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LWR Fuel Recycle Program Quarterly Progress Report April Through June 1977

**Compiled by
J. H. Jarrett**

August 1977

**Prepared for the Energy Research
and Development Administration
under Contract EY-76-C-06-1830**

Battelle
Pacific Northwest Laboratories

BNWL-2080-5

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BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 999352

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INTRODUCTION

This is the sixth in a series of quarterly progress reports on research and development studies performed by Pacific Northwest Laboratory (PNL) for the LWR Fuel Recycle Program. The LWR Fuel Recycle Program, sponsored by the U.S. Energy Research and Development Administration (ERDA), is designed to provide information needed by industry to close the back end of the commercial light water reactor (LWR) fuel cycle. Included in this program are activities in support of specific design studies as well as activities with more general application to fuel recycle technology. The overall program, managed by Savannah River Laboratory (SRL) under the direction of ERDA Savannah River Operations (SRO), is divided into the following categories. PNL conducts research in all but one of these categories.

- Economic and Environmental Studies*
- Spent Fuel Receipt and Storage*
- Head-End Processes*
- Off-Gas Treatment*
- Purex Process (solvent extraction)*
- Finishing Processes*
- Waste Management*
- Environmental Effects*
- Safeguards
- General Support*

This quarterly report summarizes PNL activities during the months of April, May and June, 1977. Previous reports in this series are BNWL-2052, BNWL-2080-1, -2, -3 and -4.

*Categories in which PNL is conducting research.

ECONOMIC AND ENVIRONMENTAL STUDIES

PROJECT TITLE: Economic Studies - Plutonium Strategy Analysis

PROJECT MANAGER: R. M. Fleischman, Nuclear Systems Analysis Section,
Energy Systems Department

PRINCIPAL INVESTIGATOR: D. F. Newman, Nuclear Systems Analysis Section,
Energy Systems Department

OBJECTIVE

The objective of this project is to provide a comprehensive evaluation of plutonium utilization strategies during the transition from a light water reactor (LWR) economy to a liquid metal fast breeder (LMFBR) economy. This comprehensive evaluation includes:

- characterization of both the LMFBR and LWR fuel cycles in detail so that the effects of the minor isotopes of plutonium on the fuel performance in each system are taken into account
- determination of the value of plutonium in either system
- evaluation of the benefits of various plutonium utilization strategies in the nuclear fuel cycle
- identification of decision points central to implementing the various plutonium utilization strategies and to avoiding logistical inadequacies in the nuclear fuel cycle for those strategies.

SUMMARY

- A computer simulation model was used to determine how changes in assumed U.S. electrical growth rates, uranium resources, and spent fuel recycle options affect the optimum pattern of future electrical supply, based on minimization of the total present-worth cost of building and operating all types of electrical power plants. The growth of the LMFBR was shown to be limited by the availability of plutonium from LWRs for starting up new plants, and by the compound-system doubling time of the LMFBR design. The installed capacities of both the LWR and LMFBR were found nearly independent of the total electrical demand.

ECONOMIC AND ENVIRONMENTAL ... contd

- An analysis was performed of the availability of plutonium to feed the LMFBF during the transition period after terminating Pu recycle in LWRs, as affected by various recycle options. The results showed that about 30% more plutonium would be available annually to supply the LMFBF between 1999 and 2006, after once-recycle of plutonium is stopped in 1999, than would be available in that period for a full-uranium-recycle scenario.
- The effect of plutonium isotopic composition on LMFBF fueling requirements was also analyzed. Plutonium to feed the startup and growth of the LMFBF during the transition from an LWR economy to a breeder economy was assumed to be supplied from LWRs. Plutonium fueling requirements for the initial cores of both a small pin LMFBF design with a two-year core lifetime and a large pin LMFBF design with a three-year core lifetime were calculated using various plutonium isotopic compositions typical of that contained in LWR spent fuel. The calculated feed requirements have been least-squares fitted to the plutonium isotopic compositions. The fitted coefficients are useful in determining the LMFBF fueling requirements for a plutonium isotopic composition that varies from those used in the study.

TRIPS AND VISITORS

- May 2 - 4, 1977 - R. M. Fleischman and D. F. Newman attended the ANS Topical Meeting on "The Plutonium Fuel Cycle" in Bal Harbour, Florida. Mr. Newman presented an invited paper on the work funded under this project. This paper was co-authored with Mr. Fleischman and M. K. White of ERDA Headquarters, Division of Waste Management Production and Reprocessing.
- June 13 - 16, 1977 - D. F. Newman attended the ANS Annual Meeting in New York, and participated in the standards committee working group, ANS19.2, to consider standardizing the definitions of the fuel cycle analysis terms "doubling time" and "breeding ratio."

TECHNICAL PROGRESSElectrical Supply Simulation

A linear programming model⁽¹⁾ was used to simulate the entire U.S. electrical economy with both mid-growth (4%/yr) and low-growth (2%/yr) assumptions for the future electrical capacity of all energy sources. The growth of LWR capacity is mainly dependent on the availability of low cost uranium (at prices less than \$100/lb U_3O_8). Figure 1 shows LWR growth capacity assuming no fuel reprocessing and high uranium resources (3.7 million short tons U_3O_8). Figure 2 shows the LWR growth capacity, also with high uranium resources, but assuming uranium recycle only. When the LMFBR is allowed to enter the electrical economy, as shown in Figure 3, it displaces high-cost fossil fueled plants. The growth of the LMFBR is limited by the availability of plutonium from LWRs for starting up new plants and by the compound-system doubling time of the LMFBR design (15 years for these studies). The cumulative economic benefits, through the year 2040, of the LMFBR entering the electrical economy in 1993, discounted at 10% per year back to 1977, are \$33 billion and \$20 billion for the mid-growth and low-growth scenarios, respectively. These benefits result from displacement of high-cost fossil fueled electrical generation.

The difference between scenarios for the mid-growth and the low-growth electrical demand projections is in the amount of fossil fueled electrical capacity installed. For the low-growth reference scenario, less than half of the electrical capacity installed between 1980 and 2040 is fossil fueled. For the mid-growth reference scenario, about two-thirds of the electrical capacity is fossil fueled. The installed capacities for both the LWR and the LMFBR are nearly independent of the total electrical demand.

Nuclear Fuel Logistics

Improved models for both uranium and plutonium recycle mass flows were incorporated into the logistic and economic analysis of typical 50-year nuclear growth projections. The logistic calculations were extended from 30 years to 50 years for the reference scenario;⁽²⁾ with full recycle of both uranium and plutonium in LWRs the cumulative fractional reduction in U_3O_8 demand increased from one-fifth for 30 years to one-third for 50 years. Both the 30-year and 50-year reductions are directly related to the assumed growth rates and the subsequent decline of LWR installed capacity during the next 50 years.

FIGURE 1. Optimum Electrical Mid-Demand Scenario with No Reprocessing, High Uranium Resources, No LMFBR

FIGURE 2. Optimum Electrical Mid-Demand Scenario with U Recycle but No Pu Recycle in LWRs, High Uranium Resources, No LMFBR

FIGURE 3. Optimum Electrical Mid-Demand Scenario with U Recycle but No Pu Recycle in LWRs, High Uranium Resources, 1993 LMFBR

An analysis was made of the availability of plutonium to feed LMFBRs during the transition period after terminating Pu recycle in LWRs, as affected by various recycle options. As shown in Figure 4, about 30% more plutonium is annually available to supply the LMFBR between 1999 and 2006 after once-recycle of plutonium is stopped in 1999 than would be available in that period for a full-uranium-recycle scenario. The transition period is completed 8 years after plutonium-recycle is stopped; the logistics follow the full-uranium-recycle scenario after the year 2007.

FIGURE 4. Plutonium Available Annually to Supply LMFBR

LMFBR Fuel Requirements

Plutonium fueling requirements for LMFBR core designs usually assume the use of equilibrium LMFBR plutonium isotopic composition as the feed material. This assumption applies, however, to the period after the growth rate in LMFBR capacity has slowed to less than the growth rate of plutonium in the LMFBR system; it does not address the effects of using plutonium from other sources during the early growth of the LMFBR system. Plutonium to feed the startup and growth of the LMFBR during the transition from a LWR economy to a breeder economy will probably be supplied from LWRs.⁽²⁾ It will even be necessary for the LWR to supply the LMFBR with plutonium for several years after LMFBR spent fuel reprocessing is in operation. This requirement is related both to the growth rate of LMFBR capacity and to the plutonium doubling time for the LMFBR design. The lower the growth rate and the shorter the plutonium doubling time for the LMFBR, the smaller the requirement for supplemental plutonium supply from LWR spent fuel.

The core design parameters used in this study were for 2500 MWt advanced oxide LMFBRs⁽³⁾ with low fissile inventory and compound system doubling time of less than 15 years. These parameters were obtained from Hanford Engineering Development Laboratory (HEDL). Plutonium fueling requirements for the initial core of both a small pin LMFBR design with a two-year core lifetime, and a large pin LMFBR design with a three-year core lifetime were calculated using various plutonium isotopic compositions typical of that contained in LWR spent fuel. The LMFBR reactor physics calculations were performed with the multi-group diffusion theory burn-up code 2DB⁽⁴⁾ in two dimensions. The core was burned using an iterative process so that the eigenvalue remained ≥ 1.0 during each annual cycle.

Table 1 compares LMFBR equilibrium plutonium requirements with LWR plutonium requirements, as feed to an LMFBR with a two-year core lifetime. The plutonium content of the fuel ranges from 15% to 19%. If the plutonium has a high ^{242}Pu content, the fueling requirements are correspondingly higher. A high ^{238}Pu or ^{240}Pu content does not increase the fueling requirements because these isotopes have appreciable fission cross sections themselves and are converted to fissile isotopes (^{239}Pu and ^{241}Pu) when they do capture a neutron. Also shown in Table 1 is the fissile content of the fuel for each

type of plutonium. Generally, the higher the fertile content (^{238}Pu , ^{240}Pu , and ^{242}Pu) of the plutonium, the lower the fissile requirements. When enriched uranium is used to start up the LMFBR, the ^{235}U atom fraction requirement is 0.1678. The calculated feed requirements have been least-squares fitted to the plutonium isotopic compositions. The fitted coefficients listed in Table 2 are useful in determining the LMFBR fueling requirements for a plutonium isotopic composition that varies from those used in this study. The coefficients indicate that ^{241}Pu is very valuable in fueling an LMFBR. ^{239}Pu , ^{238}Pu , ^{240}Pu , and ^{242}Pu are of decreasing value.

TABLE 1. Comparison of LMFBR Equilibrium Pu Requirements with LWR Pu Requirements as LMFBR Feed

Reactor	Source of Plutonium		Pu Atom Fraction	Fissile Pu Atom Fraction
		Discharged Fuel		
LMFBR		Equilibrium	0.1550	0.1142
PWR		1st UO_2	0.1488	0.1133
PWR		Equilibrium UO_2	0.1562	0.1082
BWR		2nd U Recycle	0.1512	0.0999
PWR		1st Pu Recycle	0.1748	0.0998
PWR		3rd Pu Recycle	0.1906	0.0927
PWR		3rd U Recycle	0.1524	0.0856

TABLE 2. Plutonium Fueling Coefficients for LMFBRs Fueled with LWR-Generated Plutonium

Isotope	A_i (a)
^{238}Pu	0.186 ± 0.005
^{239}Pu	0.121 ± 0.004
^{240}Pu	0.254 ± 0.014
^{241}Pu	0.061 ± 0.013
^{242}Pu	0.342 ± 0.017

(a) The plutonium fueling requirement from these coefficients, for a specific plutonium isotopic composition, is given by:
 atomic fraction Pu in LMFBR core =

$$\sum_{\text{all Pu isotopes}} A_i (\text{Pu isotopic fraction})_i$$

The results for the three-year core lifetime with the advanced large pin design are similar to the results presented in Tables 1 and 2 for the two-year core lifetime with the small pin design. The coefficient for ^{241}Pu is somewhat larger (0.070 vs. 0.061) because the longer residence time results in more ^{241}Am buildup. The effect of increased ^{241}Pu decays is a decrease in the number of atoms available for fissioning and an increase in fissile content to overcome the poison effect of ^{241}Am .

Open-cycle breeding ratios⁽⁵⁾ were calculated during the approach to equilibrium using either LWR-produced plutonium or enriched uranium to start up the LMFBR. Although the breeding capability of the LMFBR design can only be determined for closed, equilibrium fuel cycle operation, the open-cycle nonequilibrium breeding ratios can be significant quantities in a growing system of breeder reactors in which the mean age of the breeder is small.⁽⁶⁾ Our analyses indicate that the fueling requirements and breeding ratios for LMFBRs are very dependent on the isotopic composition of the plutonium. The fertile isotopes reduce the fissile requirement and increase the breeding ratio significantly.

PLANS FOR NEXT QUARTER

A summary of a paper entitled "Effect of Plutonium Isotopic Composition on LMFBR Fueling Requirements" will be submitted for presentation at the ANS Winter Meeting to be held in San Francisco, California November 27 to December 2, 1977. Work on plutonium fuel strategies in LWRs and LMFBRs is being redirected toward establishing systems studies in support of the Integrated Fuel Cycle Development Program. A topical report covering the work to date in FY-77 is expected to be issued at the end of the fiscal year.

PROJECT TITLE: Economic and Environmental Studies
in Support of LWR Fuel Cycle

PROJECT MANAGER: W. A. Reardon, Economic Analysis Section,
Energy Systems Department

PRINCIPAL INVESTIGATORS: Task 1 - W. A. Reardon, Economic Analysis
Section, Energy Systems Department
Task 2 - L. E. Addison, Computers and Infor-
mation Systems Section, Engineering Physics
Department

OBJECTIVE

The objective of this project is to conduct economic analysis of specific components of the LWR fuel cycle as required to support the SRL LWR Fuel Recycle Program. Initial tasks are to provide updated estimates of U_3O_8 costs, and to describe and use the "NUCOST" computer code.

SUMMARY

Attempts at disaggregating the linear program solution into component costs continued to lead to boundary problems that make the solution matrices quite unstable. Work on these problems constituted the effort on this project during this reporting period. Work on this particular approach has been stopped and will remain so unless new data provides a clue as to why the approach yields fluctuating results.

TRIPS AND VISITORS

None.

TECHNICAL PROGRESS

Recent work has centered on examining the mill construction component of uranium mine capital costs in Klemenic's \$15 COULD uranium mine case.⁽⁷⁾ Mill construction cost is one of four component costs used in this study. The object of this work is to reestablish the costs to the degree possible, on a yearly and plant basis. In Klemenic's paper, costs were allocated as spent rather than as capitalized. Reduction back to the basic costs will then allow the costs to be capitalized and used for modeling.

Our study approach was to allocate to the new production the funds spent for mill construction, in a manner consistent with the mill construction time schedule. The cost of the mill is a function of ore grade, mill size, and type of plant (i.e., whether the mill is "conventional," or its equivalent for "nonconventional" processes). The basic data was adjusted to a common basis using data on mill costs. The common basis chosen was a "conventional" mill producing one kiloton U_3O_8 concentrate per year from ore of grade 0.15%. Table 3 presents this adjusted basic data. The capital costs shown in the table are taken directly from Klemenic's table.⁽⁷⁾ The new production data was obtained by difference from cumulative production data.⁽⁸⁾ The values in parentheses for 1972 and 1980 are assumed to be extra new production to make up for a marked drop in ore grade of Class 1 reserves and are consistent with statements made by Klemenic⁽⁷⁾ in his paper.

TABLE 3. New Concentrate Production by Production
Center Class - \$15/lb COULD Case

Year	Production, Tons U_3O_8 in Concentrate, ($\times 10^3$)					Mill Construction Cost, Mega \$
	Class 1	Class 2	Class 3	Class 4	Total	
1976	-	-	-	-	-	131
1977	2	1	-	-	3	124
1978	2	2	1	-	5	84
1979	1(1.2)	1	0	1	3	96
1980	-	-	1	1	2	99
1981	2(.5)	1	1	1	5	51
1982	1	1	4	1	7	45
1983	1	-	-	-	1	56
1984	-	-	-	2	2	61
1985	1	-	-	2	3	65
1986	-	1	1	2	4	63
1987	-	-	-	3	3	43
1988	1	1	2	1.8	5.8	27
1989	-	.4	-	1.4	1.8	25
1990	-	-	.4	.2	0.6	19
TOTAL	11	8.4	10.4	16.4	46.2	989

Figure 5 shows the total capital cost for the basis plant for the various years of Klemenic's data. The fluctuation represents the variation which would be encountered if such a plant were built, assuming the corrections made were adequate. For completeness, the correction methods are given below. They can also be employed to reconstruct mill costs for plants other than the basis plant.

- Plant size: Plant sizes were only allowed to vary over three sizes, 0.5, 1.0, and 2.0 kilotons U_3O_8 per year. The size allocation was based upon guesses which were consistent with new production and with reducing the fluctuations observed in Figure 5. The cost of 1/2-kiloton plants was taken as 1.2 times as large as kiloton plants and as 1.2 times less than 2-kiloton plants.⁽⁸⁾
- Nonconventional Plants: Nonconventional plants were assumed to have mill costs 1.2 times greater because, generally, exploration and the equivalent of mining costs should be less. The total cost usually is competitive. Thus mill construction equivalent should be expected to be larger. The amount of new "nonconventional" production was available from Klemenic data.⁽⁷⁾
- Ore Grade: Corrections to the cost were made for ore grade on the basis of a constant cost of the mills on a tons-of-ore basis. This assumption is shown to be the case by Klemenic⁽⁸⁾ for grades from about 0.05 and up for the range of grades studied. The ore grade of new production was obtained from ore grade data given by Klemenic.⁽⁷⁾

PLANS FOR NEXT QUARTER

Next quarter we will complete and document the NUFUEL computer code.

FIGURE 5. Mill Construction Capital Cost (Adjusted to a common basis of size, type plant, and ore grade.)

SPENT FUEL RECEIPT AND STORAGE

PROJECT TITLE: Survey Current Technology of Fuel Handling Techniques

PROJECT MANAGER: R. S. Kemper, Materials Development Section
Materials Department

PRINCIPAL INVESTIGATORS: D. E. Blahnik, Applied Engineering and Development
Section, Physics Department
G. S. Allison, Materials Development Section,
Materials Department

OBJECTIVE

The objective of this project is to identify and assess potential improvements for spent fuel handling in predissolver equipment. This will be done by evaluating the spent fuel handling process and the conceptual design development, and by evaluating the demonstration and operation of prototype equipment. Alternative processes will be developed where significant improvements are identified in either plant efficiency or waste management.

SUMMARY

- Procurement action continues for the arc saw for fuel bundle end fitting removal development. Arrangements to perform additional end fitting removal trials on the manufacturer's prototype arc saw are being finalized.
- Fuel bundle design information from EXXON is being reviewed for release to public domain.
- The laboratory-scale blast equipment is being upgraded to perform decontamination cleaning tests on coupons with simulated contamination and subsequently with radioactive contamination. A search for a facility to perform radioactive decontamination tests is in progress.

TRIPS AND VISITORS

None.

TECHNICAL PROGRESS

Preprocurement planning for the arc saw has been completed and approved by ERDA-RL. Functional specifications have been sent to the manufacturer and he has responded with a proposal for the arc saw equipment. This proposal is now in the process of being negotiated. The appropriation request for the capital equipment items for the arc saw, namely the 3000 Amp DC power supply, the hydraulic drive power supply, and the arc saw control console, has been approved and funded from the Chop-Leach Cladding Residues Program budget. The prototype arc saw will be funded from the Spent Fuel Receipt and Storage Program FY-77 budget.

Arrangements are being completed for additional fuel bundle end fitting removal trials on the manufacturer's prototype arc saw in the FY-77 fourth quarter. Dummy fuel bundles, both PWR and BWR types, are being procured for these trials.

The fuel bundle design data that EXXON has agreed to furnish have been collected and are being reviewed by EXXON management. The data are expected to be released by the end of June.

The PNL laboratory blast equipment was modified to produce larger CO₂ crystals and to increase test run times. The modifications worked successfully, but further modifications are necessary to increase the blast pressure above 50 psi and to make the equipment perform more reliably. Components are being procured and facilities are being established to provide blast pressures up to 250 psi and improve equipment reliability. Test runs at 50 psi removed both lacquer and enamel paint coatings from metal plate. Similar tests to remove heavy oxide from stainless steel and inconel plate were not as successful.

A search has revealed that activated zinc sulfide or soap (e.g., Tide, Ivory Snow) may be suitable to use as stand-ins for radioactive materials during preliminary decontamination tests. The degree of cleanup with CO₂ blasting can be determined by examination with a black light.

Facilities at Hanford are being surveyed to determine where lab-scale decontamination tests can be made using specimens with radioactive contamination.

PLANS FOR NEXT QUARTER

Procurement activities for the arc saw and the fuel end fitting removal trials will be completed. The available fuel design data will be assembled into a document.

Other plans for next quarter include:

- completing the facility for lab-scale CO₂ blast equipment and improving its reliability
- performing preliminary decontamination tests using stand-in materials
- arranging for use of a facility to perform lab-scale tests with radioactive materials
- initiating design of full-scale blast cleaning equipment
- beginning negotiations for full-scale tests on shipping casks at a commercial facility.

HEAD-END PROCESSES

PROJECT TITLE: Alternatives to Voloxidation

PROJECT MANAGER: H. H. Van Tuyl, Applied Chemistry Section,
Chemical Technology Department

PRINCIPAL INVESTIGATOR: L. L. Burger, Applied Chemistry Section,
Chemical Technology Department

OBJECTIVE

The objective of this project is to review and evaluate alternative approaches to voloxidation for the removal of tritium and other volatile fission products from irradiated fuel prior to aqueous feed preparation.

SUMMARY

- The water recycle study being carried out by the General Electric Company under contract to PNL is progressing well. PNL water recycle studies are also continuing. The G.E. and PNL research show the use of water distillation as an isotopic enrichment adjunct to be an attractive concept.
- Dissolution of UO_2 in liquid N_2O_4 was examined further by using mixtures with polar solvents, primarily acetonitrile and propylene carbonate. Parameters measured were temperature and concentration of N_2O_4 . The dissolution rate appears adequate for process use, but practical problems of separating the solvent and removing the HTO that forms seriously limit the applicability. Current work emphasizes the use of molten nitrates for dissolution. Experiments using both metal nitrates and ammonium nitrate are reported.

TRIPS AND VISITORS

May 5, 1977 - L. L. Burger visited G.E. in San Jose to discuss the water recycle studies.

Water Recycle (L. L. Burger)

Previous reports^(9,10,11) have noted various factors which control the degree of water recycle attainable, and the levels of tritium that may be permitted. With these limitations, a variety of recycle schemes are possible. Some of the schemes described previously include the option of isotopic enrichment of the water storage stream. This option appears to be an attractive adjunct since with recycle it is not necessary to produce a highly depleted water stream, and the separative effort can be applied to the low volume "heads" stream.

Figure 6 is a diagram of a simplified recycle scheme without enrichment, for a Purex reprocessing plant. The major modification is contained in the first cycle aqueous waste (HAW) system. This includes HAW holdup tanks, an HAW concentrator and acid fractionator, acid storage tanks, a calciner, and an NO_x absorber.

FIGURE 6. Simplified Water Recycle Scheme (Circled letters refer to streams listed in Table 4.)

Table 4 shows flows and concentrations for three of many options considered. Tritium concentrations reach a maximum of about 2 Ci/ℓ. As shown, the maximum tritium loss is 5%, into the dissolver off-gas (DOG), and into the second cycle via the organic (HAP) uranium stream. It should be fairly easy to control this loss to below 5% through the use of a fresh water scrub, although even a 10% T discharge is probably acceptable. A second leakage term results from the ventilation system. To maintain a tritium loss of less than 5% would require limiting the 0°C saturated airflow to $1.5 \times 10^4 \text{ m}^3$ per day through the first cycle components.

TABLE 4. Simplified Water Recycle Flowsheet

Stream	Daily Quantities of Aqueous, Acid and Tritium											
	Case I				Case II				Case III			
	Stream Volume, ℓ	NO ₃ , kmole	HNO ₃ , M	Tritium Activity, Ci	Stream Volume, ℓ	NO ₃ , kmole	HNO ₃ , M	Tritium Activity, Ci	Stream Volume, ℓ	NO ₃ , kmole	HNO ₃ , M	Tritium Activity, Ci
A	12,000	78	3.0	25,200	12,000	78	3.0	22,140	12,000	78	3.0	10,270
B	17,400	48	2.76	32,680	17,600	48	2.73	28,500	17,600	48	2.73	10,840
C	4,000	12	3.0	7,580	4,000	12	3.0	6,410	1,000	3	3.0	620
D	12,000	36	3.0	22,200	12,050	36	3.0	19,240	12,050	45	3.75	7,370
E	1,400	0	0	2,900	1,550	0	0	2,850	4,600	0	0	2,850
F	--	48	0.15	100	--	48	0.15	50	--	48	0.15	50
G	0	0	0	0	50	--	--	100	50	--	--	100
H	2,000	6	3.0	0	2,000	6	3.0	0	5,000	15	3.0	0

The final water waste stream, stream E, with a volume of about 3000 ℓ/day for the illustrated cases, could be sent to an isotopic separation unit. The enriched stream would be sent to storage, and a depleted stream routed to stream C or H depending on the concentration. Even without enrichment, the 3000 ℓ/day volume is not an unreasonable quantity to store.

The General Electric recycle study, being carried out by G.E. under contract to PNL, is nearly completed. This study includes enrichment by water distillation.

Alternate Head-End Methods (R. D. Scheele, L. L. Burger)

Further studies were carried out on the dissolution of UO₂ using N₂O₄ systems. When certain organic solvents are added to N₂O₄, its reactivity is enhanced. This increased reactivity of N₂O₄ may be due either to ionization, if a high dielectric solvent is used, or to ion pair formation with donor solvents.

HEAD-END PROCESS contd

Several solvents have been examined in the present study, but attention has been primarily directed towards two very stable ones, acetonitrile, which acts primarily as an ionizing solvent with some donor properties, and propylene carbonate (PC), a highly polar donor molecule. In the latter case a possible reaction path can be represented by,

A high concentration of nitrate ion would aid in dissolving the oxidized UO_2 and increasing the reaction rate.

The chemistry of liquid N_2O_4 systems has been extensively investigated by C. C. Addison and co-workers, and reported in the Journal of the Chemical Society from 1949 to 1963. Addison's work indicates that the ratio of N_2O_4 to solvent is critical in influencing its rate of reaction with metals.⁽¹²⁾ Unfortunately, it does not appear possible to predict the optimum N_2O_4 concentration for a given system. PNL conducted two types of experiments to study concentration effects. The first employed solvent mixtures of the mole ratios, 2/8, 4/6, 6/4 and 8/2, at temperatures from 20°C to the maximum for each mixture. The second involved bubbling NO_2 through the solvent at temperatures varying from roughly 30° to 100°C. Typical results gave 30%-60% of a UO_2 pellet dissolved in 60 minutes for CH_3CN and PC, and 5%-20% for MeNO_2 . Attempts to optimize the conditions were generally unsuccessful due to poor reproducibility. In some cases the reaction product appeared to be a solvated uranyl nitrate. In other experiments a solid that was insoluble in water precipitated during the reaction. Attempts to remove the tritium from the solution by distillation were only partially successful due to the strong affinity of the uranium reaction product for water.

Thus, although an adequate dissolution rate is probably achievable with N_2O_4 solutions, the problems of potential hazardous reactions and the difficulty of separating the uranium from the solvent and from tritium make the applicability questionable.

To remove all the water would probably require calcination. If this step is considered, dissolution in a "minimum water" nitric acid dissolver may be more reasonable. Brief experiments indicate that, while UO_2 pellets dissolved very slowly in pure HNO_3 , the dissolution was reasonably rapid in 90% HNO_3 . Dissolution in 85-95% HNO_3 would keep the water content to less than 2 moles H_2O per mole of U and calcination would produce 15 ℓ H_2O per MTU.

Another possible method of dissolving fuels is through the use of molten salts to oxidize UO_2 to a uranate and effect fission gas release. This method has been discussed in past quarterly reports.^(9,10,11) Thermogravimetric analysis (TGA) of the $\text{NH}_4\text{NO}_3 - \text{UO}_2$ and $\text{NaNO}_3 - \text{UO}_2$ systems in air indicates reaction between the two components at $\sim 260^\circ\text{C}$ and 450°C , respectively. The reaction between NH_4NO_3 and UO_2 occurs after decomposition of the NH_4NO_3 has begun, while the reaction between NaNO_3 and UO_2 occurs prior to decomposition of NaNO_3 .

In recent experiments, a sample of the sodium uranate was treated with 1 M HCl to determine the extent of a reaction of the UO_2 with NaNO_3 . The sample dissolved completely, indicating complete oxidation of the UO_2 .

As was mentioned in the last report, x-ray diffraction did not positively identify the reaction products of UO_2 and molten nitrates with the exception of the silver uranate. The color, hardness, x-ray spectra and infrared spectra only indicate that the various uranates are greatly different from each other.

PLANS FOR NEXT QUARTER

- Water recycle studies will continue. G.E. will issue a report on their phase of the research in September or October.
- Plans also include a study of the reduction of the uranates with hydrogen or ammonia as a means of separating the uranium, e.g., from the sodium in Na_2UO_4 . V. I. Spitsyn⁽¹³⁾ has shown that the latter compound can be reduced to $\text{NaOH} + \text{UO}_2$.

OFF-GAS TREATMENT

PROJECT TITLE: Investigation of Air Cleaning Processes for Removing Tributyl Phosphate (TBP) from Off-Gas Streams

PROJECT MANAGER: L. C. Schwendiman, Particulate & Gaseous Waste Research, Atmospheric Sciences Department

PRINCIPAL INVESTIGATOR: G. B. Parker, Particulate & Gaseous Waste Research, Atmospheric Sciences Department

OBJECTIVE

The objective of this project is to provide a suitable air cleaning process for removing low concentrations of tributyl phosphate (TBP) vapors from nuclear fuel reprocessing off-gas streams.

SUMMARY

- Parametric studies investigating promising sorbents for removing TBP vapors have been initiated. Bench-scale experiments using granular alumina catalyst sorbent indicate a high retention of TBP vapor at 100°C bed temperature.
- A Hewlett-Packard 5736A gas chromatograph has been used for back-up analysis of airborne TBP. Chromatographic analyses indicate that quantities of TBP are being generated in the laboratory experimental apparatus as predicted by theory. Analysis is by direct injection of vapor samples from bench scale TBP generation system.
- Consideration has been given to broadening the scope of the work in 1977, in the area of TBP measurement in process streams. Several tasks have been identified and discussed with personnel at SRL and SRO, and are being evaluated.

TRIPS AND VISITORS

None.

TECHNICAL PROGRESS

Parametric experiments investigating promising sorbents for removing airborne TBP vapors were initiated in early April. Initial experiments are being conducted on a granular alumina catalyst material identified in early experiments as a promising sorbent for TBP. Three 1-in. diameter glass columns packed with 1.5 in. of the alumina material are being loaded with TBP vapors in an air stream flowing at 0.40 ℓ /min. The bed temperature is 100°C in the initial run. Inlet TBP concentration, calculated from vapor pressure data (and confirmed by gas chromatographic analysis), was 5×10^{-5} g/ ℓ at the start of the run. The inlet airborne TBP concentration has been subsequently increased to 3×10^{-4} g/ ℓ to accelerate breakthrough and decrease run time.

The alumina material has shown excellent retention of TBP at these conditions. No detectable breakthrough of TBP has been observed in the first 1.5 in. of material after 0.9 g was metered to the column. Minimum detectable concentration is approximately 0.01% of inlet concentration using the Meloy PA460 analyzer.

Preliminary work with the Hewlett-Packard 5736A gas chromatograph in analyzing TBP vapors has shown the chromatograph to be highly sensitive to TBP when using the nitrogen/phosphorus selective option to the flame ionization detector. The chromatograph has been used to confirm the calculated airborne TBP concentration generated in the laboratory experiments. The gas chromatograph will be used for back-up analysis to the Meloy PA460 phosphorus analyzer currently in use.

Several areas of work have been identified for consideration in 1977 that would broaden the scope of the current work plan. Three main tasks were identified from current work process involving measuring TBP. These tasks were designed to explore alternative methods to successfully measure TBP in actual process streams. These tasks are: (1) pursuing development of a prototype laser analysis instrument for in-situ measurement of TBP, (2) investigate methods and materials for sampling TBP in actual process streams using other

analysis techniques, and (3) a comprehensive study of flowsheets and consultation with engineers involved with effluent measurement and control at the Purex plant at Hanford and the LWR Waste Preparation Facility pilot plant at PNL. Initial development work with laser measurement techniques has shown promise.

PLANS FOR NEXT QUARTER

Parametric studies with promising sorbents will continue in the laboratory. Work with the gas chromatograph will continue with the development of a complete quantitative calibration of TBP vapor. Initial development work will begin to measure TBP vapors in actual streams by studying possible sources and methods for measurement.

PUREX PROCESS -- SOLVENT EXTRACTION

PROJECT TITLE: Study of Iodine Chemistry in Process Solutions

PROJECT MANAGER: H. H. Van Tuyl, Applied Chemistry Section,
Chemical Technology Department

PRINCIPAL INVESTIGATOR: J. L. Swanson, Applied Chemistry Section,
Chemical Technology Department

OBJECTIVE

The objective of this project is to study the chemical species and behavior of the iodine not volatilized from the dissolver solution, and to develop means of managing iodine in the various plant streams to minimize its release to the environment.

SUMMARY

Work this quarter again emphasized studies in the area of removal of iodine from dilute HNO_3 solutions simulating the low level aqueous waste stream.

- The use of a macroreticular polymeric adsorbent impregnated with manganese dioxide shows considerable promise. Good iodine removals were achieved with such materials with either iodide ion or elemental iodine in the feed; thus, no feed adjustment step is needed.
- Oxidative pretreatment of a macroreticular polymeric adsorbent was found to give an adsorbent that was less reactive with the sorbed elemental iodine, thus reducing the concentration of hydrogen peroxide needed for high iodine removals from solution.
- Additional consideration was given to analytical methods involving spectrophotometry, precipitation, and chromatography.

TRIPS AND VISITORS

None.

TECHNICAL PROGRESS

Process Chemistry (J. L. Swanson)

Emphasis continues to be directed at reducing the amount of iodine which is released with the low level aqueous waste stream. Work this quarter centered on removal of iodine from simulated waste streams using MnO_2 -impregnated macroreticular polymeric adsorbents. Data were also obtained using macroreticular polymeric adsorbents that had been treated in an effort to improve their effectiveness in removing iodine.

MnO_2 -Impregnated Polymeric Adsorbents

Adsorbents impregnated with manganese dioxide and tested previously were prepared using porous inorganic materials (e.g., porous glass and zeolites). We have recently been performing studies with MnO_2 -impregnated macroreticular polymeric adsorbent (Amberlite XAD-4) and have obtained very promising results. The first test involved material prepared by simply contacting the XAD-4 with a water solution of KMnO_4 for an hour or so, then rinsing and drying the MnO_2 -impregnated material. When this material was used in preliminary column tests, iodine DFs* in excess of about 200 were obtained with 0.05 M HNO_3 containing either 1×10^{-5} M KI or 0.5×10^{-5} M I_2 at flow rates of about two column volumes per hour. The fact that high DFs were obtained with iodide ion as well as with elemental iodine is very encouraging in that it means that no feed adjustment step would be required to obtain high iodine DFs for the iodine species expected to be present in the low level aqueous waste stream. Later tests also showed that the iodine DFs were affected little by flow rate (in the range 1-10 column volumes/hr, at least).

One disadvantage to the use of MnO_2 -impregnated macroreticular polymeric adsorbents is that the MnO_2 gradually dissolves, adding manganese to the waste solution and necessitating periodic regeneration of the column to maintain its effectiveness. Test results indicate, however, that the concentration of added manganese would be low, and column regeneration would be required only two or three times a week. Regeneration is easily accomplished by reaction of the polymeric adsorbent with potassium permanganate solution.

*DF = the ratio of activity in feed to activity in effluent.

Initial concerns about the safety of MnO_2 -impregnated polymeric adsorbents were greatly alleviated by the results of some differential scanning calorimetry tests. Gradual heating (in air) of MnO_2 -impregnated XAD-4 did not result in noticeable reaction occurring until the temperature reached about 220°C , which is well above that required for use in the proposed application. Similar tests with untreated XAD-4 did not result in noticeable reaction until the temperature reached about 310°C .

Long-term, continuous column tests with MnO_2 -impregnated Amberlite XAD-4 have been performed in which feed solutions were passed through the columns until the MnO_2 had dissolved off most of the column. The column was then regenerated with dilute KMnO_4 solution and feed flow was as resumed. The columns were initially prepared and regenerations were usually performed by 1) rapidly flowing the KMnO_4 solution to displace most of the residual solution, 2) stopping the flow and allowing an hour for reaction to occur (at room temperature,) 3) then rapidly displacing the solution again with fresh KMnO_4 solution and allowing another hour for additional reaction to occur.

Results of these long-term tests are summarized in Table 5, in the context of the conceptual flowsheet for low level aqueous waste treatment and discharge presented in Figure 7. In this flowsheet, the solutions resulting from the regeneration cycles and the post-regeneration rinse solutions are recycled back to a waste concentrator because they contain relatively high concentrations of iodine. All of the manganese added in the regeneration solutions ends up in the waste concentrator; the excess permanganate is first destroyed by addition of a reductant (e.g., hydrogen peroxide or hydroxylamine nitrate) to avoid oxidation of ruthenium in the concentrator. A small fraction of the effluent from the adsorption step returns to the waste concentrator in the vaporizer bottoms blowdown stream; this stream will contain the manganese resulting from dissolution of MnO_2 during the iodine-removal cycles. The bulk of the effluent is discharged to the atmosphere as water vapor. The relative flows of excess water, vaporizer bottoms blowdown, and intermediate level liquid waste used in this flowsheet are comparable to those planned for the proposed Exxon Nuclear reprocessing plant.

TABLE 5. Iodine Removal with MnO_2 -Impregnated Polymeric Adsorbent

Cycle	Volume of Composite Effluent, CV(a)		Iodine DF ^(b)		Length of Cycle, Days	Flow Range, CV/hr	Regeneration Solution		Rinse Solution Volume, CV	Iodine in Recycle, % of I in Feed	Manganese Concentration In Composite ILLW, M	
	In Cycle	Sum of All Cycles	During Cycle	Summed Over Cycles			KMnO ₄ Conc., M	Volume, CV			During Cycle	Summed Over Cycles
Feed = 0.05 M HNO ₃ Containing 1 x 10 ⁻⁵ M Added I ₂												
1	380	380	75	75	6	1-9	0.01	2.5	2.8	1.9	0.0066	0.0066
2	313	693	77	76	5	1-8	0.01	2.5	2.5	2.7	0.0040	0.0054
3	303	996	49	66	3	3-5	0.01	2.5	2.6	1.4	0.0041	0.0050
4	219	1215	44	60	4	1-5	0.01	2.5	2.9	3.6	0.0057	0.0051
5	348	1563	49	57	3	3-10	0.01	2.5	3.0	2.8	0.0036	0.0048
6	117	1680	30	54	3	1-10	0.01	2.5	2.9	7.1	0.0107	0.0052
7	247	1927	26	49	2	4-10	0.01	2.5	2.3	4.6	0.0051	0.0052
8	206	2133	26	45	2	4-8	0.01	2.5	3.4	5.5	0.0061	0.0053
9	110	2243	19	43	3	1-6	0.003	2.5	2.5	4.1	0.0034	0.0052
10	93	2336	29	42	1	4-6	0.01	2.5	2.8	11.4	0.0134	0.0055
11	240	2576	22	39	2	4-9	0.01	2.5	1.9	5.4	0.0052	0.0055
12	118	2694	13	36	1	4-9	0.01	2.5	2.1	9.2	0.0106	0.0057
13	95	2789	16	35	3	1-9	0.01	2.5	2.3	9.6	0.0132	0.0060
14	207	2996	16	33	2	4-8	0.01	2.5	2.7	6.2	0.0060	0.0060
Feed = 0.01 M HNO ₃ Containing 1 x 10 ⁻⁵ M Added I ₂												
1	982	982	160	160	12	1-10	0.01	1.5	1.8	0.7	0.0020	0.0020
2	359	1341	48	106	4	1-10	0.01	2.5	3.9	4.3	0.0035	0.0024
3	521	1862	55	88	3	4-9	0.01	2.5	8.3	2.2	0.0024	0.0024
4	243	2105	58	84	4	1-10	0.01	2.5	3.9	5.3	0.0051	0.0027
5	378	2483	42	75	3	4-9	0.01	2.5	3.8	3.0	0.0033	0.0028
6	114	2597	37	72	3	1-9	0.01	2.5	3.5	9.7	0.0110	0.0032
7	289	2886	35	67	4	1-5	0.01	2.5	2.9	4.0	0.0043	0.0033
8	91	2977	25	64	4	1-7	0.01	2.5	4.0	12.0	0.0137	0.0036
9	260	3237	38	62	3	2-5	0.02	2.5	3.3	9.8	0.0096	0.0041
10	225	3462	21	57	4	1-9	0.02	2.5	3.0	11.2	0.0111	0.0045
11	436	3898	26	52	3	4-9	0.02	2.5	3.5	7.1	0.0057	0.0046
12	241	4139	22	50	4	1-8	0.02	2.5	4.0	10.9	0.0104	0.0050

(a) CV = column volumes

(b) DF = (amount of iodine in feed) : (amount of iodine in effluent)

FIGURE 7. Conceptual Flowsheet for Low-Level Aqueous Waste Treatment

The most striking features of the data in Table 5 are that the iodine DFs tend to decrease as the columns are used longer (but the DFs are still substantial after thousands of column volumes), and that the use of the MnO₂-impregnated polymeric adsorbent is more attractive with a feed solution containing 0.01 M HNO₃ than one containing 0.05 M HNO₃. These columns were generally regenerated when the MnO₂ had dissolved off about 75% of the column, but variations were sometimes made to accommodate work schedules. The volume of solution that could be put through before regeneration was required increased with the increasing flow rate. Since relatively low flow rates were required to run through weekends in the equipment used in these tests, the throughputs achieved here were not optimized.

PUREX PROCESS contd

Potassium permanganate concentrations of 0.003 to 0.02 M were used in the regeneration solutions; the one test with 0.003 M indicated it to be too dilute to be very effective. During the regeneration and rinsing operations a small amount of iodine was removed from the column for recycling. This amount was usually less than 10% of the iodine in the feed during the cycle; the total volume of the solution to be recycled was also low.

According to the conceptual flowsheet in Figure 7 the concentration of manganese to be added to the composite intermediate level liquid waste (ILLW) was generally below 0.01 M during any one cycle, and even lower over all cycles. The manganese concentrations in the effluent samples were highest (about 3×10^{-4} M) immediately after regeneration, and then gradually decreased to low values (about 5×10^{-6} M).

More detailed presentations of some of the results are given in Figures 8 and 9, where the iodine concentrations in effluent samples are plotted against column volumes for several of the cycles listed in Table 5. The horizontal lines attached to some of the points indicate the volume span covered by that sample; the other samples were taken over relatively small volume spans.

In both of these figures it is seen that the concentration of iodine in the effluent is far from constant during a given cycle. It is especially interesting that the iodine removal near the end of the cycle (when relatively little of the column is impregnated with MnO_2) is considerably better than it is shortly after the column is regenerated. This fact gives hope that much better overall removals can be achieved when the chemistry of the system is better understood. Some flow rate data are also included in Figure 9, to illustrate the small effect of flow rate on iodine removal observed in these tests.

The last cycle shown on Figure 8 is of special interest. These data, which were obtained with no iodine present in the feed, indicate that much of the iodine found in the effluent in later cycles was iodine that had been sorbed in earlier cycles and was then washed off, rather than iodine which was not being sorbed from the feed during the later cycle. One importance of this fact is, of course, that the iodine DF observed in later cycles would decrease with decreasing concentration of iodine in the feed, possibly reaching unity

FIGURE 8. Iodine Removal from 0.05 M HNO_3 with MnO_2 -Impregnated Polymeric Adsorbent

FIGURE 9. Iodine Removal from 0.01 M HNO_3 with MnO_2 -Impregnated Polymeric Adsorbent

(or less) if the iodine concentration in the feed suddenly dropped appreciably lower than it had been in earlier cycles. Since the concentration of iodine in the effluent would not increase over what it was in earlier cycles, however, the rate of release from the plant would still be satisfactory.

Pretreated Polymeric Adsorbents

Earlier work with macroreticular polymeric adsorbents demonstrated that reduction of iodine (the sorbed species) to iodide occurs on the adsorbent, and that the presence of an oxidant (hydrogen peroxide) was necessary to attain high iodine DFs. It has now been found that pretreatment of the commercially available adsorbent with an oxidizing agent results in a decrease in the hydrogen peroxide concentration required to achieve a given iodine DF.

In the first test of this approach, the column was prepared by using hydroxylamine nitrate (in HNO_3) to remove the MnO_2 from the column used in the first test of the MnO_2 -impregnated approach described earlier. On subsequent passage of a feed solution containing $1 \times 10^{-5} \text{ M I}_2$ in $0.05 \text{ M HNO}_3 + 0.1 \text{ M H}_2\text{O}_2$, an iodine DF of about 350 was obtained at a flow rate of about two column volumes per hour. This compares with a DF of 32 reported in the previous quarterly report for untreated XAD-4 under somewhat similar conditions. Later results cast some doubt, however, on the magnitude of DF improvement attributable to the pretreatment.

Long-term, continuous column runs were then performed with both pretreated and as-received XAD-4 to obtain data at different H_2O_2 and HNO_3 concentrations and at different flow rates. In these runs data were obtained under one set of conditions, usually after operating for a day or so under the given conditions. The conditions were then adjusted, and data were again obtained. As apparent inconsistencies began to develop in the results of these runs, it was found that here, too, (as in the MnO_2 -impregnated tests) the results were being strongly affected by the washing-off of iodine which had been adsorbed earlier in the runs. The washing-off of iodine appears to increase as the amount of freshly adsorbed iodine increases; as the time since adsorption increases, the effect becomes less pronounced. Because of this effect, it appears that quantitative comparisons of pretreatment effectiveness (and of compositional variations) will require use of a number of columns, each run at a given set of conditions, rather than using one column under a variety of conditions.

Analytical Methods (J. L. Cox)

The investigation of a spectrophotometric technique to determine iodide (I^-) and iodine (I_2) in the ppb range has been initiated. The method involves the formation of crystal violet through selective oxidation of leuco crystal violet by hypiodous acid (HOI) that is produced from iodide or iodine by oxidation with N-chlorosuccinimide in a buffered solution. The high sensitivity of the method is due to the high molar absorptivity (4.2×10^6 at 592 nm) of the crystal violet. Five cm cells have been used to extend the sensitivity even further; good adherence to Beer's Law was found at concentrations as low as 5×10^{-9} M I^- .

A theoretical assessment of a precipitation technique in combination with atomic absorption analysis for I^- determination has been carried out. The crux of the hypothetical analytical method is that the initial I^- concentration could be calculated (assuming equilibrium was established) by measuring the difference between the initial and final Ag^+ concentration when added to an unknown concentration of I^- . Calculations revealed that successful application of the technique depended upon an a priori knowledge of the approximate I^- concentration. Furthermore, in the 10^{-7} to 10^{-8} M I^- range it would be necessary to accurately and reproducibly determine the Ag^+ concentration differences of less than 10%. In view of these difficulties, further pursuit of this approach has been abandoned.

Finally, a photoionization gas chromatography detector has been ordered. This detector is reported to be superior to the flame ionization, flame photometric, thermal conductivity, and electron capture detectors commonly employed. It has sensitivities down to 0.001 mg, and a wide dynamic range and response to most organics and some inorganics, including HI, I_2 , and organic iodides. It is anticipated that some of the problems previously encountered with the electron capture detector in measuring volatile iodine compounds will be alleviated with this detector.

PLANS FOR NEXT QUARTER

Efforts will be continued in the areas discussed above.

FINISHING PROCESSES

PROJECT TITLE: Mixed-Oxide Studies

PROJECT MANAGERS: Tasks 1 and 5 - S. Goldsmith, Fuels Design and Development Section, Materials Department
Task 3 - W. S. Kelly, Applied Engineering and Development Section, Engineering Physics Department

PRINCIPAL INVESTIGATORS: Task 1 - D. W. Brite and W. J. Baily, Fuels Design and Development Section, Materials Department
Task 3 - D. E. Blahnik, Applied Engineering and Development Section, Engineering Physics Department
Task 5 - W. J. Coleman, Physical Measurements and Electro-Optic Systems Section, Engineering Physics Department

OBJECTIVES

The objectives of the three tasks are:

- Task 1 - to establish the data base required to formulate product specifications for mixed-oxide fuels
- Task 3 - to evaluate development needs for mixed-oxide fuel fabrication processing and materials handling equipment
- Task 5 - to develop a conceptual automatic fuel pellet inspection process.

SUMMARY

- Studies under Task 1 were resumed during this reporting period in an effort to make the draft of the data base report on mixed-oxide (UO_2 - PuO_2) fuels more extensive.
- For Task 3, the fuel supplier survey summary was updated to incorporate responses received since February.
- Under Task 5, effort concentrated on identifying fuel pellet quality assurance/quality control parameters, and on investigating equipment and techniques for fuel pellet inspection.

FINISHING PROCESSES contd

TRIPS AND VISITORS

June 10, 1977 - S. Goldsmith (PNL) was at SRL for program discussions with R. T. Huntoon.

TECHNICAL PROCESS

Task 1 (D. W. Brite and W. J. Bailey)

Efforts in Task 1 were resumed during this reporting period to make the draft of the data base report on mixed-oxide ($\text{UO}_2\text{-PuO}_2$) fuel more extensive. In particular, work was restarted in the area of the effects of impurities in UO_2 and PuO_2 powders and in the $\text{UO}_2\text{-PuO}_2$ fuel pellet.

Task 3 (D. E. Blahnik)

The fuel supplier survey summary was updated to incorporate responses received since February. Thank-you letters with copies of the survey summary were mailed to all survey participants. Work has been initiated on a final report.

Task 5 (W. J. Coleman and F. R. Reich)

During the initial part of this reporting period, program progress was cut back due to a delay in receiving full funding for this task. Efforts during this reporting period have been concentrated on:

- literature search to identify fuel pellet QA/QC (Quality Assurance/Quality Control) parameters and basic inspection method and techniques
- identification of capital equipment to support conceptual gauging/inspection techniques
- formulation of specific fuel pellet inspection parameter QA/QC tolerance descriptions.

Current fuel pellet inspection techniques use both a sampling and 100% throughput inspection format. With the current pellet inspection, surface integrity inspection and large PuO_2 particle detection are the only 100% throughput inspection tasks. All other fuel pellet parameters are verified through programmed sampling from each pellet batch. To support the selection of gauging/inspection concepts, generic characteristics of nuclear fuel

pellet inspection parameters are being identified. For example, in visual inspection for surface condition, the inspection criteria is separated into chip and crack designations. For chips the accept/reject descriptor is defined by area and depth limits while for cracks the primary accept/reject descriptor is length with depth as a threshold condition.

Current homogeneity inspection task methods are being analyzed for potential automation and simplification. The current inspection method includes macro- and micro-homogeneity sampling of select pellets representative of a batch with alpha autoradiography, microprobe scanning and microscopy analysis. These processes are slow, expensive and do not lend themselves directly to automation and large volume manufacturing.

PLANS FOR NEXT QUARTER

- Task 1 - We plan to complete the data base report during the next quarter.
- Task 3 - Plans are to continue working on the final report.
- Task 5 - During the next reporting period the identification and classification of fuel pellet parameters and conceptual gauging schemes will continue. Conceptual system designs for visual and dimensional inspection will be initiated and homogeneity inspection techniques identified.

WASTE MANAGEMENT

PROJECT TITLE: Electropolishing to Decontaminate LWR
Fuel Recycle Metallic Waste

PROJECT MANAGER: R. D. Nelson, Metallurgy Research Section,
Materials Department

PRINCIPAL INVESTIGATORS: R. P. Allen, H. W. Arrowsmith, Metallurgy
Research Section, Materials Department

OBJECTIVE

The objective of this project is to develop and provide the concepts, information and engineering data needed by the LWR Fuel Recycle Program to design and operate an electropolishing facility that will decontaminate surface-contaminated solid metallic waste. Major program tasks include:

- the transfer of compatible technology from the commercial electropolishing industry
- the development of solution treatment procedures to extend electrolyte life and minimize secondary waste volume
- the development of removal techniques for entrained contamination.

SUMMARY

- Technology transfer activities continued with a visit to a large electroplating facility with the type of automated equipment and control system required for the high-volume electropolishing of surface-contaminated metallic waste.
- Liquid-liquid solvent extraction techniques appear very promising as a purification method for plutonium-contaminated phosphoric acid electrolytes. Tests using methyl isobutyl ketone show reductions of more than 80% in plutonium concentration.
- Removal of copper from spent electrolytes by a sulfide precipitation process was demonstrated.

WASTE MANAGEMENT contd

- Progress was also made in developing enhanced removal techniques for the potentially contaminated recast layer on cut metal edges. An edge removal rate more than six times the corresponding uniform surface removal rate was achieved by using high current densities and a close cathode spacing.

TRIPS AND VISITORS

April 12, 1977 - R. P. Allen and H. W. Arrowsmith inspected automated plating equipment and discussed automation technology with H. Holden from Hewlett-Packard Company, Palo Alto, CA.

April 14, 1977 - R. P. Allen and H. W. Arrowsmith visited M. D. Boersma and P. H. Permar at SRL to discuss applications of electrochemical decontamination techniques.

April 21, 1977 - M. D. Boersma of SRL visited PNL to review program progress and inspect the recently completed 400-gallon electropolishing facility.

June 6, 1977 - P. H. Permar and T. S. McMillan of SRL visited PNL to discuss the design of electrochemical decontamination systems.

TECHNICAL PROGRESS

Technology Transfer

Electropolishing is a well established industrial process widely used to clean, deburr and surface finish various metals and alloys. One of our program objectives is to use existing electropolishing and electroplating technology and equipment wherever possible in the design, construction and operation of electrochemical decontamination systems. Our technology transfer activities continued during this reporting period with a visit to a large, automated electroplating facility used by the Hewlett-Packard Company for the high-volume electroplating of circuit boards and other electronic components. This facility has 46 separate stations serviced by three programmed hoists and can process almost 2000 ft² of material per shift with only 2 1/2 operators to load and unload the plating racks. The entire operation is computer controlled, including the independent movement of the racks through

the plating stations, monitoring of all process variables, addition of chemicals as required, and preparation of the production log. Plant personnel noted that the automated equipment, despite its size and complexity, is relatively maintenance-free and that an automated electropolishing system of the type required for the high-volume decontamination of metallic waste could be controlled by an inexpensive desktop computer system.

Solution Treatment Studies

Solution treatment studies are in progress to extend electrolyte life and minimize secondary waste volume by developing techniques for removing dissolved metal and contamination from the electrolyte and rinse solutions. This work includes an evaluation of both energy separation (distillation, electrolysis) and mass separation (solvent extraction, precipitation/neutralization, ion exchange) methods. Of the studies to date, liquid-liquid solvent extraction appears to be the most promising approach. A series of single extractions using methyl isobutyl ketone to separate the impurities from 75% phosphoric acid shows the following results:

- approximately 60% to 79% reduction in dissolved metal concentration (Fe, Cr, Ni) for solutions containing up to 6 wt% metal
- greater than 80% reduction in plutonium concentration for electrolyte containing dissolved metal and up to 1.0 g/l of plutonium either in the form of oxide or introduced by anodic dissolution
- recovery of 58 to 65 vol% purified acid for recycle with only 35 to 42 vol% of the original acid rejected as process residue.

These results suggest a reprocessing system that would be capable of purifying the contaminated electrolyte at ambient temperature using a suitable organic solvent. The process would (1) recycle the raffinate, or waste acid fraction, with final neutralization and disposal of the raffinate, (2) water strip the purified acid fraction from the organic phase, (3) recycle the organic, (4) concentrate the purified acid by distillation and (5) recycle the distilled water to the organic stripper and the concentrated acid back to the decontamination facility.

Studies were also conducted to evaluate sulfide precipitation using hydrogen sulfide to remove selected metal ions from spent 75% phosphoric

acid electrolyte. The pH of the acid was varied from 0 to 2.5 by dilution and partial neutralization to enhance the precipitation of sulfides. The test results showed only a minimal reduction in the iron content of the electrolyte and no reduction in the nickel content. The only precipitates that did form were principally copper sulfides. The ability to remove copper from spent electrolyte is important, however, since the erosion of copper current contacts often introduces this ion into the electrolyte and some waste treatment systems (as at Hanford) cannot accept liquid waste containing copper.

Entrained Contamination Studies

Metallographic studies have shown that a potentially contaminated recast layer up to 12 mils thick forms on cut edges produced by arc saw and plasma torch cutting operations. The ability to minimize or remove this recast layer will directly affect the design of electrochemical decontamination systems with respect to tank size versus the amount of component disassembly and sectioning work required to prepare the metal waste for processing. The metal dissolution rate at the edges is higher than that for a planar surface (edge-effect), for normal electropolishing conditions. A series of edge-effect tests was conducted using the 400-gallon electropolishing facility to measure the metal removal rates at the edges and center of a 2 ft x 1 ft x 0.125-in. thick stainless steel plate to determine if standard electropolishing treatments can remove this recast layer.

The initial edge-effect tests were conducted in 75% phosphoric acid electrolyte at a current density of 100 A/ft^2 . The metal plate was positioned vertically with one edge 2 in. from the tank wall (cathode). The other edges and the center of the plate were more than 1 ft from the nearest cathode surface. The metal removal rate for the edge nearest the cathode was 3.6 times the rate at the center of the plate, as compared with an edge/center metal removal rate ratio of 1.8 for the edge in the center of the tank.

The same tests were repeated with the current density increased from 100 to 300 A/ft^2 . This increased the edge/center metal removal rate ratio to 6.2 for the close cathode spacing and to 4.0 for normal cathode spacings. Operating at increased current densities thus produced a substantial enhancement in the relative edge erosion rate. More importantly, these results show

that it should be possible through proper control of electropolishing variables to preferentially remove the recast layer produced by arc saw and plasma torch cutting operations while only removing the 1 or 2 mils from planar surfaces needed for decontamination.

PLANS FOR NEXT QUARTER

The solution processing studies will continue, with emphasis on precipitation-neutralization and electrolysis methods. An assessment of ion exchange and adsorption techniques will also be initiated. Further solvent extraction tests will be deferred until plutonium-contaminated electrolyte is available from the operation of the 400-gal electropolishing facility. Additional edge-effect tests will be conducted to investigate the effect of cathode geometry and electrolyte composition on the relative edge erosion rates.*

*Additional information on electropolishing as a decontamination technique is available in the PNL Nuclear Waste Management Quarterly Progress Reports. The latest issue is BNWL-2290.

ENVIRONMENTAL EFFECTS

PROJECT TITLE: U.S. Scale Transport, Dispersion,
and Removal Model Comparisons

PROJECT MANAGER: C. L. Simpson, Manager
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PRINCIPAL INVESTIGATORS: L. L. Wendell, W. E. Davis
Atmospheric Chemistry & Physics Section

OBJECTIVE

The overall objective of this work is to provide estimates of the confidence limits for radionuclide surface air concentrations and deposition amounts computed by U.S. scale assessment models. These models simulate the processes of atmospheric transport, diffusion, dry deposition and precipitation scavenging. The approach to this problem involves using parallel models that incorporate state-of-the-art treatments of the basic parameters in short-term comparison tests.

SUMMARY

- Comparisons have been made of the rates of wet and dry radionuclide removal in U.S. scale assessment models for April 1-15, 1974. Tests were made measuring hourly precipitation, precipitation over a period of six hours, and semimonthly precipitation. The results show a significant decrease in the amount of wet deposition when using the average hourly precipitation versus the average semimonthly precipitation.
- Two assessments were made of an SRL simulated radionuclide release using the U.S. scale assessment mode. One assessment was made from January 26 to February 25, 1975 and the other from June 4 to July 3, 1975. The results of these cases included data on depositing and non-depositing pollutants, and will be compared with ARL and LLL results.
- The method of estimating vertical diffusion in the U.S. scale assessment model was modified. Eimutis' and Konicek's parameters were used out to 100 km. Beyond 100 km, vertical dispersion was derived from

ENVIRONMENTAL EFFECTS contd

$\sqrt{2K_z T}$. The results showed a decrease in the surface air concentration to 100 km. Beyond 200 km the air concentrations were generally higher.

TRIPS AND VISITORS

April 26, 1977 - John Switch and Jack Corey from SRL visited W. E. Davis to discuss tape formats.

TECHNICAL PROGRESS

Precipitation Effects

One of PNL's research objectives is to identify, through computing wet deposition, those elements that would significantly alter results of assessment models. Previously, annual precipitation averages have been used in looking at U.S. scale assessments.⁽¹¹⁾ When these averages were applied to an assessment for April 1-15, 1974 for SRL, deposition values >90% of the amount released were obtained. To test how representative these values were, hourly precipitation tapes were produced. Using these hourly values, a reassessment was made in which the amount of radionuclide deposited by precipitation alone decreased to 52%. The next tests were made to determine the effect of using intermediate time averages. A first test using the average of the hourly data for April 1-15, 1974 resulted in a deposition increase from 52% to 75% of the amount released. A second test using six-hour averages showed a deposition increase from 52% to 56.6% of the total amount released. Other tests will be conducted incorporating the differences in the station density between the hourly cooperative observer network and the synoptic network. The cooperative observer network has significantly higher station density.

Table 6 summarizes the results of these tests. The purpose of these analyses is to estimate the variability in wet deposition rates caused by time and space averaging of precipitation data. Averaging of precipitation data causes error mainly because, with long-term average precipitation, wet removal occurs continuously throughout the averaging period rather than less frequently when interaction between precipitation and plumes actively occurs.

TABLE 6. Effects of Precipitation Averaging Time on Wet and Dry Deposition

Precipitation Averaging Time	Washout Ratio	Dry Deposition Velocity, cm/sec	Total Deposited, %	Total Wet Deposited, %	Total Dry Deposited, %
Dry Case	0	1	82	0	82
1 hr	5×10^5	0	52	52	0
1 hr	5×10^5	1	84.4	-- (a)	-- (a)
360 hr	5×10^5	0	75	75	0
360 hr	5×10^5	1	95.5	-- (a)	-- (a)
6 hr	5×10^5	0	56.6	56.6	0
6 hr	5×10^5	1	90.6	30.0	60.6
6 hr ^(b)	5×10^5	1	79.7	31.0	48.7

(a) Yet to be determined.

(b) No dry deposition allowed over oceanic areas.

Wet Versus Dry Deposition

Another problem addressed this quarter was the percentage of the total radionuclide deposition that can be attributed to precipitation scavenging in single layer models. The series of comparisons shown above in Table 6 indicates the relative importance of wet removal during a short test period. The first case compared wet removal averaged over a six-hour period of precipitation with the dry removal. The results indicated that wet removal was ~50% of dry removal for the period listed as the second entry from the bottom of Table 6. This value appeared to be biased, since precipitation values for six-hour periods were only available over land while dry deposition occurred continuously over land and water. When any dry removal was excluded over ocean areas, the amount of wet removal increased by 1%, while dry removal decreased from a deposition of 61.6% to 48.7% of the total assumed radionuclides released (see last entry in Table 6). With this decrease, the wet removal was 64% of the dry removal.

ENVIRONMENTAL EFFECTS contd

A gross calculation was made to compare the effective wet deposition velocity for the reference time period, April 1-15, 1977. Assuming that the pattern of removal was uniform, the rainfall rate was constant, and the air concentration was uniformly distributed over a 1000 meter layer, an estimate was made of an effective wet deposition velocity. This was done by first using a value of 82% for radionuclides deposited over the grid by the dry deposition velocity of 1 cm/sec. Next, an effective time of 1.7×10^5 seconds was calculated. Applying this same time to the wet deposition of 52% of the total released, a wet deposition velocity of $\sim .4$ cm/sec was calculated. Using the wet deposition combined with 1 cm/sec dry deposition of 30% of the total released, the effective wet deposition velocity was reduced to $\sim .3$ cm/sec. This contrasts sharply with the wet deposition velocity of 2 cm/sec calculated using the climatological annual average precipitation. More work is planned in this area to obtain a more statistically representative result.

A question yet to be answered is whether a model that includes vertical motions will produce results significantly different from those of the simpler model, which does not include vertical motions. Current test runs use a model which incorporates vertical motion through isentropic considerations. This model will be used to further clarify the plume interactions with precipitation as well as dry deposition. A plume moving vertically over a front should result in less dry deposition.

Assessment Runs

Assessment runs were made for January 26 - February 25, 1975 and for June 4 - July 3, 1975 from data on a simulated release at SRL with a release rate of 1 Ci/sec at a height of 62 m. The single layer model was run with a mixing depth of 650 m⁽¹⁴⁾ for the January 26 - February 25, 1975 case, and with a mixing depth of 1500 m for the June 4 - July 3, 1975 case. Both cases were run for a nondepositing tracer and a depositing tracer with a dry deposition velocity of 1 cm/sec. Figures 10 and 11 show the sharp seasonal contrast in the air concentration patterns for these assessment computer runs. Results were written to tape and delivered to SRL for comparison with ARL model results. Later, the data will be investigated to determine if interface problems exist between the U.S. scale models and the regional scale model of LLL.

FIGURE 10. Surface Air Concentrations for Jan. 26 - Feb. 25, 1975 for a Continuous Hypothetical Release of 1 Ci/sec of a Non-depositing Tracer from the Savannah River Laboratory

FIGURE 11. Surface Air Concentrations for Jun. 4 - Jul. 3, 1975 for a Continuous Hypothetical Release of 1 Ci/sec of a Non-depositing Tracer from the Savannah River Laboratory

ENVIRONMENTAL EFFECTS contd

Diffusion Code

A modification in the diffusion code was completed on the U.S. scale model. Assessment runs were made using the Eimutis and Konicek representation of vertical diffusion to a distance of 100 km. Beyond 100 km vertical diffusion was represented using $\sqrt{2K_z t}$ where K_z is the vertical diffusion coefficient and t is time since release. The main effect of this new formulation was a decrease of ~25% in surface air concentrations to 100 km. Beyond 200 km, the surface air concentrations were ~20% higher with the new formulation. This increase was strictly due to the change in formulations.

PLANS FOR NEXT QUARTER

Work next quarter will include:

- formulating an effective washout ratio for use in long-term precipitation averages
- continuing sensitivity analyses of the U.S. scale assessment model results to different averaging times and station densities of the available precipitation data
- running the updated isentropic model for the April 1-15, 1974 test period using hourly precipitation
- reviewing new developments in dry deposition and re-entrainment.

GENERAL SUPPORT

PROJECT TITLE: Criticality Safety and Shielding

PROJECT MANAGERS: E. D. Clayton, Criticality Experiments
C. L. Brown, Criticality and Shielding Analysis

PRINCIPAL INVESTIGATORS: R. C. Lloyd, Criticality Experiments
D. R. Oden, Criticality Analysis
M. G. Zimmerman, Shielding Analysis

OBJECTIVE

The objective of the criticality safety support task is to:

- utilize the PNL Critical Mass Laboratory to provide confirmatory criticality data required to support alternative fuel cycle R&D
- establish benchmark data for code validation and safety assessment
- provide criticality safety parameters to support design concepts and equipment specifications
- provide support in the area of radiation shielding analysis.

SUMMARY

- Preparation of materials and equipment for criticality experiments continued this quarter.
- Plutonium source term data were developed and have been presented in a document now in publication.

TECHNICAL PROGRESS

Criticality Experiments

The major activities during this period were materials planning, the fabrication of experimental equipment, and preparation of the laboratory equipment for use in carrying out the experimental program. The engineering design drawings were completed for the criticality vessels and lattice templates. Fabrication of this equipment is about 90% complete.

Planning for the materials to be used in the experiments is progressing. Criticality safety specifications were completed for handling fissile materials during dissolution of the UO_2 into uranyl nitrate solution. The process equipment for carrying out the dissolution has been completed. Criticality safety specifications have also been completed for storage, handling and shipping of the $\text{U}(4.3)\text{O}_2$ rods to be used in the experimental investigations. The storage units for these rods are now finished.

Laboratory equipment is now being prepared for use in the experimental program. Such equipment includes hoods, liquid level measuring equipment, pumping equipment and associated piping. Planning and analysis for storage of the fissile solutions are in progress.

Shielding

A review of the results of past work and of literature available at PNL has been performed to obtain the data needed to develop plutonium source terms. These data include isotopic composition as a function of number of plutonium recycles, and radiation source terms for each plutonium isotope. The final plutonium isotopic compositions are averaged from some twelve sources for the equilibrium uranium fuel cycle and from four plutonium recycles. These results are shown in Table 7. Radiation source terms based on these isotopic compositions and used as the basis for shielding calculations should provide reasonable estimates of dose rates from recycled plutonium. The plutonium isotopic compositions, radiation source terms and a discussion of plutonium shielding are being documented.

TABLE 7. High Burn-Up Plutonium Isotopic Compositions Recommended for Use in Shielding Analysis

Isotope	Composition at End of Cycle, wt%				
	Equilibrium Uranium Cycle	First Pu Recycle	Second Pu Recycle	Third Pu Recycle	Fourth Pu Recycle
^{236}Pu	0.08 ppm	0.06 ppm	0.06 ppm	0.06 ppm	0.06 ppm
^{238}Pu	2.	2.5	3.5	4.	5.
^{239}Pu	57.	43.	37.2	29.	26.
^{240}Pu	24.	28.5	30.2	29.	26.
^{241}Pu	12.	16.	16.1	19.	19.
^{242}Pu	5.	10.	13.	19.	24.

This task is complete and a document is in the publication process. Comments on an initial draft have been received from SRL and PNL internal review and have been incorporated into the document. Plutonium source term data from three additional references brought to our attention during the literature review have been included and discussed in the document.

PLANS FOR NEXT QUARTER

Fabrication of experimental equipment for use in the initial program will be completed and preparation of laboratory equipment for experiments will continue.

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