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AFCT/TFCT/ISFS PROGRAM

TECHNICAL PROGRESS REPORT
FOR THE PERIOD
JANUARY 1, 1978 - MARCH 31, 1978

COMPILED BY
O. F. Hill

MASTER

April 1978

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

BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WA 99352

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ABSTRACT

This is the ninth in a series of quarterly progress reports on research and development studies performed by 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 (SRL) under the direction of the Fuel Cycle Program Office (FCPO) of the Savannah River Operations Office (SR) 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
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	Electropolishing to Decontaminate Metallic Waste from Alternate
650.2	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 January, February, and March, 1978. Previous reports in this series are BNWL-2052, BNWL-2080-1, -2, -3, -4, -5, -6, and -7.

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PROJECT TITLE: THORIUM RESOURCE PRICE ANALYSIS

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

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 Savannah River Laboratory 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

- A preliminary supply curve was developed for domestic thorium resources and the high, low, and mean costs were estimated.
- The critical variables which influence production costs were identified for each thorium resource.
- Data on thorium are incomplete; thus, resource and cost estimates are uncertain.

TRIPS AND VISITORS

January 6, 1978

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

February 13, 1978

- Mr. Walt Enderlin traveled to Denver to see Mr. Staats of the USGS. He also contacted the Bureau of Mines concerning their research on domestic thorium resources.

TECHNICAL PROGRESS

This quarter we completed a preliminary analysis of domestic thorium resources and prepared a preliminary supply curve for reactor grade thorium oxide. Figure 1 presents the supply as a block diagram including individual deposits. Figure 2 presents the supply curve representing the high, low, and most likely cost estimate.

FIGURE 1. Supply of Thorium Oxide from Domestic Resources

FIGURE 2. Supply Curves for Thorium Oxide from Domestic Resources

Resource and Cost Estimates

Early in the project a maximum demand (700,000 tons by 2070) for thorium oxide was calculated using a computer model of the nuclear fuel cycle. Domestic resources sufficient to meet this demand (Table 1) were identified and a range of costs was developed for each resource. We characterized the resources with relation to the type, grade, deposit and size of the ore, and the mining, milling and refining method. For each type of resource, we developed conceptual mining, milling and refining processes and estimated the associated capital costs, operating costs, fixed charge on capital, and depletion allowance (using normal industry cost estimating procedures). Detailed cost estimates were developed for mining, milling, and refining costs for stockpiles, thorite deposits (Lemhi Pass, Idaho), carbonatite deposits (Iron Mountain, Colorado), conglomerates and beach

placers. Preliminary cost estimates have been completed for the more costly fluvial placers and Conway granite.

TABLE 1. Domestic Thorium Resources and Production Cost

<u>Deposit</u>	<u>Size in Short Tons of ThO₂</u>	<u>Cost (in \$/lb ThO₂)</u>			<u>Critical Variable</u>
		<u>High</u>	<u>Most Likely</u>	<u>Low</u>	
Nitrate and Metal Stockpiles	1,500	4.0	3.0	2.0	Facility Size
Sluge Stockpiles	2,500	5.0	3.0	2.0	Facility Size
Hall Mountain (Thorite)	3,000	4.0	3.0	2.0	Ore Grade
Wet Mountain (Thorite)	4,500	8.0	6.0	4.0	Ore Grade
Lemhi Pass (Thorite)	250,000	29.0	7.0	2.0	Ore Grade
Iron Mountain (Carbonatite)	33,000	10.0	6.0	2.0	By-product Credit
Mountain Pass (Carbonatite)	28,500	32.0	17.0	2.0	By-product Credit
Conglomerates	48,200	90.0	42.0	16.0	Ore Grade
Beach Placers	16,000	88.0	48.0	9.0	By-product Credit
Fluvial Placers	96,000	138.0	64.0	39.0	By-Product Credit
Conway Granite	<u>250,000</u>	84.0	69.0	46.0	Mining Method
TOTAL	733,200				

For most deposits, the critical variable is ore grade, where the high cost represents the poorest exploitable ore grade, and low cost represents the best ore grade. The most likely cost represents the mean ore grade.

For several deposits the critical variable is facility size, where the high cost estimate represents the small facility production cost, the low cost estimate represents the large facility production cost, and the most likely cost represents a mean production cost. In the case of Conway granite, the most likely production cost is based on average mining and milling conditions.

For resources that occur in conjunction with other valuable materials, the critical variable is the credit given for by-products. The major by-products are currently produced on a relatively small scale. Large scale production of thorium would result in large increases in the amount of marketable by-products, which would tend to reduce the market price. For this study two cases were considered, full current market value for by-products (low cost estimate) and no value for the by-products (high cost estimate). The most likely cost is the mean between no by-product credit and full by-product credit.

Uncertainties in the Estimates

The preliminary supply curve is based on current information and represents a conservative estimate of resource size and ore grade. However, since information on domestic thorium is incomplete, there are many uncertainties surrounding both its quantity and production cost.

Most thorium deposits were discovered in the 1940s and 1950s. In most cases the deposits were not extensively explored, so estimates of deposit size and ore grade are not completely reliable. The uncertainty in domestic thorium resources produces uncertainty in the quantity of thorium available (the horizontal axis of the supply curve). This impacts the preliminary supply curve in several ways. First, currently undiscovered deposits may be identified. Second, known deposits may prove to be larger than those currently estimated. This is particularly true of thorite and conglomerate deposits.

Uncertainties in the production cost estimates (vertical axis of supply curve) may result from several factors. The uncertainty in ore grade can cause a large uncertainty in the production cost estimate in addition to uncertainty in by-product credit. Also the production cost estimates were developed from conceptual flow diagrams and designs for thorium processing facilities. With the exception of beach placers, no domestic thorium deposit has been commercially processed; therefore, the conceptual designs have been based on the results of laboratory testing. Since little research has recently been done on thorium processing, optimum processing procedures probably have not been developed. In addition, a

cost estimate based solely on a laboratory process with no commercial or pilot plant operating experience is extremely uncertain.

PLANS FOR NEXT QUARTER

The supply curve will be updated as new information becomes available and uncertainties are resolved. We will continue the identification of domestic thorium deposits, including new deposits in the supply curve. The detailed analysis of smaller thorite deposits, fluvial placer deposits and Conway granite will be completed. Detailed analysis of the deposits will be updated and refined as new information becomes available. In addition, we will analyze the elasticity of the by-product market so that a more confident estimate of by-product credit can be developed. We will complete a detailed statistical analysis of production cost uncertainty and a topical report on the domestic thorium resources will be prepared in FY 1978.

PROJECT TITLE: INVESTIGATION OF AIR CLEANING PROCESSES FOR
REMOVING TRIBUTYL PHOSPHATE (TBP) FROM OFF-GAS
STREAMS

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

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

OBJECTIVE

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

SUMMARY

- TBP loading of a granular alumina catalyst in a laboratory scale system was terminated when the breakthrough reached 100%. The capacity of the catalyst for retaining TBP was 377 mg TBP/g material at a bed temperature of 100°C and TBP concentrations of 1×10^{-4} to 4.5×10^{-4} g/l.
- A spherical 4 x 8 mesh catalyst material, identified as a candidate material in early experiments, was loaded with TBP in the laboratory scale system. The material showed nearly instantaneous breakthrough of TBP with an ultimate retention of 11 mg TBP/g material at a bed temperature of 100°C and TBP concentration of 1×10^{-4} g/l.
- A second TBP loading experiment under identical conditions was initiated on the spherical catalyst material. In the second experiment, the material was crushed and sieved to 6 x 1 mesh to increase the surface area. Preliminary results indicate little improvement in TBP retention due to the crushing.
- We have started laboratory studies to assess the feasibility of using laser analysis for TBP vapor. We want to determine the possible interfering species present in the off-gas streams and measure their effect upon analysis using a small laboratory laser.

- The laboratory demonstration unit for measuring the effectiveness of TBP sorbents to protect the silver loaded beds is complete and we are conducting initial checkout procedures. The analysis method for methyl iodide in these experiments has been demonstrated to be reliable and accurate.

TRIPS AND VISITORS

March 19-22, 1978	G. B. Parker attended the ANS sponsored topical meeting, "The Back End of the LWR Fuel Cycle," in Savannah, GA.
March 23, 1978	G. B. Parker toured the AGNS fuel reprocessing plant at Barnwell, SC.

TECHNICAL PROGRESS

Loading of TBP onto a granular alumina catalyst material was terminated in early January after a total of 8.96 g of TBP was metered to the material in an air stream flowing at 2 ℓ /min. We maintained the bed at 100°C and inlet TBP concentration ranged from 1×10^{-4} to 4.5×10^{-4} g/ ℓ . The 3.8 cm (1.5 in.) of material packed in a glass column 2.54 cm (1 in.) diameter was loaded with 2.9 g TMP before we observed any measurable breakthrough. The capacity of the granular catalyst to retain TBP was calculated from breakthrough data to be 377 mg TBP/g material.

An experiment in 1977 was run on the same material in an identically packed column using an air flow of 0.38 ℓ /min; all other conditions were the same. In the present experiment, we measured detectable breakthrough after metering 1.8 g of TBP to the material, approximately 1.1 g less than in the run just completed. This indicates that the higher air flow was beneficial in increasing the amount of TBP retained before detectable breakthrough occurred.

We began TBP loading of a 4 x 8 mesh spherical catalyst material in late February. A 3.8 cm (1.5 in.) deep x 2.54 cm (1 in.) diameter bed of material at 100°C was loaded with TBP in air flowing at 2 ℓ /min. The TBP concentration

was 1×10^{-4} g/% throughout the run. Nearly instantaneous breakthrough was measured and 100% breakthrough was achieved after loading 0.45 g TBP to the material. The capacity of the spherical catalyst to retain TBP was 11 mg TBP/g material, which is considerably less than the granular alumina catalyst material tested under identical conditions.

This spherical catalyst was identified in previous experiments as a candidate for further testing but showed poor retention in the test. We subsequently crushed the spherical material to 6 x 14 mesh to increase the surface area for reaction and ultimate TBP retention. We then packed the crushed material in an identical glass column and began a TBP loading experiment under identical conditions. Preliminary results indicate little increase in TBP retention of the crushed form of the material compared to the spherical form of the material.

An assessment of all possible major constituents in off-gas streams containing TBP is underway to determine interferences with a laser analysis method for TBP. We are developing a laboratory experiment to generate a simulated gas stream to examine possible interferences. A helium cooled laser will be used in this experiment. A preliminary feasibility study indicates that selective laser analysis of the low concentration of TBP expected in off-gas streams is achievable using current state-of-the-art instrumentation.

The laboratory demonstration unit to determine the effectiveness of TBP sorbents to protect silver loaded beds is complete. Experimental procedures are being finalized and the initial experiment using the granular alumina catalyst material will begin in April. We have developed methyl iodide analysis procedures using the gas chromatograph and they proved reliable and accurate.

PLANS FOR NEXT QUARTER

We will continue to develop the laser analysis techniques and will start laboratory experiments. Selected sorbent material will be loaded with TBP to identify the best material for the demonstration unit. Also, we will

begin experiments with the granular alumina catalyst material in the demonstration unit, where it will be used as a protective material for the silver sorbent. As other sorbents are identified from long-term TBP loading experiments, they will be similarly tested in the demonstration unit as protective beds.

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

A major effort of the quarter was writing an interim report describing the results of this project. Highlights of the experimental data obtained during the quarter are:

- in the use of MnO_2 -impregnated polymeric adsorbents for removal of iodine species from simulated excess water, a lower concentration of iodine in the feed solution was found to decrease the iodine DF and to increase the effective throughput capacity;
- in the use of an anion exchange resin for removal of elemental iodine from simulated excess water, a lower concentration of iodine in the feed solution gave a lower iodine DF;
- in the use of an inactive iodine-loaded polymeric adsorbent for removal of iodine species from simulated excess water, promising results were obtained in the first cycle but not in the second cycle; and
- in the suppression of iodine volatilization during vaporization of simulated excess water, good suppression was achieved by adding sufficient mercuric ion to the acidic solution or by adding sufficient sodium hydroxide to make the solution basic.

TRIPS AND VISITORS

March 20-22, 1978 J. L. Swanson attended the ANS sponsored topical meeting, "The Back End of the LWR Fuel Cycle," in Savannah, GA.

TECHNICAL PROGRESS

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

We also obtained additional data on the suppression of iodine volatilization during vaporization of simulated excess water. Good volatilization suppression was achieved by adding sufficient mercuric ion to the acidic solution or by adding sufficient sodium hydroxide to make the solution basic.

An interim report describing the results of this project was written and started through review and clearance procedures. It is entitled "Control of Radioiodine in the Excess Water Stream of a Nuclear Fuels Reprocessing Plant" (PNL-2618).

MnO_2 -Impregnated Polymeric Adsorbents

Latest results with the MnO_2 -impregnated polymeric adsorbent system show that the concentration of iodine in the feed solution affects both the iodine DF and the rate of MnO_2 wash-off (which controls the effective throughput capacity). In an experiment using a feed I_2 concentration of $1 \times 10^{-6}\text{M}$, the iodine DF was about 30 until nearly 5000 column volumes of feed had been processed. Earlier experiments with $1 \times 10^{-5}\text{M}$ I_2 in the feed gave iodine DFs of about 100 until 2000 to 3000 column volumes of feed had been processed under similar conditions.

Anion Exchange Resin and Elemental Iodine

We found that the iodine DF obtained when using an anion exchange resin to remove elemental iodine from a simulated excess water stream was lower at a lower concentration of iodine in the feed. The DF values were lower by a factor of about three with $1 \times 10^{-6}\text{M}$ I_2 than with $1 \times 10^{-5}\text{M}$ I_2 in the feed. We further evaluated elution of the sorbed iodine using alternating treatments with NaOH and HNO_3 solutions to define the minimum amount of NaOH required. The minimum amount required for quantitative elution appears to be about 15 millimole NaOH/ml resin.

Inactive Iodine-Loaded Polymeric Adsorbent

An adsorbent prepared by loading inactive elemental iodine on a polymeric adsorbent was found to be effective in the removal of radioiodine species from a simulated excess water stream. Presumably the mechanism of radioiodine removal is simply one of isotopic exchange with the preloaded inactive iodine. The radioiodine DF was affected little, if at all, by flow rate (up to a rate of 10 column volumes/hr, at least) and was the same whether the radioiodine species was elemental iodine or iodide ion. The radioiodine DF was greater than 100 during the processing of the first 700 column volumes of feed ($1 \times 10^{-5}\text{M}$ I_2 or $2 \times 10^{-5}\text{M}$ I^- in 0.01M HNO_3) and was greater than 10 for about 2000 column volumes.

The polymeric adsorbent column was preloaded to an average loading of about 0.85 millimole I_2 /ml adsorbent by flowing a solution containing 0.001M I_2 in 0.01M HNO_3 until all of the adsorbent was dark from the color of the adsorbed iodine. About one-third of this iodine washed off of the adsorbent during the processing of 2400 column volumes of the hot feed.

Following elution of the sorbed iodine with 0.05M NaOH, we put the polymeric adsorbent through a second cycle of inactive iodine loading and subsequent operation with hot feed. Radioiodine DFs were very poor (2 to 5) in this second cycle, which involved an inactive iodine loading only about half that achieved in the first cycle. We do not understand this tremendous drop in DF from the first cycle to the second.

Suppression of Iodine Volatilization

An alternative or supplemental way of decreasing the release of radioiodine with the excess water stream is to suppress the volatilization of iodine when the stream is vaporized for discharge. During the quarter, we measured the volatilization of iodine from acidic solutions containing mercuric ion and from basic solutions. The conditions used in these tests simulated the vaporization of a 0.01M HNO_3 excess water stream in a vaporizer running at a feed solution to bottoms solution flow ratio of 50. The procedure simulated continuous vaporizer operation in that feed solution was added to the boiling bottoms solution in small increments at about the same rate as solution was distilled away. The equipment used and the results of some preliminary scouting experiments were described in a previous report.⁽¹⁾

The results of the tests using mercuric ion in acid solution are presented in Table 2. Entries 1 through 4 of this table simulated the case where $1 \times 10^{-6}\text{M}$ I_2 is present in the excess water stream. Entry 1, in which water containing no iodine was the feed, shows that revolatilization of iodine from the bottoms solution was insignificant when 0.01M Hg^{++} was present. Entries 2 and 3, in which iodine-containing feed was introduced below the surface of the bottoms solution, show only a slight increase in the amount of iodine volatilized. The distillate was decontaminated from iodine by a factor of about 100 (relative to the feed) in these tests. An additional slight increase in the amount of iodine volatilized occurred in Entry 4, where the feed solution was added to the surface of the boiling bottoms solution. This is in contrast to an earlier test⁽¹⁾ where addition of the feed to the surface of the bottoms solution resulted in a large increase in the amount of iodine volatilized. Recent attempts to duplicate the earlier result also indicated only a slight effect of feed addition point; thus, we conclude that the earlier result must have been in error.

Entries 5 through 12 of Table 2 simulate the case where a tenfold greater concentration of iodine ($1 \times 10^{-5}\text{M}$) is present in the excess water stream. Entry 5 shows that 0.01M Hg^{++} is not sufficient to prevent excessive iodine volatilization at this higher iodine concentration. Increasing the Hg^{++} con-

TABLE 2. Suppression of Iodine Volatilization by Mercuric Ion

Entry	Bottoms Composition, <u>M</u>			I/Hg	Feed Composition, <u>M</u>		DF (feed) ^(a)	DF (bottoms) ^(b)
	Hg	I	HNO ₃		I ₂	HNO ₃		
1	0.01	1x10 ⁻⁴	0.5	0.01	--	--	--	3300
2	0.01	1x10 ⁻⁴	0.5	0.01	1x10 ⁻⁶	0.01	100	5300
3	0.01	1x10 ⁻⁴	0.5	0.01	1x10 ⁻⁶	0.01	94	5000
4 ^(c)	0.01	1x10 ⁻⁴	0.5	0.01	1x10 ⁻⁶	0.01	69 ^(c)	3900 ^(c)
5	0.01	1x10 ⁻³	0.5	0.1	--	--	--	520
6 ^(c)	0.01	1x10 ⁻³	0.5	0.1	1x10 ⁻⁵	0.01	9 ^(c)	430 ^(c)
7	0.03	1x10 ⁻³	0.5	0.033	--	--	--	2200
8	0.03	1x10 ⁻³	1.0	0.033	--	--	--	2300
9 ^(c)	0.03	1x10 ⁻³	0.5	0.033	1x10 ⁻⁵	0.01	27 ^(c)	500 ^(c)
10 ^(c)	0.03	1x10 ⁻³	1.0	0.033	1x10 ⁻⁵	0.01	33 ^(c)	1900 ^(c)
11	0.06	1x10 ⁻³	1.0	0.017	--	--	--	3300
12 ^(c)	0.06	1x10 ⁻³	1.0	0.017	1x10 ⁻⁵	0.01	58 ^(c)	3200 ^(c)

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

(b) DF (bottoms) = (concentration of iodine in bottoms)/(concentration of iodine in distillate)

(c) Feed solution added to the surface of the bottoms solution (as opposed to below the surface in the other cases)

concentration to 0.06M did decrease the iodine volatilization but not to a value as low as that obtained at the lower iodine concentration. Feed addition did not affect iodine volatilization under these less efficient retention conditions as much as at the lower iodine concentration.

Results of tests measuring the volatilization of iodine from basic solutions are presented in Table 3. These tests simulated the case where enough NaOH was added to neutralize the input acid and to provide enough excess acid to maintain an NaOH concentration of 0.5M in the bottoms solution. Entries 1 through 5 simulate the case where 1 x 10⁻⁶M I₂ is present in the excess water. The most important result of these

tests is that higher iodine volatilization occurred when the feed solution was made basic than when it was kept acidic until being added to the vaporizer. Presumably, this effect is related to the formation of hypoiodous acid (which is volatile) as an intermediate in the basic feed solution.

Entries 6 and 7 of Table 2 simulate the case where $1 \times 10^{-5} \text{M}$ I_2 is present in the excess water. Revolatilization from the bottoms solution was also insignificant in this case, but more iodine was volatilized when it was present in the feed solution.

TABLE 3. Iodine Volatilization from Basic Solution

Bottoms Composition: 0.05M NaOH + 0.5M NaNO ₃							
Entry	Total Iodine in Bottoms, M	Feed Composition, M				DF (feed) ^(a)	DF (bottoms) ^(b)
		I ₂	HNO ₃	NaOH	NaNO ₃		
1	1x10 ⁻⁴	--	--	--	--	--	9,000
2	1x10 ⁻⁴	1x10 ⁻⁶	0.01	--	--	200	8,600
3 ^(c)	1x10 ⁻⁴	1x10 ⁻⁶	0.01	--	--	120 ^(c)	5,600 ^(c)
4	1x10 ⁻⁴	1x10 ⁻⁶	--	0.001	0.01	32	1,400
5	1x10 ⁻⁴	1x10 ⁻⁶	--	0.001	0.01	57	2,600
6	1x10 ⁻³	--	--	--	--	--	10,000
7 ^(c)	1x10 ⁻³	1x10 ⁻⁵	0.01	--	--	51 ^(c)	3,200 ^(c)

^(a) DF (feed) = (concentration of iodine in feed)/(concentration of iodine in distillate)

^(b) DF (bottoms) = (concentration of iodine in bottoms)/(concentration of iodine in distillate)

^(c) Feed solution added to the surface of the bottoms solution (as opposed to below the surface in the other cases)

PLANS FOR NEXT QUARTER

Continue evaluation of various sorption processes and select one for testing on a larger scale.

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 and J. E. Mendel, Nuclear Process Technology Section and Analysis Section, Chemical Technology Department

OBJECTIVE

The objective of this project is to develop process design information for the conversion of high-level liquid waste from reprocessing thorium fuel to a vitrified waste form.

SUMMARY

Initial runs were made in a laboratory-scale spray calciner using a simulated Thorex waste feed. Fluoride volatility was between 3 and 30%. We believe that the lower number has greater validity, but improved sampling will be required to verify the data. Initial measurements indicated that corrosion in the apparatus is low.

Initial melt development studies with materials simulating Thorex-type wastes indicate that at least 20 wt% refractory oxides characteristic of those in thorex waste are soluble in glass formed at 1050°C. Up to 33 wt% of those oxides appear to be soluble in glass formed at 1150°C.

TRIPS AND VISITORS

January 12-13, 1978	O. F. Hill, J. H. Jarrett, and J. E. Mendel visited the Savannah River Laboratory to participate in budgetary discussions.
January 25, 1978	S. F. Peterson from duPont, SRL, visited PNL to discuss the waste vitrification and related programs and to tour facilities.

February 8, 1978	O. F. Hill attended a meeting at the Operations Office on reporting format and distribution requirements.
February 20-22, 1978	O. F. Hill, J. H. Jarrett, and J. E. Mendel attended an ANS/DOE sponsored meeting in Savannah, GA, on the "Back End of the LWR Fuel Cycle."

TECHNICAL PROGRESS

We completed installation and initiated operation of a laboratory-scale spray calciner. Studies with the unit included measurements of fluoride volatility and corrosion rates. Related investigations included glass melt compositions and methods of fluoride analysis.

Fluoride Volatility

The laboratory-scale spray calciner was operated for 7.9 hr with a simulated Thorex waste feed. The feed composition approximated a diluted solution of TW-2, Simulated, shown in Table 4. Feed fluoride concentration was measured to be 1.82 to 1.91 g/l. Aluminum/fluoride mole ratio was analyzed to be about 2.5, and the average feed rate was 0.8 l/hr.

Analyses of the water discharge from a spray chamber and of a caustic scrubber solution through which an off-gas sample had been passed determined fluoride volatility. A simple schematic flow diagram of the laboratory-scale spray calciner showing sampling points is presented in Figure 3.

Based on the analyses of spray chamber samples, fluoride volatility was calculated to be 3 to 4%; the range was from 0 to 9%. The measured fluoride concentrations in the spray chamber were low (<0.2 ppm) after correcting for a blank of about 0.1 ppm. For efficient operation, the contactor requires a relatively high water flow (3 gpm), compared to an off-gas flow of 2 scfm. Nitrate concentrations in these samples indicated an NO_x flow of 1.7 to 2.7×10^{-2} scfm, and steam flow was estimated at 0.57 scfm. Fluoride concentration before entering the spray chamber was estimated at 4 to 6×10^{-4} g/scf and after the spray chamber, 6×10^{-6} g/scf. The

TABLE 4. Composition of TW-2

Constituent	TW-2, Nominal		TW-2, Simulated	
	Molarity at 378 g/MTHM	Kg Oxide/MTHM	To make 1 liter Combine:	Grams Oxide/g
HNO ₃	2.0		127 ml con. HNO ₃	
Na	0.01	0.117 Na ₂ O	0.85 g NaNO ₃	0.31
Fe	0.18	5.440 Fe ₂ O ₃	88.9 g Fe(NO ₃) ₃ ·9H ₂ O	17.6
Cr	0.02	0.575 Cr ₂ O ₃	8.00 g Cr(NO ₃) ₃ ·9H ₂ O	1.52
Ni	0.009	0.254 NiO	5.53 g Ni(NO ₃) ₂ ·6H ₂ O	1.42
PO ₄	0.10	2.686 P ₂ O ₅	6.81 ml 85% H ₃ PO ₄	7.10
F	0.18	1.294 F	8.64 g KF	3.42
K	0.18	3.209 K ₂ O	as KF, included above	8.48
Al	0.36	6.947 Al ₂ O ₃	135.05 g Al(NO ₃) ₃ ·9H ₂ O	18.4
Zr	0.3	<u>13.992</u> ZrO ₂	113.1 g Zr(NO ₃) ₂ ·2H ₂ O	<u>52.1</u>
		34.5		110.
Rb	0.021	0.743 Rb ₂ O	3.10 g RbNO ₃	1.96
Sr	0.040	1.569 SrO	8.47 g Sr(NO ₃) ₂	4.15
Y	0.021	0.898 Y ₂ O ₃	52.87 g RE nitrate mix	0.04
Zr	0.123	5.737 ZrO ₂	[included above]	---
Mo	0.090	4.904 MoO ₃	15.98 g MoO ₃	15.98
Tc	0.021	1.232 Tc ₂ O ₇	substitute Mo, included above	---
Ru	0.040	2.015 RuO ₂	substitute Fe, included above	---
Rh	0.004	0.192 Rh ₂ O ₃	1.16 g Co(NO ₃) ₃ ·6H ₂ O (a substitute)	0.32
Pd	0.010	0.463 PdO	substitute Ni, included above	---
Te	0.013	0.785 TeO ₂	2.07 g TeO ₂	2.07
Cs	0.071	3.787 Cs ₂ O	13.84 g CsNO ₃	10.0
Ba	0.026	1.509 BaO	6.79 g Ba(NO ₃) ₂	3.98
La	0.023	1.418 La ₂ O ₃	as RE mix, included above	4.94
Ce	0.048	3.127 CeO ₂	as RE mix, included above	9.90
Pr	0.024	1.547 Pr ₆ O ₁₁	as RE mix, included above	1.03
Nd	0.064	4.076 Nd ₂ O ₃	as RE mix, included above	3.50
Pm	0.001	0.065 Pm ₂ O ₃	as RE mix, included above	
Sm	0.011	0.726 Sm ₂ O ₃	as RE mix, included above	0.62
Eu	0.002	0.132 Eu ₂ O ₃	as RE mix, included above	0.16
Gd	0.002	<u>0.137</u> Gd ₂ O ₃	as RE mix, included above	<u>0.41</u>
		35.1		59.
Th	0.1	9.994 ThO ₂	as RE mix, included above	
U	0.01	1.063 U ₃ O ₈	as RE mix, included above	
Np	0.0004	0.041 NpO ₂	as RE mix, included above	
Pu	0.006	<u>0.616</u> PuO ₂	as RE mix, included above	
		<u>11.7</u>		<u>170.</u>
		81.3		

fluoride concentration in the caustic scrub solution was also measured to be less than 0.2 ppm.

FIGURE 3. Alternative Fuel Cycle Waste Program Lab-Scale Calciner Corrosion and Volatility Studies

Based on fluoride concentrations in calcine samples, the overall fluoride loss would have been 30 to 40%. However, if this much fluoride volatility would have occurred, it should have been detected in the water and caustic scrubber samples; therefore, fluoride volatility is believed to be closer to the 3 to 4% estimate.

A summary of calcine location versus measured fluoride concentration is given in Table 5.

TABLE 5. Fluoride Concentration in Calcine

<u>Calcine</u>	<u>Total, g</u>	<u>Wt. F</u>
Filter	526.8	1.17
Barrel	115.0	0.23
Seal Pot	33.1	1.36
Pan	47.7	1.07
Lid and Nozzle	<u>5.0</u>	
	727.6	

Corrosion

During the above run, coupons for corrosion measurement were placed in the feed tank, at the bottom of the calciner, and on the sintered metal filter. These locations are noted in the schematic flow diagram of Figure 3.

Table 6 lists the corrosion rates for this run. An exposure time of 7.9 hr (from feed-on to feed-off) was used for calculations.

TABLE 6. Equipment Corrosion Rates

<u>Metal</u>	<u>Corrosion Rates</u> <u>(mils per month)</u>	
	<u>Feed Tank</u>	<u>Calciner Barrel</u>
304	0.	---
304-L	0.1	0.1
310	0.3	0.2
316	0.0	---
316-L	0.2	0.2
601	---	0.6
690	---	0.6

The weight losses were very small during this short run. These rates are simply a qualitative indication that the corrosion rates are expected to be low throughout the calciner equipment. Rates given for the coupons in the calciner barrel are probably slightly conservative due to a thin oxide layer formation on these coupons which was not removable during cleaning.

The slightly higher Inconel corrosion rates are not yet indicative that nickel-base alloys are less corrosion resistant. These coupons had more cross-grain surface than the stainless steel coupons. This surface is often less corrosion resistant than longitudinal surfaces and may well account for the slightly higher rates. During future tests, all coupons will have identical geometries, and a more precise comparison of Inconels and stainless steels may be made.

Microscopic examination of the 316SS sintered-filter discs indicated that no significant corrosion took place during the 7.9 hr exposure. A net weight gain did occur in all but the coarsest sintered material after cleaning the discs ultrasonically in distilled water and acetone. Cross-sectional mounts of these samples are currently being examined by scanning electron microscopy (SEM) and X-ray fluorescence for elemental analysis to ascertain the chemical composition of a bronze-colored deposition on the sintered material.

Analytical results of a second run of several hours are not yet available. A 50-hr run to study ruthenium behavior was completed at month end; details of the run and analytical data will be reported next quarter.

Thorex Waste Melt Development

One of the characteristics of a calcine produced from high-level waste from the Thorex process is its high refractory oxide content, particularly CeO_2 , ZrO_2 , and ThO_2 . We conducted a series of melts using mixtures of these these oxides^(a) and 76-101 frit;^(b) the levels of the materials in the glasses were equivalent to oxide loadings of 21 to 33 wt%. When melted at 1050°C, only the glass containing the lowest waste loading was free of insolubles. When the glasses were re-melted at 1150°C, all products appeared homogeneous. We will try to determine whether compositional changes in the glass frit would increase this waste loading while still maintaining a lower melting temperature.

^(a) 51.5 wt% ZrO_2 , 11.7 wt% CeO_2 , 36.8 wt% ThO_2 .

^(b) 59.7 wt% SiO_2 , 14.2 wt% B_2O_3 , 11.2 wt% Na_2O , 7.45 wt% ZnO , 3.0 wt% CaO , 4.45 wt% TiO_2 . This frit composition was chosen since other studies have shown it to have good capacity for refractory oxides.

Fluoride Analysis Development

Samples of calcine and glass were used in evaluating the steam distillation procedure. In this procedure, the samples are first fused in sodium carbonate to convert the fluoride to a water soluble form. The fused melt is leached with distilled water and the fluoride distilled from an H_2SO_4 - H_3PO_4 mixture at 160°C . The fluoride in the distillate is determined colorimetrically using "Amadac F," a commercial lanthanum-alizarin complexant dye.

The calcine analyzed at approximately 1% fluoride; the glasses analyzed at approximately 0.2% fluoride. When the samples were spiked with a weighed amount of NaF and fused, spike recovery by the steam distillation procedure was 96% for the calcine and 101 to 108% for the glasses.

We tried a pyrohydrolysis procedure on the calcine sample. Fluoride recovery was 77% of the value obtained by steam distillation.

PLANS FOR NEXT QUARTER

The laboratory-scale spray calciner is being moved to a new location. Improvements are planned in processing and sampling of the off-gas system, and we anticipate increased reliability in the data on fluoride volatility. Nevertheless, existing samples from past runs will be analyzed. Also, corrosion coupons exposed during these runs will be inspected and measured to obtain corrosion data.

Additional data will be obtained on the solubility of refractory oxides in glass formulations. Studies will include variation in frit composition to increase the solubility.

With the assistance of the Savannah River Laboratory, we will continue in our efforts to establish a reference composition for Thorex high-level waste.

An NBS standard of opal glass (SRM No. 91) containing 5.72% F, 67.53% SiO_2 and 6.01% Al_2O_3 has been ordered. This standard, when

received, will be used for checking both the steam distillation and the pyrohydrolysis analytical procedures.

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

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; and
- 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

- Technology transfer activities continued with visits to and from program consultants and manufacturers of electropolishing, surface finishing, and plasma arc torch cutting equipment. Some of the subjects covered in these activities include liquid blasting techniques, barrel electropolishing techniques, vibratory finishing techniques and the solution handling system.

- In entrained contamination studies, tests using a vibratory finisher with a medium-wear-rate ceramic media reduced the thickness of the recast layer created by plasma arc torch sectioning by about one-half.
- Evaluation of membrane technology for purification of spent phosphoric acid electrolyte indicated that this technology would not be applicable to our needs without a major research effort.
- Studies relative to disposing of contaminated electrolyte by absorbing it on a commercial absorbant concluded that the acid-loaded absorbant would require neutralization.
- Neutralization efforts produced a hard solid product with a volume ratio of 3.72 gal product to 1 gal acid. However, the product is water soluble.
- Evaluations of titanium and tantalum as low-erosion-rate rack, basket, and liner materials for decontamination applications indicate that the tantalum may be a better material than titanium for basket- and barrel-type decontamination applications.

TRIPS AND VISITORS

December 19, 1977	- R. P. Allen and H. W. Arrowsmith discussed plasma arc torch cutting technology with W. J. Hance, Linde Division, Union Carbide Corporation, San Francisco, CA.
January 12, 1978	- R. P. Allen and H. W. Arrowsmith discussed mechanization and automation of plasma arc cutting equipment with R. L. Bullard, Advanced Systems Division, Union Carbide Corporation, Indianapolis, IN.
January 13, 1978	- R. P. Allen reviewed technical progress and budgetary status of the AFCT/TFCT program activities with L. H. Meyer, M. D. Boersma and P. H. Permar (SRL), and R. E. Erickson (SRO) at Savannah River.

- January 13, 1978 - H. W. Arrowsmith discussed large vibratory polishing equipment with R. Sermo, Ultra-Matic Equipment Company, Addison, IL.
- January 14, 1978 - H. W. Arrowsmith discussed vibratory finishing equipment with R. J. Hauser and J. K. Deede, Automated Finishing, Inc., West Allis, WI.
- January 14, 1978 - H. W. Arrowsmith discussed wet blasting techniques with A. Lienemann and R. S. Morgan, Vapor Blast Manufacturing Company, Milwaukee, WI.
- January 14, 1978 - H. W. Arrowsmith discussed electropolishing techniques with L. S. Winter and D. Skvara, Hydrite Chemical Company, Milwaukee, WI.
- January 30, 1978 - H. W. Arrowsmith and R. A. Lundgren observed and photographed a flow-through vibratory finisher at Peco Manufacturing Company, Portland, OR.
- January 31, 1978 - H. W. Arrowsmith discussed and photographed a large-scale electropolishing operation at the Coors Brewery, Golden, CO.
- February 1, 1978 - H. W. Arrowsmith discussed and photographed vibratory finishing equipment with Automated Finishing and wet blasting techniques with Vapor Blast Company in Milwaukee, WI.
- February 2, 1978 - H. W. Arrowsmith discussed and photographed automated electropolishing equipment with A. Marelli, National Plastics and Plating Supply Company, Terryville, CT.
- February 3, 1978 - H. W. Arrowsmith discussed and photographed automated barrel plating with the Racine Plated Company, Racine, WI.

- February 22, 1978 - Program consultants L. S. Winter and D. Skvara of Hydrite Chemical Company visited PNL.
- March 29, 1979 - D. G. McIntyre of American Machinery and Engineering Corporation visited PNL to discuss automated equipment for plasma arc torch cutting.

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 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. These technology transfer activities continued during this reporting period with visits to and from program consultants and manufacturers of electropolishing, surface finishing and plasma arc torch cutting equipment. Some of the technology areas covered in these discussions and plant visits included:

- Liquid Blasting Techniques. Liquid blasting techniques may offer important advantages over dry abrasive blasting methods as a pre-treatment approach for the removal of paint, grease, corrosion layers and gross contamination to prepare sectioned material for final decontamination by electropolishing. Arrangements are in progress to obtain a liquid blasting system for evaluation and comparison with dry abrasive techniques with respect to speed, contamination containment, and secondary waste generation.
- Barrel Electropolishing Techniques. Automation concepts used in the commercial electropolishing and electroplating industries will be adapted for electropolishing decontamination applications to facilitate

the processing of large volumes of surface-contaminated metallic waste while minimizing personnel exposure and processing costs. Studies are in progress as part of this effort to develop optimum rack materials and procedures to hold the material to be decontaminated. We are investigating an alternate approach: the use of industrial barrel electropolishing techniques to decontaminate sectioned material without the need to individually rack each part. The barrels rotate to ensure that all areas are uniformly electropolished (decontaminated) and to minimize electrical contact problems. Based on our discussions with operators of commercial barrel lines, we have ordered a small barrel system that will permit evaluation of different liner materials (Ti, Ta, etc.), as well as demonstrate the feasibility of this approach. The combination of an automated barrel-type electropolishing system with a vibratory finisher could provide an efficient and effective system for the pretreatment and decontamination of large volumes of sectioned metallic waste.

- Vibratory Finishing Techniques. During discussions with experts from the metal surface finishing industry, we identified a simple approach that will help minimize the amount of secondary waste generated by the vibratory finishing pretreatment process. A significant fraction of the wear of the ceramic media used in vibratory finishing occurs during the initial break-in period. For example, the wear rate of a typical media is 1.2 wt%/hr during the first two hours and only 0.3 wt%/hr after complete break-in. More than 7% of the total weight loss occurs during the first 8 hr of operation. This initial loss during operation can be eliminated by preprocessing the media in a noncontaminated system. We have made arrangements to procure prerun media for our subsequent vibratory finishing pretreatment studies.

Our discussions with manufacturers of vibratory finishing equipment also helped us identify another possible decontamination application of vibratory finishing. Some industries routinely use vibratory finishing to remove flashings from die-molded plastic and rubber parts. The softer parts are processed at subzero temperatures.

This would suggest that vibratory finishing techniques potentially could be used to decontaminate selected nonmetallic components such as thermoplastic panels from glove boxes.

- Solution Handling System. We met with program consultants from the Hydrite Chemical Company to discuss the design of a solution transfer and storage system for the contaminated electrolyte and rinse solutions. Topics covered included tank design, pumps for phosphoric acid service, safety systems and the possible use of less expensive, lined construction materials like those in the phosphoric acid industry. The use of a multiport valve system rather than a jumper arrangement was suggested for the solution transfer station.

Other technology transfer activities have included evaluation studies of titanium and tantalum as low-erosion-rate rack, basket and barrel liner materials for decontamination applications using the test fixture shown in Figure 4. Although various items were successfully decontaminated using titanium, there were indications of poor electrical contact between the test parts and the surface of the titanium sheet. The titanium was replaced with a tantalum sheet and similar tests were conducted with no evidence of arcing. These results are consistent with earlier test results that led to the selection of tantalum as the contact material for a remote electropolishing system used to decontaminate fission product storage capsules.⁽²⁾ Although the rack that supported the capsules was made of titanium, the tantalum used for the spring contacts that transmitted the electropolishing current to the Hastelloy C and stainless steel capsules successfully reduced arcing problems.

These observations indicate that tantalum may be better material than titanium for basket and barrel-type decontamination applications. However, we encountered a problem in using tantalum sheet as the contact surface for decontaminating mild steel process tube end caps from the Hanford N-Reactor. Decontamination proceeded normally when the rim of

the cap was in contact with the tantalum sheet. Placing the closed end of the caps in contact with the sheet resulted in the formation of a corrosion layer in the contact area that limited the electropolishing current. We should be able to eliminate this problem by using a perforated sheet to provide better access of the agitated electrolyte to the contact area.

FIGURE 4. Basket Test Fixture with a Tantalum Contact Surface. The stainless steel support structure is electrically isolated.

Entrained Contamination Studies

Some of the contamination on metal surfaces will be incorporated in the recast layer that is formed on cut edges by fusion-type metal cutting operations. We hope to minimize the entrained contamination problem by: 1) developing precleaning treatments to remove most of the surface contamination before sectioning; 2) identifying cutting techniques and operating procedures that will minimize the thickness and contamination content of the recast layer; and 3) developing pretreatment and electropolishing techniques for the sectioned material that will preferentially remove all or part of the recast layer.

As part of this effort, tests were conducted using the vibratory finisher to determine if this pretreatment technique would remove at least part of the recast layer from cut edges. The initial studies using plastic media were not encouraging with respect to elimination of the recast material because most of the metal removed by vibratory finishing is from sharp edges and corners. However, subsequent tests using medium-wear-rate ceramic media demonstrated a reduction of more than one-half in the thickness of the recast layer (Figure 5). It should be noted that the recast layer produced by the small dry torch is discontinuous, suggesting the possibility of almost eliminating the recast layer through careful control of the torch operating parameters.

FIGURE 5. Use of Vibratory Finishing to Remove the Recast Layer Produced by Plasma Arc Torch Cutting: As Sectioned Using a 100-A, Dry Plasma Arc Torch (Left) and After Vibratory Finishing for 3 hr (Right)

Solution Treatment Studies

As part of our efforts to develop solution treatment procedures to extend electrolyte life, membrane separation methods have been studied.

These methods for separation of impurities from an aqueous media take advantage of semipermeable ion selective membranes. These separation processes depend on the permeation of the membrane by some of the chemical species in contact with it. Typical membrane processes include:

- dialysis - both cation and anion diffuse from a colloid or waste solution to a "pure" water side of an ion permeable membrane;
- reverse osmosis - pressure-assisted diffusion of water from waste side of a membrane to the "pure" water side;
- Donnan dialysis - application of a cation exchange membrane to effect separation of metal ions from a salt solution to an acid solution; and
- electrodialysis - electrolytic-assisted separation of cations through a cation permeable membrane and anions through an anion permeable membrane.

An evaluation of the applicability of membrane technology for purification of spent phosphoric acid electrolyte was conducted by program consultants from the Hydrite Chemical Company. They noted the following problems: 1) the relevant processes are not developed industrially; 2) high viscosity reduces mobility, throughput rates and efficiency; and 3) the high acid concentration and complexing characteristics of the phosphate ion result in an unfavorable equilibrium situation. We concluded that membrane technology would not be applicable without a major research effort.

Also of interest in our solution treatment studies is development of ways to handle and reduce the volume of spent electrolyte. An initial attempt to dispose of contaminated electrolyte involved absorbing it on a commercial absorbant. The ratio used was about three volumes of absorbant to one volume acid. The material absorbed the acid well, but gas was generated which would make it impossible to package the loaded absorbant for disposal. We therefore concluded that the acid-loaded absorbant would require neutralization prior to packaging for disposal.

There are several agents available to neutralize the loaded absorbant. Solutions of either sodium hydroxide, trisodium phosphate, or sodium carbonate could be used for this process. We conducted several laboratory experiments using absorbant loaded with clean acid in what was assumed to be the proper ratio (0.31 gal acid/gal absorbant). From these experiments, we determined that if two volumes of the loaded absorbant were mixed with one volume of an approximately 30% sodium hydroxide solution, the absorbant would be neutralized. The neutralization produced a large amount of heat, enough to cause boiling. This mixture solidified when cooled to room temperature, which could make it an excellent material for disposal. The solid has a density of about 93 lb/ft³. The volume produced by the neutralization is about 3.72 gal product/gal acid. A major disadvantage of the solid is that the material is water-soluble; the binding material in the solid is a waste-soluble sodium-phosphate compound.

Figure 6 shows the before, during and after phases of the neutralization process. On the right, the beaker has the Quick Sorb® containing phosphoric

FIGURE 6. Various Steps in the Neutralization of Acid-Loaded Absorbant. The product is on the left, the reacting material in the middle, and the load absorbant on the right.

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acid; the beaker in the middle contains the reacting mixture, shown here during the cooling process. On the left is the product of the neutralization process. The thermometer in the solid was accidentally left there overnight and could not be removed the following morning.

This technique was next tried on the contaminated absorbant. Due to the nonuniform distribution of acid in the absorbant, the barrel-size neutralization did not proceed as in the smaller scale test. The neutralization of this material required adjustment in the ratio of sodium hydroxide to absorbant. Each batch acted differently. We even used straight acid to ensure a neutral product in some batches. Due to the larger volumes of material, these batches took much longer to solidify. When neutralized, the material filled 90% of four polyethylene-lined drums. The remaining 10% volume was filled with the commercial absorbant to ensure that there would be no free liquid in the barrels. There were no material balances kept on these neutralizations. The barrels were sealed and cleaned. Presently, they are awaiting transfer to Rockwell Hanford Operations Company for disposal.^(a)

PLANS FOR NEXT QUARTER

One of the major activities during the coming quarter will be the public information meeting on electropolishing as a decontamination technique that will be held in Richland on April 18-19, 1978. The program will include in-depth discussions of the technology transfer, solution treatment and entrained contamination removal studies. Preliminary registration data indicate widespread interest in electropolishing decontamination technology and a good representation from industry as well as other DOE sites and government agencies is expected. The installation and on-site testing of the reciprocating acid adsorption system for purification of the phosphoric acid electrolyte should be completed by mid-May. The technology transfer activities and edge effect studies will continue as presently scheduled.

(a) Additional information on electropolishing as a decontamination technique is available in the PNL Nuclear Waste Management Quarterly. The latest issue is PNL-2377-4.

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, 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 multi-layer models that incorporate state-of-the-art modifications of the basic parameters in short-term comparison tests.

SUMMARY

- An assessment for Carlsbad, NM, has been made using a maximum σ_y of 3 grid elements for 1975.
- An assessment for Morris, IL, has been made using a maximum σ_y of 3 grid elements for 1975.
- Preparations are ready to perform an annual assessment for the NE United States using Brookhaven Laboratory as the source site. Gridded wind fields and gridded hourly precipitation have been collected for 1975.
- Deposition results between the case with the model computing dry deposition with the case of the model computing dry and wet deposition have been compared for the SE assessment.

TRIPS AND VISITORS

January 23, 1978

Dr. Ron Drake attended a meeting at the Savannah River Laboratory to discuss status of the program and finances.

March 29-30, 1978

Dr. David Powell and W. E. Davis attended a meeting discussing the format of the SE assessment report. Also discussed were future plans for research.

TECHNICAL PROGRESS

Carlsbad, NM, Assessment

An air concentration assessment in Carlsbad, NM, has been completed. We used fields and hourly gridded precipitation to make the assessment, and tapes of the results have been sent to SRL for evaluation and study. The model was run in three modes for this assessment: no deposition, dry deposition, and wet and dry deposition. Figures 7 - 11 show air concentrations and cumulative ground deposition on a monthly and annual basis for this assessment.

Morris, IL, Assessment

The third assessment in this series, Morris, IL, has been completed. We are presently producing a tape of the results to be sent to SRL. Gridded wind fields and gridded hourly precipitation tapes were produced for the assessment.

The model was run for the same three modes indicated above. Air concentrations and cumulative ground deposition were produced in both monthly and annual simulations (Figures 12 - 16).

Preparation for NE Assessment

We have started collecting data for our last assessment. Gridded winds and gridded hourly precipitation have been prepared and work is now underway to determine the proper mixing height to be used for modeling. Since the site is near the ocean, strong gradients of mixing layer heights are

FIGURE 7. The Air Concentration for a Simulated Release from Carlsbad, NM, for 1975. Concentrations are in Ci/meter³.

FIGURE 8. The Air Concentrations for a Simulated Release from Carlsbad, NM, for 1975 with a Dry Deposition Velocity of 1 cm/sec. Concentrations are in Ci/meter³.

FIGURE 9. The Air Concentration for a Simulated Release from Carlsbad, NM, for 1975 with a Dry and Wet Deposition Using Hourly Precipitation

FIGURE 10. The Total Ground Deposition for a Simulated Release from Carlsbad, NM, for 1975 with a Dry Deposition Velocity of 1 cm/sec. Deposition is in Ci/meter².

FIGURE 11. The Total Ground Deposition for a Simulated Release from Carlsbad, NM, for 1975 Using Both Dry and Wet Deposition. Deposition is in Ci/meter².

FIGURE 12. The Air Concentrations for a Simulated Release from Morris, IL, for 1975. Concentrations are in Ci/meter³.

FIGURE 13. The Air Concentration for a Simulated Release from Morris, IL, for 1975 with a Dry Deposition Velocity of 1 cm/sec. Concentrations are in Ci/meter³.

FIGURE 14. The Air Concentration for a Simulated Release from Morris, IL, for 1975 with a Dry and Wet Deposition Using Hourly Precipitation

FIGURE 15. The Total Ground Deposition for A Simulated Release from Morris, IL, for 1975 with a Dry Deposition Velocity of 1 cm/sec. Deposition is in Ci/meter³.

FIGURE 16. The Total Ground Deposition for a Simulated Release from Morris, IL, for 1975 Using Both Dry and Wet Deposition. Deposition is in Ci/meter².

Comparison of Deposition: Mode 2 and Mode 3

We compared the results of simulations using Modes 2 and 3 on an annual basis to calculate the cumulative ground deposition. Mode 2 calculates ground deposition based on the use of a dry deposition velocity of 1 cm/sec, while Mode 3 uses the same dry deposition velocity but also includes an estimate of wet removal. In the wet removal estimate, a washout ratio of 5×10^5 is used and a conservative assumption on deposition is made that the air in the mixing layer is subject to incloud scavenging. Figure 17 presents the results of the comparison, showing that increased deposition occurs near the source of Mode 3 over Mode 4. At the source an increase of ~20% occurred in the Mode 3 results over Mode 2 results. To the Northwest over the Appalachians we noted an increase of 80%. This increase correlates quite well with the maximum annual precipitation shown in Figure 18. However, these increases became smaller until the Mode 2 values exceeded those of Mode 3. Further investigation is required of the other assessment results, as well as a comparison of the monthly values, before comments on a general parameterization of the hourly precipitation can be made. However, from these initial results it appears, at least on the annual basis, that the removal code may be simplified to avoid the use of hourly precipitation.

Documentation of the PNL U.S. Scale Assessment Model

This quarter we started to document the PNL model and peripheral programs used for data input. The introduction was drafted and technical discussion sections were completed. The document is presently being edited and coded to aid the potential model user.

PLANS FOR NEXT QUARTER

- Test the incorporation of time and space variation of the mixing height for the NE assessment.
- Finish NE assessment and mail results to SRL.
- Work on incorporation of surface temperatures into estimates of deposition of tritiated water vapor.
- Investigate the effects of vertical motions on surface deposition.

FIGURE 17. The Net Increase in Per Cent of Dry and Wet Deposition over Dry Deposition Alone (Note: Only positive values are shown)

FIGURE 18. The Annual Precipitation for the United States

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 objectives of the criticality safety support task are to:

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

SUMMARY

- The series of criticality experiments to determine the effect of neutron absorbers on water-moderated lattices of 4.3 wt% ^{235}U -enriched fuel rods was completed and the data is being analyzed.
- Survey calculations for the 4.3 wt% ^{235}U rods in uranyl nitrate solution were completed using gadolinium as the neutron absorber.
- Experimental equipment is being installed for the experiments with the 4.3 wt% ^{235}U rods in the uranyl nitrate solution.

TRIPS AND VISITORS

None

TECHNICAL PROGRESS

Criticality experiments were conducted to obtain benchmark data for validating criticality control calculations on the use of soluble neutron absorbers.

The completed experiments on lattices of fuel rods immersed in water include measurements at each of three different lattice spacings with soluble neutron absorbers. The center-to-center fuel rod spacings were 22.9 mm, 27.9 mm and 33.0 mm. These fuel rods contained UO_2 pellets of 4.3 wt% ^{235}U -enrichment in stainless steel tubes, with a clad OD of 14.4 mm (clad thickness of 0.183 mm). A detailed drawing is shown in Figure 19. The measurements on the 33.0 mm lattice include critical experimental data for two different concentrations of gadolinium and three concentrations of boron in the water.

FUEL TUBE CANNED LENGTH*	(34.421 in.)	874.29 mm
FUEL ROD CORE LENGTH	(32.394 in.)	822.81 mm
FUEL PELLET LENGTH	(0.625 in.)	15.88 mm
CLAD - OD	(0.567 in.)	14.40 mm
CLAD THICKNESS (304L SS)	(0.032 in.)	0.813 mm
FUEL COLUMN DIAMETER	(0.498 in.)	12.649 mm
UO_2 DENSITY (% THEORETICAL)	94 - 98	
ENRICHMENT	4.3 w/o ^{235}U	

* MEASURED FROM BOTTOM OF END SPADE TO BOTTOM OF TOP SPADE

FIGURE 19. Description of 4.3 wt% ^{235}U Enriched Fuel Rods

Analysis of these data show that the critical number of fuel pins varies from almost 187 (water without neutron poison) to about 317 in water containing 0.044 g Gd/l, and 434 in water containing 0.22 g b/l for the 33.0 mm lattice spacing. Figure 20 shows a graph of the critical number of rods plotted against the neutron absorber concentration for the 33.0 mm lattice.

FIGURE 20. Effect of Neutron Absorber on 1.3 in. (33 mm) Lattice

Survey Calculations

Survey calculations were made on the criticality of three lattices of 4.3 wt% ^{235}U rods in uranyl nitrate solution containing varying amounts of gadolinium as the neutron absorber. Table 7 lists the results of these calculations.

TABLE 7. Calculated Effect of Gadolinium

<u>Hexagonal Pitch (cm)</u>	<u>No. of Fuel Rods</u>	<u>Gd Concentration (g/.)</u>	<u>Bm^2 (cm⁻²) x 10⁻³</u>	<u>Hc (cm)</u>
2.286 (0.9")	480	0	7.91	37
2.286 (0.9")	480	0.10	6.50	53
2.286 (0.9")	480	0.15	5.87	68
2.286 (0.9")	480	0.175	5.55	80
2.286 (0.9")	480	0.130	5.50	82
2.794 (1.1")	429	0	8.22	32
2.794 (1.1")	429	0.05	6.81	40
2.794 (1.1")	429	0.10	5.54	54
2.794 (1.1")	429	0.125	4.98	66
2.794 (1.1")	429	0.146	4.51	82
3.302 (1.3")	409	0	7.04	33
3.302 (1.3")	409	0.02	6.12	39
3.302 (1.3")	409	0.06	4.62	57
3.302 (1.3")	409	0.07	4.30	65
3.302 (1.3")	409	0.085	3.80	82

A graph of critical solution height against the gadolinium concentration is shown in Figure 21 for the three lattice spacings, 22.9, 27.9 and 33.0 mm.

Experimental Preparation

Experimental equipment needed for the experiments with the lattices of 4.3 wt% ²³⁵U rods in uranyl nitrate solution is being installed. The previously used obsolete storage tank system was removed and two slab storage tanks have been installed for containing the uranyl nitrate solution. Liquid level measurement instrumentation is now being installed.

PLANS FOR NEXT QUARTER

- Complete preparation of the system for experimentation with fuel rods in uranyl nitrate solution and perform experiments with rods in the solution containing gadolinium as a neutron absorber.

- Complete survey criticality calculations with mixed oxide fuel rods in water and in water containing neutron absorbers.
- Complete analytical analysis of the experimental data from the 4.3 wt% ^{235}U rods in water and in water containing neutron absorbers.

FIGURE 21. Effect of Gd on Critical Height

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

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

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

OBJECTIVES

The criticality research program is directed towards 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 criticality data needed in the chemical reprocessing of the TFCTP fuels.

SUMMARY

Funding authorization was received immediately preceding the quarter. Task breakdowns and staff assignments were made to accomplish the objectives of the program. Our first efforts were limited to document reviews and literature searches pertinent to chemical reprocessing of the TFCTP fuels. These were essentially completed and a draft document was prepared delineating data requirements.

TRIPS AND VISITORS

Documents, reviews and literature searching pertinent to chemical reprocessing of the TFCTP fuels were completed. Contacts were also made with Savannah River Laboratory, Oak Ridge National Laboratory, and Rocky Flats personnel involved with criticality research and chemical reprocessing development.

TECHNICAL PROGRESS

A basic process flow diagram has been formulated for use as a guide in planning criticality experiments of the TFCTP. This flow diagram closely follows the Reference Thorium Fuel Cycle covered in DPST-TFCT-77-101 and will

be included in a document which summarizes existing light water experimental data on ^{233}U , ^{233}U -Th mixtures, ^{235}U - ^{238}U -Th mixtures, and Pu-Th mixtures.

A summary of the expected material forms requiring benchmark criticality data is given below:

Powders:	UO_2 - ThO_2 and PuO_2 - ThO_2 at low and medium neutron moderations.
Pellets:	UO_2 - ThO_2 and PuO_2 - ThO_2 both dry and moderated (may only be needed in fuel fabrication).
Rods and Fuel Rod Bundles:	UO_2 - ThO_2 and PuO_2 - ThO_2 mixtures in acid solutions with and without soluble neutron poisons.
Solutions:	$\text{UO}_2(\text{NO}_3)_2$ - $\text{Th}(\text{NO}_3)_4$ and $\text{Pu}(\text{NO}_3)_4$ - $\text{Th}(\text{NO}_3)_4$ solutions at both over and undermoderated conditions, and with and without soluble poisons.
Fluidized Beds:	$\text{UO}_2(\text{NO}_3)_2$ - $\text{Th}(\text{NO}_3)_4$ with oxide spheres and possible plutonium.

Some criticality data currently exist that can be used to satisfy some of the above requirements. We are developing an experimental program based on this data and the above requirements.

In anticipation of the experimental program on criticality research in support of the TFCTP, several sources of ^{233}U (5 kg to 150 kg of varying ^{232}U content) and ^{235}U have been identified that may be useful in conducting the critical experiments. These include some ^{235}U (20)-Th rods to be fabricated next year at Hanford as part of the Thorium Fuel Refabrication Program. Purification of ^{233}U solutions can be accomplished locally in PNL facilities.

PLANS FOR NEXT QUARTER

Continued development of program plan to meet objectives of project.

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