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NUCLEAR WASTE MANAGEMENT
AND TRANSPORTATION
QUARTERLY PROGRESS REPORT
OCTOBER THROUGH DECEMBER 1974

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

FEBRUARY 1975

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OCTOBER THROUGH DECEMBER 1974

Compiled by

G. J. Dau
Nuclear Waste Technology Department

February 1975

On January 19, 1975, research and development programs of the U.S. Atomic Energy Commission (AEC) became part of the newly formed Energy Research and Development Administration (ERDA). In this report, since it refers to work done in 1974, most references are to AEC programs.

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RICHLAND, WASHINGTON 99352

FOREWORD

This document is one of a series of technical progress reports designed to report on all Division of Waste Management and Transportation (DWMT) funded programs at Battelle, Pacific Northwest Laboratories, with the exception of the Commercial Radioactive Waste Fixation Program. These reports are issued on a quarterly basis.

The prior report in this series is Nuclear Waste Management and Transportation Quarterly Progress Report July through September 1974, BNWL-1876.

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1.0 ADVANCED METHODS FOR MANAGEMENT AND DISPOSAL OF RADIOACTIVE WASTES

J. W. Bartlett* - Project Manager

The objectives of this program are to 1) develop comprehensive plans for a program to select and implement methods for disposal of radioactive wastes and 2) extend the base for the plans and program via selected topical studies.

Work in the Advanced Waste Management program is concerned with extending initial assessments of waste management alternatives⁽¹⁾ to programs and evaluations designed to select preferred disposal methods. As a result of progress in defining disposal options and data needs, studies previously allocated to six tasks were consolidated into three tasks during the quarter. The new tasks are:

TASK 1. Development of Advanced Waste Management Plans

Objective: Prepare comprehensive plans for a program to select and implement preferred disposal methods.

TASK 2. Development of Evaluation Methodology

Objective: Develop and apply methods to characterize and compare alternative waste management systems.

TASK 3. Special Evaluations

Objective: Identify and perform special-topic studies in support of plans development (Task 1) and ongoing programs.

Criteria for this work and its relationship to other waste management programs remain as stated in the quarterly report for July through September of 1974.

SUMMARY

An initial workshop on development of R&D plans for disposal of commercial radioactive waste was held in December. An outline of a plans document was developed to serve as the basis for future workshops and planning activities.

A model for migration of actinide-chain isotopes through a soil column was developed. The model is being used to evaluate consequences of escape of waste materials from a generic underground geologic disposal site.

To provide data for the nuclide migration evaluations, sorption of americium on arid northwestern and humid southeastern U.S. soils was measured as a function of sodium and calcium concentrations.

*J. W. Bartlett has replaced K. J. Schneider as Project Manager.

Methods to measure public attitudes toward nuclear waste management were developed and subjected to comprehensive peer review.

Four special studies to support development of R&D plans for disposal and other ongoing waste management programs were identified: waste management for long-lived fission products, screening comparisons of waste management options, irretrievable loss of waste to the sea, and assessment of pilot plant testing.

Task 1 - Development of Advanced Waste Management Plans -

J. W. Bartlett

The output from this task is to be a comprehensive plan for R&D to investigate and evaluate alternatives for disposal of commercial radioactive wastes. Spent fuel from reactors is the starting point for defining alternatives for waste forms and their disposal. Waste forms and alternative management schemes are summarized in Figure 1. The plans will consider all waste forms and options.

The basic procedure for developing the plans will be to convene a series of workshops composed of persons who collectively possess the spectrum of expertise needed to formulate the plans. Need for a broad range of expertise in the physical and social sciences is anticipated.

Most of this quarter was used to prepare background information for the workshops and to begin planning workshop activities. Information from previous studies (e.g., as reported in BNWL-1900⁽¹⁾) was reviewed and summarized as the basis for the workshops.

An initial, pilot workshop was held in Salt Lake City on December 11, 1974. Participants were:

Battelle-Northwest: A. M. Platt,
J. W. Bartlett, M. R. Kreiter
Sandia Laboratories: W. P. Bishop,
D. R. Anderson
Oak Ridge National Laboratory:
W. C. McClain
Woods Hole Oceanographic Institute:
Charles D. Hollister

Excellent cooperation and intensive effort by the participants produced a proposed outline of the R&D plans (Table 1) and a plan for future action to further develop the plans. Participants in this workshop were assigned responsibilities for preparation of portions of the first draft of the R&D plans.

Development of future plans will involve more workshops with wider circles of participants and interests represented. At present, the following activities and outputs are anticipated:

January 16-17, 1975 - Second plans workshop; Denver, Colorado
February 1 - Second draft of proposed plans
Late March - Waste management symposium; Tucson, Arizona (not a formal planning effort)
Early April - Third plans workshop; Airlie House, Virginia
June - Third version of first-draft plans
September - International symposium for presentation and critique of first-snapshot plans.

Participants in the January and April workshops will be more numerous than those for the Salt Lake City

FIGURE 1. Radioactive Wastes and the Basic Alternatives for Their Management

TABLE 1. Basic Outline of Proposed Disposal R&D Plans Document

1. INTRODUCTION
2. SCOPE
3. PROGRAM ALTERNATIVES
 - Reference Plan
 - Crash Plan
 - Maximum Alternatives Plan
4. R&D PROGRAM DESCRIPTION
 - A. Isolation Methods
 - Land
 - Seabed
 - Ice
 - B. Elimination Methods
 - Partitioning
 - Extraterrestrial
 - Transmutation
 - C. Waste Forms
 - HLW
 - α Waste
 - Hulls
 - β - γ Waste
 - Decommissioned Equipment
 - Spent Fuel
 - D. Evaluations
 - Disposal Method
 - Characterization
 - Disposal Method Comparisons and Selections
 - Private Sector Contributions
5. PROGRAM MANAGEMENT
6. PROGRAM EVOLUTION
 - Each discussion on disposal methods (sections 4A,B) will include the following topics:
 - Historical Resume
 - Status of Concepts
 - Future Work
 - Site Characterization
 - Emplacement
 - Transport
 - Waste Forms
 - Summary of Major Problem Areas

workshop. They will also represent a broader scope of backgrounds and interests. They will be expected to contribute their expertise to formulation of the plans.

The initial draft, which will serve as input to the January workshop, will have three basic purposes:

1. To report current thinking on the R&D content (e.g., extension of physical data) of the program on radioactive waste disposal;
2. To indicate the scope of activities, problems, and concerns thus far recognized to be significant to development of radioactive waste disposal methods; and
3. To provide triggers for thought and contributions from the future participants in development of the plans.

This draft will therefore serve only as a baseline. It will suggest programs, raise questions, and discuss problems. Preparation of an appropriate first-snapshot plan will require the multidisciplinary, multi-interest efforts of the forthcoming workshops.

Task 2 - Development of Evaluation Methodology

Actinide Chain Migration

Calculations - H. C. Burkholder and G. Jansen

The theory for the transport of a dissolved species through a column of adsorbing medium has been established by many previous workers (Wicke 1939, Lapidus and Amundson 1952, Vermeulen 1958). From this theory, which includes the effect of axial dispersion, breakthrough curves are calculated which describe the concentration of the dissolved species and its lag behind the flow of the liquid in which it is dissolved. This theory forms the basis for analyzing chromatographic and ion-exchange separation

processes and has been applied for many years to describe the retardation of single nuclides by soil (Ionue and Kaufman 1962 and Routson 1973). Radionuclides in the actinide portion of the nuclide spectrum decay by first-order chains and their migration through the soil has not been modeled either analytically or numerically. The solution to this problem has become important in evaluating the risk associated with the underground storage and disposal of waste materials from the U.S. nuclear power economy.

The model to be analyzed is an underground nuclear waste disposal site and a one dimensional soil column that connects the site with a surface water body. At some arbitrary time after the waste is deposited, the contents of this site are released at a specified rate to an underground water stream which flows at a constant velocity directly through the soil column and discharges into the surface water body. The soil is uniform and has ion exchange properties independent of radionuclide concentration. The radionuclides which are dissolved in the water have a constant dispersion coefficient and are in adsorption equilibrium with the soil at all points. The nuclides decay both while adsorbed on the soil and while in solution.

In terms of a rectangular coordinate system stationary at the soil column inlet, the migration of the "i"th member of a radionuclide chain is described by the set of linear partial differential equations which describe the material balances of the

"i"th chain member and all preceding chain members over a differential volume of the soil column. These are:

$$D \frac{\partial^2 N_1}{\partial Z^2} - V \frac{\partial N_1}{\partial Z} - K_1 \frac{\partial N_1}{\partial t} - K_1 \lambda_1 N_1 = 0$$

$$D \frac{\partial^2 N_2}{\partial Z^2} - V \frac{\partial N_2}{\partial Z} - K_2 \frac{\partial N_2}{\partial t} - K_2 \lambda_2 N_2 + K_1 \lambda_1 N_1 = 0$$

⋮

$$D \frac{\partial^2 N_i}{\partial Z^2} - V \frac{\partial N_i}{\partial Z} - K_i \frac{\partial N_i}{\partial t} - K_i \lambda_i N_i + K_{i-1} \lambda_{i-1} N_{i-1} = 0$$

where N is the nuclide inventory, Z is distance, t is time, D is the dispersion coefficient, V is the water velocity and λ is the decay constant. K is the equilibrium constant and is defined by $K = 1 + K_d \rho / \epsilon$ where K_d is the distribution coefficient, ρ is the soil density, and ϵ is the soil porosity. Focusing on the equation for the "i"th member, the first and second terms represent the net change in the "i" inventory from dispersion and convection. The third term represents accumulation, and the fourth and fifth terms represent disappearance of "i" by decay and appearance of "i" from decay of the preceding chain member, respectively. We note that the K 's are present in the fourth and fifth terms because decay occurs in both the adsorbed and dissolved states.

The equations are solved (by the Laplace transform technique) subject to the boundary conditions which describe the pre-release state of the system, the mode of release from the repository at the soil column inlet, and the state of the system at the soil column outlet. Two release modes are of practical interest: impulse release and decaying band release. Impulse release assumes that the entire contents of the site are dissolved in the underground water stream at the time of release, while band release assumes that the contents are leached from the site at a

constant rate from the time of initial release until the source is depleted. For illustration, we consider the impulse release described by the boundary conditions in Table 2.

TABLE 2. Boundary Conditions for Impulse Release

Initial $t = 0, \text{ all } Z$	Inlet $t > 0, Z = 0$	Outlet $t > 0, Z = L$
$N_1 = 0$	$N_1 = N_1^0 \delta(t)$	$N_1 = \text{finite}$
$N_2 = 0$	$N_2 = N_2^0 \delta(t)$	$N_2 = \text{finite}$
$\vdots$	$\vdots$	$\vdots$
$N_i = 0$	$N_i = N_i^0 \delta(t)$	$N_i = \text{finite}$

$\delta(t)$ is the unit impulse function and N^0 is the inventory at the time of release

The analytical results for a two-member chain with impulse release are:

$$\begin{aligned}
 N_1 &= N_1^0 \frac{V\eta}{2L} \sqrt{\frac{K_1 P}{\pi \Theta}} \exp \left[-R_1 \Theta - \frac{P\Theta}{4K_1} \left(\frac{K_1 \eta}{\Theta} - 1 \right)^2 \right] \\
 N_2 &= N_2^0 \frac{V\eta}{2L} \sqrt{\frac{K_2 P}{\pi \Theta}} \exp \left[-R_2 \Theta - \frac{P\Theta}{4K_2} \left(\frac{K_2 \eta}{\Theta} - 1 \right)^2 \right] \\
 &+ N_1^0 \frac{V}{2L} \frac{K_1 R_1}{(K_1 - K_2)} \exp \left[-\frac{R_1 K_1 - R_2 K_2}{K_1 - K_2} \Theta + \frac{P\eta}{2} \left(1 - \sqrt{1 + \frac{4K_1 K_2 (R_2 - R_1)}{P(K_1 - K_2)}} \right) \right] \\
 &\left\{ \operatorname{erfc} \left[\frac{\eta}{2} \sqrt{\frac{K_2 P}{\Theta}} - \sqrt{\left(\frac{P}{4K_2} + \frac{K_1 (R_2 - R_1)}{K_1 - K_2} \right) \Theta} \right] \right. \\
 &\quad \left. - \operatorname{erfc} \left[\frac{\eta}{2} \sqrt{\frac{K_1 P}{\Theta}} - \sqrt{\left(\frac{P}{4K_1} + \frac{K_2 (R_2 - R_1)}{K_1 - K_2} \right) \Theta} \right] \right\}
 \end{aligned}$$

Figure 2 shows numerical results for the two-member chain, $^{237}\text{Np} \rightarrow ^{233}\text{U}$, with impulse release. The dimensionless discharge rate is defined as the discharge rate divided by the release rate of ^{237}Np at the disposal site. The two peaks in Figure 2 show the contributions from the initial inventories of the parent (^{237}Np) and the daughter (^{233}U). Superimposed on these peaks is the contribution to the ^{233}U discharge that results from decay of ^{237}Np . Because the adsorption equilibrium constant for the daughter is greater than that for the parent, the parent's peak appears first and the daughter trails along behind.

The most important predictions of the analysis are: 1) the difference in adsorption characteristics between nuclides causes the inventory of a given nuclide to exit the soil column over the entire period between the times its precursors exit and the time the initial inventory of that nuclide exits, thereby greatly reducing the discharge rate of the nuclide at any one time; 2) axial dispersion significantly reduces the maximum discharge rate of the first chain member and the slowest decaying chain member but only slightly reduces the discharge rate of other chain members with significantly faster decay rates; 3) increasing soil column length and decreasing water velocity increase the migration times and decrease the maximum discharge rates of all nuclides; 4) the maximum discharge rate of a given nuclide occurs close to the exit time of its initial inventory if its half-life is long and

FIGURE 2. Dimensionless Discharge Rate as a Function of Time for the $^{237}\text{Np} \rightarrow ^{233}\text{U}$ Chain

occurs close to the exit time of its slowest decaying precursor if its half-life is short; 5) the situation with the greatest total nuclide discharge rate at any one time for a given water velocity and path length occurs when release is by the impulse mode, axial dispersion is negligible, and all chain members have the same equilibrium constant; and 6) the impulse release analysis is usually sufficiently accurate to describe chain migration, but increasing water velocity and leach time and decreasing path length, axial dispersion coefficient, and equilibrium constants all cause band analysis to be increasingly accurate relative to impulse analysis.

During the next quarter, a paper will be presented at the March 1975 meeting of the American Institute of Chemical Engineers in Houston which describes the detailed mathematical model used in the actinide chain migration calculations. The calcula-

tional models will be used to determine the effect of waste form (leach rate) and the degree of isolation (path length) on the dose to a population from a simulated release at a geologic disposal site. Application of the results to incentives for partitioning will be conducted in the Partitioning Program.*

Sorption of Americium on Soils -

R. C. Routson

Introduction

Plant uptake and sorption studies suggest that americium (Am) may be relatively mobile in certain soil systems. However, there is a general lack of data on Am sorption in soil systems of widely varying properties. The objective of this study was to measure the sorption of ^{241}Am as a function of sodium and calcium concentrations over the maximum concentration ranges expected in soil systems. Two soils were selected for the study: one soil, a Burbank sand subsoil from the arid Northwest and the other soil, a South Carolina subsoil from the humid Southeast. These soils represent the range in weathering stage common to the United States. The distribution coefficient (K_d) was chosen as the sorption parameter and is defined as the equilibrium activity sorbed on the soil in cpm per gram (g) divided by the equilibrium activity of the effluent per ml.

Methods

Sorption was measured in a batch equilibrium system at a 2.5:1 (25 ml

to 10 g) solution to soil ratio. Am values were measured on solutions composed of calcium nitrate [$\text{Ca}(\text{NO}_3)_2$] in concentrations of 0.002M, 0.02M, 0.05M, 0.10M, 0.20M, and sodium nitrate (NaNO_3) in concentrations of 0.015M, 0.030M, 0.30M, 0.75M, and 3.0M.

Tare weights were determined on 250-ml polyethylene screw cap centrifuge bottles. Approximately 10.0 g of quartered and well mixed soil were weighed into the tared centrifuge bottles. Twenty-five ml of the Na and Ca solution (without Am) were then placed in each bottle. Bottles were then placed on a shaker and moderately agitated for 20 to 30 min. The bottles were next centrifuged at 20 to 30°C for approximately 20 min at 4000 rpm and the supernatant carefully aspirated from the soil. Two additional prewashes of the matrix solution (solution without Am) were performed. After the third aspiration, the bottle was reweighed and the residual solution calculated by difference.

Twenty-five mls of the Na and Ca solution were then pipetted into the bottles, and the soil was swirled to assist mixing. Twenty-five μl spikes of ^{241}Am were added individually to each bottle following solution and soil mixing. Initial activities per bottle and solution pHs were approximately 3.7 μCi and 2.5 pH units, and 1.0 μCi and 3.1 pH units for the Burbank sand and South Carolina soils, respectively. The bottles were then agitated at moderate speed for 21 to

*See Section 2.0, "Separation of Actinide Elements from Radioactive Waste," in this Quarterly Report.

24 hr. Following agitation, the bottles were centrifuged as before and the supernatants placed in 50-ml polyethylene bottles for subsequent use.

Aliquots of 10 microliters (μ l) of supernatant were placed on 1-in. planchettes. Also added to the planchettes were 300 μ l of distilled water. Plates had been previously baked at high temperatures for several hours to remove any adhering grease or oil. Planchettes were then dried on a low heat setting of a hot plate and subsequently counted in a 2π gas flow system along with suitable backgrounds and standards.

The pH of selected samples was monitored with a single electrode pH meter. All data was run as four replicates per point.

Results and Discussion

An alpha energy analysis was performed on the ^{241}Am influent solution to determine the ^{241}Am purity, and no measurable amount of any other α -emitter was found present.

The equilibrium pH of the Burbank sand Na and Ca supernatants varied from 7.7 to 8.1. In contrast to the equilibrium pH, the South Carolina subsoil Na and Ca supernatants varied from 4.1 to 4.5.

Calculations of Kd values were based on the following equation:

$$K_d = \frac{(r C_o - C_e)}{C_e} \frac{A}{B} = \frac{\text{ml}}{\text{g}}$$

where A = total mls of solution in last treatment (25.0 mls + x residual mls), B = total grams (g) of soil present, r = ratio of trace solution

to total solution ($25/25 + x$ residual mls), C_o = concentration of trace constituent in effluent solution in cpm/ml, C_e = concentration of trace constituent in effluent solution in cpm/ml.

KdAm values for the South Carolina soil are tabulated in Tables 3 and 4 and plotted on Figure 3 for the Ca and Na solutions, respectively. In both cases, it can be seen that the KdAm decreased with increasing concentration of either Na or Ca. It can be seen from Table 3 and Table 4 that divalent Ca has a much greater effect on Am sorption than does monovalent Na. Linear regression analysis of the data for the South Carolina soil and Ca system (Table 3) gave a correlation coefficient (r) of -0.999, a standard error of the estimate of 0.08, and an equation of the line: $y = -1.34 - 0.89 x$ where y is the ln of the concentration and x is the ln of the KdAm. For the South Carolina subsoil and the Na⁺ system (Table 3), $r = -0.984$, $s = 0.47$, and $y = 1.11 - 1.02 x$. Inspection of Figure 3 and the r values shows a high degree of correlation in both cases; however, the variance of data about the Na line is much greater than the Ca variance.

Sorption of ^{241}Am by the Burbank sand subsoil was greater than anticipated, which resulted in only greater than values being determined. Calculated values of the KdAm ranged from 1200 to 8700 ml/g with the values being essentially independent of concentration of Na and Ca. At Kd values of this magnitude, small errors

TABLE 3. KdAm as a Function of Calcium Nitrate Concentration for a South Carolina Subsoil

Concentration of Ca(M)	KdAm (ml/g)
0.002	1.0 $\pm$ 0.04 ^(a)
0.02	2.2 $\pm$ 0.2
0.05	3.6 $\pm$ 0.6
0.10	8.4 $\pm$ 1.0
0.20	67 $\pm$ 5.0

a. $\pm$ standard deviation.

TABLE 4. KdAm as a Function of Sodium Nitrate Concentration for a South Carolina Subsoil

Concentration of Na(M)	KdAm (ml/g)
0.015	1.6 $\pm$ 0.1
0.030	2.9 $\pm$ 0.2
0.300	6.3 $\pm$ 0.7
0.750	130 $\pm$ 2.0
3.00	280 $\pm$ 24

FIGURE 3. Kd Am as a Function of the Na and Ca Solution Concentrations for a South Carolina Subsoil

in experimental procedure or counting variability result in a large change in the Kd value. For example, the percentage of Am sorbed at KdAm of 1200 and 8700 ml/g are 99.8% and 99.9%, respectively, with background counts being of the same order of magnitude as equilibrium solution counts. Thus, relatively small variations in count rate result in large KdAm changes at high Kd values, and the KdAm for the Burbank sand subsoil is best reported as >1200 ml/g although the actual values may be somewhat higher.

No further experimental work is planned. A topical report summarizing the work to date, entitled "Sorption of ^{99}Tc , ^{237}Np , and ^{241}Am on Two Subsoils from Differing Weathering Intensity Areas," will be issued.

Public Attitudes Toward Nuclear Waste Management - William S. Maynard (HARC)

Work completed during this quarter included 1) preparation of an informative, educational presentation highlighting the major issues in high-level nuclear waste disposal; 2) preparation of a survey questionnaire for obtaining public response to a set of issues associated with waste disposal; and 3) review of the entire research program by an external peer review committee which met at the Battelle Seattle Research Center on December 6 and 7, 1974. Participants in the peer review are listed in Table 5. As a result of the review, the videotape and questionnaire will be revised.

Work planned for the next quarter includes: modifying of methodology to take into account recommendations made by the review panel; pretesting of procedure; data gathering and analysis of results.

TABLE 5. External Peer Review
Conference Participants

External Review Group

Dr. Stephen Andersen
Staff Economist
Sierra Club

Dr. Kermit Garlid
Professor and Chairman
Department of Nuclear Engineering
Professor, Social Management of
Technology Program
University of Washington

Dr. Kai Lee
Research Assistant Professor
Political Science
Social Management of
Technology Program
University of Washington

Dr. Leon Rappoport
Associate Professor
Department of Psychology
Kansas State University

Dr. Keith Stamm
Associated Professor and Director
Communications Research Center
University of Washington

Battelle

Human Affairs Research Centers

William S. Maynard
Stanley M. Nealey
John A. Hebert

Pacific Northwest Laboratories

Jim Litchfield
John W. Bartlett

Informative Presentation. A script was prepared and used as a basis for a 30-min videotape presentation. The objective of the information presentation is to furnish respondents with a uniform level of information concerning nuclear waste disposal and to highlight a number of issues which are incorporated in the questionnaire that follows presentation. The review panel

acknowledged the difficulties in preparing a message on this topic which contains no biases, does not predetermine participant's responses, and accurately conveys the level of uncertainty surrounding many of the substantive aspects of nuclear waste disposal. A number of suggestions were made during the review which will be incorporated into the final form. It was also suggested that a narrated slide presentation be used rather than a film or videotape.

Questionnaire. A prototype questionnaire was presented to the review panel for consideration. The major sections of this questionnaire include: direct measurement of the relative importance of the four factors (long and short-term safety, costs, and control by man); indirect measurements via a policy evaluation procedure; measurement of attitudes toward related issues; and respondent evaluation of this procedure as a means of obtaining public input. This questionnaire is being revised slightly to take into account the review panel's recommendations. Changes will aim at streamlining the instructions and broadening the range of issues posed in the response instrument. The peer review committee has agreed to review and comment upon the revised procedures.

The review committee generally endorsed our proposal to sample respondents from intact groups. Committee members suggested a number of sampling dimensions and groups to consider in this study. Some of the sampling dimensions are age, geographic area, rural versus urban, and educational level. Groups that might be

sampled include those involved in relevant decision making processes (e.g., public utility officials), professional organizations, and civic action groups. Methods to obtain appropriate samples are being developed.

Task 3 - Special Evaluations

J. W. Bartlett

These studies support and result from plans development work (Task 1) and other waste management programs. Some studies are intended to provide definitive responses to specific problems or questions. Others are to initiate investigation of topics of concern but not yet under study in other programs.

Task 1 emphasizes developing background information for the waste disposal R&D program planning workshops. Thus effort within this special evaluations task was aimed primarily at identifying studies to assist the planning effort and guide future programs. Four topics were selected for further investigation.

1. Waste Management for Long-Lived

Fission Products. In contrast to the actinides, long-lived fission products such as ^{129}I , ^{99}Tc , and ^{79}Se may transport readily in geologic formations and the biosphere because of their relatively high solubility in water. Concepts similar to actinide partitioning, i.e., processing to isolate the nuclides in order to use special waste management procedures, may be useful or necessary for these isotopes.

At present, programs are under way at various laboratories to provide an initial assessment of incentives and

feasibilities for special waste management procedures for these isotopes. When results from these ongoing studies are sufficient, they will be used to scope and initiate the study within this task.

2. Irretrievable Loss of Waste to the

Sea. Ultimate disposal to space, an ice sheet, or the seabed share the common possibility of irretrievable loss of a waste package to the oceans. Waste disposal via any of these options might therefore require that the waste be packaged to satisfy this contingency; the packaging requirements might be so stringent that these disposal methods would be unattractive in comparison with geologic disposal on land. An evaluation of this topic might therefore be an important element in decisions on programs concerning space, ice, and seabed disposal.

Investigation of this topic requires the expertise of a variety of disciplines. It also requires evaluation of problems such as packaging tradeoffs (e.g., designing the package to prevent failure may inhibit detection and therefore promote irretrievability), probability that loss will be irretrievable, and the consequences of loss.

Preliminary evaluation of packaging requirements for disposal to space were reported in BNWL-1900.⁽¹⁾ Initial discussions of irretrievable loss at sea were held at the planning workshop at Salt Lake City in December (cf. Task 1). These discussions indicated that losses might actually be retrievable under most circumstances.

Gathering of information on this topic will be renewed at the Denver planning workshop in January. The study effort will be defined after that meeting.

3. Screening Comparisons of Waste

Management Options. Because of alternatives for disposal modes (e.g., land or seabed), disposal sites, and pre-disposal processing and handling, hundreds of specific options are being considered. Highly detailed evaluation and comparison of all such options is not practical; methods are needed to validly and efficiently winnow such options to those worthy of detailed study.

These screening comparison methods should incorporate concepts that are typical of cost-benefit analysis and risk evaluation methodology. They should not, however, be as complex and detailed as these methodologies usually are. Yet they should be capable of identifying, evaluating, and comparing the important characteristics of alternative waste management options.

Methods for making such comparisons are not available today. If analytical rather than highly judgmental comparisons are to be made when many options are being considered, the methods will have to be developed.

The need for these capabilities will be considered in the planning workshops.

4. Pilot Plant Testing. Development programs usually contain a pilot plant testing phase in which operation of the system in question is observed and adjustments are made as necessary. If such testing is to be used within the disposal development program, it will be necessary to demonstrate that it is adequate and meaningful. Problems such as extrapolation of short-term data to long time periods, feasibility of use of stand-in materials, requirements and feasibility for data, and site selection will have to be evaluated. Possibilities for using simulated disposal environments should also be investigated.

The need for results from such a study may be many years in the future. However, a preliminary consideration of the problem may affect the R&D program plan for disposal. This topic will therefore be considered in the planning workshops.

REFERENCES

1. K. J. Schneider and A. M. Platt, Editors, Advanced Waste Management Studies: High-Level Radioactive Waste Disposal Alternatives, USAEC Report, BNWL-1900, Battelle, Pacific Northwest Laboratories, Richland, WA, May 1974.

2.0 SEPARATION OF ACTINIDE ELEMENTS FROM RADIOACTIVE WASTE

E. J. Wheelwright - Project Manager

The primary FY75 objective is to determine the effect of actinide partitioning upon advanced waste management concepts and thereby demonstrate the incentives or lack thereof for partitioning. A secondary objective is to develop the necessary technology for performing the partitioning processes and thus determine the technical feasibility.

SUMMARY

The program was redirected during this report period by the addition of two tasks, 5 and 6, designed to evaluate the safety implications of partitioned waste compared with nonpartitioned waste and to determine the incentives for partitioning. These two tasks have been given a higher priority than the first four tasks, and they should provide answers to the question, "Should partitioning be adopted as an integral part of waste management?"

Task 3 was not assigned until late in the period because of its low priority. As a result, there is no definitive work to report.

The design, fabrication, and testing of equipment for preparing dissolver solutions from irradiated fuel are nearly completed. Cleanup of "B" cell in which the dissolution studies are to be made is underway.

The calcine dissolution studies have shown that simulated calcine is very difficult to dissolve in a sin-

gle system but can be solubilized by a combination of chemical treatments.

A conceptual partitioning process has been outlined for removing americium and curium from the high-level waste and decontaminating them from selected fission products. The waste from this process would be suitable for solidification by spray calcination and/or conversion to a glass. A key step in the process is a "no additives" solvent extraction process to separate americium and curium from some of the lanthanide as well as other fission products. Dibutyl butylphosphonate shows promise as the extractant for this process; other extractants are also being tested as they become available.

A rationale has been developed for defining incentives for partitioning. Preliminary results for a simulated release by leaching from a geologic disposal site indicate that daughters in actinide chains may be important relative to inventories of single nuclides.

TECHNICAL PROGRESS

Task 1 - Characterization and Treatment of Solids in High-Level Wastes -

F. P. Roberts and J. H. Westsik

Objectives of this task are to characterize the solids found in reprocessing-plant dissolver solution and the solids formed during the reprocessing of irradiated fuel. The development of technology for treating these solids to allow the removal of long-lived nuclides is also an objective of this task.

Laboratory-scale studies of dissolver solution solids are to be performed in B-cell in the 325 Building High-Level Radiochemistry Facility. This cell and the associated equipment are now undergoing renovation and cleanup. A bench-scale dissolver has been fabricated for preparation of small batches (100 to 200 g U) of dissolver solutions using irradiated Yankee fuel as feed. The fuel has been cut into three-fourths to one-inch segments suitable for dissolution.

A flowsheet for the dissolution has been developed which will simulate the plant-scale process expected to be used in future fuel reprocessing plants. This flowsheet, which can be closely controlled, will provide a source of dissolver solids for the characterization.

An eight-stage centrifugal contractor has been obtained and is being overhauled for use in preparation of aqueous high-level wastes from the irradiated Yankee fuel. This aqueous waste is to be used in evaluating the effects of the solids (formed during the solvent extraction and subsequent high-level waste concentration) on the partitioning process.

Task 2 - Removal of Long-Lived Nuclides from Solidified Waste -

J. A. Partridge

The objective of this task is to develop a flowsheet for partitioning solidified high-level waste, e.g., a calcine. The work this quarter has been directed toward developing a dissolution scheme for getting the calcined waste into solution for subsequent partitioning processing. Some of the fission products in the waste, e.g. molybdenum and zirconium, are capable of forming highly refractory compounds during the calcination process. These are difficult to dissolve.

The laboratory work during this period has been done with simulated calcine prepared by pot calcination of a synthetic high-level waste referred to as PW-4b. This is a low-sodium fission product waste prepared with the following chemical substitutions: Fe for Ru, Co for Rh, Ni for Pd, Mo for Tc, K for Rb, rare earths for Y + Am + Cm, and U for Pu + Np. The synthetic waste contains solids; i.e., not all of the constituents completely dissolve during preparation of the "solution."

Results of acid digestions on the simulated calcine are shown in Table 6. The results show that nitric-hydrofluoric acid digestion is better than nitric acid alone, but it is not adequate for dissolution of the calcine.

Dissolution of the calcine by means of molten salt fusions was also investigated. The fusions were done in nickel crucibles. After the fusion treatment, the cooled melt was dissolved in nitric acid. Results of

TABLE 6. Dissolution of Simulated Calcine by Acid Digestion

Acid Test No.	Acid	ml Acid/ g Calcine	Temp.	Time, hr	% Calcine Dissolved
1	15.7M HNO ₃	10	Boiling	4	28
2	Con HCl	10	Boiling	4	75
3	15.7M HNO ₃ -0.3M HF	10	80°C	5	57
4	15.7M HNO ₃ -0.03M HF	10	80°C	24	60
5	15.7M HNO ₃ -0.03M HF	30	95°C	5	70
6	15.7M HNO ₃	5	Boiling	2	
	followed by				
	4M HNO ₃	30	Boiling	1	35
7	15.7M HNO ₃ -0.003M HF	5	Boiling	2	
	followed by				
	4M HNO ₃ -0.01M HF	35	Boiling	1	55
8	4M HNO ₃ -0.03M HF	25	100°C	1	53

these tests are shown in Table 7. A caustic-peroxide fusion was the best of the three types investigated, but even it would not completely solubilize the calcine.

The results of dissolution tests on the simulated calcine thus far indicate that a combination or series of chemical treatments will be required to dissolve all of the calcine. For example, neither aqueous acid digestion nor molten salt fusions resulted in complete dissolution of the calcine. However, when a nitric-hydrofluoric acid digestion was performed on the residual solids from fusion test No. 3 (Table 7), an overall dissolution of 99% was achieved; and when a caustic-peroxide fusion on the residual solids from acid test

No. 8 (Table 6) was performed, an overall dissolution of 98% was achieved.

A second batch of simulated calcine was prepared from filtered PW-4b synthetic high-level waste. Nitric-hydrofluoric acid digestion tests on this calcine were more successful than those on calcine prepared with unfiltered synthetic high-level waste, as shown by the data in Table 8. This indicates that the solids filtered from the waste solution prior to preparation of this calcine are refractory or are converted to refractory compounds in the calcination process.

Characterization tests of residual solids from dissolution experiments are planned in an effort to further evaluate dissolution processes.

TABLE 7. Dissolution of Simulated Calcine by Molten Salt Fusion

Fusion Test No.	Fusion Mixture	Temperature, °C	Time, hr	% Calcine Dissolved
1	Sodium hydroxide-sodium peroxide	600	1	77
2	Sodium hydroxide-sodium peroxide	600-650	2	90*
3	Sodium hydroxide-sodium peroxide	600-650	2	87
4	Alkali carbonate	600	2	33
5	Sodium-Potassium Nitrate	600	1-1/4	40

*The cooled melt was dissolved in 4M HNO₃ containing 0.006M HF in this test.

TABLE 8. Acid Dissolution of Simulated Calcine Prepared from Filtered Synthetic Waste

Acid Test No.	Acid	ml Acid/g Calcine	Temp., °C	Time, hr	% Calcine Dissolved
1	15.7M HNO ₃ -0.007M HF	40	80	6	90
2	15.7M HNO ₃ -0.045M HF	40	80	6	92
3	15.7M HNO ₃ -0.15M HF	40	80	6	93

Work is proceeding toward preparation of some actual calcine from "typical" high-level waste in the 325 Building High Level Radiochemical Facility. This calcine will be used to test the most promising dissolution procedures obtained with the simulated calcine. The waste to be used for preparation of this calcine will be that obtained from processing irradiated power reactor fuel through TBP cycles simulating the Purex process in conjunction with the work in Task 1.

Further work is necessary to get an optimum calcine dissolution procedure, but the results obtained with

nitric-hydrofluoric acid digestion followed by peroxide fusion of the residual solids indicate that complete dissolution of the calcine is possible.

Task 3 - Removal of Long-Lived Nuclides from Non-High-Level Wastes - J. H. Westsik

This task was assigned in late December, and there is no progress to report. The objective will be to survey the general area of non-high-level waste to determine the scope of the problem and define the need, if any, for partitioning.

Task 4 - Development of Low-Additive
Flowsheets for Removing Long-Lived
Nuclides from the Fission Products -
J. L. Swanson

The objective of this task is to develop and test methods for purifying the high-level fission product waste from potentially hazardous long-lived nuclides (primarily actinides) by processes which do not add excessive amounts of additives to the waste. The major concern at the moment is the development of methods for the separation of the americium and curium from the fission products since this is thought to be the most difficult problem to be solved.

One very important aspect in determining what level of additives in the waste is excessive is the effect of the additives on the process used to convert the liquid waste to solid. An acceptable (perhaps even desired) additive content for one waste solidification process may be unacceptable for another process; for example, the presence of a moderate amount of sodium in the liquid waste is deleterious if the liquid is to be solidified by fluidized bed calcination but not if it is to be solidified by spray calcination or by incorporation in a glass. Results obtained in the Waste Fixation Program indicate that suitable low-melting glass compositions can contain significant amounts of such potential partitioning process salt additives as:

sodium - Na_2O to fission product
oxide weight ratio of about
0.6,
calcium - CaO to f.p. oxide ratio of
about 0.3,

zinc - ZnO to f.p. oxide ratio of
about 1.0,

aluminum - Al_2O_3 to f.p. oxide ratio
of about 0.06.

It would appear to be much easier to develop processes acceptable to such a waste solidification process than to one requiring the absence of such additives.

A conceptual partitioning process for purifying the high-level waste from americium and curium and also purifying the Am and Cm from the fission products while providing a waste suitable for solidification by spray calcination and/or conversion to glass is as follows (highly simplified):

- Extraction of Am and Cm from the high-level waste solution into dibutyl butylphosphonate (DBBP) and scrubbing with $\sim 5\text{M}$ HNO_3 to effect significant separation from less extractable elements (including lanthanum and cerium).
- Stripping of Am and Cm from the DBBP into $\sim 8\text{M}$ HNO_3 to effect significant separation from more extractable elements (including many of the heavier lanthanides as well as other fission products).
- Purification of the stripped Am and Cm from similarly extractable elements (likely praseodymium, neodymium, and promethium) by ion exchange or solvent extraction processes employing complexants (e.g., DTPA) to improve separation.
- Stripping of the more extractable elements from DBBP (by dilute acid) for recombination with the less extractable elements.
- Solvent cleanup as required.

In this process the step which adds the bulk of the salts to the waste is the purification of the Am and Cm from the similarly extractable elements by processes employing complexants. Such processes require at least partial neutralization of the complexants, generally by sodium or ammonium hydroxide so that sodium or ammonium ion is added to the waste. The use of cation-exchange chromatography (CEC) also involves use of a barrier ion (generally zinc) which also ends up in the waste unless it can be recycled. We estimate that a CEC process using DTPA completely neutralized with NaOH (Na_5DTPA) to produce a purified Am + Cm fraction would add to the waste only about 0.5 g Na_2O per g of fission product oxide if the feed to the CEC process is largely purified from all contaminants except praseodymium, neodymium, and promethium. The addition of zinc would amount to only about 0.1 g ZnO per g of fission product oxide. If the feed to the CEC process were to contain all of the fission product lanthanides, about twice as much sodium and zinc would be added to the waste, and the sodium content would then be higher than desired for incorporation into glass.

The DBBP solvent extraction process outlined above to accomplish separation of the Am and Cm from the bulk of the fission products is based primarily on data obtained from the literature. Other phosphonates and perhaps phosphinates may be more suitable for the application, and efforts are in progress to obtain such extractants for evaluation. In the

meantime, we plan to proceed with development of detailed flowsheets using DBBP, which is readily available. Tests using simulated waste solutions in our miniature centrifugal contractors will begin in the near future.

Two main reasons for choosing DBBP as the first extractant for in depth study are: 1) its ability to extract Am and Cm from HNO_3 concentrations which are easy to achieve and which would not likely lead to formation of troublesome solids and 2) the fact that appreciable separation from many of the lanthanides can be achieved without the use of added complexants. Data illustrating these facts and comparing DBBP with other neutral organophosphorous extractants are shown in Figures 4 and 5.

The bidentate compound, dibutyl-N, N-diethyl carbamylmethylene-phosphonate (DBDECMP), is seen (Figure 4) to extract Am very strongly but to give relatively little separation from Ce. The phosphinate, butyl dibutylphosphinate (BDBP), is seen to extract Am quite well, but nothing is known of the separation from Ce. The phosphate, tributyl phosphate (TBP), gives appreciable separation from Ce but does not extract Am sufficiently well except at high HNO_3 concentrations. The phosphonate, dibutyl butylphosphonate (DBBP), extracts Am sufficiently well for a purification process at both low and high HNO_3 concentrations and also gives appreciable separation from Ce. Additional data relative to the separation of Am from some other lanthanide elements by DBBP extraction from HNO_3 solutions are presented in Figure 5.

FIGURE 4. A Comparison of the Distribution Coefficients of Selected Lanthanide and Actinide Ions with Potential Extractants

Task 5 - Evaluation of Incentives for Partitioning -

G. Jansen, H. C. Burkholder, and
D. A. Baker

The purpose of this task is to determine under what conditions incentives exist for partitioning high-level wastes before ultimate disposal/storage. The scope is limited to an evaluation of the effect of partitioning on the safety of various storage

FIGURE 5. Distribution Coefficients of Selected Lanthanide and Actinide Ions with DBBP

forms and storage modes after the interim storage phase is completed and after all handling operations for ultimate disposal are completed.

Task 5 also deals with the incentives for partitioning by considering the health risk over an indefinite future period. For wastes generated by the U. S. Nuclear Fuel Economy through the year 2000, this period begins in the year 2100 (interim storage is assumed between 2000 and 2100).

Storage/disposal is to be compared for three source terms:
a) unpartitioned high-level wastes,
b) long-half-life (primarily the actinide) fraction of the partitioned wastes, and

- c) the short-half-life (primarily fission product) fraction of the partitioned waste.

In the initial evaluation, the decontamination factors of each waste fraction from the other are assumed infinite. The storage modes will be: 1) geologic storage in a nonsalt, western U.S. desert geologic formation, and 2) surface storage in a western desert for each of the three source terms.

The risks associated with storage/disposal are a function of the source radioactivity, the waste form, the probability of occurrence of release incidents, and the remoteness of the source from man's environment. The risk of storage/disposal of unpartitioned waste can be described approximately by the equation:

$$\text{Risk} = \sum_{\text{All Incidents}} \left(\begin{matrix} \text{Radioactive} \\ \text{Source} \end{matrix} \right) \times \left(\begin{matrix} \text{Probability} \\ \text{of Incident} \end{matrix} \right) \times \left(\begin{matrix} \text{Waste} \\ \text{Form} \end{matrix} \right) \times \left(\begin{matrix} \text{Remote-} \\ \text{ness} \end{matrix} \right)$$

If the waste is partitioned into a short-lived fraction and long-lived fraction, the risk is the sum of the risks associated with the storage/disposal of each fraction.

$$\text{Risk}_{\text{Mixed Waste}} = \text{Risk}_{\text{Long-Lived}} + \text{Risk}_{\text{Short-Lived}}$$

The purpose of Task 5 is to determine under what conditions partitioning and separate storage/disposal can decrease the risk over the value for mixed waste. For example, the long-lived waste fraction might be stored

geologically as a glass and the short-lived waste fraction might be stored on the ground surface as a salt. A complete risk analysis is not possible within the time frame of the study. Therefore, analysis of only a single, but important, pathway for each of the two storage concepts will be attempted. The leach incident pathway with chronic exposure to a population will be used for geologic storage, and the direct atmospheric release pathway with acute exposure of a population will be used for surface storage. Parameters to be varied in the comparison are:

- 1) waste form (typified by leach rate in the leach incident, and fraction released and particle size in the acute atmospheric release),
- 2) time of the release incident, and
- 3) degree of geologic isolation (typified by the path length to discharge to a river).

The results will be expressed in terms of radiation dose to a population.

The incentives for partitioning are evaluated by comparing the trade-offs between various parameters. For example, if mixed waste stored geologically required a significantly longer path length to the river to give the same dose as the worst partitioned fraction, then there might be an incentive for partitioning. Likewise, if mixed waste required a less leachable material than the long-lived fraction, then there might be an incentive for partitioning. And, finally, if complete elimination of

one waste fraction by space disposal or transmutation greatly reduces the population dose, then there might be an incentive for partitioning followed by these disposal schemes.

To date, partial results have been obtained using methodology developed in the Advanced Waste Management Program for the simulated release of the contents of a high-level waste geologic disposal site by contact with ground water.⁽¹⁾ For the initial calculation, ground water from a typical, arid western U. S. region is assumed to leach all the unpartitioned wastes from the year 2000 over a 333-year period into a 10-mile soil column through which water travels in 145 years. Soil retardation factors for radionuclides and other properties of the system are given in BNWL-1900.⁽²⁾ The waste inventories (also given, in Section 2 of BNWL-1900) include all manmade radioactivity generated in the cladding, fuel, and fuel impurities as calculated by the ORIGEN code.

The rates of discharge of radionuclides from the soil column into a river are shown in Figure 6. Depending upon their specific chemical properties, each radionuclide is delayed relative to the water travel time, separating them into sharp chromatographic concentrations; peaks are reduced by axial dispersion in the soil column and by the finite leach rate. Familiar nuclides such as ^{90}Sr , ^{137}Cs , ^{239}Pu and ^{241}Am decay to insignificance while traveling to the river. The time scale for complete discharge of radionuclides to the river is 7 million years.

This is clearly beyond any time for which an attempt to predict the course of human civilization might be made, although some nuclides such as ^{99}Tc and ^{129}I , which are not retarded, discharge in a few hundred years. Nuclides such as ^{10}Be , ^{14}C , ^{41}Ca , ^{59}Ni , ^{93}Mo , and nonfission product ^{93}Zr originate in fuel impurities and cladding. The earliest actinide to appear in the river is ^{237}Np at 15,000 years, but most actinides travel much more slowly. The effect of the soil column on migration of radioactive actinide decay chains is important and unique. The discharge of the daughters in these chains is spread over a very long time, reducing the concentration levels in the river. Although nuclides such as ^{241}Am and ^{241}Pu do not survive to discharge to the river, they do contribute significantly to the quantities of daughters such as ^{237}Np .

Figures 7, 8, 9, and 10 show the neptunium, curium, plutonium, and uranium peaks in detail. The results show the low discharge rates of daughters in actinide decay chains, and the complexity of the chain interaction suggests that partitioning actinides from each other will not improve the safety of ultimate disposal.

The significance of the discharge rates in terms of dose to population is not yet certain; such calculations will be made during the next report period. The results will then be used to estimate the value of partitioning in trade-offs with reduced leachability and varying path length.

FIGURE 6. Geologic Disposal of High-Level Radioactive Wastes Generated by the U.S. Nuclear Power Economy Through the Year 2000. Discharge of radionuclides from a ten-mile soil column after a hypothetical leach incident at year 2100.

FIGURE 7. Neptunium Peak

FIGURE 8. Curium Peak

FIGURE 9. Plutonium Peak

FIGURE 10. Uranium Peak

Task 6 - Comparison of Waste Management System With and Without Partitioning -

J. W. Bartlett

The scope and content of work in this task is highly contingent on the results of work in Task 5. It is also contingent on capability to characterize flowsheets and other parameters of the alternatives with detail sufficient to make the necessary comparisons.

Significant effort in this task will not start until results of Task 5 are sufficient to decide if incentives for partitioning justify the effort. Initial Task 5 results are expected during the next quarter.

Preliminary effort has been aimed at two areas: compilation of information concerning partitioning

processes, and development of methodology for comparing waste management systems. Information from other laboratories and progress in developing R&D plans for waste disposal are being monitored. Development of the methodology has focused on identifying criteria for comparison and identifying system parameters to be evaluated.

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1. G. J. Dau, Compiler, Nuclear Waste Management and Transportation Quarterly Progress Report, July through September 1974, BNWL-1876, Battelle, Pacific Northwest Laboratories, Richland, WA, December 1974.
2. BNWL-1900, High-Level Waste Management Alternatives, May 1974, pp. 352-356. Note that an error in the calculations for this report (which presents methodology only) spreads the chromatographic bands in Figure 3.13 over far too wide a time scale.

3.0 DECONTAMINATION AND DENSIFICATION OF CHOP-LEACH CLADDING RESIDUES

R. L. Dillon and R. S. Kemper

Fuel element cladding residues from the chop-leach head end process constitute a low-density, high-surface area metallic waste with a substantial actinide element contamination. The objective of this study is to determine the feasibility of and to develop methods for removal of long half-life transuranics from the cladding residues and for consolidation of the zirconium and stainless steel residues. The study involves:

1. *Selection and laboratory testing of methods for transuranic removal by*
 - a) *aqueous reagents*
 - b) *molten salt reagents*
 - c) *fluxing during melting.*
2. *Consolidation of the fuel element hulls by melting and casting Zircaloy, stainless steel, Inconel or alloys of the three.*
3. *Testing and evaluation the consolidated fuel hulls for long-term waste management.*
4. *Absorption and fixation of tritium into the waste ingots.*
5. *Consolidation of transuranics removed.*

SUMMARY

An accurate knowledge of the details of transuranic contamination in cladding hulls is necessary in developing an effective decontamination process. We made an initial determination of the type, amount and location of transuranic contamination on a short piece of irradiated Zircaloy-4 cladding from a Saxton (Pu, U) O₂ fuel rod. Most of the contamination was superficial to the internal oxide. A lesser amount was incorporated in the oxide and on the Zircaloy metal surface while an amount predictable from the initial uranium and thorium contamination of the metal was found

in the metal matrix. All of these contamination levels were more than an order of magnitude larger than the maximum amount allowed for nonrecoverable storage (i.e. 10 nci/g).

Development of decontamination and densification processes for cladding hulls continues. Mini-scale investigations of fluoridation reactions of oxidized Zircaloy showed that hydrogen fluoride at about 600°C for 30 to 45 min, followed by treatment in a solution of ammonium oxalate, citrate, and fluoride containing hydrogen peroxide at 90°C for 1 hr produced the most effective oxide removal at ac-

ceptable metal corrosion losses of the processes tested. This process was applied to one short piece of irradiated fuel cladding with reasonably good results. Boron trifluoride, carbon tetrafluoride, dichlorodifluoromethane and mixtures of HF and BF_3 were explored as fluorinating agents. Modifications of fluoride, citrate, and hydrogen peroxide concentrations in the aqueous reagent were also explored, but none of the alternative solutions led to a better overall process.

The chemical and metallurgical properties of the Zircaloy-Inconel stainless steel billets representing densified, leached fuel residues were determined. Chemical analyses of the raw material used and 5 of the 6 heats made to date have been received. Major alloying elements agree well with the design compositions. Only slight increases in C, H, O, and N were noted.

Microprobe scans of specimens taken from center and edge ingot locations show no differences in chemical compositions. Microprobe analyses of the three major metallographic constituents have been performed.

TECHNICAL PROGRESS

Actinide Distribution in Chop-Leach Fuel Hulls - B. Griggs

The design of surface decontamination and subsequent molten metal decontamination processes for fuel hulls requires an accurate knowledge of the type, chemical state, and distribution of transuranic contamination of the hull material. A sample for analytical determination of these

factors was obtained from a fuel rod (F-2 [HQ]) exposed in Saxton Reactor about 6 yr ago. Fuel composition was 6.6% PuO_2 in UO_2 . A 0.6-cm piece of F-2 (HQ) cladding was lightly surface abraded to remove corrosion product oxide and etched 30 sec total in two nitric-hydrofluoride acid etch solutions. The decontaminated piece was moved to an uncontaminated area and etched 20 sec more in two etches, and briefly leached in concentrated nitric acid for complete surface decontamination. Less than 25 μm was removed per surface in the total treatment sequence. The sample was then dissolved in etch solution in two approximately equal steps. One sample contained the dissolved outer and inner 0.014-cm surface layer and the other sample the residual 0.028-cm center. Radiation readings with survey instruments after decontamination showed no detectable α and 20 mR γ at contact. A second sample from the same section of fuel rod cladding (not abraded) was treated in 0.4 M ammonium oxalate - 0.16 ammonium citrate - 0.1 M ammonium fluoride - 0.3 M hydrogen peroxide solution at 90°C for 2 hr and subsequently treated again under the same conditions with fresh solution for an additional 2 hr. The residual 1.1-cm piece of cladding was dissolved in 6 M HNO_3 - 1.7 M HF solution. An adjacent 0.43-cm piece of cladding which had been abraded and etched clean was dissolved to use as a correction for the previously dissolved piece with surface contamination. The results of these analytical determinations are shown in Table 9.

	Surface Leach ^(a)	Surface Oxide ^(b) +25.4 μ of Metal	Base Metal ^(c) Outer & Inner 0.14 mm	Base Metal ^(d) Center 0.28 mm	Contamination Ratio ^(e) Total Activity/ Base Metal Activity
U	3.81 10 ⁻³ g/dm ² 2.61 10 ⁻⁶ ppb(F)	3.51 10 ⁻³ g/dm ² 1.75 10 ⁻⁶ ppb(F)	2.98 10 ⁻⁵ g/dm ² 1.68 10 ⁻³ ppb(F)	2.80 10 ⁻⁵ g/dm ² 1.58 10 ⁻³ ppb(F)	1.3 10 ²
Pu	3.20 10 ⁻⁴ g/dm ² 2.20 10 ⁻⁵ ppb	4.12 10 ⁻⁴ g/dm ² 2.05 10 ⁻⁵ ppb	1.02 10 ⁻⁶ g/dm ² 5.50 10 ⁻¹ ppb	1.04 10 ⁻⁶ g/dm ² 5.62 10 ⁻¹ ppb	3.6 10 ²
Total α (g)	9.94 10 ⁻⁵ Ci/dm ² 6.82 10 ⁻⁵ Ci/g	9.88 10 ⁻⁵ Ci/dm ² 4.92 10 ⁻⁵ Ci/g	1.29 10 ⁻⁷ Ci/dm ² 7.26 10 ⁻⁹ Ci/g Zr	1.38 10 ⁻⁷ Ci/dm ² 7.46 10 ⁻⁹ Ci/g Zr	7.4 10 ²
²³³ U	Not determined	Not determined	3.60 10 ⁻¹⁰ Ci/dm ² 2.03 10 ⁻¹¹ Ci/g Zr	3.47 10 ⁻¹⁰ Ci/dm ² 1.87 10 ⁻¹¹ Ci/g Zr	Not determined
Pu	4.4 10 ⁻³ Ci/dm ² 3.02 10 ⁻³ Ci/g	6.1 10 ⁻³ Ci/dm ² 3.04 10 ⁻³ Ci/g	3.66 10 ⁻⁶ Ci/dm ² 2.06 10 ⁻⁷ Ci/g Zr	3.90 10 ⁻⁶ Ci/dm ² 2.1 10 ⁻⁷ Ci/g Zr	1.4 10 ³
²³⁸ Pu	9.35 10 ⁻⁶ Ci/dm ² 6.41 10 ⁻⁶ Ci/g	2.53 10 ⁻⁵ Ci/dm ² 1.26 10 ⁻⁵ Ci/g	1.12 10 ⁻⁸ Ci/dm ² 6.30 10 ⁻¹⁰ Ci/g Zr	1.18 10 ⁻⁸ Ci/dm ² 6.37 10 ⁻¹⁰ Ci/g Zr	1.5 10 ³
²⁴¹ Pu	4.35 10 ⁻³ Ci/dm ² 2.98 10 ⁻³ Ci/g	6.0 10 ⁻³ Ci/dm ² 2.99 10 ⁻³ Ci/g	3.73 10 ⁻⁶ Ci/dm ² 2.10 10 ⁻⁷ Ci/g Zr	3.81 10 ⁻⁶ Ci/dm ² 2.05 10 ⁻⁷ Ci/g Zr	1.4 10 ³
²⁴¹ Am	Not determined	Not determined	5.25 10 ⁻⁸ Ci/dm ² 2.96 10 ⁻⁹ Ci/g Zr	5.25 10 ⁻⁸ Ci/dm ² 2.83 10 ⁻⁹ Ci/g Zr	Not determined
²⁴⁴ Cm	Not determined	Not determined	1.37 10 ⁻⁹ Ci/dm ² 7.70 10 ⁻¹¹ Ci/g Zr	1.40 10 ⁻⁹ Ci/dm ² 7.53 10 ⁻¹¹ Ci/g Zr	Not determined
¹³⁷ Cs ^(h)	9.43 10 ⁻² Ci/dm ² 6.47 10 ⁻² Ci/g	9.19 10 ⁻² Ci/dm ² 4.58 10 ⁻² Ci/g	2.40 10 ⁻⁵ Ci/dm ² 1.35 10 ⁻⁶ Ci/g Zr	2.51 10 ⁻⁵ Ci/dm ² 1.35 10 ⁻⁶ Ci/g Zr	3.8 10 ³
¹²⁵ Sb ^(h)	5.07 10 ⁻⁴ Ci/dm ² 3.48 10 ⁻⁴ Ci/g	1.03 10 ⁻³ Ci/dm ² 5.13 10 ⁻⁴ Ci/g	5.13 10 ⁻³ Ci/dm ² 2.89 10 ⁻⁴ Ci/g Zr	5.51 10 ⁻³ Ci/dm ² 2.97 10 ⁻⁴ Ci/g Zr	1.1 10 ⁰
⁶⁰ Co ^(h)	Not determined	5.21 10 ⁻⁵ Ci/dm ² 2.60 10 ⁻⁵ Ci/g	1.11 10 ⁻⁴ Ci/dm ² 6.25 10 ⁻⁶ Ci/g Zr	1.19 10 ⁻⁴ Ci/dm ² 6.39 10 ⁻⁶ Ci/g Zr	1.2 10 ⁰
⁵⁴ Mn ^(h)	Not determined	2.59 10 ⁻⁵ Ci/dm ² 1.29 10 ⁻⁵ Ci/g	1.49 10 ⁻⁵ Ci/dm ² 8.42 10 ⁻⁷ Ci/g Zr	1.61 10 ⁻⁵ Ci/dm ² 8.67 10 ⁻⁷ Ci/g Zr	1.8 10 ⁰
²³³ U	Not determined	Not determined	0.1233 atom %	0.1222 atom %	
²³⁴ U	0.0060 atom %	0.0067 atom %	0.0099 atom %	0.0099 atom %	
²³⁵ U	0.5533 atom %	0.5917 atom %	0.5590 atom %	0.5556 atom %	
²³⁶ U	0.0473 atom %	0.0481 atom %	0.0414 atom %	0.415 atom %	
²³⁸ U	99.3933 atom %	99.3535 atom %	99.2664 atom %	99.2708 atom %	
²³⁸ Pu	0.25 atom %	0.25 atom %	0.07 atom %	0.07 atom %	
²³⁹ Pu	70.37 atom %	69.41 atom %	84.29 atom %	84.88 atom %	
²⁴⁰ Pu	18.10 atom %	18.44 atom %	11.63 atom %	11.02 atom %	
²⁴¹ Pu	9.63 atom %	10.17 atom %	3.68 atom %	3.70 atom %	
²⁴² Pu	1.64 atom %	1.74 atom %	0.34 atom %	0.33 atom %	

- Radioactivity removed by leaching fuel cladding piece in 0.4 M ammonium oxalate - 0.16 M ammonium citrate - 0.1 M ammonium fluoride - 0.3 M hydrogen peroxide solution for two 2-hr periods at 85°C. The 1.1-cm piece of clad (6.97 cm² total area; 3.08 cm² ID area) had been leached previously in boiling 4 M HNO₃ for 1 hr followed by overnight soak in the same solution to simulate dissolver treatment. The ID area was used for all areal calculations on the assumption it was the contaminated area. Weight losses on leaching of 29.6 mg and 15.3 mg for the first and second leach, respectively, were used to normalize to weight even though these values contain some corrosion of exposed end metal.
- The surface oxide contamination was determined by dissolving the oxide and metal piece completely and then correcting for the dissolved metal by subtracting the base metal results. Surface areas are related to ID surface area of 3.08 cm². Weight data is assumed to be an estimated ID oxide thickness⁽⁴⁾ of 6.35 μ m plus 25.4 μ m of ID metal for a total of 61.8 mg. The metal layer immediately adjacent to the oxide layer has not been analyzed separately. This layer may have the same low concentrations of transuranics as the base metal. The thin metal layer is included here because in any practical decontamination scheme removing the oxide layer will result in some minimal removal of base metal.
- Cladding which had been completely surface decontaminated by abrasion and etching was dissolved in etch solution to produce a nearly uniform removal of metal from all surfaces. Results stated in units/unit area use the ID surface area of 1.59 cm².
- This is the residue from the dissolution previously described in note c. The same ID surface area is used for comparison purposes only.
- The ratio of surface normalized activities is calculated to show a rough measure of the decontamination factor that could be expected by removing surface oxide from a cladding hull.
- ppb is the weight fraction times 10⁹.
- Total α measurements are affected by self shielding and are only an approximation to insure that all α emitters of significance have been included in separate analyses.
- Decay corrected to mid-November 1974.

TABLE 9. Analyses of Irradiated Saxton Fuel Clad

The data of Table 9 show that the surface oxide is substantially contaminated; however, little if any of the fuel material diffuses more than a few microns into the Zr-4 substrate. Some of this surface fuel contamination is removable by extended leaching with a fluoride decontaminant, but complete decontamination requires removal of the oxide (and possibly removal of a few microns of metal). Future analysis will attempt to determine how the fuel material is incorporated into the oxide crystal structure. Analyses of the base metal showed the major actinides that the computer analysis had predicted to arise from neutron activation or fission of impurity uranium in the Zircaloy-4 cladding. The computer prediction of the expected activation and fission products can be improved by adjusting the ORIGEN computation code to better approximate the flux in the Saxton reactor during this irradiation and the impurity concentration of the cladding alloy.

Preliminary Decontamination and Densification Process

We have reviewed and selected potential processes for decontamination and melting of chop-leach cladding residues. They are discussed in previous publications.⁽¹⁾ Briefly stated, the process selected for development consists of: 1) a gas phase treatment of cladding hulls by a fluoride to render the corrosion product oxide soluble in dilute aqueous fluoride solutions, 2) oxide removal and light etching in dilute aqueous fluoride solutions (nitric-hydrofluoric acid or oxalic-

hydrofluoric acid-hydrogen peroxide solutions), 3) melting of the hulls into an eutectic stainless steel-Zircaloy alloy by induction heating in graphite crucibles with or without a flux or solid adsorbent. If the metal is to be reused for reactor applications, a more conventional cold wall crucible with arc or induction melting of the segregated stainless steel, Inconel, and zirconium alloys would be required. The process steps requiring the most immediate attention were the fluoridation or zirconiumdioxide corrosion product film so that it may be removed in a standard aqueous process and a test of the feasibility of the alloy melting process.

Decontamination Process - B. Griggs

As previously described,⁽²⁾ treatment of zirconium dioxide corrosion films in HF gas at 550°C to 620°C produces films that are removable in dilute aqueous reagents. The characteristics of this reaction have been explored to aid in improving the process. Metallography of samples of oxidized and unoxidized Zircaloy after HF treatment has shown that HF reacts with metal through film imperfections. The oxide is subsequently undercut and removed by the aqueous process. Metallography of a sample showing this laterally progressing reaction under the oxide is shown in Figure 11. Reaction rate studies of HF and Zircaloy show an increasing rate of reaction with declining temperature in the 650°C to 500°C range (Figure 12). The fact that there is minimal reaction of oxidized zirconium with HF at 500°C shows that the

714X

714X

FIGURE 11. Autoclaved Zr-2 (47 mg/dm²) Reacted with Anhydrous HF at 600°C for 5 min. (Note apparent zirconium HF reaction product forcing zirconium oxide from metal surface.)

FIGURE 12. HF-Zr-2 Reaction
50% HF 50% Argon

550°C to 600°C temperature is apparently needed to produce film imperfection. Through these pores or cracks the HF can attack the metal and spread laterally to push the oxide layer off the metal. Microprobe studies of the metallographic sections of the reacted Zircaloy are being conducted to examine the role of fluoride in the process.

Fluoride containing gases such as boron trifluoride, carbontetrafluoride, hexafluoroethane, and difluorodichloromethane are alternative fluoridizing agents. They are of special interest because they do not produce water as a reaction product. The reaction of zirconium with water is an undesirable side reaction. Tests with CF_4 showed it to be nearly inert to Zircaloy and ZrO_2 at 550 to 650°C. Boron trifluoride reacted with Zircaloy, but not as uniformly as HF. Dichlorodifluoromethane produced severe reaction with the metal which was not limited to a surface coating. C_2F_6 has not been tested yet. Mixtures of HF and BF_3 were similar in effect to HF alone.

The performance of the aqueous decontaminating solution was evaluated as a function of its constituent concentrations. No significant improvements were found over the original function of 0.4 M ammonium oxalate, 0.16 M ammonium citrate, 0.1 M ammonium fluoride and 0.3 M hydrogen peroxide adjusted to pH 5 at 90 to 95°C.

An initial test of the hydrogen fluoride-aqueous strip process on a small piece of Saxton fuel cladding has been completed, but not all the analytical data have been obtained.

Briefly the 1.3-cm piece of Saxton Zircaloy-4 fuel cladding (from the same section used for transuranic element distribution studies) was treated in HF at 600°C for 30 min. This produced a weight gain of 886 mg/dm^2 . The piece was subsequently stripped in the ammonium oxalate, citrate, fluoride peroxide reagent for 1.2 hr at about 85°C, producing a weight loss of 3614 mg/dm^2 . The piece appeared to be almost completely stripped of oxide, leaving only a few islands on the surface (a few mm^2). The finished piece did not smear α contamination, and its gross radiation reading was reduced to a value similar to the completely decontaminated pieces used for analyses. Analyses of solution and piece will more firmly establish the degree of α decontamination. In general the piece stripped more completely than anticipated by work with autoclaved standing pieces, indicating a possible effect of radiation on film integrity. Additional tests on irradiated material will be made when suitable cladding can be obtained.

Densification Process - R. G. Nelson

Heat Number 9-12-74* a 90 wt% Zircaloy-2 - 6.7 wt% 304L stainless steel - 3.3 wt% Inconel 718 composition was poured at approximately 1415°C. The pour was mushy and the ingot consisted of blobs of metal fused together. Even at the $\sim 1415^\circ\text{C}$ pouring temperature, crucible reaction was severe. This coupled with the higher pouring temperatures required for good ingot practice makes this alloy a poor candidate for graphite melting.

Heat Number 9-18-74* an 80 wt% Zircaloy-2 - 13.4 wt% 304L stainless steel - 6.6 wt% Inconel 718 exhibited good fluidity and good pouring characteristics with a mild crucible reaction at a pouring temperature of 1315°C. The ingot, however, was full of cracks.

Heat Number 9-26-74* was an attempt to produce slow cooling of the 80 wt% Zircaloy-2 alloy by allowing it to cool in the crucible from 1300°C. This resulted in what appears to be a crack-free ingot; however, crucible reaction was severe, and the crucible had to be broken to remove the ingot.

Mold pre-heating with controlled cooling may be a requirement for crack-free ingots of these compositions.

Table 10 shows the chemical analyses of the major alloying elements of 5 of the 6 heats melted to date. All of the results agree reasonably well with the design compositions except for chromium in Heat Number 9-26-74.

Carbon, hydrogen, oxygen and nitrogen values for the melting stock and the resulting 5 heats are shown in Table 11. Carbon pick up from melting in the graphite crucible is minimal. The 250 ppm carbon level was the result of a high pouring temperature (1415°C). The melting of pure zirconium in graphite at temperatures of 1820 to 1920°C will result in carbon contents as high as 3000 ppm.⁽⁵⁾ Hydrogen and oxygen showed little or no increase after melting and nitrogen was slightly raised.

Rockwell C hardness values of the as-cast ingots range from 31 to 40.

Overall microprobe scans of specimens taken from center and edge ingot locations of Heat Number 9-5-74, 85 wt% Zircaloy-2 - 10 wt% stainless steel - 5 wt% Inconel 718 showed no difference in chemical compositions. Microprobe analyses of the three major microconstituents were performed on an as-polished specimen from this same heat and are shown in Figure 16.

TABLE 10. Chemical Analyses (wt%) of Major Alloying Elements

Element	Aim ^(a) Comp	Heat Number		Aim Comp	Heat Number 9-12-74	Aim Comp	Heat Number	
		8-29-74	9-5-74				9-18-74	9-26-74
Zr	83.52	84.8	84.9	88.43	86.5	78.61	78.8	78.2
Fe	7.91	8.15	8.15	5.34	5.92	10.54	10.33	10.72
Ni	3.57	3.59	3.64	2.39	2.64	4.72	4.76	4.64
Cr	2.94	2.80	2.83	1.99	2.06	3.88	3.76	2.82

a. The aim composition is calculated from the mid-range of the ASTM specifications of the raw materials used and the amounts used to make up the heats.

* Heat Numbers 9-12-74, 9-18-74 and 9-26-74 were melted and reported⁽²⁾ during the previous reporting period. Figures 13 through 15 are photographs of these ingots that were not available at that time. The melting results are repeated here for continuity.

TABLE 11. Chemical Analyses (ppm) of Interstitials

Heat No.	Composition of Ingots, ppm					Melt Stock Composition, ppm		
	85% Zr 2 Alloy		90% Zr 2 Alloy		80% Zr 2 Alloy	304L		Inconel 718
	8-29-74	9-5-74	9-12-74	9-18-74		Zircaloy 2	Stainless Steel	
C	100	160	250	160	90	100	420	430
H	24	14	8	60	80	22	80	17
O	1060	1010	980	980	870	1150	50	10
N	380	120	200	180	70	50	350	280
Rockwell C Hardness								
		38	31	40	30

FIGURE 13. 90 wt% Zircaloy-2 - 6.7 wt% 304L Stainless Steel - 3.3 wt% Inconel 718

FIGURE 14. 80 wt% Zircaloy-2 - 13.4 wt% 304L Stainless Steel -
6.6 wt% Inconel 718

FIGURE 15. 80 wt% Zircaloy-2 - 13.4 wt% 304L Stainless Steel -
6.6 wt% Inconel 718

FIGURE 16. Microprobe Analyses of As-Cast 85 wt% Zircaloy-2 - 10 wt% 304SS - 5 wt% Inconel 718

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4.0 PYROLYSIS-INCINERATION OF COMBUSTIBLE ALPHA WASTES

P. C. Walkup and L. K. Mudge - Project Managers

L. K. Mudge, R. A. Walter, and R. G. Sullivan - Principal Investigators

The principal program objective is to develop a pyrolysis-incineration process for safe and economical reduction of the volume of alpha wastes from 10- to 50-fold as a precursor to long term storage and/or disposal. The technical and economic merits of this process will be defined to allow its comparison to other processes being considered for volume reduction of alpha wastes. The program objective will be pursued via 1) a phased program of bench-scale research leading to supporting pilot plant development activities, and 2) systematic and continuing analytical studies to assess the technical and economic merits of the process. Bench-scale and pilot plant activities will be conducted first with noncontaminated simulated wastes and later with actual wastes from Hanford operations.

SUMMARY

The design of equipment for demonstration of the pyrolysis-incineration of simulated alpha wastes was completed during the reporting period. Purchase requisitions for the needed equipment were sent out. The equipment should be ready for operation in March 1975 as scheduled, barring unforeseen delays in procurement.

Ash from pyrolysis-incineration of simulated alpha wastes in laboratory studies was submitted for studies on conversion to a glass. Small glass samples are being prepared from the ash. Processes for volume reduction of alpha wastes now under development were reviewed to determine advantages of the different processes and possible process combinations.

TECHNICAL PROGRESS

Equipment for the pyrolysis-incineration of about 20 lb/hr of simulated combustible alpha wastes

was designed during the second quarter of FY75. All designs were approved and procurement of equipment components was begun. Assembly of the equipment is expected to proceed on schedule to allow operation of the equipment to evaluate air channeling through the bed of material in the reactor and to measure tar formation.

Figure 17 is an equipment flow schematic of the system being fabricated to meet the program objectives. All components are on hand with the exception of the reactor, which has been the object of the design effort.

The reactor is a refractory lined vertical cylinder with a working height of 6 ft and an inside diam of 11 in. Waste is distributed and packed into the reactor by a rabble arm capable of vertical and rotational motion. The stroke of the mechanism is 3 ft, and rotational speeds are from 0 to 5 rpm. Maximum

FIGURE 17. Schematic of Pyrolysis-Incineration Demonstration Unit

compaction pressure is 50 psi. Simultaneous vertical and rotational movements will be used to prevent channeling of air through the bed of refuse. Ash is removed from the reactor into a star valve feeder through a rotary grate which rotates at speeds from 0.125 to 1.875 rpm and exerts a maximum torque of 155 ft lbs.

Waste is fed into the reactor through a lock hopper equipped with a 9 in. diam, 9 in. pitch screw conveyor. The screw is driven with a variable speed hydraulic motor. The

volume of the lock hopper is approximately the same as the internal capacity of the reactor.

The reactor is instrumented to monitor temperatures, pressures, and flows of all streams and internal operating temperatures and pressures. Use of the rabble arm distributor will allow determination of the operating level in the reactor.

Tars and other liquid wastes will be removed in a caustic scrubber to determine quantity and character. Evaluation of the tar formation prob-

tem and the channeling problem will be completed by the end of FY75.

Studies on process combinations and ash fixation started during the current reporting period. Glass samples being prepared from ash generated during laboratory studies on pyrolysis-incineration should be available early in the third quarter of FY75. Processes now being considered for volume reduction of alpha waste were reviewed to determine the characteristics of the residues from the processes. Very little information was found on residue characteristics.

Review of alpha waste processes was started to determine advantages that might accrue from use of process combinations either in parallel or in series. It may be advantageous to use two processes each capable of handling a different type of waste. Also, use of a molten salt to destroy tars formed by pyrolysis-incineration may be advantageous since tars are low in ash content and ash is difficult to remove from molten salts. Work on the study of process combinations is continuing.

5.0 TRITIUM SEPARATION AND FIXATION

L. L. Burger - Project Manager

The goals of the program are to investigate isotopic separation as a means of concentrating tritiated aqueous wastes, to consider alternative procedures such as increased recycle of water, and to examine methods of fixing tritiated water for permanent storage.

SUMMARY

Two types of tritiated organic polymers have been synthesized and are being tested. One is a resorcinol-acetaldehyde polymer, the other, poly(acrylonitrile). Although both polymers appear promising, the latter has more restrictions in the synthesis. Leach tests in water are under way to examine the materials for tritium loss. Other polymers are being considered.

Experiments are being performed to attempt selective dissociation of HDO molecules through a combined laser excitation to the 110 level followed by a flash lamp excitation to the unstable electronic state. Calculations of the concentrations of various free radical and molecular species following flash dissociation have been made. No D_2 has been found in the first experiments possibly because of reaction with oxygen present.

TECHNICAL PROGRESS

Tritium Fixation -

E. C. Martin, J. A. Franz, and D. C. Elliott

In a previous Quarterly Progress Report⁽¹⁾ we described the development of a resorcinol-acetaldehyde-formaldehyde condensation polymer for the incorporation of tritium-containing water. Tritium has since been incorporated in the polymer in the form of tritiated acetaldehyde, which can be prepared on an industrial scale by the mercury-catalyzed hydration of acetylene⁽²⁾ in a tritium-containing aqueous medium.

Tritiated acetaldehyde was more conveniently prepared on a laboratory scale by the acid-catalyzed hydrolysis of ethyl vinyl ether in a tritium-containing medium. Material prepared by this route

was identical (nmr) and IR) to an authentic sample of acetaldehyde. When tritiated acetaldehyde was heated with resorcinol and finally with formaldehyde, an insoluble, crystalline polymer resulted. In this manner tritium was incorporated at sites where the tritium is inert to exchange with water, and, when combined with the insolubility of the polymer itself, eliminates the loss of tritium into the environment during storage.

A sample of about 10 g of the polymer exhibiting a total count of 5×10^4 disintegrations/min was washed with several ml of water. The water gave a total count of 750 disintegrations/min, due to surface contamination of the polymer with monomer. After an additional 4 weeks of exposure to the polymer by the water sample, no further increase in the tritium content of the wash solution was detectable. We conclude that the polymer successfully fixes tritium.

The search for inexpensive polymers with suitable properties for the fixation of tritium led us to consider poly(acrylonitrile). Acrylonitrile ($\text{CH}_2 = \text{CH} - \text{C} \equiv \text{N}$) is prepared industrially by the copper-catalyzed addition of hydrogen cyanide to acetylene.⁽³⁾ If the reagents undergo reaction in a catalyst mixture containing THO, tritiated acrylonitrile results.

Polymerization of tritiated acrylonitrile provides a polymer inert to hydrogen exchange with the environment.

The tritium level in the catalyst mixture for the condensation of HCN and acetylene can be maintained by the introduction of TCN, prepared as indicated in the following equations using sulfur trioxide, THO and KCN.

Acrylonitrile and tritiated acrylonitrile were prepared in this laboratory to evaluate synthetic procedures and properties of the labeled polymer by a procedure similar to that of P. Kurtz.⁽⁴⁾ Hydrogen cyanide was prepared according to a standard procedure.⁽⁵⁾ Commercial acetylene was purified by passage through concentrated H_2SO_4 and a cold trap ($\sim 78^\circ\text{C}$). Purified acetylene was introduced at 0.6 liter/min into a rapidly stirring catalyst mixture maintained at 85° , consisting of 500 ml of tritiated water ($10.4 \mu\text{Ci/ml}$), 300 g of CuCl , 100 g of NH_4Cl , 10 ml of concentrated HCl , and 10 g of Cu powder. Hydrogen cyanide (40-60 g) was introduced over a period of approximately 6 hrs. An organic phase (40 g) and an aqueous phase were collected by passage of

the excess acetylene stream through a vertical condenser and a receiver maintained at -78°C . The organic phase was distilled; the portion boiling from 72 to 77°C was found to be identical by nmr and IR to an authentic sample of acrylonitrile, and exhibited 1.28×10^4 disintegrations/min/g. In general, yields of acrylonitrile prepared in this fashion were comparable to those achieved by Kurtz.⁽⁴⁾ The acrylonitrile prepared in this manner contains trace quantities of divinylacetylene, which significantly reduces the molecular weight of the poly(acrylonitrile) prepared from it.⁽³⁾

Unlabeled poly(acrylonitrile) was prepared by emulsion polymerization.⁽⁶⁾ A sample of unlabeled acrylonitrile was polymerized in quantitative yield by treatment under nitrogen of a stirring emulsion of 80 g of acrylonitrile, 120 g of water, and 2 g of sodium lauryl sulfate with potassium persulfate (0.1 g) and sodium sulfite (0.04 g). The polymer, an insoluble white powder of high molecular weight, precipitated after 20 min.

When a sample of the tritiated acrylonitrile prepared as above was used, larger quantities of persulfate initiator (1 g) were necessary to induce polymerization. The yield of crystalline polymer was approximately 40% based on unrecovered acrylonitrile; the remaining 60% consisted of low molecular weight noncrystalline polymer. It is apparent that purification procedures to remove trace impurities in industrial preparation of polymer-grade acrylonitrile⁽³⁾ cannot be compromised for use in a waste-disposal scheme.

Leach tests of tritiated poly (acrylonitrile) are under way. The polymer is insoluble in water and is not expected to show loss of tritium on exposure to water.

Poly(acrylonitrile) has suitable properties for tritium storage. But the necessity of an added step to replenish tritium in the catalyst mixture, the necessity of using several starting materials in the preparation of the monomer, and the relatively high purity of the monomer required to achieve a crystalline polymer may restrict the procedure to disposal of highly concentrated waste tritium. A final evaluation must await results of leach tests and the determination of minimum purification procedures.

Efforts are under way to evaluate poly(vinylacetate) and poly(vinyl ether) for tritium fixation.

Molecular Excitation Experiments -

T. J. Whitaker, R. D. Scheele, and J. R. Morrey

We have described previously several schemes for separating deuterium from a $\text{HDO}/\text{H}_2\text{O}$ mixture. In present experiments we are using a tunable infrared laser to selectively excite HDO molecules. The laser system consists of a Chromatix Nd:YAG laser with tuning accomplished via an optical parametric oscillator. Spectra of HDO vapor at approximately 100°C have been taken using this laser system, and the $000 \rightarrow 110$ vibrational transition has been identified. Selective dissociation of HDO is accomplished by holding the frequency of the laser system at this frequency while simultaneously firing a Xenon flash-lamp system to pump the excited HDO molecules to an unstable elec-

tronic state. The intensity of the flash-lamps as a function of frequency is important since a high intensity at the frequency required to pump the vibrationally excited HDO molecules to the dissociation band is desirable, but low intensity at the frequency required to pump ground state H_2O molecules to the dissociation is needed if the process is to be selective. Work has been continuing to test certain rare earth aqueous solutions for their usefulness as filters. Depending on the success of H_2O filtered flash-lamps, these may be incorporated into the system.

A computer study of the kinetics of free radicals generated by the dissociation of HDO has been completed. The study indicates that in an environment of H_2O , HDO, and D_2O , deuterium from dissociated HDO molecules would appear mostly in the form of HD and D_2 . These species lend themselves easily to analysis by a mass spectrometer. Initial experiments to separate deuterium have not shown any D_2 . It is believed that O_2 in the cell containing HDO caused the formation of peroxides instead of HD and D_2 . This is supported by the kinetics study made. By filling a cell with HDO vapor in a vacuum oven purged with N_2 this problem should be avoided. Results using this method are not yet available.

It should be noted that workers at Los Alamos⁽⁷⁾ have attempted a very similar experiment. Using a DF laser, they have excited HDO molecules to the first vibrationally excited state. Since the ground state of H_2O is approximately $1,245\text{ cm}^{-1}$ (8) higher in

energy than the ground state of HDO, the energy difference between the unexcited H_2O and the excited HDO molecules in their experiment was insufficient to allow efficient selective pumping of the HDO to an unstable state. However, by pumping the HDO molecules to the 110 state, an energy difference of about $2,900\text{ cm}^{-1}$ is produced. This is believed to be a wide enough energy gap to allow selective pumping of the HDO. If experiments prove otherwise, there are higher energy HDO transitions which may be used so that the second excitation would require even less energy. These are not being used presently because of the lower cross-section at these frequencies.

If the current experiments prove encouraging, work will begin on using this technique to separate tritium from $\text{HTO}/\text{H}_2\text{O}$ mixtures.

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6.0 CHARACTERIZATION OF 300 AREA AND WYE BURIAL GROUNDS

J. R. Eliason - Project Manager

This study is designed to evaluate and define the present status of waste materials disposed to the 300 Area and Wye burial grounds and to evaluate the alternatives of designating the sites for permanent storage and/or removal of all or part of the materials.

SUMMARY

An ecological evaluation of 300 and Wye burial grounds is continuing. This evaluation has delineated uptake of various chemical constituents by specific plant species.

Magnetometer surveys have been conducted in the 300 North burial ground. Results from this study have demonstrated applicability of magnetic surveys in burial ground trenches. Design and testing of acoustic systems used in subsurface exploration of burial grounds is continuing.

Laboratory calibration and testing of psychrometric systems is nearly complete. Thermocouple psychrometers and temperature sensors will be installed during drilling activities at various locations in and around burial grounds.

A review and analysis of existing historical records was completed. A preliminary document containing inventory analysis data has been distributed to task investigators.

TECHNICAL PROGRESS

Evaluation of Biological Transport Mechanisms - W. H. Rickard

Ecological characterization of the 300 and Wye Area burial grounds began in autumn of CY-1974. The season was appropriate to observe growth performance of rabbitbrush, Russian thistle, and bursage as autumn is their normal season of flowering and maturation. However, most plants growing there are spring flowering, and observation of species composition and abundance will need to be made during the spring, April and early May, of CY-1975.

Chemical analyses were made on tissue of whole plants collected from burial grounds and plants from native habitats. The results are shown in Tables 12-15. Russian thistle rapidly accumulated potassium and calcium; bursage accumulated calcium (Table 15). Rabbitbrush was not as efficient at accumulating potassium and calcium as were Russian thistle and bursage. Russian thistle is a known accumulator of ^{90}Sr and ^{137}Cs

TABLE 12. Mineral Composition of Russian Thistle Growing in Waste Burial Grounds in the 300 and Wye Areas

	Burial Ground Identification						Avg
	2-3	4	7	300N	WYE	Control	
% Ash	10.5	10.4	11.1	11.5	10.8	11.8	11.0
% P	0.20	0.23	0.34	0.18	0.24	0.24	0.24
% K	3.0	2.0	3.3	3.7	3.4	3.4	3.1
% Mg	1.2	1.0	0.41	0.44	0.70	0.75	0.74
% Ca	1.4	1.0	1.5	1.5	1.7	1.6	1.5
ppm Ba	20	62	207	35	42	32	66
ppm B	1.8	18	44	34	29	25	25
ppm Cu	6	12	12	9	10	9	10
ppm Zn	9	21	26	33	21	13	20
ppm Mo	1.2	0.07	2.0	0.28	0.10	0.04	0.62

TABLE 13. Mineral Composition of Bursage Growing in Burial Grounds in the 300 and Wye Burial Areas

	Burial Ground Identification					Avg
	4	2-3	7	Wye	Control	
% Ash	9.7	15.4	10.6	10.4	11.2	11.5
% P	0.38	0.26	0.23	0.22	0.30	0.28
% I	1.5	1.7	1.8	1.6	1.7	1.7
% Mg	0.25	1.0	0.36	0.38	0.35	0.47
% Cu	1.6	2.2	2.1	1.9	2.0	1.6
ppm Ba	217	100	30	50	57	91
ppm B	50	124	58	28	57	63
ppm Cu	13	11	9	12	13	12
ppm Zn	38	17	18	75	83	46
ppm Mo	0.58	1.2	2.0	0.6	0.4	0.96

TABLE 14. Mineral Composition of Rabbitbrush Growing in 300 and Wye Area Burial Grounds

	Burial Ground Identification			Avg
	7	4	Control	
% Ash	4.5	4.4	4.6	4.5
% P	0.16	0.15	0.17	0.16
% K	1.0	0.92	1.1	1.0
% Mg	0.20	0.15	0.25	0.20
% Ca	0.67	0.58	0.84	0.70
ppm Ba	42	37	25	35
ppm B	65	37	40	42
ppm Cu	6	6	8	7
ppm Zn	21	21	15	19
ppm Mo	0.20	0.15	0.24	0.20

TABLE 15. Summary of Mineral Composition of Rabbitbrush, Russian Thistle, and Bursage Growing in the 300 and Wye Burial Grounds

	Russian Thistle	Bursage	Rabbitbrush
% Ash	11	11.5	4.5
% P	0.24	0.28	0.16
% K	3.1	1.7	1.0
% Mg	0.74	0.47	0.20
% Cu	1.5	1.6	0.70
ppm Ba	66	91	35
ppm B	25	63	42
ppm Cu	10	12	7
ppm Zn	20	46	19
ppm Mo	0.62	0.96	0.20

and also has the noxious habit of mature plants breaking off at ground level and being carried by wind. Russian thistle plants accumulate and grow in large numbers in depressions in the ground surface protected from wind action.

The mineral content of burial ground species was not very different from "control" plants not growing on burial grounds, except for barium in bursage from burial ground 4 (Table 13) and Russian thistle in burial ground 7 (Table 12).

Burial ground surfaces appear to provide poor habitat for small burrowing mammals but at least some reside in burial grounds, judging from tracks seen in the dust. To confirm whether or not appreciable populations of small mammals reside in the burial grounds, a grid of traps was set up in the Wye burial ground. The program will consist of a live trap-mark-release and recapture experimental design. It should yield information on population density, activity areas, weights of individual animals, sex, age, reproductive condition and general health, time of summer torpor of adults and the onset of winter torpor. This information would be compared with a similar trap grid set up in a nonburial ground habitat nearby. Data from this study should begin to accumulate with the emergence of pocket mice in March.

Waste Detection and Mapping - G. A. Sandness

Recent and current work in this task has been directed toward identification of useful and effective

techniques for detecting and mapping buried wastes.

Test surveys with a proton precision magnetometer have been conducted in the 300-N burial ground. This method is potentially useful for detecting buried ferrous materials such as oil drums and other steel containers, vehicles, tools, and miscellaneous steel scrap. The results are difficult to interpret in areas of large concentrations of steel objects. The method may be useful, however, in preliminary surveys of trench areas where concentration of steel objects is not too high.

A study has determined the availability of metal detectors for both ferrous and nonferrous metal objects. An instrument has been obtained and is being used in the field.

An additional study is being conducted to design an acoustic system for detecting objects and trench boundaries in the alluvial sediments in the burial grounds. Recent work has included maintenance on an existing seismic exploration system to be used for conducting preliminary measurements of the acoustic properties in sedimentary material.

Instrument Wells and Characterization of Sediments - S. J. Phillips

To determine quantitative information on migration of meteoric waters and contaminated radionuclide waste fluids through the vadose zone in burial ground facilities, various analytical detection systems have been identified. Procurement and fabrication of instrument sensors and monitoring apparatus is nearly com-

plete. Laboratory calibration and testing of thermocouple psychrometers used in determining water potential in Hanford sediments is proceeding. Thermocouple psychrometers are being tested for stability, equilibration time, and failure characteristics.

Computer data analysis techniques have been developed to determine partially saturated flow characteristics in sediments. Computer programs are used for calculation and display of capillary pressure, relative hydraulic conductivity, fluid retentivity, and fluid content variables.

Review of Historical Records -

A. E. Reisenauer

A review of existing records concerning burial ground inventories is complete. A draft document containing information involving waste material type, origin, quantity, location of interment, date of shipment, containment structure, and method of burial, derived from historical record review analysis, has been distributed to task investigators. Engineering drawings of burial ground facilities have been procured and copies distributed to task investigators. In addition, included in the draft document are photographs of existing burial ground facilities, illustrating negative cover, enclosures, etc.

7.0 DISPOSITION (D&D) OF RETIRED CONTAMINATED FACILITIES AT HANFORD

K. M. Harmon - Project Manager

The program has the following objectives for FY 1975-1976: to establish methods, costs and priorities for the disposition of retired (excessed) Hanford facilities which will be transferred to WMT; to formulate plans and specifications for a project to accomplish permanent disposition of a specific Hanford facility; and to identify required facility disposition R&D.

SUMMARY

Published information on retired liquid radioactive waste disposal sites in the 200 Areas has been tabulated for inclusion in the Appendices of the Resource Book, and the Atlantic Richfield Hanford Company has supplied a list of ten cribs as candidates for demonstration of disposition methods. Simple soil characterization studies showed that there is little chance that crib soils can be decontaminated adequately by simple washing of fines from the coarse soil particles.

Assembly of the disposition information center has continued; the 550 documents received to date have been catalogued and indexed using a computer printout. The areas where additional R&D is needed were identified and reported in a letter to the AEC.

The Plutonium Concentration Building (233-S) of the Redox Plant was recommended as the facility for the first D&D demonstration project. Work to estimate the plutonium content of the process hood was initiated, alternative disposition strategies were identified, and preparation

of procedures, preliminary cost estimates and risk evaluations were started.

TECHNICAL PROGRESS

Task 1 - Develop Methods, Costs and Priorities of D&D at Hanford

Subtask 2 - Site Characterization - B. W. Mercer

Published information on retired liquid radioactive waste disposal sites in the 200 Areas has been tabulated for inclusion in the appendices of the Resource Book currently being prepared for the program. The disposal sites consist of 133 cribs, three ponds, and one ditch. The information tabulated for each site includes 1) the facility name and number designation, 2) location and period of service, 3) function, 4) physical description, 5) radionuclide content, 6) incentives for disposition, 7) current site characterization status, 8) additional work required to complete characterization, 9) disposition alternatives, and 10) disposition history. It is anticipated that future site characterization activities will consist of a

limited well drilling program coupled with computer modeling techniques to confirm the reported radionuclide content of each site and determine the distribution of radionuclides in the soil beneath the disposal sites.

At the request of Battelle, Pacific Northwest Laboratories, the Atlantic Richfield Hanford Company has supplied a list of ten radioactive liquid disposal cribs in the 200 Areas as candidates for demonstration of disposition methods. Reference drawings were made available along with information concerning disposal history, crib construction, radionuclide inventory, and chemical composition of waste discharged. The candidates were selected to provide a variety of different types of cribs and a range in quantity and type of radionuclides disposed.

A preliminary study is being conducted to determine the feasibility of decontaminating radioactive soil by size fractionation. A sieve analysis of soil samples was performed on soil removed from two experimental columns which had received B-Plant process condensate. Samples of each size fraction of the contaminated soil were counted for ^{137}Cs activity to evaluate the effectiveness of sieving as a means of reducing the cesium content of the bulk of the soil. The ^{137}Cs content of each particle size range is illustrated in Figure 18.

As expected, the finest size fraction, that with the sediment and clay phases (0.03 mm mean particle size), contained the highest concentration of ^{137}Cs . The difference in activity levels between the fine and coarse

FIGURE 18. Concentration of ^{137}Cs in Soil as Related to Particle Size

size fractions is, however, not sufficient to effect significant removal of ^{137}Cs activity by simple washing of the fines from the coarse material. Eighty-three to 88% of the soil consists of sand in the range of 0.125 to 2 mm (see Table 16). The fraction of soil finer than 0.125 mm contains only 30% of the total activity.

TABLE 16. Sieve Analysis of Soils

Particle Size Range, mm	Percent Soil in Each Size Range	
	Column 1	Column 2
<0.062	6	12
0.062 - 0.125	6	5
0.125 - 0.250	11	9
0.25 - 0.50	19	17
0.50 - 0.84	26	23
0.84 - 2.00	32	34

Characterizing retired solid waste burial sites, process buildings, and support facilities in the 200 Areas has started. The task of collecting historical information concerning process buildings and support facilities has been assigned to personnel of the Atlantic Richfield Hanford Company.

Subtask 3 - Disposition -

M. R. Kreiter

Potential ultimate uses were reviewed for a variety of retired Hanford sites under consideration for the first D&D demonstration project. The general consensus of the ten reviewers was that the most probable ultimate use for any 200 Area sites and for the 300-Wye Burial Ground area was as a nuclear industrial park; and for the 100 Area sites, as a navigation/port facility or as a nuclear or non-nuclear industrial park.

A list of disposition strategies for the retired production reactor facilities was developed in consultation with UNI personnel. The alternatives include:

- Leave the facility as is and continue surveillance.
- Construct a fence around the facility and entomb it.
- Decontaminate the facility and leave it standing as a shell.
- Dismantle the facility and store the removed equipment and material in a retrievable manner pending future decisions as to ultimate disposal.

Subtask 5 - Procedures and Techniques -

H. T. Fullam, R. F. Maness,
C. H. Allen, and W. Sande

Work continues on assembling the information center on the disposition of nuclear facilities. Approximately 550 documents, reports, and papers have been obtained and catalogued. An additional 100 or so documents have been ordered but not yet received. The documents have been catalogued and indexed using a computer printout. Entries are indexed by report number, author, date, and key words. The printout will be updated on a semi-annual basis as additional references are obtained. Copies of the printout are available to Hanford personnel as needed.

Work is under way on preparation of an annotated bibliography of pertinent references. When completed, the bibliography will be issued as a BNWL report.

After a preliminary review of the literature and discussions with various individuals, the areas where additional R&D is needed to provide improved procedures for the disposition of nuclear facilities have been identified. The problem areas include:

1. Soil decontamination, especially the removal of actinides.
2. Concrete decontamination, removal of the contamination so that the bulk of the concrete can be disposed of in uncontrolled fashion. Also applies to other structural materials.

3. Dry decontamination procedures for use in areas where liquid wastes can not be handled.
4. Size reduction and volume reduction. Improved methods are needed for dismantling and segmenting large equipment, particularly remote methods. Methods for reducing the bulk volume of contaminated equipment, prior to storage, are also needed.

In addition to these specific problem areas, improved aqueous methods are needed for decontaminating all types of materials to uncontrolled release levels.

Task 2 - Formulate Plans and Specifications for a D&D Project

Subtask 1 - Planning and Evaluation - K. M. Harmon

The six candidate facilities for the first D&D demonstration project were compared in terms of 1) the incentives for their disposition in the near future, 2) what could be learned from each project, 3) the complexity and cost, and 4) the feasibility of having project planning completed early in FY76. The 233-S (Redox Plutonium Concentration Building) was selected for the following reasons:

1. It is highly contaminated with plutonium. Shutdown of the building ventilation system could allow contamination spread to the outside. Hence this facility, although within the 200 West Area fence, requires constant attention and will represent a potential hazard until something is done with it.

2. With adequate planning, D&D of 233-S could be used to test and demonstrate a variety of new techniques for disposition of plutonium-contaminated facilities.
3. Project cost and schedule appear reasonable.

Subtask 2 - Site Characterization - B. W. Mercer

Preliminary measurements of the thermal neutron flux were made at various points around the 233-S process area to establish the approximate location of the bulk of that area's residual plutonium. The neutron flux was measured using copper and copper-cadmium discs which were exposed for 24 hr in the 233-S Building, then counted for radioactivity induced by thermal neutron capture. The highest neutron flux was found at the face of the first level viewing area. The flux values obtained were relatively low (0.08 to 0.21 n/cm²/sec) and tend to confirm the prevalent belief that much of the residual plutonium is located on the floor of the process hood and in or around the lower portions of the process equipment. Additional neutron flux measurements are planned to more accurately define the source of the neutrons and determine the need to quantify the amount of residual plutonium in the process hood.

Subtask 3 - Disposition - M. R. Kreiter

This list of disposition strategies for the 233-S Building was developed as a result of meetings with BNW and ARHCO personnel:

- Leave the facility in its present status with continued surveillance.
- Enclose (cocoon) the building after isolating the building service and processing connections to the Redox Building.
- Entomb the facility using the existing building as the form.
- Decontaminate the facility, remove the processing equipment and leave the walls as a standing shell (can be carried out to various degrees).
- Dismantle the facility and store the removed material and equipment

in a retrievable manner pending future decisions as to ultimate disposal of waste materials.

The procedures required for each of the disposition alternatives for the 233-S Building have been defined in general terms. Work is now under way to define the specific methods and schedules required for the two most promising disposition alternatives: 1) decontaminate the facility with removal of all equipment, and 2) complete dismantling of the facility.

8.0 DEVELOPMENT OF PACKAGING TECHNOLOGY FOR $\text{Pu}(\text{NO}_3)_4$ SHIPMENT

H. H. Van Tuyl - Project Manager

The USAEC has announced that the shipment of $\text{Pu}(\text{NO}_3)_4$ solution will be prohibited as of June 17, 1978. After that date, the only solid Pu form that can be shipped to fuel fabricators as a plutonium fuel material is PuO_2 .

The objectives of this program are:

1. *To determine if a solid form of plutonium nitrate can be found that is reasonably stable and directly soluble in nitric acid;*
2. *To define new packaging technology for the shipment solid plutonium form; and*
3. *Based on the success of (1 and 2) to define a commercially feasible process to make the solid plutonium form.*

If plutonium nitrate in powder form can be stored and shipped safely, made economically, and is readily soluble in nitric acid, then plutonium nitrate powder is likely to be superior to PuO_2 for shipment to plutonium fuel fabricators.

The compounds investigated are $\text{Pu}(\text{NO}_3)_4 \cdot x\text{H}_2\text{O}$, $\text{K}_2\text{Pu}(\text{NO}_3)_6$, and $(\text{NH}_4)_2\text{Pu}(\text{NO}_3)_6$.

SOLID PLUTONIUM SHIPPING FORMS -

G. H. Bryan, Principal Investigator
SUMMARY

The compound $\text{Pu}(\text{NO}_3)_4 \cdot x\text{H}_2\text{O}$ has been selected for further evaluation as a shipping form from the three compounds-- $\text{Pu}(\text{NO}_3)_4 \cdot x\text{H}_2\text{O}$, $(\text{NH}_4)_2\text{Pu}(\text{NO}_3)_6$, $\text{K}_2\text{Pu}(\text{NO}_3)_6$ --originally under consideration. The compound $(\text{NH}_4)_2\text{Pu}(\text{NO}_3)_6$ was eliminated due to excess gas evolution, and the potassium salt ($\text{K}_2\text{Pu}(\text{NO}_3)_6$) due to waste disposal problems that would be created because of the potassium. Pilot plant scale studies are determining optimum parameters for production of $\text{Pu}(\text{NO}_3)_4 \cdot x\text{H}_2\text{O}$.

Results of radiolytic gas production from a 17% ^{238}Pu sample indicate the gases produced are the same as formed using 85% ^{238}Pu , with oxygen being the single largest component. G_T values (molecules per 100 ev) varied somewhat between the two samples.

TECHNICAL PROGRESS

Radiolytic gas evolution identification and determination of G_T values (molecules per 100 ev) on a 17% ^{238}Pu sample are complete. G_T values for the 17% ^{238}Pu sample were found to be very similar to those found using 85% ^{238}Pu , with some variations noted during the early stages of the experi-

ment. An average G_T value of about 0.16 was found with the 85% ^{238}Pu material during the first 5×10^{17} Mev/gm Pu alpha radiation dose, whereas with the 17% ^{238}Pu the average G_T value was about 0.07 over this range. Both samples were found to approach a G_T value of about 0.02 after the initial higher values (see Figure 19).

FIGURE 19. G_T Values as a Function of Accumulated Dose

The gases evolved in each sample were about the same, with oxygen the predominant species. Hydrogen evolution in each case was less than 3% of the total gases evolved (see Table 17).

TABLE 17. Composition of Radiolytic Gases(a) Evolved from $\text{Pu}(\text{NO}_3)_4 \cdot \text{XH}_2\text{O}$

	17% ^{238}Pu	85% ^{238}Pu
$\text{O}_2\%$	71	86
$\text{N}_2\%$	13	8
$\text{H}_2\%$	2.4	0.4
$\text{NO}_x\%$	4	4
$\text{N}_2\text{O}\%$	6	- - -
$\text{CO}_2\%$	4	3

a. Dose rate of 1×10^{17} Mev/g Pu

The production of $\text{Pu}(\text{NO}_3)_4 \cdot \text{XH}_2\text{O}$ is presently under study, with parameters being determined for optimum production. Rates of evaporation have been determined during a simulated pilot plant run at 80°C on a 231.5 g/l plutonium solution at 5 mm Hg (see Figure 20). The resultant solid was completely soluble in 4M HNO_3 . Production of the intermediate compound detected during thermogravimetric analysis of $\text{Pu}(\text{NO}_3)_4 \cdot 3\text{H}_2\text{O}$ was attempted by heating of the compound produced by the above method to 150°C . The resultant solid was found to be insoluble in nitric acid.

FIGURE 20. Percent of Original Sample Weight Loss Versus Time

9.0 TRANSPORTATION SAFETY STUDIES

L. D. Williams - Project Manager

The objective of the transportation safety study program is to develop and use a model for calculating the risks associated with the shipping of radioactive materials.

SUMMARY

The final draft of BNWL-1846, The Risk of Transporting Plutonium Oxide and Liquid Plutonium Nitrate by Truck, was completed and transmitted to the USAEC for comment.

The computer code HEAD, used for the risk calculations, has been modified to increase its area of applicability.

Work toward assessing the risks of UF_6 and non-high-level waste shipments has started. System descriptions and fault trees for the shipments have been developed.

TECHNICAL PROGRESS

Risk Assessment for Plutonium Shipments - T. I. McSweeney

The final draft of BNWL-1846, The Risk of Transporting Plutonium Oxide and Liquid Plutonium Nitrate by Truck, was completed and transmitted to the AEC Division of Waste Management and Transportation for review and comment. The report describes the methodology developed to assess the risk, presents the calculated risk for the two plutonium shipment forms, and compares them to other risks to which society is exposed.

Risk Model Development - D. K. Davis

The computer code HEAD, which performs the risk assessment calculations, was originally formulated specifically for plutonium shipment analysis. The code has been further developed to increase its generality. Given the proper input data, air pathway risk calculations can now be made for any radioactive material. In addition, modifications to clarify the presentation of results were made in the code output. The next code modification will be to incorporate chemical toxicity risks into the analysis.

Risk Assessment for UF_6 Shipments - R. J. Hall

Two UF_6 shipping vessels are being considered in the analyses. One is a 2 1/2-ton container and the other is a 14-ton container. The tonnage refers to the maximum UF_6 weight which may be transported in the vessel. The 2 1/2-ton UF_6 vessel is nominally 30 in. diam by 81 in. long. The 14-ton vessel is nominally 48 in. diam by 150 in. long. The 2 1/2-ton vessel is used for shipment of up to 5% ^{235}U enriched uranium. The maximum enrichment for the 14-ton vessel is 4.5%. For shipment of greater

than 1% enriched uranium a protective outer packaging is required on the cylinder.

At present only truck transport of UF_6 is being analyzed. Applicable shipping condition assumptions from the plutonium analysis (e.g., year of shipment, projected population density) are being used in the UF_6 analysis. Fault trees showing the relationship of conditions and events necessary to release UF_6 during transport have been constructed. Effort is being directed toward determination of the information available to describe the UF_6 shipment accident environment. The detail and accuracy of the risk assessment is a strong function of the knowledge of the accident environment.

Work is progressing toward evaluation of the consequences of a UF_6 release. UF_6 readily reacts with water to form hydrofluoric acid and UO_2F_2 . Even in an atmospheric release, there will be interaction of the UF_6 with the moisture in the air forming the toxic acid. Thus a portion of the consequences of a UF_6 release will be chemical effects not related to the radioactive characteristics of the material.

Risk Assessment of the Non-High-Level Waste Shipments - R. J. Hall

Non-high-level wastes, which consist of diverse solids (e.g., paper, plastics, glass, rubber, metal) generated in various stages of the fuel cycle, are shipped in 55-gal. drums. For this study the waste is divided into three categories: alpha waste, alpha-beta-gamma waste, and beta-gamma waste. Alpha waste and alpha-beta-gamma waste are packaged in a rigid polyethylene liner which fits inside the 55-gal. drum. Alpha waste contains actinide elements in concentrations greater than $10 \mu\text{Ci/kg}$ but has sufficiently low external radiation levels (surface dose rate $< 10 \text{ mrem/hr}$) that the drum can be handled without shielding. Alpha-beta-gamma waste is the same as alpha waste except that it has a gamma radiation level high enough (10 to 1000 mrem/hr) to require radiation shielding. An overpack is used on the drum to provide shielding during transportation. Beta-gamma waste, which contains less than $10 \mu\text{Ci/kg}$ actinide elements, is shipped in a 55-gal. drum without a polyethylene liner.

Fault trees showing ways that waste could be released during truck transport have been developed and are currently undergoing internal review.

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