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SEMIANNUAL REPORT
KK PROCESS DEVELOPMENT AND TECHNOLOGY
NOVEMBER 1, 1975 THROUGH APRIL 30, 1976

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ABSTRACT

This document represents the final report on a semiannual basis of the activities supported by KK funds in the areas of process development. These research and engineering activities have the goal of improving the performance of the plutonium processing and waste management programs being operated by the Atlantic Richfield Hanford Company.

RESEARCH AND ENGINEERING DIVISION
SEMIANNUAL REPORT

KK PROCESS DEVELOPMENT AND TECHNOLOGY
NOVEMBER 1, 1975 THROUGH APRIL 30, 1976

INTRODUCTION

This document describes the scientific investigations and engineering application programs being sponsored by KK funds under the direction of Atlantic Richfield Hanford Company's Research and Engineering Division. The document has been divided into 11 program areas--1 associated with plutonium production, 1 with Purex fuel reprocessing, and 9 with managing the wastes generated during production. Each of these program areas is divided into the activities necessary to effectively present the technical information.

The report titles are those identified in the program plans and the ARHCO code listings. The program manager responsible for defining the program objectives and ensuring that adequate resources are available to perform the work is identified, as well as the principal investigators who are responsible for directing the daily work.

This report is intended to present progress and is not a total program plan for each activity. Interested individuals are invited to communicate with the program managers for planning information and to offer comments.

Each report is presented with sufficient background for the reader to understand the scope and objective of the program, a brief summary of prior work, a review and analysis of the work conducted during the report period, and work

planned for the next six months. Pertinent reports issued during this period are listed.

Previous documents issued include ARH-ST-128 A and ARH-ST-128 B.^{1,2} Future documents will be consolidated with quarterly process technology and process development reports.

SUMMARIES

Page

PUREX FUEL REPROCESSING

CONTROL OF IODINE-129 DURING FUEL REPROCESSING

12

The majority of ^{129}I associated with fuel processing at Purex will exit in the organic wash. The ratio of inert iodine to ^{129}I trapped by silver is high; and, therefore, silver reactors would be too expensive.

¹Research and Engineering Division Semiannual Report, KK Process Development and Technology, November 1, 1974 Through April 30, 1975, R. D. Fox, ed., US-ERDA Report ARH-ST-128 A, Atlantic Richfield Hanford Company, Richland, Washington, August 1975.

²Research and Engineering Division Semiannual Report, KK Process Development and Technology, May 1, 1975 Through October 31, 1975, R. D. Fox and C. W. Manry, eds., US-ERDA Report ARH-ST-128 B, Atlantic Richfield Hanford Company, April 1976.

PLUTONIUM FINISHING PLANT

PLUTONIUM PROCESS TECHNOLOGY

Plutonium Reclamation Facility Waste Solidification 14

The corrosion problems of the drum drier can be controlled by proper choice of construction materials. Vapor-condensate Pu DF of 10^5 to 10^6 can be achieved across each stage of concentration equipment, and using two stages of concentrating equipment produces condensate that is well below environmental release guidelines.

Plutonium Solvent Extraction Model 16

Computer modeling has enabled evaluation of operating conditions affecting process efficiency and should improve control of the process.

Waste Treatment Column Improvement 18

The need for an improved method of detecting column flooding and accuracy of cerium measurement as a simulation of americium was demonstrated. A special differential pressure indicator for detection of flooding was ordered.

Plutonium Oxide Sinterability Characteristics 20

Sintered densities increased 1 to 3% upon aging for one week, although all PuO_2 powders sintered to $>90\%$ of theoretical density at temperatures $\leq 750^\circ \text{C}$.

PLUTONIUM PROCESS DEVELOPMENT

Plutonium Oxide Dissolution 22

A scouting study indicates that several characteristics for fusions on incinerator ash must be met to be acceptable. Methods to identify and

classify actinide-bearing scrap types are being developed.

Plutonium Oxide Dissolvers

24

The use of thermal energy to provide continuous agitation in conjunction with continuous feed capabilities is an improvement over the batch dissolvers presently in use.

Amine Solvent Extraction of Plutonium

26

Acquisition of reliable data was completed for distribution of Pu and HNO_3 between 30% LA-2- CCl_4 solvent and 1 to 4M HNO_3 -0 to 1M $\text{Al}(\text{NO}_3)_3$ -1 to 20 g/liter $\text{Pu}(\text{NO}_3)_4$ solutions. The SOLVEX computer program for calculating steady-state and transient performance of solvent extraction systems has been compiled on the Hanford computer for use with the LA-2 distribution data.

Bidentate Organophosphorus Extraction of Americium and Plutonium

29

The bidentate Am-Pu extraction process is a practical, viable scheme suitable for use in the Hanford PRF. Bidentate process flowsheets for efficient removal of Am and Pu from acid waste solution--even to the point of producing actinide-free (<10 nCi/g) solutions--have been devised and demonstrated in mixer-settler tests.

FRACTIONIZATION

REMOVING CESIUM FROM PUREX CURRENT ACID WASTE

33

A laboratory program has been defined for evaluating Cs removal from Purex acid waste by ion exchange.

PageSLUICING AND B PLANT FEED PREPARATION

34

A market search was made for a substitute for the discontinued flocculating agent XFS 4111L. Several satisfactory substitutes were found and recommended to be tested under actual operating conditions.

STRONTIUM SOLVENT EXTRACTION

35

High Sr losses in B Plant were not caused by substituting citric acid for part of the more expensive HEDTA and EDTA complexants.

STRONTIUM PURIFICATION

37

A lead carrier precipitation process was developed which recovered at least 90% of the Sr during purification. A process test will be conducted during the next sulfate purification campaign.

ENCAPSULATIONCESIUM ENCAPSULATION

39

The tilt-pour melt caster and pan drier prototype equipment were fabricated and installed for testing. Mock-ups of this equipment to be used for concurrent remote testing have been fabricated and are being assembled along with a remote test station equipped with manipulators.

CAPSULE DECONTAMINATION

40

The electropolishing equipment for in-cell operation was fabricated. Construction of a cell mock-up which will be used in cold testing the electropolishing equipment is about 50% complete.

PageALTERNATIVE PRODUCT FORMS FOR ENCAPSULATION

41

Cesium can be converted to material for long-term and/or final storage. This can be accomplished by converting either the process feed solution from B Plant or with CsCl after conversion from the carbonate or after dissolution from the stored capsules.

CONCENTRATION, TERMINAL LIQUOR, SALT CAKE,
AND SLUDGE CHARACTERIZATIONWASTE MANAGEMENT PLANNING SUPPORT

45

The Waste Management Model provides an insight into the operating parameters which have the greatest impact on future waste volumes. The model was modified to incorporate the shutdown of 242-T Evaporator and also to reflect the effects of partial neutralization of the waste.

INTERSTITIAL LIQUOR REMOVAL

47

Instrumented Salt Well facilities provided data to compare with the flow predictions of a mathematical model. Tests of a deep well jet pump have warranted a recommendation that they be implemented as standard salt well pumping equipment. Steam injection into a salt well was found to stimulate flow.

TERMINAL LIQUOR DIPOSITION

52

The wiped film evaporator samples obtained from Savannah River were characterized. Work on the clay process for waste solidification was terminated. A partial neutralization process test was completed successfully, and the design for a prototype installation was completed.

PageALUMINUM REMOVAL FROM HIGH-LEVEL RADIOACTIVE WASTE

56

It has been shown that aluminum removal from Hanford wastes is not feasible for interim waste storage because of the time required to develop and demonstrate an acceptable process.

SALT CAKE CHARACTERIZATION (ANALYTICAL DEVELOPMENT)

58

The laboratory is now in a position to receive and analyze waste tank core samples on a routine basis.

SALT CAKE CHARACTERIZATION (CORE SAMPLER DEVELOPMENT)

60

The salt cake coring equipment has been tested and is a viable method of obtaining salt cake samples.

TANK OPERATIONAL CRITERIA

62

The underground waste storage tanks which were originally designed for liquid storage are now to be used for the storage of salt cake. Criteria have been prepared for the 271-U and 241-SX Tank Farms. Data are available for the preparation of a criterion for the 241-BX, 241-BY, 241-TX, and 241-A Tank Farms.

WASTE TANK LEAK DETECTION

65

Air-purged weight factor dip tubes, flushed weekly with steam, have proved the only reliable method of determining the liquid level in a waste tank when the liquid level is below the surface of the salt cake. Corrosion monitoring probes and ultrasonic thickness measurements of the steel are being tested to determine their utility for monitoring tank liner corrosion.

Page

CONTAMINATED SEDIMENT CONTROL AND BIOLOGICAL UPTAKE

SITE CHARACTERIZATION

67

A split tube sampler developed for use in the 216-U-10 Pond was successfully demonstrated and assures that no cross contamination of the sediments occurs. Results from U Pond samples tended to substantiate that radionuclides are concentrated in the top few centimeters of soil.

SOIL STABILIZATION

70

A soil conductivity system installed at the Tank 102-AX vent leak immobilization test showed increases in soil conductivity during the water addition and first addition of asphalt. The final two additions showed little change. A drilling technique has a potential for supplying a quick and low-cost method of installing monitoring wells, injection points for grouting, segmented electrodes, and core sampling.

DECONTAMINATION/IMMOBILIZATION

75

Modification of the mobile vacuum system is about 90% completed. A characterization study of the 200 West Area Laundry effluent was initiated.

RADIONUCLIDES IN SOIL-PLANT SYSTEMS

78

Concentration ratios of ^{99}Tc were measured for cheatgrass and tumbleweed. An experiment was initiated to measure the biological availability of Sr and Cs as a function of soil properties and waste composition.

RADIONUCLIDES IN TERRESTRIAL PLANT-ANIMAL SYSTEMS

81

Potential radiocontaminant pathways through the food chain are discussed. Techniques were developed for assessing dose rates to native

animals, and body-burden levels of animals living near a radioactive waste pond were determined.

RADIONUCLIDES IN AQUATIC BIOTA AND SEDIMENTS

86

A two-year field sampling study of seasonal body burden levels and diets of coots living on radioactive waste ponds was completed. There does not appear to be any seasonal cycling of radionuclide levels in coots.

ACTINIDE SEDIMENTS

ACTINIDE TRENCH CHARACTERIZATION

89

Tests indicate that two tools have been developed which are capable of obtaining representative samples of sludge within Tank 361-Z. Preliminary analyses of samples from the 216-Z-12 Trench indicate that the bulk of the Pu lies just below the point of waste release. Efforts to determine Pu "bonding strength" in trench samples were initiated.

HYDROLOGY

GROUNDWATER HYDROLOGY

95

Field verification of structural data for 200 area wells was initiated. Maps of the Hanford water tables were issued. A rise in the water table in the vicinity of U Pond is being investigated.

OFFSITE MIGRATION

98

Program plans were drafted for this new program.

PageTANK FARM GEOLOGY

100

Completion of the tank farm geology maps and Ringold Formation coring have aided in understanding the structure and stratigraphy under the 200 Areas. *In situ* analyses of radionuclides in sediments will benefit future tank-leak studies.

TRANSPORT STUDIES

MOISTURE TRANSPORT STUDIES

104

Lysimeters were valuable in determining the depth to which meteoric water moves down, but more information is needed to characterize areas with different sediments and with various amounts of water or liquid waste being added routinely. Progress has continued on the development of a computer code that will model nonisothermal unsaturated moisture transport in porous media.

PUBLIC PROTECTION ASSURANCE

ENVIRONMENTAL INSTRUMENTATION

107

The beta assayer shows promise in assaying environmental samples. Gamma surface assay equipment in vans for characterizing and assisting in current problems are likely alternatives to costly laboratory analyses.

HAZARDS AVAILABLE TO MAN

112

Work continued on the HI portion of the HA equation. Estimates were made on the meteorological transport factors, resuspension, dilution, and deposition.

ANALYTICAL METHODS DEVELOPMENT

ACTINIDE ANALYTICAL PROGRAM

115

The separation alpha counting procedure for determination of Pu and Am has an analytical precision of $96 \pm 12\%$ and $83 \pm 19\%$, respectively. More work is needed on the U, Th, Np, and Am procedures.

RADIATION ANALYSIS SYSTEMS DEVELOPMENT

119

Phase 3 of the GXDR update has been completed for one NS-660 only with a resultant four-fold reduction in gamma spectral data reduction time. A Canberra Scorpio 3000 is being purchased to provide a data path from existing data acquisition systems as well as to provide for future 7000+ channel acquisition system expansions.

CALIFORNIUM MULTIPLIER SUBCRITICAL ASSEMBLY

122

During the report period the program received approval and funding. The principal contracts were signed and purchase orders were issued. A schedule of milestones and safety reviews has been established together with the personnel to implement them.

PUREX FUEL REPROCESSING

CONTROL OF IODINE-129 DURING FUEL REPROCESSING

Program Manager: R. E. Felt
Section Managers: R. E. Felt
H. Babad
Principal Investigators: D. W. Jeppson
D. M. Strachan
Activity Code: B111A

Objectives and Scope

The purpose of this activity is to determine the routes of iodine-129 through the Purex Plant and to develop methods to control its release during fuel reprocessing to as low as practicable.

Prior Work

None.

Progress During Report Period

The ^{129}I content of the Purex recovered nitric acid, washed organic, uranium product, and main stack off-gas has in the past been determined for periods of plant operation. Based upon the data collected to date, the majority of the ^{129}I in fuel to be processed during future campaigns at Purex (assuming the new condensate recycle system is adopted and no new provisions for removal of iodine in the dissolver off-gas are implemented) will exist in the organic wash waste.

Analyses of the dissolver off-gas silver reactor beds indicate that the use of silver to trap ^{129}I would be expensive because of the high ratio of inert iodine to ^{129}I trapped on the beds during past operations.

Laboratory studies using ^{131}I as a tracer and using irradiated fuel elements to determine the iodine split in the dissolver during decladding and uranium dissolution have been started.

Evaluation of Effort

The paths of ^{129}I leaving the Purex Plant during future operations have been determined. Further definition will be obtained by analyzing the exiting process streams when the plant again begins operation.

Future Work

Refinements in procedures are being developed to provide the capability of measuring the iodine split in the dissolver during decladding and uranium dissolution.

Additional analyses of the used silver reactor Berl saddles are being made to determine if other elements such as fluoride are significant competitors to iodine for silver.

If additional ^{129}I control is warranted, then some alternatives to the silver reactor will be evaluated.

PLUTONIUM FINISHING PLANT

PLUTONIUM PROCESS TECHNOLOGYPlutonium Reclamation Facility Waste Solidification

Program Manager: R. E. Felt
Section Manager: R. E. Felt
Principal Investigators: T. R. Haddad
D. A. Reynolds
M. J. Schliebe
Activity Code: B04T3

Objectives and Scope

This project's goal is to develop a concentrating and drying process for converting Plutonium Reclamation Facility (PRF) aqueous salt waste to a solid suitable for retrievable transuranic storage.

Prior Work

A double-drum drier system will be used to dry the PRF aqueous salt waste. A pilot unit was purchased and installed to study operating parameters.

Initial studies on the evaporator showed that the first experimental evaporator did not have enough vapor deentrainment room. A new unit was designed and built to alleviate this problem.

Progress During Report Period

The data from the drum drier indicate that a production rate of 300 g (bone-dry) product/hr/m² can be expected. This product varies in dryness from 12 to 17% moisture. The three most important variables in the production rate are drum revolutions per minute (r/min), steam pressure, and percent solids in the feed. Drum r/min is by

far the most important variable. Steam pressure and percent solids have less impact on the production rate.

After a period of running the drum drier, corrosion pits appeared on the 316 stainless steel drums. Battelle Northwest Laboratories was asked to evaluate the cause of the corrosion. Their tests have shown that 316, 304L, and E-Brite 26-1[®] (Airco Alloys and Carbon Division, Air Reduction Co., Inc.) are attacked at the grain boundaries. Hastelloy C[®] (Stellite Division, Cabot Corporation) and tungsten carbide corrode even faster. However hard chrome plate over 316 stainless steel, Hastelloy G[®] (Stellite Division, Cabot Corporation) and Inconel 690[®] (Huntington Alloys Products Division, International Nickel Co., Inc.) are resistant to corrosion by the feed solution. Since those initial runs, the drums were resurfaced and run with many different combinations of feeds and blades but no sign of pit corrosion has occurred.

Metals have been ruled out as doctor blade materials for stainless steel drum driers. Bronze, nickel, and tantalum were tried as blade materials. Nickel and tantalum scored the drum surface. Bronze did not damage the drums but corroded badly. Several plastics have been tried for doctor blades. Glass-filled Teflon has been the most successful blade yet. Ryton-4[®] (Phillips Products Company, Inc.), a polyphenylene oxide, also worked very well.

Vapor-condensate-decontamination factors for Pu in the PRF aqueous salt waste were tested in the laboratory. Limiting the liquid entrainment in the off-gas system yielded a decontamination factor (DF) of 10^6 for condensate produced from the evaporation of the PRF high-salt waste solution. A DF of $\sim 10^5$ was also demonstrated in the second evaporation cycle. The overall DF of $\sim 10^{11}$ exceeds the

minimum design requirement of the PRF Waste Solidification program.

Evaluation of Effort

The PRF aqueous salt waste can be dried adequately on a drum drier. The corrosion problems can be controlled by proper choice of construction materials.

Vapor-condensate Pu DF's of 10^5 to 10^6 can be achieved each stage of concentration equipment. An average DF of 10^4 is adequate using two stages of concentrating equipment to produce process condensate which meets environmental release limits.

Future Work

The drum drier will be run extensively to get better corrosion data on the equipment. A chrome-plated drum will be tried to test corrosion and operating difficulties. These trials will also show if it is possible to treat the waste stream from the Hanford Engineering Development Laboratory acid digestion process on the drum drier.

Plutonium Solvent Extraction Model

Program Manager:	R. E. Felt
Section Manager:	R. E. Felt
Principal Investigator:	M. J. Klem
Activity Code:	B04T1

Objectives and Scope

The objectives are to develop a mathematical model of the PRF solvent extraction process using the SEPHIS

program, study the existing flowsheets, and evaluate changes for improved process performance. The model will be used to develop flowsheets as comprehensive equilibrium data for the distribution of plutonium and nitric acid into new process solvents become available.

Prior Work

The PRF solvent extraction system has been modeled. Alternative flowsheets have been constructed for improving process efficiency, reducing the quantity of liquid wastes discharged, and reducing the extractable Pu content of the waste stream by a factor of 10.

Progress During Report Period

The standard Pu(IV) flowsheet was documented. A statistical design of stream flow rates and feed concentrations was completed and a response equation developed for predicting extraction and stripping column Pu losses. Also the effect of changing the 20 vol% tributylphosphate (TBP) extractant concentration to 18 and 22% was determined.

Evaluation of Effort

Computer modeling has enabled evaluation of operating conditions affecting process efficiency. The computer method of analyzing complex solvent extraction problems should improve control of the process.

Future Work

Document the plutonium-uranium partitioning flowsheet and develop flowsheets as comprehensive equilibrium data for new process solvent become available.

Waste Treatment Column Improvement

Program Manager: R. E. Felt
Section Manager: R. E. Felt
Principal Investigator: M. J. Klem
Activity Code: B04T3

Objectives and Scope

The objective of this work is to evaluate potential changes in the Continuous Waste Treatment facility equipment and flowsheet for improved solvent extraction recovery of americium and plutonium. New extraction columns and process solvents will be tested.

Prior Work

Installation and preliminary testing of the three-column pilot facility have been completed. Two problems common to the plant columns have been identified:

- The capacitance probe for detecting the position of the aqueous-organic interface in the column gives erroneous indications because air registers the same as the organic.
- The interface controller on the upstream column(s) does not provide a smooth, continuous flow of organic to the cascade downstream column(s).

The pilot pulse columns could not be packed tightly enough with 2.5-cm Kynar[®] (Pennwalt Corporation) plastic Intalox saddles to prevent the packing from moving up and down with the pulse wave, thus decreasing the column's efficiency.

Progress During Report Period

An improved interface controller was installed on the pilot extraction column for smoothing the flow of organic to the downstream columns. A hold-down plate was installed in the top portion of the same column to prevent movement of the Kynar packing during the pulsing action. Test runs were started using synthetic high-salt waste (Elf) as the feed. The feed contained natural Ce(III) as a substitute for Am(III). Aqueous-organic phase mixing efficiency was evaluated for columns containing 2.5-cm Kynar plastic Intalox saddles and 1.6-cm stainless steel pall ring packing. Laboratory problems in analyzing for Ce have prevented quantitative evaluation of the test runs.

Evaluation of Effort

Work has demonstrated the need for an improved method of detecting column flooding and accuracy of Ce measurement. A special differential pressure indicator for detection of flooding has been ordered.

Future Work

Column efficiencies will be determined by varying pulse conditions, using other types of column packing material, using different sieve and nozzle plates, changing flowsheet conditions, and using new process solvents. The value of the previous work has been reduced because of the poor accuracy of Ce analyses.

Plutonium Oxide Sinterability Characteristics

Program Manager: R. E. Felt
Section Manager: H. Babad
Principal Investigator: A. L. Dressen
Activity Code: B03T0

Objectives and Scope

Failure of Fast Flux Test Facility (FFTF) fuel-grade plutonium dioxide batches to meet the reactor development technology specification for sinterability to 90% of theoretical density or greater results in costly reprocessing and delays in shipping schedules. The objective of this study is to develop the technology to consistently produce sinterable PuO_2 . This task involves identification of powder characteristics which determine PuO_2 sinterability and definition of operating parameters to control these characteristics.

Prior Work

Other studies have identified a number of relationships between powder characteristics, operating conditions, and sinterability. However, in some cases, the conclusions are contradictory and inadequate to explain the variations in ceramic qualities of PuO_2 which occur within a continuous production run.

Progress During Report Period

Evaluation of the effect of recalcining PuO_2 of poor sinterability was completed. All powder recalcined at $\leq 750^\circ \text{C}$, as well as the original PuO_2 , sintered to $\geq 90\%$ of theoretical density, suggesting the existence of uncontrolled variables. No statistically significant correlations were observed in these tests between sintered density and powder characteristics.

The characteristics of PuO_2 from one production run have been examined. Sintered densities increased 1 to 3% upon aging for one week although all samples initially sintered to >90% of theoretical density. This aging phenomenon has been observed previously in powders of poor sinterability. Stoichiometry of the PuO_2 was confirmed by measurement of the lattice parameters. The powders consisted of two types of agglomerates: small clusters of randomly oriented plates and larger rosettes which appeared similar to the small clusters consisting of columnar growth radiating from a core.

Evaluation of Effort

Incomplete data make meaningful evaluation impossible at this time.

Future Work

Further examination of powder characteristics will be conducted to define relationships between these and sinterability. In addition a bench-scale model for semi-continuous PuO_2 production will be utilized to select and define the operating parameters of feed adjustment, precipitation, and calcination which have important effects on powder characteristics and sinterability.

PLUTONIUM PROCESS DEVELOPMENTPlutonium Oxide Dissolution

Program Manager: R. E. Felt
Section Manager: H. Babad
Principal Investigator: D. A. Dodd
Activity Code: B1350

Objectives and Scope

The purpose of this study is to better understand the dissolution behavior of plutonium dioxide alone and when contained in miscellaneous scrap forms. This work will define dissolution conditions which will effectively reclaim remaining Pu and are also compatible with the PRF. The system must minimize effluent waste which contains actinide contaminants. The waste streams produced must not introduce additional hazardous conditions under storage conditions.

Prior Work

The technology for optimum HNO_3 -HF dissolution of PuO_2 and actinide-bearing scrap was reported in the last semiannual report, ARH-ST-128 B. The major contribution made was in the design and testing of the continuous counter-flow solid-solvent reaction vessel. This laboratory-scale vessel was proven to significantly increase dissolution rates of scrap by maintaining the uncomplexed fluoride ion within the optimum concentration range (0.05 to 0.2M) and in intimate contact with the less-soluble PuO_2 .

Preliminary tests were made on actinide scrap such as incinerator ash and centrifuge sludge with dissolution solutions other than HNO_3 -HF and fusion techniques.

Progress During Report Period

Preliminary oxidizing fusions were made on incinerator ash in a scouting study. To be acceptable, the fusion must have several characteristics including:

- Must be controllable.
- Minimum scrap-to-salt ratio.
- Minimum melt-to-acid ratio such that the aqueous product-bearing stream is greater than 10 g/liter Pu.
- Process must be more efficient than the current HNO_3 -HF leaching techniques.
- Must be applicable to continuous processing environment.

Methods to identify and classify actinide-bearing scrap types are being developed. This technology will aid in determining processing conditions required to recover Pu from the scrap matrix. To be considered a viable tool for process control, techniques for analyses without prior time-consuming preparation are necessary.

Evaluation of Effort

The alternative aqueous systems to HNO_3 -HF tested thus far have not produced a consistent improvement over the HNO_3 -HF counter-current dissolver system. These tests have been directed specifically toward dissolution of incinerator ash.

The limited fusions performed to date, although effective on laboratory-scale batches, as yet have not been identified as viable routine processing techniques.

Future Work

Additional fusion and molten salt formulations will be tested and measured.

Plutonium Oxide Dissolvers

Program Manager:	R. E. Felt
Section Managers:	R. E. Felt H. Babad
Principal Investigators:	D. A. Dodd C. E. White
Activity Code:	B1350

Objectives and Scope

The objective of this study is to design and test a dissolver that will represent a significant improvement over the currently used dissolver pots. Present flowsheet parameters necessitate the use of an HNO_3 -HF system. Because of the highly variable nature of stored scrap, this study considers as many forms as practicable.

Prior Work

Controlled batch and semicontinuous dissolution tests were conducted on PuO_2 prepared from oxalate, metal, and oxide sintered to 1400°C . These tests showed that the continuous system is superior to the batch and that 0.1 to 0.2M HF in 12M HNO_3 is sufficient to dissolve Pu.

Alternative dissolver solutions and nonaqueous methods have not shown significant improvement over the HNO_3 -HF system.

Progress During Report Period

Continuous agitation was obtained in the prototype dissolver using a thermal gradient as the driving force. Design modifications established a satisfactory "quiet zone" from which product solution can be withdrawn.

Continuous feed testing was initiated using simulated ash and water to check flow characteristics. The dissolver is capable of handling up to two liters per hour of liquid throughput before the turbulent zone and the quiet zone began to merge.

Evaluation of Effort

The use of thermal energy to provide continuous agitation in conjunction with continuous feed capabilities is a significant improvement over the batch dissolvers presently in use.

Future Work

Further testing of continuous feed capabilities is necessary. Testing will also be performed to determine satisfactory methods of undissolved solids removal, internal flow measurement, and solids buildup measurement.

Hot testing will be conducted after the dissolver hydraulics have been worked out with cold testing.

Amine Solvent Extraction of Plutonium

Program Manager: R. E. Felt
Section Manager: H. Babad
Principal Investigators: W. W. Schulz
D. G. Bouse
Activity Code: B1350

Objectives and Scope

The objective of this program is to develop an amine plutonium extraction process to replace the tributylphosphate (TBP) process currently used in the PRF to realize improved performance and minimize operating costs.

Prior Work

Plutonium extraction capacity and other chemical and physical properties of CCl_4 and trichlorobenzene (TCB) solutions of Amberlite LA-2[®] (Rohm and Haas Company), a high-molecular weight secondary amine, were studied intensively. Comprehensive equilibrium data were obtained for the distribution of Pu and HNO_3 between 30% LA-2- CCl_4 solvent and aqueous 0.1 to 4.0M HNO_3 -0.0 to 1.0M $\text{Al}(\text{NO}_3)_3$ -0.0 to 0.25M HF solutions containing 1, 10, 30, and 60 g/liter Pu. These data were used to devise a new amine Pu extraction process. This new process involves operation of only the CA (extraction), CC (strip), and CO (solvent wash) Columns; an HNO_3 -HF solution is used in the CO Column to remove small amounts of Pu which are not stripped in the CC Column.

Initial attempts were made to model the LA-2 extraction system for subsequent computer-assisted flowsheet development activities. Equilibrium organic phase Pu and HNO_3 concentrations were compared with those expected from known secondary amine extraction mechanisms and

stoichiometries. Acidities of LA-2 phases containing high (>20 g/liter) Pu concentrations appeared self-consistent and in reasonable agreement with expected concentrations. At lower organic phase Pu concentration, however, HNO_3 concentrations of the LA-2 phases were higher than expected. This discrepancy was believed to have arisen from failure to completely convert the LA-2 from free base-form to nitrate-form before extraction of Pu and HNO_3 .

Progress During Report Period

Progress toward development of a reflux amine Pu flowsheet continued at a minimal level because of a continuing deliberate decision to utilize available resources to complete studies of the bidentate extraction process. A key task completed, however, was acquisition of reliable data for distribution of Pu and HNO_3 between 30% LA-2- CCl_4 solvent and 1 to 4M HNO_3 -0 to 1M $\text{Al}(\text{NO}_3)_3$ -1 to 20 g/liter $\text{Pu}(\text{NO}_3)_4$ solutions. These data were obtained with LA-2 which had been completely converted to the nitrate-form.

A new computer program, SOLVEX, for calculating steady-state and transient performance of solvent extraction systems has been recently published by W. Scotten of the Savannah River Laboratory. This program can accommodate tabular arrays of distribution data and thus appears easier to use, at least in initial flowsheet modeling, than the SEPHIS program. The SOLVEX deck was obtained and has been successfully compiled on the Hanford computer in preparation for using the program with the LA-2 distribution data.

Secondary amines such as LA-2 can undergo reaction with nitrous acid to form insoluble nitrosoamines. Earlier scouting studies indicated that reaction of LA-2 with nitrous acid was highly unlikely under normal PRF operating

conditions. However, because such reaction represents a potential way of losing LA-2, a contract has been negotiated with Professor Celestine Tillman of Southern University to determine exactly what conditions of temperature, HNO_3 , and nitrous acid concentration are required for LA-2 to react with nitrous acid.

Evaluation of Effort

The demonstrated superior chemical and radiolytic stability of Amberlite LA-2, compared with that of TBP, makes an amine extraction process particularly well-suited for eventual use in the PRF with scrap containing significant concentrations of ^{241}Pu and ^{240}Pu . The potential benefits to be derived from replacing the TBP solvent with LA-2 are considered great enough to warrant a continuing low level of effort to computer-model the LA-2 extraction system and to devise optimum chemical flowsheet conditions.

Future Work

A major effort in the next report period will be devoted to application of the SOLVEX program in computer-assisted flowsheet modeling of LA-2 extraction systems. Some of the resulting flowsheet conditions will be tested in mixer-settler runs. Detailed laboratory studies of the reaction of LA-2 with nitrous acid by Professor Tillman are also scheduled.

Bidentate Organophosphorus Extraction of Americium and Plutonium

Program Manager: R. E. Felt
Section Manager: H. Babad
Principal Investigators: W. W. Schulz
D. G. Bouse
Activity Code: B1350

Objectives and Scope

The principal objective of the activity is to develop technology for reducing the americium and plutonium content of the PRF aqueous waste to ≤ 10 nCi/g levels. A secondary objective is to develop and demonstrate a bidentate organophosphorus solvent extraction process to recover both Am and Pu directly from the PRF acid waste stream, thereby avoiding the pH adjustment step required by the present dibutylbutylphosphonate (DBBP) extraction scheme.

Prior Work

Extensive data were obtained for the equilibrium distribution of Am, Pu, HNO_3 , and various impurity metals (e.g., Ca^{2+} , Mg^{2+} , etc.) between aqueous HNO_3 - $\text{Al}(\text{NO}_3)_3$ -HF solutions and 30 to 40 vol% dihexyl-N,N-diethylcarbanylmethylenephosphonate (DHDECMP)- CCl_4 [and trichlorobenzene (TCB)]. The bidentate organophosphorus reagent DHDECMP is available from Wateree Chemical Company, Lugoff, South Carolina.

Technical-grade DHDECMP contains an unidentified acidic impurity that has a very high affinity for Am(III) and Pu(IV) at low-acid ($\leq 0.25M$ HNO_3) concentrations. Conditions for removal of this impurity by acid (HCl) hydrolysis techniques were devised. Work completed near the end of

the prior work period indicated that the offending acidic impurity could also be removed from commercial DHDECMP by batch-washing with ethylene glycol. Some 15 mixer-settler extraction column runs were completed employing both 30 and 40 vol% purified (HCl hydrolysis) DHDECMP-TCB solvents and two different batches of plant acid waste as feed. Flow-sheet conditions (40 vol% DHDECMP; 8 stages, aqueous/organic flow = 1/2) for reproducible reduction of the soluble ^{241}Am concentration of acid waste solution to <10 nCi/g were established. Behavior of Pu in these mixer-settler runs was not established; but, from previous work, it is known that Pu(IV) is at least a factor of 10 more extractable than Am(III). These mixer-settler runs thus established conditions for producing an actinide-free waste.

Also during the report period, a cyclic alpha radiolysis-solvent wash experiment was begun to determine more fully the useful life of a DHDECMP extractant under the PRF conditions. This experiment involved allowing a 30 vol% DHDECMP-TCB solvent containing ^{241}Am to stand at 25°C until an absorbed dose of 15 to 20 watt-hr/liter had been accumulated; subsequently the solvent was washed with HNO_3 and Na_2CO_3 and its Am extraction and stripping properties compared with those of virgin solvent. This cyclic irradiation-wash pattern approximates that expected for a DHDECMP solvent in the PRF.

Progress During Report Period

The effectiveness of ethylene glycol washes in removing deleterious acidic impurities from DHDECMP-TCB solvents was convincingly demonstrated in tests with several batches of technical-grade DHDECMP. [Dilution of the DHDECMP with TCB or other appropriate diluent is necessary prior to washing with ethylene glycol since undiluted

DHDECMP is miscible with ethylene glycol.] Wash variables studied intensively included number of washes, wash time and temperature, and ratio of ethylene glycol to DHDECMP solvent. Optimum purification conditions are three equal volume washes at 60° C.

The cyclic alpha radiolysis-solvent wash experiment was completed with very satisfactory results. In the first irradiation cycle a 30 vol% DHDECMP-TCB solvent containing $\sim 2 \times 10^5$ $\mu\text{Ci/liter}$ ^{241}Am and $\sim 0.5M$ HNO_3 was allowed to stand 96 days at 25° C to an accumulated absorbed dose of 16 watt-hr/liter. Over this irradiation period the amount of ^{241}Am retained by the solvent after three equal volume contacts with 0.1M HNO_3 increased from 0.002 to 4.0%. Washing the irradiated solvent once with an equal volume of 5 wt% Na_2CO_3 solution reduced the amount of "unstrippable" Am from 4.0 to 0.047%. In the second irradiation cycle the carbonate-washed DHDECMP solvent was loaded with 6.6×10^5 $\mu\text{Ci/liter}$ ^{241}Am and allowed to stand at 25° C to an accumulated dose of 20 watt-hr/liter. Three equal-volume 0.1M HNO_3 strips removed only about 96% of the Am from the final irradiated solvent; however, after a single 0.47M Na_2CO_3 wash, three such strips removed more than 99% of the ^{241}Am . These tests thus not only provided information about the resistance of DHDECMP to alpha radiolysis but also demonstrated the beneficial effects of periodic carbonate washing in overcoming deleterious radiolysis effects.

Two different bidentate flowsheets are being developed for possible use in the PRF. One, which employs a 40 vol% DHDECMP-TCB extractant, is designed to reduce soluble Am and Pu values in acid waste solution to or below 10 nCi/g. A second flowsheet, which employs a 30 vol% DHDECMP-TCB extractant, is intended for application

with the same flows and equipment used in the present DBBP extraction flowsheet. Extensive mixer-settler tests were completed this report period to develop, demonstrate, and optimize the 40 vol% DHDECMP flowsheet. Mixer-settler runs simulated both extraction and partition column conditions and were executed with 40 vol% DHDECMP-TCB solvent purified either by ethylene glycol washing or by hydrolysis with 6M HCl at 60° C. Flowsheet performance was not dependent on the solvent purification method. With the 40 vol% DHDECMP flowsheet, Am(III) was satisfactorily partitioned from co-extracted Pu(IV) by operating with four mixer-settler stages at 50° C and 0.1M HNO₃ as the partitioning agent. Under these conditions 80 to 90% of the Am accompanied by only 4 to 5% of the Pu reported to the aqueous product stream.

Comprehensive data for the equilibrium distribution of Am(III), Pu(IV), and HNO₃ between aqueous HNO₃-Al(NO₃)₃-HF solutions and purified 30 and 40 vol% DHDECMP-TCB solvents were obtained for use in subsequent computer-assisted flow-sheet calculations.

Evaluation of Effort

Progress toward achieving all the objectives of this research task continues on a steady scheduled course. Bidentate process flowsheets for efficient removal of Am and Pu from acid waste solution--even to the point of producing <10 nCi/g solutions--have been devised and demonstrated in mixer-settler tests. Auxiliary laboratory studies have established a simple, effective wash procedure for purifying technical-grade DHDECMP; these studies have also demonstrated the radiolytic stability of DHDECMP extractants. All in all, the bidentate Am-Pu extraction process is a practical, viable scheme suitable for use in the Hanford PRF.

Future Work

Mixer-settler tests to define and demonstrate satisfactory bidentate process Am-Pu partition (S1) column and Pu-strip (S2) column flowsheet conditions will be conducted with 30 and 40 vol% DHDECMP-TBP extractants purified by both acid hydrolysis and ethylene glycol washing procedures. Initial attempts to apply the computer program SOLVEX with DHDECMP equilibrium Am and Pu distribution data to define bidentate process flowsheet conditions will be made. Work to establish various physical properties (viscosity, flash point, etc.) of 30 and 40 vol% DHDECMP-TCB extractants will be performed.

FRACTIONIZATION

REMOVING CESIUM FROM PUREX CURRENT ACID WASTE

Program Manager:	R. E. Felt
Section Manager:	H. Babad
Principal Investigator:	J. S. Buckingham
Activity Code:	B07T1

Objectives and Scope

Cesium in Purex current acid waste (CAW) has been removed by a phosphotungstic acid precipitation flowsheet followed by ion exchange. The objective of this program is to find an alternative method for removing Cs from CAW.

Prior Work

In the past, phosphotungstic acid precipitation has been used to remove Cs from CAW. Phosphotungstic acid is an expensive reagent and the performance of Cs removal has not been satisfactory.

Progress During Report Period

A laboratory program has been defined for evaluating Cs removal by ion exchange. An alternative method of using a scavenging agent will also be investigated.

Future Work

Ion exchange flowsheets for removing Cs from CAW will be studied. Evaluation of alternative processes will continue.

SLUICING AND B PLANT FEED PREPARATION

Program Manager:	R. E. Felt
Section Manager:	H. Babad
Principal Investigators:	J. E. Horton J. S. Buckingham
Activity Code:	B07T4

Objectives and Scope

The objectives of this activity are to characterize sludges from underground waste tanks and to provide engineering laboratory assistance for the AR Vault/Sluicing program.

Prior Work

Samples of sludge from those tanks scheduled to be sluiced were characterized. The use of flocculating agents to improve liquid-solid phase separation was investigated and used with some success. The flocculating agent, XFS 4111L[®] (Dow Chemical Company) was the best of those tested.

Progress During Report Period

Dow Chemical Company stopped producing the flocculating agent XFS 4111L; therefore a market search was made for a substitute. Fifty-two flocculants were tested in the laboratory on synthetic Purex sludge and supernatant liquid. Six of the flocculants tested were comparable to XFS 4111L. Another three, although less effective than XFS 4111L, had high enough settling rates so they could be used. The six, in order of effectiveness, were S4058[®] (American Cyamid), S4057[®] (American Cyamid), 1130[®] (Betz Corporation), 03-L[®] (Calgon Corporation), 5004[®] (Olin Corporation), and E597[®] (American Cyamid). The other three were 5000[®], 5008[®], and X557[®] (all marketed by the Olin Corporation). It was recommended that the flocculating agents be tested in the AR Vault under actual operating conditions.

Evaluation of Effort

Satisfactory substitutes for the Dow Chemical Company flocculating agent XFS 4111L were found.

Future Work

Goals have been met; therefore no additional laboratory work is planned for this program.

STRONTIUM SOLVENT EXTRACTION

Program Manager:	R. E. Felt
Section Managers:	R. E. Felt H. Babad
Principal Investigators:	J. S. Buckingham D. G. Bouse J. E. Horton S. S. Koegler
Activity Code:	B07T0, B07T1

Objectives and Scope

The purpose of this work is to improve the efficiency and cost of operation of the solvent extraction process for strontium removal from Purex Acidified Sludge (PAS). Further purification of strontium as feed for the encapsulation plant is also included in this activity.

Prior Work

The synergistic effect of tributylphosphate (TBP) was investigated for the solvent extraction of Sr into di(2-ethylhexyl)phosphoric acid (HDEHP). Equilibrium data for strontium, sodium, calcium, iron, and aluminum were determined and incorporated into the solvent extraction computer program SEPHIS. A computer program designed to predict the chelating ability of different chelating agent blends for B Plant solvent extraction was developed.

Progress During Report Period

Laboratory work showed that lead contamination found in purified B Plant Sr product could be removed by a precipitation process.

Additional laboratory work on the effect of chelating agents on B Plant Sr solvent extraction indicated that neither the chelate blend nor the amount of excess chelate used adversely affected the extraction of Sr. The chelate blends tested were mixtures of hydroxyethylenediaminetriacetic acid (HEDTA), ethylenediaminetetraacetic acid (EDTA), and citric acid. Excess of chelates tested were 10 and 30%. Higher metallic decontamination factors were obtained with a larger excess of chelating agent.

Evaluation of Effort

A viable method for removing lead from Sr product was developed.

High Sr losses in B Plant were not caused by substituting the less-expensive citric acid for part of the HEDTA and EDTA complexants.

Future Work

At this time no additional laboratory work is planned on these projects.

STRONTIUM PURIFICATION

Program Manager:	R. E. Felt
Section Manager:	R. E. Felt
Principal Investigator:	S. S. Koegler
Activity Code:	B07T1

Objectives and Scope

The purpose of this work is to decrease strontium waste losses from the Strontium Purification process.

Prior Work

None.

Progress During Reporting Period

Laboratory tests were conducted to improve the first strike of the Sr sulfate purification process. Neither increasing the pH nor increasing the sulfate concentration with sulfuric acid measurably decreased Sr losses. Since improvements to the sulfate purification process itself were not feasible, a process was developed to reclaim the Sr from the first sulfate strike waste. A lead carrier precipitation process was developed which recovered at least 90% of the Sr with good calcium and magnesium decontamination from synthetic first-strike waste.

Evaluation of Effort

The Sr lead carrier recovery process appears to be directly applicable to B Plant operation. A process test will be conducted during the next sulfate purification campaign.

Future Work

This project has been completed.

ENCAPSULATION

CESIUM ENCAPSULATION

Program Manager: R. E. Felt
Section Manager: R. E. Felt
Principal Investigator: J. D. Moore
Activity Code: B08T0

Objectives and Scope

A flowsheet and equipment for achieving desired cesium chloride encapsulation production rates will be developed in tests with full-scale equipment.

Prior Work

Two CsCl solidification processes were selected for parallel development. Design drawings for the tilt-pour melt caster and pan-drier equipment were completed.

Progress During Report Period

The tilt-pour melt caster and pan drier prototype equipment was fabricated and installed for testing. Mock-ups of this equipment to be used for concurrent remote testing have been fabricated and are being assembled along with a remote test station equipped with manipulators.

Mechanical testing with the tilt-pour melt caster prototype has indicated the need for installation of a counterweight to obtain proper balance for tilting operations.

In tests with the pan drier and melt caster prototypes, water boil-up rates of 15 and 4 liters/hr, respectively, were achieved. The melt caster boil-up rate is

unsatisfactory but can be increased by modification and insulation of the off-gas line.

Evaluation of Effort

During the report period the development program predicted completion schedule was delayed by approximately two months. Delays resulted primarily from limited availability of maintenance craftsmen for installation, and machine shop services to respond to needed equipment modifications.

Future Work

Development testing of the tilt-pour melt caster and pan drier prototypes and mock-ups will be completed. The better solidification equipment will be recommended for use in the encapsulation plant. A decision concerning the necessity for remote testing with the more favorable prototype equipment will be made.

CAPSULE DECONTAMINATION

Program Manager:	R. E. Felt
Section Manager:	R. E. Felt
Principal Investigator:	D. W. Jeppson
Activity Code:	B08T0

Objectives and Scope

The purpose of this activity is to provide an efficient in-cell method for decontaminating inner capsules of strontium and cesium.

Prior Work

Decontamination of waste capsules by electropolishing has been demonstrated in the laboratory to be very effective.

Progress During Report Period

Design prints of the electropolishing equipment for in-cell operations were drawn and the equipment fabricated.

A cell mock-up which will be used for cold testing the electropolishing equipment is about 50% constructed.

Evaluation of Effort

The scope of this project was expanded to include construction of a mock-up cell to allow testing of equipment handling with manipulators. Delays in construction of this mock-up has delayed the cold-testing of the equipment.

Future Work

Completion of the cell mock-up and cold testing of the assembled electropolishing equipment are planned.

ALTERNATIVE PRODUCT FORMS FOR ENCAPSULATION

Program Manager:	R. E. Felt
Section Manager:	H. Babad
Principal Investigator:	D. M. Strachan
Activity Code:	B0820

Objectives and Scope

Alternative product forms which exhibit more desirable long-term storage characteristics will be developed for cesium chloride and strontium fluoride.

Prior Work

Initially the direction of this project was to develop a glass formulation using cesium carbonate which would have a low enough viscosity at temperatures less than 900° C to allow glass to be produced by the Encapsulation facility using existing equipment. Several glasses appeared to meet these criteria. However, a quantitative measurement of the viscosity demonstrated these glasses to be unsuitable. It also became apparent that such a product would not be produced at the Encapsulation facility. Thus, while several acceptable glasses were developed, the direction of the project changed and other materials were investigated.

Progress During Report Period

Four waste forms have been investigated. These materials are glass, glass-ceramics, cesium-loaded zeolites, and pollucite ($\text{CsAlSi}_2\text{O}_6$). Glass formulations containing up to 45 mass% Cs_2O have been developed. Some of these glasses were prepared using a simulated Cs_2CO_3 feed from B Plant. All glass-making components (boron trioxide, silica, etc.) were mixed directly with the Cs solution, dried, and melted. Conventional mixing of dry components yielded similar products. Leach rates between 3×10^{-2} and 5×10^{-6} g Cs/cm²-day have been observed in distilled water at room temperature. A cesium-lithium-borosilicate glass had the lowest leach rate.

Glass-ceramics were considered as viable for long-term storage of Cs wastes. Glass-ceramics combine the processing advantages of glass plus the thermodynamic stability of a crystalline product. In this process a mixture of glass-forming materials is melted to form a glass which is then poured and cast. This cast form is then heat-treated to cause crystallization. High crystallinity can be achieved (~90%) and the products are very hard and thermally shock-resistant. Corning Ware[®] (Corning Glass Works) is a typical example of a glass-ceramic. Jet engine turbine parts are also made using a glass-ceramic process. For high Cs wastes pollucite would be crystallized from a glass formulation.

Approximately 25 different formulations were tried and although several yielded very acceptable glasses, none could be crystallized. Not all formulations have been investigated. Leach rates for the resulting glass products were 1×10^{-6} to 1×10^{-8} g Cs/cm²-day in distilled water at room temperature. These rates compare with that of pollucite, 6×10^{-8} g Cs/cm²-day.

If CsCl is removed from stored capsules it may be necessary to remove the chlorine so that it is more acceptable to some future immobilization process. Ion exchange using zeolites could be used not only to prepare the Cs for further use, but also to immobilize it for long-term or final storage. This concept was investigated. Zeolite columns of AW-500[®] (Linde Company) or Zeolon-900[®] (Norton Company) were loaded to 100% breakthrough using a CsCl solution. These were then washed with water, dried, and fired. Leach rates of the resulting products decreased with increasing firing temperature up to 1100° C. Both zeolites gave similar results-- 1×10^{-4} g Cs/cm²-day at 500° C to 3×10^{-6} g Cs/cm²-day at 1200° C in distilled water at room temperature and a cesium oxide content of ~23 mass%.

Pollucite is a very insoluble Cs compound which has existed for millennia. By mixing a CsCl solution with a bentonite clay, then drying and firing the mixture, pollucite can be formed at temperatures as low as 600° C. Little or no loss of Cs was observed during the firing process. Chlorine as an unidentified molecular species was lost between 800 and 1000° C. Leach rates of these products were not determined because of their high porosity. Natural pollucite was found to have a leach rate in distilled water of 6×10^{-8} g Cs/cm²-day at room temperature.

Of the products investigated only cesium glass and pollucite are viable alternatives. The other products were temporarily dropped from consideration because the Cs storage densities were quite low. Cesium chloride has a Cs storage density of 3.0×10^3 kg of Cs/m³ of stored product volume. Glass-ceramics and zeolites had storage densities to 200 to 700 kg Cs/m³. Little improvement could be expected by increasing the density of the zeolite product to theoretical (500 kg Cs/m³). Glass with 45 mass% Cs₂O has a Cs storage density of 1.4×10^3 kg Cs/m³. Pollucite as produced by the above method has a storage density of $\sim 0.2 \times 10^3$ kg Cs/m³. Densification could raise this to 1.2×10^3 kg Cs/m³.

Evaluation of Effort

Work over the past period has demonstrated that Cs can be converted to an acceptable material suitable for long-term and/or final storage. This conversion can be accomplished with either the process feed solution from B Plant, with CsCl after conversion from the carbonate, or after dissolution from the stored capsules.

Future Work

Work over the next year is likely to be curtailed somewhat due to higher priority projects. Attempts will be made to determine the amount of Cs which is volatilized during processing. Cold-pressing of the dried mixture of CsCl and bentonite clay followed by firing will be used in an attempt to increase the density above 90% of theoretical. Some work on the Sr alternatives will also be attempted.

CONCENTRATION, TERMINAL LIQUOR, SALT CAKE, AND SLUDGE CHARACTERIZATION

WASTE MANAGEMENT PLANNING SUPPORT

Program Manager:	R. J. Thompson
Section Managers:	R. J. Thompson H. Babad
Principal Investigators:	J. S. Buckingham R. E. Van der Cook
Activity Code:	B06T0

Objectives and Scope

The program provides waste management planning support for other activities in the Concentration, Terminal Liquor, Salt Cake, and Sludge Characterization programs.

Prior Work

The Waste Management Model (WAMM), a computer simulation of the solidification portion of the Waste Management program, was used to make sensitivity analyses to provide an insight into the parameters which have the greatest impact on the projected waste volumes.

Eight process parameters and seven operational modes of Hanford facilities were varied and the effect of each change was identified by comparison with the model's base case. Results of the initial studies showed that the parameters which cause major variances in the projected volumes are reduction factors, volume of interstitial liquid per gallon of wet salt, and volume of terminal liquid formed per gallon of wet salt. All of these are process-oriented.

Progress During Report Period

The WAMM was updated to reflect the current volumes of dilute liquid, concentrated liquid, and salt cake which are in the tanks. The model was modified to reflect the shut-down of the 242-T Evaporator and also the effects of partial neutralization of the liquid waste; *i.e.*, the reduction of the NaOH content with HNO_3 . The modification of the model for the 242-T Evaporator is operational. The partial neutralization portion is still in the debugging stage.

Additional experiments were undertaken in the laboratories to define more precisely the volume reduction achieved by the evaporators when processing a variety of feeds. The results of these studies may lead to a revision of the boildown factors currently incorporated in the WAMM.

Measurement of the elevation of the domes of three waste storage tanks (103-A, 106-A, and 103-AX) had been taken for a six-month period beginning in August 1975. The maximum difference between successive measurements was 0.0024 meters. This change does not exceed specifications.

An internal report was issued which compiled the total estimated amounts of chemical stored in the underground waste tanks.

The data were assembled based on the flowsheet values for the Bismuth Phosphate, Uranium Recovery, Redox, Purex, and Waste Fractionization processes.

An internal report was issued defining the volumes and composition of specific types of liquid wastes in the waste storage tanks.

Evaluation of Effort

The work carried out within the scope of this project will permit the other activities to be accomplished in a timely manner.

Future Work

The debugging of the partial neutralization option to the WAMM will be completed. Other modifications to the model will be made as required. The effort to characterize the waste tanks contents by sampling, calculation, and inventory determination will continue.

INTERSTITIAL LIQUOR REMOVAL

Program Manager:	R. J. Thompson
Section Managers:	R. J. Thompson H. Babad
Principal Investigators:	W. P. Metz O. B. Schlippacke D. M. Strachan
Activity Code:	B06T2

Objectives and Scope

The objectives are to design, develop, and demonstrate the technology, methods, and equipment to remove as much

interstitial liquid and to immobilize residual liquid as is technically and economically practicable.

Five areas of investigation were identified for the Interstitial Liquid Removal/Immobilization Development program:

- Characterization of the flow of interstitial liquids in salt cake.
- Improvement of salt well pumping equipment.
- Stimulation of flow of interstitial liquids.
- Evaluation of feasibility of residual liquid evaporation as a removal or immobilization technique.
- Evaluation of the feasibility of *in situ* fixation of interstitial liquids.

Prior Work

A mathematical model has been developed which appears to adequately describe the liquid flow in salt cakes.

Instrumented Salt Well facilities for Tanks 241-BY-107 and 241-S-111 were designed, fabricated, and installed for testing. The facilities were to provide sufficient data to verify a model of interstitial liquid flow through salt cake. Data from the 241-BY-107 Instrumented Salt Well facility have been obtained. The 241-S-111 Instrumented Salt Well facility has been installed and data collection initiated.

Dr. Lyman L. Handy, Chairman of the Petroleum and Chemical Engineering Departments, University of Southern California, was retained as a consultant on the program; and his expertise provided an independent approach to the flow characterization problem.

A market survey was conducted among manufacturers of deep-well pumping equipment, and a deep-well jet pump manufactured by Goulds Pumps, Inc., was procured for testing. Cold-testing of a modified commercial deep-well jet pump was completed. The results were sufficiently encouraging to warrant the initiation of design and fabrication of a unit for in-tank installation. A vertical-mount, low-capacity, prototype Moyno Pump[®] (Moyno Pump Division, Robbins & Myers, Inc.) assembly was purchased for testing. The use of a 75.7 liters/min turbine pump (mini-pump) for salt well installations was recommended until a more suitable pump has been proven.

A study was performed on the feasibility of modifying tank bottom configuration to enhance liquid drainage and showed that any such modifications would be very costly. A test to demonstrate the applicability of oil field, shaped-charge jet perforating techniques for the stimulation of salt well flow, and the safety of these techniques, was performed. Work on the imbibition displacement of interstitial liquids from salt cakes was completed. The effects of compressed air displacement of interstitial liquor were evaluated. Neither process was found to be effective for removing interstitial liquid. Bench-scale tests conducted to evaluate the feasibility of vibrating the drained salt cake to free additional liquids for removal produced a significant increase in removable liquid.

A preliminary evaluation of the potential for evaporation to reduce the amount of interstitial liquor was completed. The technique appeared to be potentially beneficial, particularly when applied to sludges.

Progress During Report Period

Data from both the 241-S-111 and 241-BY-107 Instrumented Salt Well facilities were obtained and were used to compare with and evaluate the flow predictions of the mathematical model. A topical report on the model, its application, and the results obtained has been written. A technical assessment of the model has been prepared by Dr. Handy.

The deep-well jet pump as-received and a modified version were tested in the Instrumented Salt Well facilities in Tanks 241-S-111 and 241-BY-107, respectively. The resultant performance of the pumps and the production from the wells during the tests warranted a recommendation to implement the jet pump as standard salt well pumping equipment.

The prototype Moyno Pump was received and installed for cold-testing. Initially some drive shaft bearings problems were encountered. They have been resolved, and cold-testing is in progress. If the tests are successful, the Moyno Pump will provide an alternative pump to the jet pump.

Steam injection into a salt well was found to stimulate the flow of interstitial liquid. Stimulation of flow by oil field shaped-charge jet perforating techniques has been dropped from the program.

A process test proposal for an *in situ* drying test was issued. The test is designed to obtain drying data on both salt cakes and sludges.

Physical data on synthetic salt cake and sludges drying, and the CO₂ immobilization study have been documented.

A salt cake liquid heel immobilization concept, a "capillary pump," has been identified. The concept utilizes

the wicking characteristics of diatomaceous earth to immobilize the heel which is expected to remain after salt well pumping.

Evaluation of Effort

One of the major benefits from the program has been a more thorough understanding of the entire scope of interstitial liquor removal and the physics of the Hanford solidified waste. The impact of that understanding is now being implemented. The theoretical and laboratory aspects of the program have progressed satisfactorily. The hardware aspects (field work and pump development work) had previously been slower than anticipated; however more effort was shifted to this area, and significant progress has been made.

Future Work

During the next report period preliminary results of the *in situ* drying test will be available, cold testing of the prototype Moyno Pump will be completed, and work on the capillary pump will begin.

Reports Issued

D. M. Strachan, *Permeability, Capillarity, and Pore Volume of Synthetic Hanford Salt Cake*, ARH-ST-129, Atlantic Richfield Hanford Company, Richland, Washington, November 1975.

D. M. Strachan, *Effect of Carbon Dioxide on the Permeability of Synthetic Hanford Salt Cake*, ARH-ST-130, Atlantic Richfield Hanford Company, November 1975.

TERMINAL LIQUOR DISPOSITION

Program Manager: R. J. Thompson
Section Managers: R. J. Thompson
R. E. Felt
Principal Investigators: R. I. Donovan
D. R. Frazier
W. P. Metz
W. E. Ogren
R. E. Van der Cook
Activity Code: B06T3

Objectives and Scope

The objectives are to develop and demonstrate on a prototype-scale a process or combination of processes to convert terminal liquids to acceptable solids for interim storage, preferably in existing underground waste storage tanks.

The activity has two principal emphases:

1. Additional volume reduction of terminal liquors in existing evaporators or in a wiped-film evaporator.
2. Partial neutralization of the terminal liquor with nitric acid and subsequent evaporation to reduce liquid volume and produce nonhygroscopic solids.

Prior Work

An engineering study compared alternative terminal liquor disposition schemes in order to identify the most promising based on operability and minimum volumes of final storable wastes generated. The study considered partial neutralization, high-temperature concentration, aluminum removal by the Bayer process, and silica addition. The results indicated that silica addition is more suitable than the Bayer process for the removal of Al. The advantage of high-temperature concentration is that Al removal and

partial neutralization would not be necessary before processing terminal liquors.

Synthetic formulations of current and expected Hanford evaporator bottoms liquors were processed in a pilot-scale wiped-film evaporator designed by Savannah River Laboratory, Aiken, South Carolina. The extensively evaporated products discharged freely out of the evaporator and solidified upon cooling.

A process test of partial neutralization of 242-S Vacuum Evaporator liquor with nitric acid was run during June 1975 and demonstrated that the partial neutralization process is a viable alternative for the reduction of terminal liquors.

Partial neutralization tests were conducted in the pilot-plant vacuum evaporator-crystallizer to:

- Determine if oxides of nitrogen (NO_x) can be generated in the pilot evaporator-crystallizer during partial neutralization under conditions similar to those in the June 1975 242-S Evaporator-Crystallizer process test.
- Compare the effects of different nitric acid concentrations and acid-slurry mixing methods on the generation of NO_x in the off-gas.
- Determine the cumulative corrosion rates on carbon steel resulting during the test.

The tests revealed that: (1) NO_x can be generated in the pilot evaporator-crystallizer during partial neutralization, (2) NO_x generation decreased as acid concentration decreased, (3) NO_x generation could be markedly reduced by improving the dispersal of the acid at the point of injection, and (4) corrosion rates in the evaporator-crystallizer

were essentially comparable to those under normal evaporator operation.

A partial neutralization process test was proposed to demonstrate that acid could be added to the 242-S Evaporator-Crystallizer without causing corrosion in the off-gas system and that acid injection could be performed with only a minimal loss of the evaporator-crystallizer production.

Progress During Report Period

Additional product sample characterizations were carried out on wiped-film evaporator product prepared in the Savannah River Laboratory unit. These included the determination of cooling curves, volume balance verification, and thermal conductivity. The cooling curves will be useful in determining the temperature versus percent solids relationship and, thus, in defining operating conditions for multiple pass mush processing. The volume verification will authenticate the current assumptions used in flowsheet calculations. The effects of feed compositions and operating temperatures on product characteristics have been determined.

The decision was made to terminate work on the clay process as a means of waste solidification.

An HNO_3 partial neutralization process test was carried out at the 242-S Evaporator-Crystallizer during November and December 1975 to demonstrate that the process was an acceptable method of reducing the volume of terminal liquor. During the test 3, 6, and 9M HNO_3 were injected via various injection nozzles into the 242-S slurry to determine the amount of NO_x evolved. A liquor volume reduction of 40 to 50% was obtained. Corrosion in the off-gas system was less than one mil per year.

The design for a partial neutralization prototype was initiated late in 1975.

Tests were carried out in bench-scale evaporator-crystallizer using a high-temperature slurry process. A slurry of aluminum, nitrate, and nitrite solids was produced. However the supernatant was supersaturated in aluminum as might have been expected from the anomalies of aluminum solubility. Operation of the evaporator-crystallizer with a fixed flowsheet at constant pressure produced a unique slurry composition with a characteristic boiling point for that pressure.

Evaluation of Effort

Projections using Savannah River Laboratory data have indicated the applicability of a wiped-film evaporator for use on Hanford wastes.

The partial neutralization process has been demonstrated to offer significant advantages in the process of converting concentrated terminal liquor to acceptable solids for interim storage. A major volume reduction of terminal liquor will be achieved.

Laboratory tests of a high-temperature slurry process have indicated sufficient promise to initiate planning for a large-scale test.

Future Work

The construction and installation of the partial neutralization prototype equipment at the 242-S Evaporator-Crystallizer is scheduled for completion in August 1976, and a prototype test is scheduled for September 1976.

The preparation of functional design criteria for partial neutralization at the 242-A Evaporator-Crystallizer will begin late in FY 1976.

A demonstration test of the high-temperature slurry process will be proposed to convert ~1.1 million liters of terminal liquor to ~900,000 liters of slurry for storage in a double-shell tank.

ALUMINUM REMOVAL FROM HIGH-LEVEL RADIOACTIVE WASTE

Program Manager:	R. J. Thompson
Section Managers:	R. J. Thompson H. Babad
Principal Investigators:	J. S. Buckingham W. E. Ogren D. A. Puryear
Activity Code:	B1420

Objectives and Scope

Aluminum will eventually interfere in the partial neutralization volume reduction program by precipitating in the process and producing a difficult-to-drain, high-liquor content material in the tank. The program objectives, development and demonstration of techniques to remove aluminum from terminal liquor, have the potential of reducing the number of double-shell storage tanks required for interim storage.

Prior Work

The solubility limits of NaAlO_2 , NaNO_3 , and NaNO_2 were determined as functions of temperature and NaOH concentration. Two aluminum removal alternatives--reaction of silica

with the dissolved aluminum and a Bayer process adaptation-- were tested in preliminary laboratories studies. Additional laboratory tests incorporating the results of engineering studies which forecasted anticipated feed compositions and processing rates were carried out.

Progress During Report Period

Engineering studies and laboratory tests provided the bases for a technical review of the proposed aluminum removal processes. A consulting agreement was arranged with Kaiser Aluminum and Chemical Corporation for discussion of the development program and the feasibility of practical application of aluminum removal.

The process chemistry for production of aluminum trihydroxide (gibbsite) by adapting the Bayer process to Hanford waste liquors was considered by the Kaiser consultant to be workable and to give predictable results. The reaction of silica with the dissolved Al in the waste liquor was also considered applicable. Aluminum removal by the Bayer process or reaction with silica would each require an agitated vessel for crystal growth and recycle of fine Al solids to the growth vessel to obtain an acceptable interim storage product. Both processes would have to be carried out on a batch basis to obtain reasonable crystal growth rates. Scaling formation of vessels and piping in contact with the growth would occur.

A review of the number of unit operations needed, the periodic descaling procedures, and the level of process control required to produce material meeting interim storage requirements indicated that the process is not viable. The limiting constraint was found to be the extended time period required to develop and demonstrate a process for interim

storage. A forecast based upon test data indicates that approximately two years would be required to remove one-half of the Al from solutions in a 190,000-liter batch tank facility.

Evaluation of Effort

The results of efforts to date indicate that Al removal from Hanford waste liquors is not applicable to interim waste storage. However, if the length of time for proper development of the Bayer process adaptation were available, application to long-term waste management may prove beneficial. The improved understanding of Al phase behavior in Hanford waste solutions has proved an adjunct to development of other processing options to achieve interim storage of Hanford wastes.

Future Work

No additional work is planned.

SALT CAKE CHARACTERIZATION (ANALYTICAL DEVELOPMENT)

Program Manager:	R. J. Thompson
Section Manager:	H. Babad
Principal Investigator:	V. L. Schuelein
Activity Code:	B06T6

Objectives and Scope

Waste tank salt cake core samples will be analyzed for physical and chemical characteristics to determine chemical inventories in the tank and the physical characteristics of the tank contents.

Prior Work

Preliminary design of a shielded glove box facility was completed and the development of analytical procedures for use on the core samples was completed.

Progress During Report Period

The design and fabrication of the shielded glove box facility continued but is behind schedule. Completion is anticipated in June 1976.

The analysis of a prototype salt cake sample has revealed some problem areas in the chemical procedures. These will be modified where possible to circumvent the difficulties. The procedures and their limitations have been evaluated.

Because of the sensitivity of the semiconductor detectors to radiation, the calorimeters for determining radioactive heat generation rates cannot be calibrated using a radioactive source and they will not be used. The heat generation rates will be calculated from the quantities of the major radioisotopes which are present.

The first sample from Tank 241-TX-116 is being analyzed. As anticipated, the top 0.3 m consisted of diatomaceous earth with the final 10 cm being salt cake. No results are available at this time.

Evaluation of Effort

Although further development of analytical techniques and sample handling methods is anticipated as the sampling program expands, the laboratory is now in a position to receive and analyze samples on a routine basis.

Future Work

The evaluation of the data from the radioactive samples will dictate if additional analytical development is required.

SALT CAKE CHARACTERIZATION (CORE SAMPLER DEVELOPMENT)

Program Manager:	R. J. Thompson
Section Manager:	R. J. Thompson
Principal Investigators:	M. D. Martin G. K. Allen
Activity Code:	B06T6

Objectives and Scope

The objectives are to design, develop, and demonstrate equipment capable of safely and efficiently retrieving representative samples of in-tank, high-level waste.

Prior Work

An integrated system test of the core sampling equipment was conducted to evaluate the equipment's performance under simulated field conditions and to develop contamination control techniques. A soft steel bit, which cannot damage the waste tank steel liner, was successfully used to drill plaster of paris and solid rock salt, but the soft steel would not penetrate portland cement.

The use of a dual probe electrical resistance system was selected as a means of establishing assured electrical contact with the waste tank liner, thereby permitting operations to be terminated prior to liner damage. Tests of the system indicate that actual waste tank experience will be required to establish its workability.

A liquid sampler was designed and fabricated. Tests on water-clay mixtures indicated the sampler will recover thin slurries or liquids. Additional study is needed to sample thick slurries or pastes.

A test plan for in-tank use of the sampler, the sampling flowsheet, the equipment information manual, and operating procedures were prepared. A summary of the expected composition of sludges produced by each of the separations processes used during the past 30 years was issued.

Progress During Report Period

Cold demonstration tests of the sampling system were performed to evaluate operating procedures and equipment and to allow the Hazards Review team to observe the overall operation. Although the system performed satisfactorily, minor changes in the operating procedures were made as a result of these tests. One in particular was the incorporation of a decontamination barrier on the sample receiver shut-off valve to prevent spread of radioactive material when moving the sample receiver from the drill rod to the cask.

The test plan for in-tank sampling and the drilling flowsheet were issued.

The equipment was set up at Tank 241-TX-116 and four samples were obtained from a riser about 1.8 m from the Tank center. The first two samples appeared to be essentially diatomaceous earth which had been added to the tank to absorb liquid. Visual examination of these samples was made in the laboratory, but chemical analyses have not been completed. The second two samples are awaiting laboratory evaluation.

Evaluation of Effort

The core sampling equipment has been tested and found capable of obtaining salt cake samples.

Future Work

The probe for the electrical resistance system that will indicate tank liner contact will be installed. Tests will be conducted on the system during the next corings. However all analyses on the first samples will be completed before further samples are obtained. At that time sampling will be completed in the tank riser currently being used. For a second sampling the drilling complex will be moved to a riser approximately 90° from the first.

TANK OPERATIONAL CRITERIA

Program Manager:	R. J. Thompson
Section Manager:	R. J. Thompson
Principal Investigators:	R. G. Geier R. E. Van der Cook F. R. Vollert
Activity Codes:	B05T0, B06T0

Objectives and Scope

The underground waste storage tanks, designed for storing liquids, will be used for interim (tens of years) storage of salt cake. The objective of this program is to develop criteria for tank usage so that operating specifications and standards can be established for filling the tanks with salt cake.

Prior Work

Thermal creep time-dependent analyses of reinforced concrete Tanks 241-SX and 241-U that are sustaining soil loads were completed for prospective temperature conditions during salt cake storage. The analyses were performed to determine whether self-limiting cracking or creep deformation in the tank structure could be expected under the conditions considered. The finite-element computer program SAFECRACK, developed for such an analysis, was used. Tank temperatures were obtained from heat transfer analyses of the underground tank structures using projected values for the heat generation rates of the salt to be stored in the tank farms. Based on the information generated by SAFECRACK, interim criteria were issued for the use of Tanks 241-U and 241-SX.

Progress During Report Period

Analyses made during this period were used to evaluate the safety factors applicable to the concrete tank structures for a number of tank farms.

- 241-U Tank Farm. If the maximum temperature at the base of the tank cylinder wall is 157° C, the safety factor for the tank dome under 2.1 m of soil would be 4.0.
- 241-BX Tank Farm. If the maximum temperature at the base of the tank cylinder wall is 157° C, the safety factor for the tank dome under 2.4 m of soil is 3.5.
- 241-BY Tank Farm. If the maximum temperature at the base of the tank cylinder wall is 121° C, the safety factor for the tank dome under 2.7 to 2.9 m of soil is at least 2.6.

- 241-SX Tank Farm. If the maximum temperature at the base of the tank cylinder wall is 177°C , the safety factor for the tank dome under 2.0 m of soil is 5.0.
- 241-TX Tank Farm. If the maximum temperature at the base of the tank cylinder wall is 121°C , the safety factor for the dome under 2.9 m of soil is 2.6.
- 241-A Tank Farm. Analyses on Tanks 241-A have been made on the presumption that the concrete tank walls temperatures have not exceeded 177°C , that rapid temperature changes have not been experienced, and that the thermal gradient across the concrete has not been excessive. If this has been the case and the maximum temperature at the base of the concrete cylinder is 177°C , the safety factor for the dome under 2.4 m of soil cover is 4.2.

Evaluation of Effort

Structural analytical results have been obtained and evaluated for 241-U, 241-BX, 241-BY, 241-TX, 241-SX, and 241-A Tank Farms. These data will permit the establishment of criteria for continued use of the tanks.

Future Work

Thermal creep and ultimate load analyses will be performed for Tanks 241-AX with 2.4 m of soil cover and a maximum temperature of 177°C . Soil cover limitations and maximum temperatures will be established for Tanks 241-AY and 241-AX.

WASTE TANK LEAK DETECTION

Program Manager: R. J. Thompson
Section Manager: R. J. Thompson
Principal Investigator: J. A. Atherton
Activity Code: B1440

Objectives and Scope

The objectives of this work are to improve existing waste tank surveillance methods and develop new techniques where needed, to increase our capability to assess waste tank integrity, and to determine modes of tank failure.

Prior Work

The liquid level gauges [Food Instrument Corporation (FIC)] have been incorporated into an automatic data recording system.

Improvement continued in dry well monitoring techniques for external leak detection. Additional dry wells have been drilled. Dry well monitoring by neutron- and gamma-ray logging techniques has continued on a weekly basis.

Progress During Report Period

Tank 241-S-111 was used as the test tank for evaluating various devices having the potential for monitoring liquid level when it is below the surface of the salt cake. Air-purged weight factor dip tubes flushed on a weekly basis by steam and water proved to be the only reliable method tested. The sleeved FIC gauge, installed in Tank 241-S-111, plugged with salt cake at immersed openings and has been temporarily abandoned. The neutron logging technique, although detecting only the liquid level to within ± 1.2 m, may hold promise; and development work on improving the sensitivity of the detection element is continuing.

Gamma monitor probes are being installed around a single-shell tank to demonstrate the efficacy for tank leak detection. The evaluation of using changes in the electrical conductivity of the soils around waste tanks as a means of detecting is continuing.

Of the nine corrosion monitoring probes that had been installed, only eight are still in place. The probe in Tank 241-S-102 was removed for observation of effects so far encountered. However, because of the high radiation level, only visual examination was possible. No evidence of corrosion could be seen.

One measurement by an ultrasonic technique was made to determine the bottom thickness of Tank 241-A-101. The thickness was found to be 375 mils--the same as when the tank was new.

Evaluation of Effort

Determining the level of liquid in a salt cake matrix with bubbler dip tubes, although difficult because the salt cake-liquid system is subject to crystallization of salt cake on slight cooling and consequent pluggage, has proved possible if care is taken to maintain the monitoring equipment.

Future Work

Development of improved interstitial liquid level monitoring devices will continue.

The second corrosion probe will be removed for examination.

A concrete core and steel plate will be removed from the Tank 241-SX-108 wall for evaluation.

CONTAMINATED SEDIMENT CONTROL AND BIOLOGICAL UPTAKE

SITE CHARACTERIZATION

Program Manager: M. W. Legatski
Section Manager: M. W. Legatski
Principal Investigator: K. R. Fecht
Activity Code: B0960

Objectives and Scope

The objectives of this program are (1) to characterize various intermediate- and low-level disposal facilities with respect to the radionuclides and stratigraphy, and (2) to determine the migration or potential migration of radionuclides in the sediments at these facilities.

Prior Work

Sediment and microorganism samples were taken at the 216-S-16 Redox Swamp, 216-S-17 Redox Pond, 216-S-19 Pond, and 216-U-10 Pond using random core and ring sampling techniques. Wet chemistry and gamma-energy analyses of the samples were initiated to determine radionuclide concentrations on the bottoms and along the shoreline for these facilities.

Under ARHCO sponsorship, BNW began development of a screening laboratory to reduce costs of analyzing organisms and pond sediments.

Progress During Report Period

A split tube sampler developed for use in the 216-U-10 Pond was demonstrated successfully. Sediment cores more than 1 m in length were obtained from the Pond bottom. The technique developed assures that no cross contamination of the sediments occurs.

The high cost of analyzing environmental samples has resulted in a survey to identify a reliable and economical method of analysis as well as a laboratory that can handle the number of samples required for the characterization program. Twenty samples have been sent to five laboratories for analysis of 6 radionuclide concentrations. The analytical data will help identify the precision and accuracy of each of the laboratories, as well as the costs.

Work was completed in the development of a screening laboratory at BNW. The objective is to be able to analyze organisms and pond sediments in the nCi/g-range of Pu and Am using lower cost techniques.

A lower cost technique has been developed and verified for determining Pu and Am concentration in sediment and microorganism samples from the 216-U-10 Pond. It was found that in 452 samples analyzed by wet chemistry a ratio existed between Pu and Am. Using this ratio, Pu and Am concentrations have been determined by sensitive and precise measurements of gamma emissions from ^{241}Am using optimal geometry. Approximately 100 back-logged samples have been analyzed at a reduced cost using this technique.

A survey with well monitoring networks was initiated of the vertical distribution of gamma-emitting radionuclides in the vadose zone beneath intermediate-level disposal cribs.

Evaluation of Effort

During the reporting period the site characterization program was being re-scoped to be more responsive to ARHCO and ERDA needs with respect to fission products now stored in the ground. The goal is to identify and measure relative mobility of the radionuclides in the ground beneath the various types of disposal facilities as a function of waste-type and environmental conditions.

The U Pond results tended to substantiate previous results that radionuclides are concentrated in the top few centimeters of soil. The activity levels decrease with increasing distance from U Pond. To complete the sampling in this Pond, additional samples are required from the delta. Additional data may also be needed at depths beneath the Pond.

Future Work

During the next reporting period the following projects are planned:

- The existing data collected from 216-U-10 Pond will be evaluated and a decision made as to what information is needed to complete the Pond characterization.
- The well logging to survey the vertical distribution of gamma-emitting radionuclides will be completed.
- The size and calcium carbonate analyses of sediment samples from crib wells to develop the stratigraphy beneath the crib facilities will be initiated.
- The crib histories will be documented (construction, types and amounts of effluents, etc).

The logs of the radiation profiles from the crib wells will be used as a data base for future logging and radionuclide migration studies. The development of the stratigraphy beneath the crib facilities will aid in well logging interpretation and radionuclide migration potential. The radiation profiles, stratigraphy, and crib histories will be used in determining which crib facilities need detailed characterization with respect to the fission products and the sediments.

SOIL STABILIZATION

Program Manager: M. W. Legatski
Section Manager: M. W. Legatski
Principal Investigator: P. J. Wiater
Activity Code: B09T1

Objectives and Scope

The objective of this program is to provide the capability to effectively control the movement of both surface and subsurface contamination.

Prior Work

Several field tests were completed using asphaltic emulsion to encompass a simulated underground water line leak. At the request of tank farm management emulsion was injected into the ground to immobilize an underground vent line leak at the AX Tank Farm. This leak was located 3 m below grade-level between Waste Tanks 101-AX and 102-AX. A total of 2500 liters of asphalt emulsion were injected into the subsurface through four injection wells evenly spaced around the area to be covered.

A new vibratory drilling tool was secured to drive rods or pipes in the ground.

An experimental trench that simulates a burial trench was constructed for use by ARHCO and BNW. The trench was covered with various media to determine which materials will restrict growth into buried radioactive soil and retard resuspension. Success will be measured by analyzing the plants for lithium which is available to the plants only if the roots penetrate the restriction zone.

Approximately 20,000 m² of land bordering the covered and seeded 216-S-16 Redox Swamp were sprayed with M166[®]

(Dowell Division of Dow Chemical Company). Observation of vegetative growth and soil containment in this area continued into this report period.

Approximately 1200 m² of contaminated S-16 Redox Swamp was affixed with an asphalt emulsion. The area was contaminated with greater than 1 nCi Pu/g of soil. The area was then covered with 1 m of new soils, mulched, and seeded with cheatgrass.

Progress During Report Period

A soil conductivity system was installed at the 102-AX Dresser Coupling[®] (Dresser Industries, Inc.) to evaluate soil conductivity changes during the addition of asphalt emulsion. The readout in millivolts showed positive increases in soil conductivity during the water addition and on the first addition of asphalt. The final two additions showed very little change, indicating that the soil conductivity had stabilized. The 11-02-12 Dry Well that was connected to this system remained stable throughout the test.

Two measurements were made to determine radionuclide movement around the 102-AX Dresser Coupling--one on November 4, 1975; and the other on March 25, 1976--to determine the effect of the leak seal test. A lithium drifted germanium [Ge(Li)] probe was used in the 15.24 cm Dry Wells E-25-112, E-25-128, and E-25-102; and an NaI probe was used to monitor the four injection wells. The readings are reported as gamma activity with the high peak being ¹³⁷Cs. The readings in the dry wells showed no large increase in activity. The readings taken in the four injection wells show that diffusion toward the surface which was occurring prior to and after injection continued to increase in the

November readings. Diffusion upward of activity had stabilized in the March readings. To improve the reliability of the data in the high-activity region of the injection wells a 2.54-cm plastic phosphorus probe will be tried in May.

A gas engine-driven mechanical vibrator is being tested as a quick and inexpensive way to drive piping into the ground. When the vibrator is tuned to the proper frequency, penetration of the pipe into sands and rocky sediments is significantly improved over an air hammer or standard well driving rig. During February several tests were conducted to evaluate drive points on the end of 2.54-cm pipe. Three points were selected--a blunt point, an open pipe, and a sharpened point. Six pipes were driven into rocky soil to a depth of 7.2 m. The 2.54-cm pipe with the sharp point was driven into the ground at a speed of 1.8 m per 30 sec.

Neutron moisture probe readings were started at the S-11 simulated waste burial trench and the 28 concrete culvert test plots located in the 200 West Area. The moisture readings will provide information that BNW will require to carry out their revegetation studies.

Six dry wells (15.24 cm x 3.05 m) were installed at the S-16 Redox Pond--three in the asphalt-treated area and three in the untreated area. These wells will be used as measurement points to determine radionuclide migration below the asphalt and if asphalt can stop the upward migration of radionuclides. Two moisture probe wells were also installed in the soil-filled Redox Pond.

A burn area test was conducted in the 200 West Area. The test was done to evaluate techniques which would keep contaminated sand, ashes, and debris from leaving a fire location. The current test was to seed the burn area with

90% cheatgrass, 5% Siberian Rye, and 5% Russian Rye, followed by a spray affixant (M166) to fix the area until the grasses grow. The site will be studied to evaluate the regrowth of vegetation and affixant stability.

Work began on two proposals associated with grouting. One was to test-grout a well with asphalt emulsion and cement mixture as a possible improvement over currently used cement grouting. A second proposal is to draw up a contract with an established subsurface applicator vendor to demonstrate techniques for rapid and complete large-volume coverage by subsurface injections.

Evaluation of Effort

The conductivity readings from the BNW d.c.-potential system at Tank 102-AX vent leak immobilization test gave an indication when the prewater injections were made but did not respond to all injections with asphalt emulsion. The value of this system will have to be measured in field tests where excavation of the grouting can be correlated with the data readout of the conductivity system.

Probe readings in the wells around the 102-AX Dresser Coupling immobilized leak showed no great increase in activity in the subsurface ground contamination that would necessitate immediate action. More data are necessary to determine if the radionuclides in the soil have been stabilized.

The vibratory drilling test showed that a capped pipe would not penetrate a rocky surface, and the open-end pipe was driven 7.2 m into the rocky soil with considerable effort. The pipe with the sharp drill point was driven into the rocky soil at a speed of 1.8 m in 30 sec. This drilling technique has a potential for supplying a quick and low-cost

method of installing monitoring wells, injection points for grouting, segmented electrodes, and core sampling.

Future Work

The Tank 102-AX leak immobilization test will continue to be studied so a formal evaluation can be made on the success of the subsurface affixation of the leaking Dresser Coupling.

Vibratory drilling techniques will be established for drilling injection points for grouting. Two different types of grout will be tried in the field (asphalt emulsion and silica) to simulate sealing a leaking line. Vibratory drilling of a 15.24-cm casing and a 7.62-cm diameter plastic pipe into a semirocky sediment will be attempted.

Russian Thistle studies will be started in the S-11 simulated waste burial trench to evaluate the effectiveness of the various Bio-Barriers.

The S-16 Redox Pond will be studied for radionuclide movement in the asphalt-affixed versus the nonaffixed area.

The growth of vegetation on the burn area will continue to be studied in FY 1977. The study will look at the cheat-grass now established in this area and determine if it has sufficient growth to propagate itself in October.

A sodium silicate-calcium chloride grouting field test will be performed to simulate stabilizing the radionuclides in the soil around a tank. An offsite vendor will be contracted to do the work.

Greenhouse studies will be started to determine the effects of surface affixants on seed germination. Three drought-resistance grasses and three chemical affixants will be tested under a variety of conditions. This work will interface with the Biological Uptake program.

Stability of a moving sand dune with a chemical affixant will be evaluated.

DECONTAMINATION/IMMOBILIZATION

Program Manager:	M. W. Legatski
Section Manager:	M. W. Legatski
Principal Investigator:	G. M. Holter
Activity Code:	B0970

Objectives and Scope

The objectives of this program are to develop concepts for cleanup of contaminated vegetation, sediments, effluents, and burial grounds; then immobilizing and storing the retrieved material.

A plan of action for treatment of contaminated areas will include use of characterization data and a hazard assessment to develop criteria, equipment, and techniques for removing and immobilizing radionuclides.

Prior Work

Approved prints were completed for the modification of the mobile vacuum system.

Several systems which could be used for pond cleanup work were studied.

Work began on reviewing alternative radionuclide removal systems for liquid effluents.

Progress During Report Period

Modification of the mobile vacuum system was initiated. The spray system and boom systems were installed. The work

is about 90% complete, awaiting fabrication and installation of the vacuum nozzles.

Alternative methods to the Mud Cat[®] (National Car Rental System, Inc.) system for pond cleanup were defined. The alternatives include other dredging systems, as well as removal of pond sediments by earthmoving equipment after allowing the pond to dry.

Sediment samples were sent to Sala Magnetics, Inc., of Cambridge, Massachusetts, for magnetic characterization studies. Their report indicates that magnetite would have to be separated from the soil, using a normal magnetic system, before radionuclide removal. Then, theoretically, PuO₂ could be removed by a high-gradient magnetic system (such as that developed by Sala and consultants from the Massachusetts Institute of Technology).

A characterization study of the 200 West Laundry effluent was initiated. Battelle-Northwest is conducting the study, and will provide a summary of treatment alternatives and design data for a treatment facility.

Evaluation of Effort

Several problems associated with using the mobile vacuum system for cleanup of contaminated areas have been identified. The modifications in progress are designed to correct some of these deficiencies.

A more complete evaluation of the pond cleanup problem is needed before equipment and method of operation decisions are made. To allow time for this evaluation and because of the priority of other items, work in this area has been deferred.

The Sala Magnetics' report concerning magnetic separation of radionuclides from soils needs further study before recommendations are made.

Laundry effluent characterization has received priority due to the fact that funds for the construction of a waste treatment facility have been entered in the FY 1977 budget.

Future Work

Modifications to the mobile vacuum system should be completed shortly. Cold tests will then be run to evaluate filter integrity and to identify any other necessary modifications. After these tests (and subject to the scheduling of further development work), the unit will be available for work in uncontaminated areas.

Laundry effluent characterization is scheduled to be completed in September 1976. To verify feasibility, this will be followed by laboratory tests of the candidate treatment method. Data from these tests will be used in design of a treatment facility.

Pond cleanup work will continue in FY 1977, when it is expected that characterization data necessary for adequate evaluation of the problem will be available.

Studies for handling contaminated vegetation and other bulky wastes (*e.g.*, by incineration) will begin in FY 1977. The objective is to reduce the volume of wastes being sent to the burial ground.

RADIONUCLIDES IN SOIL-PLANT SYSTEMS

Program Manager: M. W. Legatski
Section Manager: M. W. Legatski
Coordinating Biologists: V. A. Uresk
E. L. Klepper, BNW
Principal Investigator: R. C. Routson, BNW
Activity Code: B09TE

Objectives and Scope

This program provides quantitative information concerning the ecological fate and behavior of radionuclides in the soil-plant system. These data will be used in the formulation and maintenance of ecologically sound waste management practices.

Prior Work

Growth chamber studies showed that the chemical form of transuranic wastes affects their uptake by plants from Hanford soils. Plant uptake of transuranics increased in the presence of the chelates ethylenediaminetetraacetic acid (EDTA) and diethylenetriaminepentaacetic acid (DTPA) and salts of several organic acids, probably resulting from the increased supply of soluble-chelated elements to plant roots.

Roots growing through a soil layer of transuranics transported the nuclear material downward, suggesting that the roots are a major transuranic sink while shoot concentration is limited.

Tumbleweed uptake of ^{90}Sr and ^{137}Cs was measured as a function of soil radionuclide concentration and plant age. Cesium and strontium uptake were measured over three and five orders of magnitude of soil concentration, respectively,

at 8 and 12 weeks of plant age. Concentration factors (ratio of $\mu\text{Ci/g}$ in the plant to the soil on an oven-dry basis) were found to be independent of soil concentration over these broad concentration ranges. Plant uptake was about 300 times higher for Sr than for Cs at both 8 and 12 weeks. Concentration factors were found double for both Sr and Cs between the 8- and 12-weeks harvests.

Progress During Report Period

Plant uptake by tumbleweed and cheatgrass was measured for five Hanford soils at the initial soil concentration of 1.0 pg/g of ^{99}Tc traced by $^{95\text{m}}\text{Tc}$ (metastable, γ -emitter) added as TcO_4^- . Samples were harvested at 1-, 2-, and 3-month intervals. Uptake at the 3-month harvest ranged from 23 to 82% and 10 to 69% of the added Tc for tumbleweed and cheatgrass, respectively. The soil order of uptake by tumbleweed was Lickskillet < Burbank < Warden < Rupert < Ritzville.

The soil order of uptake by cheatgrass was Burbank < Lickskillet < Rupert < Warden < Ritzville. Shoot concentration of ^{99}Tc ranged from 0.06 to 0.33 ng/g and from 0.05 to 0.36 ng/g for tumbleweed and cheatgrass, respectively. Shoot concentration generally decreased with increasing time of harvest, probably due to the reduction in soil concentration. Concentration ratios (CR) were calculated based upon the estimated soil concentration during each harvest period. The CR values ranged from 76 to 357 and from 54 to 421 for tumbleweed and cheatgrass, respectively. The CR values were generally constant with increasing harvest time except for the Lickskillet soil which decreased from the first to the third month for both tumbleweed and cheatgrass.

In a separate experiment the tracer $^{95\text{m}}\text{Tc}$ was increased by a factor of 50 over that used in the primary experiment.

Good agreement was found to the percent uptake, shoot concentration, and CR values between high and low ^{95m}Tc tracer concentrations. This demonstrated that no measurable Tc toxicity or radiation effects were measurable in the experiment and that ^{95m}Tc is a good tracer for the ^{99}Tc .

An experiment was initiated to measure the biological availability of Sr and Cs as a function of soil properties and waste composition. The study has been delayed due to loss of part of the samples during vandalism in the laboratory.

A greenhouse has been completed and is now available for use. Special equipment suitable for use in studies with radionuclide-contaminated soils has been received for use in the greenhouse.

Evaluation of Effort

Concentration factors determined for ^{137}Cs , ^{90}Sr , and ^{99}Tc will be useful in predicting plant uptake of these radionuclides by measuring their soil concentrations. The nearly two-fold increase in CR values from 8 to 12 weeks for both ^{137}Cs and ^{90}Sr suggests that plants whose roots have the potential for invading buried waste should be eradicated early in the growth cycle. The high uptake values for ^{99}Tc suggest that its concentration in the groundwater and biota should be monitored periodically.

Future Work

Work will continue to measure the biological availability of ^{90}Sr , ^{137}Cs , ^{239}Pu , and ^{241}Am as a function of soil properties and waste composition. These data will be used to develop predictive capability for estimating biological availability on the basis of simple chemical tests.

Previously contaminated soils will be used to test and verify the developed predictive equations.

A new project to be initiated in Transition Year 1976 will be concerned with determining if herbicides used in the 200 Areas waste sites may form complexes with radionuclides present in the soil, thus leading to enhanced uptake by plants.

RADIONUCLIDES IN TERRESTRIAL PLANT-ANIMAL SYSTEMS

Program Manager:	M. W. Legatski
Section Manager:	M. W. Legatski
Coordinating Biologists:	V. A. Uresk E. L. Klepper, BNW
Principal Investigators:	<u>BNW</u> J. F. Cline R. E. Fitzner K. A. Gano J. D. Hedlund E. L. Klepper W. H. Rickard L. E. Rogers D. W. Uresk
Activity Code:	B09TE

Objectives and Scope

The purpose of these studies is to provide information about the ecology and behavior of plants and animals inhabiting waste management zones and their actual or potential role in exposure, concentration, transport, or dispersal of radioactive wastes.

Prior Work

Events associated with the penetration, over 12 years ago, of buried radioactive wastes by animals resulted in the dispersal of Cs and Sr over several thousand m² of land within the Hanford Reservation.

Study sites near the contaminated area and near a former radioactive waste pond were selected for radioecological studies of the vegetation, insects, and small mammals. Initially ecological characterization studies of the two sites were conducted. Characterization of the vegetation provided information concerning species frequency, shrub size, canopy cover, micro- and macronutrient composition, and soil composition.

The invertebrate populations were also studied for two years to provide information about species, abundance, and food habits.

Habitat requirements, food habits, burrowing behavior, position in food chain, and potential for long-distance dispersal of contamination was summarized for the 25 species of mammals living on the Hanford Reservation.

Sampling of small mammals for radiochemical analysis was begun in April 1975 at five areas near U Pond representing different habitat types.

A study of the birds of prey on the Hanford Reservation was begun in April 1975. The objectives were to determine their diet, body burden level, nesting density, reproductive success, resident status, hunting range, and migration pattern.

A program was initiated to characterize the plants growing in burial grounds of different ages and soil types. Among the five burial grounds sampled, cheatgrass and

tumbleweed were the most abundant plants, although a total of 19 species of plants was present.

A study was established to assess the potential for invasion of buried wastes and subsequent uptake of radionuclides by roots of specific plants. The deepest rooted plants found were sagebrush, rabbitbrush, tumbleweed, and hopsage which rooted to depths exceeding 2 m.

Vegetative cover plots were established in November 1974 on soil fill overlying a portion of a former radioactive waste pond to determine treatments beneficial in establishing a dense, self-perpetuating vegetative cover which would protect the soil against wind erosion and prevent invasion by undesirable deep-rooted plants. A bentonite/straw combination was most effective in establishing a cheatgrass cover.

Progress During Report Period

A study of birds of prey has shown that 21 different raptor species occur on the Hanford Reservation. Ten of these species nest here, while the remaining 11 are either winter visitors or migrants. The Swainson's Hawk is the most abundant diurnal nesting raptor, while the Great Horned Owl appears to be the dominant nocturnal raptor.

Dietary analyses for the Swainson's Hawk nestlings have revealed that snakes make up 61% of the diet, while small mammals comprise 20% and small birds 18% of the diet.

Infertile eggs taken from raptor nests and raptor bones were radiochemically analyzed; the results were not significantly different from levels found in birds not associated with the waste management facilities.

A study of the species composition, abundance, and food habits of grasshopper populations in two areas on the 200 Areas Plateau was completed. Seventeen species were found within the study areas:

Although 28 species of vascular plants were discovered in the study areas, only 7 species were major components in the grasshopper diets. Big sagebrush was the plant most heavily utilized (41%), followed by green rabbitbrush and turpentine cymopterus (15%), Carey's balsamroot (13%), Jim Hill mustard (7%), and western tansymustard (4%). Some plants were apparently avoided since they were encountered at a much lower diet frequency than would be expected based on their natural abundance. Included in this group was cheatgrass, the most abundant plant and one receiving considerable attention as being suitable for revegetation of disturbed sites in the 200 Areas.

A partial listing of insect species inhabiting the 200 Areas Plateau was completed, showing identification of 123 species of insects to date. This number will probably double as further classifications are received.

Sampling of mice in five different habitat types near U Pond has shown differences in body-burden levels as a function of species and habitat type, probably reflecting differences in feeding and watering behavior. Detectable ^{137}Cs has been found primarily in the kidney, muscle-bone, lung, liver, testes, and gastrointestinal tract fractions. At this time only 2 of 18 composite lung-liver samples have shown detectable Pu.

Thermoluminescent dosimeters (TLD) were implanted beneath the skins of native mice which were then released to the U Pond. The mice will be retrapped in the Fall and the TLD analyzed radiochemically. Readings from the TLD will be

compared with the mouse tissue radioanalysis data to determine if a correlation factor exists.

Evaluation of Effort

Documentation of the studies ecologically characterizing the vegetation, mammals, and insects near two representative radioactive waste sites is nearly complete. Potential radiocontaminant pathways through the food chain are being identified by dietary analysis of black-tailed hares, birds of prey, common insects, and selected waterfowl. Techniques for assessing dose rate to native animals have been developed. Body-burden levels of animals living near a radioactive waste pond have been determined.

Future Work

Radiotelemetry will be used to delineate the territories and hunting ranges of birds of prey on the Hanford Reservation. A study of the burrowing behavior of 200 Areas animals will be initiated. Deer fecal pellets will be collected in the vicinity of radioactive waste sites to determine dietary and radiological composition. Characterization of species and radionuclide levels in plants growing on burial grounds will be continued.

Reports Issued

J. K. Sheldon and L. E. Rogers, *Grasshopper Populations Inhabiting the B-C Cribs and Redox Pond Sites, 200 Area Plateau, United States Energy Research and Development Administration's Hanford Reservation*, US-ERDA Report BNWL-1943, Battelle, Pacific Northwest Laboratories, Richland, Washington, February 1976.

RADIONUCLIDES IN AQUATIC BIOTA AND SEDIMENTS

Program Manager: M. W. Legatski
Section Manager: M. W. Legatski
Coordinating Biologists: V. A. Uresk
E. L. Klepper, BNW
Principal Investigators: R. E. Fitzner, BNW
R. G. Schreckhise, BNW
Activity Code: B09TE

Objectives and Scope

The purpose of these studies is to define the pathways of radioactive materials previously discharged to a cooling water pond through the aquatic food web, including waterfowl.

Prior Work

Experimental goldfish confined to areas over sediments of relatively high radioactivity (~ 24 ng/g) accumulated over twice as much ^{137}Cs as did fish confined over sediments of relatively low activity (~ 12 ng/g). A study on radioactivity in native and experimental biota at Gable Mountain Pond showed that the main source for uptake was from ^{137}Cs associated with the Pond sediments. Measurement of dose rates by TLD placed at the sediment-water interface ranged from 4 to 230 mR/day.

The ^{137}Cs concentrations in experimental ducks restricted to the Pond were approximately the same as for resident coots, but higher than those for transient waterfowl.

A 2.5-year survey of waterfowl usage of radioactive waste ponds showed that 126 species of birds were observed

using the ponds and their associated vegetation. Waterfowl feeding habits offer the opportunity for ingestion of radionuclides from the vegetation and aquatic sediments. They have the potential for migrating away from the area and becoming part of the human diet during the hunting season.

A study determined the seasonal body-burden levels and dietary habits of coots living on Gable Mountain, B, and U Ponds. Residence times and migratory distances were studied by banding the birds with U.S. Fish and Wildlife Service tags.

The first year's average concentrations for total body burden levels of adult coots collected at Gable Mountain, B, and U Ponds were approximately 155, 12, and 97 pCi/g, respectively; while control coots sampled at the Columbia Wildlife Refuge near Othello, Washington, averaged background ^{137}Cs concentration levels of 0.26 pCi/g.

Progress During Report Period

The field sampling portion of a two-year study of seasonal body-burden levels and diets of coots living on radioactive waste ponds was completed. During this report period 30 coots were collected at Gable Mountain, B, and U Ponds while 7 control coots were collected at the Columbia Wildlife Refuge.

The average whole body counting results for ^{137}Cs were 131, 39, and 6.4 nCi/total body count for Gable Mountain, B, and U Ponds samples, respectively. When the same samples were counted with the gastrointestinal tract and contents removed, the average whole body counting results for ^{137}Cs were 61, 23, and 4.3 nCi/total body for Gable Mountain, B, and U Ponds, respectively. The average total body burden for ^{137}Cs in the control coots was 0.058 nCi. Comparison of control values with values from samples with the

gastrointestinal tract removed showed that the total body-burden levels for ^{137}Cs were 1052, 397, and 74 times those of the controls for Gable Mountain, B, and U Ponds samples, respectively. Equations used to assess dose rate to man show that a human adult consuming one Gable Mountain coot per day would reach an equilibrium level of less than 3% of his maximum permissible body burden.

Radiochemical analysis to determine Sr concentrations in coot bone showed values of 2.94 and 4.67 pCi/g for Gable Mountain and B Ponds samples, while the control was 0.627 pCi/g. Analysis of coot liver samples for total Pu showed values of 0.026, 0.103, and 1.88 pCi/g for Gable Mountain, B, and U Ponds, respectively.

Evaluation of Effort

There does not appear to be any seasonal cycling of radionuclide levels in coots. Most probably this can be explained by the fact that the availability of radionuclides in coot dietary items does not change significantly throughout the year. Differences in total body levels of ^{137}Cs between coots with and without the gastrointestinal tract indicate that approximately half of the whole body radiation results from the presence of ingested contaminated diet items and associated sediments.

Future Work

Analyses of the remaining samples collected during this study will be completed. Identification of the coots' major food items will be made by examination of the contents of stomachs saved during the field collections.

During Transition Year 1976 the effects of chronic irradiation on waterfowl productivity will be made by comparing clutch size and nesting success on the 200 Areas ponds with that found in a control area.

ACTINIDE SEDIMENTS

ACTINIDE TRENCH CHARACTERIZATION

Program Manager: M. W. Legatski
Section Manager: M. W. Legatski
Principal Investigators: S. M. Price
M. M. Kiilsgaard
Activity Code: B0965

Objectives and Scope

The objective of this program is to characterize the chemistry and distributions of the actinides and their compounds, particularly plutonium and americium, present in sediments underlying liquid waste disposal facilities in the 200 Areas of the Hanford Reservation. The scope includes the following tasks:

- Locate the zones of greatest actinide concentrations and assess their nuclear reactivities.
- Delineate the distribution of actinides beneath the facilities.
- Characterize the mineralogical locations of actinides within the sediment samples.
- Characterize the potential migration behavior of Pu and Am in the ground under present and possible future environmental conditions.
- Determine the immediacy and priority of implementing migration control measures for approximately 20 trenches containing the highest actinide inventories.

Prior Work

The Actinide Trench Characterization program was formally initiated in FY 1973. Since then, three test holes

have been drilled: one through the bottom of the 216-Z-9 Trench and two through the bottom of the 216-Z-1A Trench. All three wells were drilled under conditions of total containment in order to prevent the release of actinides to the uncontrolled environment.

Assessment by neutron pulsing of the reactivities at both the 216-Z-9 and 216-Z-1A Trenches indicates that the probability of attaining criticality at these sites is essentially nonexistent if they are left in an undisturbed state.

Analyses of samples acquired by drilling revealed that the greatest measured concentration of ^{239}Pu , $\sim 10^6$ $\mu\text{Ci/liter}$ of sediment, occurs just below the bottom of both the 216-Z-9 and 216-Z-1A Trenches. This concentration decreases to $\sim 10^3$ $\mu\text{Ci } ^{239}\text{Pu/liter}$ of sediment (approximate limit of ^{239}Pu detection) at the 10-m depth. The ^{241}Am levels decrease similarly from a maximum concentration of $\sim 10^5$ $\mu\text{Ci/liter}$ of sediment just below the bottom of both facilities to < 10 $\mu\text{Ci/liter}$ of sediment at the 10-m depth. Studies have indicated that the detection of actinides below the 10-m depth requires new sample preparation facilities and the utilization of a more sensitive analytical technique.

Alpha-sensitive autoradiography of relatively undisturbed sediment cores from the 216-Z-9 and 216-Z-1A Trenches revealed two types of Pu occurrence--the "particulate" and the "nonparticulate." The particulate-type consists of discrete PuO_2 particles varying from 5 to 100 μm in diameter. These particles were released to the trenches as part of the original waste stream and were trapped in the upper 1 m of the sediment column by filtration. The nonparticulate-type of Pu was originally disposed of as soluble Pu(IV) . This second type penetrated deeper within

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the sediment profile and was deposited in association with silicate hydrolysis of the sediment fragments. Results are discussed in more detail in ARH-SA-232 (see "Reports Issued"). Tests were conducted to determine the level of background contamination in the containment hood. The results indicated that the detection of actinide contamination below certain values would require new hood facilities and the utilization of a more sensitive analytical technique.

Sludge from Tank 361-Z was sampled using a compartmentalized valve sampler. The sampler proved to be highly matrix-dependent and was not adequate for obtaining a representative sample from certain locations in the tank.

Progress During Report Period

The first three months of the report period were primarily spent in an effort to provide new tools capable of obtaining representative samples of the contents of Tank 361-Z. Two tools were designed and constructed. These tools were subsequently tested to verify their capabilities in bentonite mixed to simulate sludge from Tank 361-Z. Although some necessary design modifications were revealed by the tests, both tools appear to be capable of obtaining representative samples from the Tank.

Characterization studies were initiated at the 216-Z-12 Trench (Pu inventory of ~25 kg) during the second half of the report period. Totally contained drilling of the first test well through the bottom of the trench was completed to a depth of 15 m (8.3 m below the point of waste release). Actinide contamination utilizing field instruments was not detected in sediments sampled below a depth of 11 m (4.3 m below the point of waste release).

Consequently, further deepening of the test well was scheduled to be carried out without utilizing the glove box. The glove box was moved to the 216-Z-1A Trench where totally contained drilling through the bottom of the "b" section was subsequently initiated.

Initial analyses of samples from the test well by the Chemical Technology Laboratory (CTL) indicate that the highest concentration of Pu occurs within the first meter below the bottom of the facility ($\sim 10^5$ $\mu\text{Ci } ^{239}\text{Pu/liter}$ of sediment). These analyses also indicate that low levels of this actinide (~ 1 $\mu\text{Ci } ^{239}\text{Pu/liter}$ of sediment) have penetrated to a depth of at least 6 m below the bottom of the facility. Sediment samples containing < 10 $\mu\text{Ci } ^{239}\text{Pu/liter}$ of sediment are scheduled to be sent to an offsite laboratory to confirm the reported concentrations.

Five core samples obtained during totally contained drilling of the first test well at the 216-Z-12 Trench were submitted to BNW for characterization of sediment-waste associations. These relatively undisturbed samples have been impregnated and polished sections prepared. Autoradiography of these samples has been initiated to determine the microdistribution of the Pu within the sediments.

Subsequential leaching studies, designed to determine the "bonding strength" of actinides in contaminated trench samples, have been initiated by BNW. Columns of sediments from a sample from the floor of the 216-Z-9 Trench have been leached in triplicate utilizing the following succession of agents: water, magnesium chloride, diethylenetriaminepentaacetic acid, hydrogen peroxide, oxalate-oxalic acid, and nitric acid. Preliminary results from this first test have been received for evaluation.

Three dual-walled core barrels (designed to provide access holes for neutron pulsing instruments) have been completed by the Diamond Drill Company of Spokane, Washington. The newly constructed core barrels are designed to be driven with a cable tool rig.

Specifications for new sample preparation facilities were completed by CTL personnel. Two hood facilities were subsequently placed on order--a compartmentalized hood and an open-faced hood. Utilization of such facilities is required in order to control background levels during sample preparation.

Evaluation of Effort

Tests indicate that two tools have been developed which are capable of obtaining representative samples of sludge such as found within Tank 361-Z. Provision of onsite capability to obtain such samples was accomplished during the report period.

The first relatively undisturbed core samples of sediment underlying the 216-Z-12 Trench were acquired. Preliminary analyses of these samples indicate that the bulk of the Pu lies just below the point of waste release, similar to the distributional patterns encountered at the 216-Z-9 and 216-Z-1A Trenches. The availability of such samples allows the first examination of the microdistribution of Pu in sediments from a trench which received high-salt wastes. Previously examined samples were from the 216-Z-9 and 216-Z-1A sites, trenches which received only acidic wastes.

Efforts to determine the bonding strength of the Pu in trench samples were first initiated during the report period. Evaluation of the results of such efforts is

expected to allow a more adequate definition of the direction of future leaching work.

The provision of dual-walled core barrels which can be driven utilizing a cable tool drilling rig rather than a pile driver is expected to reduce installation costs. Unlike the pile driver, provision of a cable tool drilling rig is available through an onsite contractor.

Efforts by CTL personnel to design and acquire new sample preparation facilities should allow the reliable preparation of samples from below the 10-m depth as soon as such facilities are available for use.

Future Work

Sampling of the contents of Tank 361-Z will be carried out utilizing the samplers developed during the report period.

Completion of totally contained drilling through the bottom of the 216-Z-1A (b section) Trench is scheduled. In addition, two wells will be drilled utilizing a cable tool rig and open drilling, to the water table adjacent to the 216-Z-12 and 216-Z-1A Trenches. Water samples from these wells will be analyzed for actinide content.

Dual-walled core barrels will be installed within the 216-Z-1A and 216-Z-12 Trenches and pulsing carried out.

Installation of the compartmentalized and open-faced hoods by CTL personnel is scheduled.

Report Issued

S. M. Price and L. L. Ames, *Characterization of Actinide-Bearing Sediments Underlying Liquid Waste Disposal Facilities at Hanford*, US-ERDA Report ARH-SA-232, Atlantic Richfield Hanford Company, Richland, Washington, September 1975.

HYDROLOGY

GROUNDWATER HYDROLOGY

Program Manager: R. A. Deju
Section Manager: R. A. Deju
Principal Investigators: R. C. Arnett
R. E. Gephart
R. K. Ledgerwood
Activity Code: B09T2

Objectives and Scope

The goals of the work are to characterize the present and future groundwater flow regimes of the Hanford Basin. This will involve characterizing the present and future flow regimes of the confined and unconfined aquifers, determining the capacity of the present well monitoring network to define present and future water tables, and defining and characterizing the major lithologic units controlling groundwater movement.

Prior Work

A controller program was written to display the Hanford map on the Tektronix 4014[®] (Tektronix, Inc.) terminal and run travel time programs including the generation of instantaneous streamlines from points selected using the terminal's cursor.

A study was conducted to predict changes in the water table over the period 1975-1995. This study was coupled with a prediction-verification study where the same mathematical model was used to predict 1968-1973 water table levels using 1968 data as input. Seven areas where the difference between the predicted and observed groundwater potential for 1968-1973 surfaces exceeds 1.5 m were

identified. In general, the areas of high difference coincide with areas of low data density or steep gradients in the water potential. Most often the model-predicted results are lower than the observed values, indicating low estimates for some inflow rates as suggested by consultants Mr. W. K. Summers and Dr. R. A. Deju.

Progress During Report Period

Field verification of structural data for 200 area wells structures was initiated. The purpose of this work is to field-locate, determine depth to bottom, and structural integrity of approximately 200 wells. This information will be used in preparing the well inventory. The final report on this task is scheduled for distribution in July 1976. The object of this report will be to recommend repairs and maintenance of selected well structures, establish a standardized groundwater monitoring network, and recommend sites for drilling new monitoring wells. A review of all monitoring wells in the 100, 300, and 600 Areas is continuing; the report on these structures will be issued before the end of Transition Year 1976.

Results of measurements of water levels taken in 300 monitoring structures during the month of December were used in preparing maps of the Hanford water table and depth to water maps for the separations areas. Water level measurement data indicate a drastic rise over the last 12 months in the 200 West Area mound in the vicinity of U Pond. The rise averages 2.5 cm per week and is being investigated.

A contract to drill five wells in three new pumping test sites went out for bids in January. The contract was awarded to Aqua Drilling and Development Company, Inc., of Spokane, Washington; and operations started on March 1, 1976.

A pathline has been developed, coded, and implemented on the computer. Work was initiated to conduct a systematic error analysis and sensitivity study on this model. If it appears adequate, pathlines will be run on the 20-year Hanford Variable Thickness Transport (VTT) model; and the results of travel time information using pathlines will be compared with the results using instantaneous streamlines.

The VTT groundwater flow model sensitivity analysis is continuing. Work continued on quantifying the error introduced by simplifying assumptions and analyzing its sensitivity to boundary and initial conditions.

The electric log study continued on schedule. A report is being prepared discussing the definition of environment of deposition and hydraulic properties based on the results of logging work conducted last fiscal year.

Work was initiated to conduct verification studies on the transport model (MMT), and in refining the transmissivity program so that it can operate on the PDP-11/45[®] (Digital Equipment Corporation) computer.

Evaluation of Effort

All work is on schedule.

Future Work

During the coming six months three additional pumping tests will be conducted in the wells that have been drilled. These tests will serve to adequately determine the hydrological characteristics of the aquifers underlying the Hanford Reservation. The effort will concentrate on conducting simulations using hypothetical scenarios.

OFFSITE MIGRATION

Program Manager: R. A. Deju
Section Manager: R. A. Deju
Principal Investigators: R. C. Arnett
R. E. Gephart
R. K. Ledgerwood
Activity Code: B09T3

Objectives and Scope

The objectives of this program are to identify and sample with a minimum number of core holes the confined aquifers underlying the Hanford Reservation, to accurately analyze the groundwater samples utilizing the best analytical techniques available and determine the possible source of all radiological contamination detected, and to evaluate the ultimate fate of all long-lived radiological contamination detected in the confined groundwater flow systems.

Prior Work

This is a new program and has been initiated during the current report period.

Progress During Report Period

Plans for the Offsite Migration program were drafted. The program plan consists of a development of the number and location of additional monitoring wells, required analyses, analytical procedures, and sampling methods. Emphasis is being placed on standardization of analytical procedures.

Work was initiated in February to evaluate the feasibility of constructing a preliminary data base to apply a modified version of existing transport codes to the topmost confined aquifer. This investigation will result in a

recommendation of the most feasible approach to the modeling problem and documentation of the available data.

All data on previous studies on the confined aquifer have been analyzed and a report has been issued. This report, ARH-SA-253 (see "Report Issued"), together with the program plans will form the bases for future work in subsequent report periods.

Evaluation of Effort

All work is on schedule.

Future Work

Work scheduled for the coming six months includes an assessment of analytical procedures which may be used for low-level analyses of groundwater from the confined aquifers.

If routine water sampling of the confined aquifers detects above background levels of long-lived radiological contaminants (e.g., ^{129}I), their concentration and areal extent will be determined. Sources of contamination will be identified using the deep well monitoring network.

Report Issued

R. K. Ledgerwood and R. A. Deju, *Hydrogeology of the Uppermost Confined Aquifers Underlying the Hanford Reservation*, US-ERDA Report ARH-SA-253, Atlantic Richfield Hanford Company, Richland, Washington, March 1976.

TANK FARM GEOLOGY

Program Manager: M. W. Legatski
Section Manager: M. W. Legatski
Principal Investigators: W. H. Price
K. R. Fecht
Activity Code: B05T2

Objectives and Scope

- Document the structure and stratigraphy of the partially saturated sediments underlying the 200 Areas tank farms for use in tank surveillance and tank leak studies.
- Identify significant hydrological and sorption properties of each stratigraphic unit for evaluating the impact of tank leaks on the environment.
- Assist Tank Farm Surveillance Section management in determining the status of tanks and determine on an as-needed basis the spatial distribution of radio-nuclides leaked to the environment.

Prior Work

Prior to November 1, 1975, 10,419 sediment samples had been sieved and analyzed for calcium carbonate. These data were input into the ROCKSAN computer program, which categorizes the sediments into 1 of 19 classes. Sediment samples for the 241-A, 241-AX, 241-SX, 241-T, and 241-TX Tank Farms were visually examined and the ROCKSAN data presented on geologic cross sections and maps.

Battelle-Northwest tested laboratory equipment for determining hydrological properties of the sediments in the vadose zone beneath the tank farms and completed 654 determinations. Oregon State University is aiding in determining the precision and accuracy of these data.

Five dry wells were drilled in the 241-T Tank Farm to determine the distribution of activity of the radionuclides leaked to the sediments from the Tank 241-T-106 leak. The Chemical Technology Laboratory (CTL) completed 60% of the gamma-energy analyses on the sediment samples from these wells. The five wells were logged with a lithium drifted germanium [Ge(Li)] detector for comparison of results with the CTL analyses.

A study was conducted to assess the Tank 241-A-104 leak. Geologic cross sections were drawn to determine the stratigraphy of the unconsolidated sediments. The leak detection laterals were logged with neutron, Geiger-Muller, and Ge(Li) detectors.

Progress During Report Period

Data were prepared to provide basic geologic information for utilization in evaluating the impact of suspected and confirmed tank leaks. Incorporated were 140 detailed geologic maps and cross sections which were made with the aid of size and calcium carbonate analyses of over 9400 sediment samples from 427 wells drilled in and around the 200 Areas tank farms.

A contract was let to upgrade all tank farm water wells to meet the State of Washington's Minimum Water Well Standards. The water wells were perforated from the ground surface to a depth of 6 m and perforated from a depth of 27 m to just above the water table (approximately 60 m in 200 West Area). An 11.43-cm well casing with a packer attached was run in each well and the packer set near the water table. The annulus between the 11.43-cm casing and the existing casing was then grouted.

Battelle-Northwest is determining the hydrological properties of the sediments in the vadose zone beneath the tank farms. Draining and imbibition curves are being measured for matrix potential versus saturation and relative hydraulic conductivity of the nonwetting phase versus saturation. To date 1,651 determinations have been completed.

Battelle-Northwest is also determining the mineralogy of sediments beneath the tank farms. Twenty-two samples have been sorted by size and are currently being analyzed by X-ray fluorescence and the electron microprobe. Upon completion of this phase of work an assessment of the variance in mineralogy between various lithologic units and within a single lithologic unit will be made. These determinations are prerequisites before sorption properties can be determined.

Boyles Bros. Drilling Company, Spokane, Washington, completed two core wells in 200 West Area. These core wells are the first known attempt to continuously core unconsolidated and semiconsolidated sediments utilizing standard mud rotary coring techniques. Core recovery was estimated to be between 85 and 90%, and data from the core has demonstrated the continuity of major lithologies within the Middle Ringold Formation between tank farms under 200 West Area. Data from the two core wells were incorporated into the tank farm geology maps and cross sections. Samples from the core wells are currently being analyzed for mineralogy and hydrologic properties.

Gamma-energy analyses have been completed on all sediment samples collected during the drilling of five dry wells through the Tank 241-T-106 leak. All wells within the vicinity of the Tank have been analyzed with a Ge(Li) detector to determine the concentrations of the radionuclides

present. A facility has been designed to better calibrate this instrument.

In January work on the Tanks 241-T-106 and 241-A-104 leaks was stopped in order to complete all tank farm geology maps and cross sections by April 26, 1976. An assessment of the completion data for these two reports is currently in progress.

Evaluation of Effort

Completion of the tank farm geology maps and Ringold Formation coring have aided in a better understanding of the structure and stratigraphy under 200 West and 200 East Areas. Hydrological and mineralogical studies are showing that the procedures selected appear to be adequate to determine the properties of the sediments. *In situ* analyses of radionuclides in sediments with the use of the Ge(Li) detector will benefit in the saving of time and money in future tank leak studies.

Future Work

Geologic maps and cross sections will be drawn for the new 241-AW Tank Farm as soon as the tank excavation is completed. Tank Farm water well grouting will continue and should be completed by the end of FY 1976. Reports for the Tanks 241-T-106 and 241-A-104 leak investigations should be nearing completion by the end of Transition Year 1976. Further mud rotary coring is planned in FY 1977 to better define the geology under the separations areas. Sufficient hydrological and geological data should be completed so that actual tank leak simulations can be run in the first half of FY 1977. These simulations will be used to assess the analyses and computer modeling of tank leaks. Studies of the Tank 241-BX-102 leak will commence in

late FY 1977 and other tank leak studies will follow. By FY 1980 the distribution of all radionuclides within tank farm sediments will be determined.

TRANSPORT STUDIES

MOISTURE TRANSPORT STUDIES

Program Manager: M. W. Legatski
Section Manager: M. W. Legatski
Principal Investigators: P. G. Easley
D. J. Brown
Activity Code: B09TC

Objectives and Scope

This program's objectives are to determine the rate and direction of moisture movement and its effect on movement of radionuclides in the partially saturated sediments underlying the 200 Areas. This includes moisture from precipitation, waste dumped to cribs, pipe and tank leaks, and water applied to prevent airborne suspension. This information will be used to make decisions on waste storage siting, control of radionuclides, and waste disposal procedures.

Prior Work

The moisture distribution in two lysimeters, located about a mile south of 200 East Area, has been determined routinely since January 1972. The data have shown that the moisture entering the ground from precipitation moves downward to a depth of 2 to 4.5 m and then back to the ground surface, probably in the vapor phase, and into the atmosphere. The maximum moisture content is usually reached in January or early February (12 to 15 vol%), and the minimum

in late September to mid-October (3 to 5.5 vol%). The average moisture content below the 5.5 m depth is 6.0 vol%.

Matric potential and temperature profiles from the surface to the water table have been recorded. Equations describing the nonisothermal flow of water, vapor, and gas in partially saturated sediments have been calculated. Progress was made on an algorithm for the solution of these equations using orthogonal collocation on finite elements and Gear's method.

Progress During Report Period

The sediment moisture profiles for the open- and closed-bottom lysimeters were measured twice a month, and weekly during periods of heavy rainfall. The profiles reflected a fairly dry winter--the rainfall from November 1 to March 31 was 23% less than normal. In both lysimeters the shallow, dry zone from 2 to 3 m that is formed by summer desiccation was not removed by winter rainfall, as is normal. The precipitation wetting front slowly descended until April when the front was fairly straight, extending from the low moisture content of about 4.5 vol% at 2 m to the high point of 9.5 vol% at 1.25 m.

Instrumentation that will utilize the psychrometers and pressure tubing in the lysimeter to automatically record matric potential, temperature, and barometric pressure has been designed and is being purchased.

Progress has continued on the development of a computer code at Boeing Computer Services Richland, Inc. (BSCR) which will model nonisothermal, unsaturated moisture transport in porous media. The equations are believed to be complete, but test cases have indicated that there should be refinements in the numerical methods.

Research has begun at the BNW hydrology laboratory on the nonisothermal properties of typical Hanford sediments. Data from this will test the assumptions made for the mathematical model about the variance with temperature of hydraulic conductivity and matric potential.

Evaluation of Effort

While the lysimeters south of the 200 East Area have been valuable in determining the depth to which meteoric water moves down, other lysimeters or test wells are needed to characterize areas with different sediments, and to characterize areas where various amounts of water or liquid waste are being added routinely.

The algorithm for the solution of the mathematical model must be improved, as the test cases did not show an adequate fit to reality. Assumptions about the nonisothermal transport properties of sediments must be validated.

Future Work

Test wells or lysimeters of a new design will be installed near 200 East and 200 West tank farms, crib sites, and solid waste storage sites. These will be more shallow (about 6 to 10 m) and smaller in diameter than the old lysimeters, or may be just test wells spaced around a planted radionuclide source.

Sediment samples from each lysimeter or test well will be characterized and their hydrological properties determined. Upon completion of the initial study of the temperature dependence of sediment properties, a decision will be made as to whether or not the new samples should be characterized over a range of temperatures. Additional research to be done in the hydrology laboratory includes investigation of heat capacity and heat of wetting as functions of

saturation, and the importance of osmotic potential as a driving force in partially saturated moisture transport.

The computer code being developed by BSCR will be modified to model two-dimensional flow, stratified systems; and osmotic potential gradients.

Report Issued

L. E. Brownell, J. G. Backer, R. E. Isaacson, and D. J. Brown, *Soil Moisture Transport on Arid Site Vadose Zones*, US-ERDA Report ARH-ST-123, Atlantic Richfield Hanford Company, Richland, Washington, July 1975.

PUBLIC PROTECTION ASSURANCE

ENVIRONMENTAL INSTRUMENTATION

Program Manager:	M. W. Legatski
Section Manager:	M. W. Legatski
Principal Investigator:	L. E. Bruns
Activity Code:	K1930

Objectives and Scope

The objective of these studies is to develop field instrumentation and auxiliary equipment that will provide information to determine the concentration, amount, type, and distribution of radionuclides in the 200 Areas Plateau. It may include any new development or concept that would benefit measurement and control of toxic substances in the environs.

Prior Work

A beta assayer was laboratory-tested and it was demonstrated that radionuclides such as strontium could be separately identified and assayed providing the waste were over one year old.

The gamma Ge(Li) surface assayer was field-tested to verify that specifications were met. Some equipment for Dev-Van-II was received along with a used van. Temporary Dev-Van-II-type in-well Ge(Li) gamma detection equipment was successfully used in the field.

Work continued with metal foils for determining Pu concentrations in high-gamma fields. Thermoluminescent detectors (TLD) were further tested in the U Pond area to obtain dose rates in low-level, highly biota populated waste sites. A directional neutron counter was tested in conjunction with National Nuclear Corporation (NNC).

A concept for a calibration facility was devised. A proposal was received from Holosonics, Inc., for testing a void-volume detector.

The following concepts were reviewed for possible application: (1) CdTe detector for Am and Pu using the 10 to 100 keV energy range; (2) a large crystal Intrinsic Germanium (IG) unit for Pu, Am, Cs, ^{235}U , Eu, Co, and Sb; (3) an in-well ^{252}Cf activation unit for estimating sediment type (U, Pu, density, and porosity); and (4) drum monitor for assaying Pu at the 10 nCi/g-level for wastes with densities greater than 1.0.

Progress During Report Period

Battelle-Northwest conducted beta assayer laboratory tests which demonstrated that ^{90}Sr could be assayed at the

10 to 100 pCi/g-level with 10-year old waste if the ^{90}Sr contribution were less than 10% of the total beta activity. With both ^{137}Cs and ^{90}Sr present at equal activity, an nCi/g sensitivity level could be attained.

The prints for Dev-Van-I were revised and checked by the Quality Assurance Department. Approved prints for final assembly of the complete van were given to Transportation Utilities Maintenance Section for their installation. Some results using Dev-Van-I instrumentation were obtained in the field and sensitivities for Cs and other gamma-emitters in the pCi/m² range were attained.

All the electronics, detectors, cords and auxiliaries are on site for the *in situ* gamma core and in-well assay system, Dev-Van-II. The used van was found inadequate for off-road use and a four-wheel vehicle will be obtained. The new vehicle will be purchased as a balanced logger-van combination that will permit lowering a lead-shielded detector down a well.

Metal foil and TLD work were delayed due to construction of the calibration facility. No action was taken on the void-volume detector proposal made by Holosonics.

A design for the neutron in-well counting system was finalized at a meeting with the developers, NNC. It will include a directional unit and a coincidence unit to help distinguish between spontaneous and α, n neutrons.

Evaluation of Effort

The beta assayer shows promise for establishing new "in-roads" into assaying beta-emitters. Current shelf item instruments can give some indication in the nanocurie range but do not have the sensitivity or accuracy required for environmental work. If the field tests support the

laboratory conclusions, the beta assayer will be a valuable instrument for detecting radionuclide changes with time on surface, for assisting characterization of a given area for filter assays, for establishing if cleaned-up areas can be removed from radiation status, and for assistance in emergencies.

The need for the vans for characterization, assistance in current problems (*e.g.*, tank farm leaks, ditch monitoring) is evident; also the very high cost of laboratory analyses makes the development vans very attractive.

Field tests of the gamma surface assay equipment (part of the Dev-Van-I equipment) showed better than expected sensitivity (nanocuries) and should be a real help to investigators in identifying problems, and characterizing and noting changes of surface contamination with time. The most questionable inaccuracy associated with surface measurements is depth corrections--is the detector "seeing" a powerful source a meter deep or a low-level source in or near the surface? The major variables (*e.g.*, varying distance, use of absorbers, and core profiling) should be reviewed and held constant. This would enable the calibrations to be corrected for these attenuation "unknowns." Angular response must also be considered although the lead collimated detector may take care of this.

Dev-Van-II has many uses such as in-van core assay; *e.g.*, in-well assaying of any surface contamination and assay of pond and ditch bottoms. A temporary unit devised by BNW has helped tank farm management evaluate leaks outside of high-level waste tanks by measuring which radionuclides move in the subsurface and the extent of that movement. Work with the temporary unit has shown other

needs (e.g., improved logger, directional ability, and improved safety) for Dev-Van-II.

Further work on new concepts such as IG detectors, ^{252}Cf in-well, and drum assayers was delayed pending investigation of needed alternatives such as other field instruments and improved laboratory instruments.

Future Work

Both the beta assayer and Dev-Van-I should be ready for field exploratory studies at the end of the next six-month period. Work will resume on use of metal foils after the calibration tank is in place (see "Hazards Available to Man"). A vendor will be selected within the next six months for the Dev-Van-II logger.

Work will be continued on new conceptual instrumentation that may help investigators make better determinations of geological characteristics, radionuclides in the environs, and other toxic substances in the environs; these will include the *in situ* IG detector, ^{252}Cf in-well sediment and radionuclide assayer, ^{252}Cf drum counter, and real-time off-gas monitor.

Specifications for a neutron directional and coincidence counter will be issued for offsite purchase. National Nuclear Corporation will demonstrate a CdTe detector for assaying Pu and Am in the field.

HAZARDS AVAILABLE TO MAN

Program Manager: M. W. Legatski
 Section Manager: M. W. Legatski
 Principal Investigator: L. E. Bruns
 Activity Code: B1430

Objectives and Scope

The program objective is to develop an equation which can be used to establish relative potential health problems to man at waste disposal sites and to help establish guidelines for retrievability and releasability of wastes at the disposal sites.

The program's scope is to identify, collect, and develop a data base which can be used with this equation to aid in management decisions as to what action should be taken to minimize the short- and long-term effects of past, present, and future practices on man's biosphere.

Prior Work

The Hazard Indexes (HI) and Transport Factors (TF) were combined into an equation titled Hazards Available (HA).

$$HA = HI + TF$$

where

$$HI = \log_{10} \frac{\text{Toxic Substance Concentration}}{\text{Maximum Permissible Concentration}} = \log_{10} \frac{TSC}{MPC}$$

$$\text{and } TF = \log_{10} TF_{R_1} + TF_{R_2} \dots TF_{R_m}$$

where $R_1 \dots R_m$ are routes to man by a series of consecutive pathways.

$$TF_{R_1} = (P_a)(P_b)(P_c) \dots (P_n)$$

P_n = a, b, c, d, etc. (mechanisms of transport);
e.g., "a" could be fraction moved from sub-surface to surface, "b" - fraction from surface to air (resuspension), "c" - fraction remaining at a given distance due to dilution, "d" - fraction remaining that did not undergo deposition at the given distance

A literature study was made which concentrated on identifying and evaluating existing information on HI's and MPC's of toxic substances.

Progress During Report Period

Further work was done on determining the variable sensitivity (MPC, TSC) of the HI portion of the HA equation. Current programs were reviewed to identify data sources and data reliability. Estimates were made on the meteorological transport factors, resuspension, dilution, and deposition. High-volume mobile air sampling units were used to gather information on the levels of radionuclides being resuspended at various locations in the 200 Areas.

Evaluation of Effort

An important result of the HI studies is to understand what the absolute value as well as the order of magnitude variations represent in predicting potential hazards of radionuclides and other toxic substances.

For example, if there are 4 grams of Pu spread over 4000 m² and to a depth of 30 cm, the HI using the most conservative MPC_{air} is about 9.5. An HI of 9.5 means it would take 10^{9.5} m³ of air to dilute 1 m³ of these sediments below MPC_{air}. If 2 more grams of Pu are added, the HI is only changed from 9.5 to 9.7; similarly, if 90% of the salt cake is removed from a high-level waste tank, the HI for ⁹⁰Sr may be changed from only 16 to 15. Since MPC for a

given substance can vary several orders of magnitude depending on form (most MPC's are predicted), its impact on cleanup costs can be truly significant. It could mean the difference between cleaning up 90% versus 99.9%. Also if large quantities are present, 90% of the remaining material must be removed for a reduction of 1 in the HI. If a reduction in the Hazard Index of 4 is required, 99.99% of the material must be removed.

A proper review of the data available from other programs will require sensitivity analysis on the variables associated with the HA equation.

The transport factor, the second variable in the HA equation, will generally have negative numbers and is the other mechanism for reducing the HA. [The first mechanism is reducing the concentration of the toxic material (see HI equation).] If the amount of toxic material moving can be reduced a factor of 10 by grouting or burying, the HA will be reduced by 1. Extrapolating from a preliminary review of HI and TF, it is likely that a combination of retrieval and immobilization will provide the best cost benefit to society. Hopefully this equation will aid management in determining how much should be retrieved and how much should be immobilized in place.

The high-volume air sampling data have identified several areas that are above fallout as a result of resuspension caused by high winds. However the levels are still well below MPC_{air} and no restriction of activity is currently necessary.

Future Work

Studies in the next six months will involve performing a sensitivity analysis on the variables associated with the HA equation. Based on this analysis the type of data, the precision and accuracy, and the level of sensitivity will be determined; and the appropriate needs communicated to the existing programs which can supply the data. Those areas where data are not available and are necessary will also be identified and proposals made to obtain these data.

ANALYTICAL METHODS DEVELOPMENT

ACTINIDE ANALYTICAL PROGRAM

Program Manager:	H.	Babad
Section Manager:	H.	Babad
Principal Investigators:	W. I. Winters	
	S. J. Johnson	
Activity Code:	B09T6	

Objectives and Scope

The objectives of this work are to develop fast and reliable analytical methods for concentrating and determining trace-amounts of actinides in various Hanford nuclear wastes.

Prior Work

During the last report period solvent extraction systems for separating actinides from Hanford nuclear waste using Aliquat-336[®] (General Mills, Inc.), dibutyl-N,N-diethylcarbamylnmethylenephosphate (DBDECMP), and theonyltrifluoroacetone (TTA) were studied in greater detail and applied to a limited number of actual samples.

A single electrodeposition cell was set up and evaluated on several samples using an established procedure. Results indicated that the method was capable of providing adequate actinide recoveries and superior mounts for alpha energy analysis (AEA).

Spectrophotometric methods for determining total Th in waste were surveyed. A spectrophotometric method for determining total U in waste was developed.

Progress During Report Period

Alkaline waste solutions from Tanks 110-BX, 109-BX, 109-TX, 118-TX, 111-S, and 102-S were analyzed for Pu, Np and Am employing a sequential separation with Aliquat-336, DHDECMP, and TTA extractants. Data from these analyses indicated that the actinide levels for all except Tank 110-BX wastes were greater than 10 nCi/g. The method was evaluated on samples of sludge, salt cake, and radio-nuclide removal test effluents.

Two cells of a four-cell electroplating system have been installed and are being used to evaluate an electrodeposition procedure for actinides. A statistically designed experiment to evaluate six operating parameters indicated that plating time was the most significant variable. Plating efficiency for ^{239}Pu standards at concentrations from 24 disintegrations/minute (d/m) to 1.5×10^5 d/m was between 80 and 90%. Waste tank samples carried through the entire procedure of extraction, stripping, bisulfate melt, and electroplating gave 70 to 80% recoveries for Am and 80 to 90% recoveries for Pu with excellent alpha energy resolution.

A total Th analysis based on the spectrophotometric determination of a thorium-arsenazo-III complex was studied.

The Th was separated from interferences by extracting into Aliquat-336 from HNO_3 and stripping into 10M HCl. In order to overcome color instability, the arsenazo complex can be formed in 8M HCl following evaporation of the 10M HCl strip. Another means of removing color instability can be achieved by extracting the Th into di(2-ethylhexyl)phosphoric acid (HDEHPA), and developing the color in an HDEHPA-isopropanol-HCl matrix. Both methods were capable of giving recoveries of about 95%.

An interfering ion study was completed for the spectrophotometric determination of total U with 2-(5-bromo-2-pyridylazo)-5-diethylaminophenol (bromo-PADAP). By using a tri-*n*-octylphosphine oxide (TOPO) preextraction for U before color development, all major interferences were overcome. The method was evaluated on alkaline solutions, sludge, and salt cake wastes. Potential environmental applications of the method were surveyed by analyzing U standards at the 10^{-4} and 10^{-5} g/liter level. Data indicated that excellent precision, $\pm 2.6\%$, could be expected at the 10^{-4} g/liter U level. Uranium spike recoveries of $106 \pm 10\%$ were recorded for waste analyses. Formal documentation of the method was completed.

All the electronics, detector chambers, and vacuum ware for implementing a new AEA system were received. Testing of the electronics and development of data-reduction software was initiated.

Evaluation of Effort

The separation alpha counting procedure for sequential determination of ^{239}Pu , ^{237}Np , and ^{241}Am has an analytical precision for Pu and Am of $96 \pm 12\%$ and $83 \pm 19\%$, respectively. Frequently cruddy alpha mounts were obtained for

the Am analyses requiring numerous reruns. Further work is needed to develop methods for ^{233}U and ^{235}U . The electrodeposition system was shown to be one means of overcoming poor AEA analyses. By operating a series of the electrodeposition cells simultaneously, it should be possible to shorten the solvent extraction procedures. With completion of the new AEA system, abilities to analyze Np and Cm isotopes will be enhanced. Development of a total U procedure is complete; however further work is needed to assess interferences on the Th procedure. Even though many new capabilities have been acquired by this program, further work is merited in order to improve analytical reliability, reduce analytical cost, and complete the transfer of technology to routine laboratory operations.

Future Work

Efforts will be directed toward developing an Am separation which will yield improved AEA's. Separation methods for ^{233}U and ^{235}U analyses will be evaluated. A four-cell electrodeposition system will be completed and evaluated on both low- and high-level actinide waste. The plating procedure will be tested further for Am, Np, and U analyses; and the effect of interferences determined. The spectrophotometric method for Th will be investigated to establish detection limits and interferences. Work will continue on implementing the new AEA system for routine analyses.

RADIATION ANALYSIS SYSTEMS DEVELOPMENT

Program Manager: H. Babad
Section Manager: H. Babad
Principal Investigators: W. H. Zimmer
V. W. Hall
C. P. McLaughlin
Activity Code: B06T8

Objectives and Scope

The objectives of Chemical Technology Laboratory's participation in this program are the design and application of data acquisition and reduction systems to all phases of nondestructive measurement instrumentation which utilize radioactive emissions.

Prior Work

Two NS-660[®] (Tracor Northern, Inc.) data reduction systems oriented to gamma energy analysis and one NS-880 (Tracor Northern, Inc.) oriented to X-ray range energy analysis have been placed in service. These three systems with their associated custom programs comprise the Gamma, X-ray Data Reduction (GXDR) system. The purpose of this system is to provide all of ARHCO with automatic data reduction of radiation spectral data.

Thirteen satellite analyzer-cassette recorders are currently producing data for GXDR system action and more acquisition systems are being developed and placed in service for energy analysis of radioactive and nonradioactive constituents in bulk environmental samples, low-level crib and tank leak soil cores, low-level filter samples,

evaporator products and effluent samples, high-level waste management research and development support, passive fissile analysis, and others.

Progress During Report Period

Six phases of update of the GXDR system are scheduled. Phase 1, the establishment of communication between the four 4096 channel pulse height analyzers in the ND 50/50[®] (Nuclear Data, Inc.) and the Nova 1220[®] (Data General Corporation) in one NS-660 has been completed. Now the ND 50/50 has been transferred to the Standards Section; therefore Phase 1 has been voided and four acquisition systems lost. Phase 2, the establishment of communication between the GXDR system, Nova 1220, and the CAMAC Nova 840[®] (Data General Corporation) has been completed. Phase 3, the establishment of communication between the GXDR system Nova's 1220 and 1200[®] (Data General Corporation) and the addition of a DG 4234[®] (Data General Corporation) cartridge disc to speed data reduction has been installed and is now in the process of utilization. Phase 4 has three functions: control of the Californium Multiplier (CFX) pneumatic transfer system, establishment of a spectral data path from all sources to the DG 4234 disc (Phase 3), and the future expansion of "bussed" pulse height analyzers for data acquisition. Phase 4 has been specified and let for bid. Phases 5 and 6 are scheduled as future work.

The GXDR system gamma program revisions now permit data reduction of 8000 channel spectrum and cartridge disc-assisted data reduction. It was found that the Phases 3 and 4 updates must interface with the NS-660 - DG 4234 system through RDOS, a Data General operating system program. This has proven difficult and time-consuming but now very nearly accomplished. Both RDOS and the assembly

language data reduction program wanted control of the system. To accommodate this requirement meant that the Nova 1220 had to be expanded to 32 K of core and program revisions made to alternate control. Temporarily, the NS-660-1200 will not be included in the program and system updates.

The well Ge(Li) detector system was not received and accepted until the end of the report period. Evaluation and utilization will be conducted in the next report period.

A 10 IG detector array with attendant amplifier-mixer was justified, specified, and let for bid. Ortec Inc. has been awarded the detector bid and Nuclear Equipment Corporation has been awarded the amplifier-mixer bid. A 30-ton shield had been designed for the incorporation and use of two such arrays operating face to face in the analysis of bulk environmental level samples.

Evaluation of Effort

Throughout the report period CTL had 6 to 7 more detector systems available than pulse height analyzers on which to acquire data. Consequently, although able to keep pace with analytical support requirements, little or no calibration or system evaluation took place. Two detector systems failed and were repaired during the report period. The well Ge(Li) detector (U) was received twice before it was accepted. Significant advances were made in the GXDR system resulting in approximately a 4-fold reduction in the time it takes to reduce an average gamma spectrum. This will improve to a net 10-fold reduction with the completion of the Phase 4 GXDR system update. This represents use of only one NS-660. When both NS-660's are on line, capacity should be doubled.

Future Work

During the next report period the Phase 4 update, a Scorpio 3000[®] (Canberra Industries) will be received and integrated into the GXDR system. This will include hardware and programming interfaces of the Scorpio by Canberra to the NS-880 (X-ray data reduction), the NS-111A[®] (Tracor Northern, Inc.) cassette tape reader, and the DG 4234 cartridge disc. Spectra needing backgrounds subtraction shall have this performed in Scorpio from background stored on floppy disc. Chemical Technology Laboratory and Systems must complete the software handshake to transfer data from Scorpio to the DG 4234 via the Nova 1220 using RDOS. Purchase of "bussed," add on, 7000+ channel analyzers to Scorpio will commence.

The GXDR system programs will be altered to reduce cyclical activation analysis data and to report all gamma results with accuracy values instead of counting precision.

The well Ge(Li) detector(U) will be evaluated and placed in service.

CALIFORNIUM MULTIPLIER SUBCRITICAL ASSEMBLY

Program Manager:	H.	Babad
Section Manager:	H.	Babad
Principal Investigator:	W. H.	Zimmer
Activity Codes:	Y8D10,	B0661

Objectives and Scope

The primary objectives of this project are the analyses of low levels of fissile material. A particular need exists at Hanford for the nondestructive assay of fissile material, in any media, at a level as low as 10 nanograms fissile

material (specifically ^{239}Pu) per gram of sample. The secondary task of this project is analysis of long-lived beta-emitting fission products and nonradioactive elements through the use of standard neutron activation analysis techniques.

Prior Work

The Californium Multiplier, CFX-40, was the neutron source chosen for this project because it is capable of multiplying the spontaneous neutron flux from 40 mg of ^{252}Cf to a flux density sufficient to attain program goals in a subcritical assembly. Samples will be irradiated briefly in the core to the assembly, causing the fissile contents of the sample to fission. A delayed neutron counter (DNC) will count the delayed fission-product neutrons to determine fissile content of the sample. The neutron flux of the CFX-40 will also activate long-lived beta-emitting fission products and nonradioactive elements, permitting detection of these waste constituents at the part-per-billion level without additional investment of money or manpower. A pneumatic sample-transfer system, using molded capsules as sample-transfer vehicles (rabbits), will connect the CFX-40 with the DNC and other detector equipment.

Several phases of long-term, high-level waste management at Hanford will benefit from this analytical capability. These include projects in waste retrieval, characterization, and immobilization, as well as efforts to gather information on the long-term fixation of radio-nuclides in soil.

Progress During Report Period

The project was approved and funded. The IRT Corp. of San Diego, California, has been awarded the contract for the design and fabrication of the CFX-DNC system. The contract date was April 21, 1976.

The U.S. National Bureau of Standards 1.3-inch diameter pneumatic transfer capsule was adopted as the sample-transfer vehicle. Marlex BMB 5040[®] (Phillips Petroleum, Incorporated) ethylene copolymer was chosen as the capsule molding material on the basis of its extremely low uranium content. Uranium in the capsule material limits the assay of sample concentrations of fissile material. A molding order for 10,000 capsules was awarded to the Wilks Precision Instrument Co., Inc., of Rockville, Maryland.

The pneumatic transfer system will be purchased from Reactor Experiments, Inc., of San Carlos, California. The system will contain an automatic sample loader, an irradiation terminus, and four detector termini. All points will be connected by flexible polyethylene tubing. A central processing unit (CPU) designed to meet project requirements will control the transfer system. Specifications for a computer to fulfill this function were released for competitive bidding. Canberra was the sole bidder with their Scorpio 3000.

Evaluation of Effort

During the report period the project received approval and funding. All the required approvals were obtained and the principal contracts and purchase orders were let. A schedule of milestones and safety reviews has been established together with the personnel to implement them.

Future Work

During the next report period:

- The CFX pneumatic transfer system will be received, installed, and tested.
- The CFX transfer system control processor, the Scorpio 3000, will be received, installed, programmed, and tested.
- The CFX site will be designed, approved, and construction initiated.
- The CFX multiplier fuel plates will be fabricated and delivered to IRT Corp.
- The CFX and DNC designs will be completed. The safety and criticality prevention design of the CFX will be approved by ARHCO and fabrication started.

ACKNOWLEDGMENTS

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