and bulk densities in vibrated Kilorod-type metal tubes ranged from 88 to 91% of theoretical. Gas release values and O/U ratios were consistently less than 0.05 cc/g and 2.2, respectively. Some particle breakup does occur during calcination, but not enough to be of concern. Bed movement is uniform, with no propensity for "layering" of either fine or coarse particles.

The most important control instrument developed during the tests was a solids-level detection system for the upper (feed) end of the calciner (Fig. 4.5). The bed-level probe was operated in a reciprocating manner, with the probe lowered to rest on the bed for 2 min and then raised for 2 min. This system was superior to simply letting the probe rest on the bed during the entire feed-discharge period. The position of the probe was recorded on a strip chart, and the feeder automatically turned on when the bed level reached a given point. In addition, the signal to the recorder was fed to a ramp-function generator (RFG) - alarm circuit. This RFG reliably detects loss of bed movement in less than 4 min. Stoppage of

ORNL DWG 65-6187

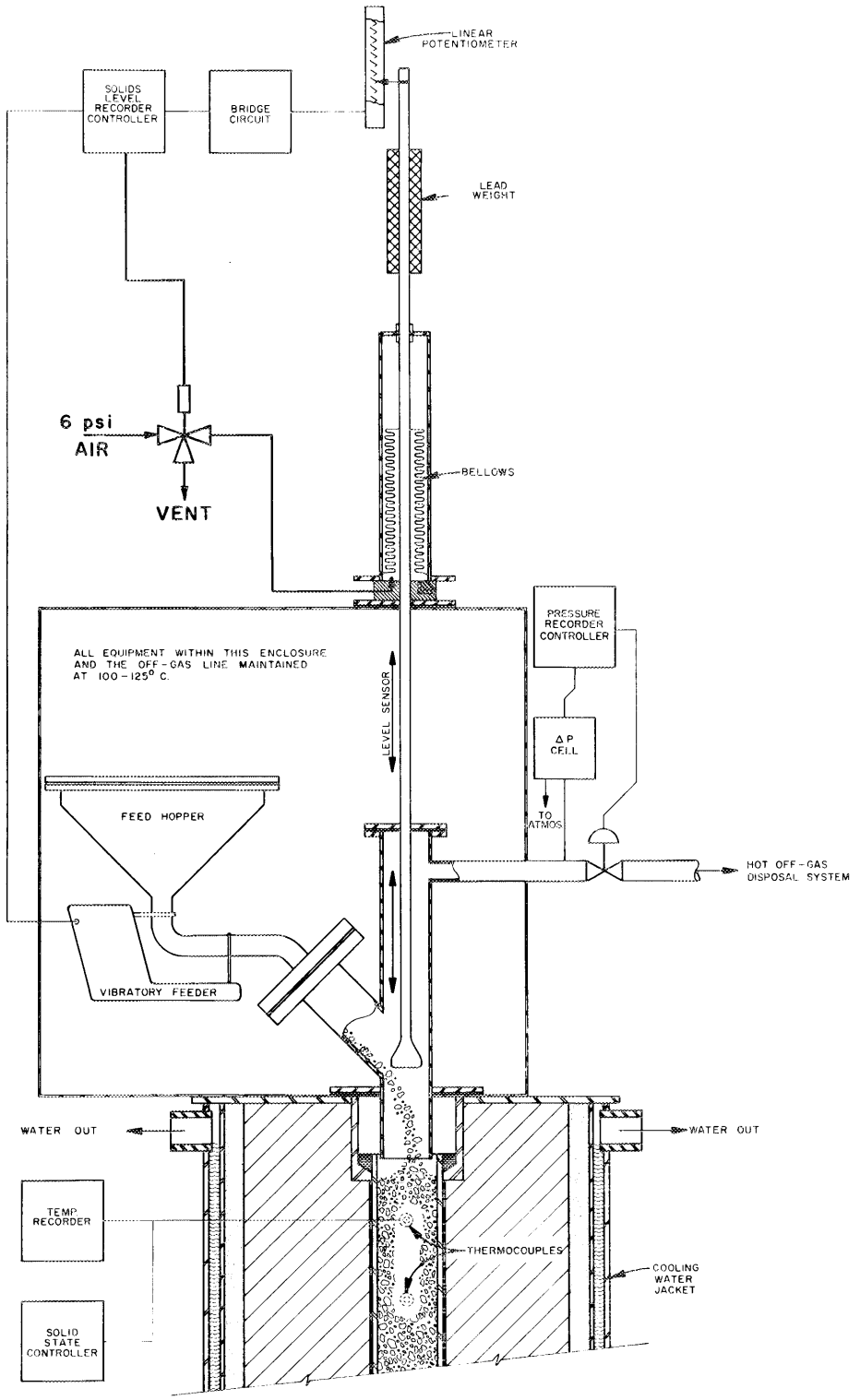

Fig. 4.5 Equipment Used in Feeding Gel Solids to Calciner.

bed movement at the feed end indicates bridging. As 1 to 2 hr is sufficient notice of bridging, this device easily meets our requirements for an indicator-alarm circuit for remote operations.

In the current sustained operating test, the performance of the entire apparatus has been good. In the first 72-hr operating period, 102 kg of product was processed. Succeeding operating periods sufficient in number to prepare about 250 kg of calcined solids for experimental use by the Metals and Ceramics Division are planned. The preparation of this quantity of product and attendant work on the furnace will conclude the testing of this prototype calciner.

5. SOL-GEL PROCESS DEVELOPMENT

P. A. Haas

A variety of sol-gel processes are being developed for preparing microspheres and other particle shapes for nuclear fuels. The original development of the sol-gel process is being continued as the thorium fuel-cycle program (see Section 4). Engineering studies of sol-gel processes for preparing microspheres of urania during the last year have just been declassified and will be summarized here. Procedures for preparing sols and forming them into gels have been developed in the laboratory for plutonium, zirconium, other actinides, and some rare earths. Some of these other sol-gel processes will require engineering development.

5.1 Preparation of Urania Microspheres

S. D. Clinton P. A. Haas

The process for converting aqueous sols into gelled oxide microspheres as previously developed for thoria sols (see Section 4) was used for urania sols without changes except where the previously developed procedures were inadequate. In this process, the sol is dispersed into drops of controlled diameters in a drying solvent, and the drops are supported in the solvent while water is extracted, causing the sol to gel. Nearly all the urania microspheres were prepared by dispersing the urania with a two-fluid nozzle and fluidizing the drops in the column during extraction of water. The preparation of urania microspheres up to June, 1965 has been reported in detail elsewhere[12,13].

During the quarter, eight 300-g batches of fully enriched urania sols were converted into gel microspheres with the 2-in.-minimum-diameter tapered column. The desired size ranges of the calcined microspheres were 40 to

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[12] R. G. Wymer, ORNL-TM-1110 (April 9, 1965) (Confidential).

[13] J. P. McBride (compiler), Preparation of UO_2 Microspheres by a Sol-Gel Technique, ORNL-3874, (in preparation).

80 μ, 125 to 177 μ, and 177 to 250 μ. With a two-fluid nozzle, yields up to 90% were obtained in each of these size ranges. About a kilogram of natural urania sols were converted to gel microspheres during the quarter.

In our column system, a surfactant must be dissolved in the organic solvent to prevent coalescence of the sol drops, sticking of the sol drops on the column walls, or clustering of partially dried drops. Surfactants also lower the interfacial tension between the sol and the solvent; the interfacial tension must be high enough to keep the drops spherical. A surfactant concentration of 0.1 to 0.5 vol % in the organic liquid seems to be sufficient. A surface-active agent is necessary to make the process work; however, the determination of the type of surfactant required for a new sol and a specified particle size range is largely empirical. With 0.5 vol % Amine O in 2-ethylhexanol, the nitrate-stabilized urania sols behaved very similarly to the regular thoria sols. With Ethomeen S/15 as the surfactant, the urania droplets dried partially and then stuck to each other. These clustered particles interfered with continuous operation of the column. Span 80 was less satisfactory because of the distortion of large drops into nonspherical ones, and because of its decomposition in the distillation system. No single surfactant was found for formate-stabilized urania sols, but a mixture, 0.5 volume % Amine O and 0.5 volume % Span 80, was satisfactory. Span 80 alone resulted in nonspherical shapes, with "cherry-pitting" or cherry-shaped particles as the most common effect. The Amine O by itself permitted excessive clustering of particles. These clusters dropped out of the column too quickly and decreased the yields of good microspheres. Only two batches of urania-20% zirconia sol were formed in the column system. The first of these sols was a coprecipitated nitrate-stabilized sol, and the second was a coprecipitated formate-stabilized sol. These sols were formed into microspheres without difficulty, using the mixtures 0.5 vol % Amine O and 0.5 vol % Span 80. The behavior in the column was as good as that of the better urania sols.

The conditions in the column oxidize the nitrate-stabilized urania sols from U^{4+} to UO_2^{2+}. With no argon atmosphere over the organic or no

resin column to remove nitrate in the solvent-recovery system, the U^{4+} content of the gelled spheres (as removed from the column) from a nitrate-stabilized urania sol was 62%. With an argon atmosphere and a resin column in the solvent recovery system, the U^{4+} content of the gelled spheres from a similar sol was 73%. For a formate-stabilized urania sol, the U^{4+} content of the gelled spheres was 82% with the argon atmosphere and a resin column.

Very little information was obtained on the long-term recycle of the organic since most of the urania sol batches were formed in new organic. There is evidence that the Span 80 loses its effectiveness upon recycling through the distillation system.

5.2 Continuous Reduction of Uranyl Ions to Uranous in the Aqueous Nitrate System

J. J. Perona

The reduction of uranium is of interest as a first step in the production of UO_2 sol[14]. A tubular flow reactor was built, tested, and proved capable of continuously producing 1 liter of 0.5 M $U(NO_3)_4$ per hour. An operating pressure of at least 400 psig was necessary to obtain complete conversion with 1 kg of catalyst pellets (platinum on alumina pellets; 0.5% Pt, 99.5% Al_2O_3). The continuous reduction of 0.2 M $UO_2(NO_3)_2$ solution with the same catalyst has been reported[15].

The flowsheet in Fig. 5.1 shows that $UO_2(NO_3)_2$ solution and H_2 are fed into the bottom of the reactor and that the liquid level is automatically maintained above the top of the catalyst bed by a solenoid valve in the product outlet line. The solenoid is triggered by an "on-off" controller which receives a signal from a thermal probe (a thermocouple probe with a small 5-w heater in it). The thermal probe temperature stays at about 65°C when the liquid level is low, and drops below 50°C

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[14] J. P. McBride (compiler), Preparation of UO_2 Microspheres by a Sol-Gel Technique, ORNL-3874, (in preparation).

[15] R. De Leone, G. Cogliati, L. Lorenzini, and R. Lang, "Studies of Uranium (IV) Nitrate", Third Nuclear Chemistry Conference, Gatlinburg, Tenn., October, 1962.

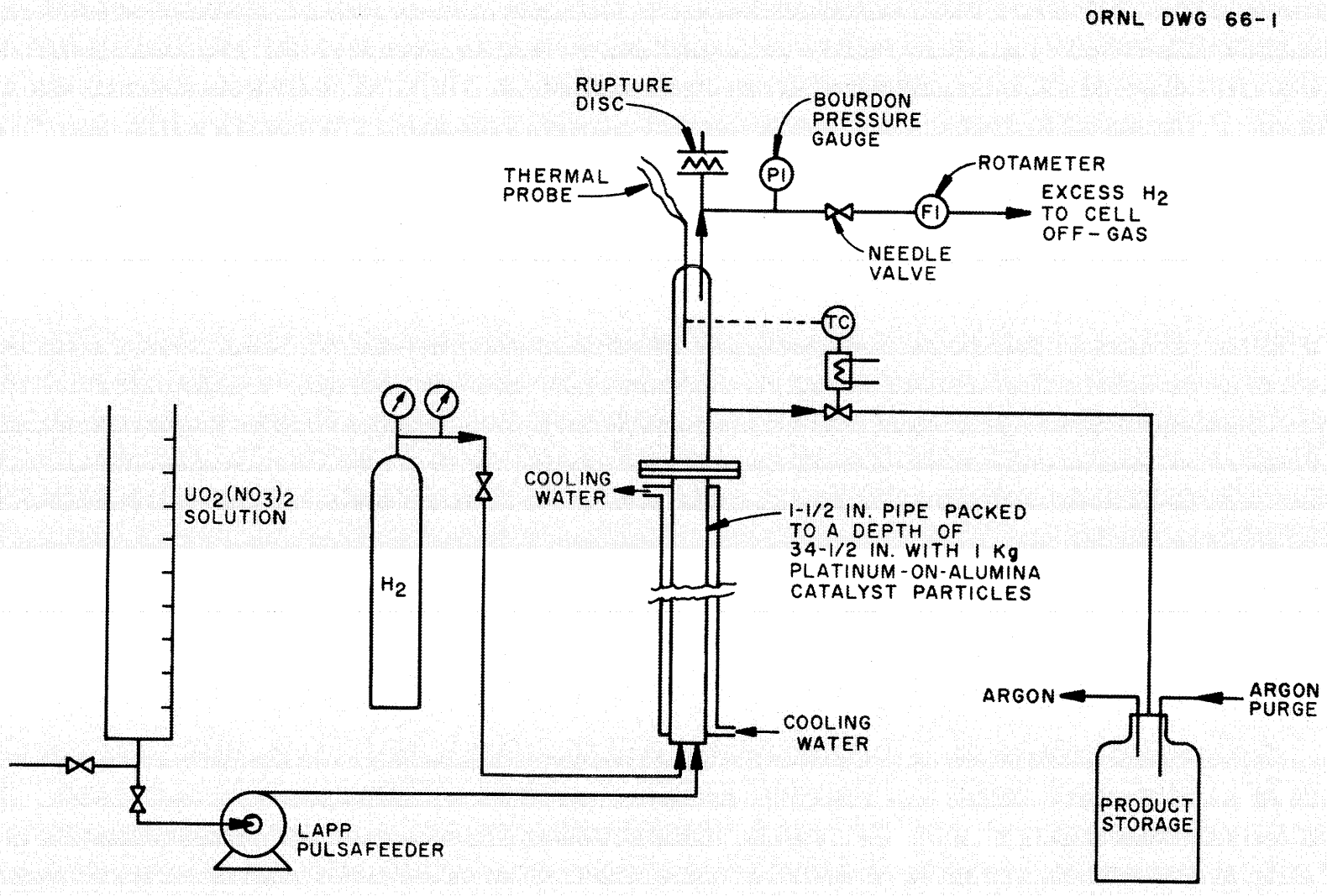

Fig. 5.1 Equipment for Continuous Production of Uranous Nitrate Solution.

in about 10 sec when the liquid reaches the probe. Attempts to control the liquid level with an electrical conductivity probe were unsuccessful, apparently because the insulation got wet. Other insulators -- Teflon, polyethylene, epoxy resin, and glass -- were tried.

The pressure on the system was controlled by the discharge-pressure-setting on the H_2 cylinder, and the H_2 flow rate was controlled by a needle valve in the off-gas line. The reactor was protected by a rupture disk, rated at 994 psig.

In the four experiments to date, conversions of 99.0 and 99.8% were obtained at hydrogen pressure of 400 and 500 psig at flow rates of nearly a liter an hour (Table 5.1). Good conversion was also obtained at 300 psig with a flow rate of 0.32 liter per hour, but at a flow rate of about one liter an hour, the conversion dropped throughout the 5-hr run. The initial reactor charge was 96% U^{4+}, and samples at 2, 3.5, and 5 hr showed U^{4+} concentrations of 78, 64, and 52%.

On a grams-per-liter basis, the feed composition for these experiments was as follows: U, 115; nitrate, 100; urea, 18.5. The solution was 1.4 N in H^+. The product analysis showed no change in nitrate or urea concentration, but hydrogen ion concentration had dropped to about 0.5 N. The product from R-23, kept in a stoppered glass jug purged with argon after filling, showed no decrease in U^{4+} concentration after one week.

5.3 Preparation of Tetravalent Uranium Sols by Catalytic Reduction of Uranyl Nitrate Solution with Gaseous H_2 in a Low-Pressure Reactor

P. A. Haas J. B. Cordiner

This study was a part of the sol-gel development work for producing fuel-oxide microspheres for use in fuel elements. It consisted in reducing a uranyl nitrate solution to a tetravalent compound dispersed in water as a colloidal sol, readily convertible to high-density, high-strength, microspheres of UO_2. Reduction was done in a 3-in.-ID reactor by gaseous hydrogen at a reactor pressure of 40 psig, using a 0.5% platinum-on-alumina catalyst.

Table 5.1 Continuous Reduction of Uranyl Nitrate Solution

Run	Flow Rate (liters/hr)	Pressure (psig)	UO_2^{2+} to U^{4+} Conversion (%)
R-20	0.32	300	96.8
R-21	0.943	300	< 52.2
R-22	0.882	400	99.0
R-23	0.900	500	99.8

Reduction was achieved over a wide range of uranyl nitrate concentration; an initial concentration of about 1.5 M is recommended, however, for ready conversion of reactor product to the desired 1.0 M sol. Critical factors for a successful reduction and sol-preparation process included active catalyst, temperature control, regulation of the nitrate ion concentration, termination of the reduction process at the optimum endpoint, and proper processing of the reactor product to prepare the sol.

Regulation of the H^+ ion concentration was not necessary, since the H^+/U ratio approached zero at the termination of reduction, independently of whether an excess or deficiency of hydrogen ions was used. The suggested reactor temperature is 45°C ± 10°, with the exact value depending on the desired degree of reduction of nitrate ions.

Convertibility of the sol to microspheres was the best criterion of optimum reduction endpoint. Results indicated an optimum endpoint corresponding to a slurry pH of 2 to 3. Electrical conductivity is recommended as a good general indication of the approach of this endpoint, with pH used as the final indication. Recommendations are included for estimating the degree of completion of the reduction and the rate of reduction at any given time.

These results will be reported in detail as an ORNL-TM report[15]; the first draft is 95% complete. The preliminary studies in an autoclave have already been published and the abstract follows[16]: "The preparation of a uranous sol from uranyl nitrate requires reduction of UO_2^{2+}, removal of nitrate, and formation of a dispersable uranous oxide. Tests in an autoclave have shown that these steps can be accomplished in a single operation using H_2 with a Pt catalyst at below 50°C. Reduction at higher temperatures without a catalyst did not give a dispersible oxide. The nitrate is removed as NO, H_2O, N_2 and other volatile reduction products.

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[15] J. B. Cordiner, Preparation of Uranium (IV) Sols by Catalytic Reduction of Uranyl Nitrate Solution with Gaseous Hydrogen in a Low-Pressure Reactor, (to be published as an ORNL-TM report).

[16] P. A. Haas, Sustained Reduction of $UO_2(NO_3)_2$ to Remove Nitrate as Volatile Reduction Products and Prepare a Urania Sol, CF-65-6-79, June 28, 1965 (Declassified).

However, control of the reaction and reproducibility of results were poor in the autoclave. The tests will be continued in a low-pressure system designed for better control of the process variables."

6. SEPARATION OF BIOLOGICAL MACROMOLECULAR SUBSTANCES

C. W. Hancher C. D. Watson H. O. Weeren

The production of t-RNA, on a kilogram scale, from E. coli cells is undergoing development. Two preliminary runs and three production runs were made. The combined nucleic acid precipitate from production runs 2 and 3 yielded 7.9 kg of the lighter RNA fraction, containing 22.4% RNA, 0.1% DNA. Additional processing steps are required before the final mixed t-RNA product is obtained.

In addition to the RNA runs, a cold room (21 x 21 ft) was completed on the second floor of Building 4505, and construction was started on a solvent storage and handling facility. The design of a process refrigeration system and a water demineralization-distillation system was begun.

The RNA extraction process flowsheet (Fig. 6.1) consists of four major steps:

Step 1. - E. coli cells are concentrated and mixed with buffer solution and phenol. The phenol destroys the cell walls and places the constituents of the cell nucleus into solution. The phenol and cell residues are removed by centrifugation, leaving the nucleic acids in the aqueous solution. The nucleic acids are then precipitated with ethanol and centrifuged.

Step 2. - In the second step, the desired RNA is separated from the bulk of the other nucleic acids by precipitating the DNA and high-molecular-weight RNA in isopropyl alcohol and centrifuging them out of the solution.

Step 3. - The desired RNA is separated from the remaining DNA and other impurities by loading the material on DEAE cellulose (organic ion exchanger) and preferentially eluting the RNA.

Step 4. - Purified RNA is separated into its several fractions by chromatographing. The entire process tapers sharply as it progresses: 1 kg of cells yields about 1 g of mixed t-RNA.

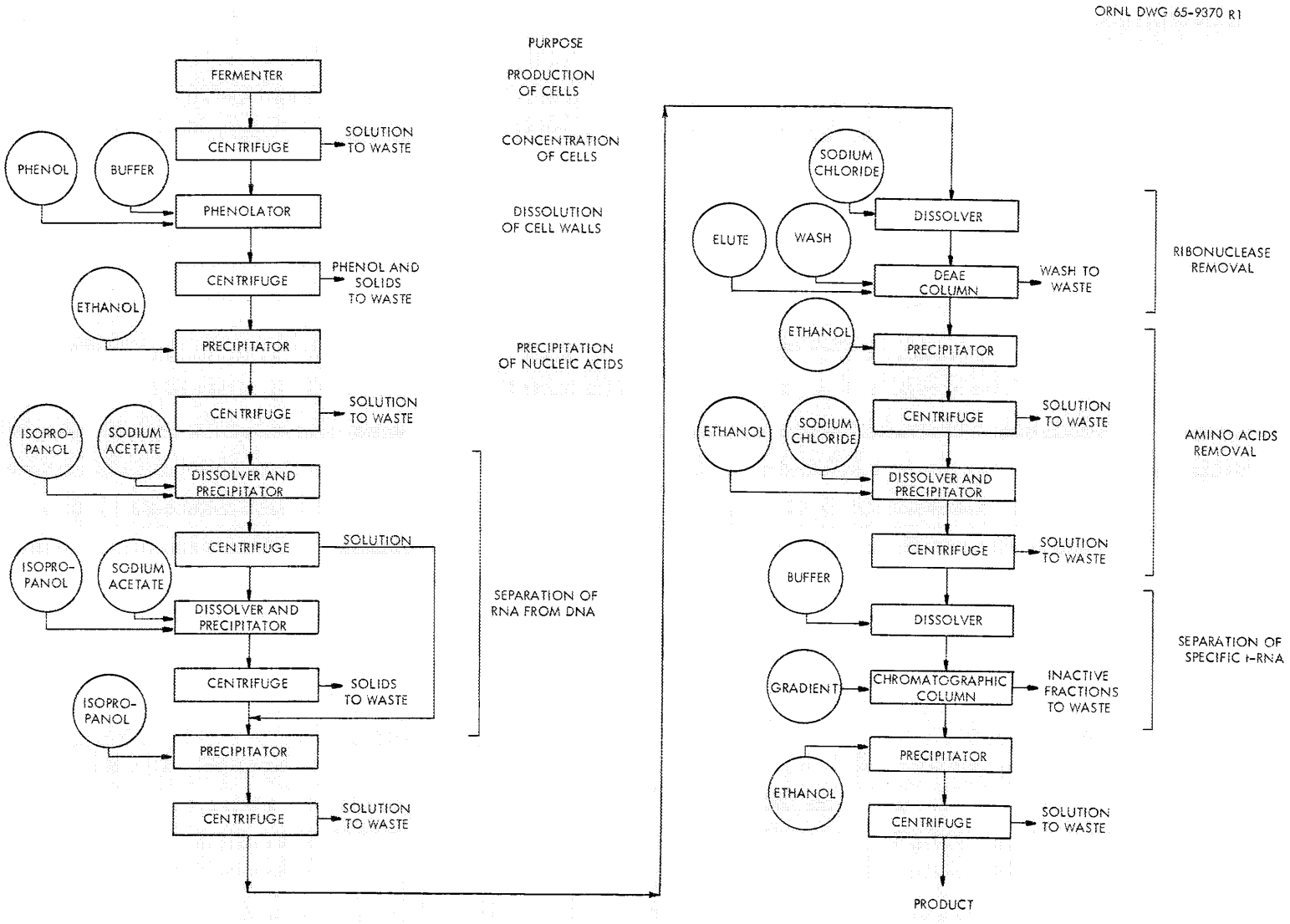

Fig. 6.1 Flowsheet for Extracting Ribonucleic Acids.

The original plan for the RNA extraction process was to grow the E. coli cells in the Biology Division's fermenter at the Y-12 Plant, concentrate the cells in a centrifuge there, bring the concentrated cells to the cold room of the Chemical Technology Division, and complete the remaining processing there. However, delivery of a centrifuge required for the handling of many of the head-end steps of the process was delayed. Therefore, the centrifuging of large volumes was done at Y-12, and the material was brought to ORNL only when the volumes had been reduced sufficiently to be further centrifuged here.

Two preliminary runs and three production runs were made at Y-12. The preliminary runs were made to obtain data on the processing variables; the runs were made with the fermenter operated batchwise. These runs showed, among other things, that the fermenter must be operated continuously to keep pace with the rest of the process, and the three production runs were made accordingly.

The first preliminary run (made on July 20) established that the Sharples centrifuge at Y-12 adequately separated the phenol and the cell residue from the aqueous solution containing the nucleic acids. It was also established that the phenol and cell residue would drain from the centrifuge bowl when the bowl was stopped and that it would not be necessary to dismantle and clean the bowl every few minutes during the run. This favorable development greatly simplified this particular operation.

The second preliminary run (made on July 27) was mainly a series of scouting tests to establish favorable procedures for cell concentration, buffering, and phenolation. These tests showed that cells concentrated on the Westfalia centrifuge were far easier to handle and gave higher yields than the cells concentrated on the Sharples centrifuge. They also indicated that the mixing of phenol and buffered cells was best done by adding the buffered cells slowly to an agitated phenol solution and that long contact between the buffered cells and the phenol was not only unnecessary but undesirable. In this run, 8.5 kg of cells (wet weight) were processed to yield 291 g of nucleic acid precipitates in ethanol (wet weight). This precipitate was taken to ORNL for further processing.

Three production runs were made. In the first run, which started August 19, 17.2 kg of E. coli cells were processed, yielding 546 g of nucleic acid precipitates in ethanol from 30.5 kg of cells processed. The second run yielded 1075 g of nucleic acid precipitate.

In the first two runs, the E. coli fermenter was operated with a continuous glucose nutrient feed and drawoff of the cells, which were concentrated and collected by a Westfalia centrifuge. Periodically, the cells were buffered, phenolated, and centrifuged. The phenol waste was discarded, and the aqueous product was treated with ethanol and stored. After about 12 hr of operation, the concentration of cells in the fermenter began to decrease steadily, despite corrective measures, and the runs were stopped.

The cessation of cell growth in the fermenter was attributed to a cell-destroying virus. Before the third run, therefore, several possible paths by which the virus could have been introduced into the culture were closed, and a more resistant strain of E. coli (strain w) was used.

The third run began August 31 and was continued without interruption for 80 hr until September 3, when it was stopped. The operation was smooth and uneventful, and 300 kg of cells were processed to yield 17.4 kg of nucleic acid precipitates in ethanol.

The nucleic acid precipitates from runs 2 and 3 were combined and dissolved in sodium acetate solution, and the DNA and heavier RNA fraction were precipitated in isopropanol, centrifuged, and discarded. The lighter RNA fraction was then precipitated by a higher isopropanol concentration and centrifuged. A total of 7.9 kg of the lighter RNA fraction was obtained. An analysis gave values for the RNA and DNA concentrations of 22.4% and 0.1%, respectively. The remainder of the precipitate is an unknown contaminant that presumably will be removed in the additional steps required to purify the product.

7. RADIATION RESISTANCE OF PROTECTIVE COATINGS (PAINTS)

G. A. West

Tests to evaluate some protective coatings (paints) for use in reactor containment vessels, spent-fuel processing plants, and laboratories were conducted by exposing (1) some specimens to gamma radiation, (2) other specimens to the fallout of radioactive fission products from the plasma-jet volatilization of irradiated UO_2 in a 1350 ft^3 vessel, and (3) selected coatings to a tentative standard decontamination test. The Nuclear Safety Pilot Plant group conducted the volatilization test with coating specimens supplied by the Unit Operations Section.

7.1 Radiation-Tolerance Testing

Protective coatings were exposed to ^{60}Co gamma radiation at an intensity of 1×10^6 r/hr at 40 to 50°C in air and in demineralized water. Several manufacturers submitted special formulations of their coatings in an attempt to discover a coating with unusual or outstanding resistance to gamma radiation in the presence of deionized water.

Three such coatings surpassed the previous high (5×10^9r) resistance in demineralized water: (1) Carboline Co's. system K, consisting of a modified phenolic (Phenoline 368) containing embedded glass cloth and Phenoline 368 seal coat. This coating failed at 8.3×10^9r by blistering and loss of adhesion, (2) Varni-Lite Corp's. No. S-0900 epoxy system failed at 8.0×10^9r by blistering, (3) Amercoat No. 1762 with No. 66 epoxy seal coat failed at 7.9×10^9r by chalking (Table 7.1).

There is evidence of greater radiation tolerance in air since 19 of 79 coatings appear to be serviceable after an exposure of 1×10^{10}r. At this time 18 other coatings have been exposed to 3×10^9r in air. Irradiation will continue to failure or to 1×10^{10}r whichever occurs first.

7.2 Studies of Contamination Due to a Simulated Reactor Excursion

Protective-coating specimens and controls were exposed to an atmosphere contaminated with radioactive fission products from the plasma-jet

Table 7.1 Radiation-Resistance Rating of Several Protective Coatings

Radiation Source: Cobalt-60 at 1×10^6 r/hr
Temperature: 40 to 50°C
Manufacturers are listed alphabetically, with the coatings listed in decreasing order of resistance to radiation

Manufacturer	Coating	Substrate	Exposure Under Demineralized Water (r)	Effect	Exposure in Air (r)	Effect
Amercoat Corporation	Epoxy, No. 1762 and No. 66	concrete	7.9×10^9	A	6.2×10^9	D
	Epoxy, No. 4000 Bondstrand pipe	fiberglas	1.0×10^{10}		1.0×10^{10}	
	Polyester, No. 5000 Bondstrand pipe	fiberglas	1.0×10^{10}		1.0×10^{10}	
	Inorganic, Dimetcote-3	steel			2.2×10^{10}	
	Inorganic, No. 1741	316 steel	9.6×10^8	C	1.0×10^{10}	F
	Inorganic, Dimetcote-4	steel	3.9×10^9	G	1.0×10^{10}	
	Epoxy, No. 1762 and No. 66 gloss	conc.	2.2×10^9	C	4.2×10^9	B
	Epoxy, No. 1762 and No. 1732	conc.	1.7×10^9	C	4.2×10^9	B
	Vinyl, No. 1762 and No. 33HB	conc.	1.2×10^9	C	3.2×10^9	A,B
	Epoxy, No. 1762 and No. 74	conc.	1.7×10^9	A,B,C	2.7×10^9	B
	Vinyl, No. 1762 and No. 88	conc.	1.2×10^9	C	2.7×10^9	A,B
	Vinyl, No. 1762, No. 88, and No. 99	conc.	1.2×10^9	C	2.7×10^9	A,D
Carboline Company	Modified phenolic, system K	conc.	5.8×10^9	C,D	1×10^{10}	
	(Phenoline 368)	steel	8.3×10^9	C,D	1×10^{10}	
	Modified phenolic, system C	conc.	3.6×10^9	C,D	8.3×10^9	B,C
	(Phenoline 300)	steel	6.3×10^9	A,B	1×10^{10}	
	Modified phenolic, system H	conc.	2.6×10^9	C	1×10^{10}	
	(Phenoline 368)	steel	6.3×10^9	C	1×10^{10}	
	Modified phenolic, system G	conc.	4.7×10^9	C	1×10^{10}	
	(Phenoline 368)	steel	4.7×10^9	C	1×10^{10}	
	Modified phenolic, system I	conc.	4.7×10^9	C	1×10^{10}	
	(Phenoline 368)	steel	3.6×10^9	C	1×10^{10}	
	Modified phenolic, system F	conc.	No test		7.8×10^9	C
	(Phenoline 300-368)	steel	3.6×10^9	C	6.8×10^9	C
	Modified phenolic, system J	conc.	2.1×10^9	A	4.7×10^9	B,C
	(Phenoline 305)	steel	2.1×10^9	A	1×10^{10}	
	Modified phenolic, system A	conc.	4.3×10^9	A,C	3.2×10^9	C
	(Phenoline 300)	steel	4.7×10^9	C	7.1×10^9	B,C
	Modified phenolic, system B	conc.	2.6×10^9	C	5.6×10^9	B,C
	(Phenoline 305)	steel	3.6×10^9	C	7.1×10^9	C
	Modified phenolic, system D	conc.	2.1×10^9	A,E	6.0×10^9	C
	(Phenoline 300)	steel	4.7×10^9	A,E	1×10^{10}	
	Modified phenolic, system E	conc.	2.1×10^9	A	6.0×10^9	C
	(Phenoline 300)	steel	2.1×10^9	A	6.8×10^9	B

Table 7.1 (Continued)

Manufacturer	Coating	Substrate	Exposure Under Demineralized Water (r)	Effect	Exposure in Air (r)	Effect
Devoe and Raynolds	Epoxy, (No. 4), 46612, 43614	steel	1.0×10^{9}	C	1.0×10^{10}	
	Cat. Epoxy, (No. 2), 46612, OX-1055	steel	1.57×10^{9}	C	1.0×10^{10}	
	Mod. Epoxy, (No. 3), ReNew Coat	conc.	2.9×10^{8}	A	1.0×10^{10}	
	Epoxy, (No. 3), 46612, SX-1346	steel	2.0×10^{8}	C	1.0×10^{10}	
	Acrylic, (No. 1), MX-140, MX-142, MX-145	steel	2.0×10^{8}	C	1.0×10^{10}	
	Chlor. Rubber, (No. 2C), SX-1662, SX-1661	conc.	3.2×10^{8}	C	1.0×10^{10}	
	Vinyl, (11-S), 46610, SX-1667	steel	8.5×10^{9}	C	1.0×10^{9}	C
	Epoxy, (1-C), SX-1655, SX-1655	conc.	1.3×10^{9}	C	8.97×10^{9}	C
	Epoxy, (FS-3C), Cat. 46000, SX-1665	steel	1.58×10^{8}	A,B,E	$>7.0 \times 10^{9}$	
	Vinyl, (FS-5), SX-1666	steel	1.8×10^{8}	C	$>7.0 \times 10^{9}$	
	Epoxy, (FS-4), RB-1304, SX-1665	steel	9.0×10^{9}	C	3.87×10^{9}	C
	Epoxy, (FC-6), SX-1665	conc.	1.4×10^{9}	A,B	$>7.0 \times 10^{9}$	
	Epoxy, (FC-3), OX-1055	conc.	1.4×10^{9}	A,B	$>7.0 \times 10^{9}$	
	Epoxy, (FC-2), Cat. 48101, OX-1055	conc.	1.4×10^{8}	A,B	$>7.0 \times 10^{9}$	
	Epoxy, (FS-3B), Cat. 46000, SX-1665	steel	9.0×10^{8}	C	$>7.0 \times 10^{9}$	
	Epoxy, (FS-2), Cat. 46610, 46613, 46614	steel	5.4×10^{9}	C	$>7.0 \times 10^{9}$	
	Epoxy, (FC-8), SX-1029	conc.	1.4×10^{8}	C	2.68×10^{9}	A,C
	Epoxy, (FS-3A), Cat. 46000, SX-1665	steel	9.0×10^{8}	C	2.8×10^{9}	C
	Acrylic, (FC-7), MX-148	conc.	1.8×10^{9}	C	2.68×10^{9}	B,D
	Epoxy, (FG-6), SX-1655(FG)	conc.	1.5×10^{9}	A	$>5.0 \times 10^{9}$	
	Epoxy, (FG-5), SX-1665(FG)	conc.	1.5×10^{9}	A,C	2.5×10^{9}	C
	Epoxy, (FG-3), OX-1055(FG)	conc.	1.5×10^{9}	A	$>5.0 \times 10^{9}$	
	Epoxy, (FC-4), SX-1665	conc.	1.4×10^{9}	A,C	1.7×10^{9}	C
	Cat. Epoxy, (No. 2), OX-1055	conc.	1.57×10^{9}	C	1.53×10^{9}	B
	Epoxy, (FG-4), SX-1665(FG)	conc.	1.5×10^{8}	C	1.5×10^{10}	C
	Epoxy, (1-S), 4600, SX-1655	steel	8.5×10^{8}	C	1.0×10^{10}	C
	Chlor. Rubber, (No. 2C), SX-1662, SX-1661	steel	3.2×10^{8}	C	1.0×10^{9}	B,C,D
	Epoxy, (FC-1), Cat. 48101, SX-1346	conc.	5.4×10^{8}	C	$>7.0 \times 10^{9}$	
	Chlor. Rubber, (No. 6), 14700, 47212, 47201	steel	2.0×10^{8}	A,C	9.5×10^{9}	B
	Epoxy, (FS-6), RB-1033, SX-1029	steel	1.8×10^{8}	C	2.2×10^{9}	C
	Vinyl, (10-C), No. SX-1667 and 1666	conc.	5.28×10^{8}	C	1.87×10^{9}	A,B
	Chlor. Rubber (No. 5), 4600, 47212N, 47201	steel	2.0×10^{8}	A,C	1.53×10^{9}	B
	Epoxy, (No. 1), OX-1055 and SX-1346	conc.	2.0×10^{8}	C,D	1.5×10^{8}	B
	Vinyl, (FC-9), L-10429, PVS	conc.	1.8×10^{8}	C,D	9.0×10^{8}	D
	Acrylic, (FS-1), 303, MX-145, MX-148	steel	9.0×10^{9}	C	5.4×10^{9}	B,D
	Epoxy, (FC-5), SX-1665	conc.	1.4×10^{9}	A,B	$>7.0 \times 10^{9}$	

Table 7.1 (Continued)

Manufacturer	Coating	Substrate	Exposure Under Demineralized Water (r)	Effect	Exposure in Air (r)	Effect
Hughson Chemical Company	Polyurethane, Z552, (B)	conc.	1.0×10^9	C	$>6.0 \times 10^9$	
		steel	1.0×10^9	D	4.6×10^9	B
	Polyurethane, Z051, (C-1)	steel	1.0×10^8	C,E	4.6×10^9	B
	Polyurethane, Z051, (C-3)	steel	1.68×10^9	C	4.6×10^9	B
	Polyurethane, Z552, (A)	conc.	1.0×10^8	C	$>6.0 \times 10^9$	
		steel	1.68×10^8	C	2.9×10^9	B
	Polyurethane, Z001, (D)	steel	5.0×10^8	C,E	4.6×10^9	B
	Polyurethane, Z452, (E)	steel	5.0×10^8	D	4.6×10^9	B
	Polyurethane, Z051, (C-2)	steel	1.68×10^8	C	1.3×10^9	C
	Polyurethane, Z051, (C-4)	steel	1.68×10^8	C	1.0×10^9	C
Varni-Lite Corporation	Epoxy, S0900	conc.	8.0×10^9	C	1.0×10^{10}	
		steel	(not in H_2O)		1.0×10^{10}	
	Epoxy, Industrial	conc.	8.56×10^8	C	4.0×10^9	B
		steel	(not in H_2O)		1.0×10^{10}	
	Epoxy, S0800	conc.	8.56×10^8	C	3.0×10^9	B,C
		steel	(not in H_2O)		3.0×10^9	B,C
	Epoxy, Industrial Special	conc.	8.56×10^8	C	1.9×10^9	C
		steel	(not in H_2O)		1.9×10^9	C
Wisconsin Protective Coating Company	Mod. Phenolic, (No. 5), 7155, FG, 7133H	steel	3.0×10^9	B,E	1.4×10^{10}	
	Mod. Phenolic, (No. 4), 7100H, FG, 7133H	steel	1.1×10^9	C	1.4×10^{10}	
	Mod. Phenolic, (No. 2), 9028, 7155, FG, 7133H	conc.	3.0×10^9	B	7.2×10^9	B
	Mod. Phenolic, (No. 1), 9028, 7100H, FG, 7133H	conc.	2.7×10^9	C	4.2×10^9	C
	Epoxy, (No. 3), 9028, 9009-1	conc.	3.0×10^9	C	2.7×10^9	B,C
	Epoxy, (No. 6), 9009-1	steel	3.0×10^9	B	2.7×10^9	B

Radiation effect: A - coating chalked; B - coating embrittled; C - coating blistered; D - loss of adhesion; E - interfacial area attack; F - rough, sand-like, surface; G - bleeding of binder materials.

volatilization of natural UO_2 that had been irradiated at an average neutron flux of 4.5×10^{10}. This resulted in about 0.6 curies of total mixed fission product gamma activity after cooling for 7 days. In tests simulating a reactor excusion and the volatilization of fuel, the amount of radionuclides deposited on the exposed surface was measured and compared (Table 7.2). Radionuclides were identified and measured by scanning the specimens with a gamma-ray spectrometer before decontamination.

Coatings exhibiting the least retention of the ten radionuclides measured are listed below:

Vinyls

No. 33, Amercoat Corporation
No. SX-1666, Devoe and Raynolds Company
Nukem-40, Amercoat Corporation

Epoxies

No. R4-74F, Plas-Chem Corporation
No. R4-74E, Plas-Chem Corporation
No. SX-1655, Devoe and Raynolds

Modified Phenolics

Phenoline 300, Carboline Company
Phenoline 305, Carboline Company
No. 7122, 7133X, Wisconsin Protective Coating Company

Polyester

Liquid Glass No. 5301, Stanley Chemical Company

Chlorinated Rubber

No. SX-1661, Devoe and Raynolds Company

Inorganics

Dimetcote-4, Amercoat Corporation
Dimetcote-3, Amercoat Corporation

7.3 Improved Test to Determine Decontaminability of Coatings

A standard (ASTM type) test for evaluating the ease and extent of decontaminability of protective coatings or paints does not now exist. A previous procedure included the use of 3 M (17.2%) HNO_3, which in some cases attacked and removed a surface layer from some of the coatings. Also, the

Table 7.2 Retention of Volatilized Radionuclides on Protective Coatings and Control Materials

Irradiated UO_2 was volatilized by a plasma torch and transported via a short connecting pipe to a 1350 ft^3 stainless steel containment vessel where samples were systematically located.
Vessel temperature: 105°C
Source material: Natural enriched UO_2 irradiated to ~ 0.6 curies (4.5×10^{10} n/cm²/sec, average)
Activity before melt (millicuries): ^{131}I - 33.4; ^{147}Nd - 26.3; ^{239}Np - not calculated; ^{132}Te - 28.3; ^{103}Ru - 16.2; ^{141}Ce - 45.6; ^{99}Mo - 30.5; ^{95}Zr-Nb - 24.6; ^{137}Cs - 0.161; ^{140}Ba-La - 69.8.

	Manufacturer*	^{131}I dpm/cm²	^{147}Nd dpm/cm²	^{239}Np dpm/cm²	^{132}Te dpm/cm²	^{103}Ru dpm/cm²	^{141}Ce dpm/cm²	^{99}Mo dpm/cm²	^{95}Zr-Nb dpm/cm²	^{137}Cs dpm/cm²	^{140}Ba-La dpm/cm²
Pyrex glass		473	51	41	32	5	3	24	1	1	0
Aluminum		1,650	123	85	67	7	5	54	0	0	1.5
304L stainless steel		1,590	143	117	60	15	33	45	0.2	3	1
Copper		2,330	140	170	67	4	44	28	0	0	0
Carbon steel		3,880	227	166	70	10	113	0	0	0	0
Galvanized steel		7,880	842	313	67	0	339	0	0	0	0
Vinyl (average)		1,960	178	100	46	11	72	34	0	0	0
No. 33	1	1,310	61	68	42	7	0	36	0	0	0
SX-1666	5	1,300	91	94	32	9	94	0	0	0	0
Nukem-40	1	1,360	167	56	39	4	0	32	0	0	0
Series-A	4	1,410	50	160	65	9	47	63	0	0	0
No. 33 HB	1	1,520	127	65	43	11	54	34	0	0	0
Chem Flex-1620	7	1,550	141	89	3	0	89	0	0	0	0
TP-216	11	1,670	101	114	74	0	74	0	0	0	0
Polyclad No. 933-1 D. G.	2	1,720	127	104	50	13	82	29	0	0	0
UL-8134	6	1,680	183	117	56	11	74	49	0	0	0
Polyclad No. 933-3	2	2,080	247	81	37	0	70	43	0	0	0
No. 88	1	2,180	227	63	26	0	91	0	0	0	0
AV-461	11	2,500	244	140	67	74	66	75	0	0	0
Polyclad No. 933-1; 1200-20	2	3,720	358	173	71	0	156	77	0	0	0
No. 99 semigloss	1	4,450	430	122.5	41.2	15.3	144	50	0	0	0
SB-312	11	9,160	120	53	49	8	35	19	0	0	0
Epoxy (average)		4,680	399	190	57	12	166	62	0	2	0
R4-74F	7	1,100	136	74	49	10	0	49	0	0	0
R4-74E	7	1,140	266	141	24	14	109	65	0	47	0
SX-1655	5	2,000	227	102	42	0	113	41	0	0	0
R3-133D-2	7	1,970	217	119	46	78	112	0	0	0	0
No. 74 Seal gray	1	2,420	213	183	54	15	59	46	0	0	0
16-210	6	2,300	261	134	63	12	105	61	0	0	0
Cp 2100 HB special	7	2,560	496	126	51	0	130	0	0	0	0
UC-9700	6	2,760	286	148	48	0	149	45	0	0	0
Chem-Pon 2310	7	2,770	349	148	74	0	94	77	0	0	0
1206 Tile cote	10	3,220	386	175	85	13	138	83	0	0	0

Table 7.2 (continued)

	Manufacturer*	^{131}I dpm/cm^2	^{147}Nd dpm/cm^2	^{239}Np dpm/cm^2	^{132}Te dpm/cm^2	^{103}Ru dpm/cm^2	^{141}Ce dpm/cm^2	^{99}Mo dpm/cm^2	^{95}Zr-Nb dpm/cm^2	^{137}Cs dpm/cm^2	^{140}Ba-La dpm/cm^2
Chem-Pon 2315	7	3,510	367	168	48	0	190	0	0	0	0
R3-80A	7	3,560	309	195	49	13	202	0	0	0	0
Colma prot. ctg.	9	3,670	136	326	95	0	163	118	0	0	0
Gilsaloid tile red	8	3,740	376	215	55	16	203	69	0	0	0
UC-9647	6	4,130	430	196	67	0	186	83	0	0	0
LOR	7	4,340	357	232	61	0	254	0	0	0	0
R3-132B	7	4,240	421	225	63	17	219	94	0	0	0
No. 66 gray	1	5,320	504	146	42	17	157	53	0	0	0
Coroline 505.2	3	7,060	689	132	33	21	86	71	0	0	0
Colma surface kote	9	6,710	662	330	71	0	228	173	0	0	0
Tygoweld 56-63	11	12,400	1,350	562	117	43	589	233	0	0	0
9009-1R	12	20,000	104	57	45	8	0	36	0	0	0
Modified phenolic (average)		3,770	452	216	620	19	200	60	0	0	0
Phenoline 300	2	1,592	70	118	47	4	44	51	0	0	0
Phenoline 305	2	1,780	212	80	61	0	50	49	0	0	0
7122-7133X	12	3,540	395	159	47	0	179	0	0	0	0
7155-7133X	12	1,995	690	328	3,390	50	405	0	0	0	0
7118 + clear	12	5,960	622	253	70	15	225	73	0	0	0
7100	12	5,980	576	292	72	25	278	123	0	0	0
7118 + 7118T	12	7,060	724	265	46	23	298	0	0	0	0
Phenoline 368	2	7,760	768	321	93	35	246	131	0	0	0
Polyester											
Liquid glass-5301	10	909	112	61	31	0	52	20	0	0	0
Chlorinated rubber											
SX-1661	5	2,930	342	168	53	0	196	0	0	0	0
Inorganic (average)		6,800	570	197	49	8	192	36	0	0	0
Dimetcote-4	1	3,770	381	108	37	10	132	46	0	0	0
Dimetcote-3	1	5,280	433	186	38	0	192	0	0	0	0
No. 1741	1	9,180	733	248	60	12	222	50	0	0	0

*Manufacturer:
1. Amercoat Corp., 201 N. Berry St., Brea, California
2. Carboline Co., 32 Hanley Industrial Court, St. Louis, Missouri
3. Ceilcote Co., 4832 Ridge Rd., Cleveland 9, Ohio
4. David E. Long Corp., 133-20 91st Avenue, Jamaica 18, Long Island, New York
5. Devoe and Raynolds Co., Inc., 116 Baver Avenue, Louisville, Kentucky
6. Pittsburg Plate Glass Co., Gateway Center, Pittsburg 22, Pennsylvania
7. Plas-Chem Corp., 6177 Maple Avenue, St. Louis, Missouri
8. Rowe Products, Inc., Niagara Falls, New York
9. Sika Chemical Co., 35 Gregory Avenue, Passiac, New Jersey
10. Stanley Chemical Co., Div. The Stanley Works, East Berlin, Connecticut
11. U. S. Stoneware Co., Akron 9, Ohio
12. Wisconsin Protective Coating Co., 1136 W. Mason St., Green Bay, Wisconsin

contaminant solution (a mixture of mixed fission products in nitric acid) became more concentrated as the solution evaporated to dryness, again resulting in surface attack.

A new approach to the evaluation of decontaminability is being attempted through tests conducted by E. I. Wyatt of the Analytical Chemistry Division. His contaminant consisted of a nitric acid solution of mixed fission products obtained from spent fuel (irradiated to 20,000 Mwd/ton and cooled for a year). The photopeaks of this solution as distinguished by a gamma-ray spectrometer were those associated with ^{144}Ce-Pr, ^{106}Ru-Rh, ^{95}Zr-Nb and ^{137}Cs- ^{137m}Ba. All were present to about the same extent. The solution was originally 8 M in HNO_3 but had been adjusted to a pH of 4 immediately before application to the specimen. (Without a complexing agent, Zr-Nb^{95} soon precipitate from weak acids.)

During decontamination, there was no visual indication of corrosive attack of the coatings by the weak acid solution of 0.4 M $C_2H_2O_4$, 0.05 M NaF, 0.30 M H_2O_2. Stepwise decontamination was performed by suspending the test specimens in (1) water, (2) mild acid at room temperature and (3) mild acid at 80°C, which was agitated by a magnetic stirrer for periods of 10 min each. The radionuclides remaining on the coupons were measured and identified by a gamma-ray spectrometer. All the dry coupons counted before decontamination had nearly the same amount of radioactivity, which was in the range 6×10^4 to 2×10^5 dis/min per nuclide.

The first water wash removed so much of the ^{137}Cs- ^{137m}Ba that they were never again found on any of the coupons. The other contaminants were removed generally to the extent of 50 to 90% for all coupons, with plate glass being the most completely decontaminated.

After the water wash, no ^{95}Zr-Nb remained on the plate glass. Other coupons were not as readily cleaned as the glass; however, epoxy SX-1346 gave up about 98% of the ^{95}Zr-Nb. After the 80°C mild-acid wash, the contaminants on the glass and Varni-Lite S-0900 had reached the lower limit of detection, providing an overall decontamination factor of 600.

Ruthenium^{106} is one of the more difficult to remove. Table 7.3 shows the decontamination results for the total nuclides and ^{106}Ru from the

Table 7.3 Decontamination of Coatings: Comparison of Results for Overall Factors with Factors for Ruthenium

Test Coupon	Initial Radioactivity (dpm)	Final Radioactivity (dpm)	Overall Decontamination Factor	Decontamination Factor for ^{106}Ru
	CONTROLS			
Plate glass	5.18×10^5	<550	>900	>800
Stainless steel, 304L (2-B finish)	5.66×10^5	1.01×10^3	560	185
Teflon sheet	6.00×10^5	2.43×10^3	247	185
Polyethylene sheet	5.30×10^5	1.81×10^3	293	112
	TEST COATINGS			
Amercoat Corp.				
No. 99 semigloss (vinyl)	5.72×10^5	3.65×10^3	156	56
No. 33 HB (vinyl)	5.20×10^5	3.56×10^4	14	29
No. 66 (epoxy)	5.26×10^5	7.64×10^4	7	7
Carboline Co. Coatings				
Phenoline 300 (mod. phenolic)	4.72×10^5	1.93×10^4	24	13
Phenoline 305 (mod. phenolic)	6.14×10^5	2.65×10^4	23	10
Phenoline 368(H), (mod. phenolic)	5.34×10^5	2.77×10^4	19	13
Phenoline 368(G), (mod. phenolic)	5.27×10^5	3.73×10^4	14	8
Devoe and Raynolds Corp.				
No. SX-1346 (epoxy)	5.97×10^5	4.75×10^3	125	52
No. SX-1667 (vinyl)	5.39×10^5	4.50×10^3	120	40
No. SX-1666 (vinyl)	5.80×10^5	5.80×10^3	100	35
No. 47201 (chlor. rubber)	5.34×10^5	5.47×10^3	97	32
No. SX-1661 (chlor. rubber)	5.34×10^5	7.12×10^3	75	25
No. SX-1665 (epoxy)	4.99×10^5	6.94×10^3	72	35
No. SX-1665(B), (epoxy)	4.88×10^5	7.03×10^3	69	39
No. 46614 (epoxy)	5.20×10^5	8.09×10^3	64	29
No. MX-145 (acrylic)	5.46×10^5	2.18×10^4	20	8
David E. Long Corp.				
Series-A (vinyl)	5.39×10^5	1.19×10^4	45	19
Pittsburg Plate Glass Co.				
No. UC-9647 (epoxy)	5.34×10^5	5.61×10^4	9	17
No. U -8134 (vinyl)	5.76×10^5	8.55×10^3	66	44
Varni-Lite Corp.				
No. S-0900 (epoxy)	5.34×10^5	<900	>590	>330
Industrial special (epoxy)	5.75×10^5	7.51×10^3	78	30
No. S-0800 (epoxy)	5.94×10^5	1.36×10^4	44	18
Industrial (epoxy)	5.84×10^5	2.25×10^4	26	26
Wisconsin Protective Coating Co.				
Plasite 7155 (mod. phenolic)	5.53×10^5	1.62×10^4	34	14
Plasite 7100 (mod. phenolic)	4.82×10^5	2.40×10^4	20	15
Plasite 7122 (mod. phenolic)	4.39×10^5	3.14×10^4	14	10

various materials and coatings after 10 min each of (1) washing in water, (2) cleaning in 0.4 M $C_2H_2O_4$--0.05 M NaF--0.30 M H_2O_2 at room temperature and (3) repetition of step 2 with the solution at 80°C. All the liquids were agitated.

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