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RESEARCH AND ENGINEERING DIVISION SEMIANNUAL REPORT, KK PROCESS DEVELOPMENT AND TECHNOLOGY, NOVEMBER 1, 1974 THROUGH APRIL 30, 1975

Edited by

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ATLANTIC RICHFIELD HANFORD COMPANY
RICHLAND, WASHINGTON 99352

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ABSTRACT

This document represents the first of a series which will report on a semiannual basis the activities supported by KK funds in the areas of process technology and 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, 1974 THROUGH APRIL 30, 1975

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 and 10 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 cost code listings. As further assurance that the cost of performing work is correlated, the cost center is included. Also listed is the program manager who has the responsibility for defining the program objectives and insuring adequate resources are available to perform the work. The principal investigator who has the responsibility for directing the daily work is also identified.

Each report is presented so that sufficient background is available to understand the reason for the program, what has transpired, a review of the current progress, and, finally, identification of the current direction.

This report includes brief summaries to acquaint the reader with information acquired during the report period.

SUMMARIES

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PLUTONIUM FINISHING PLANT

PROCESS TECHNOLOGY PLUTONIUM

Waste Solidification 10

A process for solidifying aqueous salt wastes containing plutonium is being developed.

Continuous Monitoring of Liquid Effluent for Plutonium at Maximum Permissible Concentration 12

A new monitor will be required as a result of new sampling data and a factor-of-10 reduction in the plutonium maximum permissible concentration (MPC) allowed in the effluent from the Plutonium Finishing Plant.

PLUTONIUM PROCESS DEVELOPMENT

Plutonium Scrap Dissolution

Plutonium Oxide Dissolution 14

Use of a continuous dissolver, rather than the present pot dissolver, has produced significant increases in plant PuO_2 dissolution capability.

Leaching Plutonium from Soil 17

An HCl-$SnCl_2$-HF leaching solution was found to be superior to the conventional HNO_3-HF solutions for removing plutonium from soil. The increased plutonium recovery was not adequate to justify installing a chloride removal process, which is necessary to make these solutions compatible with the plutonium reclamation process.

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Fused Salt Alternatives 19

A process was developed which can recover plutonium from solids that are not easily acid-leached. The process is applicable to small quantities of material that are relatively free from silica and organic material.

Plutonium Scrap Processing

Amine Solvent Extraction of Plutonium 20

A new reflux-type solvent extraction process which employs the secondary amine Amberlite LA-2® (Rohm and Haas Company) is being developed to recover plutonium from dissolved metal scrap. Alpha radiolysis products of Amberlite LA-2 are relatively innocuous compared with those of tri-*n*-butyl phosphate (TBP).

Bidentate Organophosphorus Extraction of Americium and Plutonium 23

The reagents dihexyl-N,N-diethylcarbonylmethylene phosphate (DHDECMP) and dibutyl-N,N-diethylcarbonylmethylene phosphate (DBDECMP) efficiently extract both plutonium and americium from acid waste solutions; applicability of these bidentate compounds to treatment of the acid waste stream generated in the Plutonium Reclamation Facility is being investigated.

Waste Organic Disposal 28

Practical schemes for disposing of the accumulated (∿1900 liters) inventory of Pu containing fabrication oil (50 vol% lard oil-50 vol% CCl_4) waste are being sought. One suitable process involves physical separation of Pu-containing solids from the lard oil-CCl_4 phase and sorption of the liquid phase onto a combustible substrate which eventually can be incinerated.

Page

ENCAPSULATION

CESIUM ENCAPSULATION 30

Processes for solidification of cesium chloride by pan-drying and melt-casting are being developed as workable alternatives to the existing process used in the Waste Encapsulation and Storage Facility.

ALTERNATE PRODUCT FORMS FOR ENCAPSULATION 32

Glasses suitable for encapsulation which contain up to 45 wt% cesium oxide have been have been made. They have leach rates in the range 10^{-4} to 10^{-5} g of cesium/day-cm^2.

CONCENTRATION, TERMINAL LIQUOR, SALT CAKE, AND SLUDGE CHARACTERIZATION

WASTE MANAGEMENT PLANNING SUPPORT 34

Sensitivity analysis studies produced by the Waste Management Model provide an insight into the parameters which have the greatest impact on projected waste volumes. Initial results indicate that the major variances in projected waste volumes are all process-oriented.

INTERSTITIAL LIQUOR REMOVAL 37

The 241-BY-107 Instrument Salt Well Facility was installed and operation initiated. The safety of oil field-shaped-jet charge perforating technique for salt well flow stimulation was demonstrated.

TERMINAL LIQUOR DISPOSITION 41

Wiped-films, 242-S and 242-A Evaporator facilities were identified as alternative methods to obtain volume reduction of terminal liquids for interim storage as a semisolid "mush" product in double-shell tanks. The conversion of radioactive waste

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solutions by partial neutralization will reduce the volume of stored residue liquors while forming an acceptable salt cake product. Clay in-tank solidification of terminal liquors provides an alternative method for rapidly converting liquids to solids which are acceptable for interim storage in existing underground tanks.

ALUMINUM REMOVAL FROM HIGH-LEVEL RADIOACTIVE WASTE 46

Controlled aluminum removal from radioactive waste solutions will allow continued evaporation-crystallization conversion.

SALT CAKE CHARACTERIZATION

Analytical Development 49

Development of satisfactory chemical and physical test procedures will assure timely analysis of salt cake samples when they become available.

Core Sampler Development 51

Completion of design and preliminary testing on wire-line rotary core drilling components have demonstrated that a promising method is available for waste characterization.

TANK OPERATIONAL CRITERIA 54

The underground waste storage tanks which were originally designed for liquid storage are now to be used for the storage of salt cake. The program will define criteria which will control tank use for interim salt cake storage.

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WASTE TANK LEAK DETECTION

Radio- and ultrasonic-frequency prototypes were installed in tanks and tested. The ultrasonic unit is being modified on the basis of initial tests. The radiofrequency unit was not suitable for the service required of it. 56

SOIL CONDUCTIVITY LEAK DETECTION 60

Field tests of the tank leak and pipeline detection systems were successful in detecting leaks of 380 liters.

CONTAMINATED SEDIMENT CONTROL

SURFICIAL CONTAMINATION--CRIBS, TRENCHES, SWAMPS, PONDS 62

Information collected to date indicates that the radionuclide buildup in algae floc and various plants and organisms does not pose a problem to man.

SOIL STABILIZATION 65

Surface affixants show promise for holding the soil, yet allow vegetative growth to prosper.

DECONTAMINATION/IMMOBILIZATION 68

Radionuclides associated with pond bottoms can probably best be reduced while the water remains in the pond.

BIOLOGICAL UPTAKE

Radionuclides in Soil-Plant Systems 71

Tests on tumbleweed indicate that concentration factors are independent of soil concentration for ^{90}Sr and ^{137}Cs over several orders of magnitude.

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Uptake is very age-dependent, the highest concentrations being measured between the eighth and twelfth weeks.

Radionuclides in Terrestial Plants and Animals 74

The plant communities in the B-C Crib and Redox Swamp areas were characterized. Studies of animals, including their food habits, populations, and behavior are nearly complete and documents describing the work have been published.

Radionuclides in Aquatic Biota and Sediments 78

Results of the study of radioactivity in biota and sediments show significant buildup of radionuclides in the resident fauna and flora. While significant numbers of avifauna are attracted to the 200 Areas Plateau by the ponds, it does not appear that transitory waterfowl build up significant amounts of ^{137}Cs.

ACTINIDE SEDIMENTS

ACTINIDE TRENCH CHARACTERISTICS

Characterization and Transport 82

Multiple chemical reactions occurred in the actinide trenches making it almost impossible to predict the actinide distribution of one trench based on data from another. The variations in the chemical reactions are due to differences in the sediments underlying the trenches and in the characteristics and volume of wastes disposed to each trench.

Drilling and Sampling Technology 87

Sampling equipment for use in tanks was developed and tested. Improved actinide trench drilling and sample recovery techniques were developed. Isochem's discovery of plutonium in water samples from Well 299-W18-5 were confirmed.

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HYDROLOGY

GROUNDWATER HYDROLOGY (Model Verification and Management) 91

The groundwater monitoring network was reviewed and new operating criteria were established. Wells were cleaned and pump tests were conducted. Gravity anomalies were mapped and used to locate potential areas of buried stream channels. Work on computer models continued to improve their accuracy and usefulness.

MOISTURE TRANSPORT STUDIES 96

Information obtained from the lysimeter test site continues to show that the movement of moisture is upward through the partially saturated sediments underlying the 200 Areas.

TANK FARM GEOLOGY 100

Oregon State University has begun a quality assurance program for the hydrologic analysis of Tank Farm samples collected by Battelle-Northwest. Geologic maps showing particulate size and calcium carbonate analyses indicate a good correlation between the actual radionuclide distribution and that which would be predicted from geologic data.

TRANSPORT

AIRBORNE RELEASES 102

Computational methods for estimating downwind surface concentrations of radionuclides from the BC Crib have been successful.

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INSTRUMENTATION

PUBLIC PROTECTION ASSURANCE

Environmental Instrumentation 107

A beta analyzer 10 times more sensitive than those currently in use was tested and calibrated for ^{90}Sr. Environmental measurement instruments are being developed for nanocurie (10^{-9}) levels. Metal foil activation methods for neutron-emitters, directional gamma, and shielded thermoluminescent dosimeters were tested.

Waste Characterization 112

The effectiveness of two different environmental sampling techniques has been verified by several different laboratories.

ANALYTICAL METHODS DEVELOPMENT

ACTINIDE ANALYTICAL PROGRAM

115

A method for separating and determining Pu, Am, and Np in Hanford wastes was developed on actual terminal liquor and salt cake waste forms.

RADIATION ANALYSIS SYSTEMS DEVELOPMENT

118

Seven new detector systems were brought on-line--three of which are intended for mobile applications. Detector failures continue to affect full equipment utilization.

PLUTONIUM FINISHING PLANT

PROCESS TECHNOLOGY PLUTONIUM

Plutonium Reclamation Facility Waste Solidification

Progam Manager:	R. E. Felt
Section Manager:	R. E. Felt
Principal Investigators:	M. R. Fox T. R. Haddad
Activity Code:	B04T0

Objective and Scope

Design and development of a waste solidification process for Plutonium Reclamation Facility effluent streams is the objective of this program. The solid product will be in a form suitable for retrievable surface storage. The process capacity will be based on the current Z Plant operating at full capacity.

Prior Work

None.

Progress During Report Period

A preliminary flowsheet for a solidification process is being developed. The design flow rate is 15 liters/min. Although the waste stream composition will vary, a design basis composition has been established as follows: 0.4*M* ANN, 0.2*M* Ca^{++}, 0.03*M* Fe^{+++}, 0.3*M* Mg^{++}, 3.3*M* Na^{+}, 0.3*M* H^{+}, 0.2*M* total F^{-}. This waste material has an initial boiling temperature of 107° C and a density of 1.31 g cc^{-1}.

Currently more extensive data on boiling and drying characteristics are being obtained. Several vendors have

been contacted concerning equipment fabrication and pilot-plant studies.

Evaluation of Effort

The current flowsheet includes a concentrator (boiler) and a dryer. The information developed thus far indicates that these processes are able to evaporate the waste stream and produce a solid with suitable handling properties. When the evaporation of simulated waste is carried beyond the precipitation point (120° C), a slurry is produced. The solids in the slurry are granular and soluble in water. Further information will be needed to complete development and establish operating conditions for these processes.

Future Work

More solubility information will be required to refine the flowsheet and to design for process control and for recovery from process upsets. Neutralization information will be required to optimize nitric acid reduction while simultaneously avoiding precipitation problems. Finally, some pilot-plant work must be done using actual Z Plant waste to reduce the uncertainty incurred in using a simulated waste and finalize the flowsheet.

Continuous Monitoring of Liquid Effluent for Plutonium at Maximum Permissible Concentration

Program Manager:	R. E. Felt
Section Manager:	D. E. Braden
Principal Investigators:	G. E. Martin D. N. Morrill
Activity Code:	B04T2

Objective and Scope

The equipment and techniques, needed to reliably monitor the Z-19 ditch effluent from Z Plant on a continuous basis, are being developed for plutonium at maximum permissible concentration (MPC).

Prior Work

An instrument was designed, built, and installed in the 2904-ZA Building as an engineering prototype to monitor the Z-19 ditch. Troubleshooting of the system was started.

Progress During Report Period

Several hydraulic problems exhibited by the monitor were resolved and the monitor calibrated. The datalogger for this equipment developed several problems after installation and was removed from service for redesign of one part.

Evaluation of Effort

During this reporting period two facts were learned about the monitor which will limit its usefulness.

First, data from the new proportional sampling

equipment (which went on line in August 1974) showed conclusively that the solids contribution to the alpha burden of the stream must be taken into consideration. When this activity was initiated in 1972, the data indicated that the solids contribution could be ignored and the design proceeded on this basis. The existing monitor will not count solids and cannot be modified to do so.

Second, in October 1974 the Energy Research and Development Administration, Richland Operations Office (ERDA-RL), issued a letter changing the status of the Z-19 ditch releases which lowered the acceptable plutonium concentration in the effluent stream below the sensitivity of the existing monitor.

Work is continuing to place this equipment in service because it is felt it can be useful in monitoring burst releases until a better design is developed.

Future Work

The existing monitor will be placed in service as soon as possible. Work has started on a new monitor design which takes into consideration the suspended solids and the lower detection limit now required.

PLUTONIUM PROCESS DEVELOPMENT

Plutonium Scrap Dissolution

Plutonium Oxide Dissolution

Program Manager:	R. E. Felt
Section Managers:	H. Babad R. E. Felt
Principal Investigators:	D. A. Dodd M. R. Fox
Activity Code:	B1330

Objective and Scope

Plutonium-bearing scrap is generated as a result of purification, fabrication, and handling operations in processing facilities. The objective of this investigation is to increase Pu recovery by determining the optimum dissolution and/or leaching conditions compatible with Plutonium Reclamation Facility's operation. Because scrap is highly variable in matrix and composition, this study considers as many scrap forms as practical.

Prior Work

Controlled batch dissolution tests were performed on plutonium dioxide calcined to a temperature of 400° C, on burned metal at 600 to 800° C, and on sintered powder at 1400° C. The optimum fluoride ion concentration to dissolve these oxides was determined to be 0.1 to 0.2*M*. These levels provided an adequate fluoride concentration without precipitating plutonium from solution as plutonium fluoride.

Rate constants for PuO_2 dissolution at elevated temperatures have been calculated for the rate law

$\frac{d[Pu]_t}{d_t} = KiS[HF]_f$, where Ki is the rate constant, S is the surface area of the PuO_2, $[Hf]_f$ is the concentration of free hydrofluoric acid, and $[Pu]_t$ is the total plutonium concentration. These constants increase with temperature from 1.7×10^{-8} liters/cm -min at 35° C to 54×10^{-8} liters/cm^2-min at 100° C.

Progress During Report Period

Continuous countercurrent dissolver concepts have been evaluated with PuO_2 and plutonium-bearing incinerator ash, recycled Pu-Al alloy, and miscellaneous accumulations of scrap. Dissolver solutions consisting of 0.1*M* HF-12*M* HNO_3 or 0.1*M* HF-15.8*M* HNO_3 were considered.

Plutonium oxide does not dissolve linearly with time in a continuous dissolver system. Dissolution rates of both the burned metal and sintered oxide were measured against time and found to follow a Gausian curve. The dissolution rate of sintered plutonium oxide pellets was increased in tests by a factor of 20, to nearly the rate for burned metal oxide. This increase was produced by grinding the pellets, thus decreasing the particle size and increasing the surface area. The nonlinear dissolution rate has been observed in dissolution tests of incinerator ash and other scrap.

Reheating the incinerator ash before making dissolution contacts significantly increased the plutonium leach rates and lowered the plutonium concentration in the residual sludge to a level no longer practical to process for plutonium.

Alternating HNO_3-HF and 50% NaOH contacts of incinerator ash did not enhance the solubilization of

plutonium. Ash residue following the NaOH contact solution was very difficult to separate from the liquor. After standing for two days a precipitate re-formed in the filtered product solution.

Preliminary studies have been completed using nonaqueous and alternate dissolver solutions. As yet these have shown no improvement over the HNO_3-HF system.

Evaluation of Effort

Tests have demonstrated significant increases in efficiency of a continuous dissolver concept over static pot dissolvers. Increased efficiency, though variable with scrap type, has reduced the level of operator attention and leach time. Lowering the fluoride concentration lessens the chance of plutonium fluoride precipitation, yet maintains effective dissolution.

Future Work

Incinerator ash and other high-silica scrap will be treated with concentrated hydrofluoric acid at ambient temperature, after which the plutonium will be dissolved in a high aluminum nitrate system.

Leaching Plutonium from Soil

Project Manager:	R. E. Felt
Section Manager:	R. E. Felt
Principal Investigators:	D. A. Dodd P. C. Ely
Activity Code:	B1330

Objective and Scope

In this study data were collected for development of a flowsheet which could be used to reclaim plutonium from the Z-9 Enclosed Trench soil.

Prior Work

Past studies indicated that plutonium is concentrated in the top 15 cm of the trench as particulate oxide, tightly bonded ion-exchanged species, and material adsorbed in cracks and fissures of the sand. Tests have shown that screening with nitric acid separates ∿85% of the plutonium and ∿20% of the soil from bulk material.

Progress During Report Period

A leaching flowsheet was developed for use on the plutonium-rich portion of the Z-9 soil. This flowsheet incorporates dry screening to separate the plutonium-bearing fines from the coarse material. Plutonium leaching is accomplished by contacting the fines successively with nitric acid, then with hot nitric acid-hydrofluoric acid.

Demonstration runs with this flowsheet indicate a plutonium recovery from the fines of 50 to 70% with a product concentration of about 1 g Pu/liter. Higher liquid-to-solids ratios yield appreciably better leaching but a more dilute product.

A continuous-type dissolver model being developed for plutonium recovered 95% of the plutonium from high-temperature dried fines with a product concentration of about 0.5 g Pu/liter (see p. 14, "Plutonium Oxide Dissolution").

A new leaching solution, 7.5*M* HCl-0.1*M* $SnCl_2$-0.5*M* HF, was tested with very good results. Contact for 1 hr with agitation at 90° C on unscreened soil yielded from 95 to 100% plutonium recovery with a product concentration of ∿0.2 g/liter.

A flash point of 105° C was determined for air-dried surface soil in a closed cup-type apparatus. There was no flash point for the normally moist soil.

Evaluation of Effort

An HNO_3-HF leaching flowsheet has been demonstrated but process optimization and equipment design studies will be required before implementation can take place. Leaching with HCl-$SnCl_2$-HF is superior to HNO_3-HF leaching for removing plutonium soil. However the improvement was not adequate to justify installation of the chloride removal equipment necessary to prepare the solution for processing through the Plutonium Reclamation Facility.

The flash point of the dried Z-9 soil is not considered a severe hazard as a fairly high temperature (105° C), poor ventilation, and an ignition source would be required to ignite the vapor phase.

Future Work

The activity will be terminated since the decision was made to store rather than recover the Pu from the Z-9 soil.

Fused Salt Alternatives

Program Manager:	R. E. Felt
Section Managers:	H. Babad H. H. Van Tuyl (BNW)
Principal Investigators:	D. A. Dodd J. A. Wheelwright (BNW) E. J. Wheelwright (BNW)
Activity Code:	B1330

Objective and Scope

The objective of this project is to develop a process to recover plutonium from existing incinerator ash, acid-leached ash, and solids collected during processing; and to demonstrate the process on a laboratory-scale.

Prior Work

A process was demonstrated in which equal parts of $NaOH$-Na_2O_2 were fused with Pu, leaving scrap in a scrap-to-salt ratio of 2:7. After fusing in a mild steel can, the can and contents were dissolved first in water, then in HNO_3. Tests conducted on two containers of stored incinerator ash resulted in plutonium recoveries of 95%. On a third container of ash, a gelatinous material formed during acidification from which entrained plutonium could not be removed. The result was a very low product recovery.

Progress During Report Period

As much as 95% of the Pu was recovered from solids centrifuged from process streams using the optimum solids-to-salt ratio of 2:7. In one attempt the melt and sample spewed out of the reaction vessel. Evaluation of the reaction indicates that an organic residue which reacted

with the caustic peroxide at the elevated temperature was present in the solids.

The experimental data have been assembled into a draft final report and evaluated for process applicability.

Evaluation of Effort

This process is not readily adaptable to scale-up, but it is practical for processing small quantities of solids low in silica and organic residues. Ejection of the molten salt during processing could be eliminated by pre-incinerating the solids, which produces the desired low-organic content. No techniques were developed to remove silica, which is probably causing the gel.

Future Work

Significant experimental work on this project has been concluded. A report will be prepared.

Plutonium Scrap Processing

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

Objective and Scope

The objective of this program is to develop an amine plutonium extraction process to replace the

tributyl phosphate (TBP) process currently used in the Plutonium Reclamation Facility to realize improved performance and minimize operating costs.

Prior Work

A conceptual plutonium recovery process was devised which employs Amberlite LA-2® (Rohm and Haas Company), a high-molecular weight secondary amine, as the extractant. The 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 solvent wash column to remove small amounts of Pu which are not stripped in the CC Column. This new amine extraction process was satisfactorily demonstrated in mixer-settler runs at 25° C employing both 30% LA-2-CCl_3 and 30% LA-2-TCB (trichlorobenzene) solvents. Because of improved kinetics, process performance with the LA-2-TCB solvent was considerably improved by operation at 50° C rather than at 25° C.

Chemical and radiolytic stability of the LA-2 extractant under proposed flowsheet conditions is superior to that of the presently used 20% TBP-CCl_4 solvent. For example, after exhaustive stripping with dilute HNO_3, an irradiated LA-2 solvent retains only about 0.3 mg Pu/liter for each watt-hour of absorbed energy, compared with retention of 56 mg Pu/liter by an irradiated, stripped 20% TBP-CCl_4 solvent.

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 4.0*M* HNO_3-0.0 to 1.0*M* $Al(NO_3)_3$-0.0 to 0.25*M* HF solutions containing 1, 10, 30, and 60 g/liter Pu.

Progress During Report Period

Having demonstrated in laboratory studies the technical feasibility and operability of a reflux amine flowsheet, current emphasis is on obtaining comprehensive data for the equilibrium distribution of plutonium, nitric acid, uranium, and thorium between a 30% LA-2-CCl_4 solvent and various HNO_3-$Al(NO_3)_3$-HF solutions. Such data will provide input for mathematical modeling of the amine systems and will allow computer-assisted design and testing (using the program SEPHIS) of optimum reflux and once-through flowsheet conditions.

Input equilibrium distribution data for modeling the system HNO_3-$Al(NO_3)_3$-HF-$Pu(NO_3)_4$-LA-2-CCl_4 were reviewed during this report period and found to be deficient in several critical concentration areas. Additional batch contacts were made to obtain the missing distribution data. Extensive measurements were made of the specific gravity of 30 vol% LA-2-CCl_4 solvents containing $Pu(NO_3)_4$ and/or HNO_3; such data are needed to account for changes in organic phase volume which occur as Pu is loaded into the amine extractant. Good progress was made toward acquisition of equilibrium distribution data for plutonium, uranium, and nitric acid for the system HNO_3-$Al(NO_3)_3$-HF-$Pu(NO_3)_4$-$UO_2(NO_3)_2$-LA-2-CCl_4. These data will be used to devise flowsheets for amine solvent extraction recovery of Pu from Pu-U scrap.

Evaluation of Effort

The superior chemical and radiolytic stability of Amberlite LA-2, compared with that of TBP, makes an amine extraction process particularly well-suited for use with Pu scrap containing significant concentrations of ^{241}Pu and ^{240}Pu. Tests of a conceptual three-column amine

extraction process in laboratory-scale counter-current equipment show it to be a workable scheme for use in the Plutonium Reclamation Facility. The potential benefits--both short-term and long-range--to be derived from replacing the TBP solvent with LA-2 appear to be great enough to warrant the additional research effort needed to develop and demonstrate a satisfactory amine extraction system.

Future Work

Initial computer-assisted design of a flowsheet for amine recovery of plutonium from Pu-only scrap will be undertaken using equilibrium distribution and specific gravity data obtained in the last year. Mixer-settler tests of one or two such flowsheets will be made to verify the accuracy of the computer model. Additional batch contacts will be made to complete acquisition of equilibrium distribution data for the HNO_3-$Al(NO_3)_3$-HF-$Pu(NO_3)_4$-$UO_2(NO_3)_2$-LA-2-CCl_4 system.

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:	B1340

Objective and Scope

The principal objective is to develop technology for reducing the actinide content of the Plutonium Reclamation Facility (PRF) aqueous waste streams to $\leq$10

nCi/g. A secondary objective is to develop a bidentate organophosphorus solvent extraction process to recover both americium and plutonium directly from PRF current acid waste (CAW stream), thereby avoiding the pH adjustment step required by the present dibutylbutyl phosphonate (DBBP) extraction scheme.

Prior Work

Comprehensive data were obtained on the equilibrium distributions of americium, plutonium, HNO_3, and various impurity metals (*e.g.*, Ca^{2+}, Mg^{2+}, etc.) between various aqueous HNO_3-$Al(NO_3)_3$-HF solutions and 30 vol% DHDECMP-CCl (and TCB, trichlorobenzene) solutions and 30 vol% DBDECMP-CCl_4 solutions. The latter bidentate organophosphorus reagents are available from Wateree Chemical Company, Lugoff, South Carolina; DHDECMP is an acronym for dihexyl-N,N-diethylcarbamylmethylene phosphate while DBDECMP is dibutyl-N,N-diethylcarbamylmethylene phosphonate.

Technical-grade (as-prepared) DHDECMP and DBDECMP both contain an unidentified impurity which has a very high affinity for Am(III) and Pu(IV) at low acid (<0.5*M*) concentrations. Removal of this impurity from DBDECMP by vacuum-distillation permits the use of dilute HNO_3 solutions to partition americium from co-extracted plutonium. Vacuum-distilled DBDECMP is available from Wateree Chemical Company. Because of its higher boiling point, DHDECMP is not particularly amenable to purification by vacuum distillation. Suitably pure 30% DHDECMP-CCl_4 (or TCB) solvents can be prepared, however, by acid hydrolysis-caustic washing procedures.

A conceptual chemical flowsheet employing purified 30% DHDECMP-CCl_4 (or TCB) as extractant was

successfully demonstrated in mixer-settler tests with actual CAW solution as feed. The new flowsheet increased overall americium recovery to 90% from the 69 to 80% obtained with the present DBBP flowsheet. Over 99% of the soluble plutonium in the CAW solution was recovered.

Stability of the 30% DHDECMP extractant to alpha radiolysis was found to be excellent over the range of exposure expected in PRF application (2 to 10 Wh/liter). Effects of cross-contamination of DHDECMP solvents with mainline 20% TBP-CCl_4 solvents were determined.

Progress During Report Period

Additional equilibrium data were obtained for the distribution of Am(III), Pu(IV), and HNO_3 between HNO_3-$Al(NO_3)_3$ solutions and 15 and 30 vol% DBDECMP-CCl_4 (and TCB) extractants. These experiments were performed with purified (vacuum-distilled) DBDECMP.

These new distribution data, together with earlier results, were used to set conditions for mixer-settler tests of DBDECMP Am-Pu extraction flowsheets. Extraction column tests with actual CAW solution employed four mixer-settler stages; equal aqueous-to-organic flow ratios; and 30, 20, and 15 vol% DBDECMP-CCl_4 extractants. The aqueous raffinate from these runs contained, respectively, 0.47, 5.1, and 15% of the Am in the CAW solution. In all cases over 99.5% of the Pu in the CAW feed reported to the DBDECMP phase. Significantly, the aqueous raffinate from the run with the 30 vol% DBDECMP extractant contained only 24 nCi/g of ^{241}Am and 4 nCi/g of Pu. In partition column tests using three mixer-settler stages, approximately 85% of the Am, accompanied by only 3% of the Pu, was

stripped from the 15% DBDECMP extract with 0.1*M* HNO_3 at an aqueous-to-organic flow ratio of 0.33.

Scientists at the Energy Research and Development Adminstration Idaho Nuclear Energy Development site are also studying the use of neutral bidentate organophosphorus reagents for removing actinides from their first-cycle high-level waste. They report that the solubility of DHDECMP in dilute HNO_3 solutions is low (∿0.5 g/liter) and comparable to that of TBP; unfortunately DBDECMP is disproportionately soluble (∿60 g/liter) in 0.1*M* HNO_3. These data emphasize the need to scrub partition and Pu-strip column aqueous products with a small volume of diluent to recover valuable DBDECMP and to prevent its carry-over into subsequent Am and Pu purification steps.

A sample of purified (vacuum-distilled) dibutyl-N,N-diethylcarbamylphosphonate (DBDECP) was obtained from Richmond Organics, Richmond, Virginia. Capacity of 30 vol% DBDECP-CCl_4 solutions was found to be markedly inferior to that of DBDECMP-CCl_4 solvents.

Because its solubility in dilute HNO_3 solutions is so much less than that of DBDECMP, DHDECMP is an attractive candidate bidentate reagent for extraction of Am and Pu from CAW solutions. Additional tests confirmed that satisfactorily pure DHDECMP-CCl_4 solvent can be prepared by hydrolysis for 24 hr at 60° C with an equal volume of 6*M* HCl followed by washing with 1*M* NaOH and HNO_3 solutions. This purification technique cannot be used with undiluted DHDECMP, however, because the DHDECMP itself degrades excessively. Other tests indicate that at 50° C HNO_3-HF solutions can be used to strip Am(III) from 15 vol% DHDECMP-TCB solvent prepared from technical-grade DHDECMP. These results indicate it may be possible to use as-received DHDECMP in a

simple coextraction-costrip cycle for removal of actinides from acid CAW solution.

Evaluation of Effort

The ability of bidentate organophosphorus extractants to reduce the soluble Am and Pu concentrations of PRF CAW solution to actinide-free ($\leq$10 nCi/g) levels has been demonstrated. Applicability of such flowsheets directly to acid CAW solution eliminates the need for difficult-to-control on-line neutralization of the waste to 0.1*M* HNO_3 by addition of NaOH. As a further advantage of not adding NaOH to the CAW solution, the volume of solidified acid waste is less than the volume of solidified neutralized waste.

Before these new extraction processes are ready for plant-scale tests the most satisfactory bidentate compound must be selected. DHDECMP is difficult to purify but has suitably low aqueous phase solubility, while DBDECMP can be readily purified but is relatively soluble in dilute acid solutions. Further research to resolve questions of solvent purification and solubility is merited to realize the potential advantages of a bidentate extraction process.

Future Work

During the next report period the remaining work-distribution ratio tests, mixer-settler runs, alpha radiolysis studies, and solubility tests required for laboratory-scale development of a DBDECMP flowsheet will be continued. It is also planned to continue, at a modest level of effort, to seek new ways to purify DHDECMP and develop methods of using technical-grade DHDECMP.

Waste Organic Disposal

Program Manager:	R. E. Felt
Section Manager:	H. Babad
Principal Investigator:	D. A. Dodd
Activity Code:	B1340

Objective and Scope

Approximately 305 cans containing about 1900 liters of waste fabrication oil (50 vol% lard oil-50 vol% CCl_4) are stored in the 234-5 Building (Z Plant). This material, produced in previous plutonium metal fabrication operations, consists of a liquid phase and Pu-bearing solids ("sludge" and "crust"). The objective of this task is to devise suitable methods for recovering the plutonium from the fabrication oil and for disposing of the organic liquid safely.

Prior Work

Limited exploratory work was performed several years ago at Hanford to determine a suitable way of disposing of the accumulated inventory of fabrication oil; these studies were terminated before achieving their objective.

Workers at both the U. S. ERDA Rocky Flats and Los Alamos sites have studied methods of handling plutonium containing fabrication oil, including sorption of the organic material on noncombustible substrates.

Progress During Report Period

Physical means (filtration and centrifugation) were used to separate Pu-bearing solids (sludge and

crystalline crust) from the liquid oil phase. Approximately 95 to 99% of the Pu sorbed on the solids was recovered by igniting the solids in air at 400° to 500° C and subsequently leaching the fired residue with 12*M* HNO_3-0.1*M* HF solutions at boiling temperature.

The liquid (lard oil-CCl_4) phase when free of solids contains only small (~0.003 g/liter) amounts of plutonium. Conventional paper towels sorb large volumes of such liquid. For example, a standard package (3400 cm^3) of Crown-Zellerbach paper towels, before becoming saturated, absorbed 1.3 liters of the lard oil-CCl_4 solution. Paper towels containing sorbed mixtures of lard oil and CCl_4 can be stored conveniently in suitable steel drums pending eventual incineration.

Various noncombustible substrates were screened for their ability to sorb lard oil-CCl_4 solutions. Of the materials tested, Zonalite®, a vermiculite insulation manufactured by W. R. Grace Company, proved most satisfactory; Zonalite sorbed up to half its volume of the 50% lard oil-CCl_4 solution before becoming saturated.

Evaluation of Effort

What appears to be a safe, technically and economically feasible scheme for disposing of stored fabrication oil wastes has been devised. Key features of this scheme include (1) physical separation of Pu-containing solids from the liquid lard oil-CCl_4 phase, (2) sorption of the liquid phase on a combustible substrate, and (3) subsequent incineration of the substrate and sorbed organic material. Existing equipment and technology can be used, if necessary, to recover plutonium from the ash that results

from the incineration step and for ignition and leaching of plutonium from the Pu-bearing sludge and crust materials.

Future Work

Potential applicability of Imbiber-Beads®, a product of Dow Chemical Company, for sorption of the lard oil-CCl_4 phase will be determined. Previous work has shown that organic liquids tightly entrapped by these combustible alkyl styrene beads do not desorb on standing or upon minor temperature changes. The suitability of other combustible sorbents for this purpose will also be investigated as candidate materials are found or suggested.

ENCAPSULATION

CESIUM ENCAPSULATION

Program Manager:	R. E. Felt
Section Manager:	R. E. Felt
Principal Investigators:	J. D. Moore T. R. Gallant
Activity Codes:	B08T0, B08T1

Objective and Scope

A workable new process for cesium solidification will be developed which will allow waste encapsulation production goals to be achieved. One or more promising process alternatives will be developed in nonradioactive tests using full-scale equipment. From these tests, a process flowsheet and equipment requirements for cesium solidification in the Waste Encapsulation and Storage Facility (WESF) will be identified.

Prior Work

None.

Progress During Report Period

Two cesium chloride solidification processes will be developed in parallel. In one process, the cesium chloride is dried to a free-flowing powder in a stirred, steam-heated pan dryer and the powder is densified in storage capsules by vibratory compaction. In the second process, cesium chloride solution (concentrated in an evaporator) is dried, melted, and cast into slugs in a tilt-pour vessel. The slugs are then placed in the storage capsules.

Equipment for process testing is now being designed.

Evaluation of Effort

In selecting a cesium chloride product form, existing WESF chlorination and off-gas systems can be used. A potential for commercial sales of the cesium chloride salt also exists.

Batch solidification processes such as those chosen for development are felt to be most compatible with the Waste Fractionization Plant cesium feed system and with the existing batch capsule welding and inspection processes.

Use of a separate evaporator and tilt-pour melt caster will reduce the feed evaporation time cycle and eliminate the need for high-vacuum seals necessary for operation of the existing WESF equipment. The pan dryer operates at lower temperatures and thus is a less corrosive process than melt casting. Limited nonradioactive development of the pan-drying process carried out by the Bethlehem Corporation has shown the process to be a promising alternative for solidification of cesium chloride solutions.

Future Work

In the next six months, of full-design scale equipment for testing the tilt-pour melt caster and pan-dryer processes will be completed and fabrication of the equipment will be initiated. Laboratory studies correlating cesium chloride solution boiling points, specific gravities, and molarities will be completed to aid in design of a concentrator for use with the melt caster.

ALTERNATE PRODUCT FORMS FOR ENCAPSULATION

Program Manager:	R. E. Felt
Section Manager:	H. Babad
Principal Investigator:	D. M. Strachan
Activity Code:	B08T0

Objective and Scope

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

Prior Work

None.

Progress During Report Period

Although all the possible compositions have not yet been investigated, several glasses with apparent viscosities low enough at 900° C to be used in the present encapsulation equipment have been selected as alternatives to cesium chloride. These glasses contain up to 45 wt% cesium oxide and one glass uses Battelle Pacific Northwest Laboratories'

73-1 zinc borosilicate glass frit. Four of these glasses have been stored at 400° C in a furnace for more than five months with no visible signs of devitrification.

Leach rates have been obtained on several glasses containing up to 45 wt% cesium oxide and were found to be 10^{-4} to 10^{-5} g Cs/day-cm^2. Taking into account the very high cesium contents of these glasses (30 to 45% as opposed to ∿1% for other silicate melt glasses), these are very low leach rates.

Evaluation of Effort

Work to date has demonstrated that glasses can be made with estimated rheological properties compatible with the cesium chloride melter. The glasses are stable for long periods of time with respect to crystallization at temperatures above the projected storage conditions. Stability in water has also been shown. These properties make the glasses far superior to cesium chloride as materials for long-term storage in capsules under water should any penetration of the capsules occur.

Future Work

Past work has demonstrated that pollucite ($CsAlSi_2O_6$) can easily be formed from a basic solution containing cesium. Attempts will be made to obtain quantitative yields of pollucite. Pollucite offers the advantages of high cesium content (42.6 wt%), low leachability, and extremely low volatility to temperatures in excess of 1200° C. Other mineral forms will be investigated as well.

Attempts will also be made to immobilize cesium on zeolitic compounds. Alternative strontium compounds such as

strontium silicate ($SrSiO_4$) will be evaluated. A number of cesium glasses will be sent to Battelle-Northwest for viscosity measurements.

CONCENTRATION, TERMINAL LIQUOR, SALT CAKE, AND SLUDGE CHARACTERIZATION

WASTE MANAGEMENT PLANNING SUPPORT

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

Objective 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 developed to calculate inventories of liquid and salt cake wastes as a function of time for a wide variety of plant operating models.

Progress During Report Period

Sensitivity analysis studies on the Waste Management Model were completed to provide an insight into the parameters which have the greatest impact on the projected waste volumes in the waste solidification program. Eight process

parameters and seven modes of operation of Hanford facilities were varied and the effect of each change was identified by comparison with the models base case. Results of the analysis showed that the parameters that 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. Consequently with additional data, specifically salt core samples, the accuracy of the model results could be improved.

Synthetic atmospheric waste evaporator terminal liquors evaporated at 41 torr in a laboratory vacuum evaporator to remove 30 and 40% of the water produced up to 0.25 liter of nondeliquescent solids per liter gallon of feed when the solids and liquids were separated at 40° C or higher. Solids separated from mother liquors at 25° C deliquesced between 9 and 16 days after being placed in a 70% relative humidity chamber at 23° C. Analysis of solids showed lower sodium hydroxide concentration in the solids separated at higher temperatures.

In the Salt Cake Characterization Program, wastes were found to contain more aluminum than had been anticipated previously. The majority of the salt cake which will form upon evaporation of the wastes is expected to consist of sodium nitrate and sodium carbonate solids.

Knowledge of the anticipated breathing rate of salt storage tanks is required as a basis for evaluating such effects as hydrogen accumulation (from radiolytic decomposition of water) and water accumulation rates by hygroscopic solids. The anticipated breathing rate due to natural fluctuations in atmospheric pressure for calendar year 1973 was found to be 0.7% per day. It was also found

that breathing rates were higher in winter months than in summer months.

Soil samples from dry wells in the 241-SX Tank Farm were categorized into eight different types and thermal conductivity measurements were made on each type. The average dry thermal conductivity was found to be 0.43 W/m °C.

A study was completed to determine the relationship between feed chemistry and the required evaporator-crystallizer system flow rates to maintain a uniform slurry residence time. Of the various feed constituents, the hydroxide and nitrate ion concentrations were found to have the greatest effect on the fresh feed and recycle feed rates required to maintain a given slurry rate.

An estimate of the total dissolved solids in the dilute waste inventory has been compiled based on waste type and available information about the composition of each waste.

The hydrogen content of tank vapor spaces above salt cakes was estimated using the known hydrogen evolution rate from aqueous nitrate solutions in radiation fields and the tank breathing rate. Based upon the assumption that all radiation energy is absorbed in water (salt cakes typically contain 4 to 24 wt% water), the hydrogen concentration will remain below 2.5 vol% (flammable range in air is 4 to 75 vol%) for up to 8.8 kW of radioisotopic heat generated.

Evaluation of Effort

The work which has been carried out within the scope of this activity will permit the other activities in the over-all program to be accomplished in a timely manner.

Future Work

The hydrogen evolution rate of salt cake will be determined. The solids formed from vacuum-evaporator crystallizer processing of atmospheric evaporator bottom liquors will be characterized in the laboratory.

INTERSTITIAL LIQUOR REMOVAL

Program Manager:	R. C. Roal
Section Manager:	R. J. Thompson
Principal Investigator:	D. R. Christensen
Activity Codes:	B0659, B06T2

Objective and Scope

In this program the objective is to develop, design, and demonstrate the technology, methods, and equipment required to remove the maximum quantity of interstitial liquid and to immobilize any residual liquid to the extent that is technically and economically practicable.

The five major areas of investigation which have been identified are to (1) characterize the flow of interstitial liquids in salt cake, (2) improve salt well pumping equipment, (3) stimulate the flow of interstitial liquids, (4) evaluate the feasibility of residual liquid evaporation as a removal and/or immobilization technique, and (5) evaluate the feasibility of *in situ* fixation of interstitial liquids.

Prior Work

In the area of flow characterization, Instrumented Salt Well facilities were designed for two types of salt cake storage tanks. Dr. Lyman L. Handy, Chairman of the

Petroleum and Chemical Engineering Departments at the University of Southern California, was retained as a consultant on the program to develop a mathematical model of interstitial liquid flow through salt cake.

In the area of pumping equipment improvement, a deep-well jet pump was procured for testing and a test loop was designed to simulate the salt well pumping environment.

In the areas of flow stimulation, residual liquid evaporation, and *in situ* immobilization, five studies were undertaken.

- Battelle Pacific Northwest Laboratories provided preliminary feasibility evaluations of various stimulation and immobilization techniques and ARHCO's Chemical Technology Laboratory investigated techniques for the removal and/or immobilization of nonpumpable liquids.
- A study was performed on the feasibility of modifying tank bottom configuration to enhance liquid drainage. Results showed that any such modifications would be very costly.
- In other programs evaporation rate of liquids to the tank vapor space ventilation air stream was calculated and found to be small.
- Bench-scale tests were performed on the feasibility of vibrating drained salt cake to free additional liquids for removal. A significant increase in removable liquid was produced.
- A proposal was made concerning the applicability of oil field shaped-charge jet perforating techniques for the stimulation of salt well flow and a test to evaluate the safety of these techniques was designed.

Progress During Report Period

The 241-BY-107 Instrument Salt Well Facility was installed and operation began. The design for the 241-S-111 Facility was finalized. The preliminary loop testing of a modified commercial deep-well jet pump was completed. The permeability of synthetic salt cake (determined by both Battelle-Northwest and Dr. Handy) was in the range of one Darcy. The development of an in-house capability for salt cake permeability determinations was begun.

A theoretical study of the retention of liquids in beds of solids disclosed that particle size range and packing can have a significant impact on the drainability of liquid from the solids. Preliminary evaluation of stimulation and immobilization techniques was completed, and additional work was initiated on the displacement of interstitial liquids from salt cake.

The safety of oil field shaped-charge jet perforating techniques for possible salt well flow stimulation was demonstrated. The heated carbon dioxide diffusion technique for salt cake drying was evaluated and found to be too slow for practical immobilization of tank liquids. Design work was initiated for construction of a Waste Management Pilot Plant to allow pilot-scale testing of various mechanical flow stimulation techniques.

Evaluation of Effort

With the exception of instrumentation and equipment problems associated with the 241-BY-107 Instrumented Salt Well Facility which have thus far precluded the generation of data suitable for permeable flow parameter prediction, efforts on other aspects of the program have shown a good rate of progress. Although the construction materials in

the standard unit are not compatible with the chemistry of interstitial liquids, the deep-well jet pump system appears capable of sustained, low-rate salt well pumping.

Displacement of interstitial liquid by an organic-surfactant fluid mixture is the only feasible stimulation of immobilization technique identified to date, although a more thorough evaluation of the technique is necessary. Oil field shaped-charge jet perforating techniques could be used safely in an installed salt well to treat permeability barrier in close proximity to the well bore.

Future Work

The 241-S-111 Instrumented Salt Well Facility will be installed and operated. Data will be obtained from both the 241-S-111 and 241-BY-107 facilities. These data will be used to develop a mathematical model suitable for predicting the flow of interstitial liquid through salt cake. The deep-well jet pump system will be retrofitted with suitable materials and additional loop testing will be performed, followed by possible hot testing in an operating salt well. Displacement and *in situ* fixing of interstitial liquid will also be evaluated.

TERMINAL LIQUOR DISPOSITION

Program Manager:	R. C. Roal
Section Manager:	R. J. Thompson H. Babad
Principal Investigators:	J. S. Buckingham R. I. Donovan D. R. Frazier W. P. Metz W. E. Ogren R. E. Van der Cook
Activity Codes:	B06T0, B06T1, B06T3

Objective and Scope

This program undertakes 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 three principal thrusts:

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

Prior Work

Evaporation

The 242-S and wiped-film evaporator volume reduction processes were studied on a pilot-scale. The liquors used in the tests were estimated to be terminal or non-evaporable by available techniques. The wiped-film evaporator operated satisfactorily. The 242-S pilot system experienced potential problems in plant operation because of

the high solids content of the liquors after extensive boildown.

Partial Neutralization

Laboratory studies were performed to investigate the chemistry of the neutralization-evaporation process. A pilot vacuum evaporator-crystallizer was used to more accurately model the 242-S system and to define more precisely the chemical behavior of simulated waste when partially neutralized. Product quality criteria for 242-S partial neutralization were defined and documented.

In-Tank Clay Solidification

Laboratory testing of the in-tank clay solidification process was completed and pilot-testing of the process with synthetic liquors was initiated.

Progress During Report Period

Engineering Studies

An engineering study of alternatives for disposition of terminal liquors for interim storage was completed. The goals of the study included identifying processing alternatives which appear feasible in terms of timely process development, thus reducing the number of double-shell tanks needed for interim storage of liquid wastes. The study identified the following feasible alternatives:

- Partial neutralization with nitric acid.
- Selective aluminum removal.
- Semisolid production by vacuum evaporation.
- Semisolid production by wiped film.
- Solidification by clay addition in-tank.

Each of these alternatives is being further developed to permit selection of the method of terminal liquor disposition which best meets interim waste storage requirements and minimizes the number of double-shell tanks required.

A computer program (NEWVAP) has been developed which calculates rates, volumes, and concentrations in all 242-S evaporator-crystallizer process streams. This program aids in optimization of current 242-S concentration and partial neutralization efforts and has been used to calculate preliminary liquor and solids volumes for the partial neutralization.

Evaporation

The quality of the material produced by evaporating waste solutions in Battelle-Northwest's wiped-film evaporator was determined. The product slurries ranged from 14 to 17 wt% solids at 93° to 94° C, increasing to 63 to 85 wt% upon cooling to room temperatures. The sodium hydroxide content of the liquid phases ranged from 11 to 16 molar while sodium hydroxide content of the solid phases ranged from 12 to 17 wt%. Thermal conductivities of the product material averaged 0.94 W/m °C.

Terminal liquor compositions were estimated on the basis of tank sample analyses. The compositions are significantly different from earlier estimates and necessitate reevaluating and rerunning of the pilot studies of terminal liquor boildown.

A proposal for testing the pilot-scale wiped-film evaporators at Savannah River Laboratory was submitted to and accepted by E. I. du Pont de Nemours and Company. The tests were scheduled for April 1975.

Use of the 242-S Pilot Plant in 221-T Building was discontinued because of its large chemical and manpower requirements.

The assembly of smaller bench- and pilot-scale evaporators is in progress to continue evaporation studies.

Partial Neutralization

Laboratory testing of four proposed partial neutralization flowsheets for terminal liquor showed that the solid phase produced was essentially sodium nitrate contaminated with a small amount of mother liquor. A significant volume of sodium nitrite was found in the solids formed when the flowsheet requiring the greatest degree of neutralization was used. The volume of solids produced ranged from 14.5 vol% (based on feed vol%). The volume of "pumpable" liquid produced ranged from 49 to 77%.

Design of a continuous flow vacuum evaporator that can be used as an aid in partial neutralization studies was completed. The unit should provide an accurate means of defining the chemistry and amount of crystal formation during neutralization-evaporation.

A partial neutralization process test is scheduled for June 1975. The test will feature double-pass processing of a batch of bottoms liquor with a settling period between passes. This will assist in obtaining representative samples of the liquor and solids from each pass and demonstrate the operating characteristics of the 242-S Evaporator-Crystallizer during acid addition.

Engineering support studies of partial neutralization of synthetic terminal liquor with nitric acid have been carried out in the pilot evaporator-crystallizer. Partial neutralization resulted in a significant volume reduction of

the terminal liquors fed to the evaporator-crystallizer. Examples of the volume reductions (based on the original feed volume) are as follows: a 15% neutralization of the feed sodium hydroxide reduced the volume to 82%, producing a slurry containing 10% solids; 43% neutralization reduced the volume to 36%, producing a slurry containing 23% solids.

In-Tank Clay Solidification

A prototype testing program has been proposed to demonstrate and evaluate the merits of the clay solidification process in a plant-scale.

Evaluation of Effort

Investigation of the three methods of treatment of terminal liquors (evaporation, partial neutralization, and clay solidification) included in this activity will define their usefulness for process service. At this point it appears that each of the three possibilities could provide reliable containment of the terminal liquor for an interim period. However large-scale testing is necessary prior to final selection and application on a Tank Farm-basis.

Future Work

Results of the wiped-film evaporator test at Savannah River Laboratory will be evaluated to aid process flowsheet development and equipment design. The operating conditions of the wiped-film evaporator will be correlated with product characteristics to insure that the material produced is suitable for storage in double-shell tanks.

Future laboratory investigations and subsequent engineering flowsheet refinements will help to better define the operating parameters that may be attained with partial

neutralization. A pilot-plant investigation will be made to more closely simulate the chemical behavior of actual 242-S-type feeds and to verify laboratory partial neutralization data.

A prototype test of the clay solidification process will be carried out by the end of calendar year 1975. The test will provide a basis for evaluating future application of the process.

ALUMINUM REMOVAL FROM HIGH-LEVEL RADIOACTIVE WASTE

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

Objective 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 objective, development and demonstration of techniques to remove aluminum from terminal liquor, has the potential of reducing the number of double-shell storage tanks required for interim storage.

Prior Work

In anticipation of a need for aluminum removal from terminal liquors, a program was undertaken to define

aluminum solubility in waste solutions and to identify methods for controlling aluminum precipitation to produce acceptable interim storage solids. Laboratory investigations and engineering studies were directed toward developing feasible alternatives for aluminum removal.

Progress During Report Period

Engineering Studies

To determine possible process application methods to the Hanford waste system, conceptual flowsheet development was pursued based upon literature information and preliminary laboratory results. Identified process alternatives include the Bayer process for removing aluminum as aluminum oxide trihydrate (Gibbsite) or as sodium aluminosilicate (cancrinite), a mineral form.

Use of major existing facilities may help to meet the goal of timely availability of aluminum removal processing. During flowsheet development the 242-S or 242-A Evaporator facilities, AR Vault-type facilities, and 3.8-million-liter tank facilities have been considered as potential process equipment.

The need for a consultant's services for Bayer process development was identified. A consulting contract, to include experimental investigation of proposed flowsheets with synthetic waste solutions, is being initiated.

Laboratory Studies

Solubilities of $NaAlO_2$, $NaNO_3$, and $NaNO_2$ were determined as functions of temperature and NaOH concentration. Aluminum is more soluble in a system containing sodium nitrate and sodium nitrite than in the pure

Na_2O-Al_2O_3-H_2O system. In addition, the maximum aluminum solubility is shifted to a lower NaOH concentration.

Laboratory work showed aluminum could be removed from terminal liquor by reacting with silica according to the following reaction:

$$NaAlO_2 + SiO_2 + xH_2O \rightarrow NaAlSiO_4 \cdot xH_2O$$
(cancrinite)

The effectiveness of the reaction depends on the surface area. For example, diatomaceous earth, which has a small particle size and a large surface area per unit weight, will remove 97 to 98% of the Al. However small particles of diatomaceous earth result in a high percentage of interstitial (nonpumpable) liquor. Fourteen sources of silica have been surveyed to date in the search for a reactive silica that will produce a large, solid cancrinite particle.

In a laboratory demonstration of the Bayer process, Al was precipitated as Gibbsite ($Al_2O_3 \cdot 3H_2O$) crystals from a solution containing only Na_2O-Al_2O_3-H_2O. In one week the particle size of the Gibbsite increased from 32 μm in the seed crystals to between 60 and 70 μm. Approximately 16% of the aluminum in solution was precipitated.

Evaluation of Effort

Development efforts to date indicate that aluminum removal from Hanford waste liquors is feasible. Additional work is required to define a plant flowsheet which can be used to determine the overall benefits of this program.

Future Work

Laboratory investigations and engineering flowsheet development will be continued and pilot-plant studies may be initiated to better define promising alternatives for plant-scale aluminum removal. The impact of aluminum removal alternatives on the Hanford Waste Management Program will be investigated.

SALT CAKE CHARACTERIZATION

Analytical Development

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

Objective and Scope

This program will develop appropriate physical and chemical techniques for analyzing the samples obtained by core drilling of salt cake in the waste tanks. The cake cores will be analyzed as they become available.

Prior Work

Past work has consisted of assessing the laboratory requirements for receiving various-sized salt cake cores and determining the modifications to existing analytical procedures or the need for the development of new and more sophisticated techniques.

Progress During Report Period

The conceptual design of a remotely operated facility for receiving and breaking down the core samples was completed, as was the initial scoping of the requirements for the temporary and permanent storage of the salt cake cores.

A number of analytical procedures and techniques have been revised and/or modified for the salt cake analyses. These include development of a satisfactory method for determining plutonium in the salt cake, completion (in draft-form) of procedures for determining the physical characteristics of the salt cake, and organization of a standards and referee program.

Several pieces of equipment specifically for the characterization have been ordered and received.

- A twin cell microcalorimeter has been received and is being calibrated.
- A Hassler cell is being modified to determine salt cake permeability. Preliminary tests indicate that the unit gives satisfactory results.
- An HIVAC particle counter has been set up to determine particle size and distribution.
- A carbon analyzer has been ordered to determine total acid and organic carbon in the salt cake.

Evaluation of Effort

The laboratory is now in a position to provide analyses of the salt cake as required by the core drilling program.

Future Work

Although minor analytical equipment and procedure revisions may be required, future work will consist principally of obtaining the analytical data.

Core Sampler Development

Program Manager:	R. C. Roal
Section Manager:	R. J. Thompson
Principal Investigators:	M. D. Martin V. L. Schuelein
Activity Code:	B06T6

Objective and Scope

Design, development, and demonstration of equipment capable of safely and efficiently retrieving representative samples of in-tank, high-level waste is the objective of this program.

Prior Work

A concept was developed to use conventional rotary core drilling equipment employing a wire line sample tube retrieval technique. Preliminary design work was started on modifications to the commercial equipment and on various support equipment, such as the shipping and transfer cask.

A tentative decision was made to utilize smaller diameter core barrels than originally planned during the concept development phases. The smaller core barrels, which produce a sample approximately 3.5 cm in diameter, would simplify handling and shielding requirements.

Progress During Report Period

Preliminary tests were completed to demonstrate the basic concept and to compare the drilling and core retrieval characteristics of the smaller size core barrels with the originally conceived larger core barrels.

Drilling was performed in synthetic salt cake, portland cement, and gypsum. There were no significant differences found in the characteristics of the two drill sizes. Successful core samples of all the materials were obtained. The parameters which influence the initial phase of core drilling include the material surface, how hard the waste is, proper stability in the drilling boom, optimum rotational speed, and downward pressure.

Additional preliminary tests were conducted to determine the conditions necessary for the drill to damage the tank bottom liner. The tests proved that serious damage could occur under certain conditions and also identified several methods to prevent such damage. It was found during the tests that a sudden change in pressure occurs when the drill bit encounters the metal bottom. This change in pressure can be identified by the altered downward rate of drill travel. Another method which has potential is to set up a resistance circuit composed of the tank liner and core drill. Upon contact of the core drill with the liner, a drop in circuit resistance would shut off the drill.

All equipment drawings were completed and work orders were issued for fabrication of support equipment and for modification to the basic core drilling equipment.

Evaluation of Effort

The preliminary tests have proven that the rotary core drill-wire line retrieval method is a viable technique for recovering representative samples from the waste tanks. They have shown that further effort is needed to develop a reliable system for detecting contact with and preventing damage to the tank bottom liner.

Future Work

Upon completion of all equipment fabrication, further preliminary tests will be made to check mechanical operability. When all equipment has been demonstrated to be mechanically operable, an integrated system cold test will be conducted with conditions set up to simulate as closely as possible the actual tank situation. Operating procedures will be developed to assure proper radiation and contamination control and to prevent damage to the tank liner. If the cold integrated system test is successful and no major changes in equipment are required, the standard operating procedures for hot waste tank sampling will be completed and the first hot demonstration of core sampling will be conducted.

TANK OPERATIONAL CRITERIA

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

Objective 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 progrm is to develop criteria for tank usage so that operating specifications and standards can be established for filling the tanks with salt cake.

Progress During Report Period

Thermal creep time-dependent analyses of the 241-SX and 241-U reinforced concrete tank structures that are sustaining soil loads have been attempted 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 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. An earth cover thickness of 1.8 m at the dome crown and heat-up rates for the tank structure of about 3° C per day were assumed.

Thermal creep analyses were carried out at the 241-SX

Tank Farm under the following conditions:

- Heat generation rate (Q) = 2.6 W/m^3.
- Q = 2 W/m^3.
- Q = 3 W/m^3 with the vapor space temperature controlled to 100° C by air ventilation.

In the first two cases the thermal creep deflections did not appear to limit with time, leading to predictions of extensive cracking throughout the concrete. In the third case temperatures above 163° C were indicated only in the base of the tank. It is unlikely, however, that the calculations will support operation, even in this case.

For the 241-U Tank Farm analyses, 2.1 m of earth cover load for the dome crown and heat-up rates of 2.8° C per day for the tank structure were assumed. The conditions used for the thermal creep analyses were:

- Salt heat generation, Q, = 2.5 W/m^3.
- Salt heat generation rate, Q, = 4.5 W/m^3 with the vapor space above the salt controlled at 100° C.

The first case (2.5 W/m^3) produced a peak wall temperature of 143° C and the second case (4.5 W/m^3) produced a peak wall temperature of 174° C. Both of these cases exceed the current temperature limitation of 110° C for 241-U Tanks. Evaluation of cracking and deflections are not yet complete.

Evaluation of Effort

It is anticipated that recommendations for updating the 241-SX temperature and loading limitations will be delayed by about six months because of the additional thermal creep analyses required.

Future Work

Based on the results obtained to date, additional analyses and evaluations of the 241-SX Tank Farm reinforced concrete structure are planned, assuming temperatures in the wall of about 170° C and heat-up rates (above 90° C) of 0.3° c per day.

WASTE TANK LEAK DETECTION

Program Manager:	R. C. Roal
Section Manager:	R. J. Thompson
Principal Investigator:	H. F. Jensen
Activity Code:	B1440

Objective 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

Experience gained in the past few years has increased our capability to monitor waste tank contents and maintain close surveillance of the waste tank environment. Instrumentation for these purposes has become increasingly sophisticated and data handling has improved.

The liquid level gauges have been incorporated into an automatic readout and reporting computer system and manual readouts have been gradually phased out. Ground surveillance is being scheduled on a weekly basis and problems associated with this accelerated pace are being resolved.

The geometry of waste tank environmental monitoring with dry wells has been improved to give a capability of detecting tank leaks of 10,000 liters. Installation of the dry wells was begun.

Progress During Report Period

A Nova 840 mini-computer became operational in January and has provided an immediate data processing facility for waste tank environmental surveillance data. Monitoring data can be taken from the paper tape, transferred to the computer, and plotted on a visual display for immediate analysis.

Several devices for determining the level of liquor remaining in the salt cakes after concentration are being evaluated: ultrasonic and radio frequency devices, weight factor methods, and the present gauge in a standleg. A dry well has been fabricated for testing the potential of a neutron probe as a liquid level monitor.

Efforts are in progress to explore means for detecting tank leaks by monitoring soil conductivity around the tanks. Both the Boeing Company and Battelle-Northwest are involved in this effort, and are using somewhat different techniques.

Corrosion measuring probes for investigating waste tank integrity have been installed in nine waste tanks. These results will allow comparison of the relative corrosivity of the wastes accumulating in the three waste concentration types [*i.e.*, the in-tank solidification, the 242-T (atmospheric pressure) evaporator, and the 242-S (vacuum crystallizer evaporator]. The feed tanks of the two evaporators and selected bottoms tanks of the three systems are being investigated.

Preparations are being made to sample and perform nondestructive surface inspections of a number of waste tanks which have been declared leakers. Development of metal surface scanning equipment is in progress at Battelle-Northwest. Ultrasonic scanners for detecting stress corrosion cracking are nearing completion. A second ultrasonic device for measuring metal thickness is being designed. A demonstration of sampling procedures and examination techniques is planned for 241-SX-108. Additional tanks to be inspected include 241-A-104, 241-BY-108, 241-BX-102, 241-T-106, 241-C-111, and 241-TY-106.

Evaluation of Effort

Availability of the Nova 840 has markedly shortened the time involved in processing raw monitoring data. The previous computer facility required one week to return processed data.

Efforts to detect salt cake interstitial liquids are proving difficult. The radio-frequency effort has been terminated due to spurious signals and interference from temperature variations. The ultrasonic development has experienced serious attenuation of signal. A number of modifications have been made to the original equipment and an in-tank prototype is being redesigned for additional testing of the laboratory equipment.

Mounting of liquid level weight factor and conductivity gauges in a water-filled standleg was abandoned due to continuous problems with plugging of sensing lines and stabilization of the standleg liquid. The test was modified to use an immiscible liquid instead of water. Experience to date indicates that the weight factor approach shows some promise and that the conductivity probe may be feasible

under different circumstances, but it is unsatisfactory in the present housing. New units are being prepared for installation in Tank 241-S-111.

Soil conductivity monitoring techniques being developed by Boeing and Battelle-Northwest have demonstrated an extremely high degree of sensitivity to changes occurring in the vicinity of the waste tanks being monitored. Since duration in signal may indicate a tank leak or may be caused by entirely unrelated moisture effects, the high sensitivity may give spurious results. Characterization tests are still being performed to generate the necessary comparative data for analyzing surveillance data.

To date sampling of waste tank concrete and steel has been confined to planning and preliminary procedures.

Future Work

Work already in progress will be continued. An additional method of monitoring leaks in waste facilities such as transfer lines is being considered. This will involve directly monitoring the operation of the extensive cathodic protection system. It has been suggested that this existing system may provide access to the same type of information being sought through the Boeing and Battelle-Northwest programs.

Waste tank component sampling will proceed as manpower and schedules permit. The in-tank corrosion probes will be left in place for at least a year before selected units are retrieved.

SOIL CONDUCTIVITY LEAK DETECTION

Program Manager:	R. E. Isaacson
Coordinating Engineer:	L. E. Bruns
Principal Investigator:	K. T. Key
Activity Code:	B05T3

Objective and Scope

In this program the sensitivity of the Boeing and Battelle-Northwest (BNW) leak detection systems which are based on the principle of potential field distortion will be determined. Quantitative correlations between the simulated leak quantities and the quantities measurable will be established. This capability will be used to predict the occurrences of small leaks and the quantity released by these leaks.

Prior Work

The Battelle-Northwest and Boeing systems were tested in a simulated Tank Farm in 200 West Area. The simulated Tank Farm was constructed with a series of dry wells 9 to 15 meters deep and cased with 100 mm black iron pipe. Wells were laid out in cylindrical patterns to simulate the tank wall (by connecting inner dry wells with #1 wire), Tank Farm dry wells, and the surrounding Tank Farm ground (by connecting outer wells with #1 wire). Based on tests using these two approaches, a logarithmic relationship was established between the leakage and the measurement.

Progress During Report Period

Initial field tests in Tanks 103-BX, 102-BX, and 101-BX revealed some unforeseen problems with interconnecting pipelines, cathodic protection systems, and other perturbations.

Efforts by BNW and Boeing to solve the problems were largely successful and five tanks have been evaluated and patterned.

The final tests were conducted with baseline measurements taken for several days before and for about a week after the tests. The results indicate that both systems are normally capable of detecting leaks of less than 380 liters of waste liquid within hours after an occurrence. Some difficulty was encountered in detecting small leaks in tanks that previously had experienced large leaks from either pipelines or neighboring tanks.

A pipeline leak was simulated to test both the BNW and Boeing systems' applicability for detecting salt or water leakage from transfer or water lines. A 102-m segment of pipeline was used. Both the Boeing and BNW equipment detected transfer line leakage as low as 38 liters; only the BNW system succeeded in detecting water line leakage.

Evaluation of Effort

Field tests of the tank leak and pipeline detection systems proved the feasibility of both the Boeing and Battelle-Northwest methods to detect small leaks. The test series and the experience gained in assisting the Tank Farm management showed the complementary nature of the two approaches in providing an early warning capability for Tank Farm operations.

Future Work

Further work on segmented electrodes to profile the leakage vertically is recommended. This technique can be used to distinguish between a tank leak and a leak from the interconnecting pipeline.

CONTAMINATED SEDIMENT CONTROL

SURFICIAL CONTAMINATION--CRIBS, TRENCHES, SWAMPS, PONDS

Program Manager:	R. E. Isaacson
Coordinating Engineer:	L. E. Bruns
Principal Investigators:	K. T. Key P. J. Wiater G. M. Holter M. Pentilla
Activity Codes:	B0961, B0962, B0964

Objective and Scope

One objective of this program is to characterize a typical waste area using existing techniques and those developed under the Public Protection Assurance program. A second objective is to measure and evaluate parameters important to transport mechanisms (*e.g.*, changes in ^{137}Cs subsurface profiles with time, plutonium properties important to transport mechanisms). A third objective is to develop methods that can reduce movement of activity in selected waste areas.

Prior Work

Aerial surveys were made with helicopters in the BC Crib environs, U Pond, Gable Mountain Pond, S-16 Redox Swamp, S-17 Redox Pond (covered), S-19 Laboratory Waste Pond, and the Z-19 Ditch to determine the gamma-emitters that contaminate these surface areas. Programs on the U Pond and Gable Mountain were cooperative efforts with the Division of Biomedical and Environmental Research (DBER) and Battelle Pacific Northwest Laboratories (BNW). Over 1,000 determinations were made from U Pond samples. Initial work on physical and chemical properties included determination

of radionuclide particle sizes in U Pond and the BC Crib and chemical species in U Pond.

Progress During Report Period

Surface material from U Pond (soil, live vegetation, and organic debris) is being characterized for radionuclides (^{241}Am, ^{137}Cs, ^{60}Co, ^{239}Pu, and ^{90}Sr). The analytical results from three of the twenty 1-m^2 plots near U Pond showed ^{60}Co to be near or at background and the other radionuclides above background levels. Concentrations at the 200 East Area Fire Station and the Hanford perimeter (average) were also at background levels.

A Redox swamp area, S-16, ($\sim$24,000 m^2) was characterized in three dimensions (X, Y, and Z) using random core and ring sampling techniques. The results from about 500 measurements and calculations were:

- About 6 Ci of ^{137}Cs was in the top 30 cm.
- Less than 1 Ci of ^{90}Sr was in the top 30 cm.
- An estimated 4 g of Pu was in the top 30 cm, and was concentrated in the channel area ($\sim$3,870 m^2).
- Less than 1 g of ^{241}Am was in the top 30 cm.
- An increase of both ^{137}Cs and ^{239}Pu occurred in several areas to 30 cm (the highest Pu value found thus far was 4.8 nCi/g of soil and the highest Cs value was 2 nCi/g of soil).
- The ^{137}Cs concentration in the Redox Swamp was slightly less than in the BC Crib area and ^{90}Sr two to three orders or magnitude below that found in the BC Crib surface contamination.

Approximately 10 random, 30-cm cores were taken from

the radioactive zone of the old S-17 Redox Pond area previously covered with dirt to obtain estimates on the subsurface radioactivity.

Initial studies were also made on the S-19 Pond area where several 30-cm core samples were taken.

Evaluation of Results

The ARHCO-BNW-DBER program indicates significant buildups in algae floc and various plants and organisms over a two-year period but nothing that, at this time, poses a problem to man. With the buildups to date and the dose rates in certain areas, it may be well to consider an in-place cleanup of the Pond (see Decontamination/Immobilization).

The work summarized here provides valuable information concerning the level, nature, and behavior of surficial contaminants in the S-16 Redox Swamp area. Highly variable concentrations show the need for careful statistical evaluation of the surface concentrations.

Information obtained from the Redox Swamp characterization, coupled with data from the Resuspension Program, suggests that with high winds (particularly if the area were burned over before the wind) fallout will be exceeded locally, but not MPC_{air}.

Future Work

The X-Y-Z-characterization work will continue in the S-16, S-17, and the BC Crib areas and will be initiated on the Z-19 Ditch. The ARHCO-BNW-DBER program will continue in fiscal year 1976. Atlantic Richfield Hanford Company will expand the U Pond program to include radionuclides other than Pu-Am (the constraint of the DBER program).

Work will continue on the Gable Mountain Pond characterization. Continuing efforts will be made on physical and chemical property studies of U Pond, BC Crib, and Gable Mountain Pond and initiated on Z-19 and S-19.

SOIL STABILIZATION

Program Manager:	R. E. Isaacson
Coordinating Engineer:	P. J. Wiater
Principal Investigators:	L. E. Bruns G. M. Holter K. T. Key
Activity Code:	B09T1

Objective and Scope

The objectives of this program are to (1) demonstrate that radionuclides on surfaces of all types (*e.g.*, pristine desert land, worked land, and burned-over areas) can be affixed by chemical agents in combination with ground covers, such as cheatgrass; (2) demonstrate that subsurface radionuclide-contaminated regions can be affixed in place by chemicals such as asphaltic emulsions; (3) demonstrate that subsurface pipe and tank leaks can be stopped by grouting; and (4) investigate new affixing agents, preadditives, and methods of injection.

Prior Work

Literature surveys were made for both surface and subsurface groutings. Laboratory investigations using many different subsurface affixants (including AM-9, concrete, and asphaltic and silica emulsions) were completed. An AM-9 acrylamide injection test was successfully made in a waste

tank region. Laboratory tests were initiated on grouting material which will be used for wells extending to the water table. Several field tests were initiated using the surface affixant over an area seeded with cheatgrass.

Progress During Report Period

Surface Affixants

After several months of testing, the five most promising field-tested affixants were reduced to three: M-166, Aerospray 70, and Coherex. The best for all-around usage appears to be M-166, a latex emulsion; it holds the topsoil and still allows growth. More experience and testing are required to determine the amount of M-166 to apply for optimum moisture retention, longevity, vegetative growth, and soil containment.

Approximately 12,000 m^2 of covered swamp and pond area seeded with various grasses have been affixed with M-166 and some $\sim$1,000-m^2 plots seeded with cheatgrass are being tested with Aerospray 70 and Coherex. Grass growth has been spotty in the affixed areas; areas treated with bentonite and covered with straw have given the best vegetative growth, though erosion is a problem when a soil affixant is not used.

M-166 was also selected for stabilizing the BC Crib contaminated area in event of fire.

Subsurface Affixants

In an underground test, simulated terminal liquor was successfully immobilized using a combination of clay and asphalt emulsion. An asphalt emulsion was field- and laboratory-tested to simulate covering a swamp area. The

field tests indicated that a tank sprayer could affix a large area with an asphalt emulsion-water mix which later can be covered with soil. With hand-spraying of such a mix on a small test plot, the asphalt penetrated 1 to 2 cm, depending on the amount used. In one day the soil-asphalt mixture had become fixed.

In approximately 20 laboratory-simulated well groutings with concrete, asphalt emulsion, and mixtures of the two, an asphaltic emulsion grout following a water preadditive was shown to be most successful.

Evaluation of Results

Surface affixants show promise for holding the soil, yet allowing vegetative growth to prosper. A combination of mulch, water-retentive additives like bentonite, proper seed, and a proper affixant should give stable areas that will cover subsurface radioactivity for many years with little attention. Affixants can also be used to hold radioactivity on the surface in case of accidents. Work is still required to identify the best affixant for the particular problem.

Work on subsurface affixants is promising for stabilizing radionuclides in the soil.

Future Work

The S-16 Swamp will be affixed with an asphaltic emulsion over the area contaminated with greater than 1 nCi Pu/g of soil. The area will be covered with about 1 m of soil, mulched, seeded, and affixed with a surface agent.

Laboratory tests will continue on surface affixations.

Subsurface soil stabilization tests are being planned.

Initial tests will involve simulated areas where radioactivity is not present. Methods of adding affixants and measuring completeness of coverage will be devised.

The well subsurface grouting will be sampled periodically.

Leak prevention tests of subsurface piping are being planned using an asphaltic emulsion. New methods of injecting the affixant will be studied.

DECONTAMINATION/IMMOBILIZATION

Program Managers:	R. E. Isaacson R. L. Dillon (BNW)
Coordinating Engineer:	L. E. Bruns
Principal Investigators:	K. T. Key P. J. Wiater W. W. Schulz N. J. Englund P. L. Kochmstedt (BNW) E. M. Woodruft (BNW)

Objectives and Scope

The objective of this program is to develop new methods of cleanup and/or immobilization of contaminated soils, including surface vegetation, detritus, and contaminated subsurface soils. For both surface and subsurface areas the research investigation will characterize selected areas, assess the hazards, and develop processes to clean up and immobilize the areas.

Work on subsurface contaminated volumes (such as cribs, trenches, and buried ponds) will concentrate initially on stabilization *in situ* (see Soil Stabilization). On the basis of soil stabilization studies, a complete waste entity

will be "cocooned" *in situ* or the contaminated soil will be removed, affixed, and stored underground.

Prior Work

The surface soil vacuum cleaning system was successfully "cold" field-tested and a spray system for reducing dusting during vacuum cleaner unloading designed. Also a boom system was designed to support the vacuum hoses and improve nozzle handling. A 12 m^2 vacuum system was demonstrated on the BC Crib area by Power Masters of Portland, Oregon. Laboratory studies demonstrated methods of affixing the waste *in situ* (see Soil Stabilization) or after soil removal. Various methods for cleaning up the ponds were reviewed including dredging, clarification, sedimentation, decanting, and affixing the removed sludge.

Progress During Report Period

Methods for determining the loss of the vacuum system filter integrity were initiated. Loss of one of the ten filter socks did not significantly change the pressure drop. The holding tank for the water spray system which will be used to control dust in the vacuum system was fabricated; the design on the boom system was finalized.

Films depicting the use of a mudcat for pond cleanup were shown. A more simplified flowsheet was devised which involved sending the sludge mixed with a fixing agent directly from a mudcat to an affixed V-ditch, separating the liquid and solids in the ditch by a sedimentation technique, and decanting the water back to the pond.

An initial study was made of removing and immobilizing surface and subsurface contamination. A unit that removes

radioactively contaminated soil and affixes it with bitumen is being investigated.

Evaluation of Results

The vacuum system is available for emergency situations (*e.g.*, following a range fire on BC Crib); however the spray system, boom-mounted nozzles, and further testing for particle size retention are needed before a routine system is available. Pressure drop changes across the vacuum filter system are inadequate as a measure of filter integrity. The boom-system design allows much easier handling of the nozzles; presently they are very tiring to operate.

Evaluation of U Pond and Gable Mountain Pond characterization studies to date indicate that the best method for preventing concentration of radionuclides in organisms and reducing high exposure areas is to remove the pond bottoms and contaminated shore areas without putting the pond out of service. This might be done by use of a mudcat or dredge system. Resuspension problems are expected to be minimal because the sludge is always associated with considerable water.

Immobilization *in situ* appears to be one of the best approaches to restricting the movement of radionuclides in the soil.

Future Work

The next six months' work will include installation of the spray system on the mobile vacuum cleaner, development of a method to ascertain filter integrity, a "cold" pond mudcat test preparation, and use of *in situ* instruments to denote decontamination cleanup.

BIOLOGICAL UPTAKE

Radionuclides in Soil-Plant Systems

Program Manager:	R. E. Isaacson
Section Manager:	R. E. Isaacson
Coordinating Engineers:	V. A. Uresk E. L. Klepper (BNW)
Principal Investigator:	R. C. Routson (BNW)
Activity Code:	B09TE

Objectives and Scope

Quantitative information concerning the fate and behavior of radionuclides in the soil-plant system is necessary to formulate and maintain ecologically sound waste management practices.

Prior Work

Studies in growth chambers have shown that the chemical form of transuranic wastes affects their uptake by plants from soils. Plant uptake was observed to increase when transuranics were complexed with organic acids or chelated with ethylenediaminetetraacetic acid (EDTA) or diethylenetriaminepentaacetic acid (DTPA).

Studies on shoot uptake by tumbleweed and cheatgrass plants ranging in age from 2 to 16 weeks showed that uptake of ^{237}Np, ^{239}Pu, ^{241}Am, and ^{244}Cm added in dilute nitrate solution to the soil decreased with increasing plant age, but tended to level out at 6 to 10 weeks. The relative uptake of the radionuclides is Np>Cm>Am>U>Pu.

Shoots of cheatgrass and tumbleweed plants grown in soil to which $^{238}PuO_2$ and $^{239}PuO_2$ were added contained barely detectable ^{238}Pu and undetectable (<6 pCi/g) ^{239}Pu.

The root tissue had measurable radioactivity, however, with cheatgrass containing more than tumbleweed.

Progress During Report Period

The concentration factors

$$CF = \frac{\mu Ci/g \text{ oven-dried plant}}{\mu Ci/g \text{ oven-dried soil}}$$

for tumbleweed were measured as a function of soil radionuclide concentration and plant age. Uptake was measured using ^{134}Cs and ^{85}Sr at four equivalent ^{137}Cs and ^{90}Sr concentrations ranging from 8.5×10^{-4} to 1.6×10^{2} μCi/g and 3.9×10^{-2} to 1.4×10^{2} μCi/g, respectively.

Tumbleweeds were planted in pots containing 1 kg of radionuclide-amended, thoroughly mixed soil and placed in growth chambers. The plants were harvested at two and three months and analyzed. Results for strontium showed CF values of 9.6 and 19 for eight- and twelve-week-old plants, respectively. Cesium results showed CF values of 0.033 and 0.053 for eight- and twelve-week-old plants, respectively. The CF values were independent of soil concentration over five orders of magnitude for strontium and three for cesium. This suggests that CF values might be extended to even lower soil concentration ranges. The CF values doubled between the eighth- and twelfth-week measurements, indicating that tumbleweed growing near waste disposal areas should be eradicated early in the growth cycle.

Uptake from a totally mixed soil system was compared with uptake from soil with a discrete layer of radioactivity. Tumbleweeds were planted in soil to which ^{85}Sr had been added as a layer and in soil with ^{85}Sr mixed throughout. In the totally mixed system CF values after four

weeks were 10.3 while in the layered system, CF values of 1.4 were found. Thus the totally mixed soil system is preferable for measuring CF values for leachable radionuclides.

Construction of a greenhouse facility for use in studies of plant uptake of radionuclides is scheduled for completion by July 1975.

Evaluation of Effort

The behavior of radionuclides in soil-plant systems is strongly influenced by their chemical forms, depth in the soil, and plant age. These studies imply that roots provide a mechanism for transporting buried radioactive waste to the surface, where it may become dispersed by wind or enter the food chain.

Future Work

Work will continue to identify factors which affect uptake and concentration of radionuclides in soil-plant systems. The effect of soil type and soil solution composition on uptake of Sr, Cs, Tc, Pu, and Am by tumbleweed and cheatgrass will be investigated in order to develop predictive capability for estimating biological availability on the basis of simple chemical tests.

Reports Issued

R. C. Routson, *The Effect of Soil Concentration on the Tumbleweed Uptake of ^{90}Sr and ^{137}Cs from a Burbank Sand*, BNWL-1905, Battelle Pacific Northwest Laboratories, Richland, Washington (1975).

Radionuclides in Terrestial Plants and Animals

Program Manager:	R. E. Isaacson
Section Manager:	R. E. Isaacson
Coordinating Engineers:	V. A. Uresk E. L. Klepper (BNW)
Principal Investigators:	J. F. Cline (BNW) J. D. Hedlund (BNW) W. H. Rickard (BNW) L. E. Rogers (BNW) R. G. Schreckhise (BNW) D. W. Uresk (BNW)
Activity Code:	B09TE

Objectives and Scope

These studies are providing knowledge about the ecology and behavior of plants and animals in the 200 Areas; and their actual and/or potential role in mobilization or transport of radioactive wastes.

Prior Work

Events associated with the penetration over 10 years ago of buried radioactive wastes by animals resulted in the dispersal of cesium and strontium over several hundred acres of unoccupied land near the B-C Cribs area. The contamination was spread primarily through the deposition by black-tailed jackrabbits of "hot" fecal pellets and urine spots. The resulting pattern of radiocontamination was determined by a ground survey, and occurrences of hot spots was mapped. Study sites near the B-C Cribs controlled zone and Redox Swamp area were established for ecological characterization studies of small mammals, insects, and plants.

Progress During Report Period

A study characterizing plant communities on the

200 Areas Plateau was completed and documented. This study analyzed the vegetation in two replicate areas at each of two study sites: near the BC Cribs controlled zone and Redox Swamp areas. Species frequency, shrub size, canopy cover, macronutrient and micronutrient composition, density, and soil composition were determined. A total of 29 species of plants was found at both areas combined, with 23 occurring at the B-C area and 22 at the Redox area. The main vegetation type was the sagebrush/cheatgrass-Sandberg bluegrass association. The amount of biomass produced by understory plants at the B-C site was 40 g/m^2 per acre, which is lower than that found in other kinds of eastern Washington plant communities. Approximately 0.25 of the below-ground biomass (roots) was found to occur in the upper meter of the soil profile. Soils at the two sites were similar and classified as sandy loam. Clay content was less than 2%, implying that moisture-holding capacity and cation exchange are low.

Small mammals have been live-trapped monthly since May 1974 in the study area adjacent to the BC Cribs controlled zone. One hundred Sherman live-traps were located on a 10 x 10 grid at 10-m intervals. Captured animals were identified, weighed, their sex and reproductive condition ascertained, and their toes clipped to permit recognition during subsequent trapping sessions. This procedure allows evaluation of reproductive success, home-range area, and population structure. During the Fall of 1974 a substantial increase in trapping success was noted due to addition of young of the year into the population. The Spring 1975 emergent population was much greater than that of 1974. The Great Basin pocket mouse comprised 95% of the 361 mice trapped to date.

Within the B-C Crib and Redox Swamp study areas, the survey of invertebrate populations was also continued. Transfer pathways are being defined through determinations of food habits and behavior of common species such as grasshoppers and ants.

A colony of harvestor ants on the 216-A-24 Crib was excavated to determine tunneling depth and chamber distribution. The deepest tunnel extended to a depth of 2.4 m and a total of 688 cm^3 of soil was excavated and brought to the surface by the ants. A collection of insects is being established to provide a taxonomic reference for future ecological studies.

Food habits of the black-tailed hare are being identified by comparing plant fragments in fecal pellets with a reference collection of plants growing in the study area. Fecal pellets were collected in two study areas--one near the BC Cribs and one near the Wye Barricade. Microscopic analysis of the pellets revealed the presence of 16 species of plants in the diet. Yarrow was the preferred species, comprising over 25% of the diet. Other important contributors to the diet were cymopterus, erigeron, needle-and-thread, and tumble mustard. Cheatgrass, the dominant herbaceous species, and tumbleweed, a known accumulator of ^{90}Sr, do not occur in the jackrabbit diet.

Information relevant to waste management on habitat requirements, food habits, burrowing behavior, position in the food chain, and potential for long-distance dispersal of contamination was summarized for the 25 relevant species of mammals living on the Hanford Reservation. The 8 species believed to be of most importance to waste management are muskrat, raccoon, mule deer, coyote, badger, ground squirrel, black-tailed hare, and Great Basin pocket mouse.

Evaluation of Effort

Ecological characterization of plants, mammals, and insects near the BC Crib controlled zone and Redox Swamp areas is nearly complete. Significant progress is being made in identifying radiocontaminant pathways through the food chain by dietary analyses of the black-tailed hares, beetles, and grasshoppers. Key mammals in waste management areas were identified.

Future Work

Food habit studies of common insects will be completed and documented. Prey items of birds of prey on the Reservation will be identified and radioactivity levels assessed. The mammal census near the BC Cribs will be documented; mammals living near waste ponds will be sampled and their body burden levels assessed.

Reports Issued

W. H. Rickard, J. D. Hedlund, and R. G. Schreckhise, *Mammals of the Hanford Reservation in Relation to Management of Radioactive Waste*, BNWL-1877, Battelle Pacific Northwest Laboratories, August 1974.

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Radionuclides in Aquatic Biota and Sediments

Program Manager:	R. E. Isaacson
Section Manager:	R. E. Isaacson
Coordinating Engineers:	V. A. Uresk E. L. Klepper (BNW)
Principal Investigators:	C. E. Cushing (BNW) R. E. Fitzner (BNW) W. H. Rickard (BNW) R. G. Schreckhise (BNW) D. G. Watson (BNW)
Activity Code:	B09TE

Objectives and Scope

In these studies the pathways of radioactive materials discharged to a cooling water pond are being followed through the aquatic food web, including waterfowl.

Prior Work

When experimental goldfish were confined to areas over sediments and plants of relatively high- and low-radioactivity, radiochemical analysis revealed that these goldfish confined in the area of higher activity accumulated over two times as much ^{137}Cs as did the fish confined over the lower activity sediments. Concentrations of ^{137}Cs in goldfish over lower activity levels were similar to those found in native goldfish. Levels of ^{95}Zr and ^{60}Co were similar in all three groups.

Experimental ducks were also restricted to the ponds for studies of radionuclide accumulation.

Progress During Report Period

A study on radioactivity in native and experimental biota at Gable Mountain Pond was completed and

documented. The main source of radioactivity uptake by the biota was ^{137}Cs associated with the pond sediments. The highest concentrations of ^{137}Cs were found in the sediments at the northwest end of the pond. Over 90% of the ^{137}Cs was in the upper 5 cm of the sediment cores. Alpha-emitting radionuclides in the sediments were higher in the northwest end of the pond with maximum concentrations of ^{239}Pu and ^{241}Am of 8 and 1.4 pCi/g, respectively. Measurement of dose rates to thermoluminescent dosimeters (TLD) placed at the sediment-water interface ranged from 4 to 230 mR/day.

Experimental goldfish confined over low and high levels of radioactivity in the sediments attained equilibrium levels of ^{137}Cs at different rates, times, and magnitudes. Fish in the higher activity area attained peak concentrations of 600 pCi/g in three months as compared with 300 pCi/g reacted in two months in the lower activity area. Algae, macrophytes and detritus, which comprise the main food item of the goldfish, usually had higher ^{137}Cs concentrations than the fish.

The ^{137}Cs concentrations in experimental ducks restricted to the pond were approximately the same as those of resident coots, but significantly higher than those of transient waterfowl. The average concentration of ^{137}Cs in the muscle tissue of experimental ducks was 290 pCi/g, while for transient ducks it was 63 pCi/g and for resident coots, 480 pCi/g.

A 2.5-year survey of waterfowl usage of radioactive waste ponds was also completed and documented. The purpose of this study was to identify all avian species using waste ponds, to estimate their seasonal abundance, and to relate pertinent features of their life histories to waste management. Observations were conducted at weekly

intervals beginning in September 1971 at B, U, Redox, and Gable Mountain Ponds and beginning in February 1972 at West Lake. The observations continued through March 1974. During this period 126 species of birds were observed using the ponds and their associated vegetation.

Those species most important to waste management considerations are waterfowl which settle in fairly large numbers on the ponds during autumn migration and in some cases nest in the area. Their feeding habits offer the opportunity for ingestion of radionuclides from vegetation and aquatic sediments; and by migrating out of the area they may become part of the human diet because of the hunting season. Measurements of ^{137}Cs in the flesh of some waterfowl collected on the Reservation during the observation period showed higher levels than occurred in samples taken on the Columbia River but several orders of magnitude below MPC limits.

The American coot is the most abundantly breeding bird on 200 Areas waste ponds and offers a "worst possible case" in studies investigating the accumulation and transport of radioactive materials by migratory birds. A study was begun in June 1974 to determine the seasonal body burden levels and dietary habits of coots living on Gable Mountain, B, and U Ponds. Residence times and migratory distances are being studied by banding the birds with U. S. Fish and Wildlife Service tags. During this report period 63 coots were collected for radioanalysis and 9 were banded and released.

The average ^{137}Cs concentration for total body burden level of 26 coots collected at Gable Mountain Pond was 141 pCi/g wet weight with a maximum of 265 pCi/g wet weight. The average body burden level of ^{137}Cs for 15 coots collected

at B Pond was 4.34 pCi/g wet weight with a high value of 5.6 pCi/g wet weight. A control coot sampled on the Yakima River showed a total body burden level of 0.483 pCi/g wet weight for ^{137}Cs.

Radiochemical analyses of muscle, liver, and bones were completed for 12 coots collected at Gable Mountain Pond and 1 control coot collected on the Yakima River. Comparison of the Gable Mountain coots with the control coot showed the ^{137}Cs in muscle tissue to be 1627 times that of the control.

Evaluation of Effort

Aquatic biota appear to accumulate ^{137}Cs in proportion to the concentration in their food.

Equations used to assess dose rate to man show that a human consuming one of the average Gable Mountain coots per day (worst case) would reach an equilibrium level of 0.610 μCi or about 2% of the his maximum permissible body burden. Coots constituted 4.7% (246,800) of the total retrieved waterfowl gamebag based on Pacific Flyway 1971 data reported by the Bureau of Sport Fisheries and Wildlife; thus the probability of a Hanford coot being among those retrieved would be ∿0.12%.

Future Work

The investigation of body-burden levels and dietary habits of coots on waste ponds will be continued. Various aquatic components of the Gable Mountain Pond ecosystem will be sampled for radioanalysis in the continuing effort to understand cycling of radionuclides through the aquatic food web.

Reports Issued

C. E. Cushing and D. G. Watson, *Aquatic Studies of Gable Mountain Pond*, BNWL-1884, Battelle Pacific Northwest Laboratories, December 1974.

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ACTINIDE SEDIMENTS

ACTINIDE TRENCH CHARACTERIZATION

Characterization and Transport

Program Manager:	R. E. Isaacson
Section Manager:	R. E. Isaacson
Principal Investigator:	S. M. Price
Activity Code:	B0965

Objective and Scope

There is a need to characterize the current and future reactivities and distributions of actinides (plutonium and americium) beneath 200 Areas liquid waste disposal facilities to ensure that no hazard to the general public is posed by actinide waste management operations. The objectives of this program are to:

- Locate the zones of greatest actinide concentrations beneath liquid waste disposal facilities and assess the nuclear reactivities of these zones.
- Delineate the distribution of actinides beneath liquid waste disposal facilities.

- Characterize the mineralogical locations of actinides within the sediment samples.
- Provide a detailed characterization of the form and migration behavior of plutonium and americium 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 at the beginning of FY 1973. Since the beginning of the program, three test holes have been drilled, all under conditions of total containment. A test hole at the enclosed 216-Z-9 Trench was drilled to a depth of 14 m below the bottom center of the Trench floor. Another test hole was drilled to a depth of 33 m through the bottom of the "a" (Tile Field) section of the 216-Z-1A Trench adjacent to the initial entry point of the waste liquids. A third well was drilled to a depth of 11 m through the bottom of the "c" (Tile Field) section of the 216-A-1A Trench at the end of the distributor pipe.

Work with these wells can be categorized under three headings: reactivity assessment, actinide distribution, and mineral waste reactions.

Reactivity Assessment. The reactivity of the sediments penetrated by the two wells at the 216-Z-1A Trench was determined by neutron pulsing. For both wells the maximum reactivity occurred in a zone $\sim$0.3 m below the bottom of

the Tile Field. Calculations based upon preliminary findings indicate that the probability of attaining criticality at the 216-Z-1A Trench is essentially nonexistent if the Trench is left in an undisturbed state. [The nuclear reactivity of the 216-Z-9 Trench had previously been assessed before the initiation of the program as summarized in ARH-2915.[1]]

Actinide Distribution. Analysis of samples from the test wells indicated that the maximum measured actinide concentration occurred approximately within 0.5 m below the bottoms of both facilities. In the 216-Z-9 test well the general distribution of actinide content with depth is outlined as follows:

ACTINIDE CONCENTRATION, "H" BORE HOLE

Depth Below Trench Floor	^{239}Pu µCi/liter of Sediment	^{241}Am µCi/liter
5 cm	$\sim 1 \times 10^{6}$	$\sim 1 \times 10^{5}$
30 cm	$\sim 6 \times 10^{3}$	$\sim 6 \times 10^{2}$
7 m	$\sim 1 \times 10^{2}$	$\sim 3 \times 10^{1}$
14 m	$< 1 \times 10$	$\sim 1 \times 10^{-1}$

In general, the concentration of ^{239}Pu in the "H" bore hole decreases by ~6 orders of magnitude from the 5-cm to the 14-m depth. The water table lies at a depth of approximately 30 m below the bottom of the "H" hole.

Analysis of cores from the two wells drilled through the bottom of the 216-Z-1A facility indicated the following approximate actinide concentrations:

Depth Below Bottom of Tile Field meters	A Test Hole		C Test Hole	
	^{239}Pu μCi/liter of Sediment	^{241}Am μCi/liter of Sediment	^{239}Pu μCi/liter of Sediment	^{241}Am μCi/liter of Sediment
0.5	$\sim 5 \times 10^{4}$	$\sim 1 \times 10^{3}$	$\sim 1 \times 10^{4}$	$\sim 2 \times 10^{2}$
2	$\sim 5 \times 10^{2}$	$\sim 5 \times 10^{1}$	$\sim 1 \times 10^{3}$	$\sim 1 \times 10^{2}$
8		$\sim 3 \times 10^{1}$	$\sim 5 \times 10$	$\sim 5 \times 10^{1}$
11		$\sim 3 \times 10^{1}$	$< 1 \times 10$	$\sim 5 \times 10^{1}$

Concentrations of ^{239}Pu below the 2-m depth at the "A" site are below the detection limit for the analytical method employed ($< 5 \times 10^{2}$ μCi/liter).

In general, the concentrations of ^{239}Pu in the "c" test well decreases four orders of magnitude from the 0.5-m depth to the 11-m depth. The water table lies at a depth of approximately 40 m below the bottom of the "c" test well.

Mineral-waste Reactions. Portions of cores from all three test wells were submitted for characterization of actinide-waste-mineral associations. Mineral-waste reactions associated with alpha activity were located by exposing prepared 216-Z-9 sediment samples to alpha-sensitive film. The alpha activity generally appears to be associated with reaction rims enclosing fragments of metamorphic and basaltic sediment grains.

An examination of the sediments indicated the presence of at least two types of actinide occurrences. One consists of discrete PuO_2 particles varying from 5 to 100 μm in diameter that originated as part of the waste stream. The other type of plutonium occurs in lesser concentrations than the PuO_2 particle type but extends to a greater depth in the sediment column. The form of this second type of Pu and its stability have not been determined.

Progress During Report Period

The "H" hole, located near the center of the 216-Z-9 Trench, was extended from 14 to a 17 m in depth. The drilling was terminated at this depth because an open hole could not be maintained beyond this level. The casing had "set" at the 14-m level due to a lapse in time of almost a year between driving operations at the "H" site and could not be driven beyond this depth. Determination of the presence of ^{239}Pu at the 17-m depth will be delayed until the counting system again becomes operational.

In addition, a hole was drilled to a depth of 6 m below the bottom of the 216-Z-9 Trench at the "D" site (∿2 m south of the "H" hole). Portions of this core will be submitted to Battelle-Northwest for further examination of actinide-sediment associations.

Evaluation of Effort

A major portion of the report period was concerned with program planning and outlining of drilling and sampling requirements. Improvements in drilling and sampling techniques are described under the Drilling and Sampling Technology Program (B05T5). Field and laboratory work were done on a limited scale. The drilling conducted at the 216-Z-9 site was important both in obtaining samples which may show the boundary of Pu contamination at the site and also in training a new crew who will carry out drilling operations in FY 1976.

Multiple chemical reactions occurred in the actinide trenches, making it almost impossible to predict the actinide distribution of one trench based on data from another. The variations in the chemical reactions are due to differences in the sediments underlying the trenches and in the

characteristics and volume of wastes disposed to each trench.

Future Work

During FY 1976 the vertical distributions (up to ∿30 m) of plutonium and americium beneath 216-Z-12 and 216-Z-18 will be determined in two wells. The mineralogical location of the actinides in the sediments obtained from these wells will also be identified. Wells at the 216-Z-1A(b), 216-Z-12, and 216-Z-18 sites will be prepared for pulsing.

Assessment of the reactivities, distributions, and mineralogical locations of actinides beneath approximately 20 disposal sites containing greater than 1 kg of plutonium is estimated to be completed by the end of FY 1981.

Drilling and Sampling Technology

Program Manager:	R. E. Isaacson
Section Manager:	R. E. Isaacson
Principal Investigators:	S. M. Price C. T. Webster (consultant)
Activity Code:	B05T5

Objectives and Scope

Drilling and sampling technologies are needed to obtain credible geologic and hydrologic data in support of Waste Management operations and surveillance activities for evaluating the suitability of the Hanford Reservation for other future uses. The specific objectives of the Drilling and Sampling Technology Program are to:

- Develop a method for recovering representative samples of high-level wastes from underground storage tanks.
- Develop method(s) for obtaining representative samples of uncontaminated and contaminated Hanford sediments.
- Identify and/or develop for monitoring purposes the drilling technology for gaining access to underground locations.
- Obtain and develop methods for obtaining representative rock and water samples.
- Develop identification techniques for making credible geologic correlations.

The scope of the work includes the development and testing of all drilling and sampling methods required in support of research programs.

Prior Work

The Drilling and Sampling Technology Program was formally initiated at the beginning of FY 1975. The first drilling technology examined under the program was the use of the subterrane penetrator to provide horizontal monitoring holes beneath the Hanford liquid waste storage tanks. [The subterrane is a system invented and patented by the scientists of the Los Alamos Scientific Laboratory to make vertical or horizontal holes in the ground by melting rocks and soils.] Field tests of the penetrator on the Reservation indicated that the tool is unworkable in the varigated Hanford sediments. Further experimentation with the tool was subsequently cancelled.

A conceptual design of a drilling method capable of obtaining representative samples from underground waste storage tanks was completed and submitted for consideration. The design was deemed to be workable and a concept is being implemented which is a modification of standard diamond core drilling techniques.

Progress During Report Period

Assistance with design, construction, and testing of the tank sampling method was provided throughout the report period.

Methods for drilling through contaminated sediments were also implemented and tested. The totally contained Z-9 drilling method, which uses a cathead-activated drive hammer to drive a split-tube core barrel, was strengthened and used to core ∿25 m at the 216-Z-9 facility. Plans were subsequently formulated to improve the "Z-9" method so that it can be applied to the sampling of contaminated sediments at other actinide trenches.

The need to maintain a minimal "open" hole while drilling through contaminated sediments (to prevent cross-contamination) was recognized during the initial phases of the Actinide Trench Characterization Program. A split-tube core barrel was subsequently developed which simultaneously drives the sampler and casing. With this method, the concentration of contamination at a specific depth level can be accurately determined.

Another need, that of obtaining representative mud or sludge samples, was met by development of a split-tube core barrel lined with a plastic sheath. During driving, the sheath enfolds the sample and "snaps" closed at the end of the drive to provide a leak-proof seal.

Design and development of a dual-walled core barrel furnished a totally contained method for drilling through the bottom of actinide trenches. Two 11-m cylindrical prototype samplers were constructed for testing. The samplers were designed to function in a manner analogous to that of an apple corer, forcing the sediments into the inner barrel during driving. To prevent contamination spread, the test indicated that the "dual-walled" method can be used to penetrate up to 9 m of contaminated sediments without the requirement of additional costly containment.

The modified Z-9 Containment and Sampling and Dual-Walled Core Barrel Techniques can be used in combination to provide test wells through the bottom of actinide trenches. The dual-walled core barrel provides a 9-m hole for pulsing and representative samples to the 9-m level can be obtained adjacent to the pulsing hole. Samples from deeper levels can be obtained by deepening the test well with the Z-9 rig or a conventional cable tool.

Work was conducted to determine if ^{239}Pu could be detected in water samples from a well adjacent to the 216-Z-12 Trench. Battelle-Northwest ran six samples from Well 299-W18-5 through a large-volume water sampler. All samples contained detectable amounts of Pu decreasing with pumping time. The sample with the greatest Pu concentration contained 0.0004 pCi/ml, approximately four orders of magnitude below the ERDA guide for drinking water.

The detection of trace-quantities of Pu in Well 299-W18-5 indicates a need for standardizing a large-volume water sampling technique; these procedures have been outlined.

Evaluation of Effort

Methods of sampling have been provided to meet the needs of on-going or proposed Hanford programs. The conceptual design for tank sampling formulated under this program was sucessfully tested and will be implemented to obtain actual tank samples. Techniques were demonstrated which can provide representative samples and test bore holes through the bottom of actinide trenches.

Future Work

Continued assistance will be provided with the sampling of liquid waste tanks. Plans will be outlined to establish and standardize methods for grouting and structurally improving monitoring wells. Procedures for taking a large-volume water sample to be analyzed for plutonium will be examined for standardization. Work will be continued on all drilling and sampling innovations required by on-going research programs.

HYDROLOGY

GROUNDWATER HYDROLOGY (Model Verification and Management)

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

Objective and Scope

The studies undertaken in this program define the Hanford groundwater flow system and measure the hydrological

properties that control the rate and direction of groundwater movement. This knowledge will be used to develop a computer model which realistically simulates the Hanford groundwater flow system and is capable of solving transient groundwater and contaminant flow problem. The program encompasses all activities required to assure comprehensive management of the groundwater beneath the Hanford Reservation.

Prior Work

Extensive research has been conducted at Hanford on the behavior of radionuclide migration through the partially saturated sediments underlying waste disposal sites and in the saturated groundwater flow system. Mathematical models have been written to describe the subsurface water movement and radionuclide transport in both the saturated and unsaturated sediments. The Variable Thickness Transient (VTT) and Partially Saturated Transient (PST) flow models for the Hanford groundwater flow system were applied to a number of groundwater flow problems. Error and sensitivity analyses on the Macro-Microion Transport Model (MMT) were completed and all of these models were converted to operate on the CYBER-74 computer. Reformulation of the MMT model has removed several limitations and significantly reduced the calculation time. Extensive testing and improvements have increased the speed and accuracy of the transmissivity interactive data model.

Development of an Information Retrieval System for storing and retrieving Hanford hydrologic, geologic, and groundwater contamination data was begun. Historical groundwater data on gross beta and tritium concentrations were transferred to punch card-form for entry into the

information retrieval system. Five biweekly groundwater table measurements were statistically evaluated for each well to define and plot a temporal rate-of-change map for the water table under the Hanford Reservation.

A new transmissivity distribution map was calculated. This new distribution is now being used in model verification.

Progress During Report Period

A review of the groundwater monitoring network, including an analysis of all well structures and setting of criteria for administering the ARHCO portion of the program, have been completed.

Pumping equipment was acquired during the reporting period and the pump-testing program was initiated as planned. Eighteen wells were cleaned and upgraded prior to testing. The first test has been completed on Wells 699-15-15A and 699-15-15B. Six new wells for pump-testing have been drilled using an air rotary technique. All tests are scheduled for next fiscal year.

Gravity readings were compiled from 500 stations throughout the Hanford Reservation to an accuracy of ± 0.1 milligals. Terrain and regional (third-order polynomial) corrections were to produce a residual gravity map. This map was used to locate potential areas of buried stream channels and possible fractures in the buried bedrock surface (basalt). Examination of the regional gravity data reveals the existence of several parallel anticlinal-synclinal systems.

The gravity program was supplemented with a geophysical logging program covering over 4,000 m of well-bore. The

logging was conducted by Washington State University personnel. Field work was completed and interpretation is proceeding on schedule.

Twice during the last six months water-level measurements have been conducted by ARHCO and BNW. A map of the January 1975 water table was issued.

The hydraulic characteristics of the Hanford Reservation were studied using 30 pumping tests conducted on the unconfined aquifer underlying the Reservation. Driller logs for these wells were calibrated for transmissivity and hydraulic conductivity using the results of the pumping tests. Calibration results were applied to over 300 driller logs from wells that have not been pump-tested. The analyses produced a comprehensive areal coverage of the average transmissivity and hydraulic conductivity of the unconfined aquifer. Vertical variations in these properties were examined using driller logs, pumping tests at various penetrations, and samples. A comprehensive analysis of the data was then used to explain the depositional history of the area and the hydrologic implications of the predicted events.

Work was continued to improve all existing models and expand the data bank. Sensitivity analyses are presently being conducted by BNW. Special emphasis has been placed during the reporting period on developing a capability for determining travel times along pathlines originating at present waste sites and a means for predicting future radiocontaminant levels in the groundwater using the MMT model.

Work is proceeding normally to develop software to use in the Tektronic terminal being installed in the 202-S

Building in the 200 West Area. This terminal will be linked directly to the PDP-11 computer.

Evaluation of Effort

Models in their current state of development do serve as useful management tools, but the output must be carefully interpreted by persons knowledgeable in the hydrological application for which they are used. Until new Hanford Reservation field data on geology and hydrology are used as input to the predictive models, their usefulness will be limited. For several of the predictive models the associated numerics can be upgraded to improve efficiency and reduce numerical dispersion. Work during the reporting period has helped greatly to enhance the data base. New data will be input to the models to improve their capabilities.

Future Work

Field work planned for the future includes continuation of the water level monitoring program, expansion of pump-testing work, and continuation of the drilling and geophysical programs.

The field measurement and monitoring activities provide a continuous surveillance of the changing stresses on the Hanford groundwater system. Changes in the regional water level must be known because of its potential impact on waste management practices and plans. To furnish this information, potential head measurements must be conducted on a regularly scheduled basis.

To determine the hydrologic character of the underlying aquifers, pumping tests will be conducted at three pumping sites that were drilled and developed on the Hanford

Reservation during FY 1975 and in two additional sites to be drilled during FY 1976. This information is needed to verify the predictive models and to provide the data required to control the release of radioactive contaminants into the surface environment.

The modeling effort will, in the future, primarily involve sensitivity analyses on the models and data base improvements.

Reports Issued

R. A. Deju and R. E. Gephart, *Hydrologic Management at the Hanford Nuclear Waste Facility*, ARH-SA-235, Atlantic Richfield Hanford Company, Richland, Washington, May 1975.

MOISTURE TRANSPORT STUDIES

Program Manager:	R. E. Isaacson
Section Manager:	R. E. Isaacson
Principal Investigators:	D. J. Brown L. E. Brownell - consultant
Activity Code:	B09TC

Objective and Scope

The objective is to determine the rate and direction of moisture movement in the partially saturated sediments underlying the 200 Areas as a result of precipitation and the effect this moisture movement has on radionuclides retained by these sediments from past waste disposal practices.

Prior Work

During the past three years, significant insight has been gained into the behavior of liquids in partially saturated sediments underlying the 200 Areas. This insight was obtained principally from data obtained from two instrumented lysimeters constructed about one mile south of the 200 East Area. One lysimeter is open at the bottom and the other is closed.

Three separate monitoring systems were installed in the lysimeters: 3 access tubes for determining moisture content with a neutron probe, 15 stainless steel tubes for measuring absolute pressure with a pneumatic scanner, and 16 thermocouple psychrometers with temperature sensors for determining water potential as a function of depth.

The neutron logging system is the only one that has been used routinely since the lysimeter test site was completed in January 1972.

The data obtained show a cyclic movement of moisture downward and then back to the ground surface in the partially saturated sediments. Autumn rains usually commence the later part of September to mid-October and become more intense during November. The moisture content in the ground normally reaches its maximum value by early February. The maximum moisture content in the lysimeter soils for the 1973-1974 rain year was 12.7% on January 24, 1974. The minimum moisture content occurs during the period September to October. During the hot summer months the moisture in the sediments, to a depth of about 5 m, begins moving upward and is eventually lost to the atmosphere by evaporation. In the upper 4 m of sediment the moisture content actually decreases to a value less than the average moisture content (6% by volume) temporarily creating a partially desiccated

zone. The 5 m-depth represents the approximate extent of the seasonal zone. Below this depth the moisture content remains essentially constant.

Progress During Report Period

The sediment moisture profile for the open- and closed-bottom lysimeters was remeasured twice a month during the report period. In these two lysimeters there are five specific aspects of the moisture distribution that are being followed on a month-by-month basis. They include the position of the new percolation front for the 1974-1975 water year; the dry (desiccated) zone remaining from the previous autumn at about the 3-m depth; the residual high moisture envelope at about the 6-m depth remaining from the 1973-1974 water year; the average moisture content below the seasonal zone; and the moisture content at the bottom of the lysimeters.

During the report period the new percolation front moved downward to a depth of 2 m. There has been essentially no change in the desiccated zone at the 3-m depth from the previous rain year. The residual high moisture envelope in the closed-bottom lysimeter has disappeared from the moisture profile. In the open-bottom lysimeter the position of the residual high moisture envelope has remained unchanged while the moisture content has decreased slightly. The average moisture content below the seasonal zone remains about the same for both lysimeters, 6% by volume. At the bottom of the two lysimeters the moisture content continues to decrease: 5.5% in the closed-bottom lysimeter and 5.1% in the open-bottom lysimeter.

Evaluation of Effort

The information obtained from the lysimeter test site shows consistently that precipitation entering the ground moves back out through the ground surface and not to the water table. Radionuclides associated with leaks which occur above and do not wet the desiccated zone will be restricted from downward movement by this zone.

Future Work

The two lysimeters will continue to be monitored for moisture movement and additional lysimeters will be constructed in the 200 East and 200 West Areas near waste management facilities.

An algorithm for solving the newly developed equation of two-phase flow will be developed for predicting moisture movement in partially saturated sediments.

Reports Issued

1. A. E. Reisenauer, *Calculation of Soil Hydraulic Conductivity from Soil-Water Retention Relationships*, BNWL-1710, Battelle Pacific Northwest Laboratories (1973).

2. R. E. Isaacson, L. E. Brownell, R. W. Nelson, and E. L. Roetman, *Soil-Moisture Transport in Arid-Site Vadose Zones*, ARH-SA-169, Atlantic Richfield Hanford Company, January 1974.

3. R. E. Isaacson, L. E. Brownell, and J. C. Hanson, *Soil-Moisture Transport in Arid-Site Vadose Zones*, ARH-2983, Atlantic Richfield Hanford Company, October 1974.

TANK FARM GEOLOGY

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

Objective and Scope

The objectives of this program are to document (using cross-sections and maps) the structure and stratigraphy of the partially saturated sediments underlying the 200 Areas Tank Farms and to determine radionuclides in the ground around tanks suspected of leaking. This will involve identifying significant hydrological properties of each stratigraphic unit and assisting in locating the sources of radionuclides leaked to the ground.

Prior Work

The Hanford Well Library, located in Igloos T-105 and T-106 behind the 200 West Area, was completed. All sediment samples from drilling on the Hanford Reservation were cataloged and stored at this location.

Procedures for analyzing sediment samples were revised to include sieving of samples into nine particle-size fractions and analysis for calcium carbonate. These data are input into the ROCKSAN computer program, which categorizes the sediments into 19 classes. Samples of the same class are then composited for hydraulic and bulk chemical analyses. All samples are visually examined and the data are presented on geologic cross-sections and maps.

Gamma log analyses requested by ERDA of wells around Tank 241-T-106 were completed. The three-month study

showed a decrease in gamma radiation proportional to the ^{106}Ru decay curve. At the edges of the leak plume where the lowest radiation levels occurred, two notable exceptions were found in the form of a slight increase in activity which can be attributed to lateral migration of radionuclides.

Progress During Report Period

To date 7839 sediment samples have been analyzed and categorized by the ROCKSAN computer program. Splits have been taken from each sample for use in hydraulic and bulk rock analyses. These samples are from all tank farms with the exception of 241-AY and 241-AX, where no monitoring wells have been drilled. Several samples have also been analyzed from deeper water wells around the tank farms.

Evaluation of Effort

The 241-T and 241-TX geologic maps show the sieve and calcium carbonate analysis routine and ROCKSAN computer program to be a workable scheme. The distribution of radionuclides within the 241-T Tank Farm correlated well with the geologic maps prepared. This provides a second independent method for determining radionuclide movement in soil as a result of a tank leak.

Future Work

Analyses of samples from the 200 West Area Tank Farms will be completed within the next report period. Current hydrologic procedures and mineralogic analysis methods will be evaluated and the 241-T-106 leak study wells completed.

Two classes of samples will be analyzed by BNW and Oregon State University for wetting and imbibing conditions

utilizing water and two synthetic wastes. The evaluation of these data will aid in determining the course of further hydraulic analyses.

The same samples will be used to develop procedures for determining the variance in mineralogic composition of Hanford sediments. This will aid in defining the future course of mineral waste reaction studies.

TRANSPORT

AIRBORNE RELEASES

Program Manager:	R. E. Isaacson
Section Manager:	R. E. Isaacson
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Activity Code:	B09TB

Objective and Scope

The objective of this project is to aid in evaluating the potential inhalation hazards arising from operating the reprocessing facilities and maintenance of the radioactive waste storate sites. Airborne releases due to changes in both physical conditions and aerodynamic processes which affect the behavior of radioactive particles in the environment are studied. Experiments are performed to measure the quantities and characteristics of airborne radioactive particles released. The influences of fire, wind, and

mechanical surface stresses on resuspension, transport, and trajectories of radioactive particles are also to be investigated.

Prior Work

Experiments conducted in the laboratory on materials from the BC Crib and U Pond areas were made to determine the effect of fire on the dispersal of ^{137}Cs from these areas. Release of ^{137}Cs at 200°, 400°, and 600° C was measured and found to be low.

Resuspension of surficially contaminated particles was measured as a function of wind speed and direction with an array of sampling towers and specially designed high-volume particle-sizing samplers located at different heights in the BC Crib and U Pond areas. Airborne ^{137}Cs concentrations downwind from U Pond were consistently higher than at the BC site by a factor of about five.

A computer program for calculating atmospheric transport and downwind dispersion from a single source was developed and tested. The results indicated the unreliability of using measurements for predicting transport beyond a few kilometers of a single point.

A computer program to reduce "motion pictures" of atmospheric transport over the Hanford Reservation was developed to the point of demonstration.

State-of-the-art diffusion models were used to predict downwind airborne concentrations of the BC Crib to a distance of 50 km. Annual exposures of individuals at various distances from the source were calculated. Model and computation improvements were made to permit greater versatility in handling contamination patterns as multiple sources.

The significance of dust devils and anomalous winds in resuspending contaminated soil was studied. An extensive literature review was completed and many observations on local dust devil frequency were compiled.

Progress During Report Period

Arrays of air samplers were used in resuspension experiments at U Pond, Z-19 Ditch, S-16 Disposal Area, and BC Crib Area. Air was sampled as a function of wind speed increment, wind direction, and sampling height. Results indicated that resuspension of ^{137}Cs, ^{241}Am, and ^{239}Pu occurred in all areas. The resuspension data indicate that while plutonium levels in the 200 and BC Crib Areas exceed fallout levels, the airborne Pu is a factor of 1000 below MPC_{air} (6×10^{-13} $\mu Ci/m^2$ for Pu).

Improvements were made to the computer program which calculates the airborne concentrations of material resuspended from the B-C Crib area. These have increased computational accuracy and reduced computer costs. The improved accuracy has allowed detailed verification of an analytical model which predicts the resuspended air concentrations upwind of the peak of the surface contamination pattern. The lower computer costs will make it possible to calculate resuspended air concentrations over extended meteorological conditions which will allow verification of theoretical predictions with actual measurements.

After much effort was made to edit the historical data recorded by the meteorological telemetry system over the past few years, it was concluded that this old telemetry system is not a good data source for producing an atmospheric transport climatology. In conjunction with ERDA-RL,

criteria for a new telemetry system were developed and a base station with one remote station was ordered.

Work was begun on a model to produce a representation of the boundary layer flow over the existing terrain.

A more realistic simulation of the diurnal behavior of the mixing layer was developed and incorporated into the transport and diffusion model. An acoustic sounding device was obtained, installed, and tested to provide observational information on the behavior of the mixing layer.

Further capability was developed in the area of computer display of data and model output, specifically for contours of terrain height, time series of exposure plots, and views of plume behavior in complex flow.

Progress continued on the study of convective vortices (dust devils). Estimates were made of dose to individuals from convection vortices moving across the BC Crib area.

Evaluation of Effort

The resuspension data indicate plutonium sources are significant and the study is on schedule. Excessive delays in obtaining the ultra-low-level analyses for ^{239}Pu and ^{241}Am could affect future progress.

The surface flux model has successfully produced computational methods for estimating downwind concentrations of radionuclides contaminating the BC Crib area. All computations require knowledge of resuspension rates and deposition velocities; hence it is vital that these quantities be known for various wind and soil conditions.

Continued development of computer graphics subprograms is providing more effective utilization of data and model results. Some of these programs have proved useful in other areas of ARHCO work.

Future Work

Field resuspension studies will be increased to include TX Tank Farm, B Pond, and Gable Mountain Swamp. Sampling arrays will be expanded to include background stations which are sufficiently upwind of test sites (see Public Protection Assurance - Waste Characterization). Multiple sampling locations will determine how the resuspension plume develops from uncontaminated areas through the contaminated areas. Emphasis will be placed on developing analytical techniques of sufficient sensitivity to detect the trace Pu and levels present. Air samples will be combined to increase sensitivity and decrease analytical costs.

The model will be applied to other contaminated areas to establish the realistic guidelines on degree and extent of surface contamination which hypothetically would result in airborne contamination exceeding permissible limits. Climatogically averaged resuspension will be calculated to take into account the possible long-term exposure and source-depletion effects and comparisons will be made between calculated and observed values.

Further work on the overall flow model will provide increasingly better representation of the transporting winds over the Hanford Reservation. Work should progress in the direction of providing a realistic three-dimensional representation of the atmospheric transport and diffusion of possible contaminants from the Hanford Reservation.

INSTRUMENTATION AND CHARACTERIZATION

PUBLIC PROTECTION ASSURANCE

Environmental Instrumentation

Program Manager:	R. E. Isaacson
Section Manager:	R. E. Isaacson
Principal Investigators:	L. E. Bruns K. T. Key G. E. Martin P. J. Wiater
Activity Code:	B1430

Objective and Scope

The objective of these studies is to develop field instrumentation and auxiliary equipment that will accurately and rapidly characterize the amount, type, and distribution of radionuclides on soil surfaces, in ponds and ditches, and subsurface. Wherever possible radionuclides will be assayed *in situ*; sensitivity is desired in the range of pCi/g of soil or water. Auxiliary equipment to be developed includes void volume detectors, airborne particle size detectors, and subsurface metal detectors.

Prior Work

Dev-Van-I was received early in calendar year 1975 and a work order was issued for equipment installation. Further calibrations and results were obtained with the argon dosimeter, neutron counter, and alpha-water meter. All Dev-Van-II equipment was received, but one germanium drifted lithium [Ge(Li)] detector had to be returned to the factory for redrifting.

Determination of plutonium by the metal foil

neutron response technique was initiated. The magnetometer was further tested for subsurface metal detection. An automated instrumentation system for isolating resuspension parameters was installed in the field. An alpha monitor for screening resuspension filters was tested. New concepts investigated included a ^{90}Sr beta assayer, picocurie-level plutonium assayers, directional neutron counters, and specially shielded TLD chips for environmental dose determinations.

Progress During Report Period

The experimental ^{90}Sr field assay instrument was calibrated in the laboratory for efficiency and attenuation in soils. Strontium-90 at the nanocurie-level was successfully measured in the presence of ^{137}Cs using absorber plates; with the correct absorber plates, ruthenium, cesium, cerium, and silver can also be beta-assayed. Laboratory results indicate a 10-fold improvement over any commercial unit. Some adaptions were made to improve the instrument's efficiency and durability.

Two types of environmental plutonium monitors were studied. One was a unit devised by National Nuclear Corporation (NNC) which uses a proprietary sensing element; the other unit, devised by Battelle-Northwest, uses a thin-window intrinsic germanium diode as the detector element. For both units the objective is to detect plutonium levels below 10 nCi Pu/g of soil. Testing of the NNC unit and initial assembly of the BNW unit should begin in FY 1976.

The newly developed water system for measuring Pu and Am values has been tried on a waste tank (105-B), in Pu storage areas, and on 234-5 Building hoods with good results. Copper, zinc, and magnesium foils were used to

determine plutonium in fabrication hoods and in a 209-liter drum. The test in the 105-B Waste Tank detected a 55-gal maximum Pu content of 7 nCi Pu/g of waste in a high gamma field.

Procedures for use of the mR/yr sensitive argon dosimeter have been written and a unit was turned over to Quality Assurance for routine surveillance.

A paper evaluation of the Holosonics subsurface void volume transfer impedance detector was made.

Dev-Van-I, the environmental *in situ* surface assayer, and Dev-Van-II, the subsurface *in situ* assayer system, have been delayed due to priority problems with vehicles. All major equipment pieces are onsite. The Ge(Li) collimated gamma surface assayer is calibrated and ready for field tests. The two Dev-Van-II detectors for the core assayer were onsite but one has been sent back for redrifting. The collimator for the core assayer is being fabricated. The Ge(Li) in-well sondes are onsite, have been checked, and meet specifications.

Temporary in-well Ge(Li) and gamma directional systems were used to assist tank farm management. These units can ascertain ruthenium and cesium presence to the μCi/liter of soil level and tell from what direction the activity is coming; hence they can help pinpoint a leak.

Specially shielded TLD's were devised and set up in three locations in the U Pond area: in the water, on the shore, and inland. Duplicate units were placed in four vertical offshore positions and three vertical positions onshore and inland. Both CaF_2 and LiF chips were used.

New concepts studies during the report period included a ^{252}Cf *in situ*, in-well monitor and a 10 μCi Pu/g

drum counter. The ^{252}Cf unit can be used as a sensitive moisture monitor, a Pu and Am monitor, and for monitoring soil composition. Both the metal foil neutron response and the ^{252}Cf techniques were considered for the drum counter.

Evaluation of Effort

The ^{90}Sr beta assayer will be somewhat bulky in the field but with the proper mounting should be a valuable asset to inexpensive environmental beta characterizations. Though feasible in the laboratory, field verification will come from testing in areas such as the BC Crib. Tests thus far show that the assayer is about 10 times more sensitive than other assayers developed to date.

An important step to effective yet inexpensive environmental monitoring is the *in situ* in-the-field measurement of plutonium and americium. The NNC and BNW surface monitors have potential. The units operate by differentiating between the various X-rays given off by Pu and Am. Sensitivity to nCi/g-levels is the ultimate goal.

In-well detection is based on the foil or water technique for sensitivity (nCi/g) while the directional unit should be good in the μCi/g range. Proper selection of metal foils should allow both Pu and Am measurements; also different foils allow distinction between Pu metal, oxide, and fluoride. The problem with metal foils is the detection time (24 hours in location) and the time and cost of measuring the activation products and equating these to the concentrations of Pu and Am. It is still faster and less expensive, however, than wet chemical analysis.

The directional unit gives a real time value but sensitivity is less than desired. Two directional units are being considered--the one tested during the last report

period (an invention of ARHCO research personnel) which measures thermal neutrons. It was successfully used in the Z-9 Trench and in a Z-1-A Well. A fast fission unit from NNC will be ready for testing during the next six months.

The gamma *in situ* assayers can be used for surface and subsurface measurement of ^{106}Ru, ^{152}Eu, ^{137}Cs, ^{241}Am, and other gamma-emitters. They have already been helpful in determining the size of a subsurface salt leak and pinpointing the location of the leak. The helicopter gamma system and various portable gamma instruments have given a good overall estimation of surface contamination. The eventual completion of Dev-Van-I and Dev-Van-II should give a powerful method of completely assaying various waste entities (*e.g.*, ditches, swamps, ponds, cribs, trenches, burial grounds, and waste tank environs).

Initial study of the ^{252}Cf in-well system indicates that this could be another strong subsurface exploration system both for radionuclides and nonradionuclides. Again such a system could replace much expensive laboratory analysis and yet furnish more complete statistical coverage of a given waste entity.

A paper study of the 10 nCi Pu/g drum monitor indicates that it is feasible. Los Alamos Scientific Laboratory has developed a gamma system for drums with a density of about 0.5, which should be considered in the final design.

Future Work

In late May or early June 1976 National Nuclear Corporation will test a directional neutron counter at the Hanford site with a goal of obtaining the approximate Pu concentration in the soil with depth and with direction at

the 1 μCi Pu/g of soil level.

Dev-Van-I will be tested with a temporary setup in the vans until the equipment and auxiliaries can be installed. The in-well Ge(Li) and NaI(Tl) systems (Dev-Van-II) will be used on a temporary basis until a van can be procured and the equipment installed.

The *in situ* ^{90}Sr-beta assayer will be adapted for field use in the coming months. Two programs involving NNC equipment will be carried out--one with their fast-fission directional neutron counter and the other with the Pu surface assayer. Further metal foil experiments are planned. The first special TLD's will be removed, measured, and the results evaluated. Further work is planned on evaluating the feasibility of the ^{252}Cf in-well system and the 10 nCi Pu/g drum monitor.

Waste Characterization

Section Manager:	R. E. Isaacson
Project Manager:	R. E. Isaacson
Principal Investigators:	L. E. Bruns K. T. Key P. J. Wiater
Activity Code:	B1432

Objective and Scope

This program will identify the locations, amounts, physical and chemical properties, and ecological reactions of radioisotopes in the environment of the 200 Areas Plateau; and the means by which radionuclides from Hanford operations can enter man's environment, and their potential hazards.

Prior Work

Characterization studies were done to demonstrate (1) effective representative sampling methods, (2) reliable environmental analyses, (3) use of environmental assay instruments, (4) important physical and chemical properties, (5) resuspension parameters, (6) local wind field patterns, and (7) overall wind field movements. Studies were made on U Pond, B-C Crib, S-16, S-17, S-19, Z-19, and Gable Mountain Pond areas.

Progress During Report Period

Representative sampling and analysis studies continued, with effort directed to S-16 Swamp, S-17 Covered Pond, S-19 Swamp, U Pond, Z-19 Pond, Gable Mountain Pond, and BC Crib. Characterization of the S-16 area showed that certain areas were in the 5 nCi Pu/g of soil range. At S-17, about 1 m of soil was first removed and a 30-cm core taken; results from the analyses of these cores will become a basis for plant uptake studies. Sample analysis of S-19 will become a basis of study for removal of the radionuclides from the soil, slurry, and water. Results for U Pond and Gable Mountain Pond were mainly from the LFE Laboratory and were part of the cooperative DBER-BNW-ARHCO programs.

Characterization studies conducted by BNW involving the $^{90}Sr/^{137}Cs$ ratio in the B-C Crib, ^{137}Cs heating release experiments, BC Crib cleanup wind tunnel experiments, and the U Pond shore are essentially completed. These studies will provide a basis for resuspension (and eventually for decommissioning) studies (see section on Airborne Release).

Resuspension research studies were conducted at the U Pond towers and a tower set up downwind of a bend in

the Z-19 ditch (Plutonium Finishing Plants to U Pond Ditch). Initial results on ^{241}Am, ^{7}Be, ^{54}Mn, ^{95}Zr, ^{95}Nb, ^{103}Ru, and Cs were obtained (see section on Airborne Release for field application studies).

A fallout study was initiated to establish a better baseline for resuspension studies. Fallout averages throughout the world and during different seasons were compared. Fallout stations west of the 200 Areas process plants are being planned.

Atmospheric models which incorporate local topographic influences are being developed to provide a more representative transporting wind for the diffusion model and to help select the most effective placement of stations for a new telemetry system. Better modeling of the behavior of the mixing layer in the atmosphere was accomplished and the phenomenon was measured with an acoustic sounding device.

Significant improvement was achieved in the model for resuspension and downwind transport of radioactive particles from the B-C Control Zone. The model accounts for release, downwind diffusion, and deposition from an extended source of surficial contamination levels.

Evaluation of Effort

Continued experimentation with different types of equipment has revealed two effective sampling techniques: a 30-cm core is taken using either a 10-cm diameter acrylic tube or stainless steel rings (rings are 2.5- and 5.0-cm deep and 20 cm in diameter). One-centimeter surface samples can be taken with the 2.5-cm ring. Live vegetation and detritus are simply pulled or hand-picked and put into separate containers before surface or in-depth soil samples are taken.

Sample preparation has been continually optimized and various equipment pieces have been purchased for grinding, drying, and mixing of soil samples before analysis. Environmental analyses are validated by other laboratories; although some problems have been experienced with various variations in ^{90}Sr analyses between laboratories, difficulty in obtaining ^{137}Cs and ^{129}I analyses, and the cost of environmental analyses, generally this procedure has proved highly satisfactory.

Use of the environmental *in situ* instruments will greatly alleviate the cost problem and allow better statistical coverage of a given area.

To date only token investigations have been made of physical and chemical properties and environs reactions. Various leaching studies of U Pond sediments have been done and the particulate sizes of radionuclides in the Pond and on the shore have been studied. Property changes with temperature and particles sizes have been investigated assuming range fire conditions using BC Crib area samples.

Future Work

Work in the next six months will include: development of representative sampling techniques, patterning of airborne releases throughout the 200 Areas Plateau by the high-volume mobile air sampler, establishment of resuspension factors, and a paper study on fallout in the Hanford area. Work will begin on a control test of interlaboratory verification of environmental analyses. The various areas which have been X-Y-Z-characterized to date, mostly by sampling and laboratory analyses, will be used as a basis to formulate transport factors.

ANALYTICAL METHODS DEVELOPMENT

ACTINIDE ANALYTICAL PROGRAM

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

Objectives and Scope

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

Previous Work

In the last report period the technical feasibility of using dihexyl-N,N-diethylcarbamylmethylene phosphonate (DHDECMP) and Aliquat-336® (General Mills) to separate actinides from synthetic waste was established.

Progress During Report Period

DHDECMP was evaluated further as an extractant for actinides from Hanford wastes. Because of the low distribution coefficient of DHDECMP for Am, it was necessary to use the reagent in its undiluted- (100%) form in order to obtain quantitative extractions. The unknown impurities present in DHDECMP presented serious stripping problems for analytical applications where 100% solutions had to be used. In addition to this, solids formed in the organic (DHDECMP) phase when contacted with aqueous waste solutions in the presence of reducing agents. It was concluded that this

reagent would be inadequate for a routine analytical procedure.

Another method for separating the actinides from waste using the quaternary amine Aliquat-336 and theonyltrifluoracetone (TTA) has been evaluated and tested on actual terminal liquor and salt cake matrixes. Plutonium-236 and ^{243}Am have been used to determine chemical yields in the procedure. Plutonium recoveries were greater than 80% and Am recoveries were about 50% for most of the samples analyzed. Methods for neutralizing the samples and stripping the actinides from Aliquat-336 with Na_2CO_3 were developed. The distribution paths of Pu, Np, Am, and U through the procedure were determined.

A shorter method for analyzing plutonium using only TTA as an extractant was also tested. Although the method had very low ^{236}Pu tracer recoveries, corrected results were usually comparable with the Aliquat-336 method. The method probably will not be utilized because of the extreme difficulties encountered in making accurate acid adjustments due to the presence of large amounts of hydrolyzable cations and anions of weak acids.

Evaluation of Effort

The Aliquat-336 separation method is the only analytical procedure presently capable of giving reliable plutonium results for high-caustic, high-salt waste. It is expected that future work will also find the method capable of giving accurate results for ^{241}Am and ^{237}Np.

Because of the long half-lives and high toxicity of the actinides, their analyses are very important in characterizing and managing Hanford waste. Therefore in order to

ensure highly reliable actinide analyses, further work is merited to develop and evaluate actinide analytical methods.

Future Work

Evaluation of the Aliquat-336 method will be completed and improved Am methods will be tested. Methods for determining thorium and uranium in the waste will be evaluated. Efforts will be made to shorten the method and obtain better alpha energy analyses by developing electrode-position mounting techniques. The best methods will be evaluated on different waste forms such as sludge, filtered solids, and decontaminated salt cake solutions resulting from radionuclide removal experiments.

RADIATION ANALYSIS SYSTEMS DEVELOPMENT

Project Manager:	H. Babad
Section Manager:	H. Babad
Principal Investigators:	W. H. Zimmer V. W. Hall
Activity Code:	B06T8

Objective and Scope

This program has as its objectives the design of data acquisition and reduction systems and their application to all phases of nondestructive measurement instrumentation involving radiation emissions.

Prior Work

Two Tracor Northern, Inc., NS-660 data reduction systems oriented to gamma energy analysis and one NS-880 oriented to X-ray range energy analysis were placed in service. These three systems with their associated custom programs comprise the Gamma, X-Ray Data Reduction (GXDR) System. This system provides all of ARHCO with automatic data reduction of radiation spectral data received on magnetic tape cassettes. Eight satellite analyzer-cassette recorders are currently producing data for GXDR system action and more systems are soon to be added.

Nine semiconductor detector-acquisition systems were developed and used for energy analysis of radioactive and nonradioactive constituents in bulk environmental samples, low-level crib and tank soil cores, low-level air filter samples, evaporator products and effluent samples, high-level waste management research and development support, passive fissile analysis, and others.

Progress During Report Period

Two well sonde detectors were designed, acquired, and tested for in-well gamma energy analysis. One has high sensitivity for annular analysis and the other has low sensitivity for collimated, directional analysis.

The two low-energy, coaxial Ge(Li) detectors, "P" and "Q", have been assembled in their collimator ready for field analysis of well core segments.

The high-activity range Duode (D) detector was replaced by a newly developed, collimated, Compton suppression system (E) which improved the peak-to-Compton ratio for ^{137}Cs from 100:1 to 650:1.

The "D" detector, operated as a single (non-Duode), was combined with "P" and "Q" to form a second three-detector array for low-activity gamma acquisition.

An X-ray fluoresence, silicon drifted lithium [Si(Li)] detector system with automatic changer and transmission target, X-ray excitation source has been placed in service.

An intrinsic germanium detector sensitive to X-ray range emissions has been placed in service for passive plutonium isotopic analysis. It will also be used for rare earth-to-transuranic K-X-ray fluorescence when the sample changer/isotopic excitation chamber is received.

The "Z" environmental level gamma acquisition system has been in the development stage throughout the report period. It was initially delayed by slow deliveries and is now experiencing component failures.

Development continued on the GXDR programs. The following capabilities are now in service (these are in addition to those developed in the last report period).

- Activate analysis data reduction.
- Second order interference correction.
- Los Alamos Scientific Laboratory version of passive plutonium isotopic analysis.
- New calibration table editor routine.
- Alternate method of sorption correction with parameter inputs compatible with activation analysis.
- An aborting check of the third of three calibration tables for gamma reduction.

Evaluation of Effort

Seven new detector systems were brought on-line--three of which are intended for mobile applications. Unfortunately six detectors involving four systems failed during the report period. Only two of the affected system (three detectors) have been repaired and returned to service. After prolonged testing one additional detector was rejected as incapable of meeting purchase specifications. The contract was reassigned.

The GXDR systems operated more efficiently and accurately than previously, due to continuing improvement in the custom programs and a minimum of down-time. The system is providing gamma data reduction on a current one-day shift basis for round-the-clock acquisition of data on all laboratory detector systems.

Future Work

Four update phases of the GXDR system are scheduled. Phase 1, establishment of communication between the four 4096 channel pulse height analyzers in the ND 50/50 and the Nova 1220 in one NS 660, is scheduled for completion. Phase 2, establishment of communication between the same Nova 1220, the CAMAC Nova 830, and the CAMAC terminal at the 234-5 Building via a laser link, is scheduled for October 1975. Phase 3 is installation of a disc with 32 K words (total) of memory and MCA linkage between the GXDR Nova 1200's by December 1975. Phase 4 is the acquisition of a second NS-880 and linkage with the GXDR system Nova computers via CAMAC.

During the next report period the following semiconductor acquisition systems will be placed in service:

- A mid-activity range detector system.
- The R_3 low-activity, three-detector system will be returned to service after repair.
- An all-new, low-activity three-detector system, S_3.
- A well Ge(Li) detector system for low-activity, sum peak analysis.
- An environmental level gamma system.
- The intrinsic germanium detector, G_x, for high Z,K-X-ray fluorescence analysis.

Reports Issued

W. H. Zimmer, *Gamma, X-Ray Data Reduction System, Volume I - Gamma*, ARH-ST-114-1, Atlantic Richfield Hanford Company, December 1974.

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