Quarterly Progress Report LWR Fuel Recycle Program

January - March 1977

April 1977

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Battelle

Pacific Northwest Laboratories

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QUARTERLY PROGRESS REPORT
LWR FUEL RECYCLE PROGRAM
JANUARY - MARCH 1977

Compiled by J. H. Jarrett

April 1977

BATTELLE
Pacific Northwest Laboratories
Richland, Washington 99352

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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 (SRL-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 Table 1.

TABLE 1. LWR Fuel Recycle Program Categories

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

* Categories where PNL is conducting research.

This report summarizes PNL activities during the reporting period of January through March. The previous reports in this series are BNWL-2052, BNWL-2080-, BNWL-2080-2, and BNWL-2080-3.

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

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

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

OBJECTIVES

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. Characterize 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. Determine the value of plutonium in either system and evaluate the benefits of various plutonium utilization strategies in the nuclear fuel cycle. Identify decision points in the various plutonium utilization strategies necessary for the implementation of the chosen strategy and/or to avoid logistical inadequacies in the nuclear fuel cycle for that strategy.

SUMMARY

The relationships between LWR recycle decisions and LMFBR fuel supply were examined with the emphasis on contingency rather than strategy optimization. In this context both the conditions where LWR plutonium recycle is and is not allowed were analyzed for typical nuclear growth projections. In order to guarantee a fuel supply for projected LMFBR growth rates, significant multiple plutonium recycle in LWRs will not be possible. However, it is possible to realize about three-fourths of the benefit of multiple plutonium recycle between now and the turn of the century by one recycle and then stockpiling spent mixed oxide for the LMFBR. This stockpile is

CATEGORY 2 (contd)

sufficient to fuel the LMFBR until commercial LMFBR reprocessing can be justified. If plutonium recycle in LWRs is not approved but the LMFBR is still pursued as an energy option, the plutonium supply to the LMFBR is limited by LWR fuels reprocessing capability. To meet the LMFBR feed requirements, approximately 27,000 MT of LWR UO_2 spent fuel must be reprocessed by 1999. This amount of fuel reprocessing would be roughly equivalent to the cumulative total for the Allied General Nuclear Services (AGNS) plant in 1999 assuming it begins continuous operation in 1981.

TRIPS AND VISITORS

January 17-20, 1977 - M. K. White, ERDA, Washington, DC visited PNL to assist in co-authoring an invited paper to be presented in Bal Harbor, FL on May 2, 1977 under this project.

TECHNICAL PROGRESS

The plutonium interface between the LWR and LMFBR fuel cycles was examined for typical nuclear growth projections both with and without plutonium recycle in LWRs. In order to guarantee a fuel supply for projected LMFBR growth rates, significant multiple plutonium recycle in LWRs will not be possible. However, about 78% of the benefit of multiple plutonium recycle between now and the turn of the century is realized by one recycle and then stockpiling spent mixed oxide to fuel the LMFBR. After that time, recycle of plutonium in LWRs will be significantly limited by a continuing LMFBR demand for LWR plutonium due to the projected high LMFBR growth rate. The LMFBR reprocessing schedules were estimated based on accumulation of reprocessing load. These schedules were used to estimate the amount of plutonium recovered from LMFBR fuels and to determine the residual LWR plutonium required to meet LMFBR demand. Before the turn of the century, the plutonium transfer price would be approximately \$23/g of plutonium in undiscounted 1977 dollars when plutonium is recycled once in LWRs.

LWR reprocessing requirements were estimated for the assumed condition that LWR plutonium recycle is not approved, but the LMFBR is still pursued

CATEGORY 2 (contd)

as an energy option. The total amount of plutonium potentially available to the LMFBR is maximized by this LWR option. Since there is no competitive market for plutonium, the LMFBR can likely have LWR spent fuel for the taking. The price of plutonium transferred to the LMFBR under this condition was determined by the cost of reprocessing, waste management and transportation of spent UO_2 fuel minus the credit for the recovered uranium that would be returned for re-enrichment. The net cost for recovery of this plutonium would be approximately \$18/g of plutonium.

If the driving force behind this strategy is to minimize plutonium availability, then there is an obvious incentive to delay reprocessing, while on the other hand the LMFBR must be guaranteed a fuel supply. In Figure 1 the annual LMFBR plutonium requirements are compared to that which can be recovered with an optimistic LMFBR reprocessing schedule beginning in year 2000. The difference between these curves must be supplied annually by reprocessing LWR UO_2 fuels. If one translates this to spent fuel reprocessing requirements, approximately 27×10^3 MT of spent fuel must be reprocessed by 1999. This amount of fuel represents 18 years of operation for a 1500 MT/yr reprocessing plant regardless of the plutonium recycle decision. This would be roughly equivalent to the AGNS reprocessing plant beginning operation in 1981. There are alternative approaches to meeting the anticipated plutonium demand. Between 2000 and 2020 an LWR reprocessing capacity of approximately 11,000 MT/yr would be required. By introducing some of this needed capacity in the early 1990s, the anticipated LMFBR fuel demands for plutonium could also be met. However, the longer this capacity is delayed, the less confidence there will be in the LMFBR as a viable option.

The conflict in this discussion is apparent. In practical terms industry must be assured of the demand for their product before investing in LWR reprocessing. On the other hand, utilities must be assured of a fuel supply to order LMFBRs. Operating AGNS with government plutonium buy-back would be contrary to the desire to avoid large stockpiles of separated plutonium. Not having the LWR as a flexible source and user of plutonium renders the plutonium market unstable and can only slow the anticipated

CATEGORY 2 (contd)

growth rate of LMFBRs. Moreover, accelerated LMFBR reprocessing could support LMFBR growth only to a maximum of its actual fuel doubling-time which is significantly lower than current projections.

FIGURE 1. Annual LMFBR Plutonium Requirements for the Reference Growth Scenario

TASKS IN PROGRESS AND PLANS

Improved models for both the uranium and plutonium recycle mass flows will be incorporated into the analysis. The plutonium and uranium logistics for typical nuclear growth projections will be recalculated. The ability to stop recycle of plutonium and/or uranium in LWRs after a specified date will also be incorporated into computer models.

CATEGORY 2 - ECONOMIC AND ENVIRONMENTAL DOCUMENTATION

(F-RL-14-007)

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 Information Systems Section, Engineering Physics 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, and description and use of "NUCOST" computer code.

TRIPS AND VISITORS

January 26, 1977 - W. A. Reardon attended the ANS Uranium Fuel Supply Meeting in Monterey, California.

TECHNICAL PROGRESS

The first attempts at disaggregating the linear program solution into component costs has led to the identification of boundary problems which make the solution matrices somewhat unstable. Work on these problems constitute the effort on this project during this reporting period. A draft of the document describing NUCOST was prepared and is now being edited.

TASKS IN PROGRESS AND PLANS

Work will continue on the previously described problems.

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, Environmental Technology Evaluation
Portion of the Generic Environmental Impact
Statement for Commercial Waste Management

OBJECTIVE

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

SUMMARY

Draft EIS material was reviewed and commented on.

TRIPS AND VISITORS

None

TECHNICAL PROGRESS

Draft EIS material was reviewed and commented on.

TASKS IN PROGRESS AND PLANS

EIS material will be reviewed and commented on as needed.

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: D. E. Blahnik, Applied Engineering and Develop-
ment Section, Physics 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 and the conceptual design development, and by evaluating the prototype equipment demonstration and operation. Alternate processes will be developed where significant improvements are identified in either plant efficiency or waste management.

SUMMARY

Plans to join ARHCO in procuring an arc saw for end-fitting removal were dropped. This was necessitated when ARHCO changed the location and design to fit their requirements which no longer made the arc saw desirable for fuel reprocessing development. Plans to procure an arc saw designed for end-fitting removal development have been made and procurement action has been initiated.

The spent fuel disassembly development has been dropped from the program since it was not developed sufficiently to be introduced into the reprocessing conceptual design.

CATEGORY 3 (contd)

The CO₂ blast cask cleaning scheme has been slowed somewhat by the vendor's extended prototype fabrication schedule. Preliminary experiments at PNL indicate a good possibility for developing a simple system expressly for cask decontamination.

Recent personal contact with Exxon has resulted in their tentative agreement to furnish some additional fuel design details for purposes of designing fuel reprocessing equipment. Specifically discussed were end-fitting dimensions in sufficient detail to design bundle handling grapples.

TRIPS AND VISITORS

February 2, 1977 - G. S. Allison visited EPRI, Palo Alto, CA
February 3, 1977 - G. S. Allison visited RETECH in Ukiah, CA
March 7, 1977 - G. S. Allison Visited RETECH in Ukiah, CA

TECHNICAL PROGRESS

Discussions with ARHCO resulted in dropping the tentative plan for joint purchase and use of their proposed arc-saw facility for several reasons: 1) they required capital monies rather than expense, 2) the installation was not amenable to handling full-size bundles, and 3) the blade size would not cut through full-size Pressurized Water Reactor (PWR) bundles. A need for an arc saw has developed in the Decontamination and Densification of Chop-Leach Cladding Residues Program, which is closely related to this program. Therefore, funds from the Chop-Leach Program are being requested to procure the capital items for an arc saw installation. The capital items comprise the electrical power supply, the hydraulic power supply, and the arc saw controls. The arc saw will be procured on this program fund. The overall installation is being scoped for end-fitting removal and metallic waste volume reduction development. Initial installation will be at PNL in a nonradioactive facility and may eventually be installed in a reprocessing pilot plant.

CATEGORY 3 (contd)

The affirmative availability of the only operating arc saw for end-fitting removal studies has been established. The saw is installed at the manufacturer's plant and will be suitable for cutting shortened dummy bundles under all pertinent conditions. Discussions with the manufacturer indicate that the parametric studies for end-fitting removal can be performed by the end of FY-77.

A contact with Exxon has resulted in their tentative agreement to supply some fuel bundle dimensions for their fuel. Specifically, we discussed their supply of end fitting designs in sufficient detail to facilitate the design of grappling devices in the reprocessing plant conceptual design. Additional bundle details have been requested, but may not be supplied for proprietary reasons.

A sample search of Preliminary Safety Analysis Reports (PSAR) and Safety Analysis Reports (SAR), which are public information, indicate that much of the desired bundle data has been included.

A preliminary test was conducted at PNL with a CO₂ lab-scale blast cleaning system. CO₂ crystals were tested as a blasting agent at pressures of 40, 25, and 15 psi on samples of lacquer-coated aluminum. The system effectly removed the lacquer.

TASKS IN PROGRESS AND PLANS

Procurement of arc-saw equipment will proceed and experimental data on the manufacturer's arc saw will be initiated. Additional fuel manufacturers will be contacted for their assistance in the fuel design catalog.

Assessment of the availability and potential delivery schedules for commercial CO₂ blast equipment is in progress. Further laboratory testing will be performed at PNL to establish blasting process parameters and to establish design criteria for full-scale test equipment.

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

OBJECTIVE

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

Increased water recycle continues to look attractive, particularly if combined with storage and a modest isotopic enrichment. An agreement has been reached with the Nuclear Energy Programs Division of General Electric, San Jose, California to continue the study. The work will include the impact on plant design and operation.

Dissolution studies of UO_2 in both molten nitrates and mixtures of N_2O_4 and polar solvents were investigated. The oxidation to U(VI) occurs readily in both cases. The major problem may be separation of the U-Pu for the dissolution step.

TRIPS AND VISITORS

March 10, 1977 - H. H. Van Tuyl visited SRO.

CATEGORY 4 (contd)

TECHNICAL PROGRESS

The alternatives for reprocessing LWR fuels were briefly discussed in the previous report.⁽¹⁾ The present study emphasizes two separate paths: 1) conventional head-end and dissolving procedures followed by water recycle, and 2) alternative head-end treatments to remove tritium prior to dissolving.

RECYCLE (L. L. Burger)

Several examples of tritium recycle schemes were discussed in the previous report.⁽¹⁾ Recycle combined with isotopic enrichment and recycle with tritium confined to the first extraction cycle looked especially attractive. Calculations were based on 2 Ci/l as the working concentration. The limiting concentration is controlled by several factors:

- (1) Radiation exposure due to tritium and a corresponding increase in other fission products,
- (2) The extent of leakage into the second extraction cycle,
- (3) The partial pressure of HTO and the imposed limit on airflow (or refrigeration of air).

Subsequent studies will define the role of (1); however, it is quite likely that (2) and (3) will impose a practical limit. For example, to restrict the HTO release to 150 Ci/day from the reference 1500 T/yr plant would limit saturated airflows at 0°C to less than $3.1 \times 10^4 \text{ m}^3/\text{day}$ with 1 Ci/l aqueous and $3.9 \times 10^3 \text{ m}^3/\text{day}$ with 8 Ci/l aqueous. To maintain leakage from the first cycle via the organic stream to an arbitrary minimum of 10% of the 150-Ci/day release rate requires a scrub stream of nearly tritium-free water. For example, an 80% metal saturated TBP stream will carry about 100 l/day of H₂O. At 1 Ci/l, 85% of the extracted HTO would have to be exchanged with H₂O and at 8 Ci/l, 98%. However, 8 Ci/l represents a concentration factor of 67 over the 0.12 Ci/l 1st cycle aqueous or an annual throughput of $1.1 \times 10^5 \text{ l H}_2\text{O}$ which would be sent to storage or to isotopic separation. This can be compared with the reference plant throughput of $3 \times 10^7 \text{ l/yr}$.

CATEGORY 4 (contd)

A combination of first cycle isolation, isotopic enrichment with the tails recycled, and the enriched stream sent to storage continues to look attractive. As was mentioned previously, isotopic separation is much more attractive if combined with recycle since there is no need to produce the high volume, highly depleted stream which would otherwise be required.

Arrangements are being made to have General Electric Company, Nuclear Energy Programs Division, conduct a parametric study to further examine these concept. The study will also consider the impact on plant design and operation.

ALTERNATE HEAD-END METHODS (R. D. Scheele, L. L. Burger)

Work on nonaqueous dissolution methods during this period has included investigations of different nitrate salts and the use of liquid N_2O_4 in polar organics as oxidizing agents.

Pure liquid N_2O_4 does not dissolve UO_2 , but if a polar compound such as TBP is added it becomes reactive.⁽²⁾ Hence, several organic liquids were examined as additives to promote the reaction $N_2O_4 \rightleftharpoons NO^+ + NO_3^-$. Desired properties of the solvent include: high dielectric constant, highly polar, stable toward oxidation, nitration and radiation decomposition, and separable from the U-Pu product or compatible with the extraction solvent. TBP fits most of the criteria, but would degrade extensively during dissolution leading to excessive metal phosphate precipitates.

The following compounds were briefly examined as solvents: propylene carbonate, acetonitrile, nitromethane, ethylacetate, tetrahydrofuran, methylal, and isopropylether. The latter four are probably too reactive toward N_2O_4 to be safely employed although several successful experiments were carried out with ethylacetate- N_2O_4 solutions. Alkylene carbonates and nitromethane are quite stable chemically, although the decomposition products of the carbonates have not been determined. Ambient temperature experiments carried out by bubbling NO_2 through the solvent gave rapid dissolution of UO_2 powder. After 1 hr, $PrCO_3$ dissolved 95%, MeCN 33%, $MeNO_2 > 99\%$ and $EtOAc > 99\%$ in the first experiments. Fuel pellets take somewhat longer to dissolve. With N_2O_4 -nitromethane better results were obtained by refluxing the solution at 30 to 40°C.

CATEGORY 4 (contd)

The principal product uranyl nitrate is quite soluble in the organics used but is precipitated at high concentrations of N_2O_4 .

Possible pyrochemical head-end treatments which could be used as an alternative to voloxidation have been described in the previous quarterly reports.^(1,3,4) The two basic techniques involve the reaction of the spent UO_2 with either a molten-reducing metal or a molten-oxidizing salt. The oxidizing method is less well-defined but may be more compatible with reprocessing plant construction.

The reaction between UO_2 and molten NaNO_3 , NaNO_2 , AgNO_3 , NH_4NO_3 , 55% LiNO_3 - KNO_3 , 50% $\text{Ba}(\text{NO}_3)_2$, and 40% KCl - LiCl was studied. The conditions and results of the experiments are presented in Table 2. As expected, each nitrate salt successfully oxidized UO_2 to the U^{+6} oxidation state. Only Ag_2UO_4 has been positively identified by X-ray diffraction. The product of $\text{NaNO}_3 + \text{UO}_2$ could not be identified by this technique. Several sodium uranates are known. Toussaint and Avogrado⁽⁵⁾ identified a product formed under similar conditions as $\text{Na}_{1.75}\text{U}_2\text{O}_7$. U_3O_8 reacted with NaNO_3 to form a product identical to that formed from UO_2 . Nitrate salts of other elements were considered; however, the remaining possibilities had water in their coordination sphere. These salts either decompose on dehydration or produce very high melting nitrates. The nitrates of Ni, Zn, Cd, and Mg did not melt up to 800°C .

Introduction of large quantities of another cation into the process stream is not desirable. NH_4NO_3 and AgNO_3 have properties which could lend themselves to easy removal of the cation from the process stream, the former by decomposition and the latter by precipitation. Silver has a number of insoluble salts, the halides, chromate, cyanide, etc. However, even if recyclable, the use of >\$1 M inventory of silver is questionable.

Ammonium nitrate decomposition producing H_2O will dilute the fission product tritium with a minimum of about 600 kg H_2O /ton which can be compared to about 2300 kg/ton for a conventional dissolver. This would result in about 3000 ℓ /day of 1 Ci/ ℓ H_2O for storage.

TABLE 2. Reactions of Molten Nitrates with UO_2

Reactants & Media	Melting Point, °C (a)	Temp, °C	Time, hr	Product	Notes
UO_2 powder + excess $NaNO_3$	308	500	1	$Na_{1.75}U_2O_7$ (b)	Reaction vigorous, red to orange brown solids formed
UO_2 pellet + excess $NaNO_3$	308	500	1	$Na_{1.75}U_2O_7$ (b)	Reaction vigorous, causes disaggregation of pellet, red-orange brown solids formed.
UO_2 powder + excess 55% $LiNO_3$ -45% KNO_3	<150	150	2.5		No reaction
UO_2 powder + excess 55% $LiNO_3$ -45% KNO_3	<150	400	1		No reaction occurred until 400°C reached. Vigorous reaction, brown solids.
UO_2 powder + $NaNO_3$ (1:1 mole ratio) in 40% $KCl-LiCl$	<350	450	1.5		brown solids
UO_2 powder + excess $AgNO_3$	208	300	1	Ag_2UO_4 (c)	Vigorous reaction, dark brown solids formed.
UO_2 powder + $NaNO_2$ in 40% $KCl-LiCl$	270	450	0.5		Vigorous reaction. Tungsten crucible used for reaction vessel.
UO_2 powder + 40% $KCl-LiCl$	350	500	1.5		No reaction.
U_3O_8 powder + excess $NaNO_3$	308	500	2.0		Vigorous reaction, solids red-orange brown.
UO_2 powder excess NH_4NO_3	170	200	1		93-96% of uranium dissolved in water, yellow solution produced.
UO_2 powder + excess 50% $Ba(NO_3)_2$ -Ca $(NO_3)_2$	<500	500	2		Green solids formed

(a) Melting point of the salt or salt mixture.

(b) By weight

(c) By X-ray diffraction

CATEGORY 4 (contd)

A neutral molten salt media such as the KCl-LiCl eutectic can be used to control conditions for the voloxidation reaction. We have been using this media to react stoichiometric quantities of molten nitrates and nitrites with UO_2 to better understand the mechanism of the reaction. Avogrado and Deplano⁽⁶⁾ propose the following:

followed by

However, we have seen no evidence of a UO_3 intermediate. The reaction with a 1:1 mole ratio $\text{NaNO}_3 + \text{UO}_2$ produced the uranate. The $\text{NaNO}_2 + \text{UO}_2$ reaction was studied and also produced the uranate.

TASKS IN PROGRESS AND PLANS

More detailed calculations for the water recycle concept will be carried out. Further laboratory studies of UO_2 dissolution using both N_2O_4 - organic systems, and molten nitrate salts will be made to determine which reactions may be given further consideration.

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 TBP vapors from nuclear fuel reprocessing off-gas streams.

SUMMARY

Calibration of the Meloy Laboratories PA-460 real-time phosphorus analyzer is complete using TBP vapors in a flowing air stream. Reproducible concentrations of airborne TBP have been consistently generated in the laboratory for use in bench scale experiments. Experiments have shown that gas chromatographic analysis of TBP samples collected in hexane is accurate and reproducible. Airborne TBP samples collected in a hexane cold trap will be used for backup analysis. Preliminary discussions were held with engineers at PNL exploring the possibility of analyzing TBP vapor concentrations using laser techniques. Laboratory equipment using nearly all glass apparatus is being prepared to perform parametric studies on promising sorbent materials for TBP removal.

CATEGORY 5 (contd)

TRIPS AND VISITORS

None.

TECHNICAL PROGRESS

Calibration of the Meloy Laboratories PA-460 real-time phosphorus analyzer was initiated in early January. The calibration apparatus was constructed primarily of standard-grade stainless steel tubing. Initial efforts revealed TBP adsorption on the interior walls of the tubing and on walls of Teflon® tubing used in the internals of the instrument. The observed adsorption interfered with the calibration procedure. Equipment was ordered to reconstruct the bench-scale apparatus using glass and chromatographic-grade stainless steel tubing. Experiments showed no TBP adsorption on glass tubing. The instrument was modified to eliminate the Teflon® tubing and provide a more direct and shorter pathway for TBP to enter the instrument detector.

Calibration of the analyzer using TBP in a predominantly glass calibration system is complete. No significant adsorption of TBP on tubing walls was noticed. A reproducible logarithmic correspondence between TBP concentration and analyzer response has been found. In addition, laboratory experiments have shown that the TBP generation apparatus produces TBP vapor concentrations which are stable and reproducible.

Backup analysis for this instrument will be performed by gas chromatography on integrated samples collected over a given time period at chosen intervals. These samples will be collected by sparging the TBP-containing gas stream through hexane in a cold trap. Laboratory experiments have shown that this method is accurate and reproducible. A Hewlett-Packard 5736-A gas chromatograph with a N_2/P sensitive detector has been ordered and will be used for the analysis. Previously, gas chromatographic analysis has been performed by the Hanford Environmental Health Foundation (HEHF) laboratories.

CATEGORY 5 (contd)

Preliminary discussions were held with engineers in the Radiological Sciences Department at PNL concerning an alternative method employing laser techniques to measure TBP vapor in gas streams. Laser methodology may be suitable as a remote measurement technique for TBP concentrations in actual PUREX off-gas streams. Some development work will be performed in this area to test the feasibility of this technique. Some difficulty may be encountered with interference by other constituents present in the off-gas. All possible constituents and their likely concentrations have not been identified; however, the major known constituents will be tested for interference.

TASKS IN PROGRESS AND PLANS

Laboratory work will continue to construct the all-glass experimental apparatus for investigating sorbents. Initiation of the parametric experiments will follow the completion of the test system.

If initial experiments on laser techniques to measure TBP concentrations in gas streams show promise, further development work toward building a prototype analyzer will be initiated.

CATEGORY 6 - PUREX PROCESS (SOLVENT EXTRACTION) PART 1
(F-RL-14-004)

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

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 testing of macroreticular polymeric adsorbents and macroreticular anion exchange resins for removal of iodine from simulated fuel reprocessing plant low-level aqueous waste continues to yield promising results. Data were also obtained on suppressing the volatilization of iodine during vaporization of the waste stream by adding mercuric ion to the vaporizer or by vaporizing from a basic solution.

TRIPS AND VISITORS

None

CATEGORY 6 (contd)

TECHNICAL PROGRESS

PROCESS CHEMISTRY (J. L. Swanson)

Emphasis of this program continued to be directed at reducing the amount of iodine which is released with the low-level aqueous waste stream. The two ways of accomplishing this which are currently under investigation are: 1) removal of the iodine from the stream by adsorption on a suitable material, and 2) suppression of the volatilization of the iodine when the waste stream is vaporized for discharge.

Earlier work on the adsorption of elemental iodine from dilute nitric acid solutions by macroreticular polymeric adsorbents emphasized the condition where $0.3M$ H_2O_2 was added to the feed to aid in stabilizing the desired iodine species. The more current work has been aimed at reducing the H_2O_2 concentration (or eliminating it completely) since addition of $0.3M$ H_2O_2 to the entire low-level aqueous waste stream from the AGNS or Exxon plants would result in substantial cost (several hundred thousand dollars per year).

On passing elemental iodine feed solutions containing no H_2O_2 through either XAD-4 or XAD-7 polymeric adsorbent, significant amounts of iodide ion were found in the effluents, more so with XAD-4 than with XAD-7. This observation demonstrates that the adsorbents act as reducing agents. Since iodide ion is retained very poorly by these adsorbents, this observation also indicates that there is little chance of obtaining high iodine decontamination factors on polymeric adsorbents without having an oxidant present in the feed. One possible exception to this indication could arise if the reducing action of the adsorbents could be eliminated by some pretreatment step; limited testing of this possibility has not been successful, however.

Results of column runs performed to measure iodine DF's with XAD-4 and XAD-7 adsorbents at different H_2O_2 concentrations are presented in Table 3. These data indicate that XAD-4 is a better iodine adsorbent than is XAD-7 when there is sufficient peroxide present to overcome the reducing action of the adsorbents, and also that a higher concentration of peroxide is required to overcome the reducing action of XAD-4.

CATEGORY 6 (contd)

TABLE 3. Effect of H_2O_2 Concentration on Iodine Sorption

Feed Make-Up: $1 \times 10^{-5} \text{ M I}_2 + 0.05 \text{ M HNO}_3 + \text{H}_2\text{O}_2$ as indicated

Run Conditions:

- Flow rate—2 column volumes/hr
- Column dimensions = 0.9 cm dia x 19 cm ht
- Temperature—20°C

Total Column Volumes Through	Feed H_2O_2 (M)	Iodine DF ^(a)	
		XAD-4	XAD-7
65	0.3	350	140
125	0.1	32	54
175	0.03	6	18
220	0.3	120	61

(a) Iodine DF = (iodine concentration in feed) ÷ (iodine concentration in effluent)

These data were obtained by running about 65 column volumes of feed containing $0.3 \text{ M H}_2\text{O}_2$, then switching to feed containing $0.1 \text{ M H}_2\text{O}_2$ for about 60 column volumes, then to $0.03 \text{ M H}_2\text{O}_2$ for about 50 column volumes, and then back to $0.3 \text{ M H}_2\text{O}_2$ for about 45 column volumes. Feed flow was continuous, at a rate of about two column volumes/hr.

The results of Table 3 indicate that there is little likelihood of achieving high iodine DF's at peroxide concentrations substantially below 0.3 M unless some other action is also taken. One such action which may be beneficial is the addition to the feed of a low concentration of ferric ion in addition to the peroxide. Kinetic studies using the iodide electrode indicate that the oxidation of iodide ion is significantly faster in $0.05 \text{ M HNO}_3 + 0.02 \text{ M H}_2\text{O}_2$ when 0.001 M Fe (III) is present [but not when $0.0002 \text{ M Fe (III)}$ is present]. This possibility has not yet been tested in column runs, however.

Results of another brief experiment indicate that the presence of dissolved organic materials (TBP and NPH diluent) has little, if any, effect on the sorption of elemental iodine by XAD-4. A 0.05 M HNO_3 solution was

CATEGORY 6 (contd)

equilibrated with 30% TBP-NPH and was then used to prepare a feed solution containing $1 \times 10^{-5} \text{M}$ I_2 and 0.3M H_2O_2 . This feed was run through an XAD-4 bed at a rate of about 5 column volumes/hr. An iodine DF of about 80 was measured after about 80 column volumes, in reasonable agreement with the value of 350 listed in Table 3 (at a rate of two column volumes/hr).

Another potential method for the removal of iodine species from aqueous process streams is to convert the species to iodide ion for removal by an anion exchange resin. Column runs were performed to compare the capacities of various macroreticular anion exchange resins manufactured by Rohm and Haas Company. With $1 \times 10^{-5} \text{M}$ I^- in 0.05M NO_3^- , the iodine concentration in the effluent reached a value 10% as high as the concentration in the feed ("10% breakthrough") after about 100 column volumes with Amberlyst A-26, Amberlite IRA-910, and Amberlite IRA-900. Lower capacities were observed with Amberlite IRA-94 (75 column volumes to 10% breakthrough) and Amberlyst A-21 (55 column volumes to 10% breakthrough). Decreasing the nitrate concentration increases the capacity for iodide; with 0.01M NO_3^- and Amberlite IRA-900 resin, the capacity to 10% breakthrough was about 500 column volumes. Thus, the attractiveness of this method for iodide removal increases as the nitrate concentration of the stream decreases. Because of the relatively low capacity of the anion exchange resins for iodide ion in the dilute nitrate solutions, use of these materials for plant application would likely require regeneration and reuse. Initial tests in this area indicate that the sorbed iodide can be removed to low levels with about 25 column volumes of one or two molar nitrate solutions.

Little effort was devoted this quarter to the use of manganese dioxide-impregnated porous materials as sorbents to remove iodine species from dilute HNO_3 . Improved iodine DF's were obtained initially in one test employing material fired at 500°C (instead of 250°C), but the improved behavior was only short-lived. Testing of such sorbents prepared by a different technique is currently underway.

CATEGORY 6 (contd)

In the work on the suppression of the volatilization of the iodine when the low-level aqueous waste stream is vaporized for discharge, data were obtained on the effectiveness of low concentrations of mercuric ion (in acid solution), and on the effectiveness of vaporizing from a basic solution. The experimental procedure employed here involved heating the bottoms solution (200 ml) to boiling and then adding the feed solution semi-continuously at approximately the same rate as solution was distilled off until about 400 ml had distilled over (the time required was about 1.5 hr). In most of the tests the feed addition point was about 1.5 in. below the surface of the bottoms solution. A test with cesium tracer indicated that the equipment and procedure employed give a de-entrainment factor of 4,000 or more.

The results of the experiments involving mercuric ion are presented in Table 4. The first set of data illustrates the effect of low concentrations of mercuric ion on the iodine volatilization; little effect was obtained with $1 \times 10^{-4} \text{ M Hg}^{++}$, but a pronounced effect was obtained with $3 \times 10^{-4} \text{ M Hg}^{++}$ and a slightly greater effect resulted with $1 \times 10^{-3} \text{ M Hg}^{++}$. The second set of data illustrates that, with $1 \times 10^{-3} \text{ M Hg}^{++}$ in the bottoms, the DF decreases as the iodine concentration in the feed increases from 0.9 to $27 \times 10^{-6} \text{ M}$. The third set of data shows that the DF decreases with increasing iodide to mercuric ratio, although part of the measured effect was probably due to an accompanying increase in nitric acid concentration.

The fourth set of data was obtained in experiments with no iodine in the feed solutions. The results thus provide an indication of the extent of volatilization from the bottoms solution. These data show that volatilization from the bottoms increases with increasing nitric acid concentration, and indicate (since the DF's are higher than in comparable experiments with iodine in the feed) that volatilization from the bottoms is less important in defining the DF than is the efficiency of conversion of the volatile species present in the feed to nonvolatile species.

CATEGORY 6 (contd)

TABLE 4. Iodine DF's During Vaporization of Water from Solutions Containing Mercuric Ion

Expt	Hg ⁺⁺ (Mx10 ³)	Bottoms Composition				Feed Make-Up		DF (feed) ^(a)	DF (bottoms) ^(b)
		[I]/[Hg ⁺⁺]		HNO ₃ (M)		I ₂ (Mx10 ⁶)	HNO ₃ (M)		
		initial	final	initial	final				
1-19	--	--	--	0.05	0.15	9	0.05	1.1	--
1-21	0.1	--	0.006	0.05	0.15	9	0.05	1.2	--
1-26	0.3	--	0.1	0.05	0.15	9	0.05	37	34
1-18	1	--	0.02	0.05	0.15	5	0.05	53	53
2-9	1	--	0.004	0.05	0.15	0.9	0.05	74	83
1-18	1	--	0.02	0.05	0.15	5	0.05	53	53
2-1	1	--	0.1	0.05	0.15	27	0.05	44	48
2-2	1	0.1	0.2	0.15	0.25	28	0.05	14	45
2-3	1	0.2	0.3	0.25	0.35	28	0.05	6	29
2-4	1	0.3	0.4	0.35	0.45	28	0.05	3	20
2-14	1	0.007	0.007	0.25	0.35	--	0.05	--	210
2-7	1	0.4	0.4	0.45	0.55	--	0.05	--	71
2-16	1	0.007	0.007	1.0	1.1	--	0.05	--	18
2-18	1	0.03	0.03	2.0	2.1	--	0.05	--	30
2-9	1	--	0.004	0.05	0.15	0.9	0.05	74	83
3-8 (c)	1	--	0.003	0.05	0.15	0.9	0.05	7 (c)	6 (c)

(a) DF(feed) = (concentration of iodine in feed) ÷ (concentration of iodine in distillate)

(b) DF(bottoms) = (average concentration of iodine in bottoms) ÷ (concentration of iodine in distillate)

(c) Feed solution added to the surface of the bottoms solution

The final set of data in Table 4 compares addition of the feed solution to the surface with addition below the surface. A markedly lower DF was obtained in the surface-addition case, indicating that conversion to non-volatile species was relatively inefficient.

The results of the experiments involving volatilization of iodine during vaporization of water from a basic solution are presented in Table 5. Comparison with the results obtained on the effectiveness of mercuric ion (Table 4) points out several important differences. With the basic solution the same DF was obtained whether the feed was added to the surface or below the surface of the bottoms solution, whereas a much lower DF was obtained in the surface addition case with mercuric ion. Also, the volatilization of iodine from the pot solution is seen to be less in the basic solution case than in the mercuric ion case. The overall DF's are slightly lower in the basic solution case, however, indicating that the efficiency of conversion to nonvolatile species was somewhat lower.

Efforts to obtain meaningful data at higher NaOH concentrations have been unsuccessful because of physical carry-over of bottoms solution into the distillate. This fact was verified by pH measurements on the distillate.

CATEGORY 6 (contd)

TABLE 5. Iodine DF's During Vaporization of Water from Basic Solutions

Expt	Bottoms Composition				Feed Make-Up			DF (feed) ^(a)	DF (bottoms) ^(b)
	NaOH - (M)		I(Mx10 ⁶)		I ₂ (Mx10 ⁶)	HNO ₃ (M)	NaOH (M)		
	initial	final	initial	final					
3-7 (c)	0.02	0.03	--	4	1	--	0.005	32 (c)	32 (c)
3-15	0.02	0.03	--	4	1	--	0.005	30	29
2-24	0.02	0.03	--	40	10	--	0.005	26	24
2-22	0.02	0.01	--	36	9	0.005	--	17	17
2-28	0.03	0.03	40	40	--	--	--	--	> 330

(a) DF(feed) = (concentration of iodine in feed) ÷ (concentration of iodine in distillate)

(b) DF(bottoms) = (average concentration of iodine in bottoms) ÷ (concentration of iodine in distillate)

(c) Feed solution added to the surface of the bottoms solution

ANALYTICAL METHODS (J. L. Cox)

The detection limit of our gas chromatography/electron capture detector system was found to be much higher ($\sim 10^{-6}$ M) for ethyl, hexyl, and heptyl iodides than that ($\sim 10^{-10}$ M) for methyl iodide. This fact seriously limits the applicability of this system to analysis of process streams for alkyl iodides and also renders less applicable a contemplated method for the analysis of low concentrations of iodide ion. In the contemplated method, iodide would be reacted with a tetra-alkylammonium carbonate to form tetra-alkylammonium iodide which would be extracted into an organic solvent. The tetra-alkylammonium iodide would then be decomposed in the injection part of the gas chromatograph (at $\sim 250^\circ\text{C}$) to give an alkyl iodide which would be analyzed.

TASKS IN PROGRESS AND PLANS

Efforts will be continued in the process chemistry areas discussed above. Other analytical methods will be tested and inquiries regarding improved gas chromatographic detector response will be pursued.

CATEGORY 6 (contd)

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

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

SUMMARY

This phase of the project was completed with the publication and distribution of the documents: R. G. Geier, L. M. Browne, Solvent Extraction Equipment Evaluation Study Parts 1, 2, 3. BNWL-2186, Battelle, Pacific Northwest Laboratories, Richland, Washington, February 1977.

CATEGORY 7 - FINISHING PROCESSES
(F-RL-14-005)

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: Tasks 1 and 5 - R. O. Lokken, S. R. Wagoner, and W. J. Bailey, 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 to: establish the data base required to formulate product specifications for mixed-oxide fuels (Task 1); evaluate mixed-oxide fuel fabrication processing and materials handling equipment development needs (Task 3); develop a conceptual automatic fuel pellet inspection process (Task 5).

SUMMARY

Essentially no work was done during this reporting period in Task 1 on improving the draft of the data base report on mixed-oxide ($\text{UO}_2\text{-PuO}_2$) fuels. An informal report for Task 3, which summarizes the mixed-oxide fuel fabrication process and materials handling equipment development needs, was completed and sent to SRL in February.

CATEGORY 7 (contd)

TRIPS AND VISITORS

January 6, 1977 - R. T. Huntoon (SRL) was at PNL to discuss the financial outlook for these studies in FY-1977.

TECHNICAL PROCESS

GENERAL

R. T. Huntoon (SRL) indicated at the meeting at PNL^(a) on January 6, 1977, that the financial outlook for FY-1977 was not clear yet. He asked about our funding status at the time which was as of December 26, 1976, \$13,800 remaining of the initially authorized \$40,000 and of the \$13,800 about \$12,000 was assigned to Task 5. Huntoon suggested that the \$12,000 be reallocated between Tasks 1, 3, and 5 to avoid curtailment of activities until the overall FY-1977 funding question is answered.

By mid-February, essentially all (~98%) of the \$40,000 initially authorized in FY-1977 had been spent. Authorization for \$115,000, the test of the FY-1977 funding for Tasks 1, 3 and 5, was received at PNL on March 16.

TASK 1 - Establish Data Base Required to Formulate Product Specifications for Mixed-Oxide Fuels (R. L. Lokken, S. R. Wagoner, and W. J. Bailey)

The draft of the data base report on mixed-oxide (UO_2 and PuO_2) fuel was completed prior to this reporting period. Because of the limited funding for Tasks 1, 3 and 5 so far in FY-1977, the general depth of the draft on the study in Task 1 was reduced. During January, limited modifications to the draft were initiated as a result of the above reallocation of funds; however, essentially no work was done on the draft during this reporting period since nearly all the remaining dollars from the \$40,000 initially authorized were eventually needed to carry on the work in Tasks 3 and 5.

(a) PNL attendees were: W. J. Coleman, W. S. Kelly, D. E. Blahnik, M. D. Freshley, and W. J. Bailey.

CATEGORY 7 (contd)

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

A literature search and survey of fuel suppliers to determine the extent of development needed for production equipment for a large mixed-oxide fuel plant (~700 MT/yr) was completed. The results of this work were incorporated in an informal report which was mailed to SRL on February 14, 1977. The report summarizes types of equipment, remoteness of operations, and extent of development needed.

TASK 5 - Develop a Conceptual Automatic Fuel Pellet Inspection Process (F. R. Reich)

Due to a delay in receiving funds for this task, program efforts were cut back in the latter part of this reporting period. This task has been designated Task 1 in the revised 189 schedule.

The effort during this reporting period has concentrated on the identification of LWR Fuel Pellet inspection parameters. A previous effort cutback in Tasks 2 and 3 has made this parameter identification effort necessary as most of this data was to come from the Task 2 and 3 results. Some commercial vendor sources of information have been located, but in general this data is difficult to obtain. RDT standards for the FFTF program were analyzed and appear to be directly applicable to LWR fuel pellets. At this time, the following fuel pellet parameters are being pursued for automatic inspection:

- length and diameter
- surface condition
- homogeneity

Other potential parameters include weight and density. Methods to measure alpha distribution on the fuel pellet surface are being examined. The on-going literature search and discussion with people involved in particle detection experiments indicates that direct 2-dimensional (2-D) alpha detection with modern solid-state detector arrays has not been attempted.

CATEGORY 7 (contd)

Information from the FFTF RDT standard indicates that fuel pellet dimensional gauging (length and diameter) which supports calculated density measurements has a much more stringent accuracy requirement (± 0.0002 in.) than the dimensional specifications (± 0.020 in. length and ± 0.0015 in. dia). Several noncontact optical gauging techniques are being studied for dimensional gauging.

TASKS IN PROGRESS AND PLANS

GENERAL

Concurrent with the receipt of the authorization for the remainder of the FY-1977 funds, supplemental work orders were issued to Tasks 3 and 5 so activities on those tasks could proceed. This raised the FY-1977 dollar totals assigned so far to Tasks 3 and 5 to \$30,000 and \$75,000, respectively. Allocation of the rest of the money is to be discussed with the sponsor's technical contact.

TASK 1

If funds are made available, suggested changes or additions will be incorporated into the draft of the data base report on mixed-oxide fuel.

TASK 3

Updating of the survey summary to incorporate responses received since the February report was completed. It is planned to complete survey correspondence and to provide follow-up consultation to SRL on production equipment and layouts.

TASK 5

Evaluation of fuel pellet inspection parameters and inspection techniques will continue. Specifications for additional fuel pellet parameters, such as weight, density and surface condition are being pursued and potential homogeneity inspection techniques will be identified.

CATEGORY 8 - WASTE MANAGEMENT

(F-RL-14-012)

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

Develop and provide the concepts, information and engineering data needed by the LWR Fuel Recycle Program for the design and operation of an electropolishing facility for the decontamination of surface-contaminated solid metallic waste. Major program tasks include: 1) the transfer of compatible technology from the commercial electropolishing industry, 2) the development of solution-treatment procedures to extend electrolyte life and minimize secondary waste volume, and 3) the development of removal techniques for entrained contamination.

SUMMARY

Technology transfer activities continued with visits to electropolishing industry consultants and equipment manufacturers. Studies are in progress based on these contacts to utilize titanium as a low-erosion rate rack material and to adapt industrial nitric acid rinsing procedures for decontamination applications. The use of electrochemical and distillation techniques to remove dissolved metal from spent electrolyte was demonstrated. The ability to decontaminate using dilute electrolytes was also demonstrated. "Throwing power" studies were initiated to develop decontamination procedures for areas that are constricted or shielded from the cathode.

CATEGORY 8 (cont)

TRIPS AND VISITORS

March 8-10, 1977 - R. P. Allen and H. W. Arrowsmith visited J. F. Jumer, Electro Glo Company at Chicago, IL, and L. S. Winter, Hydrite Chemical Company; H. Fishman, Baker Bros.; and N. Campbell, National Plastics and Plating Supply Company at Milwaukee, WI.

TECHNICAL PROGRESS

Electropolishing is a well established industrial process used on a large scale to clean, deburr, and surface finish a variety of metals and alloys. One of our program objectives is to adapt as much of the existing electropolishing and electroplating technology as possible for use in decontamination applications. These technology transfer activities continued during this reporting period with visits to electropolishing industry experts and manufacturers of industrial electropolishing equipment. These contacts included:

- J. F. Jumer, President, Electro Glo Company, Chicago, IL - we discussed this firm's techniques and equipment for electropolishing large quantities of small metal objects without racking and for the in situ electropolishing of pipes, tubes and chemical reaction vessels using a minimum of electrolyte. For example, the interior of a 95,000-gal tank can be electropolished using only 8000-gal of electrolyte.
- L. S. Winter, Senior Vice President, Hydrite Chemical Company, Milwaukee, WI - topics discussed with this program consultant included industrial solution processing techniques, rinsing techniques for the removal of phosphate films, development of electrolytes with enhanced throwing power, and design of electropolishing systems for decontamination applications.
- H. Fishman, Executive Vice President, Baker Bros., Stoughton, MA - discussion included this firm's systems engineering and manufacturing services for more than 400 automated plating and electropolishing installations. Valuable information was obtained concerning: 1) the design and cost of industrial-scale electropolishing systems, 2) automation and

CATEGORY 8 (contd)

modularization concepts for decontamination applications, and 3) advanced filtration techniques for the processing of rinse solutions.

An investigation of titanium as an anode holder and electrical contact material for decontamination applications was initiated based on its use as a long-life rack material in the commercial electropolishing and electroplating industries. Titanium and similar passivating metals such as tantalum, niobium and tungsten form a thin, insulating surface oxide film that inhibits anodic dissolution. However, this film is easily disrupted by racking operations to provide electrical contact to the part to be electropolished.

Our tests using phosphoric acid electrolytes have confirmed the low erosion rate of titanium. The metal loss from titanium was less than one tenth that of copper or 304 stainless steel for equivalent voltages and cell geometries. More importantly, these tests demonstrated that titanium can be decontaminated by electropolishing despite its low erosion rate. However, longer electropolishing times and higher voltages are required to decontaminate titanium as compared with rack materials that do not form a protective surface film. For example, the radiation reading for titanium contaminated with plutonium oxide was reduced from ~30,000 c/m to less than 100 c/m by electropolishing for 30 min at 20 Vdc. Copper and stainless steel under comparable conditions, however, were decontaminated in less than 5 min at 12 Vdc.

The ability to decontaminate rack surfaces and tank walls by electropolishing is just as important as long life for decontamination applications. This permits cleaning of the facility and permits a reduction of contamination levels, whenever required, by reversing electrode polarity. A titanium rack for 8-in. diameter pipe connectors has been fabricated and will be used in an operating decontamination facility to further test the suitability of titanium as a rack material for decontamination applications.

Studies were initiated to adapt nitric acid rinsing procedures used in the commercial electropolishing industry for use in decontamination applications. Preliminary results indicate that rinse solutions containing nitric acid are more effective in removing residual electrolyte from electropolished surfaces than hot water rinsing. These results will be particularly useful for electrolytes containing substantial contamination.

TASK 2. 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. These studies show that precipitation-filtration techniques using methyl alcohol are effective in reducing the dissolved metal content of phosphoric acid electrolytes. A similar procedure using methyl alcohol and ammonia has been developed for the purification of wet-process phosphoric acid (McCullough, et al., U.S. Pat. No. 3,975,178). Precipitation of dissolved metal also can be affected by diluting phosphoric acid electrolyte with water, but this requires dilution ratios greater than 40 to 1. It would be possible, however, to use rinse water as the diluent so that a filtration/distillation procedure would serve both to purify the electrolyte and recycle the rinse solutions.

Studies were initiated to investigate the use of electrolytic techniques to remove dissolved metal from the spent electrolyte. The preliminary results show that iron and nickel can be completely removed from phosphoric acid electrolytes by using a mercury cathode with a high hydrogen overvoltage. For example, an energy input corresponding to 40 A-hr at 12 Vdc was sufficient to reduce the iron content of electrolyte containing anodically dissolved stainless steel from 14 to 0.2 g/l and the nickel content from 2 to less than 0.05 g/l.

Solution-treatment studies conducted for this program by the Hydrite Chemical Company have shown that direct distillation techniques are effective in purifying spent phosphoric-sulfuric acid electrolytes. Additional electropolishing tests using the reclaimed electrolyte are planned to verify the ability to reuse the processed electrolyte.

Other solution-treatment activities during this reporting period included the preparation of a series of standard metal-containing solutions and the calibration of a tube excited X-ray fluorescence system to permit the rapid (<5 min) determination of the dissolved metal content of electrolytes. Precipitation-filtration studies using plutonium contaminated electrolytes were initiated, and the ability to remove plutonium surface

CATEGORY 8 (contd)

contamination using dilute (12 versus 75%) phosphoric acid electrolytes was demonstrated. This latter work represents an alternative to solution processing for extending electrolyte life.

TASK 3. ENTRAINED CONTAMINATION STUDIES

Metallographic studies showed that a potentially contaminated layer up to 300 μm thick is formed on surfaces produced by arc saw and plasma-torch cutting operations. Tests are in progress using a 1500- A electropolishing facility to determine if the normally higher metal dissolution rate at edges (edge effect) is sufficient to remove the disturbed metal layer at cut surfaces.

Further, tests were initiated to compare the effect of electrolyte composition, electropolishing variables, and component geometry on the ability to electropolish inside small tubes, slots, holes, and other areas that are constricted or shielded from the cathode. The "throwing power" of the electropolishing process will directly influence the amount of cutting and component disassembly required to insure decontamination of all shielded surfaces.

TASKS IN PROGRESS AND PLANS

Component-scale testing of titanium as a rack material for decontamination applications will continue. The effectiveness of nitric acid rinsing and dilute electrolyte decontamination techniques will be evaluated using recently completed surface-contamination measurement facilities. Solution-treatment studies will continue with emphasis on precipitation-filtration and electrolytic techniques for the removal of plutonium from contaminated electrolytes. The first series of edge-effect and "throwing power" tests will be completed and evaluated.

CATEGORY 9 - ENVIRONMENTAL EFFECTS

(F-RL-14-008)

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

PROJECT MANAGER: C. L. Simpson, Atmospheric Sciences Department

PRINCIPAL INVESTIGATOR: L. L. Wendell, Atmospheric Physics Section,
Atmospheric Sciences Department

OBJECTIVE

The overall objective of this work is to provide estimates of the confidence limits on the surface air concentrations and deposition amounts of radionuclides 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 the utilization of parallel models, incorporating state-of-the-art treatments of the basic parameterizations in short term comparison tests.

SUMMARY

Previous model comparisons have demonstrated the importance of using time-changing fields of precipitation rather than climatological-averaged data in regional transport and assessment models.⁽⁷⁾ Tapes of hourly precipitation in synoptic field format have been produced this quarter for more than 3000 precipitation stations in the United States for 1974 and 1975. Programs have been adapted so that gridded fields for use in assessment models can be produced from these tapes. With the gridded fields, tests can be made to establish the sensitivity of assessment model results to space and time averaging of precipitation data.

CATEGORY 9 (contd)

Work has also continued on the isentropic model. A test of the model with a release from SRL showed good agreement between observed and computed air concentrations at Detroit, Michigan.

A literature search was made during the quarter on methods for calculating the wet removal of HTO. A recommendation was made to vary "washout ratio" based on surface temperature variations.

TRIPS AND VISITORS

January 31, 1977 - Telephone discussion with George Slinn on the treatment of HTO in wet removal.

February 15-16, 1977 - L. L. Wendell, W. E. Davis and P. J. Dionne participated in a LWR Fuel Recycle Environmental Effects meeting held for discussion of data handling and exchange.

TECHNICAL PROGRESS

HOURLY GRIDDED PRECIPITATION MAPS

As reported last quarter, a computer code was adapted for the producing of hourly synoptic precipitation data. This quarter the code was tested and used to produce tapes for 1974 and 1975. A computer code for gridding the precipitation data was utilized to produce maps for April 1974 on a fine mesh subset of the NMC grid. A second interpolative computer code has been written, tested and run which will convert the fine mesh NMC gridded values to the LWR grid of 0.5 degrees latitude-longitude. An example of the gridded hourly precipitation is shown in Figure 2.

SHORT TERM COMPARISONS

The PNL model was run for the first 15 days of April using: 1) no deposition, 2) dry deposition ($v_d = 1$ cm/sec), 3) wet deposition

FIGURE 2. Map of Gridded Hourly Precipitation for 0.600 GMT April 4, 1974. Areas of precipitation greater than 0.1 in. outlined.

CATEGORY 9 (contd)

($v_w = 2$ cm/sec), and 4) total deposition (VTOTAL = 3 cm/sec). The purpose was to demonstrate the differences which are produced in the air concentrations (see Figures 3 through 6). These comparisons point out the relative roles of wet and dry removal for these parameter specifications. The v_w of 2 cm/sec, calculated for this comparison, was based on an average rainfall of 50 in./yr and a washout ratio of $5 \times 10^{5(8)}$. The results shown here seem to indicate that, for areas which have a moderate rainfall, wet removal can play an important role in the overall environmental assessment.

ISENTROPIC MODEL

The isentropic model was tested for a series of releases from the Savannah River Plant from April 10-14, 1974. A comparison was made of computed concentrations from the model and observed concentrations at Detroit, Michigan. The comparison revealed that when the plume in the model was present at Detroit, an observed increase above background occurred.

The eight layer model has been modified to allow the averaging of winds over layers based on the vertical diffusion. Another change in the model is the inclusion of a simulation of the diurnal cycle of potential temperature. This simulation will be tested to ensure the prevention of over damping of the vertical motion in the vicinity of frontal storms.

RECOMMENDATIONS ON HTO

Recommendations were made this quarter in the treatment of HTO. A review of literature on HTO indicated that from the work of Hales, 1972, and Renne, et al., 1975, that the use of equilibrium scavenging could be used at distances >100 km. Another point brought out in their papers is the dependency of HTO on temperature. Variations in the equilibrium scavenging coefficient ("washout ratios") from $\sim 2 \times 10^5$ at 273°K to $\sim 5 \times 10^4$ at 300°K can occur. These "washout ratios" are lower than the earlier suggested interim values. Strong season variation can be expected with the removal as a function of temperature. Assessment models should take into account the variability.

CATEGORY 9 (contd)

SURFACE AIR CONCENTRATION PC1/M³

NO DEPOSITION

FIGURE 3. Average Surface Air Concentrations (pCi/m³) for April 1-5, 1974 with No Wet or Dry Removal. A continuous source used with a release rate of 1 Ci/hr.

CATEGORY 9 (contd)

SURFACE AIR CONCENTRATION PCi/M**3

VD=1.0(CM/SEC)

FIGURE 4. Average Surface Air Concentrations (pCi/m³) for April 1-15, 1974 with Dry Deposition of 1 cm/sec (see Figure 3).

CATEGORY 9 (contd)

SURFACE AIR CONCENTRATION PCi/M**3

$V_W = 2$ (CM/SEC)

FIGURE 5. Average Surface Air Concentrations (pCi/m^3) for April 1-15, 1974 with Wet Deposition of 2 cm/sec (see Figure 3)

CATEGORY 9 (contd)

SURFACE AIR CONCENTRATION PC1/M**3

VTOTAL=3(CM/SEC)

FIGURE 6. Average Surface Air Concentrations (pCi/m^3) for April 1-15, 1974 with Combined Wet and Dry Deposition of 3 cm/sec (see Figure 3)

CATEGORY 9 (contd)

With temperature $<273^{\circ}\text{K}$ one could expect snow removal of HTO . Previous assumptions made by Hales, 1972, for raindrops (i.e., of rapid mixing within the drop) could no longer be assumed in the case of snowflakes. Thus, a cumulative removal of HTO with height could not be discounted. This could cause a greater washout ratio than one used with equilibrium scavenging. Further work will be necessary in this area.

TASKS IN PROGRESS AND PLANS

- Comparisons of the hourly precipitation with other time and space average precipitation will be made.
- Quality assurance will be run on the deposition results using hourly precipitation.
- A study will be carried out on regional scale-U.S. scale interface problems. This study will also be used to calibrate the ARL and PNL models.
- Include a section in the isentropic model for limiting vertical diffusion during vertical lifting. This model will be compared with results from single layer models.

CATEGORY 11 - GENERAL SUPPORT
(F-RL-14-006)

PROJECT TITLE: Criticality Safety and Shielding
PROJECT MANAGER: Task 1 - Criticality Safety Support - C. L. Brown, Nuclear
Safety Section, Energy Systems Department
Task 2 - Criticality Shield Support - C. L. Brown, Nuclear
Safety Section, Energy Systems Department
Task 3 - E. D. Clayton, Nuclear Criticality Section, Energy
Systems Department

TASKS 1 AND 2

Based on discussions with SRO and SRL staff, no effort has been applied to these tasks during the reporting period.

OBJECTIVE (TASK 3) (R. C. Lloyd)

Provide confirmatory criticality data in support of the LWR Fuel Recycle as needed in code validation, safety assessments, and licensing, to close the fuel cycle safely and economically with assurances.

Pursuant to the LWR Fuel Cycle, it must be emphasized that:

- 1) Operations can be significantly affected through criticality concerns; i.e., by the limits or restrictions that must be applied to insure criticality prevention.
- 2) The licensees must provide assurances that adequate margins of safety have been included in equipment designs and on process controls to account for uncertainties in criticality data and unlikely, but credible, operating errors prior to the granting of a license.

TECHNICAL PROGRESS

A detailed experimental plan was written which provides for a series of experiments to obtain benchmark data for validating calculations pertaining to criticality control limits on the use of soluble neutron absorbers. Fuel rods containing low enriched UO_2 (4.3 wt% ^{235}U) will be used in these experiments. Three lattice spacings of 2.3, 2.8 and 3.3 cm were selected to be immersed in solution. Criticality data will be taken with the fuel

CATEGORY 11 (contd)

rods in solutions of water and neutron poisons and with poison systems where the solution will be uranyl nitrate. Pre-experiment calculations were completed to assist in the choice of the above lattice spacings and in the experiment planning.

With reference to Figures 7 and 8, and Table 6, the most reactive system for either $U(4.3)O_2$ rods in water or $U(4.3)O_2(NO_3)_2$ solution is for a rods-in-water system of ~ 2.5 cm hexagonal pitch. The maximum value of k_∞ is ~ 1.312 , occurring at a ~ 2.3 -cm pitch. The minimum critical cylinder diameter, ~ 17.90 cm, occurs near 2.5-cm hex pitch for a rods-in-water system (infinite-length rods, fully water-reflected) (see Figure 9 and Table 7). With the addition of $U(4.3)O_2$, the material buckling changes from a nearly linear decreasing function at a 2.032 cm pitch to a concave-downward-shaped function at 3.302 cm hexagonal pitch (see Figure 8). From an experiment design point of view, uranyl nitrate solutions having uranium concentrations ≤ 520 g/l uranium are best to work with, as these solutions remain in aqueous form at room temperature. Above this concentration, solutions must be heated to prevent their becoming a solid.

FIGURE 7. B_m^2 and K_∞ Versus Hexagonal Pitch

CATEGORY 11 (contd)

FIGURE 8. B_m^2 Versus Uranyl Nitrate Concentration

TABLE 6. U(4.3)O₂ Rods in Water^(a) B_m^2 and K_∞
 Versus Hexagonal Pitch

HEXAGONAL

PITCH (cm)	$B_m^2 \text{ (cm}^{-2}\text{)}$	K_∞
2.794	8.3248×10^{-3}	1.2814
3.048	7.3012×10^{-3}	1.2443
3.302	6.0064×10^{-3}	1.2001

(a) CLADDING IS 304-L STAINLESS STEEL,
 0.117 cm THICK.

CATEGORY 11 (contd)

FIGURE 9. Critical Cylinder Diameter Versus Hexagonal Pitch

TABLE 7. U(4.3)O₂ Rods in Water^(a) Reflected by 20 cm of H₂O Infinite Rod Height (Lattice Critical Size Versus Hexagonal Pitch)

HEXAGONAL PITCH (cm)	H/U	CRITICAL CYLINDRICAL RADIUS R _c (cm)	NUMBER OF RODS AT THIS PITCH IN A CYLINDER OF DIAMETER R _c
1.778	2.8	25.67	756
2.032	4.8	19.42	331
2.286	7.2	17.92	223
2.54	9.9	17.94	181
2.794	12.8	18.84	165
3.048	16.0	20.52	165
3.302	19.5	23.19	179

(a) CLADDING IS 304-L STAINLESS STEEL 0.117 cm THICK.

CATEGORY 11 (contd)

Figure 10 and Tables 8 and 9 describe the behavior of the material buckling with the addition of excess nitric acid and $\text{Cd}(\text{NO}_3)_2$. For the range of hexagonal pitches studied, the addition of excess nitric acid and $\text{Cd}(\text{NO}_3)_2$, both have a negative effect on B_m^2 . For example at a uranium concentration of 200 g/l uranium in uranyl nitrate and at an enrichment of 4.3 wt% ^{235}U , B_m^2 changes from a $7.9966 \times 10^{-3} \text{ cm}^{-2}$ value at a 2.54-cm pitch, 0 M excess nitric acid, to $7.3037 \times 10^{-3} \text{ cm}^{-2}$ at 2 M excess nitric acid, a difference of $\sim 0.693 \times 10^{-3} \text{ cm}^{-2}$. This difference increases to $\sim 0.743 \times 10^{-3} \text{ cm}^{-2}$ at a 3.302-cm pitch, suggesting that the nitric acid is a more effective poison as the lattice spacing opens up. A similar, more radical, change is also observed with the addition of $\text{Cd}(\text{NO}_3)_2$ (see Table 10). The value of B_m^2 is $7.3037 \times 10^{-3} \text{ cm}^{-2}$ for a 2 M excess nitric acid solution, 200 g/l U in solution and 2.54 cm pitch, and B_m^2 changes to $4.4863 \times 10^{-3} \text{ cm}^{-2}$ with the addition of 2 g/l $\text{Cd}(\text{NO}_3)_2$. This amounts to a change of $2.817 \times 10^{-3} \text{ cm}^{-2}$. Again, as the lattice spacing opens up, the change in B_m^2 increases to a value of $4.490 \times 10^{-3} \text{ cm}^{-2}$ at 3.302 cm pitch.

These data were used in designing the experiment vessels.

Pre-experimental planning and design of experimental equipment have been completed. The engineering drawings are in the final stages of completion. Fabrication and ordering of equipment needed to carry out the experiments is now in progress. Planning for the dissolution of the UO_2 into uranyl nitrate solution required for the experiments continued. Laboratory-scale dissolution tests were made in planning for the larger volume dissolution necessary for these experiments.

CATEGORY 11 (contd)

FIGURE 10. B_m^2 Versus Concentration of Cd(NO₃)₂

TABLE 8. U(4.3)O₂ Rods in Uranyl Nitrate Solution^(a) B_m^2 and K_{∞} Versus Uranyl Nitrate Concentration

SOLUTION CONCENTRATION (g/l U)	HEXAGONAL PITCH (cm)	B_m^2 ($\times 10^{-3} \text{ cm}^{-2}$)	K_{∞}
100	2.032	6.9437	1.2735
50	2.032	7.2306	1.2819
900	2.54	5.9342	1.2519
500	2.54	7.1704	1.2751
200	2.54	7.9966	1.2873
100	2.54	8.3389	1.2937
900	2.794	6.3604	1.2629
500	2.794	7.3631	1.2754
200	2.794	7.8084	1.2741
100	2.794	7.9653	1.2743
500	3.048	7.3244	1.2696
200	3.048	7.3033	1.2529
100	3.048	7.2222	1.2457
50	3.048	7.2163	1.2435
500	3.302	7.1551	1.2606
200	3.302	6.6181	1.2275
100	3.302	6.2603	1.2117
50	3.302	6.0865	1.2043

(a) 0 M EXCESS NITRIC ACID IN THE SOLUTION. CLADDING IS 304-L STAINLESS STEEL, 0.117 cm THICK.

CATEGORY 11 (contd)

TABLE 9. U(4.3)O₂ Rods in Uranyl Nitrate Solution. Poisoned with Cd(NO₃)₂ (a) B_m² and K_∞ Versus Poison Concentration.

Cd(NO ₃) ₂ CONCENTRATION (g / l Cd(NO ₃) ₂)	HEXAGONAL PITCH (cm)	B _m ² (x 10 ⁻³ cm ⁻²)	K _∞
0	2.286	7.20 ^(b)	--
1	2.286	6.18 ^(b)	--
2	2.286	5.05 ^(b)	--
0	2.540	7.3037	1.2720
1	2.540	5.8323	1.2166
2	2.540	4.4863	1.1664
0	2.794	7.0853	1.2579
1	2.794	5.2648	1.1910
2	2.794	3.6292	1.1320
0	3.048	6.5656	1.2359
1	3.048	4.4327	1.1587
2	3.048	2.5505	1.0915
0	3.302	5.8751	1.2095
1	3.302	3.4686	1.1232
2	3.302	1.3849	1.0492

(a) 2 M EXCESS NITRIC ACID IN THE SOLUTION; U CONCENTRATION OF 200 g / l U.

(b) EXTRAPOLATED VALUES.

TABLE 10. U(4.3)O₂ Rods in Uranyl Nitrate Solution
Change in B_m² Due to Cd(NO₃)₂ Addition (a)

HEXAGONAL PITCH (cm)	B _m ² (x 10 ⁻³ cm ⁻²) NO Cd(NO ₃) ₂	B _m ² (x 10 ⁻³ cm ⁻²) 2g / l Cd(NO ₃) ₂	ΔB _m (x 10 ⁻³ cm ⁻²) (UNPOISONED - POISONED)
2.286	7.20 ^(b)	5.05 ^(b)	2.15
2.540	7.3037	4.4863	2.817
2.794	7.0853	3.6292	3.456
3.048	6.5656	2.5505	4.015
3.302	5.8751	1.3849	4.490

(a) 200 g / l U IN U(4.3)O₂(NO₃)₂ SOLUTION, 2 M EXCESS NITRIC ACID.

(b) INTERPOLATED VALUES

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