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AFCT/TFCT/ISFS PROGRAM

TECHNICAL PROGRESS REPORT FOR THE PERIOD APRIL 1, 1978 - JUNE 30, 1978

COMPILED BY
O. F. Hill

August 1978

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AFCT/TFCT/ISFS PROGRAM

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O. F. Hill

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Prepared for the
Department of Energy
Fuel Cycle Program Office
Under Contract EY-76-C-06-1830

Pacific Northwest Laboratory
Richland, Washington 99352

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~~FUEL CYCLE~~

ABSTRACT

This is the tenth in a series of quarterly progress reports on research and development studies performed by the Pacific Northwest Laboratory (PNL) for the Alternate Fuel Cycle Technologies/Thorium Fuel Cycle Technologies/International Spent Fuel Receipt and Storage (AFCT/TFCT/ISFS) Program. This program, sponsored by the Department of Energy (DOE), provides information needed by industry to close the back end of the power reactor fuel cycle. Included in the program are activities supporting specific design studies, as well as activities for general fuel cycle technology. The overall program is managed by Savannah River Laboratory under the direction of the Fuel Cycle Program Office of the Savannah River Operations Office of the DOE.

During the past quarter, PNL conducted studies in the following tasks:

<u>Subtask</u>	<u>Project Title</u>
130.4	Thorium Resource Price Analyses
210.1	Spent Fuel Receipt and Storage
340.3	Investigation of Air Cleaning Processes for Removing Tributyl Phosphate (TBP) from Off-Gas Streams
350.5	Study of Iodine Chemistry in Process Solutions
610.2	High-Level Waste Treatment
650.1- 650.2	Electropolishing to Decontaminate Metallic Waste from Alternate and Thorium Converter Fuel Cycles
810	U.S. Scale Transport, Dispersion and Removal Model Comparison
840.2	Safety Criticality Experiments
840.3	Criticality Research in Support of Thorium Fuel Cycle Technology Program

This quarterly report summarizes PNL activities during the months of April, May, and June 1978. Previous quarterly reports in this series are BNWL-2052 and BNWL-2080-1 through -8.

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AFCT/TFCT/ISFS PROGRAM
TECHNICAL PROGRESS REPORT FOR THE PERIOD
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INTRODUCTION

In fiscal year 1976, the Fuel Cycle Program Office (FCPO)^a in the Savannah River Office (SR) requested the Pacific Northwest Laboratory (PNL), operated by Battelle Memorial Institute, to conduct studies in support of the back end of the Light Water Reactor (LWR) fuel cycle. Steps in the back end of the fuel cycle include spent fuel receipt and storage, spent fuel reprocessing, uranium and plutonium conversion for recycle, recycle fuel fabrication, and waste management. The Savannah River Laboratory (SRL), administered by E. I. duPont de Nemours and Company, was assigned lead responsibility for technical management of the programs.

In April 1977, the administration announced its policy of indefinite deferral of commercial reprocessing and recycle to permit a reevaluation of alternatives to the normal LWR fuel cycle, alternatives that would provide proliferation-resistant systems. Alternative fuel cycle systems include coprocessing, spiking, thorium/²³³U cycles and other variants. Thus, changes in direction of investigations in the back end of the fuel cycle result in programs in alternative fuel cycle technology (AFCT), thorium fuel cycle technology (TFCT), and international spent fuel receipt and storage (ISFS).

Our investigations under these programs include studies related to:

- the availability and economics of thorium ores suitable for mining, milling and processing
- receipt and storage of spent fuels
- investigations of the chemistry of iodine in off gasses and low activity aqueous streams from reprocessing plants to eliminate these materials as sources for release of radioiodine to the environment
- conversion of high-level liquid wastes from thorium fuels to vitrified solid waste forms
- electropolishing of metallic wastes to reduce the quantity of waste that must be disposed of in a federal repository
- dispersion and deposition of radionuclides using meteorological models
- assessment of criticality parameters as they apply to the alternative fuel cycles being considered.

The following sections discuss the progress made in each of these areas during the past quarter.

(a) The FCPO was formerly part of the Energy Research and Development Administration, presently the Department of Energy.

PROJECT TITLE: THORIUM RESOURCE PRICE ANALYSIS

PROJECT MANAGER: R. M. Fleischman, Fuel Cycle Analysis,
Energy Systems Department

PRINCIPAL INVESTIGATOR: C. H. Bloomster, Advanced Energy Analysis,
Energy Systems Department

OBJECTIVE

The large-scale use of thorium in thermal power reactors is being evaluated by SRL and PNL. The possible ranges of costs for reactor grade thorium oxide are needed to calculate fuel cycle costs and to determine the economic viability of the various proposed cycles. The purpose of the thorium resource price analysis is to provide the data needed for the fuel cycle economic studies. The supply curve will provide the basis for estimating the price and sources of thorium and will provide the input of thorium price projections needed for the nuclear fuel cycle cost analysis.

SUMMARY

- Information for the supply curve was updated for domestic thorium resources.
- The potential amount of byproducts associated with large-scale thorium production was determined.
- Data on thorium is more complete; however, resource and cost estimates are still being evaluated.

TRIPS AND VISITORS

May 22, 1978 - Dr. Roy Post, University of Arizona, visited PNL to discuss collaboration on thorium resource evaluation and conceptual facility design.

TECHNICAL PROGRESS

Deposit Analysis

This quarter we have updated the preliminary analysis of domestic thorium resources.

The changes to date are discussed in the following sections.

Stockpiles

Several stockpiles of thorium currently exist in the U.S. These stockpiles are in a semiprocessed state and thus have potentially low production costs. Information on stockpiles is available from the National Lead Co. of Ohio. However, DOE approval is necessary before PNL can gain access to the information. We are waiting for this approval.

Thorite Deposits

Although thorite is not currently mined in the United States, thorite deposits offer a promising source of thorium. Conceptual recovery methods were developed for three domestic thorite deposits: Wet Mt., Colorado, Hall Mountain, Idaho, and the Lemhi Pass Range, located in Idaho and Montana. The thorite recovery facilities consist of a mine, mill, and refinery.

The services of a consulting agency were obtained to do a detailed mining cost evaluation of the Lemhi Pass resource. This resource was chosen because it provided a good model for underground mining, which could then be applied to other deposits.

The quantity of identifiable and mineable resources for Lemhi Pass is estimated at 120,000 tons of thorium oxide. Additional reserves (130,000 tons) are hypothesized, but insufficient information is available upon which to base cost estimates. The estimated mining cost for the largest veins of Lemhi Pass containing about 20% of the ore is \$4.43/lb ThO₂. The additional cost associated with mining the other smaller thorium

veins is currently being determined. When information is complete, the cost of refining will be added to the mining cost and the Lemhi Pass production cost estimates will be completed.

After the Lemhi Pass mining model is completed, the associated mining costs will be adapted to the mining conditions at Hall and Wet Mountains. Thorium reserves from these sources are estimated at 7500 short tons of ThO_2 . Mining costs are expected to range from \$1 to \$7/lb ThO_2 .

Conglomerate Deposits

Monazite conglomerate is mined by surface mining methods. The conglomerate then undergoes extensive grinding and crushing followed by gravity and magnetic separation. A monazite concentrate is obtained, which is re-fined using acid leaching. Three facilities complete the conglomerate processing stream: the mine, the mill, and the refinery. Two conglomerate deposits were studied: Palmer, Michigan, and Bald Mountain, Wyoming.

The services of a consulting agency were obtained to do a detailed mining cost estimate for Palmer, Michigan. Palmer was chosen because it provided a good model for surface mining. Results of this study indicate that 72,000 short tons of ThO_2 can be recovered from Palmer at an average unit cost of under \$20/lb ThO_2 . Additional ThO_2 is recoverable at a much higher price.

Based on Palmer, Michigan, cost estimates, a production cost for Bald Mountain will be derived that reflects the associated resource and mining conditions. These conditions were evaluated after contacting a consulting geologist who described the topography and recommended mining methods. Bald Mountain contains an estimated 2200 short tons of ThO_2 .

Placer Deposits

Thorium-bearing monazite sand occurs as beach placers and stream placers. The specific mining techniques will be determined by the nature of the deposit; however, existing monazite mines utilize a dredge. The sand is concentrated in a mill and the monazite obtained is sent to a refinery, where rare earths are removed and reactor grade thorium oxide is produced. Monazite deposits include Bear Valley, Idaho, and the Piedmont placers in North and South Carolina.

Production costs for the Bear Valley thorium deposit are being determined. A former dredging operator at a mine located

in Bear Valley was contacted, and he has agreed to furnish information on previous mining methods and costs.

Piedmont placer deposits have been characterized. Total mineable reserves are estimated at 11,000 short tons. A great quantity of additional reserves are also located here, but large-scale methods of mining the deposits are not feasible.

Carbonatite Deposits

Carbonatite deposits represent another potential source of thorium. The conceptual carbonatite processing stream consists of three facilities: the mine, the mill, and the refinery. Two carbonatite deposits were studied, Iron Hill, Colorado, and Mountain Pass, California.

Surface mining techniques were studied for removing carbonatite from Iron Hill. A mill concentrates the thorium and pyrochlore contained in the ore. The mill concentrate is mixed with hydrochloric acid, and a niobium-containing residue remains, while the thorium and rare earths are leached from the ore. The rare earths are separated by fractional precipitation and the thorium is purified using solvent extraction. Thorium production costs are dependent on the value of by-products.

Information on Mountain Pass is currently insufficient to estimate mining costs; thus, evaluation of this resource does not seem practical at this time.

Conway Granite Deposits

Conway granite is a large but expensive source of thorium. While granite has not been mined commercially, the processes necessary for mining and refining the granite were studied previously by the Atomic Energy Commission (AEC). The conceptual granite processing stream consists of a surface mine followed by a refinery. Costs were also determined by the AEC and will be reevaluated for determination of current production costs.

Byproduct Credit

In an effort to determine the potential value of byproducts, the amount of rare earths, titanium and zirconium potentially available as a result of thorium production has been reevaluated. As much as 100,000 tons/yr of contained rare earth oxides, 46,000 tons/yr of titanium, and 6000 tons/yr of zirconium could be produced in association with large-scale thorium production.

Results indicate that the rare earth market would be severely oversupplied and the value of the rare earth byproducts would be greatly reduced. Zirconium and titanium markets would be affected to a much lesser degree. Niobium byproduct production must still be determined.

PLANS FOR NEXT QUARTER

The supply curve will continue to be updated as new information is obtained and current information is evaluated. Detailed cost analyses for Palmer, Michigan, and the Lehmi

Pass deposits will be completed. This data will be applied to estimate the mining costs of similar deposits. The analysis of the elasticity of byproduct credit will be completed so a more confident estimate of byproduct credit can be determined. This information shall be presented in a more accurate supply curve which will replace the preliminary curve presented last quarter. The USGS is currently completing a new assessment of domestic thorium resources. We hope to obtain this information as soon as practical so that the thorium supply curves can be formulated from the latest resource information.

PROJECT TITLE: SURVEY CURRENT TECHNOLOGY OF FUEL HANDLING TECHNIQUES

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

PRINCIPAL INVESTIGATORS: K. R. Ames, Applied Engineering and Development Section,
Physics Department

G. S. Allison, Materials Development Section,
Materials Department

OBJECTIVE

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

SUMMARY

- The arc saw was delivered in mid-May 1978.
- Preliminary acceptance cutting tests on the arc saw delineated only a few, easily corrected problems.
- The report of cutting trials on RETECH's prototype saw is ready for distribution pending DOE approval.
- Attempts to procure dummy fuel bundles from fuel vendors have been essentially unsuccessful.
- Blast tests were run on contaminated coupons.
- In our negotiations Lockheed was willing to help by renting equipment, but technical problems with the equipment have prevented it so far.
- The CO₂ blast test was terminated in June.
- The Fuel Design Catalog is ready for distribution pending written release by affected fuel manufacturers.

TRIPS AND VISITORS

January 3, 1978 - Ken Ames and Chuck Conrad visited Cal Fong and Lowell Turner at Lockheed to discuss prospects of using Lockheed's equipment.

January 8, 1978 - G. S. Allison visited Savannah River Laboratory to discuss AFCT/ISFS programs.

January 26, 1978 - Ken Ames and Chuck Conrad visited Cal Fong and Lowell Turner at Lockheed to witness pelletizer operation and further discuss potential use of Lockheed equipment.

February 16, 1978 - G. S. Allison and Wayne Weihermiller visited RETECH, Ukiah, CA.

TECHNICAL PROGRESS

The arc saw was delivered in mid-May 1978; this late date was due to delays in manufacturing some of the component parts, such as the hydraulic power supply. The preliminary acceptance cutting tests showed only minor problems. The controls operated satisfactorily in spite of a stick-slip problem with the main feed cylinder. Several cuts were made without the longitudinal head clamp, which permitted some wandering of the cut. Both of these problems are being corrected. The arc saw is anchored in place and the plumbing and wiring are progressing.

Attempts to procure fabricated dummy fuel bundles from fuel vendors have been disappointingly unsuccessful. There were several "No-Bids". The few positive

responses entailed high cost (>\$20,000), long delivery (6 to 8 months), and proprietary restrictions on the use of the bundles and data generated from cutting them.

Reject end-fittings and spacers have been offered. These will be procured and fitted with zirconium tubing with end plugs to simulate the bundles for cutting trials on the arc saw. Most, if not all, the configurations expected to ultimately be cut will thus be included in the cutting trials. The limits of the expected envelopes will be covered.

The publication of the Fuel Design Catalog report entitled "LWR Nuclear Fuel Bundle Data for Use in Fuel Handling" was delayed for a final data check and to get releases before submitting the final draft for SR approval.

The report of the September 1977 cutting trials on the vendor's prototype arc saw is ready for distribution pending SR release.

The primary thrust of work in the past quarter has been to demonstrate that a blast of CO₂ particles is capable of removing low-level contamination similar to that found on fuel shipping casks.

To accomplish this goal, work continued on the PNL-developed lab-scale blast equipment. When it appeared that the equipment would soon be ready, a search was made for a suitable facility for tests on radioactive contamination. Ideal

accommodations were found at U.S. Testing, and a glovebox/hood was prepared for use. Contaminated basin water was obtained from UNI at the 100 K basin. Tests on contaminated coupons began in early March and demonstrated that a CO₂ blast would remove contamination. Decontamination factors as high as 342 and as low as 3 were recorded on coupons that ranged from 1.4 to 4.8 x 10⁴ disintegrations/min per 100 cm² smearable. More tests will be run to determine what factors influence the decon factor. Control tests using standard wipedown methods yielded decontamination factors of 28 and 30.

An effort was made to use Lockheed's CO₂ pellet blasting equipment for testing, but Lockheed experienced technical problems with the equipment, so they were unwilling to loan it at that time. Negotiations are continuing and their equipment will be tested when it is available.

In June, at the request of SR, work on the CO₂ task was terminated for the remainder of fiscal year 1978.

PLANS FOR NEXT QUARTER

- Install and check out operation of arc saw.
- Fabricate simulated dummy bundles for cutting trials.
- Prepare demonstration of PNL CO₂ blast equipment for SR and SRL personnel.

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

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

SUMMARY

- The first experiment is complete using the bench-scale demonstration unit to determine the effectiveness of the alumina catalyst (material H) to protect an iodine sorbent. Results indicate that the iodine retention of columns with and without the protective sorbent (H) was nearly identical and that the retention was near the stoichiometric iodine retention capacity of the sorbent. The calculated airborne TBP concentration of approximately 8×10^{-4} g/l had little effect in reducing the capacity of the 7 wt% Ag iodine sorbent used in the experiment.
- A TBP loading experiment of 6 x 14 mesh crushed catalyst (material D) was completed. The TBP capacity of 3.8 cm of material D was calculated to be 39 mg/g material at a bed temperature of 100°C, air flow of 1.9 l/min, and TBP concentration of 1×10^{-4} g/l. The TBP retention is 3.5 times greater for crushed material (D) than for the spherical form, which was loaded in an earlier experiment in late February 1978. However, retention is less than the alumina catalyst (material H), which retained 377 mg/g in 3.8 cm.
- A granular 3 x 30 mesh silica-alumina-magnesia sorptive clay (material L) was loaded with TBP at a bed temperature of

100°C, air flow of 2.0 l/min, and TBP concentration of 6×10^{-5} g/l. Total TBP capacity of 3.8 cm of material was 70 mg/g material, which is greater than material D but less than material H.

- The work to develop an alternative analysis technique using an optoacoustic laser has been delayed due to equipment problems. This laser analysis method appears feasible although work in the area is relatively new and undeveloped.

TRIPS AND VISITORS

June 20, 1978 - Bruce Wilson, FCPO/SR, visited G. B. Parker to discuss program progress and to tour PNL.

TECHNICAL PROGRESS

An experiment was completed in late April using the bench-scale demonstration unit to evaluate the alumina catalyst (material H) as a sorbent to remove TBP and protect iodine sorbent beds. Two 2.5 cm diameter stainless steel columns were prepared: Column I, containing five 5 cm segments of a 7% Ag iodine sorbent; and Column II, containing two 5 cm segments of material H followed by three 5 cm segments of the 7% Ag iodine sorbent.

Specified experimental conditions in each column were column bed segment temperatures 130°C, air at 2% relative humidity flowing at 1.9 l/min, and average methyl iodide concentration of 1.6×10^{-4} g/l. Methyl iodide was analyzed using a gas chromatograph. The calculated airborne TBP concentration during the experiment was approximately 8×10^{-4} g/l. The TBP was introduced by sparging the feed stream through a mixture of 30% TBP/dodecane.

At the end of the experiment, 8.5 g of methyl iodide had been metered to each column. During the experiment, the breakthrough progression of methyl iodide was followed through each segment of the silver sorbent. This breakthrough is represented as a graph of C/C_0 vs grams methyl iodide metered, where C represents the concentration of methyl iodide leaving a 5-cm column segment of iodine sorbent, and C_0 represents the concentration of methyl iodide in the feed stream to the column. The breakthrough curves for Column I are in Figure 1 and those for Column II are in Figure 2.

To determine the effectiveness of the TBP sorbent to protect the iodine sorbent, a comparison of breakthrough curves is made between Section A in Column I and Section C in Column II, Section B in Column I and Section D in Column II, and Section C in Column I and Section E in Column II. Table 1 summarizes breakthrough data.

The iodine retention of the iodine sorbent is nearly the same in Sections A and B of Column I, and Sections C and D of Column II. This indicates that the presence of material H in Sections A and B of Column II had little effect on the iodine retention of the iodine sorbent located downstream. The data also indicate that iodine retention was near the stoichiometric maximum (within the uncertainty), and therefore the presence of airborne TBP did not have a deleterious effect on the iodine sorbent.

A TBP loading experiment using 15.1 g of 6 x 14 mesh catalyst (material D) was con-

ducted in a single glass column under the following conditions:

- Bed temperature: 100°C
- Average airflow: 1.9 l/min
- TBP concentration: 1×10^{-4} g/l.

Breakthrough of TBP was noted nearly instantaneously; the run was terminated after 0.97 g TBP was metered to 3.8 cm of material in 73.1 hours. The breakthrough curve for this material is shown in Figure 3. Calculated TBP retention of the crushed material is 39 mg TBP/g material.

The TBP retention of the crushed form of material D was 3.5 times greater than the spherical form of the material loaded in an earlier experiment under identical conditions. However, the TBP retention is considerably less than that achieved with the alumina catalyst material H. Further experiments with material D will not be conducted due to the very low TBP retention.

An experiment was conducted to load TBP onto an 8 x 30 mesh granular silica-alumina-magnesia sorptive clay, which indicated the third highest TBP retention in the early screening studies. Two 3.8 cm x 2.5 cm glass column segments were packed with material L; segment 1 contained 13.6 g, and segment 2 contained 14.0 g. The following conditions were employed:

- Bed temperature: 100°C
- Average airflow: 2.0 l/min
- TBP concentration: 6×10^{-5} g/l.

FIGURE 1. Breakthrough History for Methyl Iodide Loading onto 7 wt% Ag Sorbent with TBP/Dodecane Present

FIGURE 2. Breakthrough History for Methyl Iodide Loading onto 7 wt% Ag Material with TBP/Dodecane Present

TABLE 1. Analysis of Iodine Retention of 7% Ag Iodine Sorbent

Sample	Measured Percent CH ₃ I Breakthrough (C/C ₀ × 100)	Calculated from Breakthrough Curves qI/g Ag(a)	Theoretical Maximums, qI/g Ag(b)
Column I			1.2
Section A	100	1.5	
Section B	98	1.4	
Section C	33	1.3	
Section D	7	0.9	
Column II			1.2
Section A	(material H)	<0.2	
Section B	(material H)	0.2	
Section C	100	1.5	
Section D	95	0.97	
Section E	76	0.77	

(a) Uncertainty is ±25% at a 95% confidence level.

(b) Based on the reaction $\text{Ag} + \text{I} \rightarrow \text{AgI}$.

A breakthrough of 0.6% in segment 1 was measured after 0.19 g of TBP was metered to the column. After 0.94 g of TBP was metered to the material, a breakthrough of 54% was measured in the first column segment, at which time the run was terminated. No breakthrough was measured in the second column segment. The breakthrough curve for the first column segment is shown in Figure 4.

Even though breakthrough was not followed to 100%, total capacity of the material can be estimated from the available breakthrough data. Approximated capacity of the material extrapolated to 100% breakthrough is 70 mg/g material, which is greater than material D but considerably less than material H.

FIGURE 3. Breakthrough History for TBP Loading onto Material D (6 x 14 mesh granular). Bed temperature 100°C, airflow 1.9 l/min, TBP concentration 1×10^{-4} g/l.

Laboratory work to develop an alternative analysis technique using lasers has been impeded by equipment problems. The special diode necessary for experimentation has been procured and will be used in the laser for preliminary experiments. The first experiment will examine the interferences by constituents present in the off gas.

PLANS FOR NEXT QUARTER

We will continue experiments with the candidate TBP sorbents in the bench-scale demonstration unit. Commercially available iodine sorbent material will be used in these experiments, which will be run under conditions likely to be found in actual off-gas streams. Experiments are planned using NO_2 in the feed stream to determine the effect of the gas in combination with TBP on iodine retention.

FIGURE 4. Breakthrough History for TBP loading onto Material L. Bed temperature 100°C, airflow 2.0 l/min, TBP concentration 6×10^{-5} g/l.

A task will be started to assess the feasibility of studying the off-gas streams from the Nuclear Waste Vittrification pilot plant at PNL. We feel that a unit can be constructed to demonstrate the selected TBP removal process using these off-gas streams.

The TBP vapor concentration in the bench-scale demonstration unit will be measured using the PA 460 phosphorus analyzer. It is postulated from initial iodine-loading experiments that the TBP concentration may not be as high as calculated and not high enough to have a deleterious effect on the iodine sorbent. The system to generate TBP vapor may have to be modified to more nearly approach the desired TBP vapor concentration (10^{-4} - 10^{-5} g/l).

PROJECT TITLE: STUDY OF IODINE CHEMISTRY IN PROCESS SOLUTIONS

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

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

OBJECTIVE

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

SUMMARY

Studies continued on the use of various adsorption processes to remove iodine species from a solution simulating the excess water stream of a fuels reprocessing plant. Good results continue to be obtained with the MnO_2 -impregnated polymeric adsorbent system; this system was chosen for demonstration on a larger scale. Initial investigations of two other systems were also promising: 1) the polyiodide ion-loaded anion exchange resin system, and 2) the iodine-pretreated polymeric adsorbent system. Additional data were obtained on the elution of iodide ion from an anion exchange resin.

TRIPS AND VISITORS

June 20, 1978 - Bruce Wilson, FCPO/SR, visited PNL to discuss this program.

TECHNICAL PROGRESS

We continued efforts in the development of methods for adsorbing iodine species from 0.01 M HNO_3 , which is the expected composition of the excess water stream of a nuclear fuels reprocessing plant. Initial data were obtained with a polyiodide-ion loaded anion exchange resin system and with an iodine-pretreated polymeric adsorbent system. Additional data were obtained on the MnO_2 -

impregnated polymeric adsorbent system and on the elution of iodide ion from an anion exchange resin.

The topical report written last quarter has been reviewed and cleared for publication. One change resulting from the review process is the report number; in response to a request for sequential report number, the number was changed to PNL-2080-9.

MnO_2 -Impregnated Polymeric Adsorbents

Last quarter we reported that a lower concentration of iodine in the feed to an MnO_2 -impregnated polymeric adsorbent column results in a lower DF but in a higher effective throughput capacity; this was verified in continued testing. Also, we found that results obtained in stainless steel equipment were comparable to those obtained in glass equipment.

With 1×10^{-4} M I_2 in 0.01 M HNO_3 as feed and an adsorbent column prepared by reaction of Amberlite XAD-4 with 0.02 M KMnO_4 for two 1-hr periods, an iodine DF of more than 15 was obtained until ~3500 column volumes (CV) of feed had been processed. This is the highest throughput capacity yet achieved with this type of adsorbent. For a brief period near the end of this experiment (from about 7000 to about 8000 CV), the iodine DF was about 100 instead of the normal 15 to 30.

Another experiment was then done under similar conditions except that the adsorbent impregnation operation used 0.005 M KMnO_4 solution instead of 0.02 M. In this case, the effective throughput capacity was only ~4000 CV and the iodine DF remained near 15 throughout the experiment.

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The MnO_2 -impregnated polymeric adsorbent system was chosen for initial demonstration on a larger scale. This system has been more fully demonstrated than any of the others and appears to be better than most (if not all) of those investigated.

Polyiodide Ion-Loaded Anion Exchange Resin

In the last quarterly report we presented results on the use of an inactive iodine-loaded polymeric adsorbent to remove radioiodine species from a simulated excess water stream, presumably by an isotopic exchange mechanism. This quarter we have examined the use of an inactive polyiodide ion-loaded anion exchange resin for the same application. Polyiodide ions (I_3^- , I_5^- , etc.) are more strongly retained than is the simple iodide ion (I^-) and thus should allow a longer period of operation before bed regeneration or discard is necessary. We also hoped that the polyiodide ion-anion exchange resin system would be more predictable and reproducible than the iodine-polymeric adsorbent system studied earlier.

This approach has indeed proven capable of long-term operation without regeneration. In the longest test to date, nearly 10,000 column volumes (CV) of 0.01 M HNO_3 , containing 5×10^{-7} to 2×10^{-6} M added I_2 , have been processed through a polyiodide ion pre-loaded anion resin over a three-month period. The radioiodine DF was initially greater than 100 but had dropped to 10 by the time 700 CV of feed had been processed. The DF improved again on continued operation, however, and has been in the range of 20 to 40 during most of the operation. These data were obtained at a flow rate of about 9 CV/hr (except that feed was not run over weekends).

In the preloading step, 1.0 mmol of total I was loaded onto a 5-ml bed of anion exchange resin (Amberlite IRA-900) from a solution prepared by dissolving 0.002 M I_2 in 0.002 M KI in 2×10^{-4} M HNO_3 . Before beginning the flow of simulated excess water, the bed was washed with 200 CV of 1 M HNO_3 to wash off the easily removable iodine species; 14% of the total I was removed by this wash.

This promising result prompted another test under somewhat different conditions. This test is being run in a stainless steel apparatus (instead of glass). It uses a higher loading of polyiodide ion than the first test; 5.4 mmol of total I was loaded onto 5 ml of resin from a solution prepared as before, and 36% was subsequently removed by washing with 1800 CV of 0.01 M HNO_3 . So far, about

2000 CV of 0.01 M HNO_3 containing 1×10^{-5} M I_2 have been processed through this column. Radioiodine DFs have ranged from 18 to 30 at a flow rate of about 9 CV/hr; they appear to be slightly lower at 15 to 22 CV/hr.

Iodine-Pretreated Polymeric Adsorbent

Earlier work¹ has shown that untreated polymeric adsorbents can reduce elemental iodine to iodide ion, thus limiting the iodine DF because the iodide ion is not retained by the adsorbent. This effect can be largely overcome by adding hydrogen peroxide to the feed, but the H_2O_2 concentration required for high DF at high flow is quite large. We earlier² attempted to eliminate this reducing action by pretreating the polymeric adsorbent with KMnO_4 solution; the early promise indicated for this approach was not substantiated by more extensive testing, however.

We have now obtained some promising results by loading the polymeric adsorbent with nonradioactive elemental iodine, then eluting the iodine off the adsorbent and washing with 0.01 M HNO_3 before processing a simulated excess water stream (0.01 M HNO_3 , containing radioactive iodine species). In one such test, a radioiodine DF larger than 100 was maintained for about 800 CV at a flow rate of about 5 CV/hr, with a feed solution containing 2×10^{-6} M I_2 and 0.03 to 0.1 M H_2O_2 . This DF is about 30-fold greater than that previously measured with untreated polymeric adsorbent under similar conditions (0.1 M H_2O_2). When H_2O_2 feed addition to the pretreated column was ceased (after about 800 CV), the DF gradually decreased and reached a value of about 10 after an additional 700 CV had been processed.

In another test of the iodine-pretreatment approach, the DF exhibited a gradual decrease from the initial value of ~100 to a value of ~20 while processing 1200 CV of 2×10^{-6} M I_2 in 0.01 M HNO_3 + 0.03 M H_2O_2 at a rate of 9 CV/hr. Obtaining such high DFs at such high flow rates and such low H_2O_2 concentrations is indeed a vast improvement over the behavior observed with untreated polymeric adsorbents.

Anion Exchange Resin and Iodide Ion

In earlier testing of this system,³ we added 0.1 M hydroxylamine nitrate (HAN) to the 1 M HNO_3 elution solution to prevent oxidation of I^- to I_2 . We have now found that 0.01 M HAN is sufficient to prevent this oxidation but that oxidation does occur when eluting with only 0.001 M HAN in the 1 M HNO_3 . Oxidation to I_2 is potentially undesirable, because the I_2

cannot be eluted without alternating NaOH and HNO₃ solutions.

PLANS FOR NEXT QUARTER

We will continue small-scale evaluation of various sorption processes and initiate

larger scale testing of the MnO₂-impregnated polymeric adsorbent system. In addition, we will select solvent extraction and volatilization methods of iodine removal for initial study.

PROJECT TITLE: HIGH-LEVEL WASTE TREATMENT

PROJECT MANAGER: J. H. Jarrett, Nuclear Process Technology and Analysis Section,
Chemical Technology Department

PRINCIPAL INVESTIGATORS: W. F. Bonner and A. A. Garrett
Nuclear Waste Process Development Section,
Chemical Technology Department
O. F. Hill, Nuclear Process Technology and Analysis Section,
Chemical Technology Department

OBJECTIVE

The objective of this project is to develop process design information for the calcination and vitrification of high-level liquid waste from the reprocessing of irradiated thorium fuel.

cessing thorium fuels. They also toured experimental facilities.

June 20, 1978 - Bruce Wilson, Fuel Cycle Program Office, SR, toured experimental facilities supporting calcination/vitrification programs.

SUMMARY

- Analytical data on fluoride confirm the previously reported low volatility on calcining Thorex waste solutions. The observed 5 to 10% volatilization is consistent with results reported by the French.
- Corrosion measurements on candidate materials of construction for the calciner continue to be low.
- The reference composition for Thorex high-level waste has been adjusted to be consistent with the flowsheets developed at Savannah River. Preliminary glass compositions have been developed for definition of frits to be used in spray calciner/in-can melter studies.

TECHNICAL PROGRESS

Preparation for spray calciner/in-can melter runs in pilot plant and laboratory scale units continued. Supporting work included definition of the reference Thorex high-level liquid waste solution, development of analytical methods for fluoride in calcine and glass, corrosion studies, and glass formulation investigations. Results of a 50-hr run in the laboratory spray calciner are reported.

Reference Thorex Waste Composition

The Savannah River Laboratory recommended modifications to the projected Thorex waste compositions, incorporating recent flowsheet changes. Compared to the former compositions, iron, fluoride, and zirconium are higher in the reference waste; aluminum and potassium are lower. Process conditions effecting these changes include:

- use of Fe(II) to reject plutonium
- a higher concentration of fluoride during dissolving, and a lower thorium end concentration during dissolving than was assumed earlier

TRIPS AND VISITORS

May 17-18, 1978 - R. G. Baxter, R. A. Clark, J. R. Fowler, L. L. Kilpatrick, J. B. Mellen, and L. W. Patrick of E. I. duPont de Nemours and Company visited PNL to discuss calcination/vitrification technology supporting the preparation of a preliminary technical data summary for the waste portion of a plant design for pro-

- use of zirconium nitrate instead of aluminum nitrate to complex and control fluoride activity
- use of HF as the source of fluoride, rather than KF, as was assumed earlier.

The waste volume for the revised reference waste is 700 $\pm$ per metric ton of heavy metal ($\pm$ /MTHM), compared to 378 $\pm$ /MTHM (100 gal/MTHM), which we assumed earlier.

Table 2 presents a new reference composition for Thorex waste (TW-3). Table 3 summarizes the differences between this composition and the earlier one (TW-2).

TABLE 2. Reference Composition of Thorex Waste, TW-3

Constituent	Molarity at 700 $\pm$ /MTHM	kg Oxide ^(a) /MTHM
HNO ₃	2	
Na	0.0054	0.12
Fe	0.41	23
Cr	0.011	0.58
Ni	0.0049	0.25
PO ₄	0.054	2.7
F	0.32	4.3
Al	0.0014	0.005
Zr	0.17	15
		46
Rb	0.011	0.74
Sr	0.022	1.6
Y	0.011	0.90
Zr	0.067	5.7
Mo	0.049	4.9
Tc	0.014	1.2
Ru	0.022	2.0
Rh	0.0022	0.19
Pd	0.0054	0.46
Te	0.0070	0.79
Cs	0.038	3.8
Ba	0.014	1.5
La	0.012	1.4
Ce	0.026	3.1
Pr	0.013	1.5
Nd	0.035	4.1
Pm	0.0005	0.065
Sm	0.0061	0.73
Eu	0.0011	0.13
Gd ^(b)	0.046	5.8
		41
Th	0.054	10
U	0.0054	1.1
Np	0.0002	0.041
Pu	0.0032	0.62
		12
Total		99

(a) Except as otherwise noted.

(b) Includes Gd added as nuclear poison.

Spray Calciner/In-Can Melter Studies

We did not perform additional runs under the TFCT program during this reporting period. The remote laboratory-scale spray calciner and in-can melter system is being installed in the engineering development laboratory with a condenser and scrubber off-gas system. Improved capability to sample and analyze for fluoride volatility is expected.

Analytical data are complete on calcines produced last quarter in the 50-hr laboratory-scale spray calciner, using TW-2 waste. Fluoride retention was 96%, based on filter blow-back calcine sample, or 90.4%, based on a composite calcine sample. Analysis of spray chamber samples (off-gas) is not complete. These retention results are consistent with the off-gas analyses of the shorter run reported last quarter. These data are also consistent with the 5 to 10% volatilization reported by the French.*

The feed for the 50-hr run contained ruthenium. (Study of its behavior was funded under another program.) Measurable (detectable) quantities of ruthenium were not found in samples from the off-gas system; however, analysis of the composite calcine sample showed only 78.5% of the ruthenium. Blackish flakes isolated from some calcine samples analyzed 5.76 wt% Ru. These data suggest the possibility that Ru plates out in the system prior to reaching the off-gas system.

Corrosion Studies

Corrosion during the above 50-hr run was measured on coupons of several candidate materials of construction. Based on coupon weight, corrosion was small, as shown in Table 4.

Filter coupons of 316 sintered stainless steel gained weight, probably because of calcine buildup. However, metallographic examination of these coupons by scanning electron microscopy and by X-ray fluorescence indicated negligible corrosion.

Glass Formulation

Preliminary glass compositions have been developed for Thorex wastes. Table 5 shows the compositions proposed for spray calciner/in-can melter runs for both the pilot-scale (TW-2) and laboratory-scale (TW-3) units.

The TW-3 composition is subject to change, depending upon the impact of the high iron oxide content. Elimination of potassium from the waste may also have a small impact on the constituents added for the glass. We will

TABLE 3. Significant Differences Between Waste Compositions TW-2 and TW-3
(Bases: One Metric Ton of Heavy Metal)

Component	TW-2 (378 μ)		TW-3 (700 μ)	
	Molarity	kg/Oxide MTHM(a)	Molarity	kg/Oxide MTHM(a)
F	0.18	1.3	0.32	4.3
Al	0.36	6.9	0.0014	0.05
Zr ^(b)	0.42	19.7	0.24	20.5
Fe	0.18	5.4	0.41	23
K	0.18	3.2	--	--

(a) Except for fluoride.

(b) Including fission product zirconium.

TABLE 4. Corrosion Rates of Candidate Material During 50-hr Calcination Run

Material	Rate, mils/month	
	Feed Tank	Barrel
Stainless steel	304	--
	304L	0.01
	310	0.01
	316	0.01
	316L	0.02
Inconel	321	0.02
	601	--
	690	--

Fluoride Analytical Development

A procedure to analyze fluoride in glass involves a sodium hydroxide fusion followed by steam distillation from a sulfuric-phosphoric acid solution at 185°C. Determination of the fluoride in the distillate is by spectrophotometric measurement at 620 m μ using the lanthanum-alizarin complexone reagent. With this procedure the fluoride recovery from two samples of an NBS opal glass standard (number 91) was 89 and 90%.

PLANS FOR NEXT QUARTER

A pilot plant-scale spray calciner/in-can melter run is scheduled for August. Feed composition will be TW-2. We are reluctant to use the TW-3 composition with its higher fluoride content and concentration until we have gained experience on fluoride volatility and corrosion effects in the laboratory-scale unit. A laboratory-scale run with TW-3 composition is planned for September.

Additional studies are planned on glass formulations. Objectives are to improve leaching and to increase waste oxide loading.

Analytical development for fluoride should be complete, dependent upon receipt of a new spectrophotometer to replace a failed and unreliable unit.

TABLE 5. Preliminary Glass Compositions for Thorex Waste Studies

Constituent	Wt% in Glass	
	TW-2	TW-3
Calcine	20.0	20.0
Al ₂ O ₃	--	1.7
SiO ₂	42.9	41.9
B ₂ O ₃	14.7	14.4
Na ₂ O	8.8	8.6
CaO	3.3	3.2
BaO	3.3	3.2
ZnO	2.5	2.5
K ₂ O	2.5	2.5
Li ₂ O	2.0	2.0

attempt to further refine the glass to lower the leach rate and possibly raise the waste loading.

PROJECT TITLE: ELECTROPOLISHING TO DECONTAMINATE METALLIC WASTE
FROM ALTERNATE AND THORIUM CONVERTER FUEL CYCLES

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

PRINCIPAL INVESTIGATORS: R. P. Allen, H. W. Arrowsmith, Metallurgy Research Section,
Materials Department
D. K. Davis, Nuclear Process Technology and Analysis Section,
Chemical Technology Department

OBJECTIVE

The objective of this work is to develop and provide the concepts, information and engineering data needed by the Alternate and Thorium Fuel Cycle Technologies Programs to design and operate electropolishing facilities that will decontaminate surface-contaminated solid metallic waste. Major program tasks include:

- the identification and transfer of compatible technology from the commercial electropolishing and related surface finishing industries
- the development of solution treatment procedures to extend electrolyte life and minimize secondary waste volume
- the development of techniques to minimize or remove contamination entrained in cut edges by component sectioning operations
- the adaptation and optimization of electrochemical decontamination techniques and secondary waste minimization approaches for thorium converter fuel cycle metallic waste, with thorium and ^{233}U as the principal contaminants.

SUMMARY

- A public information meeting on electropolishing as a decontamination technique, which included several presentations, a film, and a tour of the electropolishing facilities, attracted 158 registrants.

- Scanning electron microscope studies have shown the effectiveness of electropolishing in prepolishing applications, where it greatly facilitates the cleaning of metal surfaces using swab and other standard decontamination techniques.
- A plasma arc cutting test system has been installed for use in developing optimum techniques for sectioning contaminated metal waste while minimizing the thickness of the recast layer on the cut edges.
- A commercial acid purification unit, which includes an ion exchange bed, acid and water metering tanks, and a control system, is being evaluated for use in electrolyte purification.
- Solution analysis studies are underway to determine the metal and acid contents of the waste and product solutions of the acid purification system. Several on-line methods for determining the phosphate content of sample solutions are being considered.

TRIPS AND VISITORS

May 23, 1978 - H. W. Arrowsmith visited the Rocky Flats Plant, CO, to discuss program progress with C. Wickland and E. Childs of the Rockwell Corporation.

June 12-16, 1978 - D. K. Davis visited Hydrite Chemical Company, Milwaukee, WI. Discussion topics included equipment and procedures for analysis of the phosphoric acid electrolytes, thin film evaporator

systems for concentration of solutions, and design of the solution handling and storage system for our solution processing studies.

PUBLIC INFORMATION MEETING

A public information meeting on electropolishing as a method for removing radioactive contamination from metal surfaces was held in Richland on April 18-19. The meeting attracted 158 registrants, including representatives from other DOE sites, utility companies, commercial electropolishing firms, manufacturers of related equipment, companies providing decontamination services, and other groups or firms with an interest in this decontamination technology. A tour of the electropolishing facilities, shown in Figure 5, and a group meeting also were well

attended. Substantial interest was expressed in the results of the SR-sponsored solution treatment studies.

Topics discussed at this information meeting included an in-depth review of electropolishing as a decontamination technique, examples of electropolishing decontamination projects at Hanford, and plans for development of the technology. Representatives of government and industry also discussed commercial electropolishing technology, decontamination needs, and potential applications of electropolishing in the nuclear industry. The PNL presentation included an 18-min film summarizing the progress of the decontamination program. A 60-page report was distributed, providing a comprehensive review and explanation of the technology.

FIGURE 5. Electropolishing Meeting Attendees Touring the Electropolishing Facilities at the Hanford 231-Z Building. Some large items decontaminated by electropolishing are displayed in the foreground.

TECHNICAL PROGRESS

The following sections describe the progress during this past quarter toward meeting our program objectives in the areas of technology transfer, solution treatment studies, and entrained contamination removal studies.

Technology Transfer

Electropolishing is a well-established industrial process widely used to clean, deburr, and surface finish a variety of metals and alloys. One of our major program objectives is to utilize existing commercial electropolishing, electroplating, and surface-finishing technology and equipment wherever possible in the design, construction, and operation of electrochemical decontamination systems.

Experience has shown that the smooth surfaces produced by electropolishing are much easier to clean than metal surfaces having a normal, as-received finish when swab and other standard decontamination techniques are used. We have conducted scanning electron micrograph (SEM) studies of the surface of an as-received stainless steel bar, shown in Figure 6, which reveals numerous microscopic imperfections capable of entrapping contamination. Figure 7 shows an SEM of a stainless steel surface after it had been electropolished for 15 min. About $13\text{ }\mu\text{m}$ of surface was removed, leaving a smooth surface free of microscopic imperfections. Figures 8 and 9 compare the profilometer readings of the two surfaces, showing how the numerous ridges and valleys of the as-received surface have been removed by electropolishing. A mechanically finished surface, although flat and easier to clean than the as-received surface, is still significantly rougher and more apt to entrap contamination than the electropolished surface, shown in Figure 10. It should be noted that the cost of electropolishing is only about one-tenth that of mechanical finishing.

The results of these studies suggest that all metallic components that will be used in a radioactive environment should be electropolished first, either before or after fabrication. Not only will the component be easier to clean using conventional wiping techniques, but if the surface is damaged in use, it can be reelectropolished to restore the initial surface conditions. Further studies are in progress to better characterize various electropolished surfaces and their response to conventional decontamination techniques.

25 μm

FIGURE 6. Scanning Electron Micrograph of a Representative As-Received Stainless Steel Surface

25 μm

FIGURE 7. Scanning Electron Micrograph of a Stainless Steel Surface Electropolished 15 min in H_3PO_4

FIGURE 8. Profilometer Trace of an As-Received Stainless Steel Surface

FIGURE 9. Profilometer Trace of Stainless Steel Surface after 15 min of Electropolishing

FIGURE 10. Scanning Electron Micrographs Comparing a (A) Mechanically Finished Stainless Steel Surface with (B) a Much Smoother Electropolished Surface

Entrained Contamination

A plasma arc cutting test system has been installed which includes a machine carriage to move the torch at controlled speeds and a water table to contain the dross. This system, shown in Figure 11, will be used to develop optimum techniques for sectioning contaminated metal waste while minimizing

the thickness of the recast layer on the cut edges. We have begun to characterize cut surfaces and to establish operating parameters to optimize cutting conditions. Further work will include adding a water muffler to further contain contamination and an electronic height sensor system to control the height of the torch, and modifying the torch to reduce the width of the cut.

Solution Treatment Studies

A commercial acid purification system, shown in Figure 12, has been acquired to evaluate acid adsorption as an electrolyte purification technique. We are evaluating the ability of acid adsorption to remove radioactive contamination, as well as dissolved metals, and to recycle the waste stream for maximum acid recovery.

In the acid adsorption purification process, spent electrolyte is forced through a column containing a suitable ion exchange resin. The phosphoric acid is preferentially adsorbed (retarded), allowing the dissolved impurities to pass through the column. The purified acid is then recovered, using water as the elutant.

The three main parts of the acid purification unit are: 1) the ion exchange bed, 2) the acid and water metering tanks, and 3) the control system for the unit. The ion exchange bed consists of two large spools each having a cylindrical void (6 in. dia by 12 in. long) in the center. These spools contain a very fine ion exchange resin. Watch glasses are located on the top and bottom of the bed so that the color and flow of the acid and water can be monitored during processing to determine the effect of the various parameters on the unit's operation. The acid and water metering tanks hold the acid to be processed and the water to be used to elute the acid from the resin. The volume of the solutions in the waste and product streams is governed by the position of the electrodes in the metering tanks. The control system automatically sequences the unit through the adsorption and elution operating stages.

This unit is currently being tested using feed streams composed of varying concentrations of metals and acids. The product, waste, acid void, and water void volumes are changed for various runs. A computer program of the purification process has been developed to assist in identifying optimum operating procedures to minimize secondary waste.

Waste and product solutions are being analyzed to determine the metal and acid

FIGURE 11. A Plasma Arc Cutting Test System for Optimizing Sectioning Procedures for Contaminated Metals

content of solutions in the acid purification system. Measurements of specific gravity, electrical conductivity, transmittance, and pH were made and evaluated. Since specific gravity is a linear function of both the metal and acid contents of the solution, it can be calculated using the equation: Specific gravity =

$$1.00085 + 0.000488H + 0.001577M,$$

where

- H • acid content in g/l, and
- M • metal content in g/l.

Electrical conductivity is not as useful a measurement because there are so many concentrations reflected in only a small range of conductivities. Transmittance is determined with a colorimeter using a wavelength of 610 nm. According to Beer's Law, the metal content of the solution is proportional

to the absorbance of the solution, which is defined as the log of the transmittance in percent divided into 100. Because the acid content does not tend to affect the transmittance of the solution, this could be a possible method for determining the metal content. The pH, even for diluted solutions, is affected by the metal content. The pH measurement gives the correct hydrogen content but not the correct phosphate content.

Several on-line methods for determining the phosphate content of sample solutions are being considered. In a modified acid base titration method, the resin adsorbs the metal ion thereby releasing hydrogen ions to convert the metal phosphate to phosphoric acid. Another method uses a liquid chromatograph unit with a resin column to separate the various species found in the solution. A special detector then permits the amount of each material in the solution to be determined.

FIGURE 12. A Commercial Acid Purification Unit to Evaluate Reciprocating Acid Adsorption as an Electrolyte Purification Technique

PLAN FOR NEXT QUARTER

Testing will continue during the next quarter using noncontaminated electrolytes, followed by testing of contaminated solution, to determine the usefulness of the acid adsorption purification system in our electrolyte recovery studies. Other efforts will

include additional testing of the plasma arc cutting test system following installation of a water muffler and a torch height sensing system. Prepolishing studies will be continued to further characterize various electropolished surfaces and their response to conventional decontamination techniques.^(a)

(a) Additional information on electropolishing as a decontamination technique is available in the PNL Nuclear Waste Management Quarterly Progress reports. The latest issue is PNL-2378-1.

PROJECT TITLE: U.S. SCALE TRANSPORT, DISPERSION,
AND REMOVAL MODEL COMPARISONS

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

PRINCIPAL INVESTIGATORS: W. E. Davis and R. L. Drake
Atmospheric Sciences Department

OBJECTIVE

The primary objective of this study is to provide air concentration assessments from a simulated release tracer, using the PNL U.S. scale model for sources in four regions of the U.S. in 1975. This model simulates the processes of atmospheric transport, diffusion, dry deposition and precipitation scavenging. Model simulations are used to assess the effects of no deposition or combinations of dry and wet deposition.

The secondary objective of this work is to estimate the confidence limits for radionuclide surface air concentrations and deposition amounts computed by U.S. scale assessment models. In meeting this objective, we use parallel single and multilayer models that incorporate state-of-the-art modifications of the basic parameters in short-term comparison tests.

SUMMARY

- Gridded mixing heights were prepared for the NE assessment.
- The climatological mixing heights were reevaluated.
- An assessment for Brookhaven, NY has been made, using a maximum of 3 of three grid elements for 1975.
- Removal using mean precipitation rates and hourly precipitation rates were compared for January 1975.

TRIPS AND VISITORS

June 21, 1973 - Pat Sellers of FCP0/SR and Jim Schubert of SRL visited PNL to discuss the AFTC Program.

TECHNICAL PROGRESS

Gridded Mixing Heights

Monthly values of mixing height, based on Holzworth's 1964 data, were interpolated to a 1° grid covering the Northeast. A series of runs was made and compared to Holzworth's analysis. A problem in the gridded values was found due to the lack of data over the ocean. After adding a series of additional "stations" near the eastern coast, lower mixing heights were preserved over the ocean. These "stations" were analyzed, mixing heights at points generated from Holzworth's 1964 analysis, as well as coastal values for Nantucket.

Reevaluation of Mixing Heights

It was necessary to reevaluate the mixing heights after we learned that the 1964 heights of Holzworth's studies were too conservative. Holzworth's findings, in 1972 showed a generally higher mixing height. The main result of incorporating his 1972 mixing heights into the regional scale assessment was a decrease in the air concentrations and deposition. In the case of the Southeast, this could mean a decrease slightly greater than a factor of two in some of the nondepositing air concentrations.

Additional work on the mixing heights will be necessary to identify individual cases of expected higher air concentrations when using long-term climatological averages. These results will be included in the sensitivity studies.

Brookhaven, NY Assessment

An annual assessment was made for Brookhaven, NY, for 1975. A release height of 62 m

was used, with a release rate of 1 Ci/sec. Two alterations were necessary in the PNL computer code to carry out the assessment. First, the code was changed so that gridded mixing height information could be used. Strong gradients in mixing height occur in coastal areas near the source; thus, a single value would render invalid results.

Second, the layer depth over which the winds are averaged was changed. For the three previous assessments, a layer of 300 to 1500 meters was used. Since the depth of the mixed layer is considerably reduced over the northeastern U.S., a reduction in the depth of averaging was necessary. The new layer averages are 100 to 600 meters for October to April, 100 to 700 meters for May

to June, 100 to 800 meters for July to August, and 100 to 700 meters for September. The result of this assessment with the two alterations is shown in Figures 13 through 17.

Comparison of Hourly Versus Average Monthly Precipitation

Previous short-term comparisons of hourly versus 15-day average precipitation rates have shown less deposition using the hourly values. We felt that this comparison was too short to conclude that hourly precipitation would cause lower depositions. Therefore, a comparison over a month was made with 1/2° gridded January 1975 hourly data and 1/2° gridded mean January 1975 data. A further comparison was made using 5° gridded climatological data;

FIGURE 13. The Air Concentration for a Simulated Release from Brookhaven, NY, for 1975. Concentrations are in Ci/m³.

FIGURE 14. The Air Concentration for a Simulated Release from Brookhaven, NY, for 1975 with a Dry Deposition Velocity of 1 cm/sec. Concentrations are in Ci/m^3 .

the result is shown in Table 6. Even with the use of monthly data, the previous lower deposition rate was confirmed with the hourly precipitation, where less than the average monthly precipitation was deposited. The 5° data provided different results than the mean monthly values because: 1) this data is a climatological mean, where the mean monthly values are for only a single month, and 2) the mean monthly values provide data only over land, whereas the climatological values extend over water, providing additional removal over ocean areas.

The results using the PNL model are now being compared with results from the ARL model to evaluate the difference in the way the two models compute wet removal.

TABLE 6. Effects of Precipitation Averaging

Type of Precipitation	Percent Deposited
1. Climatological 5° grid	39.3
2. Hourly $1/2^\circ$ grid	52.9
3. Monthly $1/2^\circ$ grid	73.7

FIGURE 15. The Air Concentration for a Simulated Release from Brookhaven, NY, for 1975 with a Dry and Wet Deposition Using Hourly Precipitation. Concentrations are in Ci/m^3 .

PLANS FOR NEXT QUARTER

- Finish comparison of 1964 versus 1972 Holzworth data.
- Complete user's guide for PNL model.
- Determine surface temperature effects in HTO scavenging.

FIGURE 16. The Total Ground Deposition for a Simulated Release from Brookhaven, NY, for 1975 with a Dry Deposition Velocity of 1 cm/sec. Deposition is in Ci/m².

FIGURE 17. The Total Ground Deposition for a Simulated Release from Brookhaven, NY, for 1975 Using Both Dry and Wet Deposition. Deposition is in Ci/m².

PROJECT TITLE: SAFETY CRITICALITY EXPERIMENTS

PROJECT MANAGER: E. D. Clayton, Criticality Analysis,
Energy Systems Department

PRINCIPAL INVESTIGATOR: R. C. Lloyd, Criticality Analysis,
Energy Systems Department

OBJECTIVE

The objective of the criticality safety support task is to:

- utilize the PNL Critical Mass Laboratory to provide confirmatory criticality data required to support Alternative Fuel Cycle R&D
- establish benchmark data for code validation and safety assessment
- provide criticality safety parameters to support design concepts and equipment specifications.

SUMMARY

- Calculation analysis was completed on the series of experiments with 4.3 wt% ^{235}U rods in water only and in water containing neutron absorbers.
- Experimental equipment has been installed for use with uranyl nitrate solution and uranium rods.

TRIPS AND VISITORS

June 13, 1978 - Joe Spencer (SRL), J. F. McCrossen (SRL) and W. F. Lawless (FCPO/SR), visited the Critical Mass Laboratory.

TECHNICAL PROGRESS

Calculational analyses have been completed on the series of experiments on lattices of fuel rods immersed in water at each of three different lattice spacings with soluble neutron absorbers.^{7,3} The center-to-center fuel

rod spacings were 22.9 mm, 27.9 mm and 33.0 mm, respectively. The UO_2 pellets of 4.3 wt% ^{235}U -enrichment were housed in stainless steel tubes with a clad OD of 14.4 mm (clad thickness of 0.813 mm). A detailed description of the fuel rods is shown in past quarterly reports.^{7,3}

Figure 18 shows a detailed schematic of the experimental system. Boron, gadolinium, and cadmium were used as neutron absorbers for the 22.9 mm lattice. Boron and gadolinium were used as neutron absorbers in the 27.9 mm and 33.0 mm lattices. The results of all the experimental data are summarized in Table 7.

For each of the critical system experimental determinations, k_{eff} values were computed to check how standard calculational techniques agree with experimental results. Values for k_{eff} were calculated using the KENO IV⁹ computer code. The calculations were made using 17 epithermal broad-group cross-sections, with the EGGNIT code,¹⁰ and FLANGE¹¹-ETOG¹²-processed ENDF/B-4 data. The single thermal group was generated using the THERMOS code¹³ and ENDF/B-3 library. All rod systems were modeled in a "smeared rod" geometry; that is, cell-averaged cross-sections were generated for fuel rods in water, and these cross-sections in turn were used across the fuel region in the model. A 1/4-in. Lexan top lattice plate, which supported fuel during the course of the experiments, was modeled as water while a bottom 5/8-in. stainless steel lattice support plate, which the fuel rods rested upon, was modeled exactly. The results of these calculations are also summarized in Table 7. On the average, calculations were 1% high in terms of k_{eff} .

FIGURE 18. Schematic of Lattice Assembly

EXPERIMENTAL PREPARATION

Experimental equipment has been installed to perform the experiments with the lattices of 4.3 wt% ^{235}U rods in uranyl nitrate solution. Operation testing and checkout is now in progress. Preparation of the uranyl nitrate solution is also in progress.

- Begin and complete series of experiments with uranyl nitrate solution and with solution containing gadolinium as a neutron absorber in a rod lattice system.

PLANS FOR NEXT QUARTER

- Complete checkout of experimental system and complete preparation and load-in of the uranyl nitrate solution.

TABLE 7. Effect of Neutron Absorber on Criticality of Heterogeneous Lattice Assembly

Experiment Number	Hexagonal Lattice Pitch (mm)	Number of Fuel Rods Required for Criticality	Neutron Absorber Concentration (g/l)(a)	Total NO ₃ (g/l)	Excess Acid Molarity M	K _{eff} (b,c)
001	22.86	223.3	-0-	--	--	1.010 ± 0.008
002	22.86	289.8	0.2307 B	--	--	1.003 ± 0.007
003	22.86	363.0	0.4514 B	--	--	0.992 ± 0.006
004	22.86	433.2	0.6053 B	--	--	1.003 ± 0.006
006	22.86	282.6	0.4293 Cd	0.722	0.004	1.013 ± 0.006 ^(d)
007	22.86	391.7	1.060 Cd	1.418	0.004	1.021 ± 0.005 ^(d)
009	22.86	283.4	0.0722 Gd	0.230	0.0023	1.005 ± 0.007
010	22.86	349.5	0.145 Gd	0.398	0.0036	0.995 ± 0.008
011	22.86	422.4	0.213 Gd	0.527	0.0044	1.016 ± 0.006
012	27.94	166.6	-0-	--	--	0.997 ± 0.005
013	27.94	225.6	0.158 B	--	--	1.006 ± 0.006
014	27.94	349.9	0.380 B	--	--	1.005 ± 0.006
016	27.94	232.3	0.0547 Gd	0.1927	0.0021	0.994 ± 0.008
017	27.94	341.8	0.1169 Gd	0.2707	0.0021	1.006 ± 0.006
018	33.02	187.4	-0-	--	--	1.024 ± 0.005
019	33.02	234.0	0.0643 B	--	--	1.012 ± 0.005
020	33.02	434	0.2154 B	--	--	0.992 ± 0.005
021	33.02	327.5	0.1507 B	--	--	1.013 ± 0.006
023	33.02	252.7	0.0257 Gd	0.1571	0.00204	1.009 ± 0.007
024	33.02	316.8	0.0440 Gd	0.1805	0.00207	1.016 ± 0.007

(a) B in the form H₃BO₃, Cd in the form Cd(NO₃)₂·4H₂O, Gd in the form Gd(NO₃)₃.

(b) Calculated using EGGNIT/ENDF/B-4 and THERMOS/ENDF-B-3.

(c) The bottom fuel rod end spade was modeled as part of the 304L stainless steel support plate and top spades were modeled as homogenized 304L stainless steel in the water region above the fuel. The top lattice plate, made of Lexan was considered to be water equivalent.

(d) Calculated using EGGNIT/ENDF/B-4 and TEMPEST/ENDF/B-4.

PROJECT TITLE: CRITICALITY RESEARCH IN SUPPORT OF THORIUM
FUEL CYCLE TECHNOLOGY PROGRAM

PROJECT MANAGER: S. R. Bierman, Criticality Analysis,
Energy Systems Department

PRINCIPAL INVESTIGATORS: S. R. Bierman, W. E. Converse, D. E. Friar,
B. F. Gore and R. A. Libby, Criticality Analysis,
Energy Systems Department

OBJECTIVE

The criticality research program is directed toward providing reliable technology for safe and economical criticality controls in chemical processing. The primary goal in FY-1978 is to establish a program plan for obtaining the criticality data needed in the chemical reprocessing of the TFCTP fuels.

SUMMARY

Draft copies of two documents were issued for review and comment. One document identified process steps and the associated criticality data needs in the chemical reprocessing of TFCTP fuels; the other summarized applicable criticality data currently available. Calculations are in progress which will aid in planning criticality experiments and in process design.

TRIPS AND VISITORS

June 13, 1978 - Joe Spencer (SRL), J. F. McCrossen (SRL), and W. F. Lawless (FCPO/SR), visited the Critical Mass Laboratory.

TECHNICAL PROGRESS

A basic process flow diagram, with possible alternative process paths, for the chemical reprocessing of TFCTP fuels was defined for use in identifying the criticality data

required in the TFCTP. A draft document entitled 'Criticality Research in Support of Chemical Reprocessing in the Thorium Fuel Cycle Technology Program - Basic Process Description' was issued for comment. Forms of material, material compositions, and equipment features were defined, to the extent currently possible, for each process step. Also, the criticality data required for each process step was identified. A second document summarizing the criticality data applicable to the TFCTP was also issued for comment.

Calculations in support of experimental design and planning were begun and are about 30% complete. The calculations involve nitrate oxide and metal-water systems of ^{230}U -Th, ^{235}U -Th, ^{239}Pu -Th, ^{233}U - ^{238}Th and ^{235}U - ^{238}U -Th mixtures in slab, cylinder, and spherical geometry. In addition to providing information for experimental planning, the results of these calculations will provide a wide range of data on criticality parameters for use in process design.

PLANS FOR NEXT QUARTER

A detailed program will be developed for providing the criticality data needed in the TFCTP. Experiments will not begin until FY-1979; however, a program plan will be issued identifying proposed experiments along with a proposed time schedule and required funding level.

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