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Quarterly Progress Report LWR Fuel Recycle Program

July - September 1976

October 1976

Prepared for the Energy Research and Development Administration under Contract E(45-1)-1830

Battelle
Pacific Northwest Laboratories

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Under Contract E(45-1)-1830

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy $4.50; Microfiche $2.25

BNWL-2080-2
UC-78

QUARTERLY PROGRESS REPORT
LWR FUEL RECYCLE PROGRAM

July - September 1976

Compiled by J. H. Jarrett

October 1976

BATTELLE
Pacific Northwest Laboratories
Richland, Washington 99352

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QUARTERLY PROGRESS REPORT
LWR FUEL RECYCLE PROGRAM
APRIL - JUNE 1976

INTRODUCTION

This is the third in a series of Quarterly Progress Reports on research and development studies for the LWR Fuel Recycle Program[a] being performed by Battelle-Northwest at the ERDA-Pacific Northwest Laboratory (PNL). The LWR Fuel Recycle Program is designed to assist in the commercialization of the LWR fuel cycle. Included in this program are both activities in support of specific design studies and other activities of more general applicability to fuel recycle technology. The overall program, which is managed by Savannah River Operations and Savannah River Laboratory (SRO-SRL),[a] is divided into the 11 categories listed in Table 1. PNL presently has research in eight of these categories as indicated by asterisks in the table below.

TABLE 1. LWR Fuel Recycle Program Categories

1 - Liaison and Program Coordination
*2 - Economic and Environmental Documentation
*3 - Spent Fuel Receipt and Storage
*4 - Head End Processes
*5 - Off-Gas Treatment
*6 - Purex Process (Solvent Extraction)
*7 - Finishing Processes
8 - Waste Management
*9 - Environmental Effects
10 - Safeguards
*11 - General Support

* Categories where PNL is conducting research.

(a) Sponsored by the Energy Research and Development Administration (ERDA) Division of Nuclear Fuel Cycle and Production.

This report summarizes PNL activities during the reporting period of April through June. The previous reports in this series are BNWL-2052 and BNWL-2080-1.

CATEGORY 2 - ECONOMIC AND ENVIRONMENTAL DOCUMENTATION
(F-RL-14-007)

PROJECT TITLE:	Economic and Environmental Studies in Support of LWR Fuel Cycle
PROJECT MANAGER:	R. M. Fleischman, Nuclear Systems Analysis Section, Nuclear Technology Department
PRINCIPAL INVESTIGATORS:	Task 1 - W. E. Reardon, Economic Analysis Section, Systems Department
	Task 2 - L. E. Addison, Information Systems Technology, Systems Department
	Task 3 - D. F. Newman, E. T. Merrill and R. M. Fleischman, Nuclear Systems Analysis Section, Nuclear Technology Department

OBJECTIVES

Conduct economic analysis of specific components of the LWR fuel cycle as required to support the SRL LWR Fuel Recycle Program. The initial tasks are to provide updated estimates of U_3O_8 costs, a description of and instructions for the use of the "NUCOST" computer code, and an analysis of economic incentives for reprocessing using PNL analysis methods.

SUMMARY

Various uranium price models were reviewed during this quarter. This review and an analyses of impacts of reprocessing delays are included in this quarterly report. The NUCOST document is in draft form.

TRIPS AND VISITORS

July 19, 1976 - Donald deHalas, Consultant, visited PNL.

August 18, 1976 - W. A. Reardon, R. M. Fleischman and D. F. Newman attended a task force meeting at SRL.

TECHNICAL PROGRESS

TASK 1. ESTIMATING U_3O_8 SUPPLY/DEMAND/PRICES (W. A. Reardon)

Several approaches to forecasting uranium prices were reviewed this quarter and some of our own forecasts were refined. The results of these reviews, which were presented at a meeting of the task forces at SRL, are detailed below.

Consultants

Jack Mommsen and Donald deHalas presented forecasts of uranium prices at the SRL meeting. Formerly of NUEXCO, Jack Mommsen is now a private consultant. He presented the format of the NUEXCO price forecasts which essentially involves a conference of knowledgeable personnel who interact and produce a consensus in the form of probability-versus-price forecast. This is shown in Table 2. The numbers in parentheses are the forecasted prices discounted at 6.5%/yr to establish constant dollar values. Note that the constant dollar price is forecasted to decline as time goes on. This will be largely due to capacity buildup and the disappearance of the current sellers market.

TABLE 2. 1975 NUEXCO Price Forecast

Chance That The Actual Price Will Be Less Than "P"	"P" A Possible Price in the Year 1976	1980	1985	1990
90%	74.90 (70.7)	79.00 (57.66)	103.00 (54.87)	126.00 (49.0)
75%	55.60 (52.2)	69.60 (50.8)	93.00 (49.5)	114.00 (44.3)
50%	47.40 (44.5)	59.10 (43.1)	75.50 (40.22)	92.00 (35.8)
25%	42.70 (40.1)	50.30 (36.7)	61.40 (32.7)	77.00 (29.9)
10%	38.50 (36.2)	42.10 (30.72)	52.10 (27.8)	66.00 (25.7)

Donald deHalas, formerly of Babcock and Wilcox, is now a private consultant. His approach to price forecasting is essentially a cost plus profit method which uses the ERDA, Grand Junction data [GJO(100)], to estimate the availability and costs required to mine the uranium. A rate of return is

then assigned and a "price" is established. This price will depend upon the use rate of uranium. That is, as the higher grade ores are mined out, the higher cost/lower-grade ores will be mined. Therefore, the use rate affects the rate of mining. This method is very detailed and may soon be published by the Electric Power Research Institute. The results of the analysis are shown in Figure 1 for two different use rates.

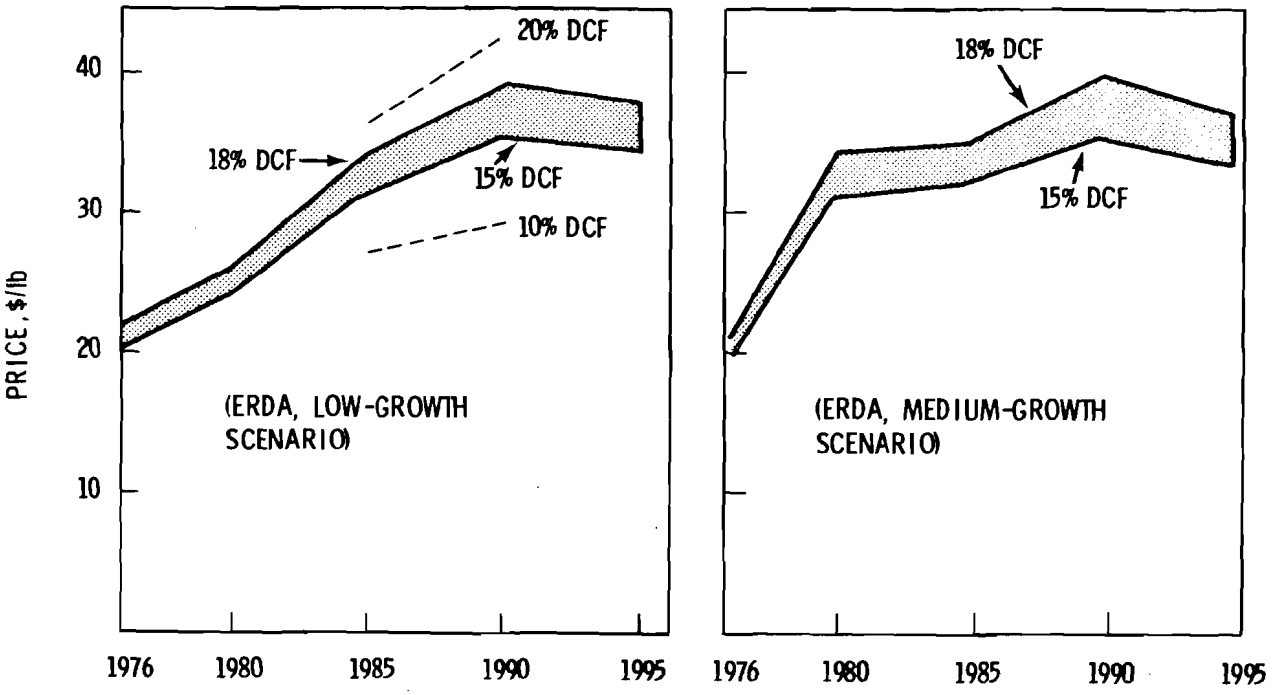

FIGURE 1. Uranium Price Forecast (deHalas)

The rationale for the seeming constancy, or limited price rise is that as the ore grade declines and the mines get deeper, it takes a larger ore body to be economically feasible. The economies of scale for the mine-mill combination are such that they permit the prices to rise only slowly or be essentially limited.

Other Approaches

Karl Peuchl, private consultant, interviewed a number of producers and buyers about uranium prices as a part of the GESMO effort. His entire report is Appendix F, Chapter XI of GESMO.[1] The relevant conclusion with some of the caveats are quoted below.

> "A price of less than \$50/lb U_3O_8 (but not less than \$25/lb) should provide adequate inducement for the necessary exploration and required production. This is not to say that the price will go no higher; it might if uncertainty continues to exist relative to adequacy of supply or if there is economic coupling to fossil fuel prices. The uncertainty relative to supply can be alleviated only by stabilization of the entire nuclear industry so that demand can be forecast with sufficient reliability to attract the necessary capital. Because of the current uncertainty, supply will be tight (and the price will be at a premium) in the near-term. This situation will be either further aggravated or alleviated in the mid-1980's depending upon overall industry developments."

PNL's Approach

One of our current approaches is patterned after that used in Chapter XI (Appendix A) of GESMO.[1] In this approach we assumed that the ERDA defined "reserves" will be mined first in short-term, followed by the "probable resources". The restriction that no more than 1/15 of any category could be mined in a given year provides approximation to the "10-year forward reserve" concept. We also assumed that the price will be set by the highest priced resource being mined (since more than one will, in general, be mined in a given year). The results of this type of analysis as shown in Figure 2 were for a low-growth scenario. The early years are a mixture of current contract prices and the new mines. The steep rise in the early 1980s is indicative of the mining beginning in the second reserve category. The average price is below the indicated reserve price because of the low current contract prices. Also note that the price remains above that of

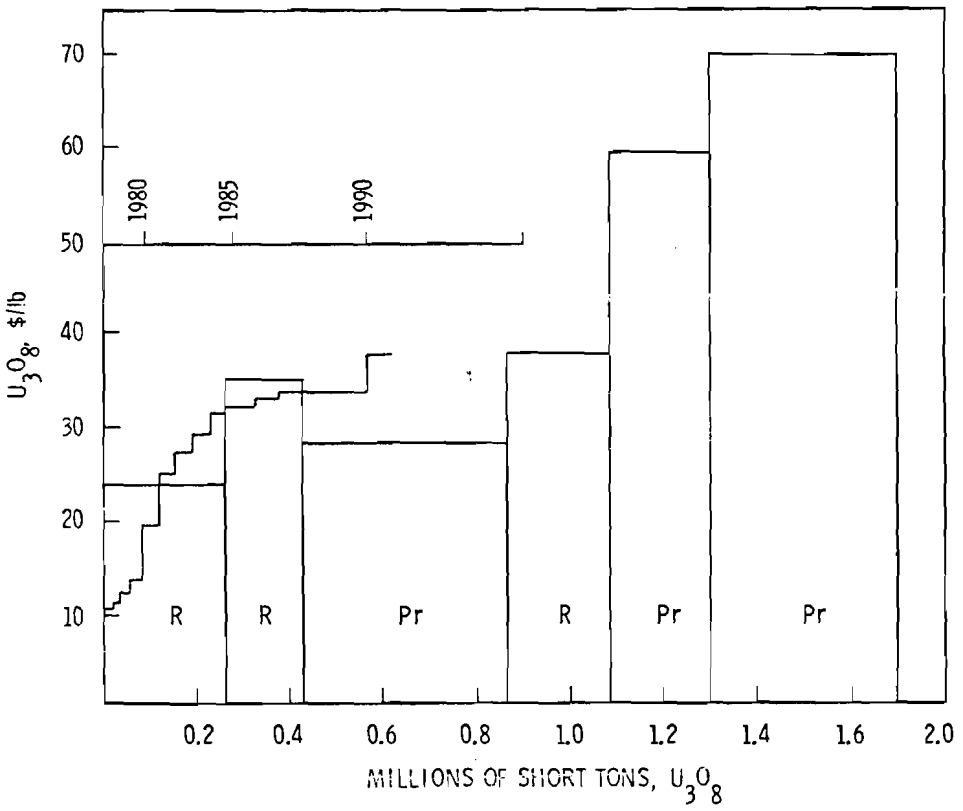

FIGURE 2. Uranium Price Forecast (1977 dollars)

the first "probable" category. Current concensus seems to be that it is just as expensive to mine known lower-grade deposits as it is to pay the extra exploration and development costs associated with the probable category. At 1990 mining will have begun in the third reserve category and the price is shown to increase correspondingly. The calculations contain corrections for imports and inventories.

A second PNL approach is to use the RAND Corp. energy model to provide for price sensitivity in the demand for power, and thus the price of uranium. This work will be reported next quarter.

TASK 2. DESCRIPTION AND USE OF NUCOST (L. E. Addison)

A report describing NUCOST is currently in draft form and will be published soon.

TASK 3. ECONOMIC IMPLICATIONS OF DELAYS IN REPROCESSING LWR SPENT FUEL

(D. F. Newman, R. M. Fleischman and E. T. Merrill)

The process flow rates and projected costs for materials and services were studied for various postulated spent fuel reprocessing schedules. The objective of this study was to evaluate the national benefits to be derived from the recycle of uranium and plutonium in spent LWR fuel, in relation to the spent fuel disposal option, for various assumed delays in the availability of reprocessing capacity. In addition, the results of this study, using the NUFUEL logistics computer model, can be compared directly with similar results from other ERDA studies using the same reference growth schedule for LWRs and the same projected unit cost factors.

A comparison of the various schedules for reprocessing capacity and the actual reprocessing load used in this study is listed in Table 3. The origin of these schedules warrants some discussion. The PNL Constrained Schedule represents our estimate of reprocessing growth assuming each plant will have a minimum backlog of 1-year's throughput at the beginning of operation. The "Reference Schedule" is the reprocessing schedule developed in other ERDA studies using apparently different logistics assumptions than are used in NUFUEL since the capacity exceeds demand in 1993 to 2000. The delay schedules reflect 2-, 5-, and 10-year delays of the reference schedules. The AGNS (Allied General Nuclear Services) startup in 1981 and 8-year Delay of Reference Schedule was designed to investigate the impact of delaying the introduction of new reprocessing but assuming the AGNS plant operates as currently planned.

The benefits accrued through the year 2000 from recycling the uranium and plutonium contained in spent LWR fuel are listed in Table 4 for the various assumed reprocessing schedules. The total benefit in 1977 dollars is $13.0 billion for the reference reprocessing schedule starting in 1981 and decreasing to $8.5 billion when the start of the reprocessing schedule is delayed 10 years. The total benefits from Table 4 are plotted in Figure 3 as a function of the number of years delay in the startup of the reference reprocessing schedule. The benefits attributable to the 1981 startup and

TABLE 3. Comparison of Schedules for Reprocessing Capacity and NUFUEL Reprocessing Load

Year	PNL Constrained Schedule (MTHM/yr)		Reference Schedule (MTHM/yr)		2-yr Delay (MTHM/yr)		5-yr Delay (MTHM/yr)		10-yr Delay (MTHM/yr)		AGNS Startup in 1981 and 8-yr Delay of Reference Schedule (MTHM/yr)	
	Capacity	Reprocess	Capacity	Reprocess	Capacity	Reprocess	Capacity	Reprocess	Capacity	Reprocess	Capacity	Reprocess
1975	0	0	0	0	0	0	0	0	0	0	0	0
1976	0	0	0	0	0	0	0	0	0	0	0	0
1977	0	0	0	0	0	0	0	0	0	0	0	0
1978	0	0	0	0	0	0	0	0	0	0	0	0
1979	0	0	0	0	0	0	0	0	0	0	0	0
1980	0	0	0	0	0	0	0	0	0	0	0	0
1981	600	599	600	599	0	0	0	0	0	0	600	599
1982	1000	999	1000	999	0	0	0	0	0	0	1000	999
1983	1500	1497	1740	1738	600	599	0	0	0	0	1500	1499
1984	2000	1996	1900	1896	1000	999	0	0	0	0	1500	1497
1985	2250	2245	2100	2095	1740	1739	0	0	0	0	1500	1497
1986	2850	2845	2700	2694	1900	1896	600	599	0	0	1500	1497
1987	3850	3847	3700	3697	2100	2095	1000	999	0	0	1500	1497
1988	5350	5346	5200	5196	2700	2693	1740	1739	0	0	1500	1497
1989	6250	6247	6700	6696	3700	3694	1900	1898	0	0	1500	1497
1990	6750	6748	8200	8196	5200	5197	2100	2095	0	0	1500	1498
1991	6750	6748	9100	9097	6700	6695	2700	2693	600	599	1740	1738
1992	6750	6748	10200	10197	8200	8196	3700	3690	1000	999	1900	1898
1993	6750	6748	10600	7281	9100	9097	5200	5194	1740	1739	2100	2098
1994	7350	7347	11100	7800	10200	10197	6700	6696	1900	1899	2700	2698
1995	7740	7748	11100	8338	10600	10598	8200	8197	2100	2096	3700	3697
1996	8350	8248	11100	8978	11100	11099	9100	9095	2700	2693	5200	5196
1997	8850	8848	11100	9523	11100	11099	10200	10197	3700	3690	6700	6697
1998	9250	9248	11100	9906	11100	11099	10600	10598	5200	5185	8200	8196
1999	9750	9748	11100	10355	11100	11099	11100	11098	6700	6696	9100	9097
2000	10350	10348	12100	10803	11100	11099	11100	11099	8200	8197	10200	10197
TOTAL		114448		126214		119189		85887		33794		65089

TABLE 4. Benefit[a] from Recycle of Uranium and Plutonium Relative to Spent Fuel Disposal

Reprocessing Schedule Description	Total Benefit to the Year 2000 (Billions of $)	Average Fuel Cycle Costs (mills/kW-hr)	Total Benefit Discounted[b] (Billions of $)	Discounted[b] Fuel Cycle Costs (mills/kW-hr)
Spent Fuel Disposal		6.22		6.27
Reference Reprocessing Schedule (starting in 1981)	13.040	5.85	1.342	6.10
PNL Constrained Schedule (starting in 1981)	12.733	5.86	1.309	6.11
Reference Schedule Delayed 2 years (starting in 1983)	12.423	5.87	1.313	6.11
Reference Schedule Delayed 5 years (starting in 1986)	10.093	5.94	1.019	6.14
Reference Schedule Delayed 10 years (Starting in 1991)	8.486	5.98	0.796	6.17
Reference Schedule Delayed 8 years (with AGNS Plant Starting in 1981)	9.193	5.96	0.837	6.17

(a) 1977 dollars
(b) Discount rate of 10%/yr

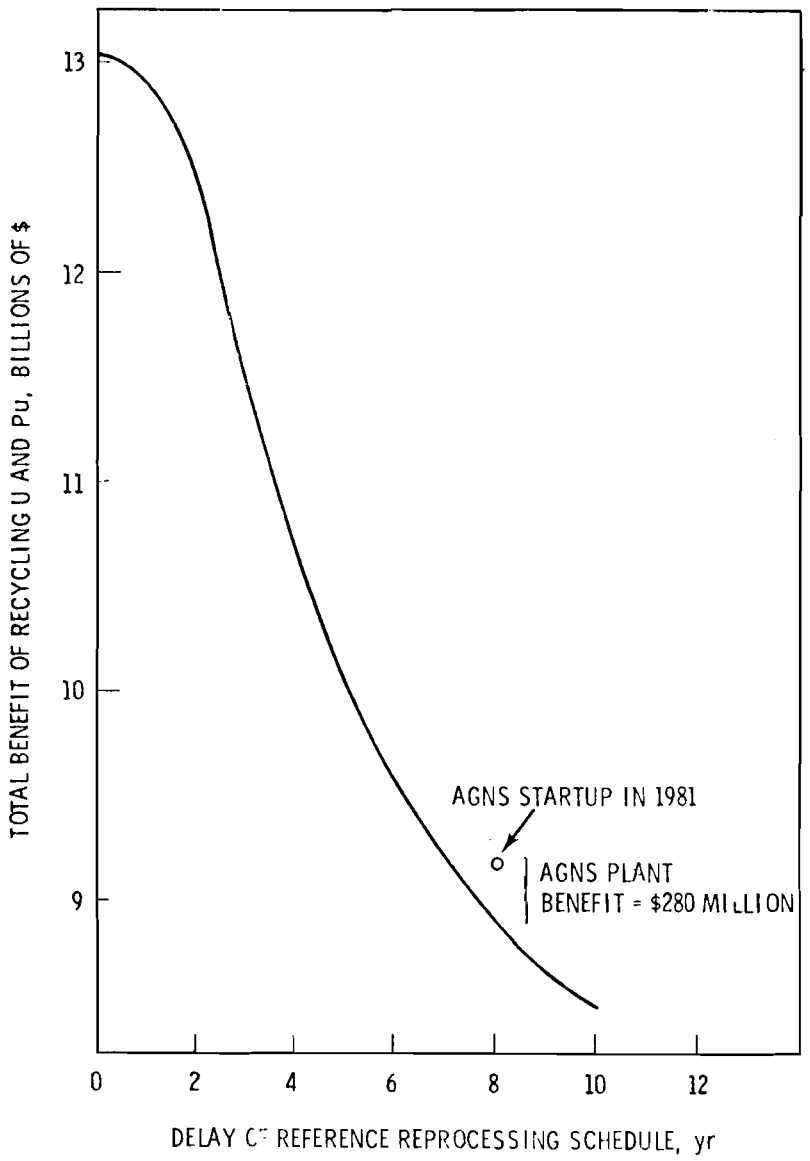

FIGURE 3. Benefit of Recycling Uranium and Plutonium as a Function of the Number of Years Delay in the Startup of the Reference Reprocessing Schedule

operation of the AGNS plant for 8 years amounts to $280 million, as can be seen in Figure 3. However, as can be seen in Figure 4, the discounted benefit of the AGNS plant operation is very small when a 10%/year discount is assumed. Finally, both the average and discounted fuel cycle costs in

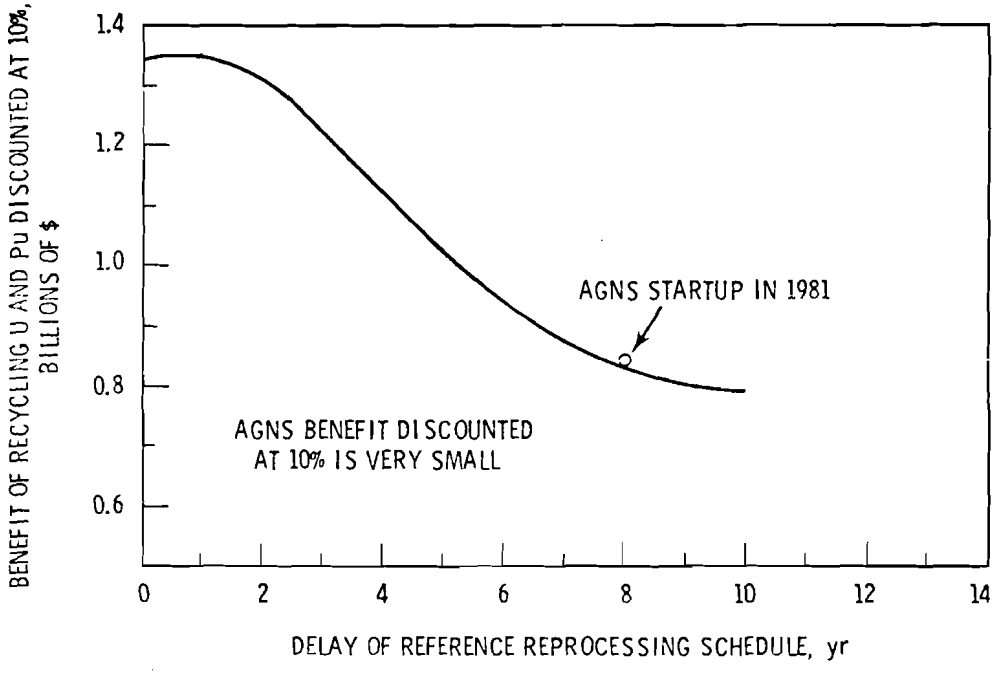

FIGURE 4. Discounted Benefits of Recycling Uranium and Plutonium as a Function of Number of Years Delay in the Startup of the Reference Reprocessing Schedule

1977 dollars are also listed in Table 4. These values provide the perspective of the large change in total benefits that accrue from rather small (~0.1 to 0.4 mill/kW-hr) differences in the fuel cycle costs.

TASKS IN PROGRESS AND PLANS

The NUCOST program will be further improved to show the difference between BWRs and PWRs. Documentation will be completed.

A model of the U_3O_8 industry has been started to assess the supply/price question of the future.

CATEGORY 2 - ECONOMIC AND ENVIRONMENTAL DOCUMENTATION

(F-RL-14-007)

PROJECT TITLE: Technical Assistance to Savannah River Laboratory

PROJECT MANAGER: I. C. Nelson, Environmental Technology Evaluation Portion of the Generic Environmental Impact Statement for Commercial Waste Management

PRINCIPAL INVESTIGATOR: I. C. Nelson

OBJECTIVE

Review and comment on Environmental Impact Statement (EIS) drafts prepared by SRL.

SUMMARY

Draft EIS material was reviewed and comments have been made.

TRIPS AND VISITORS

None

TECHNICAL PROGRESS

Draft EIS material was reviewed and comments have been made.

TASKS IN PROGRESS AND PLANS

None

CATEGORY 3 - SPENT FUEL RECEIPT AND STORAGE
(F-RL-15-001)

PROJECT TITLE: Survey Current Technology of Fuel Handling Techniques

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

PRINCIPAL INVESTIGATORS: J. F. Nesbitt, Engineering Technology Department, G. S. Allison, Materials Development Section, Materials Department

OBJECTIVES

Identify and assess potential improvements for spent fuel handling in predissolver equipment. This will be done by evaluating the spent fuel handling process, providing input to the conceptual design development, and by designing and demonstrating the prototype equipment. Alternate processes will be developed where significant needs are identified.

SUMMARY

Evaluation of existing and planned process equipment and concepts was continued through discussion with industry personnel and plant visits and by reviewing process documents. A draft report has been completed, covering process evaluation, spent fuel handling, and predissolver equipment operation.

TRIPS AND VISITORS

August 2, 1976 - J. F. Nesbitt visited Dr. E. Voiland at General Electric Company in Morris, IL

August 4, 1976 - J. F. Nesbitt visited R. W. Wiesner at Programmed and Remote Systems Corporation in St. Paul, MN

August 31, 1976 - G. S. Allison visited Carl King at General Electric Company in San Jose, CA

August 31, 1976 - G. S. Allison visited Max Schleinger at RETECH in San Rafael, CA

TECHNICAL PROGRESS

The operation of and the experience with the head-end mechanical handling tasks and equipment in General Electric's Midwest Fuel Recovery Plant and the Nuclear Fuel Services Plant (NFS) were reviewed with various plant personnel. These informal discussions indicated that operations would be made easier (assuming expenditure of more capital and operating dollars) with more space, facilities, and staff to service and maintain process equipment, and to provide increased general support activities.

The NFS plant has a single cask receiving area for both trucks and railroad cars which limits the rate at which material can be received and handled. This built-in time limitation and the time required to receive, monitor, unload, decontaminate and turn a cask around is a factor in plant operating cost.

NFS has changed their fuel receipt and storage area. Some of the major changes were:

- adding to the ventilation and humidity control system,
- adding a major cask decontamination facility, and
- making provisionts to reduce external cask contamination while unloading fuel elements.

The operation of the cranes, tools, canisters, grapnels, etc., in the fuel receipt and storage operation at NFS has been very satisfactory. Radiation and contamination are not major problems and, after 10 years of operation, all aspects of their basin water and storage pool appear to be very satisfactory.

As with the NFS design, the General Electric compact plant concept used at Morris, Illinois could benefit if additional space were available for the ongoing operations. Changes have been made to the fuel receipt and storage area in the General Electric Plant. Some of the major changes were:

- enclosing the truck and railroad car receiving area,
- increasing the storage capacity by changing the canister and canister rack design,
- using torque wrenches in place of impact wrenches for cask operations, and
- building a new cask and carrier service facility adjacent to the original plant.

Other than the relatively long time needed to unload casks, the operation of the fuel receipt and storage facility has proven to be workable. One unloading area for both rail and car shipments is a bottleneck as is the servicing and decontamination of the various casks.

There have been no problems with leaking fuel elements or off gases. Similarly, control of the temperature, purity and activity levels of the water have all been possible within desired limits. Personnel exposure has been low and contamination control has not been a problem. The operation of the various cranes, cradles, grapnels and tools has been satisfactory.

Dr. Voiland arranged for us to witness the unloading and handling of a cask at the General Electric Facility as well as portions of the cask loading at a reactor site. This operation took a total elapsed time of 43 hours to perform. Approximately 9 hours were required to monitor and check out the cask, and prepare it for the underwater operations. A total of 12 hours were required to unload the 18 BWR fuel elements and store them, while 18 hours elapsed time was taken to install the lid, decontaminate the external cask surfaces, and to flush and monitor the interior. Another 4 hours were required to load the cask on the railroad car, and to prepare the car and cask for shipment.

This total time is typical of that required at Morris for the IF-300 casks. This particular operation included several delays such as temporary rigging of lights and scaffolding and the malfunction of an underwater gate. Most of the operations were proficient in performing the various remote operations, but the provision of better arrangements and services could be applied to reduce the required operating time.

The small NAC-1 cask was turned around in 12 hours elapsed time. This was relatively fast since the average time to handle this cask has been 18.1 hours on the more recent deliveries.

Programmed and Remote Systems Corporation (PAR), who designs and manufactures the special- and product-line remotely operated equipment and systems for the nuclear industry was also visited. Discussions with PAR indicated they agreed that improvements could be made in the operation of remote processing equipment by 1) increasing initial plant design work, 2) designing the building to fit the process equipment rather than vice-versa, 3) using the process equipment only in the manner it was originally intended, and 4) providing increased support services. They stressed the need for increased conceptual design work on any equipment operated or maintained remotely, for redundancy and service provisions; and for use of mockup and cold checkouts before going into hot or remote operations.

At both reprocessing plants the problems associated with the predissolver equipment in the mechanical cell could have been minimized by more prototype operation and development work. In both cases the operations would have been more trouble-free had fully developed provisions been made to remove the end fittings and spacers so that the remainder of the processing (chopping and dissolving) handled only the rods. This, plus the obvious incentives for removing the hardware (end fittings and spacers) from the dissolver and TRU-contaminated hull streams, prompted the conception of a method to disassemble the spent fuel bundles prior to chopping. This concept would employ a means of cutting the end fittings from the bundle and stripping the spacers from the rods or vice-versa. It is desirable to disassemble the bundle under water to more easily control the ^{60}Co and other radioactive constituents of the crud buildup on the fuel rods.

One of the most promising methods of cutting the end fittings from the fuel bundle is a recently developed arc-saw. It was demonstrated that this saw is capable of cutting end fittings from a bundle of rods with the following characteristics:

- fast (about 1 minute through a BWR bundle),
- narrow kerf with no sticking or welding of adjacent parts,
- essentially no burr,
- performs well under water, and
- no pyrophoric residue.

It is being proposed that this saw plus some hydraulic spacer stripping equipment be designed into a prototype facility for developing the required disassembly procedures.

Progress toward cataloging the fuel bundle design parameters which affect fuel disassembly has been limited by reluctance of fuel manufacturers to furnish what is considered proprietary information. Efforts are currently underway to design a questionnaire which, if filled out, would furnish the required details without divulging the proprietary information. There are indications that this approach to industry may prove successful.

TASKS IN PROGRESS AND PLANS

A report covering the first or evaluation phase of the program has been drafted and is being reviewed internally. This report covers details on site visits and discussions with various industrial organizations.

Presently, various cleaning means and methods are being searched and evaluated for their use on the decontamination of large casks. If this decontamination time can be reduced, it will result in a cost reduction in both cask rental charges and in operating costs. Also, development in blast cleaning methods have the potential for reducing the volume of contaminated wastes.

CATEGORY 4 - HEAD-END PROCESSES
(F-RL-14-002)

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

OBJECTIVES

Review and evaluate alternate approaches to voloxidation for the removal of tritium and other volatile fission products from irradiated fuel prior to aqueous feed preparation.

SUMMARY

Three pyrochemical methods for head-end release of tritium are discussed and some of the problems associated with each method are identified. Several modifications of the general concept of water recycle in a reprocessing plant are also described. A combination of recycle and storage with the possible addition of isotope separation appears to be a promising method.

TRIPS AND VISITORS

July 11-15, 1976 - L. L. Burger attended the International Conference on Management of Waste from the LWR Fuel Cycle in Denver, CO.

August 5-6, 1976 - L. L. Burger attended the ANS Topical Meeting on Controlling Airborne Effluents from Fuel Cycle Plants, Sun Valley, ID.

September 8, 1976 - W. J. Brumley, SRO and J. A. Kelley, SRL visited PNL to review program progress and plans.

TECHNICAL PROGRESS (R. D. Scheele and L. L. Burger)

The pathways of tritium and alternatives for tritium control during reprocessing were discussed in a recent review.[2] As discussed in this review, pyrochemical methods offer one possible route for tritium removal prior to solvent extraction.

To utilize pyrochemical reduction/oxidation processes, the oxide fuel must be adequately exposed to the reducing or oxidizing conditions necessary to disintegrate the fuel and release the volatile fission products. The methods studied include: 1) chopping the fuel into small pieces,[3] 2) decladding the fuel by a melt-pour technique,[4] and 3) alloying the cladding with another metal.[5,6] The first method, chopping, is a tested procedure, while the last two have been studied primarily for stainless steel-clad mixed-oxide fuels. The melt decladding technique which has been studied for stainless steel (melting point of 1450°C) probably has little application to zirconium-clad fuel due to the higher melting point of zirconium (1800°C). Zirconium can be alloyed with various metals; therefore, the alloying technique could be feasible.

Following the chopping/decladding step, the oxide fuel can be oxidized or reduced. In principle, this should allow easy escape of the volatile fission products from the oxide fuel. We considered three "head-end" processes viable alternatives to voloxidation. Two processes use molten metals to reduce the oxide fuel to the metal, and the third uses molten salts to oxidize the fuel to the uranate (plutonate). A brief description of each was given in the previous quarterly report.[7]

The first alternative was presented by Wade and Wolf.[8] This method uses a two-phase system (Ca-$CaCl_2$) to obtain plutonium metal from PuO_2. The principal reaction is $2Ca + PuO_2 \rightarrow 2CaO + Pu$. The $CaCl_2$ dissolves the CaO which forms on the surface of the plutonia and prevents further reaction with the calcium. Thus, it is necessary to agitate the mixture to insure contact of all phases. This system which has massive metal yield efficiencies in excess of 99.96% is also applicable to UO_2 using the exothermic chemical reaction $2Ca + UO_2 \rightarrow 2CaO + U$.

The behavior of americium was also studied and was found by Wade and Wolf to behave much the same as plutonium in this system. This behavior can be extrapolated to the other actinides. Thus, the Ca-$CaCl_2$ waste stream should be relatively free of actinides.

Uranium and plutonium are insoluble in Ca and $CaCl_2$. This insolubility combined with differences in density, could be used to separate the product metals from the reaction mixture. Criticality control is a necessary consideration if filtration is used to separate the product metals.

Besides difficulties in separating the product, adequate contact of the oxide fuel with both reactant phases must be insured. Wade and Wolf[8] used agitation to provide the necessary interaction; their final apparatus was a prototype to be used at semiproduction level.

A second alternative is the "head-end" process developed at Argonne National Laboratories (ANL) which involves reduction and dissolution of the oxide fuel in a Zn-Ca-Mg molten alloy. In this process calcium is used as the reductant, and composition adjustment is performed to insure complete dissolution of the product uranium. The dissolved product metals are separated from the alloy by evaporation of the Zn-Mg or by precipitation of the insoluble nitrides. However, difficulties were encountered for both methods. The latter proved to be inefficient and the former had a limited maximum rate; exceeding this maximum evaporation rate caused entrainment of the product metals.

Most of the steps of this process except for the separation step have been demonstrated. Volatility of some of the heavier fission products may also occur. However, ruthenium should not volatilize under the reducing conditions used. The volatilization of iodine, xenon, and krypton have been demonstrated.[9] The large volume of waste metals and salts contaminated with actinides is a potential problem. The lack of industrial experience in working with corrosive materials at high temperatures may also hinder the development.

The third alternative uses oxidizing salts to effect the release of volatile fission products. Nitrate salts have been studied the most and would appear to be the most logical for use in a "head-end" step for the Purex process. The compound formed is the metal uranate (plutonate) which is insoluble in the molten salt. The metal uranate, following separation, would become the feed to the Purex process. This would introduce large quantities of inert metal which would become part of the high-level waste (HLW). Large amounts of alkali metals would pose a problem for glass fixation of HLW.

An alternative to a head-end tritium control step would be aqueous stream recycle. In the conventional purex plant some recycle is involved. Condensate water is used for makeup of nitric acid, and nitric acid from concentrators is reused. Various waste streams are recycled to reduce uranium and plutonium losses, etc. Thus, one concept for tritium control is to increase the water recycle, letting the tritium concentration build up so that a much smaller volume of tritiated water is to be disposed.

Many different approaches to aqueous recycle can be used. Similarly, there are many potential limitations, but the controlling ones cannot yet be identified. About 10^5 liters (ℓ) of water are used per day for a typical 5 tonne/day reprocessing plant. With no special flowsheet modification this would be the amount discharged, and the tritium concentration would be about 3×10^{-3} Ci/ℓ. The most simple concept is probably total recycle of the water. For example, if the recycle water stream could be brought up to 2 Ci/ℓ, then the discharge rate would be reduced from 3×10^7 to 4.5×10^4 ℓ/yr; the latter is not an inconceivable quantity for storage.

A modification of this concept is to consider the stored water as plant inventory water. As noted by Cooley and Richardson,[10] the volume maintained would be large enough that the decay rate (about 5.6%/yr) would equal the input rate (about 10^6 Ci/yr). This would require a plant inventory of 9×10^6 ℓ at the 2 Ci/ℓ concentration. Assuming that about 10% of the tritium could be released (calculated for a site boundary concentration of 0.2 mpc), they calculated that for a typical plant the vent air would have

to be refrigerated to about -18°C if the tritium concentration in the process water was 1.7 Ci/ℓ. Referring to the limitations listed above, water leakage is clearly an important factor.

The maximum tritium concentration level which is permitted in the plant will depend on: 1) severity of plant design changes required to prevent tritium radiation exposure to plant personnel and to control the humidity in the vent gas systems, 2) difficulty from other recycled materials such as fission products and detrimental chemical species, 3) compatibility with the plant needs for recovered nitric acid, and 4) the ultimate fate of the tritiated waste.

An attractive aspect of water recycle is that isotopic separation may be usable. Isotopic separation of tritium to produce a waste water low enough in tritium to be discharged is very costly because of the large amount of separative work that must be done to reduce the large volume of water to mpc (3×10^{-6} Ci/ℓ). However, if the separation is applied to a recycle stream, the problem is much less formidable. A "waste" stream of still moderately high tritium concentration can be recycled to the plant and the more concentrated stream can be removed for storage. The isotopic separation package does not have to be part of the original recycle scheme, but can be added at a later date. Thus, water storage, water recycle, and hydrogen isotope separation appear to fit together in the general recycle picture.

Of the tritium that stays in the dissolver, 99% or more is retained by the first cycle aqueous waste stream. Isolation of the first cycle would produce an aqueous system containing essentially all of the process tritium in about 20 to 25% of the total plant water. Although the idea has merit, a brief consideration of the complex flow pattern of water and nitric acid indicates that the application is not easy.

The general concept of water recycle has been briefly discussed by several authors[11-13] while one study[14] has considered the flowsheet in more detail.

TASKS IN PROGRESS AND PLANS

Continued evaluation of the pyrochemical methods is planned including laboratory experiments. Water recycle studies will focus on specific approaches in an attempt to identify the process or processes that have the most merit when adapted to the Purex flowsheet.

CATEGORY 5 - OFF-GAS TREATMENT

(F-RL-14-003)

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

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

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

OBJECTIVE

Provide a suitable air cleaning process for removing low concentrations of tributyl phosphate vapors from nuclear fuel reprocessing off-gas streams.

SUMMARY

Two methods of collecting and determining TBP vapor have been investigated:

- An impinger system with ethylene glycol to collect TBP vapor was studied. In this system the TBP is extracted from ethylene glycol into hexane and concentrated. The concentrate is then analyzed using a gas chromatograph. Analysis of standards proved this method to be promising.
- The use of charcoal filters to capture low concentrations of TBP vapor and the subsequent analysis of the filters for phosphorus using X-ray fluorescence was investigated. A lower detection limit of about 0.1 mg phosphorus per filter sample was established. This method also shows promise, but further development work is needed.

Purchase of a real-time phosphorus detector/analyzer was initiated.

TRIPS AND VISITORS

September 8, 1976 - J. A. Kelly and W. J. Brumley of SRL visited PNL.

TECHNICAL PROGRESS

A series of standards of known mass of TBP (liquid TBP, 99% pure) ranging from 4.9×10^{-5} to 9.8×10^{-2} g TBP was prepared in ethylene glycol solution and extracted with hexane. The extract was concentrated and analyzed using a gas chromatograph with flame ionization phosphorus detector. The extraction efficiency of hexane was approximately 80% (based on eight extractions) in the range 2×10^{-5} to 2×10^{-3} g of TBP. Initial analysis showed good reproducibility (<10%) for TBP detection between 0.005 to 10 mg.

Two series of standards of known mass of 99% pure TBP were made by delivering known aliquots to charcoal filters. These filters were analyzed by X-ray fluorescence. Phosphorus in these standards ranged from 2.3×10^{-5} to 1.1×10^{-2} g.

The first set established the lower detection limit of about 0.1 mg phosphorus per filter sample. The first standards showed erratic results, the front and back readings not uniform, and contamination was suspected.

A second series consisted of two samples each 0.565, 2.26, and 5.65 mg phosphorus per filter blank, and reagent blank. The reagent blank was acetone, since the TBP was dissolved in acetone for preparing the standards. One set was analyzed by direct X-ray fluorescence; the other was macerated, then pressed and analyzed.

The second set showed more promising results. The front and back counts showed reasonable agreement. The 0.565 mg phosphorus/filter sample counted about five times more than the blank. There was no difference in counts between the blank untreated and acetone treated filters. The macerated and homogenized filters showed higher counts than the directly measured filter, indicating the maceration is necessary to distribute the phosphorus uniformly in the sample as analyzed.

Arrangements have been made to purchase a real-time continuous gas analyzer with the capability to detect phosphorus near 0.001 ppm. The analysis is based on a patented flame photometric detector.

TASKS IN PROGRESS AND PLANS

Inorganic sorbent materials from several manufacturers have been received and will be tested in a column for TBP removal efficiency. Samples of TBP in air upstream and downstream of the material will be passed through ethylene glycol traps using an impinger system to collect the TBP. The retention of TBP will be calculated from the difference. Further work will be done to determine the accuracy of the impinger system analysis.

CATEGORY 6 - PUREX PROCESS (IODINE CHEMISTRY) PART 1
(F-RL-14-004)

PROJECT TITLE: Study of Iodine Chemistry in Process Solutions

PROJECT MANAGER: J. H. Jarrett, Chemical Development Section, Chemical Technology Department

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

OBJECTIVE

Study the chemical species and behavior of the iodine which is not volatilized from the dissolver solution, and develop means of managing iodine in the various plant streams to minimize its release to the environment.

SUMMARY

Laboratory tests have been conducted with commercially available macroreticular polymeric adsorbents. The adsorbents continue to provide good iodine removal from solutions simulating fuel reprocessing plant concentrator condensates after hydrogen peroxide has been added to stabilize molecular iodine. Test results with a macroreticular anion exchange resin were inferior to those obtained with the polymeric adsorbents. Experimental testing was begun on some of the analytical methods identified in the literature search as being promising methods to identify and quantify iodine species present in fuel reprocessing plant streams.

TRIPS AND VISITORS

September 8, 1976 - W. J. Brumley, SRO and J. A. Kelly, SRL, visited PNL to review program progress and plans.

TECHNICAL PROGRESS

SORPTION OF IODINE FROM SIMULATED PROCESS STREAMS (J. A. Partridge)

The iodine which is not volatilized from the dissolver solution prior to solvent extraction recovery of the uranium and plutonium will likely be distributed among a variety of plant streams. The majority of such iodine could ultimately end up in the low-level aqueous effluent from the plant. The release of iodine in this effluent could be the release which limits the overall plant retention factor. Our initial studies are emphasizing the removal of iodine from such streams (e.g., condensed concentrator overheads) by commercially available adsorbents. Column runs are being performed with four polymeric adsorbents and one anion exchange resin.

Tests are continuing on the performance of the polymeric adsorbents when hydrogen peroxide is added to the feed containing a mixture of iodide ion and molecular iodine. The peroxide is added to convert iodide ion to molecular iodine which is sorbed by the polymeric material; see Table 5 for results of these tests.

Encouraging results are being obtained with each of the four adsorbents. The adsorbents XAD-2 and XAD-4 continue to give a better iodine D.F. than do XAD-7 and XAD-8. In addition, based on the length of the colored band in each of the columns, it appears that the iodine loading capacity on XAD-2 and XAD-4 is considerably greater than on XAD-7 and XAD-8. For example the colored band on the XAD-4 columns listed in Table 5 are approximately 1 cm, whereas the colored band on the XAD-8 column has advanced until it is approximately 10 to 11 cm long, and the band on XAD-7 is currently about 5 to 6 cm long. Data for a quantitative determination of the iodine loading capacity of the adsorbents have not been obtained.

TABLE 5. Iodine Removal from Aqueous Solution

Sorbent Material[a]	Column Vol. (ml)	Column Length (cm)	Feed Composition (Make-up molarities) I^-	I_2	HNO_3	H_2O_2	Flow Rate (col. vols/hr)	Total Flow (col. vols)	Nominal Instantaneous DF[b,c]
A-26 Resin	14.1	22.6	1.6×10^{-6}	2×10^{-7}	0.01	--	2	29	6
--- ---									
XAD-2 Adsorbent	13.7	22.0	4×10^{-6}	8×10^{-6}	0.05	0.3	1.5 - 2.5	988	80
XAD-4 Adsorbent	14.8	23.8	4×10^{-6}	8×10^{-6}	0.05	0.3	1.5 - 2	1285	105
XAD-4 Adsorbent	10.0	11.6	4×10^{-6}	8×10^{-6}	0.05	0.3	4 - 7	2280	80
XAD-7 Adsorbent	13.4	21.6	4×10^{-6}	8×10^{-6}	0.05	0.3	1 - 2	824	35
XAD-8 Adsorbent	14.9	24.0	4×10^{-6}	8×10^{-6}	0.05	0.3	1 - 2	641	60
A-26 Resin	9.5	18.4	4×10^{-6}	8×10^{-6}	0.05	0.3	1 - 2.5	389	22
								519	24
								570	<10 (Iodine Breakthrough)

(a) All are products of Rohm and Haas.

(b) DF = (Iodine content in Feed) ÷ (Iodine content in Effluent)

(c) Instantaneous DF = Nominal DF ± 0.5 (Nominal DF)

The iodine D.F.'s measured with the polymeric adsorbents have generally remained quite constant at the values indicated in Table 5. During one period of time, however, an abnormal behavior was observed in the performance of five of the columns. The iodine concentration in the effluent from each increased slowly to about 6 to 8% of the feed concentration (D.F.'s of approximately 15) after 120 to 270 column volumes of solution had passed through them. Since this behavior occurred simultaneously in each of these columns and the iodine concentration in the effluent subsequently decreased, we suspect that it was caused by abnormalities in the feed make-up during that period. Additional runs will be made to test this.

As indicated by the relative constancy of the D.F. values during a run, the behavior observed here with the polymeric adsorbents is not typical of normal ion exchange operations. Here, we have a relatively constant "leak-through" of iodine, but there is no indication of breakthrough capacity

having been reached (or even closely approached) on these columns based on the iodine concentration in the effluent, the lengths of the colored iodine bands in the adsorbents' beds, and gamma readings down the columns as measured by a Geiger-Mueller counter. However, the run with A-26 anion exchange resin, which appears as the last set of entries in Table 5, has reached iodine breakthrough. This indicates a much lower iodine breakthrough capacity on this resin than on the polymeric adsorbents with the feed composition being tested.

Iodine sorption tests in which nitrite ion was added to iodide-molecular iodine feed solutions in place of hydrogen peroxide showed that nitrite was not effective in enhancing iodine adsorption under the conditions tested (feed make-up molarities: 1.6×10^{-6} M I^-, 2×10^{-7} M I_2, 0.01 M HNO_3, 0.001 M $NaNO_2$).

Analytical Methods (J. L. Cox)

The objective of this subtask is to review, evaluate, and devise suitable techniques to identify and quantify iodine compounds present in liquid fuel-reprocessing streams. The ability to routinely, quantitatively determine the types of iodine compounds present before and after process stream treatment to remove iodine is of fundamental importance to the development and monitoring of effective methods for its removal.

During this quarter laboratory work has been initiated on some of the analytical methods identified in the literature survey conducted during the previous quarter. For the most part these studies have been directed at measuring iodide ion concentration with an iodide-specific ion electrode.

The response of the iodide-specific ion electrode (Orion 94-53-00), coupled with an Orion 90-01-00 reference electrode in a neutral aqueous system, is Nernstian over the iodide concentration range of 1×10^{-3} to about 3×10^{-8} molar. Below 3×10^{-8} molar iodide concentration the electrode response deviates from Nernstian behavior rendering it less effective as an anlytical tool at these low concentrations.

CATEGORY 6 (contd)

Solutions containing molecular iodine as well as iodide ion were prepared and tested with the iodide-specific ion electrode. The readings consistently indicated the iodide ion concentration to be greater than the amount added. Since the experiments were performed in neutral solution, it is likely that the hydrolysis reaction

$$I_2 + OH^- = HOI + I^- \qquad (1)$$

contributed to the observed result.

A solution prepared to contain 1.4 M HNO_3 and 3×10^{-7} M iodide ion was tested with iodide-specific ion electrode and was found to contain less than 3×10^{-8} M iodide. This result indicates that oxidation to iodine and/or iodate occurred under these conditions.

The iodide-specific ion electrode appears to be a useful tool to be used in our efforts to identify and quantify the iodine species present in fuel reprocessing streams.

TASKS IN PROGRESS AND PLANS

Column runs are in progress comparing the polymeric adsorbents for iodine capacity and decontamination factors. Plans call for expanded efforts to study the effects of variations in the concentrations of total iodine, hydrogen peroxide, and nitric acid. Attempts will be made to identify the iodine species which is "leaking-through" the adsorbent columns. Scouting experiments will also be initiated with other potential iodine sorbents.

In the analytical area, developmental efforts using the iodide-specific ion electrode will be continued. Experimental evaluation of the applicability of other selected approaches will be initiated; included here are techniques such as gas chromatography (using an electron capture detector), spectrophotometric analysis (using complexation to form more highly colored species), and the iodine catalyzed reaction between Ce(IV) and As(III).

Work will be initiated on minimizing the volatilization of iodine during vaporization of waste water streams.

PUREX PROCESS (SOLVENT EXTRACTION CONTACTORS) PART 2

(F-RL-14-004)

PROJECT TITLE: Evaluation of Solvent Extraction Equipment

PROJECT MANAGER: J. H. Jarrett, Chemical Development Section, Chemical Technology Department

PRINCIPAL INVESTIGATOR: D. H. Lester, Chemical Development Section, Chemical Technology Department

OBJECTIVE

Provide an integrated study of solvent extraction equipment types. Such information will provide information needed in optimizing reprocessing plant design. Where voids in existing understanding are identified, additional R&D will be proposed.

SUMMARY

The literature search was completed. A draft literature review document was completed and is being edited and typed. A workshop was held on August 5 and 6 with 18 participants from laboratories involved in solvent extraction equipment development. The proceedings of the workshop have been prepared in draft form, and the draft is now undergoing review and revisions. A draft of an annotated bibliography has been completed and a summary of the results of the workshop proceedings and the literature review is in progress. Key areas for R&D work have been identified and plans to propose some areas for further work are under way.

TRIPS AND VISITORS

August 5 and 6, 1976 - Program personnel traveled to Battelle Seattle Research Center for participation in the Solvent Extraction Equipment Workshop

September 8, 1976 - W. J. Brumley, SRO and J. A. Kelley, SRL, visited PNL to review program progress and plans.

TECHNICAL PROGRESS

An extensive literature search both from computer data bases and hand searches of abstracts turned up applicable citations in excess of 1,000. These citations were sorted for direct applicability to operation and design of solvent extraction equipment, and then further refined to a list of 130 literature citations which were directly applicable to the purposes of this program.

The condensed list of 130 references formed the basis for a literature review which was completed in draft form during the quarter. The 130 references were read and reviewed, and the applicable information was summarized in a review document. This document will be issued as a part of a larger report. In addition, an annotated bibliography was developed for the 130 references. The annotated bibliography consist of four and five line citations for each literature source describing the content and represents a brief abstract of the material in the form of an extended title.

The workshop was held at the Battelle Seattle Research Center on August 5 and 6, 1976. Eighteen people participated in the workshop representing Pacific Northwest Laboratory, Hanford Engineering Development Laboratory, Argonne National Laboratory, Oak Ridge National Laboratory, and Savannah River Laboratory. The object of the workshop was to bring out information which was either unpublished or published in obscure sources concerning operation and design experience with various major types of solvent extraction equipment. A proceedings of that workshop has been prepared in draft form and is currently undergoing review and comment from the participants. Subsequently this draft will be developed into a final proceedings which will also form a part of a larger document.

TASKS IN PROGRESS AND PLANS

Currently in progress is a summary document (of approximately 20 to 30 pages) which will summarize the findings of a literature review and the workshop discussions. This will form a summary part of a large document which will contain the literature review, the workshop proceedings, and the annotated bibliography.

FY-1977 planned work includes completion of the document and proposed work in some of the identified R&D areas.

CATEGORY 7 - FINISHING PROCESSES

(F-RL-14-005)

PROJECT TITLE:	Mixed-Oxide Studies
PROJECT MANAGERS:	Tasks 1, 2 and 4 - S. Goldsmith, Fuels Design and Development Section, Materials Department
	Task 3 - W. S. Kelly, Applied Engineering Development Section, Engineering Technology Department
PRINCIPAL INVESTIGATORS:	Task 1 - W. J. Bailey, Fuels Design and Development Section, Engineering Technology Department
	Task 2 - D. W. Brite, Fuels Design and Development Section, Engineering Technology Department
	Task 3 - D. E. Blahnik, Applied Engineering and Development Section, Engineering Technology Department
	Task 4 - M. D. Freshley and D. W. Brite, Fuels Design and Development Section, Materials Department

OBJECTIVE

The objectives of the four tasks are to: 1) establish the data base required to formulate product specifications for mixed-oxide fuels, 2) formulate reference quality assurance/quality control (QA/QC) requirements for mixed-oxide fuel fabrication, 3) evaluate mixed-oxide fuel fabrication plant equipment and material handling, and 4) investigate the effect of microstructural parameters on fuel performance limits.

SUMMARY

Progress on this task and the proposed modifications to milestones were discussed at the recent SRL-PNL meeting.[a] Efforts to develop a

(a) The meeting was held at PNL on September 1, 1976, and the attendees were: SRL - R. T. Huntoon; PNL - S. Goldsmith, M. D. Freshley, D. E. Blahnik, and W. J. Bailey.

data base report on mixed fuel are continuing; it is presently planned to have a draft of the report completed in CY-1976. The review of available literature in Task 3 was completed during this reporting period. A survey has been initiated to obtain information from other laboratories and commercial fuel suppliers.

TRIPS AND VISITORS

September 1, 1976 - R. T. Huntoon (SRL) visited PNL.

TECHNICAL PROGRESS

TASK 1 - Establish Data Base Required to Formulate Product Specifications for Mixed-Oxide Fuels (W. J. Bailey)

We are continuing to compile and evaluate the available information to facilitate the development of a data base report on mixed-oxide fuel. Currently, the studies on mixed-oxide fuel include the following areas: fuel density, thermal expansion, melting temperature, fuel creep, chemical interaction (e.g., fuel-Zircaloy), fuel swelling and relocation, densification, fission gas release, moisture content, sorbed gas content, and the effect of oxygen/metal ratio. During the recent SRL-PNL meeting we discussed progress on this task, manpower problems (other programs that were expected to diminish in magnitude actually increased; hence, the anticipated manpower for this task was not readily available), and proposed modifications to the milestones. The draft data base report on mixed-oxide fuel is expected to be completed in CY-1976 and the final version of the report is to be completed by March 1, 1977.

TASK 2 - Formulation of Reference QA/QC Requirements for Mixed-Oxide Fuel Fabrication (D. W. Brite)

As a result of discussions with Richard T. Huntoon during his visit of September 1, 1976, the QA/QC requirements and procedures being formulated in

this task activity are being related to the reference LWR mixed-oxide fuel fabrication process being prepared by Westinghouse Hanford (HEDL). These activities are part of the same SRL-ERDA LWR Fuel Cycle Support Program. Completion of a detailed outline of QA/QC requirements, originally scheduled for this reporting period, will be delayed until FY-1977 in order to incorporate significant details of the HEDL reference process in the outline.

TASK 3 - Evaluate and Determine Mixed-Oxide Fuel Fabrication Process and Materials Handling Equipment Development Needs (D. E. Blahnik)

A literature search and a review of the literature was completed. Commercial fuel vendors and other laboratories (foreign and domestic) are being contacted for additional information.

TASK 4 - Investigation of Effect of Microstructural Parameters on Fuel Performance Limits (M. D. Freshley and D. W. Brite)

Work on this task is to be initiated in FY-1977.

TASKS IN PROGRESS AND PLANS

TASK 1

Compilation and evaluation of available data on mixed-oxide fuel for the data base report are to be continued. A draft of the data base report is to be completed by December 31, 1976. A final version of the data base report is to be completed by March 1, 1977.

TASK 2

The reference set of QA/QC requirements will be completed in FY-1977.

TASK 3

The survey will be completed and an informal report will be written which identifies the areas in which further equipment development work is needed.

TASK 4

Initiation of work on this task in the first quarter of FY-1977 is anticipated.

CATEGORY 9 - ENVIRONMENTAL EFFECTS

(F-RL-14-008)

PROJECT TITLE:	Removal and Re-entrainment of Plutonium and Other Radionuclides
PROJECT MANAGER:	C. L. Simpson, Atmospheric Sciences Department
PRINCIPAL INVESTIGATOR:	L. L. Wendell, Atmospheric Physics Section, Atmospheric Sciences Department

OBJECTIVE

The primary objective during this quarter was to compare the U.S. scale transport, dispersion and removal models of Air Resources Laboratory (ARL) and PNL. The purpose of the comparison was to provide a basis for selecting one of the models for performing the relative diffusion calculations and deposition calculations from five regional sites.

SUMMARY

A common meteorological data base for a 1-month period was selected for the model comparison. Trajectories of particles released at 6-hr intervals during the period were calculated with each model at each of the two laboratories. The trajectory sets were exchanged between the laboratories for comparison calculations. A statistical comparison of the separation distances at 3-hourly intervals along the trajectories indicated the discrepancies were 5 to 10%. This is good agreement considering the difference in trajectory construction techniques. The time-averaged air concentration patterns produced by the models for the 30-day period with similar removal parameters also compared quite favorably, despite the differences in computational techniques. Model results are also in the process of being checked against measured concentrations. This test will help to determine how much more physical reality should be included in the models.

TRIPS AND VISITORS

September 21-22, 1976 - L. L. Wendell participated in an LWR Fuel Recycle Environmental Effects Meeting held at SRL to discuss present progress and future plans of each of the participating laboratories.

TECHNICAL PROGRESS

A comparison between the U.S. scale transport dispersion and removal models of ARL and PNL was carried nearly to completion during this quarter. Both models utilize standard radiosonde data to produce a layer average wind at each measurement site, but use different techniques of trajectory construction and somewhat different techniques in sampling the released material to produce average air concentrations at ground level. The differences in the models have resulted from the fact that these models have different sources. The ARL model has evolved from models designed to handle global-scale transport, whereas the PNL model is an adaptation of a mesoscale model. The resulting major difference is in the time and space interpolation processes. The ARL model had to be slightly refined, while the resolution of the PNL model was decreased from that required by the mesoscale. The purpose of the comparison briefly described below was to determine if the models were producing different results, and to provide a basis for the selection of a model for performing the relative diffusion calculations and deposition calculations from five regional sites.

TRAJECTORY COMPARISON

The PNL model under normal circumstances has a release rate of 1 particle per hour and uses a 1-hour advection step. A copy of the model was modified to produce punched cards which contained 3-hourly positions along the trajectories of particles released every 6 hours. This was done to match the format of the cards received from ARL. The ARL model had been used to produce 6 hourly trajectories for the month of April 1974.

The fundamental difference in trajectory construction in the two models is in the technique to obtain the wind at each particle position. The ARL model interpolates directly from the randomly spaced wind measurement locations to the particle position at each time step. The PNL model interpolates from the randomly spaced stations to points on a regular rectangular grid. The winds at the particle positions are then obtained through a bilinear interpolation in space and a linear time interpolation between maps. This method is less expensive for cases in which many particles are on the grid at one time, but a certain amount of accuracy is sacrificed.

A statistical comparison of the particle separations at successive times along the trajectories is shown in Table 6. We may note from the quantity "average separation/distance traveled" the discrepancy is about 10% for the first 3 days and then drops to about 5%. The initial error of 24% could be caused by a wind station being located near the source which would be weighted much more heavily by the ARL model. Since these models are being evaluated for their performance on a regional U.S. scale, this near source discrepancy is probably not serious. Considering the uncertainty of the trajectories due to layer average assumption, the discrepancies between the trajectories produced by the two models would seem to be insignificant. Examples of these trajectory comparisons are shown in the trajectory plots in Figure 5.

COMPARISON OF AVERAGE CONCENTRATION FIELDS

The major concern in the dose calculations is the reliability of the average concentration fields. The concentration calculations were accomplished with both models running in their design mode using the same meteorological conditions, source rate, dry removal rate, and same average rainfall. The models use a different formulation for the dispersion coefficients and different sampling techniques.

Time average concentration fields were produced by each model for the month of April 1974. In spite of the differences in the models, the resulting patterns were quite similar in shape and magnitude.

TABLE 6. Comparison Between ARL and PNL Trajectories

Time After Release, hr	Number of Cases	Number of Cases of Greater Distance Traveled[a]	Average Separation, Nautical Miles	Standard Deviation, Nautical Miles	Average Separation/ Distance Traveled
3	111	69	8.82	6.34	0.24
6	111	66	10.20	10.34	0.14
9	111	63	14.89	15.05	0.13
12	111	57	16.52	15.70	0.11
15	111	55	20.43	16.98	0.10
18	110	52	23.33	17.53	0.10
21	106	53	25.80	18.12	0.09
24	97	44	26.29	17.74	0.09
27	88	39	30.94	23.93	0.10
30	78	31	33.18	27.04	0.10
33	72	30	34.47	27.17	0.10
36	65	26	36.86	27.81	0.10
39	61	23	40.38	30.00	0.10
42	58	20	41.61	28.42	0.10
45	57	17	44.06	30.96	0.09
48	52	11	46.68	32.43	0.09
51	47	10	46.25	33.60	0.09
54	44	9	49.64	38.36	0.09
57	41	10	56.63	44.47	0.10
60	38	11	57.52	49.12	0.10
63	36	9	61.79	58.16	0.10
66	35	9	65.65	65.17	0.10
69	33	9	70.44	75.99	0.10
72	32	8	74.34	83.04	0.10
75	29	8	75.57	91.42	0.10
78	29	8	78.45	98.33	0.10
81	26	9	77.32	108.32	0.09
84	25	8	68.99	102.11	0.08
87	24	8	71.54	107.93	0.08
90	21	7	43.43	20.88	0.06
93	20	7	43.19	21.23	0.05
96	19	6	43.37	22.85	0.05
99	19	6	45.65	23.43	0.05
102	17	5	47.64	24.37	0.05
105	15	4	49.86	25.53	0.05
108	15	5	53.02	33.63	0.05
111	14	4	53.99	29.77	0.05
114	14	4	56.84	31.48	0.05
117	12	3	64.22	33.06	0.05
120	12	3	69.37	44.42	0.05

(a) This compares total lengths of trajectories and the number indicates how many of the PNL trajectories that were greater in total distance traveled than the ARL trajectories

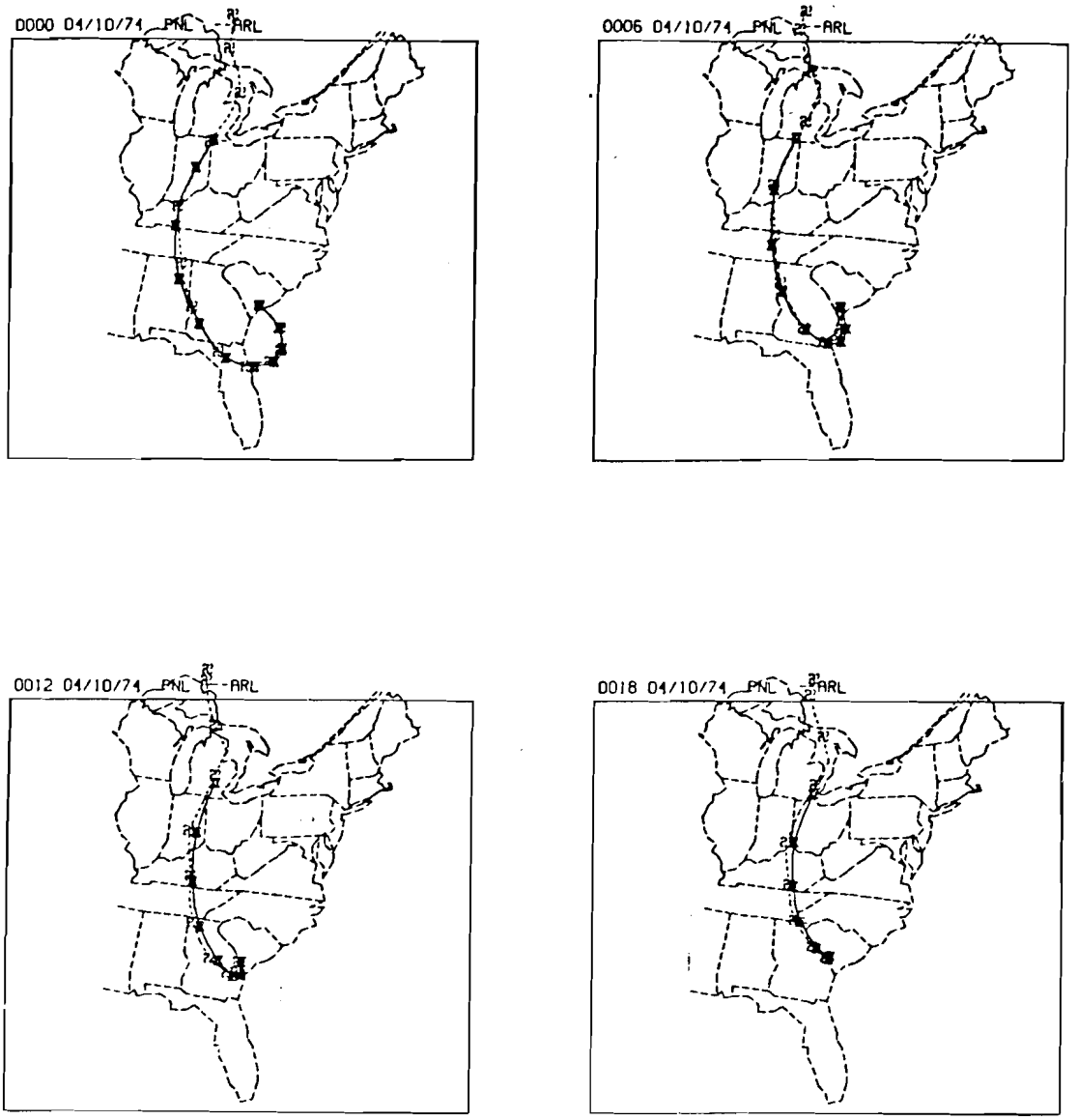

FIGURE 5. Comparison of Trajectories Calculated with Two Different Advection Techniques. The solid lines and symbols were generated with the PNL model. The dashed lines and symbols were generated with the ARL model. The symbols along each trajectory are separated by a 6-hour time interval.

VERIFICATION AGAINST MEASURED DATA

Measurements of 12 hourly average concentrations of ^{85}Kr at Indianapolis, Indiana, and Detroit, Michigan, were available by ARL during the month of April 1975. Also 3 hourly averaged values of the source term were also used to try to simulate the 12 hourly average concentrations at the measurement locations. Because of the variability and uncertainty of the source term as well as some meteorological complications due to a frontal system, it would be premature to draw firm conclusions without further investigation of these cases.

CONCLUSIONS AND RECOMMENDATIONS

From the comparison of the PNL and ARL models conducted at PNL, we would conclude that, despite some differences in their structure, they produce very similar results when compared under the same configuration; e.g., layer averaged flow, homogeneous average precipitation conditions, etc. This fact may be comforting; however, it does not prove very much about the accuracy with which the results approximate reality. The comparison with real measurements provides some hope for progress in this direction.

Further effort should be expended on comparing the results for the April case with results from a model incorporating isentropic principles as well as the actual time and space distribution of the precipitation over the period. This should help refine the wet deposition estimates as well as suggest methods for including precipitation statistics into the simpler models.

TASKS IN PROGRESS AND PLANS

Cases will be more closely examined involving actual measurements of ^{85}Kr through the use of the four-or eight-layer isentropic models. This work should provide a basis for evaluating and improving the performance of the less expensive models. The precipitation data set will be completed for the southeast U.S. for April 1974, for use in testing the wet removal portions of the transport and dispersion models.

CATEGORY 11 - GENERAL SUPPORT

(F-RL-14-006)

PROJECT TITLE: Criticality Safety and Shielding

PROJECT MANAGERS: Tasks 1 and 2 - C. L. Brown, Criticality Safety and Shielding Analysis, Nuclear Technology Department

Task 3 - E. D. Clayton, Criticality Experiments and Operations Section, Nuclear Technology Department

PRINCIPAL INVESTIGATORS: Tasks 1 and 2 - D. R. Oden and M. G. Zimmerman, Criticality Safety and Shielding Analysis Section, Nuclear Technology Department

Task 3 - R. C. Lloyd, Critical Experiments and Operations Section, Nuclear Technology Department

OBJECTIVE

Provide support studies for criticality prevention and process shielding. Review new facilities designed by SRL for fuel reprocessing, plutonium and uranium conversion, plutonium fuel fabrication, and plutonium storage. Provide benchmark criticality experiments to support criticality safety analyses.

SUMMARY

The fuel reprocessing section of the resource book is about 40% complete. Work on the PuO_2-UO_2 fuel fabrication section was started in order to provide criticality safety guidance to Category 7 - Finishing Processes. Work to assess the application of event tree/fault tree metholodology to criticality safety analysis is about 20% complete.

The PNL shielding calculation package has been transmitted to SRL for use in shielding design calculations. The package includes the PNL multigroup Monte Carlo code, a cross-section manipulation code, and cross-section

data. Plutonium source terms for recycled plutonium have been estimated for use in shielding assessment calculations and will be sent to SRL in October.

Calculations were begun in September to identify equipment and fuel configurations to be used in the soluble neutron absorber experiments. These experiments are scheduled to begin in FY-1977.

TRIPS AND VISITORS

None

TECHNICAL PROGRESS

TASK 1 - Criticality Safety Analysis

Task 1a - Develop Resource Book on Experience in Design of LWR Fuel Recycle Facilities from Criticality Safety Viewpoint (D. R. Oden)

A proposed outline of the overall book content was submitted to SRL for comment. Part 1 (Fuel Reprocessing) of the resource book is about 40% complete and other portions of this part will be sent to SRL in draft form as they are completed. This information will be correlated directly with SRL to supplement the design effort. Part 2 (PuO_2-UO_2 Fuel Fabrication) was started earlier than planned in order to provide needed input to Category 7 - Finishing Processes.

Task 1b - Investigate Application of Event Tree Analysis to Criticality Safety in Fuel Processing Plants (D. A. Kottwitz)

Work to develop a risk analysis model for use in criticality safety appraisals is about 20% complete. The objective of this project is to determine if the event tree/fault tree logic can be successfully applied to criticality safety. The method must identify criticality safety limits and controls that block the various paths to potential criticality.

The essential criticality parameters to be used in the logic model have been identified. The double contingency principle of criticality safety analysis states that "process designs should, in general, incorporate sufficient factors of safety to require at least two unlikely, independent, and concurrent changes in process conditions before a critical accident is possible". It is possible to identify each change, or event, as a violation of some criticality parameter limit. Although there is a large variety of possible parameters, they can be put into a limited number of categories as follows:

A. Material quantities
   1. Mass
   2. Volume

B. Material form
   3. Chemical composition
   4. Physical state
   5. Concentration
   6. Enrichment

C. Geometry
   7. Shape of units
   8. Dimensions of units
   9. Spacing of interacting units

D. Processes affecting criticality
   10. Moderation
   11. Reflection
   12. Absorption (by poison)

By incorporating these violation events into fault tree diagrams, it should be possible to determine systematically the combinations of events that could produce criticality. The validity of the above categorization has been checked by reviewing the documented descriptions of actual criticality accidents in nuclear facilities.

Development of the model is now in progress.

TASK 2 - SHIELDING ANALYSIS (M. G. Zimmerman)

Task 2a - Transfer PNL Shielding Calculational System to SRL

The PNL shielding calculational system has been sent to SRL for conversion to the SRL computing system. This consisted of the BMC-MG (Battelle Monte Carlo-Multigroup version code) and the cross-section data and cross-section manipulation codes. As part of this task work is continuing on combining the ORIGEN code with the BMC-MG code and expanding the albedo options in the code for SRL.

The ORIGEN code will provide source terms for the BMC-MG code based on reactor core parameters, and the cooling and reprocessing history of the LWR fuel. The albedo option expansion will provide more flexibility and reduce computer time for many streaming problems.

Task 2b - Prepare Plutonium Source Term Information for Use in Shielding Calculations

The source terms for recycle plutonium have been estimated and will be sent to SRL in October. These source terms include the actual gamma and neutron source intensity data which was updated and included in the BMC-MG code. Also included are PNL estimates of the plutonium isotopic compositions for the first, second, and third LWR fuel cycles. Additional cycle compositions will be added as needed.

TASK 3 - CRITICALITY EXPERIMENTS (R. C. Lloyd)

Task 3a - Plan Soluble Neutron Absorber Benchmark Experiments

This series of experiments is designed to procure benchmark data for validating calculations pertaining to criticality control limits on the use of soluble neutron absorbers (poisons). To determine the effects of soluble poisons [$Gd(NO_3)_3$, and $Cd(NO_3)_2$] on pellet dissolution, a series of experiments will be performed on a lattice of low-enriched uranyl nitrate solutions containing these adsorbers.

Low enriched UO_2 fuel rods of 4.3 wt% ^{235}U have been procured for use in these experiments. The rods are made up from 0.472-in. diameter UO_2 pellets contained in stainless steel fuel tubes; the fuel tube OD (304 SS) is 0.567 in. with the clad thickness being 0.046 in.

Criticality calculations were initiated in connection with pre-experiment planning to define the optimum lattice configurations and neutron absorber concentrations that will be used during the course of the experiments.

TASKS IN PROGRESS

TASK 1

Work will continue toward completion of the fuel reprocessing section of the design resource book. Development of risk analysis methodology as applied to criticality safety analysis will continue to obtain a working model for demonstration purposes.

TASK 2

A benchmark shielding problem will be calculated at PNL and SRL to demonstrate satisfactory operation of the calculational code systems at the two sites. Source terms for recycle plutonium will be documented for use by SRL in future shielding calculations.

TASK 3

The benchmark criticality experiments to measure the poisoning effects of soluble GD and Cd poison in a lattice of 4.3 wt% ^{235}U enriched UO_2 fuel rods will be started.

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