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LWR Fuel Reprocessing and Recycle Program Quarterly Report for Period April 1 to June 30, 1976

B. L. Vondra

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LWR FUEL REPROCESSING AND RECYCLE PROGRAM

QUARTERLY REPORT FOR PERIOD APRIL 1 TO JUNE 30, 1976

B. L. Vondra, Program Manager

Contributing Authors:

ORNL: W. D. Bond, C. H. Brown, D. O. Campbell, R. B. Fitts, J. H. Goode, W. S. Groenier, D. C. Hampson, S. V. Kaye, J. C. Mailen, R. E. Moore, O. K. Tallent, L. M. Toth, W. E. Unger, V. C. A. Vaughen, C. D. Watson

ORGDP: J. M. Morrison, J. H. Pashley, M. J. Stephenson

JULY 1976

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FOREWORD

Eight tasks under the LWR Fuel Reprocessing and Recycle Program have been assigned to UCC-ND with major efforts centered at the Oak Ridge National Laboratory. Because of the close relationship of the reprocessing activities with the ongoing LMFBR Fuel Reprocessing Program at ORNL, those tasks will be managed within a combined LWR/LMFBR Fuel Reprocessing Program under the direction of W. D. Burch, Program Director. B. L. Vondra, as LWR Program Manager, will coordinate all activities reported here. Responsibility for the coordination and collation of reports of all work at Oak Ridge was assigned to ORNL by SRO/SRL, who is designated as the lead organization for the LWR Fuel Recycle Program.

Projects for which budget and work plans have been established are as follows:

F-OR-02-001	SRO/LWR:	Assistance in Shear Development and Related Problems
F-OR-02-002	SRO/LWR:	Head-End Studies: Voloxidation and Dissolution
F-OR-02-003	SRO/LWR:	Off-Gases Fluorocarbon Absorption Studies
F-OR-02-004	SRO/LWR:	Purex Process Studies (Solvent Extraction)
F-OR-02-005	SRO/LWR:	Finishing Processes - MOX Fuel Fabrication
F-OR-02-006	SRO/LWR:	Uranium Hexafluoride Conversion
F-OR-02-007	SRO/LWR:	Economic Studies
F-OR-02-008	SRO/LWR:	Radiological Techniques for Environmental Assessments of the LWR Fuel Cycle

SUMMARY

This quarterly report is the second in a series to be issued on the LWR Fuel Reprocessing and Recycle Program at ORNL as part of the national program administered by Savannah River Operations - Savannah River Laboratory.

Several more dissolutions were made using irradiated fuel from the H. B. Robinson II reactor. Characterization of the solids and the resulting solutions is in progress and will be reported in the next quarterly.

Requests for bids have been issued for the design and fabrication of a model 1/2 ton-per-day voloxidizer.

Two mixer-settler units have been installed in the laboratory, and reference flowsheet studies using synthetic solutions have been initiated.

The Fluorocarbon Absorption Studies program was expanded during this quarter to include acceleration of the pilot-plant development, the initiation of plant engineering design criteria, continuance of a system reliability analysis, and a study of the chemical effects of impurities in the fluorocarbon solvent on the process and equipment.

Calculations have been made to provide mass balance data for a three-region PWR through successive recycle of self-generated plutonium.

1. ASSISTANCE IN SHEAR DEVELOPMENT AND RELATED PROBLEMS

C. D. Watson (Chemical Technology Division, ORNL)

The LWR reprocessing head-end was discussed with commercial (or potential commercial) reprocessors to identify problem areas requiring the development of new or advanced technology. These problem areas are being reviewed in a program scope now in rough draft. In general, the work areas fell into the following categories: licensing problems, mechanical problems, and safety and hazard problems.

Licensing problems. The overriding thought in all the discussions was that the shear development and related problems research and development program should be conducted in such a manner that the resulting technology would provide the appropriate necessary support for the licensing of later commercial equipment and attendant facilities.

Mechanical problems. There was basic agreement that a new mechanical shear system and a materials transport system need to be developed and tested extensively. A simpler alternative to shearing is also acceptable if a superior competitive method(s) can be developed.

Safety and hazard problems. Interlaced throughout the interview sessions were a variety of "catch-all" problems associated with safety and hazards considerations. In general, these could be lumped together as follows: (1) Zircaloy safety, (2) criticality, (3) fire fighting, (4) accountability, (5) heat dissipation, (6) seismic analysis, (7) containment of shear off-gas particulates and volatile fission products, (8) the possibility of a thermite-type reaction between Zirconium and UO_2, and (9) facility containment.

The submittal of a detailed program scope is scheduled for July 1, 1976.

2. HEAD-END STUDIES: VOLOXIDATION AND DISSOLUTION

2.1 Voloxidation

D. C. Hampson (Chemical Technology Division, ORNL)

The voloxidation process is being developed as a head-end processing method for removing tritium from the fuel prior to aqueous processing. Based on experimental work, it appears that this objective can be met by heating the oxide fuel to about 450°C in flowing oxygen or air. Volatilization of tritium and, to a lesser extent, some of the other fission products is effected as the UO_2 becomes restructured during oxidation to U_3O_8. Early removal of tritium from the fuel into a relatively small volume is desirable to avoid extensive dilution of the tritium with water in the subsequent fuel dissolution step.

2.1.1 Experimental engineering studies

W. S. Groenier and C. H. Brown (Chemical Technology Division, ORNL)

The type of equipment being considered for voloxidation is a rotary kiln to which the sheared fuel is fed continuously and oxidized from solid UO_2 to a freely flowing U_3O_8 powder. During the oxidation and pulverization, a large fraction of the tritium should be released to a filtered off-gas system.

Residence time--distribution studies were performed in a 6.5-in. (0.17-m)-diam x 7.5-ft (2.29-m)-long rotary kiln manufactured by the C. E. Bartlett and Snow Company. This experimental apparatus has been described previously.[1] Heat-transfer tests using this kiln are currently being run.

Experimental procedure. In order to accurately predict kiln performance for design purposes and to interpret heat-transfer data, it is necessary to know the residence time distribution of process material in the kiln. The kiln could be characterized as a plug flow system if all particles had the same residence time. In actual practice, however, significant deviations from idealized plug flow occur, and there is a distribution or range of particle residence times. This residence time distribution can be compared to axial dispersion occurring in a tubular gas-liquid or liquid-liquid reactor.

The experimental approach used to measure residence time distributions is similar to that described by Levenspiel.[2] While the kiln was operating at steady-state conditions (i.e., a steady flow rate of material in and out of the kiln) with untagged material, the feed was suddenly changed to material which had been tagged. The product stream from the kiln was then monitored and analyzed for the fraction of tagged particles as a function of time.

Ten tests were performed by using this technique. A summary of the experimental conditions for each of these tests is given in Table 1. With one exception, all of the runs were made with a solids flow rate of 100 g/min (0.00167 kg/sec). Run 21 was performed with a flow rate of 200 g/min (0.0334 kg/sec). The feed material used in all of the tests was 1/4-in. (0.00635-m)-diam, 1-in. (0.00254-m)-long porcelain, clad with stainless steel tubing. The tagged feed material for the stimulus-response tests was prepared by applying several coats of red spray lacquer. Tests were made at each of three kiln slopes of 0.28 in./ft (0.024 m/m), 0.159 in./ft (0.0132 m/m), and 0.0965 in./ft (0.00804 m/m).

Table 1. Experimental conditions for the residence time--distribution tests
Nominal solids flow rate = 100 g/min (0.00167 kg/sec)

Run number	Kiln slope		Kiln rotational speed	
	in./ft	m/m	rpm	rps
15	0.28	0.024	5	0.083
16	0.28	0.024	8	0.13
17	0.28	0.024	2	0.033
18	0.159	0.0132	8	0.13
19	0.159	0.0132	5	0.083
20	0.159	0.0132	2	0.033
21[a]	0.159	0.0132	8	0.13
22	0.0965	0.0804	8	0.13
23	0.0965	0.00804	5	0.083
24	0.0965	0.00804	2	0.033

[a]Solids flow rate was 200 g/min (0.00334 kg/sec).

At each kiln slope, either one or two tests were made at three different kiln rotational speeds: 2, 5, and 8 rpm (0.033, 0.083, and 0.13 rps). After each run was performed, the total bed mass was determined by emptying the kiln and weighing the contents.

Experimental results. The residence time distribution data obtained in the ten experimental tests can be represented concisely in a single graph by plotting the mass fraction of red pieces in the product as a function of the reduced effective time. Let the mean residence time of material in the kiln be defined as

$$\bar{t} = \frac{V_T}{q} , \tag{1}$$

where

$\bar{t}$ = mean residence time, sec,

V_T = total bed mass, kg,

q = solids mass flow rate, kg/sec.

Then the reduced effective time is given by:

$$\theta = \frac{t}{\bar{t}} , \tag{2}$$

where

t = time since the step change in feed, sec,

θ = reduced effective time.

The data obtained in the runs are presented in Fig. 1, which is a plot of the mass fraction of red pieces in the kiln product as a function of the reduced effective time. These data show that, for the parameters varied in these tests, a given mean residence time results in the

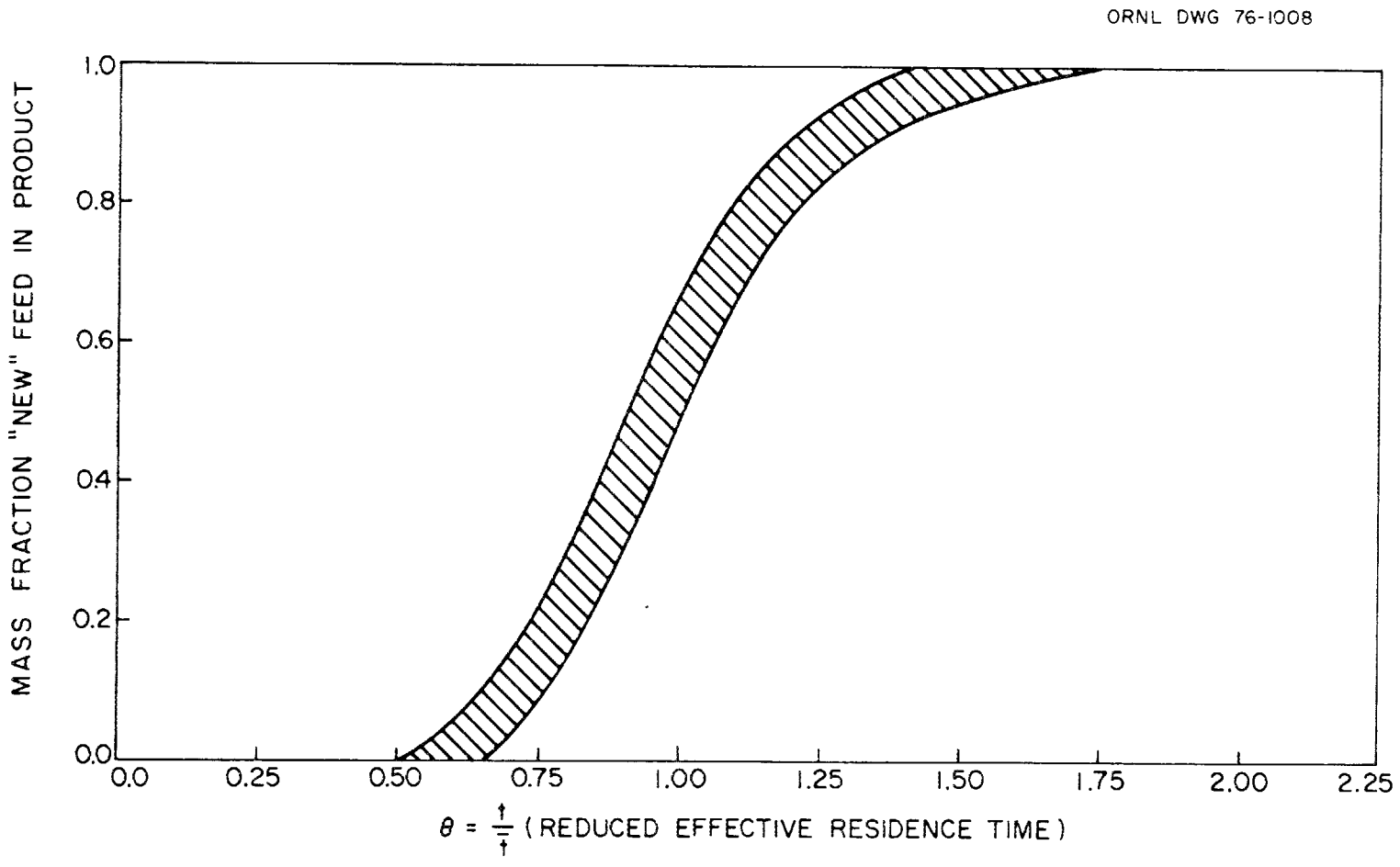

Fig. 1. Residence time--distribution data obtained from all the experiments in the 6.5-in. (0.17-m)-diam x 7.5-ft (2.29-m)-long rotary kiln.

following range of residence times of particles in the kiln:

$$0.5\ \bar{t} \leq t_R \leq 1.75\ \bar{t}\ , \tag{3}$$

where

t_R = residence time of material in the kiln, sec.

Conclusions. It can be concluded that for the variables tested (kiln slope, kiln rotational speed, and solids flow rate), the residence time distribution curve for solids in the kiln can be compactly represented in one curve as shown in Fig. 1. The objectives of this work are to determine the amount of dispersion that occurs in a rotary kiln and to ascertain what steps can be taken to minimize the dispersion which takes place. The variables that will be tested in the future are: (1) kiln diameter, (2) kiln length, (3) size distribution of the solids, and (4) mixing flight configuration. It appears qualitatively that the first two of these will not have any effect on the amount of dispersion which occurs in the kiln.

2.1.2 Component development

D. C. Hampson and E. L. Nicholson (Chemical Technology Division, ORNL)

The requirements for the larger-scale experimental rotary kiln have been discussed with representatives of commercial kiln manufacturers. A request for bids for the design and fabrication of this unit has been issued. This unit will be sized to handle 1/2 ton of fuel per day and will be more flexible, both in heating capability and in instrumentation, than would be required in a production-type unit.

2.2 Hot-Cell Studies

D. O. Campbell and S. R. Buxton (Chemical Technology Division, ORNL)

An experimental program centered around hot-cell tests of the high-radiation-level fuel reprocessing operations was initiated in January 1976 and continued throughout this quarter. This work is supported by "cold" tests with synthetic solutions having compositions. Approximately one complete hot run is conducted each month, with each experiment including most or all of the following operations: fuel dissolution, rinsing, uranium and plutonium extraction, solution clarification, raffinate (waste) concentration, and waste processing. Problems which receive special attention include the volatilization and distribution of ^{3}H, ^{14}C, and ^{129}I during dissolution, the characterization of insoluble material in the fuel and also of precipitates which may form subsequently during processing, the extractability of plutonium, and solution stability.

Four experiments have been completed, but the progress of the work is presently decelerating because of limited personnel assignments. Although much of the analytical data are not complete, results thus far suggest several significant but tentative conclusions. These conclusions are discussed in the paragraphs that follow.

Dissolution of fully irradiated fuel under conditions approximating projected operations at Allied-General Nuclear Services (AGNS) is generally satisfactory; however, a somewhat longer dissolution time will be necessary.

Approximately 0.2 to 0.3 wt % of the irradiated fuel is insoluble during dissolution in nitric acid. This residue, which is extremely

radioactive due to fission product ruthenium, may contain up to a few tenths percent of the plutonium and the other actinides. Leaching with nitric acid reduces the actinide content of the residue to a few thousandths percent, although the bulk of the residue remains insoluble. In full-scale reprocessing, this material will require attention because of its heat generation properties.

The fraction of ^{129}I remaining in the dissolver solution may be reduced to less than 1% of the amount present. Nitrogen oxides produced during dissolution remove about 98% of the iodine, and air sparging either during or following dissolution will transfer additional iodine to the off-gas.

Incomplete data for ^{14}C indicate that most of the carbon reports to the first caustic scrubber in the off-gas system. There is no evidence for organic carbon in the off-gas. The total amount of ^{14}C found is consistent with a nitrogen content in the fuel of about 10 ppm, which is reasonable.

The dissolver solution is probably stable to formation of the insoluble zirconium-molybdenum compound for several days, provided the temperature is not too high or the acidity too low. This compound would carry a significant fraction of the plutonium if it did precipitate prior to extraction.

Solids are more readily produced when the extraction raffinate is aged. Well-crystallized solids form slowly from 3 to 4 $\underline{M}$ HNO_3 raffinate, and x-ray diffraction shows them to have the same structure as the zirconium-molybdenum compound prepared previously in "cold" work. These solids, which are highly enriched in ^{125}Sb, carry several percent of the plutonium present in the raffinate.

No significant problem has been encountered in regard to plutonium extractability. Batch extractions were carried out a few days after dissolution with no valence adjustment. In tests made this quarter, the distribution coefficient for plutonium did not decrease during extraction of at least 99.9% of the plutonium.

Evaporation of the extraction raffinate at 7 to 10 M HNO_3 concentration resulted in the appearance of solids at a waste concentration of about 325 gal per metric ton of fuel, a significantly lower concentration than has been achieved with synthetic waste. The solids are not yet characterized. The concentrated waste supernate solution has been stable for over 1 month.

2.2.1 Hot-cell dissolution

Two additional dissolution experiments have been completed with fuel, discharged in May 1974 from the H. B. Robinson reactor (Carolina Power and Light Company). Clad fuel was used in Run III to study the effect of cladding on dissolution rate, solids holdup, zirconium content of the dissolver solution, hull rinsing, and other factors. In Run IV, no air sparge was used during the period of active dissolution, and a hot CuO bed was inserted in the off-gas system to eliminate the possibility of losses via volatile organic compounds.

Clad fuel from the upper 18-in. section of Rod P-r of Assembly B05 was used in Run III. The rod was cut into nine approximately equal pieces, and about 90% of the fuel remained in the cladding. Burnup varied from about 7400 MWd/metric ton at the upper end to 26,600 MWd/metric ton at the inner end; analytical data indicate that the average was about 22,000 MWd/metric ton.

Dissolution progressed smoothly but was somewhat slower than with unclad UO_2 fragments. The sequence included dissolution for 1 hr after the last fuel addition, rinsing of the dissolver and hulls twice with 3 M HNO_3 at cell temperature, secondary dissolution for 150 min with 3 M HNO_3 at 90°C, rinsing again, and finally a tertiary dissolution for 4 hr with 7 M HNO_3 at 90°C (Table 2). The secondary dissolution recovered approximately 0.5% of the uranium, plutonium, americium, and curium, showing that the 1-hr dissolution time was inadequate. The tertiary dissolution removed less than 10^{-3}% of these actinides, indicating that dissolution was complete. However, the decrease in concentration with successive rinsing was less than expected for simple rinsing, suggesting that a very small amount of material was either being dissolved or desorbed from surfaces at these low concentrations.

A 2-in.-long piece of Zircaloy cladding was dissolved in hydrofluoric acid, leaving an appreciable amount of insoluble residue. The residue was leached with hydrochloric acid and with nitric acid, and finally dissolved by fusion in Na_2CO_3. Analyses of all these solutions indicate that the quantities of actinides associated with the cladding are acceptable low (about 2×10^{-3}%), and that the gamma activity associated with cladding consists of a number of isotopes including, in decreasing order, ^{125}Sb, ^{106}Ru-Rh, ^{144}Ce, ^{137}Cs, ^{134}Cs, ^{60}Co, ^{95}Zr-Nb, and ^{54}Mn.

The fourth dissolution experiment (Run IV) was completed using a larger quantity of UO_2 fuel fragments (380 g) from the high-burnup, central portion of rod A-1 of the Robinson reactor. Fuel was added in three increments, with conditions approximating the AGNS flowsheet except that no air sparge was used during dissolution.

Table 2. Rinsing and secondary dissolution of hulls

HNO_3 (M)	Time (min)	Temp (°C)	U (mg/ml)	$^{239-240}Pu$ (count min^{-1} ml^{-1})	^{244}Cm (counts min^{-1} ml^{-1})	Relative dissolution (%)
3.45	Dis.[a]	90	322.6	9.64×10^7	1.51×10^8	100
3.0	70	35	1.80	3.46×10^5	5.42×10^5	0.4
3.0	30	30	0.087	8×10^3	5.78×10^4	0.03
3.0	150	90	1.49	5.13×10^5	7.82×10^5	0.5
3.0	30	30	0.015	3.9×10^3	7.16×10^3	0.005
7.0	240	90	0.002	5.9×10^2	9.62×10^2	0.0006

[a]Total time for dissolution, 140 min; 60 min after last fuel addition. Volume of dissolver solution, 720 ml; all others, 800 ml.

One purpose of this experiment was to obtain more definitive data for ^{129}I and ^{14}C. The equipment was modified by inserting a bed of CuO at 750°C in the off-gas train between the two NaOH scrubbers to ensure that no volatile organic material escaped via the off-gas. Mock-up tests in the laboratory in the absence of radioactive materials showed that such an oxidizing bed not only would oxidize organic material to CO_2 and H_2O, but also would convert organic iodides to iodine species which are trapped in the caustic scrubber.

The dissolution procedure was also modified by eliminating the air sparge during dissolution. The dissolver was maintained at a vacuum of about 3 in. H_2O, and air was drawn into the off-gas immediately downstream from the condenser to sweep evolved gases through the scrubbers. Approximately 50 min after the final fuel addition, dissolution appeared to be complete as indicated by a cessation of bubbling in the dissolver. The solution was sampled at this time, and an air sparge at the rate of 0.4 dissolver solution volume per minute was initiated. The solution was sampled after air sparging, for 70, 140, and 200 min at 85 to 90°C, to determine the rate of ^{129}I removal. Off-gas left the dissolver vessel at about 60°C; thus was considerable reflux within the dissolver.

2.2.2 Radioactivity in off-gas

Analytical results for ^{129}I and ^{3}H are now available for most solutions of interest; ^{14}C results are available for about half the solutions. The data appear reasonable, for the most part, and support the contention that analytical methods will be adequate for these determinations.

Data for ^{129}I in Run III agree, in general, with previously reported data for the first two runs, in that about 0.5% of the iodine found was left in the dissolver solution. However, the material balance was lower, with the amount of ^{129}I found being only about half the amount calculated for this fuel. Since this was an end piece with a range of burnups, the calculated amount of iodine is subject to some question, and it is conceivable that some iodine migrated away from this zone to a hotter region.

Run IV was modified to yield more definitive data for ^{129}I, as described in the previous section. The iodine content in the dissolver solution decreased steadily with air sparging (Table 3). It has not yet been possible to make a material balance because iodine analyses for the scrubbers will not be available for some time. The calculated amount of ^{129}I present is about 50 mg; therefore, the results indicate that about 98% was swept out of the dissolver solution by NO_x during dissolution and that air sparging reduced the iodine left in the dissolver to values as low as a few tenths percent.

Table 3. Effect of air sparging on ^{129}I content of dissolver solution in Run IV

Time of air sparge (min)	^{129}I remaining in solution[a] (mg)
0	0.87 ± 0.074
70	0.245 ± 0.050
140	0.138 ± 0.017
200	0.096 ± 0.011

[a]Average of duplicate determinations ± standard deviation.

All data obtained thus far clearly demonstrate that iodine is effectively evolved into the off-gas during dissolution under these conditions. The primary remaining uncertainty is whether or not some organic material or other impurity present in recycle solutions might prevent the volatilization of iodine. This would be a function of the acid recycle system rather than of the basic chemistry of the process. Volatile organic iodide would be found in this case.

Tritium analyses have been obtained for all four runs (Table 4). The distribution of ^{3}H is reasonably consistent with water transport through the system if it is assumed that ^{3}H appears initially as water in the dissolver solution as the fuel dissolves. However, the increase in ^{3}H in the second scrubber in Run IV suggests that about 0.5% of the ^{3}H originated as hydrogen gas which was oxidized to water in the CuO bed between the two scrubbers. Such an increase in concentration in the second scrubber, compared with the first, was not observed for any other activity, including ^{14}C and ^{129}I.

As is usually the case with ^{3}H, the material balances are low compared with the theoretical value calculated by ORIGEN.* The burnup for Run I is less certain, and the three runs with Robinson reactor fuel give recoveries of greater than 50% of the fission product tritium. A more accurate estimate of burnup will be possible when more complete fission product data are obtained.

Although some data have been obtained for ^{14}C, none are yet available for the dissolver solution. An analytical development program is being carried out to eliminate certain questions about the possibility of the

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* The ORIGEN code is described in ORNL-4628.

Table 4. Tritium behavior during LWR fuel dissolution

Run Number	Total ^{3}H found in solution (dis/min)			
	I	II	III	IV[a]
Dissolver solution	7.18×10^{10}	1.18×10^{11}	1.35×10^{11}	2.17×10^{11}
Condensate	8.90×10^{8}	1.56×10^{9}	1.39×10^{9}	6.15×10^{9}
First NaOH scrubber	3.06×10^{8}	1.38×10^{8}	5.32×10^{7}	3.77×10^{8}
Second NaOH scrubber	6.72×10^{5}	3.43×10^{5}	8.84×10^{6}	1.05×10^{9}[a]
Total	7.30×10^{10}	1.20×10^{11}	1.37×10^{11}	2.24×10^{11}
ORIGEN-FP[b]	$\sim 1.8 \times 10^{11}$	2.17×10^{11}	$\sim 1.76 \times 10^{11}$	3.4×10^{11}

[a]Prolonged air sparge transported more water vapor through off-gas system. Hot CuO trap was placed in off-gas train upstream of the second NaOH scrubber.

[b]Tritium is formed by fission and by neutron capture in ^{6}Li. This figure is for fission product tritium. Lithium at 2 ppm would yield an equal contribution.

existence of stable organic compounds in the dissolver solution and to provide the extremely high degree of separation from fission products required for ^{14}C determination. The same approach will also provide more definitive data for ^{3}H and ^{129}I in the dissolver solution.

Results obtained thus far appear reasonable, with about 95% of the ^{14}C being found in the first NaOH scrubber. The total amount found corresponds to that projected by ORIGEN calculation for fuel containing about 10 ppm of nitrogen. We have requested Westinghouse to provide analytical data, if available, for the nitrogen content of the UO_2 used for this assembly.

2.2.3 Insoluble dissolution residue

Approximately 0.2 to 0.3% of the weight of fully irradiated LWR fuel is not readily soluble in HNO_3; even less material is insoluble at lower burnups (less than 30,000 MWd/metric ton). Prolonged leaching with nitric acid slowly dissolves part of the residue at a decreasing rate, yielding a rose- or red-colored solution containing large amounts of ruthenium. Part of the residue is apparently not soluble in nitric acid below 8 M in concentration. As a result, the amount of residue found in practice is dependent on the severity of dissolution conditions (time, temperature, and acid concentration). There is also some indication that a larger fraction of the fuel, perhaps about 1%, may dissolve significantly more slowly than the bulk of the UO_2.

With reasonably complete dissolution, the residue contained from a few hundredths to about 0.1% of the plutonium, americium, and curium. Leaching with nitric acid under practicable conditions reduced the

actinide content of the residue to a few thousandths of a percent in all cases, even when most of the residue was not dissolved. The residue was much more soluble in concentrated hydrochloric than in nitric acid concentrations below 8 M (at higher nitric acid concentrations, ruthenium volatilization is probably unacceptable), and fusion in Na_2CO_3, followed by dissolution of the Na_2CO_3 in nitric acid, was used to dissolve the last remnants for material balance purposes.

Table 5 summarizes the results of five experiments: the first Sandia run, which was made by the Isotopes Development Laboratory, and the four subsequent dissolutions made as part of the present program. In all cases, the fuel was dissolved in nitric acid at a final temperature of about 90°C (the temperature was sometimes lower at the start of dissolution) to yield a dissolver solution 2 to 4 M in HNO_3 and about 1.4 M in $UO_2(NO_3)_2$. Attempts were made to approach steady-state conditions representative of those employed at AGNS by (1) adding the acid slowly as dissolution progressed and/or (2) adding the fuel incrementally in two or three batches, each followed by the addition of an appropriate amount of HNO_3.

The quantity of solids recovered after the initial dissolution was significantly lower in Runs III and IV. The conditions for these runs differed from those normally encountered in that the burnup was lower for Run III and the dissolution time was longer for Run IV. This sort of correlation is probably general.

The residue consists of very finely divided black solids, much of which is less than 1 μ in size. The settling rate of the finest solids, as indicated by solution clarficiation, was about 1 in./hr. The solids

Table 5. Insoluble residues from irradiated LWR fuel dissolution

Run[a]	Initial weight of UO_2 (g)	Total solids (mg)	Leach solution	Weight dissolved (mg)	Total Pu in solids (%)	Pu in leached residue (%)
IDL-Sandia[b]	667	3130	8 M HNO_3; reflux	2415	0.11	0.003
I[b]	250	556	6 M HNO_3; reflux	~60	0.037	~0.005
II[c]	240	673	6 M HNO_3; 90°C	141	0.014	~0.005
III[c]	290 clad	382	12 M HCl; 90°C	>90%	0.002	--
IV[c]	380	544	12 M HCl; 90°C	>90%	~0.004	--

[a]IDL-Sandia Run: Acid was added as dissolution progressed, and the concentration may have dropped below the desired value during part of the dissolution. Conditions were probably too mild or the time too short, since nitric acid leaching of the residue dissolved over 1 g of uranium and exactly proportionate amounts of soluble fission products such as cesium, strontium, etc.

Run I: Dissolution conditions approximated AGNS specifications. Fuel was added in two increments. The temperature was maintained near 90°C for approximately 1 hr after the last addition of UO_2.

Run II: Conditions were similar to those of Run I, except that Robinson reactor fuel was used (31,360 MWd/metric ton). The temperature was maintained at 90°C for 40 min after the second fuel addition and the dissolver was allowed to cool for 1 hr.

Run III: Conditions are described in Sect. 2.2.2. A temperature of 90°C was maintained for 1 hr after addition of the last UO_2, and the dissolver solution was transferred.

Run IV: This dissolution, described in Sect. 2.2.2, resembled Run II except that more fuel was used. It was added in three increments; and the total dissolution time was much longer.

[b]IDL-Sandia Run and Run I - Westinghouse fuel, cooled 5 to 6 years.

[c]Runs II-IV - Robinson Reactor Fuel, cooled 2 years.

will eventually settle, or they can be easily centrifuged. The gamma activity is extremely high, typically greater than 99% ^{106}Ru-Rh and reading greater than 10,000 R/hr per gram with the in-cell monitor. More complete composition data are not yet available.

Some size fractionation normally occurs during the experiment. Over 60% of the solids are usually recovered from the dissolver solution after it has been transferred out of the dissolver. The remainder, containing any larger particles which settle more rapidly, is easily washed out of the dissolver vessel during rinsing. (An exception was the Westinghouse fuel, which contained some larger particles that were not easily washed out.) The actinides associated with the solids are preferentially (>95%) in the finer particles recovered from the dissolver solution. Thus, the composition of the solids probably varies with particle size, with the smaller particles carrying most of the actinides associated with solids.

2.2.4 Plutonium extraction into 30% TBP*

In connection with studies to investigate the nature and behavior of "inextractable" plutonium in high-level fuel reprocessing and waste streams, plutonium extraction behavior was evaluated with radioactive feeds prepared from highly irradiated UO_2. In each experiment, the UO_2 dissolver solution, which was approximately 3 M in HNO_3, was batch contacted with equal volumes of 30% tributyl phosphate (TBP)--dodecane. Seven consecutive extraction stages were used.

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* Studies carried out by M. H. Lloyd as part of a program on Waste Stream Processing Studies funded by the Molecular Sciences Branch of the Division of Physical Research, ERDA.

The initial UO_2 dissolver solution was clarified by centrifugation and stored for about two weeks prior to extraction. Tests to determine plutonium valence indicate that the dissolver solution contained only Pu(IV) and that the plutonium remained quantitatively in the tetravalent state throughout the storage period and extraction cycles.

With three different feed solutions, more than 99.99% of the plutonium was extracted in seven stages; in one stripping experiment, more than 99.99% of the plutonium was recovered in five stripping stages. Phase separations were rapid in all cases, and interfacial crud formation was minimal. Plutonium extraction data for a typical run is given in Fig. 2, which shows the plutonium concentration (in counts min^{-1} ml^{-1}) remaining in the aqueous raffinate after each stage. The straight line illustrates the extraction behavior that would be obtained with a constant plutonium distribution coefficient of 14. In the first two stages, the plutonium distribution coefficient was less than 1 as a result of uranium saturation in the organic phase. After most of the uranium had been extracted, plutonium extraction was normal except for a very small amount of plutonium which appeared to be inextractable. This small plutonium loss also occurs when highly purified tetravalent plutonium in 3 $\underline{M}$ HNO_3 is extracted with TBP-dodecane. The reason for this behavior is not known; however, it appears to be some function of the extraction process itself because the "inextractable" plutonium from both the feed prepared from dissolution of high-burnup fuel and from pure nitric acid solution can be completely recovered by anion exchange processing.

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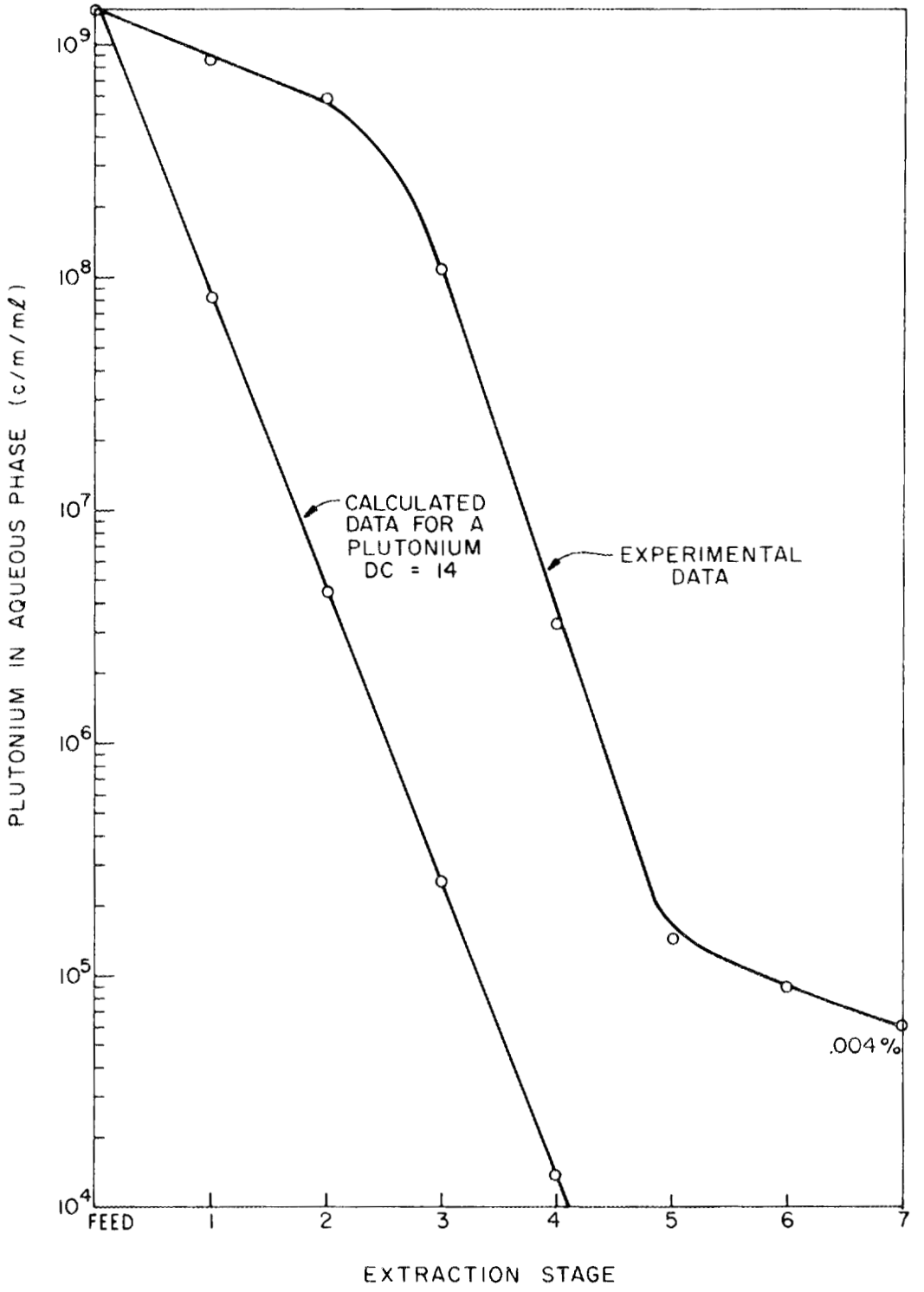

Fig. 2. Plutonium extraction behavior from feed prepared by dissolution of irradiated UO_2 feed adjusted to 3 M HNO_3 was contacted with equal volumes of 30% TBP--dodecane.

2.2.5 Solution stability

Solids appear in extraction raffinate solutions after aging at an elevated temperature. The rate of formation of such solids is dependent on the solution history, the acid concentration, and temperature during storage. The formation of solids is slower at higher acidities, and there is a greater tendency for the solids to adhere to the glass walls. Well-formed crystals have been observed in some cases. Figure 3 is an example of crystals which grew on the glass wall from a 3.7 M HNO_3 raffinate solution aged at about 80°C for 1 month.

Microprobe results show the presence of zirconium, molybdenum, and a very small amount of tellurium. Although the solid was rather radioactive, it was possible to obtain an x-ray diffraction pattern on a small sample. This clearly demonstrated that the crystals have the same structure as the zirconium-molybdenum compound which has been studied for some time and is readily prepared from nonradioactive, synthetic waste solutions.* These crystals are completely different in appearance from solids with the same structure which have been seen previously.

The gamma activity of the solid was almost entirely due to ^{125}Sb, which is a very minor contributor to the total fission product gamma activity (about 5% of the ^{137}Cs, for example). Thus, it is clear that antimony selectively follows the zirconium-molybdenum compound. Alpha analyses indicate that the precipitate also carried a few percent (less than 5%) of the plutonium present in the raffinate solution; however, less than 10^{-2}% of the curium (which is present in much greater amounts

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* X-ray diffraction studies were made by R. G. Haire, ORNL Chemistry Division.

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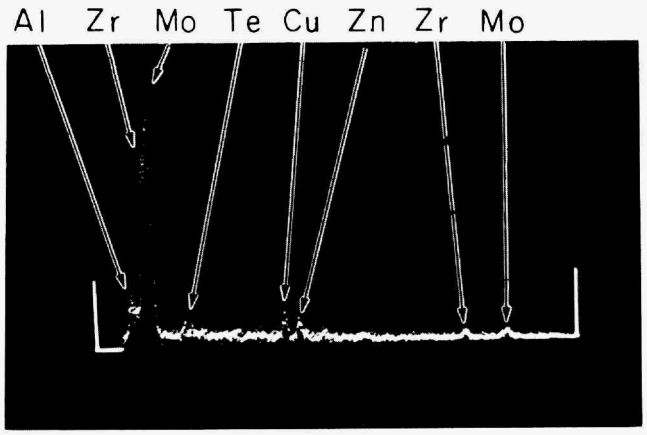

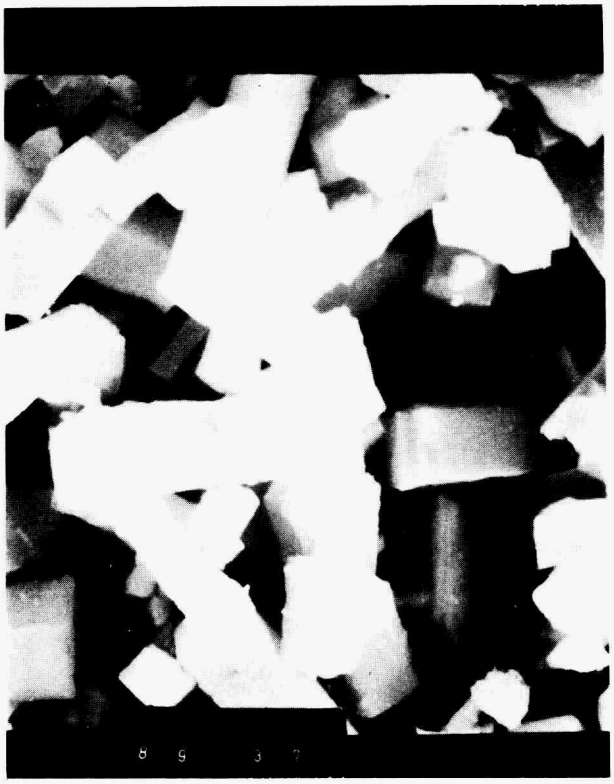

G 3000 x 10 μ

Fig. 3. Solids which formed in extraction raffinate (HAW) solution stored at approximately 80°C for 1 month.

than plutonium in this solution) followed the solid. It was previously reported that this same precipitate similarly carries a few percent of the plutonium from solutions containing much higher plutonium concentrations than the raffinate.

From the point of view of actinide losses, it is essential that formation of the zirconium-molybdenum compound be avoided until after adequate plutonium recovery has been accomplished. Both "cold" and "hot" work indicate that this should be practicable by judicious choice of storage time, temperature, and acid concentration. Fortunately, the compound apparently forms less readily in fresh dissolver solution than in aged radioactive solutions or synthetic dissolver solutions. During operations subsequent to extraction, however, it is not certain that formation of this compound can be avoided (i.e., during waste storage or waste evaporation). This problem could cause operational difficulties if not recognized.

Waste concentration. A portion of the extraction raffinate from Runs II and IV was concentrated in an evaporator, using a procedure (developed for synthetic waste studies) which minimized formation of zirconium-molybdenum compound. Concentrated nitric acid was added to a fraction of the raffinate to yield an acid concentration of about 8 *M*, and the solution was then concentrated by boiling in an evaporator immersed in silicone oil at 130°C so that no refluxing could occur. Additional raffinate was added as condensate was collected. As evaporation proceeded, the acidity of the still pot remained in the range

8 to 10 M and the fission product concentration increased. The acidity of the condensate increased to about 3 M near the end of the evaporation.

In each experiment, a precipitate appeared in the solution after concentration, by a factor of 2 to 2.5, to about 300 to 350 gal per metric ton of fuel. Evaporation was continued to final concentrations of 140 and 96 gal/metric ton for Run II and Run IV raffinate respectively. Substantial amounts of precipitate, much of which adhered to the glass walls of the evaporator, were found to be present at the end of the evaporation step. The amount of solids recovered from Run IV was 2.3 g per liter of raffinate, and additional solids were on the glass walls.

Few data for characterization of these solids have been obtained to date. Microprobe results indicate that lead is the predominant metal present in the solids, but this has not been confirmed. The source of the lead is not known. Evaporation of synthetic waste containing added lead results in precipitation of a mixed phase of barium, strontium, and lead nitrates; all of these metal nitrates are insoluble in high concentrations of nitric acid and have the same crystal structure.

The supernate from the concentrated raffinate from Run II was a clear, red solution which appeared to be stable for long perios of time (greater than 1 month) at room temperature. The acid concentration of the solution (9.6 M HNO_3) did not change appreciably during storage for 1 month. However, subsequent air sparging of the solution for 7 hr reduced the acidity to 9.1 M HNO_3. A more careful study of the effect of air sparging on acid loss will be made with the Run IV concentrate since this rate of acid loss could be quite important during long periods of waste storage.

It appears that raffinate (waste) storage and waste concentration, whether for storage or as a preliminary step in waste solidification, include some potential problem areas which have not been adequately recognized.

2.2.6 Characterization methods

The status regarding analytical data has changed only slightly. As more results are obtained, the limitations of some of the data become more visible and several areas which require more work are identified. Little improvement has been achieved to date. Certain deficiencies are discussed here.

Mass analysis for plutonium for fully irradiated Robinson reactor fuel (Run IV) has been completed, and there are significant discrepancies from the ORIGEN calculation for that particular irradiation history (Table 6). Calculated values for the heavier isotopes appear to be low.

Table 6. Plutonium isotopic composition of Robinson fuel

Isotope	Composition (wt %)	
	Found in Run IV	Calculated by ORIGEN Code
^{238}Pu	1.94	1.72
^{239}Pu	55.55	60.6
^{240}Pu	24.83	24.1
^{241}Pu	12.23	10.1
^{242}Pu	5.45	3.55
^{244}Pu	0.0005	--

Alpha pulse-height analysis with alpha counting is the simplest method for determining many actinides. Unfortunately, this method appears to have inherent errors which cause lower-energy alpha peaks to be larger than they should be. For example, the 5.15-MeV peaks of $^{239,240}Pu$ typically contribute 25% and the 5.48-MeV peak of ^{238}Pu contributes 75% of the total pultonium alpha activity; but the mass analysis corresponds to 22% and 78% for the two peaks.

Scanning electron microscopy (SEM) and microprobe results are being obtained rather slowly. The SEM photos are spectacular in some cases, while microprobe results are less satisfactory, largely because of problems in interpretation. For example, K x-rays of light elements common as impurities and M x-rays of lead interfere with L x-rays of important fission products. As a result, ambiguities have been noted in much of the x-ray microprobe data. In addition, many samples are too radioactive to permit the x-ray spectra to be measured. There are possible ways to improve the situation, but little progress has been achieved so far.

Only a few sets of data have been obtained from spark-source mass spectrometry (SSMS) analysis, and these indicate that much improvement is necessary. The method has the advantage of being general for all elements present in appreciable amount; however, as presently applied, it also has severe limitations. In particular, accuracy appears to be in the range of ±50% for most isotopes, and the range of concentration which can be measured is narrower than desired.

Work will continue on development of procedures which result in more accurate and useful analytical data. In addition, efforts will be made to decrease the time required to obtain analytical results, particularly for SSMS and SEM x-ray fluorescence measurements.

2.3 Building 4507 Hot-Cell Operations

J. H. Goode, R. G. Stacy, V. C. A. Vaughen, E. C. Hendren and O. L. Kirkland (Chemical Technology Division, ORNL)

The major effort continues to be cleanout of the hot cell (Sect. 2.3.2). Fuel procurement and testing and modification of in-cell equipment are also being accomplished as time permits.

2.3.1 Development of in-cell equipment

Intermediate-scale fuel shear tests have been made on unirradiated stainless-steel-clad (0.34 in. OD; clad thickness, 0.021 in.) and on Zircaloy-clad (0.5 in. OD) UO_2 rods. Shear pressures less than 700 psig were sufficient to chop this material into 1-, 1-1/2-, and 1-3/4-in.-long pieces with minimal deformation (less than 25% closure of the ends). The effect of cut length on the amount of UO_2 dislodged was measured by collectively weighing and sizing the loose fuel from each series of cuts. The results are shown below.

Cut length (in.)	Fuel dislodged (g/cut, avg.)	Fuel dislodged (% of total fuel)
1	1.04	9.6
1.5	1.21	7.3
1.75	0.87	4.4

The size distribution for the loose fuel was: 15 wt % < 75 μm; 75 μm ≤ 20 wt % < 150 μm; 150 μm ≤ 30 wt % < 500 μm; and 30 wt % ≥500 μm.

The 100-g-scale voloxidizer, which was received with damaged seals, has been repaired. The unit has been put into operation under process operating conditions for further calibration and endurance testing. Deposition temperature profile measurements in the region 400 to 500°C are under way.

The stirred dissolver has shown a tendency for the paddles to stall when loaded with sheared fuel and cladding. A removable, rotating basket has been fabricated to replace the paddles. The unit is being assembled for further testing.

Progress on the development of the continuous tritium monitor has been reported elsewhere.[3]

2.3.2 Decommissioning the Curium Recovery Facility

Cells 3 and 4 were entered to cap open tubing and pipe prior to sandblasting and to confirm true dimensions needed for the fabrication of new floor pans, support structures, and working surfaces. The interior of the Equipment Maintenance Box was spray-painted to fix residual loose contamination, and the windows were taped and covered with plywood in preparation for removal of the glove box in late June. A radiation survey showed little or no contamination between the Penthouse floor and the bottom of the glove box. The process water monitor sleeve extending through the roof of Cell 4 and the americium target chute extending through the roof of Cell 3 were bagged-out and sent to Solid Waste Storage. The design of pedestals for joining shipping casks to "slug chutes" for Cells 3 and 4 is in progress.

2.3.3 Miscellaneous

The modified skid for the Garden Carrier No. 2 (AEC-OR-USA-6088/BLF) was delivered, and the cask was shipped to Battelle-Columbus Laboratories to pick up a second shipment of irradiated H. B. Robinson reactor fuel (UO_2 in Zircaloy). The fire shield for the Garden Carrier No. 2 is awaiting final painting.

A work order was issued for fabrication of cans for storing excess irradiated fuel samples and a model concrete shielding plug for the Hot Garden storage wells. A work order was also issued to fabricate cell exhaust filter housings, structural supports for a work surface, and a fuel storage array for Cell 3. The completed Cell 2-3 intercell transfer assembly was delivered from the Central Shops.

The stainless steel fire barriers in the cell ventilation duct at Building 4556 were replaced, and the roughing and HEPA filters were bagged-out and replaced; DOP-testing indicated that the efficiency of the new filters was satisfactory.

3. PUREX PROCESS STUDIES (SOLVENT EXTRACTION)

B. L. Vondra (Chemical Technology Division, ORNL)

This program involves the integration of laboratory development studies, flowsheet tests employing synthetic solutions, and confirmatory investigations in a hot-cell system using irradiated LWR fuels.

3.1 Laboratory Studies

3.1.1 Solvent cleanup

O. K. Tallent and Karen Williams (Chemical Technology Division, ORNL)

The solvent from the Purex process is presently treated with sodium carbonate and sodium hydroxide to remove impurities before it is reused. However, the lack of complete effectiveness of such caustic wash methods, along with attendant reagent recycle problems, has lead us to investigate alternative solvent cleanup methods.

An experiment is currently in progress to study the utility of macroreticular resins for removing small quantities of plutonium complexed with di-n-butyl phosphate [DBP] from 30% TBP--70% dodecane solvents. Conditions for the portion of this experiment that has been completed thus far may be described as follows: Solvent samples were first prepared by adding appropriate microliter volumes of a 0.045 or 0.0045 M DBP stock solution to 30% TBP--70% dodecane solvent in order to obtain solutions ranging from 4.2×10^{-5} to 26.2×10^{-5} M in DBP. Next, 75 μl of 0.153 M Pu(IV)--12.3 M HNO_3 stock solution was added to 10-ml volumes of each of the samples. The plutonium-bearing organic samples were then equilibrated

with three equal volumes of 0.01 M HNO_3 aqueous solution. The plutonium content retained in the organic phase after the third wash was determined from gross alpha pulse-height analyses.

The data obtained are plotted in a log-log plot of retained plutonium concentration vs original DBP concentration in Fig. 4. Data reported previously by other investigators[4] for higher concentrations of DBP in 20% TBP-dodecane as a result of ^{60}Co gamma irradiation are also plotted for comparison. Both sets of data are described well by the equation

$$\ln [Pu] = 1.00 \ln [DBP] + \ln [0.54 \times 10^{-5}],$$

where [Pu] denotes molar concentration of retained Pu(IV) and [DBP] denotes initial DBP concentration. The results show that the retained plutonium concentration is directly proportional to the DBP concentration and that plutonium retention in the irradiated solvent can be closely duplicated by adding the same amount of DBP to unirradiated solvent as that formed by TBP degradation in the irradiated solvent.

The solvent samples from this experiment will be equilibrated with macroreticular Amberlyst A-29 resin next quarter. Solvents containing other cations [Zr^{4+}, UO_2^{2+}, etc.] and other solvent degradation products will be included in resin tests (batch and colum) in future work.

3.1.2 Plutonium valence adjustment

M. R. Bennett (Chemistry Division, ORNL)

In solvent extraction and solvent stripping steps, the valence of the plutonium is repeatedly adjusted from 6 to 4, 4 to 3, and 3 to 4. The adjustment of the plutonium valence to 4 from either 3 or 6 has commonly been done by addition of sodium nitrite; however, the addition of sodium

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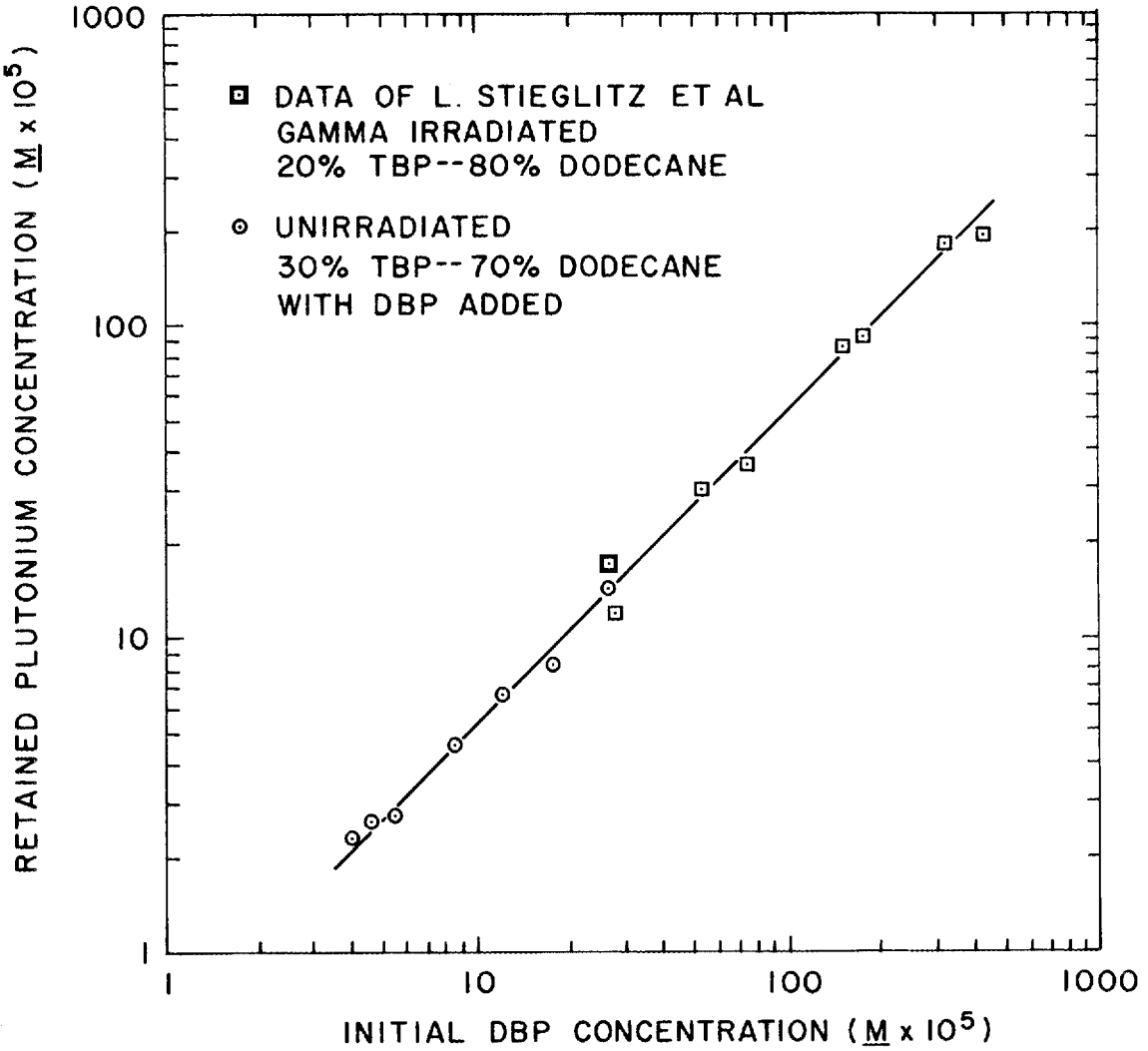

Fig. 4. Retained plutonium concentration as a function of DBP concentration in irradiated and in unirradiated TBP--dodecane.

ion has disadvantages since it increases the waste volume and can adversely affect waste solidification. A virtually uncontaminated nitrogen oxide stream from product denitration will be available in a reprocessing plant, and use of this gas stream for the plutonium valence adjustment steps would avoid introduction of an additional chemical into the plant. The composition of this stream, after condensation of water and nitric acid, can vary from nearly pure NO to a mixture of NO_2 and air, depending on the operation of the denitration step.

A system to supply typical denitration gas mixtures is presently being installed. The 6, 4, and 3 valences of plutonium will be monitored via a Cary Model 14 spectrophotometer during experimental valence adjustment steps.

3.2 Purex Studies with Irradiated Fuels

V. C. A. Vaughen (Chemical Technology Division, ORNL)

3.2.1 Purex process studies

H. C. Savage (Chemical Technology Division, ORNL)

The Purex process is used to recover, decontaminate, and separate uranium and plutonium from irradiated uranium fuel. It includes all the solvent extraction steps between the head-end reprocessing and the product finishing operations. In the reference* Purex process flowsheet, 30% TBP coextracts uranium and plutonium from 3 M HNO_3 in the first column, the plutonium is selectively stripped from the TBP in the second column (by a valence change), and the uranium is stripped into dilute HNO_3 in the

* The reference Purex flowsheet is shown in Fig. 4.1-2, AGNS Plant FSAR.

third column. While the Purex solvent extraction process has been successfully used for years with fuels of low burnup, significant process changes will be required for fully irradiated LWR fuels due to the increased inventories of fission products and higher plutonium contents.

A program was initiated last quarter to develop and test a Purex flowsheet to process high-burnup LWR fuels on a modest scale. We will construct a small-scale Purex solvent extraction system in a hot cell to test and demonstrate the proposed flowsheet and flowsheet modifications for improved process performance, and to study other problem areas in processing these fuels.

The system of first choice consists of three banks of mixer-settlers of the type used in the Curium Recovery Facility (CRF).* These mixer-settlers have been proved in extensive hot operation and are physically large enough to sample individual stages with manipulators. They are easy to clean out remotely, if necessary, because all of the settlers face one side and are open to the top. Each stage volume is about 60 ml. Two of these units are installed in a hood for cold testing (see Sect. 3.4).

An alternative system would be comprised of mixer-settlers of the SRL type, which have individual stage volumes of about 16 ml. The SRL units are based on the Knolls Atomic Power Laboratory (KAPL) design. These units have been extensively tested at SRL in shielded glove-box operations but are designed so that settlers from every other stage face on the rear side, where direct manipulator access is not possible. They require less solution volumes than the CRF models.

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* These are enlarged, modified units based on the Belgonucleaire models.

We are evaluating the design parameters of run times and solution volume requirements based on SEPHIS code predictions (see Sect. 3.3.2) for the reference flowsheet (as adapted to three banks of 16-stage mixer-settlers). Preliminary results of these calculations indicate that run times of about 20 to 30 hr may be necessary to attain 95% of the steady-state stream composition for the plutonium product stream (IBP) using CRF contactors. However, to attain 99% of the steady-state values for this stream would require about 85 hr of operation. Estimates of run times to steady state for the SRL contactors are more than twice as long. These projected run times are subject to change as alternative modes of operation are considered.

At the present time, the CRF contactors remain the first choice; these contactors have predicted run times of about 1 to 2 days (to 95% conditions above) and fuel requirements of 2 to 4 kg per run.

3.2.2 Solvent extraction flowsheet development

R. H. Rainey (Chemical Technology Division, ORNL)

The flowsheet published in the FSAR for the AGNS plant is an equipment diagram and specifies only the capacity of 5 tons/day. To provide a basis for the design of the hot-cell solvent extraction system, a set of optimized operating conditions was calculated for a reference flowsheet using the SEPHIS code. These values are presented below and in Fig. 5.

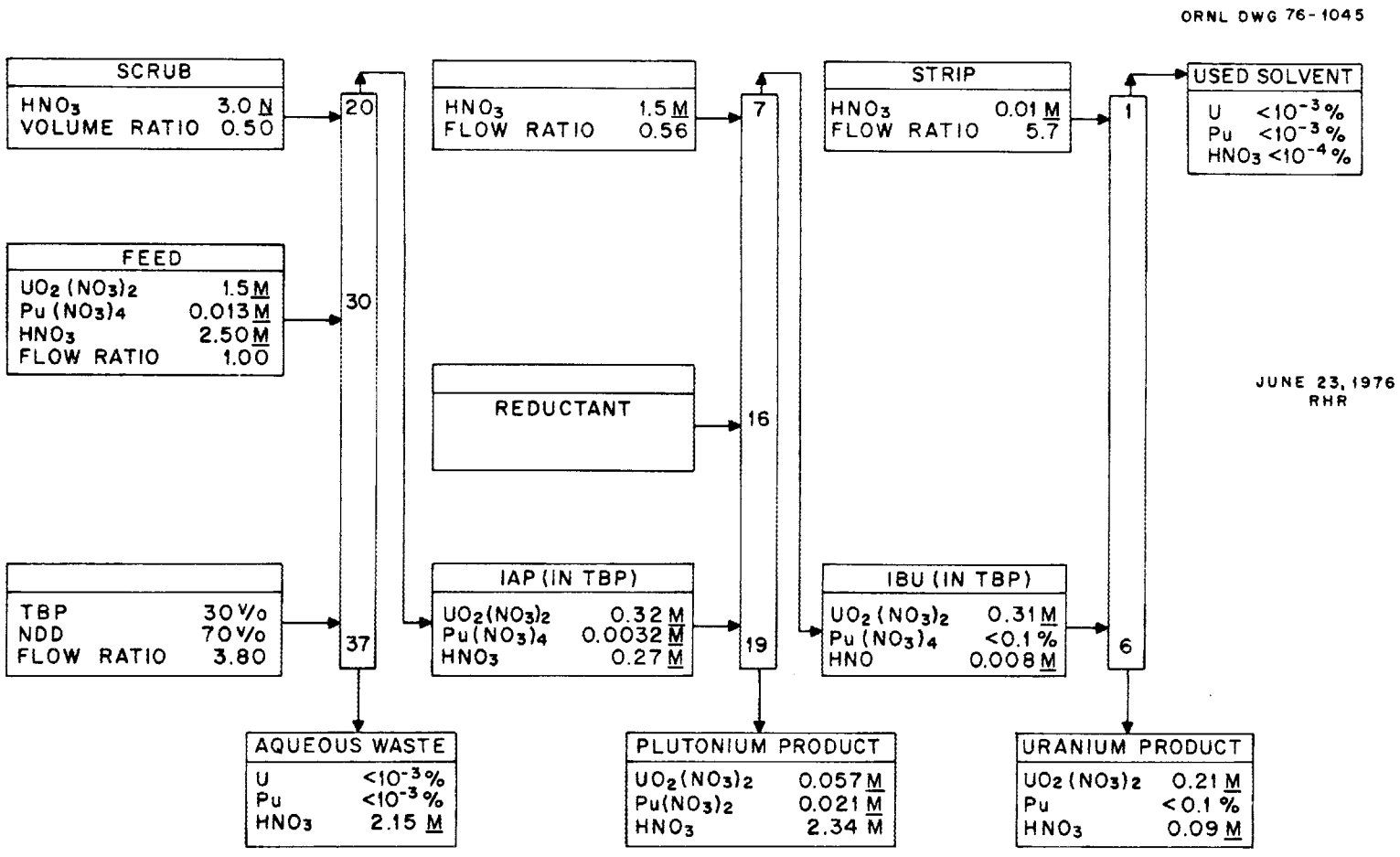

Fig. 5. LWR flowsheet based on Barnwell principles but not non-proprietary, as of June 1, 1976.

Stage number	Entering stream	Stream composition	Relative flow ratios
1	1CX	0.01 M HNO_3	5.7
7	1BX	1.5 M HNO_3	0.56
16		Reductant (electrocolumn)	-
20	1AS	3.0 M HNO_3	0.50
30	1AF	1.25 M $UO_2(NO_3)_2$; 0.013 M $Pu(NO_3)_4$; 2.5 M HNO_3	1.00
37	1AX	30 vol % TBP--n-dodecane	3.80

This flowsheet uses the number of theoretical stages estimated to be available in the Barnwell plant to recover greater than 99.8% of the uranium and plutonium. The stage numbers are listed in the table and Fig. 5. It should be remembered that actual plant operation may not attain these calculated values.

While the above flowsheet will serve as a reference starting point, the actual hot-cell pilot plant may be based on three banks of 16-stage mixer-settlers. This necessitates additional optimization calculations using the SEPHIS program. Operable (but not optimized) operating conditions for three banks of mixer-settlers containing 16 theoretical stages each are given below (stages are numbered consecutively for the three banks against the direction of flow of the organic) and also in Fig. 6.

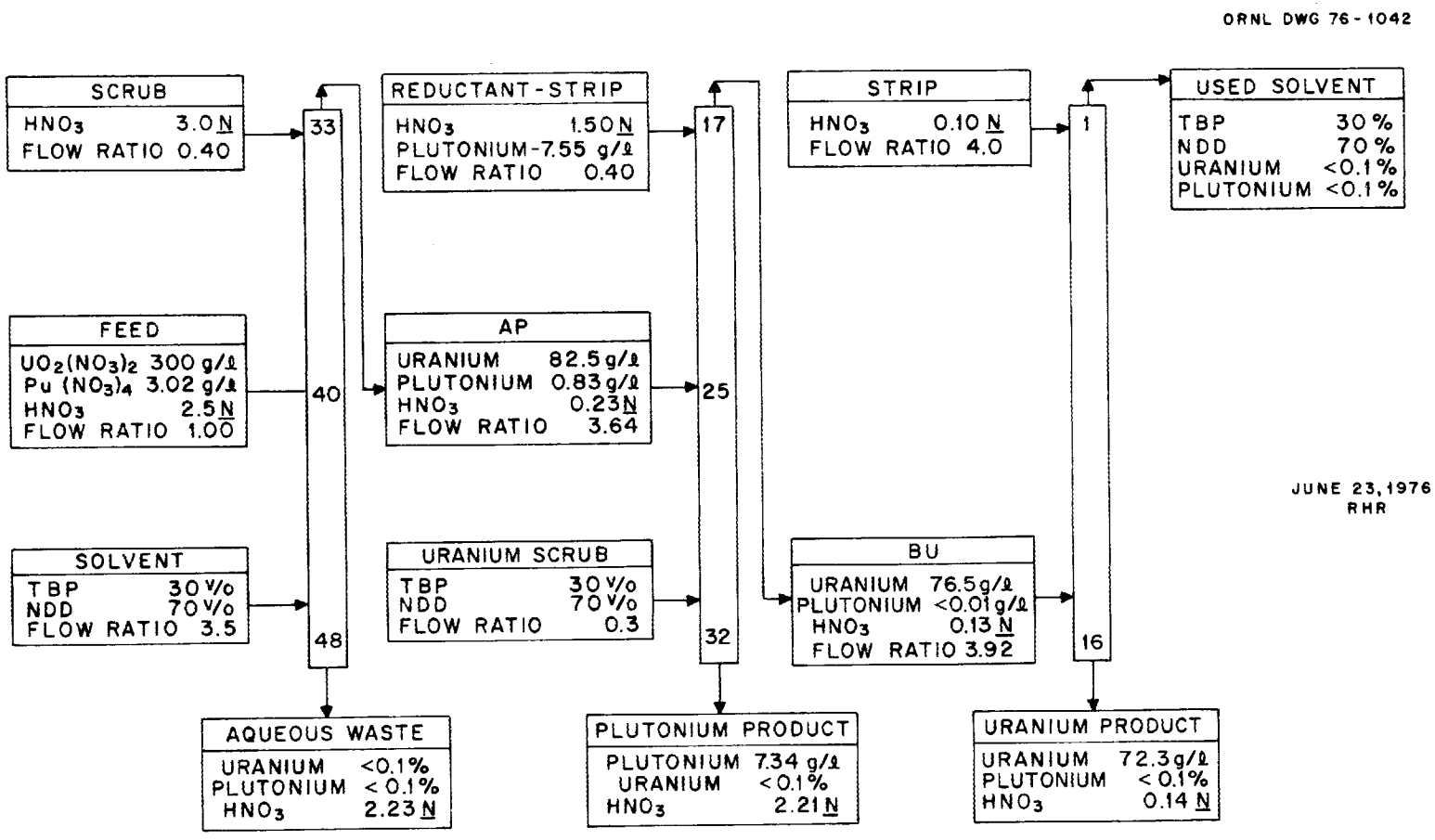

Fig. 6. Preliminary flowsheet for hot-cell experiments as of June 15, 1976.

Stage number	Entering stream	Stream composition	Relative flow ratio
1	1CX	0.01 N HNO_3	4.0
17	1BX	-7.55 g/liter Pu; 1.5 N HNO_3	0.4
32	1BS	30% TBP--n-dodecane	0.3
33	1AS	3.0 N HNO_3	0.4
40	1AF	300 g/liter U; 3.02 g/liter Pu; 2.5 N HNO_3	1.0
48	1AX	30%--n-dodecane	3.5

The negative plutonium concentration entering stage 17 simulates plutonium reductant. The computer calculations indicated that these operating conditions would result in less than 0.1% loss of uranium and plutonium to the organic and aqueous waste streams and less than 0.1% of the uranium in the plutonium product and vice-versa. The system reached 90% of steady-state operation in 48 time units, 95% in 81 time units, and 99% in 233 time units. The conversion of computer time units to real operating times and to process solution volumes depends on the contactor design and total flow rates. These values have not been finalized.

The optimized flowsheet will probably use somewhat lower flow rates of solvent and strip solutions. The lower flow rates and a larger internal reflux of the optimized flowsheet will probably require more time units to reach steady-state operation.

3.3 Cold Studies

W. D. Bond, F. A. Kappelmann, and F. M. Scheitlin
(Chemical Technology Division, ORNL)

Installation of two mixer-settler banks in Building 3508 was completed. Shakedown tests were satisfactory when the extraction of nitric acid by 30% TBP--dodecane at 25 and 45°C was carried out. A run with a simulated feed solution (fission product content equivalent to 33,000 MWd(t)/metric ton), using the flowsheet given in ORNL-5012, is planned for evaluation of the units in the extraction and stripping of uranium.

Batch extractions are being carried out to determine equilibrium distribution coefficient data as a function of uranium saturation of the 30% TBP--dodecane. The distribution coefficients of uranyl nitrate between 30% TBP--dodecane and nitric acid were measured (Table 7). Our values are in reasonable agreement within the range of the values reported in the literature, which assures us that our experimental technique and analytical methods are satisfactory. Similar measurements were made on the distribution of uranium, zirconium, ruthenium, and molybdenum between 30% TBP and simulated feed selections. However, analytical results are not available at this time.

Table 7. Uranyl nitrate distribution between 30% TBP--dodecane and nitric acid

Aqueous phase		Organic phase[a]		Distribution coeff.
HNO_3 (M)	Uranium (M)	HNO_3 (M)	Uranium (M)	(organic/aqueous)
2.42	0.00065	0.497	0.0127	19.5
2.64	0.00065	0.494	0.01256	19.3
2.41	0.0134	0.330	0.169	12.5
2.56	0.01298	0.297	0.1765	13.6
2.40	0.171	0.108	0.412	2.41
2.26	0.183	0.078	0.3996	2.18
2.05	0.116	0.122	0.358	3.09
2.07	0.118	0.100	0.357	3.03
1.96	0.062	0.162	0.319	5.14
1.96	0.077	0.132	0.329	4.27

[a]100% uranium saturation is 0.52 M.

4. OFF-GASES - FLUOROCARBON ABSORPTION STUDIES*

W. S. Groenier
(Chemical Technology Division, ORNL)

This program is concerned with the development of the Krypton Absorption process on an accelerated basis. Funding is provided for expansion of the pilot-plant development, initiation of plant engineering design criteria, performance of a proposed system reliability analysis, and a study of the chemical effects of impurities in the fluorocarbon solvent on the process and equipment.

4.1 Fluorocarbon Absorption Process Development

M. J. Stephenson (ORGDP)

4.1.1 Pilot-plant operation

The $^{85}Kr/^{14}C$ removal process will be the final step in an integrated chain of processes designed to collectively decontaminate fuel reprocessing plant off-gas. It is essential to the overall development of the fluorocarbon process to determine: (1) the consequences of the failure of upstream primary removal equipment causing fission products and chemical contaminants to be passed downstream; (2) the ability of the fluorocarbon process to act as a short-term backup system to remove the other radioactive components from the reprocessing plant off-gas in case of such a failure; and (3) the feasibility of the fluorocarbon process to function as the primary removal facility for iodine, other fission products (including ruthenium oxides), and chemical contaminants such as nitrogen

* Jointly funded by LWR Recycle and LMFBR Reprocessing programs.

dioxide. Pilot-plant work is currently being directed toward exploring these points by establishing the general process behavior of feed gas components such as carbon dioxide, nitrogen dioxide, iodine, methyl iodide, and water, and by defining the effects of these components on the general operability and overall performance of a process designed for ^{85}Kr and ^{14}C removal.

Recent pilot-plant tests continue to support hypotheses drawn from earlier scoping tests and performance calculations.[5] Preliminary results are available for experiments completed with nitrogen dioxide, iodine, and methyl iodide. The vapor pressures of these three components are given in Fig. 7, along with that of refrigerant-12 (R-12). The vapor-liquid distribution coefficients for xenon and carbon dioxide in R-12 are also shown to put the component vapor pressures into perspective. Those components more volatile than R-12 end up in either the process vent with nitrogen and oxygen or the krypton product stream, while the less volatile components collect in the solvent still bottoms receiver. Water, not shown in Fig. 7, is more volatile than iodine but significantly less volatile than CH_3I. Because krypton and xenon removals in excess of 10^3 were achieved previously,[5] high process removals were projected for the even less volatile feed gas components. These expectations have been essentially realized in that quantitative removal of iodine, methyl iodide, and nitrogen dioxide was achieved in the pilot facility.

The results of the pilot-plant tests with iodine are summarized in Table 8. The methyl iodide tests were the easiest to perform, whereas elemental iodine was reluctant to move through the feed gas circuit even though the gas lines were heated. Once in the solvent,

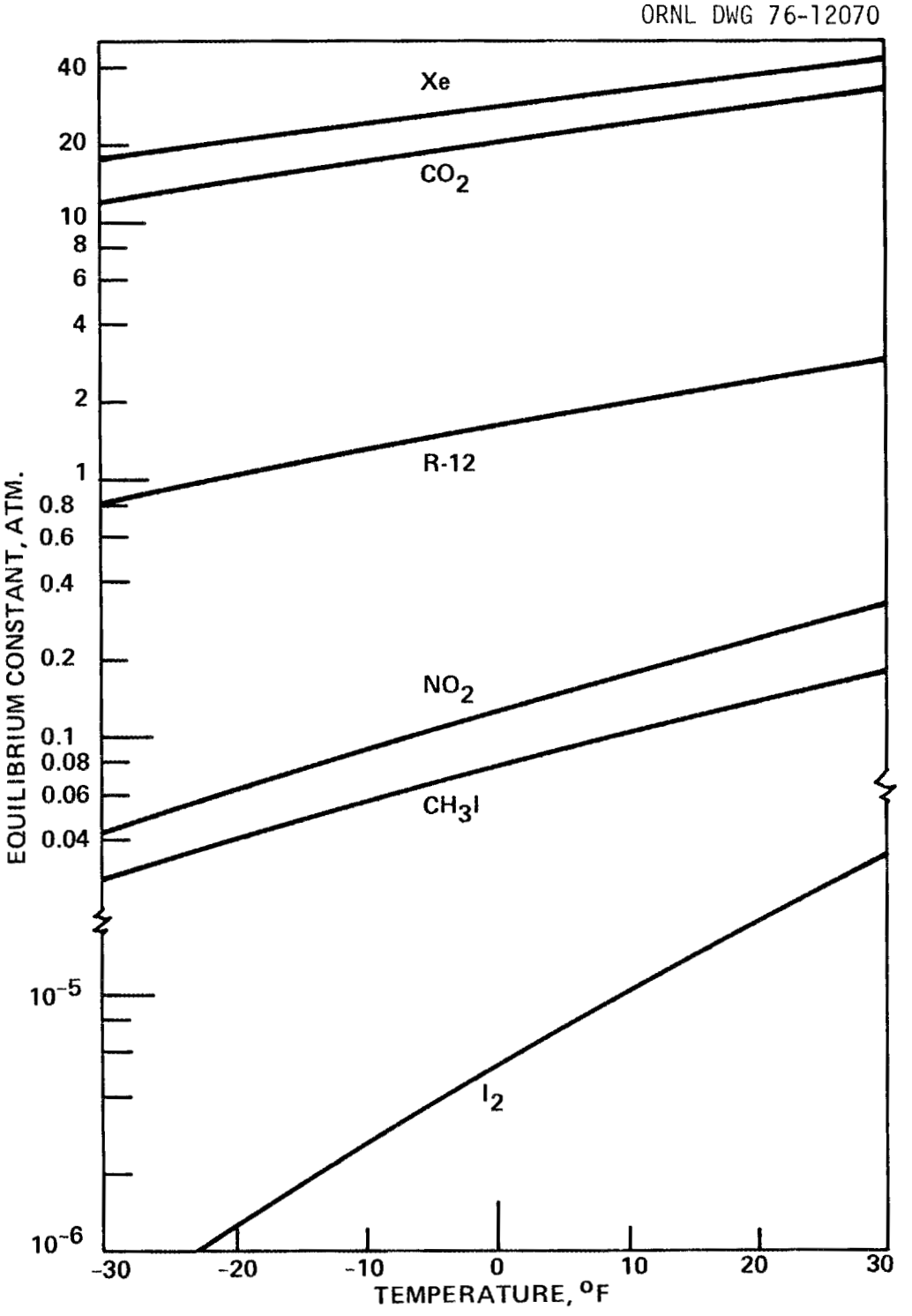

Fig. 7. Vapor pressures of important reprocessing plant off-gas components and R-12.

Table 8. Summary of iodine removal tests[a]

Test number	Iodine form	Solvent recycle	Still reflux ratio	Absorber feed gas concentration (ppm)	Measured decontamination factor[b]	Measured decontamination factor[c]
1a	I_2	No	--	~0.001	$>10^4$	--
1b	I_2	Yes	d	~0.001	$>10^3$	--
1c	I_2	Yes	0.01	~0.001	$>10^4$	--
2a	CH_3I	No	--	136	$>6 \times 10^4$	--
2b	CH_3I	Yes	0.01	7	$>2 \times 10^3$	1.5×10^4
3a	CH_3I	Yes	0.34	248	$>2 \times 10^4$	5.9×10^4
3b	CH_3I	Yes	0.34	28	$>2 \times 10^3$	4.0×10^4
3c	CH_3I	Yes	0.13	28	$>2 \times 10^3$	1.0×10^4

[a]General test conditions: Absorber pressure, 300 psig; absorber temperature, -10°F; solvent flow, 1 gpm.

[b]Based on gas stream analyses.

[c]Based on inlet gas stream and recycled solvent stream analyses and on the assumption that absorber off-gas is in equilibrium with the incoming solvent.

[d]Solvent still partially by-passed.

I_2 became mobile and ultimately collected in the solvent still reboiler. Elemental and organic iodine removals in excess of 10^4 were achieved. Each of the three tests was conducted with 5 mCi of ^{131}I, and gamma scintillation analytical techniques were used to evaluate process performance. The small amount of activity relative to the plant size and long duration of the tests largely limited the quantitative capability of the counting equipment. Process removals were quite high, and the off-gas generally contained an undetectable amount of iodine. In some cases, the absorber performance could be calculated from the amount of activity in the recycle solvent by assuming that the absorber off-gas was in equilibrium with the incoming solvent.

The test results clearly show that the efficiency of the process to remove methyl iodide is limited by the performance of the solvent purification still. Elemental iodine, on the other hand, was much easier to remove from the recycle solvent. The relative volatility of R-12 to iodine and methyl iodide is shown in Fig. 8. At the conditions of the still (i.e., -10°F), this value is greater than 10^5 for I_2 and 24 for CH_3I. Increasing the reflux ratio in the solvent still from 0.13 to 0.34 (tests 3b and 3c, Table 8) resulted in a significant reduction in the amount of methyl iodide in the recycle solvent and improved the process removal efficiency by a factor of 4. The effect of any recycled iodine on the process removal efficiency could not be determined because the iodine recycle concentrations were below the level of detection. Additional rectifying stages and/or higher reflux ratios should significantly improve the recovery capability of the process. It is important to point

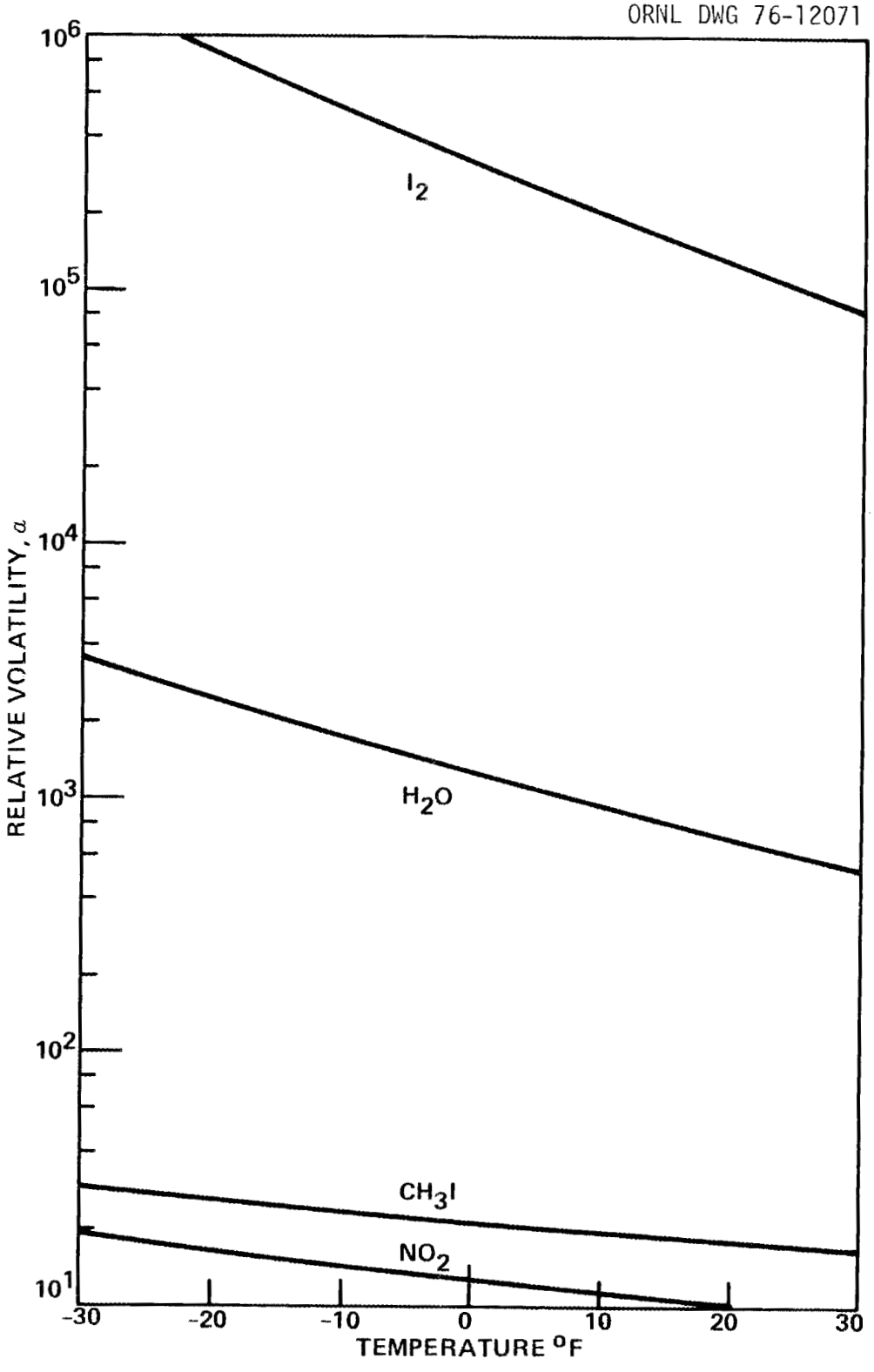

Fig. 8. Vapor pressure of R-12 relative to several reprocessing plant off-gas components.

out that water and elemental iodine are significantly less volatile than methyl iodide; consequently, these two components are much easier to remove from the solvent. Therefore, a process designed to achieve a methyl iodide decontamination factor of 10^6 should be capable of even higher DFs for elemental iodine and water.

The results of the NO_2 removal experiments are summarized in Fig. 9. More than 2 months of the current test series has been devoted to studying the process behavior of NO_2. A Du Pont Photometric Model 411 Analyzer having the capability of detecting NO_2 levels as low as 1 to 2 ppm was used for direct in-line concentration determinations for the more recent experiments. Process removal efficiencies of between 97 and 99.9% were achieved. The data suggest that the removal of NO_2 is strongly dependent on the feed concentration. This is contrary to previous experience with both less-volatile (I_2 and CH_3I) and more-volatile (krypton and xenon) components. For one series of tests, the concentration of NO_2 in the absorber off-gas remained around 20 ppm, regardless of the absorber feed gas flow or inlet concentration. This, of course, again suggested inadequate removal of high-boiling components from the recycle solvent. The absorber off-gas concentration did decrease with an increase in the solvent still reflux, but not to the extent that was found for the CH_3I tests. This was not surprising since NO_2 is more volatile than CH_3I and thereby more difficult to remove from the solvent.

It is appropriate to point out that when the solvent still was designed, quantitative recovery of the feed gas NO_2 was not a design objective. Instead, the purpose of the unit was to prevent bulk buildup of high-boiling components in the recycle solvent. Consequently, this

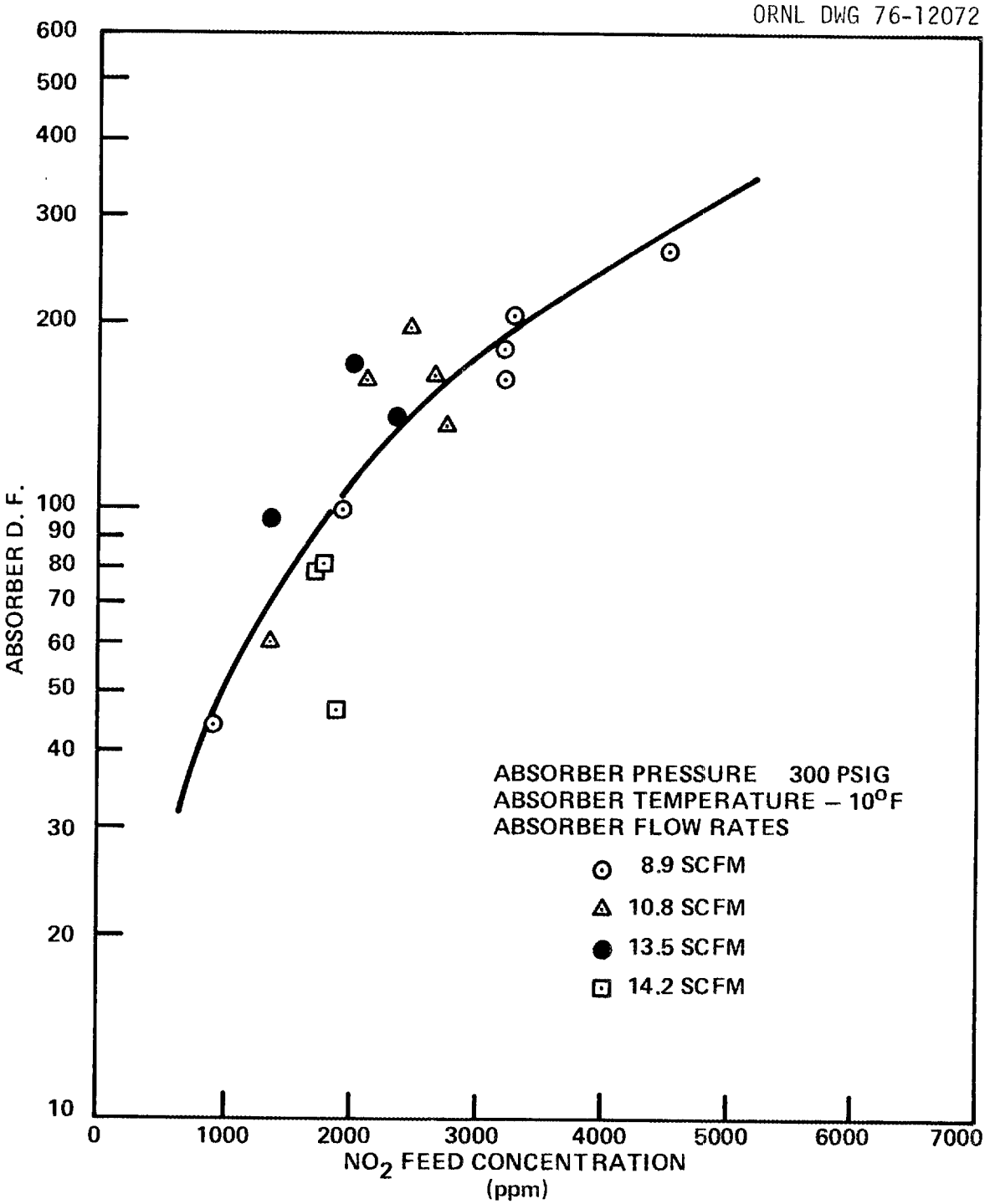

Fig. 9. Fluorocarbon process removal of NO_2 as a function of feed gas composition.

piece of equipment was built to remove only 90% of the dissolved NO_2 from the solvent.[6] Higher NO_2 removals can be obtained by enlarging the enriching section of the solvent purification column. This option is being considered for improving both CH_3I and NO_2 recovery efficiencies. Solvent washing is another option, but would require additional equipment.

Preliminary pilot-plant tests have been completed with xenon, and other tests are currently being made with CO_2. Carbon dioxide removals in excess of 10^4 are being achieved. More complete data will be available for the next progress report. Tests with water have been temporarily postponed until necessary analytical equipment is available for measuring ultra-low water concentrations. High water removals are anticipated because of the vapor pressure of water relative to R-12.

4.1.2 Process application

Continued modeling studies have indicated that the overall efficiency and economics of the process can be significantly improved by lowering the solvent/feed gas absorption ratio and increasing the height of the absorber column. This absorber trade-off will not only reduce the work load on the fractionator, but will also help to minimize the size of the fractionator gas recycle stream. The pilot-plant fractionator reboiler heater control is not responsive enough to process load changes that might be encountered in actual plant application, and the tight column control required for efficient fractionation would be difficult, if not impossible, to maintain. An automatic reboiler control circuit has been designed to maintain the necessary fractionation control and will be pilot-plant-tested next quarter. The heart of the system is an SCR (silicon-controlled rectifier) power controller which precisely proportions electrical power

to the reboiler heater according to a 4-20 mA controller signal. The feedback control signal will be provided by a thermocouple installed on the fractionator column.

4.2 Chemical Studies of Contaminants in LWR Off-Gas Processing

L. M. Toth, D. W. Fuller, and J. T. Bell
(Chemistry Division, ORNL)

A laboratory-scale investigation of various off-gas contaminants (e.g., I_2, CH_3I, H_2O and NO_2) in chlorofluorocarbon solvents was initiated during April using absorption spectrophotometry. Much of the apparatus was already in operation because of previous work on some of these contaminants in a KALC[7] process study. Several aspects of the physical and chemical behavior of I_2 in R-12 have been examined in a high-pressure optical cell and are described below.

Iodine in R-12 has an electronic absorption band in the visible region of 520 nm (ϵ = 832 liter $mole^{-1}cm^{-1}$) arising from solvated I_2 molecular species. In the absence of any added H_2O, dilute solutions of I_2 in R-12 show no sign of chemical reaction with the system, namely, the 304 stainless steel vessel, Teflon seals, or the R-12 solvent. The solubility of I_2 has been measured as a function of temperature by monitoring its 520-nm band. These data are shown in Fig. 10. The data have been fit to a least-squares line, which is given by:

$$\log_{10}(I_2) = 2.244 - (1544)/T, \qquad (1)$$

where (I_2) is the mole fraction and T is °K. This expression supersedes a similar one which was given previously.[8]

In addition to the I_2 species, we have found a small band at 270 nm which can be assigned to an I_4 species in solution. Uncertainties in the

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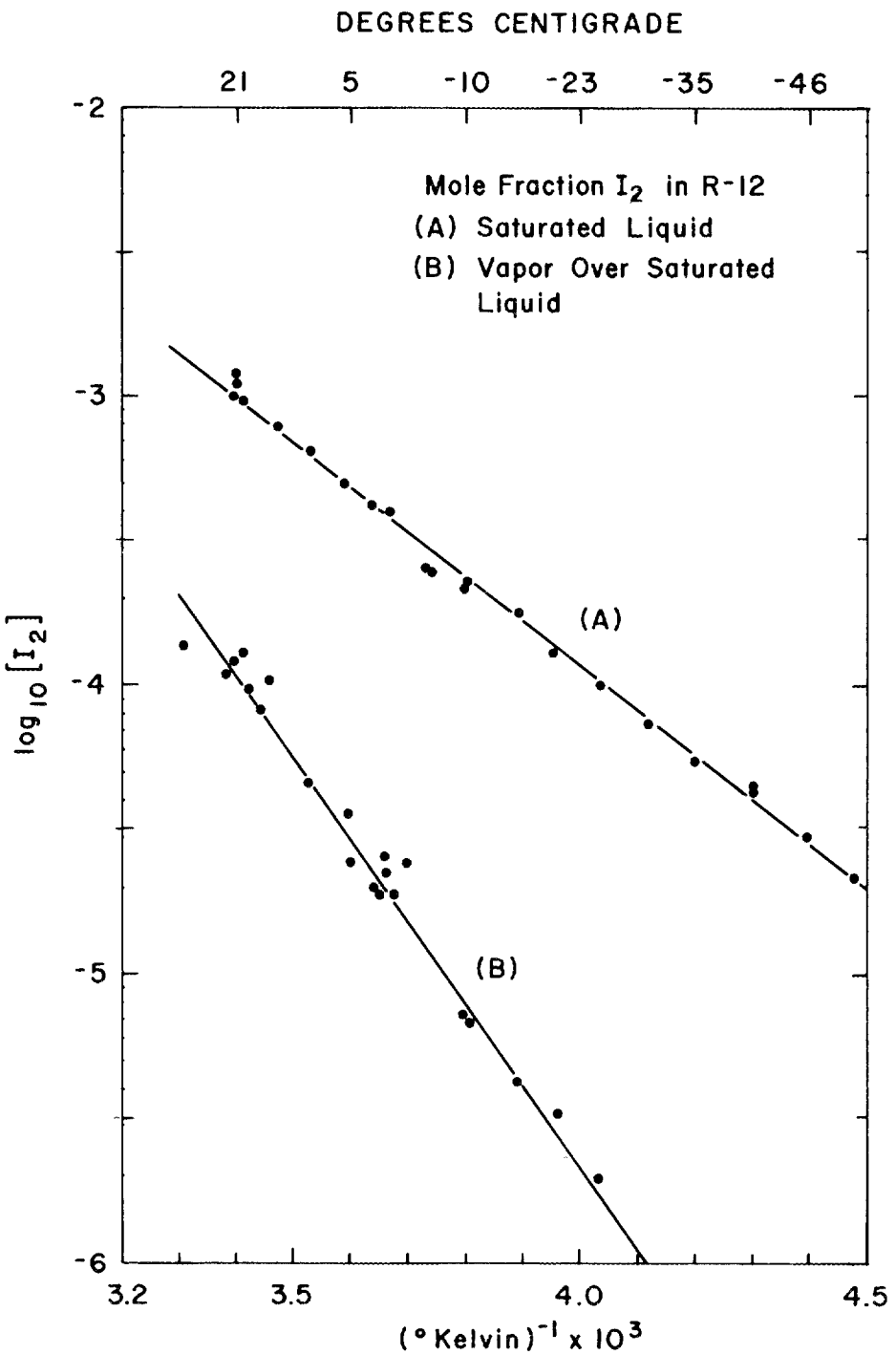

Fig. 10. Mole fraction of I_2 in R-12 (A) saturated liquid and (B) vapor over saturated liquid.

reported equilibrium constant for the reaction,[9] $2I_2 \rightleftarrows I_4$, and in the molar extinction coefficient for I_4 prevent an accurate measurement of the amount of iodine present in solution as I_4. However, we estimate its concentration to be no more than 1 to 2 % of the total soluble iodine.

Distribution coefficients for I_2 between liquid and gaseous R-12 have been measured from +21 to -42°C by monitoring the concentration of I_2 in the vapor over a solution which was saturated with I_2. The mole fraction of I_2 in the vapor as a function of temperature is also given in Fig. 10 and is represented by the expression:

$$\log_{10}(I_2)_v = 5.706 - (2840)/T. \tag{2}$$

The difference between Eqs. (1) and (2),

$$\log_{10}(D) = -3.462 + (1296)/T, \tag{3}$$

is simply the distribution coefficient expression, $D = (I_2)_l/(I_2)_v$, for a saturated I_2 solution. Based on the results of an earlier study of I_2 in liquid CO_2,[7] this expression should also be valid for dilute I_2 solutions. Nevertheless, this will be verified before the conclusion of the I_2– R-12 experiments.

R-12 solutions of I_2 containing 100 ppm or less of added H_2O corrode the 304 stainless steel cell at 0°C with the subsequent formation of an insoluble iron-iodide type of product. The limited solubility of H_2O in R-12 (25 ppm at 0°C) presents a means of obtaining fundamental rates for the corrosion reaction which is dependent on the concentration of water and iodine. For example,

$$\frac{d\,(I_2)}{dt} = -k(H_2O)(I_2). \tag{4}$$

With a constant concentration of water due to the saturated solution, $k(H_2O) = k'$, and Eq. (4) reduces to:

$$\frac{d(I_2)}{dt} = -k'(I_2). \tag{5}$$

A first-order rate process has been measured for I_2 and is being checked as a function of added water to demonstrate that, as long as the R-12 remains saturated, the rate of I_2 loss from solution remains first order and independent of the amount of water.

4.3 Reliability Studies

W. E. Unger (Chemical Technology Division, ORNL) and
D. E. Wood* (Kaman Sciences Corporation)

Progress by Kaman Sciences Corporation (subcontract No. 11x-81721) during this report period included:

(1) Estimation of failure data and repair times for the process components.

(2) Completion of the GO model and of availability runs using this model for the Selective Absorption Pilot Plant.

(3) Modification of the GO model to include process redundancy and bypass suggestions followed by additional availability runs.

(4) Performance of sensitivity runs to determine highest impact components.

Increased reliability and availability of process components are sought by examining the components in order of increasing availability. Thus, active components are considered first, with the most complex items (and highest failure rates) at the top of the list. The GO analysis and

* Senior Research Scientist, Nuclear Services Program, Kaman Sciences Corporation.

sensitivity runs guided by the above consideration led to the following suggestions relative to system improvement:

(1) The compressor, other refrigeration components, pumps, and fast-acting control valves should be redundant.

(2) Coolers, filters, slow-acting control valves, and the solvent still section should have bypass capability. (Some of these already exist.)

(3) All components (including console modules) should have spares readily available to minimize repair and replacement times. However, since failures of tanks and columns will be rare, the spares could include raw materials (pipe and headers) instead of complete units.

Some tentative conclusions from the model and results are as follows:

(1) Outage survival time (time that the system can survive with a particular component or section not in service) is important.

(2) Repair time is important if it exceeds the outage survival time.

(3) Redundancy reduces the need for accurate failure data.

(4) Relatively simple system design alterations can reduce the outage time to the point where the accuracy of failure data is not critical. (A relatively large error still allows availability criteria to be met.)

(5) Gas storage at the system input is not necessary.

(6) Gas storage at the output in any single container (or any system subject to a single failure) should not exceed an amount equivalent to the allowable emission per year.

(7) Redundancy will increase the total amount of repair work while reducing system outage time.

(8) Availability can be high enough to eliminate the need for experimental determination of failure and repair data. However, the fuel reprocessing plant may have such requirements since even short outages should probably be avoided in certain process steps.

(9) Common mode failure is not as important as in high-reliability systems since nominal outages can be tolerated.

Completion of the study and the issue of a final report are scheduled for August 1, 1976.

4.4 Engineering Design Criteria Studies

R. W. Glass (Chemical Technology Division, ORNL)

Engineering design criteria studies for a conceptual krypton removal system have been completed with the issuance of a set of check prints (CD-SK-550 through CD-SK-559) that include an overall flowsheet and equipment for feed treatment, gas compression, absorption, fractionation, stripping, product separation, off-gas treatment, solvent purification, refrigeration, and instrumentation. Comments are being received and a final set of prints will be issued in the near future. Since this study applies to both the LMFBR Reprocessing and the LWR Fuel Reprocessing and Recycle Programs, the results have been provided to Lew Landon and Dr. Vince Van Brunt of the Savannah River Laboratory who will have their engineering cost analysis people provide cost estimates for this system.

5. FINISHING PROCESSES - MOX FUEL FABRICATION

R. B. Fitts, A. R. Olsen, J. D. Sease
(Metals and Ceramics Division, ORNL)

The objective of this program is to provide technical assistance to SRL in the development of an overall plan in support of LWR-MOX fuel fabrication processes and facilities.

5.1 MOX Fabrication Processes

Information obtained during site visits[10] regarding the present status of MOX fabrication processes has been added to a preliminary summary of such technical factors. Additional information needs are now being identified. When these are satisfied, the listing of technical factors will be published for use in defining needed research and engineering development efforts and in establishing a basic flowsheet which can be used in economic and plant design analyses.

5.2 MOX Fabrication Facilities

Information has been collected and is being evaluated in preparation for the establishment of draft design philosophy and design criteria for a MOX Commercial Demonstration Fabrication Facility (CDFF).

5.2.1 Remote manipulator and viewing development

The need for remote handling and viewing development has long been recognized. Two recent papers defining the design and performance requirements in a generic fashion are available,[11,12] together with a survey of

commercially available equipment. The detailed study of the MOX-CDFF requirements is being delayed pending definition of process and equipment flowsheets.

6. URANIUM HEXAFLUORIDE CONVERSION

J. H. Pashley, R. P. Milford, and J. Dykstra (ORGDP)

Collection of information relating the various conversion processes was continued during the quarter. Collation of these data is expected next quarter.

7. ECONOMIC STUDIES

J. M. Morrison (ORGDP)

This task which was authorized on March 25, 1976, will support the fuel cycle development and demonstration programs being conducted by ERDA contractor organizations. Primary emphasis will be placed on the LWR fuel cycle, its relationship to the other nuclear fission cycles, and the interfaces among the various ERDA contractors as well as private industrial organizations involved.

7.1 LWR Recycle Data

E. H. Gift (ORGDP)

Calculations have been made to provide mass balance data for a three-region PWR through successive recycle of self-generated plutonium. The PWR model selected was based on the Westinghouse 17x17 lattice design currently being used, for example, in the Trojan plant of Portland General Electric (DOCKET 50-344). Computations were made using the LEOPARD code,[13] a spectrum dependent, nonspatial depletion code.

The three refueling regions were assumed to be initially loaded at 2.02, 2.57, and 3.2 wt % ^{235}U and were estimated to yield respective average burnups of 16,400, 25,500, and 32,850 MWd/metric ton (MT). All successive reloads were required to match the 32,850-MWd/MT burnup achieved by the 3.2 wt % ^{235}U region. Plutonium from each of the three initial regions was then assumed to be recycled through five cycles. At the end of each cycle, plutonium from the mixed oxide portion of the reload batch was recovered and mixed with the plutonium from the 3.2 wt % slightly

enriched ^{235}U portion of the reload batch. This plutonium (after losses in reprocessing and refabrication and decay of ^{241}Pu) was then mixed with natural uranium to provide the mixed oxide fuel for the succeeding reload batch. The results of these calculations are shown in Tables 9, 10, and 11 for each of the three initial regions for five successive recycles of the plutonium. The fissile enrichment (^{235}U + ^{239}Pu + ^{241}Pu) of the mixed oxide gradually increases from about 3.2% for the first recycle of plutonium to about 3.5% for the fifth recycle, as a result of the buildup of higher plutonium isotopes. Mixed oxide is seen to provide about 30 to 40% of the fissile requirements of a reload batch.

Continuing work will provide similar data for a BWR and for a PWR in cases where the mixed oxide is formulated using recovered spent uranium rather than fresh natural uranium.

Table 9. PWR fuel needs for self-generated plutonium recycle of reload region initially at 2.02 wt % ^{235}U.

[kg/1000 kg (U + Pu) charged]

Isotope	Initial Loading 1st Cycle		1st Recycle of Plutonium				
			Charged		Discharged		
	Charge	Discharge	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	20.20	7.9468	1.3654	25.64217	0.62741	6.81096	7.43837
U-236	0	2.0683	0	0	0.14859	3.20936	3.35795
U-238	979.80	965.7647	190.6794	775.67553	186.12018	755.47247	941.59265
Pu-239	0	4.3964	4.3524	0	2.24928	4.46358	6.71286
Pu-240	0	1.4795	1.4647	0	1.26538	1.79303	3.05841
Pu-241	0	0.7100	0.6400	0	0.99303	1.09813	2.09116
Pu-242	0	0.1822	0.1804	0	0.52863	0.43455	0.96318
Burnup, MWd/MT	0	16,427	0	0	32,854	32,854	32,854
Frac. Fissile	0.0202	0.0132851	0.03200	0.032			0.016828
Total Pu	0	6.7810	6.63750	0			12.82561
Frac. Fiss. Pu	0	0.754481	0.752151	0			0.686441
Total U	1000	975.7798	192.0448	801.3177	186.89618	765.49279	952.38897
E-25	0.0202	0.008144	0.00711	0.032	0.0033570	0.0088975	0.0078103
E-26		0.0021196			0.00079504	0.0041925	0.0039395

Table 9 (continued)

	2nd Recycle of Plutonium				
	Charged		Discharged		
Isotope	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.28232	21.32754	1.08251	5.66492	6.74744
U-236	0	0	0.24442	2.66934	2.91376
U-238	318.71976	645.15812	311.15828	628.35448	939.51276
Pu-239	6.64573	0	3.80762	3.71253	7.52015
Pu-240	3.02783	0	2.24789	1.49133	3.73922
Pu-241	1.88514	0	1.91005	0.91335	2.82340
Pu-242	0.95355	0	1.52485	0.36144	1.88629
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.032422	0.032			0.017708
Total Pu	12.51225	0			15.96906
Frac. Fiss. Pu	0.681801	0			0.647724
Total U	321.00209	666.48566	312.48521		949.17396
E-25	0.00711	0.032	0.00346420		0.0071088
E-26			0.00078218		0.00306979

Table 9 (continued)

	3rd Recycle of Plutonium				
	Charged		Discharged		
Isotope	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.57827	19.89807	1.25654	5.28524	6.54177
U-236	0	0	0.27208	2.49043	2.76251
U-238	360.04752	601.91670	351.48093	586.23932	937.72025
Pu-239	7.44495	0	4.44579	3.46369	7.90948
Pu-240	3.70183	0	2.65033	1.39137	4.04170
Pu-241	2.54523	0	2.32128	0.85226	3.17354
Pu-242	1.86743	0	2.41257	0.33721	2.74978
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.0332336	0.032			0.0182659
Total Pu	15.55944	0			17.87450
Frac. Fiss. Pu	0.642066	0			0.620046
Total U	362.62579	621.81477	353.00955		947.02453
E-25	0.00711	0.032	0.0035595		0.0069077
E-26			0.00077074		0.0029170

Table 9 (continued)

	4th Recycle of Plutonium				
	Charged		Discharged		
Isotope	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.65627	19.48765	1.32652	5.17623	6.50275
U-236	0	0	0.27623	2.43906	2.71529
U-238	370.93971	589.50154	362.30876	574.14752	936.45628
Pu-239	7.83039	0	4.76663	3.39225	8.15888
Pu-240	4.00128	0	2.84271	1.36267	4.20538
Pu-241	2.86088	0	2.53326	0.83456	3.36782
Pu-242	2.72228	0	3.16192	0.33025	3.49217
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.034136	0.032	363.91151		0.0186853
Total Pu	17.41483	0	0.0036452		19.22425
Frac. Fiss. Pu	0.613918	0			0.599592
Total U	373.59598	608.98919			945.67432
E-25	0.00711	0.032			0.0068763
E-26			0.00075906		0.0028713

Table 9 (continued)

Isotope	5th Recycle of Plutonium				
	Charged		Discharged		
	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.67190	19.37511	1.36088	5.14633	6.50721
U-236	0	0	0.27445	2.42498	2.69943
U-238	373.12206	586.09704	364.50439	570.83169	935.33609
Pu-239	8.07729	0	4.97070	3.37266	8.34336
Pu-240	4.16333	0	2.95592	1.35480	4.31072
Pu-241	3.03602	0	2.66366	0.82974	3.49340
Pu-242	3.45725	0	3.78882	0.32835	4.11717
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.03494104	0.032	366.13972		0.0190131
Total Pu	18.73389	0			20.26465
Frac. Fiss. Pu	0.593220	0			0.5841088
Total U	375.79396	605.47215			944.54273
E-25	0.00711	0.032	0.0037168		0.0068893
E-26			0.00074958		0.0028579

Table 10. PWR fuel needs for self-generated plutonium recycle of reload region initially at 2.57 wt % ^{235}U

[kg/1000 kg (U + Pu) charged]

Isotope	Initial Loading 1st Cycle		1st Recycle of Plutonium				
			Charged		Discharged		
	Charge	Discharge	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	25.70	7.6452	1.6337	24.37915	0.76146	6.47548	7.23694
U-236	0	3.0501	0	0	0.17651	3.05128	3.22779
U-238	974.30	953.6570	228.1496	737.46934	222.7161	718.2614	940.9774
Pu-239	0	5.0711	5.0204	0	2.71105	4.24372	6.95477
Pu-240	0	1.9793	1.9595	0	1.56424	1.70471	3.20895
Pu-241	0	1.1093	1.0000	0	1.26572	1.04404	2.30976
Pu-242	0	0.3921	0.3882	0	0.80485	0.41315	1.21800
Burnup, MWd/MT	0	25,516	0	0	32,854	32,854	32,854
Frac. Fissile	0.0257	0.014211	0.03214	0.032			0.017098
Total Pu	0	8.5518	8.3681	0			13.69148
Frac. Fiss. Pu	0	0.722702	0.719446	0			0.676664
Total U	1000	964.3523	229.7834	761.8485	223.65407	727.78816	951.4421
E-25	0.0257	0.007928	0.00711	0.032	0.0034046	0.00889748	0.007606
E-26		0.0031628			0.00078921	0.00419254	0.0033925

Table 10 (continued)

	2nd Recycle of Plutonium				
	Charged		Discharged		
Isotope	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.38556	20.83611	1.13574	5.53440	6.67014
U-236	0	0	0.25497	2.60783	2.86280
U-238	346.48606	630.29227	325.25509	613.87583	939.13092
Pu-239	6.88522	0	3.98619	3.62698	7.61317
Pu-240	3.17686	0	2.34887	1.45696	3.80583
Pu-241	2.08220	0	2.01009	0.89231	2.90240
Pu-242	1.20582	0	1.77319	0.35311	2.12630
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.0325416	0.032			0.017807
Total Pu	13.35010	0			16.44770
Frac. Fiss. Pu	0.671712	0			0.639333
Total U	335.52152	651.12838	326.64580		948.66386
E-25	0.00711	0.032	0.0034769		0.0070311
E-26			0.00078057		0.0030177

Table 10 (continued)

Isotope	3rd Recycle of Plutonium				
	Charged		Discharged		
	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.58897	19.83495	1.27140	5.26847	6.53987
U-236	0	0	0.27197	2.48253	2.75450
U-238	361.54246	600.00732	353.07815	584.37967	937.45782
Pu-239	7.53704	0	4.52284	3.45271	7.97555
Pu-240	3.76777	0	2.69559	1.38696	4.08255
Pu-241	2.61645	0	2.37075	0.84943	3.22018
Pu-242	2.10504	0	2.61403	0.33614	2.95017
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.03351848	0.032			0.0183792
Total Pu	16.02630	0			18.22845
Frac. Fiss. Pu	0.633552	0			0.614190
Total U	364.13143	619.84227	354.62152		946.75219
E-25	0.00711	0.032	0.0035852		0.0069076
E-26			0.00076693		0.0029094

Table 10 (continued)

	4th Recycle of Plutonium				
	Charged		Discharged		
Isotope	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.71066	19.22887	1.35166	5.10749	6.45915
U-236	0	0	0.28208	2.40667	2.68875
U-238	378.53596	581.67325	369.72480	566.52313	936.24793
Pu-239	7.89579	0	4.83076	3.34721	8.17797
Pu-240	4.04173	0	2.87242	1.34458	4.21700
Pu-241	2.99307	0	2.57783	0.82348	3.40131
Pu-242	2.92067	0	3.335364	0.32590	3.67951
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.0340755	0.032			0.0186952
Total Pu	17.85126	0			19.47579
Frac. Fiss. Pu	0.609977	0			0.594547
Total U	381.24662	600.90212	371.35854		945.39583
E-25	0.00711	0.032	0.00363977		0.0068322
E-26			0.00075959		0.0028440

Table 10 (continued)

	5th Recycle of Plutonium				
	Charged		Discharged		
Isotope	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.66406	19.40250	1.36262	5.15361	6.51623
U-236	0	0	0.27289	2.42841	2.70130
U-238	372.02780	586.92570	363.44997	571.63877	935.08874
Pu-239	8.09619	0	4.99453	3.37743	8.37196
Pu-240	4.17483	0	2.96596	1.35672	4.32268
Pu-241	3.06621	0	2.68044	0.83091	3.51135
Pu-242	3.64271	0	3.95736	0.32881	4.28617
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.0351218	0.032			0.0190709
Total Pu	18.97994	0			20.49216
Frac. Fiss. Pu	0.5881157	0			0.579895
Total U	374.69186	606.32820	365.08548		944.30627
E-25	0.00711	0.032	0.0037323		0.0069006
E-26			0.00074747		0.0028606

Table 11. PWR fuel needs for self-generated plutonium recycle of reload region initially at 3.2 wt % ^{235}U.

[kg/1000 kg (U + Pu) charged]

			1st Recycle of Plutonium				
			Charged		Discharged		
	Initial Loading 1st Cycle						
Isotope	Charge	Discharge	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	32.00	8.4997	1.83065	23.45672	0.85446	6.23047	7.08493
U-236		4.0051	0	0	0.19764	2.93583	3.13347
U-238	968.00	942.7877	255.64475	709.56578	249.55891	691.08460	940.64351
Pu-239		5.5703	5.5146	0	3.01880	4.08316	7.10196
Pu-240		2.2376	2.2152	0	1.74615	1.64021	3.38636
Pu-241		1.3704	1.2354	0	1.43269	1.00453	2.43722
Pu-242		0.5423	0.5369	0	0.99689	0.39752	1.39441
Burnup, MWd/MT	0	32,854	0	0	32,854	32,854	32,854
Frac. Fissile	0.032	0.016000	0.03214	0.032			0.0172238
Total Pu	0	9.7206	9.5021				14.31995
Frac. Fiss. Pu	0	0.714020	0.71037				0.666146
Total U	1000	955.2925	257.4754	733.0225	250.61101		950.86191
E-25	0.032	0.008897	0.00711	0.032	0.0034095		0.007451
E-26		0.00419254			0.00078863		0.0032954

Table 11 (continued)

	2nd Recycle of Plutonium				
	Charged		Discharged		
Isotope	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.45966	20.48303	1.18352	5.44061	6.62413
U-236	0	0	0.26110	2.56364	2.82474
U-238	343.48478	619.61153	335.16456	603.47328	938.63784
Pu-239	7.03094	0	4.17522	3.56552	7.74074
Pu-240	3.35250	0	2.48026	1.43228	3.91254
Pu-241	2.19709	0	2.13729	0.87719	3.01448
Pu-242	1.38047	0	1.97853	0.34712	2.32565
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.032474	0.032			0.0180082
Total Pu	13.96100	0			16.99341
Frac. Fiss. Pu	0.660986	0			0.6329053
Total U	345.9444	640.09456	336.60918		948.08671
E-25	0.00711	0.032	0.00351601		0.006987
E-26			0.00074884		0.00297941

Table 11 (continued)

	3rd Recycle of Plutonium				
	Charged		Discharged		
Isotope	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.62731	19.64544	1.27716	5.21813	6.49529
U-236	0	0	0.27838	2.45881	2.73719
U-238	366.89619	594.27444	358.15392	578.79611	936.95003
Pu-239	7.66333	0	4.58496	3.41972	8.00468
Pu-240	3.87341	0	2.73992	1.37371	4.11363
Pu-241	2.71749	0	2.42106	0.84132	3.26238
Pu-242	2.30239	0	2.80654	0.33293	3.13947
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.0336951	0.032			0.0183060
Total Pu	16.55662	0			18.52016
Frac. Fiss. Pu	0.626989	0			0.6083673
Total U	369.52350	613.91988	359.70946		952.67780
E-25	0.00711	0.032	0.0035505		0.0068179
E-26			0.00077390		0.0028732

Table 11 (continued)

	4th Recycle of Plutonium				
	Charged		Discharged		
Isotope	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.65010	19.49519	1.31655	5.17823	6.49478
U-236	0	0	0.27370	2.44001	2.71371
U-238	370.07892	589.72962	361.50311	574.36966	935.87277
Pu-239	7.92463	0	4.85410	3.39356	8.24766
Pu-240	4.07249	0	2.88994	1.36320	4.25314
Pu-241	2.94097	0	2.59225	0.83488	3.42713
Pu-242	3.10808	0	3.48549	0.33038	3.81587
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.0345865	0.032			0.0188320
Total Pu	18.04617	0			19.74380
Frac. Fiss. Pu	0.6021001	0			0.5913142
Total U	372.72902	609.22481	363.09336		945.08126
E-25	0.00711	0.032	0.0036259		0.006872
E-26			0.00075380		0.0028714

Table 11 (continued)

Isotope	5th Recycle of Plutonium				
	Charged		Discharged		
	Mixed Oxide	3.2 wt % Slightly Enriched	Mixed Oxide	3.2 wt % Slightly Enriched	Total
U-235	2.66730	19.37953	1.36967	5.14751	6.51718
U-236	0	0	0.27258	2.42553	2.69811
U-238	372.47935	586.23085	363.90389	570.90202	934.86591
Pu-239	8.16518		5.04326	3.37343	8.41669
Pu-240	4.21061		2.99300	1.35511	4.34811
Pu-241	3.08948		2.70757	0.82993	3.53750
Pu-242	3.77771		4.05840	0.32842	4.38682
Burnup, MWd/MT	0	0	32,854	32,854	32,854
Frac. Fissile	0.0353	0.032			0.0191459
Total Pu	19.24298	0			20.68912
Frac. Fiss. Pu	0.584871	0			0.580607
Total U	375.14664	605.61038	365.54614		944.08120
E-25	0.00711	0.032	0.0037469		0.0069032
E-26			0.00074568		0.00285792

8. RADIOLOGICAL TECHNIQUES FOR ENVIRONMENTAL ASSESSMENTS OF THE LWR FUEL CYCLE

R. E. Moore and D. C. Kocher (Environmental Sciences Division, ORNL)

The objective of this project is to evaluate the influence of "indoor living" on the estimated radiation dose to human population exposed to atmospheric releases of radioactivity.[14] This factor is thought to be significant because, in developed countries, man spends most of his time inside structures such as homes, schools, stores, and office and factory buildings. Population doses estimated for indoor exposure will be less than those estimated for outdoor exposure because concentrations of radionuclides inside a structure will be less than outside, and because walls of the structure attenuate radiation from outside sources.

This project will employ data and research results reported in the literature in areas of surface deposition, radiation shielding, building characteristics, building air turnover, properties of radionuclides, and man's activities. Based on this type of information, a general computer-implemented model will be developed to estimate indoor-to-outdoor population dose ratios. The model will consider all significant exposure pathways using site-specific demographic data.

The ratio of the indoor dose to the outdoor dose, d_i/d_o, can be expressed as follows:

$$\frac{d_i}{d_o} = \frac{d_i(\text{inh}) + d_i(\text{imm}) + d_i(\text{surf})}{d_o(\text{inh}) + d_o(\text{imm}) + d_o(\text{surf})} , \qquad (1)$$

where inh, imm, and surf refer to the exposure pathways of (1) inhalation of air containing radionuclides, (2) immersion in air, and (3) exposure

to surfaces contaminated by deposited radionucles, respectively. Equation (1) can be written as:

$$\frac{d_i}{d_o} = \frac{f(inh)d_o(inh) + f(imm)d_o(imm) + f(surf)d_o(surf)}{d_o} , \qquad (2)$$

where the f values are dose reduction factors for each of the three exposure pathways. To evaluate d_i/d_o for atmospheric releases from an LWR fuel cycle facility, it is necessary to determine these f values for each of the various kinds of man-made structures in the area for each radionuclide released.

The value for f(inh) is simply the ratio of the concentration of the radionuclide in indoor air to that in outdoor air. An expression was derived for f(inh):

$$f(inh) = \frac{\lambda_b (1 - e^{-\lambda_a t})}{\lambda_a} , \qquad (3)$$

where

$$\lambda_a = \lambda_b + \frac{V_{df}S_f}{V_h} + \frac{V_{dw}S_w}{V_h} + \frac{V_{dc}S_c}{V_h} + \lambda_r,$$

t = time,

λ_b = air turnover rate,

λ_r = radioactive decay constant,

V_h = volume of building,

V_{df}, V_{dw}, V_{dc} = deposition velocities of particulates on floor, walls, and ceiling,

S_f, S_w, S_c = surface areas of floor, walls, and ceiling.

Ranges for λ_b for various types of structures can be found in a report by Handley and Barton.[15] The theoretical and wind tunnel studies of Sehmel[16] on particulate deposition velocities are being examined for applicability to indoor environments.

Values for f(imm) may closely approximate f(inh) values corrected to account for gamma radiation emanating from outside air and penetrating the walls of the structure.

Evaluation of f(surf) requires an estimation of exposures from finite planes (walls, floor, and ceiling) indoors and from an infinite plane on ground surfaces outside the building. A literature survey of shielding effects of structural materials was recently completed. Shielding factors for various types of structures will be used in a computer subroutine to estimate f(surf) values.

A computer code is being written to estimate d_i/d_o, including all exposure pathways. This code will be used for a sensitivity analysis of basic parameters to refine the general model to evaluate actual source terms for LWR facilities.

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109. T. B. Hindman, Jr., Director, Fuel Cycle Project Office, U. S. Energy Research and Development Administration, Savannah River Operations Office, P. O. Box A, Aiken, SC 29801
110. A. H. Hines, Jr., President, Florida Power Corporation, P. O. Box 14042, St. Petersburg, FL 33733
111. C. A. Hirenda, Director of Marketing, Proposal Management, Inc., 121 N. Orionna Street, Philadelphia, PA 19106

112. P. Hogroian, Division of Nuclear Fuel Cycle and Production, U. S. Energy Research and Development Administration, Washington, DC 20545
113. R. Hoskins, Tennessee Valley Authority, 217 Electric Power Board Building, Chattanooga, TN 37401
114. C. H. Ice, Director, Savannah River Laboratory, E. I. du Pont de Nemours and Company, Aiken, SC 29801
115. R. H. Ihde, Manager, Contracts and Marketing, Babcock and Wilcox, P. O. Box 1260, Lynchburg, VA 24505
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120. H. Kouts, Nuclear Regulatory Commission, Washington, DC 20545
121. C. Kuhlman, Division of Nuclear Fuel Cycle and Production, U. S. Energy Research and Development Administration, Washington, DC 20545
122. J. A. Kyger, Associate Director, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439
123. F. W. Lewis, President, Middle South Utilities, Inc., Box 61005, New Orleans, LA 70161
124. W. H. Lewis, Nuclear Fuel Services, 6000 Executive Blvd., Rockville, MD 20952
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128. R. D. Oldenkamp, Rockwell International, Atomics International Division, 8900 De Soto Avenue, Canoga Park, CA 91304
129. G. B. Pleat, Division of Nuclear Fuel Cycle and Production, U. S. Energy Research and Development Administration, Washington, DC 20545
130. G. K. Rhode, Vice-President-Engineer, Niagara Mohawk Power Corporation, 300 Erie Boulevard, West, Syracuse, NY 13202
131. L. M. Richards, Nuclear Commercial Development Coordinator, Atlantic Richfield Company, Box 2679 - TA, Los Angeles, CA 90071
132. R. W. Roberts, Assistant Administrator for Nuclear Energy, U. S. Energy Research and Development Administration, Washington, DC 20545
133. A. E. Schubert, President, Allied-General Nuclear Services, P. O. Box 847, Barnwell, SC 29812
134. J. Shefeik, General Atomic Company, P. O. Box 81608, San Diego, CA 92138
135. N. F. Seivering, Jr., Assistant Administrator for International Affairs, U. S. Energy Research and Development Administration, Washington, DC 20545

136. L. E. Smith, Manager-Fuel, Carolina Power and Light Company, P. O. Box 1551, Raleigh, NC 27602
137. D. Spurgeon, Division of Nuclear Fuel Cycle and Production, U. S. Energy Research and Development Administration, Washington, DC 20545
138. A. Squire, Director, Hanford Engineering Development Laboratory, P. O. Box 1970, Richland, WA 99352
139. C. Stephens, Virginia Electric Power Company, Nuclear Fuel Service Department, 512 Franklin Building, P. O. Box 26666, Richmond, VA 32361
140. N. Stetson, Manager, U. S. Energy Research and Development Administration, Savannah River Operations Office, P. O. Box A, Aiken, SC 29810
141. S. Stoller, Western Reprocessors, The S. M. Stoller Corporation, 1250 Broadway, New York, NY 10001
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145. Dr. Uhrig, Florida Power and Light Company, P. O. Box 013100, Miami, FL 33101
146. E. J. Wahlquist, Deputy Assistant Director for Terrestrial Programs, U. S. Energy Research and Development Administration, Washington, DC 20545
147. A. K. Williams, Director, Nuclear Technology Division, Allied-General Nuclear Services, P. O. Box 847, Barnwell, SC 29812
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