2F

Nuclear Waste Management and Transportation

Quarterly Progress Report
January through March 1975

June 1975

Prepared for the U.S. Energy
Research and Development Administration
under Contract AT(45-1):1830

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
U.S. ATOMIC ENERGY COMMISSION
Under Contract AT(45-1)-1830

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy \$7.60; Microfiche \$2.25

NUCLEAR WASTE MANAGEMENT AND
TRANSPORTATION QUARTERLY PROGRESS
REPORT JANUARY THROUGH MARCH 1975

Compiled by

A. M. Platt
Nuclear Waste Technology Program

June 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.

Battelle
Pacific Northwest Laboratories
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 October through December 1974, BNWL-1899.

CONTENTS

FOREWORD	iii
LIST OF FIGURES	vii
LIST OF TABLES	viii
1.0 ADVANCED METHODS FOR MANAGEMENT AND DISPOSAL OF RADIOACTIVE WASTES .	1
TASK 1 - DEVELOPMENT OF ADVANCED WASTE MANAGEMENT PLANS	1
TASK 2 - DEVELOPMENT OF EVALUATION METHODOLOGY	5
RADIONUCLIDE MIGRATION FROM NONSALT FORMATIONS	5
RADIONUCLIDE MIGRATION FROM SALT FORMATIONS	6
PUBLIC ATTITUDES TOWARD NUCLEAR WASTE MANAGEMENT	8
TASK 3 - SPECIAL EVALUATIONS	8
WASTE MANAGEMENT FOR ^{129}I AND ^{99}Tc	9
LOSS OF HIGH-LEVEL WASTE AT SEA	9
IMPROVEMENT AND DOSE MODELS	9
2.0 SEPARATION OF ACTINIDE ELEMENTS FROM RADIOACTIVE WASTE	13
TASK 1 - CHARACTERIZATION AND TREATMENT OF SOLIDS IN HIGH-LEVEL WASTES	14
TASK 2 - REMOVAL OF LONG-LIVED NUCLIDES FROM SOLIDIFIED WASTE	14
TASK 3 - REMOVAL OF LONG-LIVED NUCLIDES FROM NON-HIGH-LEVEL WASTES .	15
TASK 4 - DEVELOPMENT OF LOW-ADDITIVE FLOWSHEETS FOR REMOVING LONG-LIVED NUCLIDES FROM THE FISSION PRODUCTS	15
TASK 5 - EVALUATION OF INCENTIVES FOR PARTITIONING	16
TASK 6 - COMPARISON OF WASTE MANAGEMENT SYSTEMS WITH AND WITHOUT PARTITIONING	19
3.0 DECONTAMINATION AND DENSIFICATION OF CHOP-LEACH CLADDING RESIDUES .	23
DECONTAMINATION PROCESS	24
DENSIFICATION PROCESS	28
4.0 PYROLYSIS-INCINERATION OF COMBUSTIBLE ALPHA WASTES	35
PILOT PLANT STUDIES	35
ASH FIXATION AND PROCESS COMBINATION STUDIES	36
5.0 TRITIUM SEPARATION AND FIXATION	39
TRITIUM FIXATION	39
LASER EXCITATION EXPERIMENTS	43
WATER RECYCLE STUDIES	44

6.0	CHARACTERIZATION OF 300 AREA AND WYE BURIAL GROUNDS	47
	TASK 1 - REVIEW OF HISTORICAL RECORDS	47
	TASK 2 - WASTE DETECTION AND MAPPING	47
	TASK 3 - DRILL SAMPLING WELLS AND ANALYZE SAMPLES	51
	TASK 4 - INSTRUMENTED WELLS AND SOIL CHARACTERIZATION	52
	TASK 5 - EVALUATION OF MOISTURE MOVEMENT MECHANISMS AND MODELING	52
	TASK 6 - EVALUATION OF BIOLOGICAL TRANSPORT MECHANISMS	53
7.0	DISPOSITION (D&D) OF RETIRED CONTAMINATED FACILITIES AT HANFORD	55
	TASK 1 - DEVELOP METHODS, COSTS, AND PRIORITIES OF D&D AT HANFORD	55
	SUBTASK 2 - SITE CHARACTERIZATION	55
	SUBTASK 3 - DISPOSITION	57
	Disposition Alternatives	57
	Criteria Development	58
	Priority Development	59
	SUBTASK 5 - PROCEDURES AND TECHNIQUES	62
	TASK 2 - FORMULATE PLANS AND SPECIFICATIONS FOR A D&D DEMONSTRATION	63
	SUBTASK 1 - PLANNING AND EVALUATION	63
	SUBTASK 2 - SITE CHARACTERIZATIONS	63
	SUBTASK 5 - PROCEDURES AND TECHNIQUES	63
	SUBTASKS 6 - SAFETY- AND 7 - COST ESTIMATING	64
8.0	DEVELOPMENT OF PACKAGING TECHNOLOGY FOR $\text{Pu}(\text{NO}_3)_4$ SHIPMENT	67
	SOLID PLUTONIUM SHIPPING FORMS	67
9.0	TRANSPORTATION SAFETY STUDIES	71
	DISTRIBUTION	75

LIST OF FIGURES

1	Overall Flow of Disposal R&D Program Activities	2
2	Use of Analyses and Evaluations	3
3	The Effect of the Time of Initial Release from the Disposal Site on the 50-yr Accumulated Dose	20
4	Hydrogen Pressure Versus Time at 700°C	27
5	Equilibrium Hydrogen Pressure Versus 1000/T°K	28
6	Melt Stock for Inductoslag Heat Number USBM 431 Chopped and Cleaned Zircaloy-4 PWR Cladding	29
7	Zircaloy-4 Inductoslag Melted Ingot Heat Number USBM 431	30
8	Inductoslag Remelted Chopped Leached Cladding Hulls Zircaloy-4	30
9	Inductoslag Remelted Chopped Leached Cladding Hulls Zircaloy-4	31
10	As Cast Zircaloy-4 Middle Center Ingot Location Inductoslag Melted Heat Number USBM 431	31
11	Experimental Alpha Waste Facility Installation Schedule	36
12	An Overlay of an Aerial Photo of the 300N Burial Ground and the Existing Map of that Burial Ground	49
13	An Acoustic Reflection Profile in the 300N Burial Ground	51
14	Release Criteria for Contamination of Surfaces	58
15	Release Criteria for Contamination of Liquids and Solids	58
16	Sequence of Operations for Prioritization Methodology	60
17	Estimated Cost for Disposition of Facility Class A	61
18	Utility Function of Risk	61
19	Probability Density Functions of Total Importance for Three Facility Classes	62
20	Curves of Cumulative Total Importance for Seven Hypothetical Facility Classes	62
21	Evaporation of Plutonium Nitrate Solution at Raised Temperatures	68
22	Evaporation Vessel	69
23	Equipment Arrangement	69

LIST OF TABLES

1	Anticipated Schedule of Major Activities in the Disposal R&D Program	4
2	Study Plan for Evaluation of Alternatives for Management of ^{129}I and ^{99}Tc	10
3	Plan for Study of Loss of Canisters of High-Level Radioactive Waste at Sea	11
4	PNL Dose Computer Programs - Atmospheric Release	12
5	Dissolution of Simulated Calcine by Acid Digestion	15
6	Centrifugal Contactor Testing of Am Extraction	16
7	Standard Usages of the Maximum Individual for Dose Analysis Purposes	18
8	Summary of Accumulated Doses to Individual from Leaching Accident Occurring at 100 Years after Burial, Base Case (LST001 Source File)	19
9	Corrosion of Autoclaved Zircaloy-2 Samples by Treatment in 37 mole % LiF - 37 mole % NaF - 25 mole % ZrF_4 Salt at Various Temperatures and Times	26
10	Salt Adhering to Tubing Samples When Removed From Corrosion Tests Shown in Table 9	26
11	Chemical Analyses - Graphite Remelted Zircaloy-2 - Stainless Steel - Inconel Alloys	29
12	Chemical Analyses - Inductoslag Remelted Zircaloy-4 Heat Number USBM 431	32
13	Estimated Waste Composition To Be Used in Study	36
14	Feed Composition	37
15	Chemical Analysis of Ash from Pyrolysis-Incineration Process	37
16	Results of Accelerated Leach Tests	38
17	Ash:Glass Volume Reduction Factors	38
18	Leaching Data	41
19	Cost Estimates for Polymers	41
20	Tritium Storage Efficiency	43
21	Status: Study of D&D at Hanford	56
22	Proposed Radioactivity Concentration Levels for Unrestricted Release	59
23	Tentative Performance Criteria	60
24	Relative Risk and Cost Estimates for ^{233}S Disposition Alternatives	65

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.

SUMMARY

A plan for research and development on disposal of high-level and transuranic wastes was completed.

Major features are:

- Disposal to geologic media in the contiguous U.S. and the midplate Pacific seabed is emphasized.
- At least 15 generic disposal system options will be initially investigated. These will be winnowed to five specific sites for detailed R&D and then to verification testing at two of these five sites.
- Work is planned to develop analysis and evaluation methods needed to assess acceptability of disposal techniques.
- Practical use of disposal can begin in 1997.

Workshops to aid development of R&D plans for disposal of commercial radioactive wastes were held at Denver, Colorado; Berkeley, California; Seattle, Washington; and within the Waste Management 75 meeting at Tucson, Arizona.

Evaluation of nuclide migration from disposal media was extended by study of the effect of nuclide concentration on equilibrium constants and by investigation of migration from salt formations.

Revisions were completed of the videotape and questionnaire to be used to obtain public opinion on waste disposal evaluation factors.

Detailed special studies began on 1) management of ^{129}I and ^{99}Tc , 2) loss of high-level waste at sea, and 3) revision of models used to estimate radiation dose to man.

TASK 1 - DEVELOPMENT OF ADVANCED WASTE MANAGEMENT PLANS -

J. W. Bartlett

The objective for this task is to prepare plans for R&D programs on disposal of commercial radioactive wastes. The basic procedure for the work is to convene workshops composed of persons who collectively possess the spectrum of expertise needed for the plans. Results from these workshops are then used in plans prepared by Battelle Pacific Northwest Laboratories (PNL).

Initially the work was directed at comprehensive plans for disposal R&D on the various types of wastes produced in the nuclear fuel cycle. Plans were to be completed in September 1975. Subsequently, Dr. Frank K. Pittman, Director of ERDA's Division of Waste Management and Transportation, requested PNL to prepare, by March 1975, plans for disposal of

high-level and transuranic wastes. Work was consequently focused on these wastes, with the expectation that the resulting plans could prototype those for other waste types.

R&D Plans for Disposal of High-Level and Transuranic Wastes

The plan prepared by PNL recommends a 20-year, 206 million (1975\$) R&D program on geologic structures in the contiguous U.S. and on the midplate, mid-gyre region of the Pacific seabed with the objective of developing an acceptable method for disposal of commercial high-level and transuranic wastes by 1997.

Geologic media recommended for investigation include:

1. intrusive igneous rocks such as granite, and crystalline metamorphic rocks such as quartzite, because of their low permeabilities and high mechanical strengths;
2. salt, either in stable domes or thick beds, because of its low permeability and self-sealing properties;
3. tuff and shale because of their low permeabilities and high capacities to retard nuclide migration by adsorption; and
4. the midplate/midgyre region of the North Pacific, because of its stability and isolation from man's activities.

Analysis of generic concepts and geologic media has been essentially

completed by Holifield National Laboratory (HNL), Battelle Pacific Northwest Laboratories (PNL), and Sandia Laboratories. Work on generic systems* and site reconnaissance can be initiated.

The program will require activities illustrated by Figure 1. Selection of disposal method(s) will be done by winnowing options on the basis of R&D results and evaluations as illustrated by Figure 2.

FIGURE 1. Overall Flow of Disposal R&D Program Activities

*Generic System--a complete but conceptual disposal system definition including the waste form and any encapsulation, transport system, geological media, emplacement technique, and characteristics during the operational mode and monitoring--only disposal phase.

FIGURE 2. Use of Analyses and Evaluations

For near-term planning use, an assessment was made of the scope of work needed to assure high probability of attaining acceptable disposal methods within 20 years. On the basis of present knowledge, this study showed:

- Within the four broadly defined generic media for geologic disposal (salt, shale-type and granite-type rocks, and seabed) 15 generic disposal systems (defined as a conceptual system wherein disposal media, waste form, and operations are identified but without detail) should be chosen and subjected to initial studies. These studies would add data (e.g., by media characterization, design development and evaluation, and impact assessments) sufficient for selection of specific combinations of sites and designs for detailed study.

- The 15 initial studies should be winnowed to five disposal system options for detailed study. These studies would include site reconnaissance and evaluation, further detailing and evaluation of facilities, and assessment of safety, system performance, and implementation impacts. These detailed studies would give bases for decisions to select certain system options for demonstration and verification testing.

- The five specific disposal systems would be winnowed to two (identified by specific sites and operational methods) where five-year verification testing of the pilot plant type will be done. The objective of this testing will be to demonstrate and confirm that the disposal system is acceptable.

The numbers of studies cited here for these three types of effort are based on current uncertainties to be resolved by R&D and assessments. As study results more sharply characterize the acceptable disposal systems, the number of detailed studies and tests may be fewer than indicated here. Specifically, fewer than five detailed site-system studies may be required, and only one system for testing may be necessary. However, it is essential that the program start with fifteen generic system studies.

The program includes R&D to advance capability for assessing feasibility, safety, performance, and impacts associated with disposal system concepts. This work includes:

1. improvement and extension of analytical models used to assess safety, and
2. development and improvement of methods used to assess technical feasibility, system performance as reflected by costs, logistics, and manpower requirements, and implementation impacts including environmental, social (local, national, and international), land use, resources, energy, and economics.

The role of these analyses and evaluations is to provide information and interpretations needed for decisions involving choices among disposal options.

The anticipated schedule for the program is shown in Table 1. Selection of the two sites for pilot plant testing should be made late in CY 1982. At this time scoping design for these operations could also be started. Scoping, detailed design, licensing, and construction of the pilot plant facilities are estimated to require 8 or 9 years. Operation of the pilot facilities is therefore projected to begin in 1992.

If this schedule is attained, routine disposal operations could be initiated in 1997.

Workshop Activities

Several workshops related to development of disposal R&D plans were held during the report period. Objectives and accomplishments of these workshops are summarized below.

1. Denver Workshop. This workshop, with 11 participants, took place in the Federal Center at Denver, Colorado, on January 16 and 17, 1975. The objective was to define the scope

TABLE 1. Anticipated Schedule of Major Activities in the Disposal R&D Program

<u>Activity</u>	<u>Anticipated Date</u>
Detailed Program Plan	1/76
Generic Selection Completed and Specific Work on Sites Begun	1/78
Scoping of Pilot Plants Begun	1/83
Design of Pilot Plants Begun	1/85
Operation of Pilot Plants Begun	1/92
Commercial Operation Begun	1/97

and content of comprehensive R&D plans for disposal of commercial radioactive wastes. Output from the workshop is reflected in the contents of the R&D plan described above for the disposal of high-level and transuranic wastes. A specific finding of this workshop was that disposal R&D plans should include work to develop methods for assessing the socioeconomic impacts of disposal system alternatives.

2. Bay Area Workshop. This meeting took place on February 20 on the Berkeley campus of the University of California. Fifteen participants, most of them representing the social sciences, addressed problems related to socioeconomic assessment of disposal methods. Discussions indicated that 1) the disposal system concepts will have to be specific with respect to site and method if socioeconomic assessments are to be meaningful; 2) improvements in assessment methods will be necessary, and 3) improved interpretive description of technical matters is needed for effective dialog between the technical and social science communities.

As follow-on to this workshop a "layman's version" of the disposal

R&D plans described above will be prepared and the social scientists will review methods and applications of socioeconomic R&D. A workshop planned for Airlie House in early April was deferred until these activities are completed and decisions for future action are made.

3. Seabed Disposal Workshop. PNL personnel attended a workshop near Seattle, Washington, on March 20-22. This was the fourth in a series coordinated by Sandia Laboratories. Objectives for the meeting were to report recent results and to plan future work. Although many uncertainties remain, information reported by the oceanographers indicates the mid-plate region of the North Pacific merits consideration as a disposal medium because of its stability, low biological activity, remoteness from man, and capacity to retain waste materials. Emplacement is feasible with current technology.

Tentative plans were made for a small workshop aimed at defining a complete generic disposal system based on seabed disposal.

4. WM 75 Workshop. A workshop on ultimate disposal took place as part of the Waste Management 75 conference held in Tucson, Arizona, on March 24-26. The workshop included description of previously investigated proposed methods for disposal but focused on questions related to assessment and acceptability of disposal methods. The discussions essentially confirmed the need for assessment factors such as are described in the PNL plan (see Figure 2) but also confirmed the current lack of consensus on how to apply them.

When an overall assessment of these workshop activities is made, four conclusions emerge:

1. A firm base exists for the purely technical aspects of disposal R&D. Promising disposal media have been identified, engineered designs for disposal systems can be developed from existing technology or reasonable extrapolations, and site qualification R&D can be identified and performed.
2. A poor basis exists for effective definition and use of assessments, particularly those related to the socioeconomic aspects of the disposal systems.
3. There is consensus, based partially on experience and partially on projections, that acceptability is a function of technical and social factors.
4. There is no consensus on how the technical and social factors can be combined and applied to decisions on acceptability of a disposal system.

A probable reason for lack of consensus on assessment methods involving technical and social factors is that none has been developed or tried. All evidence indicates that development of appropriate assessment methods should be a near-term goal of disposal R&D programs.

TASK 2 - DEVELOPMENT OF EVALUATION METHODOLOGY RADIONUCLIDE MIGRATION FROM NONSALT FORMATIONS - H. C. Burholder and M. O. Cloninger

Work on this subtask is reported in Section 2.0, "Separation of Actinide Elements from Radioactive Waste," in this quarterly report.

RADIONUCLIDE MIGRATION FROM SALT FORMATIONS - H. C. Burkholder and G. E. Koester

The previously developed methodology for evaluating the consequences of a leach incident at an underground nuclear waste disposal site makes a number of assumptions to reveal the effects of adsorption, dispersion, and decay on the movement of nuclides through soil.^(1,2) One of these assumptions involves the use of "adsorption" equilibrium constants for each of the nuclides which are independent of both position and time. This assumption may be realistic for a nonsalt geologic formation, but its accuracy for a salt formation has not been determined. It is well known that ions present in the leachant (e.g., sodium ions) can strongly affect the "adsorption" equilibrium constant.^(3,4) Thus we need a methodology to evaluate the effect of salt on the migration of nuclides from an underground waste disposal site.

Previous work aimed at developing this methodology considered the effect of salt (sodium chloride) on the migration of single nuclides.⁽⁵⁾ A continuous line source injection into a two-dimensional soil column was assumed for the saltwater, but an impulse release into a one-dimensional soil column was assumed for the nuclides. These assumptions are incongruous because it seems unreasonable for the salt and the waste to be released by two different mechanisms.

We can develop an alternative model by considering the release of the salt and the waste on the same basis. There are at least three possible generic release situations.

Situation I -- Salt Formation with Saltwater Injection Before, During, and After Waste Release

At some time after the waste is deposited, water comes into contact with the salt formation. Since the salt is much more soluble than the glass waste form, it seems likely that salt-saturated water will flow away from the site for some time before significant quantities of waste are released. Next the waste is released over some other period of time along with additional saltwater until all of the waste has been leached away. Finally if the bedded salt formation is large relative to the underground disposal facility, additional saltwater may be released after the waste release has been completed.

Situation II -- Salt Formation with Saltwater Injection Before Waste Release Only

Since a high salt content in the leachant may inhibit the solubility of the waste, all of the salt may be dissolved from around the waste before any waste is released. In this situation we have two distinct release periods--salt release without waste and waste release without salt. However, the migration of the waste may be affected by the salt which has gone before.

Situation III -- Salt Formation with Saltwater Injection During Release Only

In this situation the salt and the waste are released together with no salt being released either before or after the waste.

We consider a waste disposal site in a salt formation and a one-dimensional soil column connecting

it with a surface water body and write the differential equations which describe the migration of the salt and an arbitrary single nuclide. We will make all of the assumptions outlined in Reference 1 except that the adsorption equilibrium "constant" will be a function of the salt concentration in the salt-waste-water solution. These equations are:

$$D \frac{\partial^2 C_s}{\partial Z^2} - V \frac{\partial C_s}{\partial Z} - K_s \frac{\partial C_s}{\partial t} = 0 \quad \text{salt}$$

$$D \frac{\partial^2 N_1}{\partial Z^2} - V \frac{\partial N_1}{\partial Z} - K_1(C_s) \frac{\partial N_1}{\partial t} - K_1(C_s) \lambda_1 N_1 = 0 \quad \text{nuclides}$$

where

C_s = salt concentration
 K_s = adsorption equilibrium constant for salt

The other quantities are defined in Reference 1. If we consider the most complicated situation (Situation I) and let our time start when the first salt is released from the formation, the boundary conditions are as follows:

$$\begin{aligned} C_s &= 0 & @ t = 0, \text{ all } Z \\ C_s &= C_s^0 & @ 0 < t < T, Z = 0 \\ C_s &= 0 & @ t > T, Z = 0 \\ C_s &= \text{finite} & @ t > 0, Z = L \end{aligned} \quad \text{salt}$$

where

C_s^0 = salt concentration at the soil column inlet

$$N_1 = 0 \quad @ t = T_1, \text{ all } Z$$

$$N_1 = \frac{N_1^0}{(T_2 - T_1)} \text{EXP}(-\lambda_1 t) \quad @ T_1 < t < T_2, Z = 0$$

$$N_1 = 0 \quad @ t > T_2, Z = 0 \quad \text{nuclide}$$

$$N_1 = \text{finite} \quad @ t > T_1, Z = L$$

We can solve the equation for the salt migration by analogy with that for a single nuclide from Reference 1 and obtain the result:

$$C_s = \frac{C_s^0}{2} \left\{ \text{erfc} \left[\frac{1}{2} \sqrt{\frac{K_s P}{D}} - \sqrt{\frac{P_0}{4K_s}} \right] - \text{erfc} \left[\frac{1}{2} \sqrt{\frac{K_s P}{D}} - \sqrt{\frac{P \left(0 - \frac{TV}{L} \right)}{4K_s}} \right] \right\}$$

where the variables are defined above and in Reference 1.

We know from the previous measurements of $K_1(C_s)$ for various nuclides that $K_1(C_s)$ is of the form:

$$K_1(C_s) = 1 + \frac{C_1}{C_2} \exp \left[C_1 - C_2 \ln C_s \right] = 1 + \frac{C_1}{C_2} \exp(C_1) C_s^{-C_2}$$

where

C_1 and C_2 are empirical constants determined for each nuclide. We can now solve the differential equation for a migrating nuclide since we have an expression for $K_1(C_s)$ valid for all positions and times of interest.

This study will explore both analytical and numerical methods of solution and develop the calculational scheme necessary to model the migration of radionuclides from salt formations. We will then have the calculational schemes necessary to compare the consequences of leach incidents at salt and nonsalt formations. Then from estimated probabilities of the

release events we will be able to compare the risks for the two types of geologic formations for this particular pathway.

PUBLIC ATTITUDES TOWARD NUCLEAR WASTE MANAGEMENT - W. S. Maynard (HARC)

Work completed during this quarter included substantial revisions of the questionnaire and film presentation material which was incorporated in the December 31, 1974 quarterly report. This integrated package of materials has been pretested and reviewed by the Human Affairs Research Center (HARC) and PNL staff for both technical accuracy and for appropriateness as a procedure for communicating with the public.

Questionnaire. The questionnaire is designed to measure respondents' judgments about the relative importance of four waste disposal evaluative factors (Parts I and II) and to obtain their judgments about various levels of each of these factors (Part III). These responses will be incorporated into a decision model which combines social value measurements with technical performance measure to yield an ordinal ranking of proposed waste disposal concepts. The other three sections of the questionnaire (Part IV, presentation of hypothetical disposal concepts; Part V, attitude-opinion-brief statements; and Part VI, respondent evaluation) will be used to gain a better understanding of how respondents view the issue of nuclear waste disposal and will furnish a context for interpreting the results obtained in Parts I, II, and III.

Film Presentation. The written script on which the film is based has undergone extensive review during this quarter to insure that the information presented is 1) technically accurate and balanced and 2) meaningful to respondents. The BNW photography department has used this script to prepare a 12-min color videotape with professional graphic material and narrative. After sponsor clearance, this videotape will be transferred to 16mm film to reduce the logistical problems in data collection.

Data Collection. The sampling procedure involves purposive sampling of 20 groups of respondents, each group composed of at least 20 individuals. Four respondent groups will be drawn from each of 5 geographical regions of the United States. University colleagues in 4 regions will serve as field representatives and will arrange for and administer the instruments to the designated groups in their respective regions. HARC staff will conduct the data collection activities in the Northwest.

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.

During this quarter, in-depth effort was started on three studies. Need for these studies was identified

in the process of preparing the R&D plans for disposal of high-level and transuranic wastes, listed below, reported as BNWL-1904, DRAFT (see Task 1). Results of these studies will be applied to evaluation of disposal method alternatives.

WASTE MANAGEMENT FOR ^{129}I AND ^{99}Tc -
H. G. Rieck

Previous work has shown that ^{129}I and ^{99}Tc may be major contributors to radiation dose rates resulting from escape of waste from a disposal repository; these isotopes transport readily in geologic formations and the biosphere because of their relatively high solubility in water.

The scope and content of work in this study are outlined in Table 2. To date, review of prior work and compilation of basic data have been completed.

LOSS OF HIGH-LEVEL WASTE AT SEA -
W. B. Silker

An accident possibility associated with seabed disposal of high-level waste is loss of canisters prior to or during emplacement. This study will assess the mechanisms and consequences of such accidents and the feasibility of recovering the canisters.

The scope and content of work in this study are outlined in Table 3. Some preliminary information on this subject was obtained at the seabed workshop (see Task 1) which included this topic on its agenda. Participants in this workshop indicated that a single canister lost during emplacement operations could be located by any one of several methods (e.g., those listed in Table 3) and that, in

principle, recovery is feasible with existing technology. Additional work is needed, however, to define detection and recovery actions for specific loss circumstances.

IMPROVEMENT AND DOSE MODELS -

E. C. Watson and J. K. Soldat

Technical evaluation of disposal system alternatives includes estimation of potential radiation doses to man. PNL has previously developed models for predicting doses, but new information permits periodic revision and updating.

These studies will update the models for atmospheric (Table 4) and other environmental pathways to man. Modifications to the atmospheric release models will include calculation of:

- Ground contamination from dry deposition of airborne radionuclides.
- Ground contamination from wet deposition of airborne radionuclides.
- Dose from direct radiation from ground contamination.
- Cloud depletion by wet deposition resulting from both rain-out and wash-out mechanisms.

Other activities during this report period included:

1. The equation used for the calculation of internal dose from inhalation of insoluble radionuclides was rederived. Dose factors were then calculated from this new equation and placed in our files.
2. The literature was scanned for more recent data on the behavior of tritiated water (HTO) in the environment. The data reconfirmed that HTO does not accumulate in the environment or in any biota,

TABLE 2. Study Plan for Evaluation of Alternatives
for Management of ^{129}I and ^{99}Tc

- I. Identify the waste management problems for ^{129}I and ^{99}Tc .
 - A. Estimate the amounts of each to be managed.
Volume, weight, and heat generation will be important factors in waste disposal.
 - B. Identify radiological hazards.
 - C. Describe chemical, physical and biological behavior of these elements.

This section intends to provide the background information required to properly evaluate a proposed waste management technique.

- II. Identify physical and chemical forms which would be acceptable for waste disposal techniques.
 - A. Evaluate behavior of chemically inert compounds.
 - B. Evaluate physical and chemical stability of mixtures such as solidification products or calcined materials.
 - C. Review isotope (^{129}I and ^{99}Tc) recovery techniques which are proposed for fuels reprocessing operations.
 - D. Evaluate problems in transforming the recovered isotope to a preferred chemical and/or physical form.

This section intends to propose a preferred physical and chemical form of the isotopes which will provide maximum stability after disposal. It should also suggest the end product of any ^{129}I or ^{99}Tc recovery phases during fuels reprocessing operations.

- III. Identify all possible alternatives for management of these isotopes.
 - A. Identify applicable disposal techniques.
 - B. Evaluate isotopic dilution for ^{129}I .
 - C. Evaluate possible commercial applications for these isotopes.
 - D. Evaluate transmutation techniques.

In addition to normal disposal methods, this section intends to propose other possibilities such as isotopic dilution, transformation and commercial applications.

- IV. Evaluate each alternative management technique on the following points:
 - A. Preparation of final disposal form--processing and packaging
 - B. Disposal operation--logistics, site preparation, and disposal
 - C. Inventory control
- V. Evaluate the cost and risk factors for all possible methods.
- VI. Recommend future work.
 - A. Research and development
 - B. Pilot-scale studies
 - C. Field studies
 - D. Additional feasibility studies

TABLE 3. Plan for Study of Loss of Canisters of High-Level Radioactive Waste at Sea

- I. Conditions under which waste canisters could be lost
 - A. Shipwreck
 - B. Canister loss at sea
 - C. Single canister loss at disposal site
- II. Means for retrieval of lost canisters
 - A. Means for locating canister on the sea floor
 - a. Acoustic
 - b. Flotation release
 - c. Measurement of activation products
 - d. Magnetic
 - e. Optical
 - B. Potential means for recovery from sea floor
- III. Assume irretrievable loss
 - A. Maintenance of canister integrity
 - a. Physical damages
 - b. Corrosion
 - 1. Material of construction
 - 2. Estimate of lifetime
 - 3. Remaining radionuclides
 - 4. Effect of self-heating
 - B. Dissolution of solidified waste
 - a. Type of process
 - calcine - ceramic - glass
 - b. Dissolution rates for each
 - c. Ion exchange on sedimentary material
 - Types of material in sediments
 - C. Transport and dilution of dissolved radionuclides
 - a. Advective transport by currents
 - b. Diffusive mixing processes
 - c. Eventual oceanic distribution
 - D. Uptake by organisms
 - a. Transfer coefficients through the marine food web
 - b. Impact on man

with the exception of the water flea, which was found by one study to have a bioaccumulation factor of 4. Thus, except for this creature, all biota will equilibrate with their water media to the specific activity of ^3H in the media. Modification of the calculation scheme to incorporate the specific activity concept for HTO appears feasible. The radiation doses calculated by the new scheme may be somewhat lower than those previously calculated. Similar considerations for ^{14}C may be necessary since this nuclide contributed a major portion of certain of the doses previously calculated for AWM projects.

- 3. The code for estimating doses from the consumption of contaminated food crops and animal products (milk, eggs, meat) was modified to print out a more comprehensive summary of the factors and intermediate media concentrations used in the estimation of doses from a new list of 190 radionuclides.

TABLE 4. PNL Dose Computer Programs - Atmospheric Release

Program	Language	Status		Purpose ^(a)
		Operational	Documented	
RACER	FORTTRAN	UNIVAC 1108	BNWL-B-69 June 1971	Tissue dose from finite size cloud, accidental release
SUBDOS	FORTTRAN	CYBER 70 ^(b)	BNWL-B-351 In press	Penetrating dose from finite size cloud, accidental release
KRONIC	FORTTRAN	CYBER 70 ^(b)	BNWL-B-264 June 1973	Penetrating dose from finite size cloud, chronic release
SUBPOPA	FORTTRAN	CYBER 70 ^(b)	Being written	Population dose from accidental releases
LONGTM	FORTTRAN	CYBER 70 ^(b)	Included in BNWL-B-264	Normalized air concentrations from chronic releases
DACRIN	FORTTRAN	CYBER 70 ^(b)	BNWL-B-389 Dec. 1974	Internal dose via inhalation following chronic or accidental releases
GRONK	BASIC	UNIVAC 1108	BNWL-1754 Feb. 1974	Annual penetrating and skin doses from semi-infinite cloud to an individual or populations. Option permits thyroid dose calculation for inhalation and ingestion.

a. All programs are for atmospheric releases.

b. Can be run at either CSC (Richland) or at Battelle Columbus.

REFERENCES

1. D. H. Lester, G. Jansen, and H. C. Burkholder, Migration of Radionuclide Chains through an Adsorbing Medium, BNWL-SA-5079, Battelle, Pacific Northwest Laboratories, Richland, WA, March 1975.
2. H. C. Burkholder and G. Jansen, Nuclear Waste Management and Transportation Quarterly Progress Report, October through December 1974, Battelle, Pacific Northwest Laboratories, Richland, WA, p. 4, April 1975.
3. G. Jansen, R. C. Routson, and A. G. Gibbs, Advanced Waste Management Studies Progress Report, BNWL-B-223-10, Battelle, Pacific Northwest Laboratories, Richland, WA, p. 31, May 1974.
4. R. C. Routson and A. V. Robinson, Advanced Waste Management Studies Progress Report, BNWL-B-223-11, Battelle, Pacific Northwest Laboratories, Richland, WA, p. 20, July 1974.
5. G. Jansen and H. C. Burkholder, Nuclear Waste Management and Transportation Quarterly Progress Report, BNWL-1876, Battelle, Pacific Northwest Laboratories, Richland, WA, p. 22, November 1974.

2.0 SEPARATION OF ACTINIDE ELEMENTS FROM RADIOACTIVE WASTE

E. J. Wheelwright - Project Manager

The primary FY-75 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

At the mid-year review with DWMT Management held on February 5, directions were received to terminate work on Tasks 1 through 4, continue the work on Task 5, revise downward the scope of work planned for Task 6, and write a report at the end of the fiscal year summarizing the work completed to that date. These directions were promptly implemented.

A sample of irradiated "Yankee" oxide fuel has been dissolved in nitric acid, and the residual solids separated and then treated by special means to dissolve the solids not normally dissolved in nitric acid, thereby permitting determination of the composition of the solids. The analytical work is incomplete, and the results cannot be given at this time.

Complete dissolution of simulated calcine has been demonstrated using a 2-step acid digestion procedure. The calcine was first digested at 100°C in 8M HNO_3 for 6 hr, and then the dissolution was completed by digestion of the remaining solid mate-

rial in 8M HNO_3 - 0.15M HF at 100°C for 4 hr. Attempts to dissolve the calcine using HNO_3 -HF only or HNO_3 -HF followed by HNO_3 were not successful.

A conceptual separation flowsheet is being prepared based upon the limited amount of laboratory and literature work completed prior to termination of work on Task 4. However, this flowsheet is not optimized, comprehensive, or tested in the laboratory. The initial results from laboratory testing of a DBBP solvent extraction process in the miniature centrifugal contactors indicated reasonable americium extraction.

Dose rate calculations have been completed for the base case soil migration model and for three variations of the initial release time. Dose rate calculations for variations in the leach rate and path length will be completed soon. Work is nearing completion on the computer program for an air-release model to permit dose rate calculations for release of activity from a surface mode.

TECHNICAL PROGRESS

TASK 1 - CHARACTERIZATION AND TREATMENT OF SOLIDS IN HIGH-LEVEL WASTES -

F. P. Roberts and J. H. Westsik

One hundred twelve grams of 2- to 3-in.-long fuel pieces, cut from two fuel rods, were heated for 4 hr at 90 to 95°C in 250 ml of 5.5M HNO_3 . The solution was filtered through a 5- μ Mitex Millipore filter. The residue, filter, and cladding hulls were then heated an additional 4 hr in 8.0M HNO_3 and refiltered. Approximately 22 g of cladding hulls and 0.3 g of solids were isolated. The two dissolver solutions and the wash solution were combined and diluted to 1,000 ml.

The filtered solids were treated to solubilize the contained actinide elements. The treatment involved leaching for 48 hr in 8M HNO_3 -0.5M HF at 40 to 60°C, followed by fusion with NaHSO_4 . After dissolving the flux in 2M HNO_3 and filtering, the filtrate and the leach solution were combined and diluted to 250 ml. A sample from the dissolver solution and a sample from the solids treatment solution are currently being analyzed for burnup and actinide content.

TASK 2 - REMOVAL OF LONG-LIVED NUCLIDES FROM SOLIDIFIED WASTE -

J. A. Partridge

The objective of this task is to develop a partitioning flowsheet based on starting with solidified high-level waste, e.g., a calcine.

The work this quarter was directed toward developing a dissolution scheme for getting calcined waste into solution for subsequent parti-

tioning processing, and toward testing and optimizing that dissolution scheme with calcined high-level waste in the 325 Building High-Level Radiochemistry Facility. The laboratory dissolution tests were done with simulated calcine prepared by pot calcination of a synthetic high-level waste which had been filtered prior to the calcination process. The synthetic waste, referred to as PW-4b, was described in the last quarterly report on this task.

During this report period, the simulated calcine was completely dissolved by using a two-step acid digestion process. The calcine was first digested at 100 to 105°C in 8M nitric acid for 6 hr. Sixty-six percent of the calcine dissolved during this treatment. Digestion of the residual solids from this treatment in 8M nitric acid - 0.15M hydrofluoric acid at 100°C for less than 4 hr resulted in complete dissolution of the solids. (See acid tests 9 and 9b in Table 5.)

As shown by the data in Table 5, attempts to dissolve the calcine using either a nitric-hydrofluoric acid digestion only or a nitric-hydrofluoric acid digestion followed by a nitric acid digestion were not successful in complete dissolution of the calcine. The results indicate that in the nitric-hydrofluoric acid treatment both the nitric acid and the hydrofluoric acid concentrations are important. Eight molar nitric acid performed better than concentrated nitric acid or 4M nitric acid when in combination with HF, and 0.15M HF gave better results than did 0.04M HF.

TABLE 5. Dissolution of Simulated Calcine by Acid Digestion

<u>Acid Test No.</u>	<u>Acid</u>	<u>ml Acid/g of Calcine</u>	<u>Temp. (°C)</u>	<u>Time (Hr)</u>	<u>% Calcine Dissolved</u>
1	15.7M HNO ₃	30	80	6	43
2	8M HNO ₃	30	80	6	45
3	4M HNO ₃	30	80	6	42
4	15M HNO ₃ - 0.15M HF	40	100-105	2	87
	followed by 4M HNO ₃	30	100-105	3	90-95
5	4M HNO ₃	30	100-105	2	41
	followed by				
	15M HNO ₃ -0.15M HF	40	100-105	3	90-92
6	8M HNO ₃ -0.15M HF	40	100-105	3	97
7	8M HNO ₃ -0.15M HF	40	100-105	3	92
8	4M HNO ₃ -0.15M HF	40	100-105	3	90
9	8M HNO ₃	20	100-105	6	66
	followed by				
	(a) 8M HNO ₃ -0.04M HF	40	100	4	98
	(b) 8M HNO ₃ -0.15M HF	40	100	<4	100

Preparations were progressing toward testing this two-step acid dissolution of calcined waste in the 325 Building High-Level Radiochemistry Facility with some actual calcined high-level waste. However, termination of the work on this task was requested early in the quarter, and those planned tests were not conducted.

TASK 3 - REMOVAL OF LONG-LIVED NUCLIDES FROM NON-HIGH-LEVEL WASTES -

J. H. Westsik

The objective of Task 3 was to evaluate the applicability of the partitioning concept to non-high-level nuclear wastes and to develop the technology necessary to apply partitioning to these wastes, where desirable, to integrate them into the overall Advanced Waste Management Program.

During Fiscal Year 1975, Task 3 was limited to surveying the general area of non-high-level waste to determine the scope of the problem and the need, if any, for partitioning. A literature search had been started but was cancelled when directions were received to terminate work on this task.

TASK 4 - DEVELOPMENT OF LOW-ADDITIVE FLOWSHEETS FOR REMOVING LONG-LIVED NUCLIDES FROM THE FISSION PRODUCTS -

J. L. Swanson

The objective of this task was 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 inert additives to the waste.

The DBBP solvent extraction process designed to accomplish separation of Am and Cm from the bulk of the fission products was in the very early stage of testing in our miniature centrifugal contactors when work on this task was terminated. The initial results were quite promising in that Am extraction followed the expected behavior reasonable well down to a low concentration.

These initial tests were aimed at maximizing Am recovery and did not include any fission products. Two six-stage contactors were connected in series to provide eight extraction and four scrub stages. The conditions employed and results obtained in the two runs are presented in Table 6.

TABLE 6. Centrifugal Contactor Testing of Am Extraction

Extractant = 60% OBBP-NPH				
Temperature = 40-45°C				
Stream	Stage Introduced	Flow (ml/min)	HNO ₃ , M	Am, %
<u>Run 1</u>				
Extractant	12	4.96	--	--
Acid-add	11	0.99	12.0	--
Feed	5	1.62	3.0	100
Scrub	1	0.80	3.0	--
Organic product			1.7	85
Aqueous waste			3.6	30
<u>Run 2</u>				
Extractant	12	4.93	--	--
Acid-add	11	0.38	12.0	--
Feed	5	1.65	3.0	100
Scrub	1	0.82	3.0	--
Organic product			1.7	96
Aqueous waste			1.7	8

From the flow ratios and acid concentrations used in Run 2 and the americium extraction coefficients measured at room temperature (reported last quarter), the Am loss in Run 2 should have been less than 1% instead of the 8% measured. Samples

taken during Run 2 from the channels connecting some of the stages showed the effective americium extraction coefficients to be about 75% as great as those observed in the batch contacts at room temperature. Since the variation was about the same in the scrub section as in the extraction section, this variation may have been caused by a decrease in extraction coefficient with increasing temperature rather than by kinetic effects. Using the lower extraction coefficients to calculate the expected loss gives a value of about 5% instead of the 8% measured. This remaining discrepancy was probably the result of less-than-optimum contactor performance; some of the organic phase samples taken from the connecting channels contained rather large amounts of aqueous phase also. Such behavior certainly reduces the number of theoretical stages achievable.

The Am concentration in the feed for these runs was about 10^{-4} g/liter, which is about 1000 times lower than that expected in commercial high-level waste solutions. The results of these runs thus suggest that Am recoveries from the high-level waste solutions at least as high as 99.99% should be attainable without encountering kinetic problems which could limit the recovery.

TASK 5 - EVALUATION OF INCENTIVES FOR PARTITIONING - H. C. Burkholder,

D. A. Baker, and M. O. Cloninger

The purpose of this task and a description of the work were reported previously (Nuclear Waste Management and Transportation Quarterly Progress Report, October - December 1974).

Radionuclide migration calculations have been performed for an assumed "base case" leach incident at an underground disposal site. For this initial calculation it was assumed that, starting in the year 2100, groundwater from a typical western U.S. desert leached all the unpartitioned, year 2000 nuclear power economy waste. The leaching occurred over a 333-year period into a 10-mile soil column through which water travels in 145 years. The results of these calculations were reported in the previously referenced quarterly.

A pathway analysis of radiation dose was performed for the "base case" on most of the radionuclides calculated to enter a river at various times after the leach incident. The indicated releases were assumed to enter and become completely diluted in a river having an average flow rate of 10,000 cfs. The radionuclides were assumed to contribute to the dose of a hypothetical adult maximum individual* over a period of 50 years through various liquid pathways. The pathways leading to an internal dose to this individual included fish and invertebrates harvested from the river, drinking water and foods produced by irrigation with river water. The irrigated foods included produce, such as leafy vegetables, potatoes, and fruits, and included animal products, such as milk, eggs, beef, pork, and poultry. External dose pathways included swimming and boating in the river and the

direct radiation from the radionuclides mixed with the shoreline sediments. The methodology, models, and assumptions used for the estimates are summarized in previous documents.⁽¹⁻⁵⁾ Standard food consumptions and usages were those of a maximum individual living today (Table 7). Although in the far distant future these parameters may change, it is felt that foods harvested from the land and waters would have essentially similar radionuclide distributions.

Table 8 summarizes the results of this analysis, giving the critical radionuclides, critical organ, and pathways which are significant contributors to the dose. The doses were calculated assuming they were received over 50 years, i.e., a 50-year dose accumulation. Radionuclides in soil and sediment were assumed to have been accumulated for a constant 50 years throughout this 50-year dose accumulation period. In addition the 50-year uptake was assumed at radionuclide concentrations contributed from the release at the peak year.

Table 8 shows that the radionuclide of most importance for this base case (leach incident 100 years after burial) is carbon-14 at 1550 years after burial. The consumption of meat (beef, pork, and poultry) and produce contribute 72 and 18% of the dose, respectively, which after 50 years of accumulation would give an individual approximately 300 rem to his bone.

*A hypothetical person whose diet and recreational habits are chosen to maximize the radiation dose potentially received.

TABLE 7. Standard Usages of the Maximum Individual for Dose Analysis Purposes

Pathway	Annual Usage	Remarks
River Fish (kg)	18	
River Invertebrates (kg)	18	Divided equally between molluscs and crustaceans
Drinking Water from River (liter)	730	
Shoreline Sediments (hour)	500	
Water Recreation (hour)	200	Divided equally between swimming and boating
Irrigated Produce		
Fresh Leafy Vegetables (kg)	30	On basis of 72 kg/yr over 5-month growing season
Other Fresh Above-Ground Vegetables (kg)	30	On basis of 72 kg/yr over 5-month growing season
Potatoes (kg)	110	
Root Vegetables (kg)	72	
Berries (kg)	30	
Melons (kg)	40	
Orchard fruit (kg)	265	
Wheat (kg)	80	
Other Grain (kg)	8.3	On basis of 20 kg/yr over 5-month growing season
Eggs (kg)	30	
Milk & Products (liter)	274	On basis of 1 l/d over 9-month grazing season
Meat		
Beef (kg)	40	
Pork (kg)	40	
Poultry (kg)	18	

The work performed in this quarter varied the time period between burial and the release at the disposal site to determine the effect of this parameter on the doses. The other "base case" parameters were held constant. The results of varying this parameter are summarized in Figure 3. The points on the graph represent accumulated doses received by an individual over a 50-year period beginning when the peak concentration of the indicated nuclide ("peak nuclide") occurred in the biosphere. The points are plotted versus the time after

burial at which the release at the disposal site occurred, rather than at the time at which the doses are received. Thus, for example, the ^{14}C and the ^{99}Tc doses are not received by the same individuals. The results show that only the ^{14}C dose decreases significantly as the initial release time increases from 100 to 10,000 years. After 10,000 years all the doses tend to decrease at an increasing rate. Carbon-14 decreases at the greatest rate and at 100,000 years becomes insignificant.

TABLE 8. Summary of Accumulated Doses to Individual from Leaching Accident Occurring at 100 Years after Burial, Base Case (LST001 Source File)

Years Since Burial	Peak (b) Nuclides	Critical Nuclide	Critical Organs	Organ Dose(mrem)(c)(d)	Significant Pathways
2.50E2 ^(a)	H-3, Tc-99 I-129	Tc-99 I-129	GI-LLI Thyroid	2.5E4 1.5E4	Irrigated produce & milk Irrigated produce & milk
1.55E3	C-14	C-14	Bone	3.1E5	Meat & produce
1.47E4	Sr-90 Ca-41 Np-237 Se-79	Np-237+D Se-79	Bone Liver	1.1E4 3.5E3	River invertebrates & produce Produce & milk
1.45E5	Cs-135	Cs-135 Cs-135 Cs-135	Bone Liver Total Body	8.5E2 8.5E2 3.3E2	River fish Fish Fish
1.61E5	Sn-126 Pd-107	Sn-126+D Sn-126+D	Bone GI-LLI	4.6E3 2.4E3	Fish & shoreline sediment Fish & shoreline sediment
1.45E6	Pu-244 Am-241 Zr-93 Cd-113m	Nb-93M	GI-LLI	6.6E2	Fish
2.07E6	U-238	Ra-226+D Ra-226+D	Bone Total Body	4.5E2 8.3E2	River invert., milk & produce River invert., milk & produce

- a. i.e., 150 years after accident.
 b. Nuclides with highest release rate at year in column 1.
 c. Total accumulated dose from continued ingestion and shoreline exposure throughout the 50 years following the accident.
 d. Doses from radionuclides present during the peak years for CM-247 (4.85E5 yr) and Th-232 (7.23E6 yr) were estimated at less than 1 mrem.

Although the doses tend to decrease as the release from the disposal site is delayed for times greater than 10,000 to 1,000,000 years, the dose continues to be controlled by the same nuclides (^{14}C , ^{99}Tc , ^{129}I , and ^{237}Np). Thus for this particular disposal situation, the time of initial release at the site has no effect on which nuclides control the dose unless the release can be delayed beyond 1,000,000 years.

TASK 6 - COMPARISON OF WASTE MANAGEMENT SYSTEMS WITH AND WITHOUT PARTITIONING - J. W. Bartlett

The scope and content of this task were given in the previous quarterly report. Significant effort in this task has been delayed until meaningful results from Task 5 become available. The output from Task 6 will take the form of identifying favorable and unfavorable factors.

FIGURE 3. The Effect of the Time of Initial Release from the Disposal Site on the 50-yr Accumulated Dose

REFERENCES

1. J. K. Soldat, N. M. Robinson, and D. A. Baker, Models and Computer Codes for Evaluating Environmental Radiation Doses, USAEC Report BNWL-1754, Battelle, Pacific Northwest Laboratory, February 1974.
2. D. H. Denham, D. A. Baker, J. K. Soldat, and J. P. Corley, Radiological Evaluations for Advanced Waste Management Studies, USAEC Report BNWL-1764, Battelle, Pacific Northwest Laboratory, September 1973.
3. J. F. Fletcher and W. L. Dotson (compilers), HERMES - A Digital Computer Code for Estimating Regional Radiological Effects from the Nuclear Power Industry, USAEC Report HEDL-TME-71-168, Hanford Engineering Development Laboratory, Richland, WA, 1971.
4. J. K. Soldat, J. F. Cline, B. Klepper, B. E. Vaughan, F. P. Brauer, J. E. Fager, W. H. Rickard, and D. G. Watson, The Radioecology of Iodine-129: An Interim Report, USAEC Report BNWL-1783, Battelle, Pacific Northwest Laboratory, September 1973.
5. J. K. Soldat, Modeling of Environmental Pathways and Radiation Doses from Nuclear Facilities, USAEC Report, BNWL-SA-3939, Battelle, Pacific Northwest Laboratory, Richland, WA, 1971.

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 evaluating 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

A piece of Saxton fuel cladding was decontaminated by treatment in the gaseous HF (600°C) followed by an aqueous fluoride solution. Analysis of the base metal established that 99.7% of the α activity was removed from the piece, with a weight loss corresponding to 42 μ m (1.6 mil) surface removal. Residual α activity in the metal (0.3%) was attributable to the irradiated tramp uranium in the Zircaloy-4 tubing material.

In an alternative molten salt decontamination process, removal of Zircaloy corrosion film in molten (H, Li, Na, Zr)F_x salt at 600 to 700°C was effective but caused problems with salt dragout and excessive Zircaloy corrosion.

The hydrogen absorption characteristics of the Zircaloy-stainless steel-Inconel alloy was compared with Zircaloy-2 to evaluate its usefulness as a tritium storage reservoir. In general, the alloy absorbed the hydrogen less rapidly than the Zircaloy-2, but equilibrium pressures of hydrogen over the alloy were similar to those of Zr-2.

Complete chemical analyses have been received for five of the six Zircaloy-2 - stainless steel - Inconel alloys melted in graphite crucibles and reported during the previous quarter.

A 10 cm diam x 94 cm long, 41-kg ingot of Zircaloy-4 has been melted by the U.S. Bureau of Mines using their cold mold Inductoslag process.

Reject PWR cladding tubing that had been chopped and cleaned was used as melt stock. Chemical analyses and hardness results from this ingot show that this material can be fabricated into nuclear components for reuse. Billets from this ingot have been machined, upset and extruded into a rod 3.6 cm diam x 97 cm long and a tube 3.8 cm OD x 2.5 cm ID x 198 cm long.

TECHNICAL PROGRESS

Preliminary Decontamination and Densification Process

The potential processes for decontamination and densification of the chop-leach cladding hulls have been discussed in a previous publication⁽¹⁾ Briefly stated, the processes selected for investigation and development consist of: 1) a gas phase pre-treatment of oxidized Zircaloy in HF at about 600°C followed by aqueous fluoride etchants. 2) Melting of the cleaned and decontaminated Zircaloy with the stainless steel and Inconel fuel element hardware to obtain a dense, corrosion resistant, alloy. A small additional decontamination during melting by flux treatment, absorption or volatilization may be possible. 3) As an alternative to 2) the Zircaloy, stainless steel, and Inconel may be melted separately with a possible reuse of the metal after short half-life isotopes have decayed. A molten salt decontamination process for zirconium oxide removal, followed by melting for densification is a backup process. The process steps requiring the most immediate attention are the oxide removal from zirconium alloys and the

melting techniques which produce additional decontamination of the alloys from transuranic contamination.

DECONTAMINATION PROCESS - B. Griggs

As reported previously,⁽²⁾ a small section of the Saxton fuel cladding was treated at 600°C in HF for 30 min followed by 1.2 hr in an ammonium oxalate, citrate, fluoride, peroxide solution⁽³⁾ at 85°C to produce an excellent decontamination and film removal. Analyses of the stripping solution and the residual metal has verified that 99.7% of the α activity was removed by the HF-aqueous decontaminant treatment. The residual 0.3% of α activity compares with the 7 nci/g found previously for the base metal which indicates the process achieved as much decontamination as possible by surface removal alone. The slow stream of effluent gas from the 600°C HF treatment was routed through two 1 M KOH absorption solutions in 250 ml flasks followed by an activated charcoal absorber. Following treatment of the piece it was found the effluent tube (1/8 in. S/S), the KOH absorption flasks, and the charcoal were all radioactive to approximately the same level. Gamma analyses of the activity showed it to be almost entirely cesium. This indicates the effluent from the HF treatment will have to be treated for cesium activity removal.

The potential for zirconium oxide removal and fuel particle dissolution with molten salts was tested by treating autoclaved (47 mg/dm² wt gain) pieces of Zircaloy-2 tubing in a molten salt bath of 37 mole % LiF - 37 mole % NaF - 26 mole % ZrF₄ at

660 to 740°C from 1 to 20 min with and without an HF gas sparge. The residual salt was removed from the pieces by treatment in the ammonium oxalate, citrate, fluoride, peroxide solution⁽³⁾ for 30 to 60 min. The corrosion experienced in the process is shown in Table 9. The process apparently produces most of the corrosion in the first 5 min at rates far in excess of those expected.⁽⁴⁾ It would be difficult to treat hulls in a production line fast enough to reduce corrosion to an acceptable level (<25 μm). The pieces were weighed after removal from the fused salt bath in a vertical position and held to drip in this position for 1/4 to 1/2 min before they cooled below 550°C, the temperature at which this salt solidifies. Even with this draining procedure, the salt dragout (see Table 10) would require the purification and retreatment of a significant fraction of the molten salt inventory. The metal which could be dissolved into the salt would amount to about 49% for the 1.08-cm diam, 0.76-mm wall Zircaloy tubing. The variation in the amount of adherent salt appeared related to the amount of corrosion, which would suggest that freezing of the surface layer of salt occurred as the Zircaloy corroded into it. This explanation for the salt adherence has not been verified.

One potential product of densification is an alloy of 85% Zircaloy - 10% stainless steel - 5% Inconel. The preparation, primarily the melting, of this alloy will release absorbed hydrogen and tritium. The approxi-

mately 100 ppm of hydrogen evolved could be reabsorbed into a previously cast ingot of the zirconium alloy intended for storage. The absorption rate and equilibrium pressure of hydrogen over the alloy ingot were studied at representative typical low pressures and low concentration. The absorption temperature was fixed high enough (700°C) that zirconium would dissolve its own oxides and nitrides, which would otherwise impede adsorption. Maintaining a large temperature gradient over the length of the absorption specimen to increase the capacity of the ingot for hydrogen while keeping its equilibrium pressure down, produced similar results to the isothermal absorption specimens if the absorbed hydrogen was assumed uniformly distributed. Absorption rates for Zircaloy-2 and 85% Zircaloy-2 - 10% Type 304 stainless steel - 5% Inconel 718 with grit-blasted surfaces are shown in Figure 4. The relationship of the equilibrium pressures is shown in Figure 5. In general the equilibrium pressures were controlled by the presence of zirconium. The rate of absorption favored the Zircaloy-2. The hydriding of the zirconium alloy always resulted in faster subsequent absorption rates even though the hydrogen was removed before the second absorption. This suggests the development of a preferred path for absorption and diffusion. Both the alloy and Zircaloy-2 absorb hydrogen rapidly enough to be useful for storage of the hydrogen removed from the densified hulls.

TABLE 9. Corrosion of Autoclaved* Zircaloy-2 Samples by Treatment in 37 mole % LiF - 37 mole % NaF - 25 mole % ZrF_4 Salt at Various Temperatures and Times

Sample No.	Salt Temp., °C	Immersion Time	HF Sparge	Metal Loss, mg/dm ²	Metal Loss, μm
-	695	1 min	No	3,300	50
J	695	2 min	Yes	3,890	59
D	700	3 min	No	5,878	90
H	670 - 680	5 min	No	9,099	139
I	720 - 740	5 min	No	9,652	147
C	710	5.5 min	Yes	11,196	171
F	680 - 690	5.5 min	Yes	10,976	168
G	680 - 690	5.5 min	Yes	10,135	155
B	660	20 min	No	10,184	155

*Weight gain during autoclaving averaged 47 mg/dm²

TABLE 10. Salt Adhering to Tubing Samples When Removed From Corrosion Tests Shown in Table 9

Sample No.	Corrosion μm	Sample Weight Before Cleaning, g	Sample Weight After Cleaning, g	Weight of Adherent Salt, g
-	50	2.5223	1.5791	0.9432
J	59	2.6340	1.5811	1.0529
D	90	2.1794	1.4211	0.7583
H	139	2.5250	1.1493	1.3757
I	147	1.9819	1.1073	0.8746
C	171	2.0973	0.9817	1.1156
F	168	2.9630	0.9397	2.0233
G	155	2.6470	1.0156	1.6314
B	155	2.9406	1.0522	1.8884

FIGURE 4. Hydrogen Pressure Versus Time at 700°C

FIGURE 5. Equilibrium Hydrogen Pressure Versus $1000/T^{\circ}\text{K}$

DENSIFICATION PROCESS - R. G. Nelson

Table 11 shows all of the chemical analyses on the melt stock and five of the six graphite melted heats. This completes the analytical results. Most significant are the carbon, hydrogen, oxygen and nitrogen values. 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.⁽⁶⁾ Hydrogen and oxygen showed little or no increase after melting, and nitrogen was slightly raised.

A demonstration of Zircaloy-4 clad hull melting by the Inductoslag process was conducted at the U.S. Bureau

of Mines, Albany, Oregon. The feed material for this melt was reject Zircaloy-4 fuel clad tubing chopped into 1-in. pieces and etched by the ammonium oxalate, citrate, fluoride, peroxide bath similar to that used in the decontamination process. The Inductoslag melting process uses an induction-heated, split, water-cooled crucible, as described by Clites and Beal.⁽⁷⁾ Melting takes place in static, one-third atmospheric pressure helium. A pool of Zircaloy-4 and CaF_2 is melted on a "starting stub" and subsequent charge material vibratorily fed into the top of the crucible as the ingot is being extracted out the bottom of the crucible. Approximately 41 kg (90 lb) of simulated shop-leached cladding residues were melted into an ingot 10 cm (4 in.) diam x ~75 cm (30 in.) long.

Data taken during this run are listed below:

Heat Number Ar-4-431

Starting Stub 6.593 kg Wah Chang

Heat Number 389392 B-5

Zr-4 Chop-Leached

Cladding Residues 42.972 kg

CaF_2 Mixed with

Cladding Residues 1.125 kg

Initial Charge Metal 0.879 kg

Initial Charge CaF_2 0.300 kg

Melting Power 95 kW

Melting Time 2.5 hr

Figure 6 shows the chopped and cleaned PWR cladding used as feed stock for the Inductoslag ingot shown in Figure 7. The scalloped appearance of the ingot sidewall is a result of the split copper crucible design.

TABLE 11. Chemical Analyses - Graphite Remelted Zircaloy-2 -
Stainless Steel - Inconel Alloys

Heat No.	Composition of Ingots, w/o					Melt Stock Composition, w/o		
	85% Zr 2 Alloy		90% Zr 2 Alloy	80% Zr 2 Alloy		Zircaloy 2	304L Stainless Steel	Inconel 718
	8-29-74	9-5-74	9-12-74	9-18-74	9-26-74			
C	0.0100	0.0160	0.0250	0.0160	0.0090	0.0100	0.0420	0.0430
H	0.0024	0.0014	0.0008	0.0060	0.0080	0.0022	0.0080	0.0017
O	0.1060	0.1010	0.0980	0.0980	0.0870	0.1150	0.0050	0.0010
N	0.0380	0.0120	0.200	0.0180	0.0070	0.0050	0.0350	0.0280
Cr	2.80	2.83	2.06	3.76	2.82	0.097	18.06	17.88
Fe	8.15	8.15	5.92	10.33	10.72	0.117	69.2	19.3
Ni	3.59	3.64	2.64	4.76	4.64	0.053	8.62	49.64
Sn	0.49	0.49	0.59	1.32	0.37	1.33		
Zr	84.8	84.9	86.5	78.8	78.2			
Cu	<0.01	<0.01	<0.01	<0.01	<0.01			<0.01
Nb + Ta	0.19	0.27	0.16	0.28	0.25			4.86
Mn	0.15	0.14	0.09	0.20	0.19		1.33	<0.01
Mo	0.20	0.20	0.10	0.26	0.20			2.92
Ti	<0.01	<0.01	<0.01	<0.01	<0.01			1.10
Si	0.069	0.082	0.056	0.108	0.086		0.499	0.238
Al	0.13	0.11	0.17	0.10	0.10			0.33

Rockwell C Hardness

38 31 40 30

Neg 750912-1

FIGURE 6. Melt Stock for Inductoslag Heat Number USBM 431
Chopped and Cleaned Zircaloy-4 PWR Cladding

Neg 751418-1

FIGURE 7. Zircaloy-4 Inductoslag
Melted Ingot Heat
Number USBM 431

Samples were taken from the center and edge of the top, middle, and bottom ingot locations for chemical analyses, metallography, and hardness

testing. Figures 8 and 9 are macro-etched sections taken longitudinally from the top and transversely from the middle ingot locations. The fine-grained area at the top of Figure 8 is the last metal to freeze and represents the extent of the molten pool during the melting process. Figure 10, from the middle center ingot location, is typical of the six areas examined under the microscope and represents the large-grained area of Figure 8.

Neg 752047-5

FIGURE 8. Inductoslag Remelted
Chopped Leached Cladding
Hulls Zircaloy-4

Neg 752047-1

FIGURE 9. Inductoslag Remelted
Chopped Leached Cladding
Hulls Zircaloy-4

Table 12 shows the hardness results and chemical analyses from the six locations on the ingot, along with the analyses of the melt stock and the ingot specifications from which the PWR tubing was originally made. These results show that this ingot is suitable for fabrication and reuse in nuclear components. It appears that melting did not increase

Neg 4K212A "B" Etch 100X

FIGURE 10. As Cast Zircaloy-4 Middle
Center Ingot Location
Inductoslag Melted
Heat Number USBM 431

carbon or hydrogen contents. Nitrogen increased 10 to 50 ppm and oxygen 36 to 430 ppm, although even oxygen is still within specification. These increases may have been due to a furnace leak in the hopper stirring shaft seal. Fluorine has increased 9 to 19 ppm, and the increase (if any) in calcium is below the analytical technique. Iron is below specification in the melt stock, and remelting seems to have lowered it further. Silicon is also low in the melt stock. Hafnium, titanium, and molybdenum are all above specification in the melt stock and are believed to be due to analytical differences between laboratories. The 200 and 300 ppm molybdenum levels in the middle center and bottom edge locations are not explainable. Rechecks will be determined on all out of specification results.

TABLE 12. Chemical Analyses - Inductoslag Remelted Zircaloy-4
Heat Number USBM 431

		Weight Percent						
	Melt Stock	Location						Zircaloy-4 Ingot Specification
		Top		Middle		Bottom		
		Center	Edge	Center	Edge	Center	Edge	
C	0.019	0.017	0.014	0.012	0.014	0.017	0.019	0.0080-0.0270
H	0.0021	0.0023	0.0021	0.0023	0.0022	0.0015	0.0020	0.0025
O	0.0970	0.120	0.131	0.106	0.123	0.140	0.123	0.0900-0.1400
N*	0.0060	0.011	0.011	0.008	0.008	0.008	0.007	0.0080
Fe*	0.1740	0.148	0.130	0.194	0.159	0.129	0.152	0.18-0.24
Cr	0.0920	0.081	0.077	0.100	0.087	0.070	0.082	0.07-013
Sn	1.3600	1.33	1.27	1.44	1.43	1.25	1.32	1.20-1.70
Hf*	0.0065	0.0060	0.0066	0.0072	0.0058	0.0065	0.0061	0.0050
Al	0.0053	0.0040	0.0040	0.0042	0.0037	0.0043	0.0049	0.0075
B	0.00003	0.00003	0.00003	0.00003	0.00003	0.00003	0.00003	0.00005
Cd	<0.00003	<0.00003	<0.00003	<0.00003	<0.00003	<0.00003	<0.00003	0.00005
Co	<0.0005	<0.00005	<0.00005	<0.00005	<0.00005	<0.00005	<0.00005	0.0020
Cu	0.0100	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	0.0050
Mg	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	<0.0010	0.0020
Mn	0.0015	0.0010	0.0010	0.0010	0.0010	0.0010	0.0010	0.0050
Ni	0.0015	0.0015	0.0015	0.0015	0.0015	0.0015	0.0015	0.0070
Pb	0.0010	<0.0005	<0.0005	<0.0005	<0.0005	<0.0005	<0.0005	0.0130
Si*	<0.0030	<0.0030	<0.0030	<0.0030	<0.0030	<0.0030	<0.0030	0.0050-0.0120
Ti*	0.0100	0.0100	0.0075	0.0075	0.0075	0.0075	0.0075	0.0050
V	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	<0.0020	0.0050
Mo*	0.0075	<0.0010	0.0030	0.0200	<0.0010	0.0050	0.0300	0.0050
W	<0.0040	<0.0040	<0.0040	<0.0040	<0.0040	<0.0040	<0.0040	0.0100
Ca	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	<0.0050	0.0030
F	0.0007	0.0016	0.0020	0.0013	0.0014	0.0012	0.0026	
U	<0.00005	<0.00005	<0.00005	<0.00005	<0.00005	<0.00005	<0.00005	0.00035
Th	<0.00035	<0.00035	<0.00035	<0.00035	<0.00035	<0.00035	<0.00035	
Brinell Hardness Number								
Range		172-183		174-185		187-204		200 Maximum
Average		178		181		195		187 Maximum

* Off Specification Results

Hardness values are all within specification except for one of seven tests made on the bottom ingot location. It is interesting to note that hardness decreases from bottom to top ingot locations. Conversations with U.S. Bureau of Mines personnel indicate that this is characteristic of Inductoslag melted material.

Two billets from this ingot have been extruded into a rod 3.6 cm diam by 97 cm long and a tube 3.8 cm OD by 2.5 cm ID by 198 cm long. This material appears to be of excellent quality and suitable for further fabrication, and mechanical and corrosion testing. The fabrication process for the rod and tube was in the following steps:

1. Machine
2. Upset in extrusion container
bare 700°C
3. Machine
4. Clad in copper using
crimped joints
5. Extrude 720°C
6. Nitric acid pickle to remove
copper

The extrusions were performed on a Lowey horizontal 1250 ton, high speed, water accumulator press under the following conditions:

	Ram Speed in./min	Force, Tons
Rod	220	595
Tube	180	705

REFERENCES

1. B. Griggs, Feasibility Studies for Decontamination and Densification of Chop-Leach Cladding Residues, USAEC Report BNWL-1820, Battelle, Pacific Northwest Laboratories, Richland, WA, July 1974.
2. G. J. Dau, Compiler, Nuclear Waste Management and Transportation Progress Report October through December 1974, BNWL-1899, p. 35, Battelle, Pacific Northwest Laboratories, Richland, WA, February 1975.
3. A. B. Meservey, "Decontamination and Film Removal," Decontamination of Nuclear Reactor and Equipment, J. A. Ayers, Editor, The Ronald Press, New York, NY, pp. 143-155, 1970.
4. P. D. Miller, E. F. Stephan, W. K. Boyd, and W. N. Stieglmeyer, The Effect of Heat Treatment and of Salt Composition on the Corrosion of Inor-8 and HyMu-80 During Hydrofluorination, BMI-X-288, Battelle Memorial Institute, Columbus, OH, May 1964.
5. E. A. Gulbransen and K. F. Andrew, "Solubility and Decomposition Pressures of Hydrogen in Alpha Zirconium," J. Metals, vol. 7, Trans. AIME 203, p. 134-44, 1955.
6. B. Lustman and F. Kerze, The Metallurgy of Zirconium, McGraw-Hill Book Company, NY, pp. 236, 450, 1955.
7. P. G. Clites and R. A. Beal, "Inductoslag Melting of Titanium Scrap and Sponge," Proceedings of Sessions 101st AIME Annual Meeting San Francisco, CA, February 20-24, 1972, Light Metals 1972, W. C. Rotsell, Editor.

4.0 PYROLYSIS-INCINERATION OF COMBUSTIBLE ALPHA WASTES

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

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

An installation schedule for the experimental alpha waste facility was prepared in January. The target date for the completion of assembly and initial startup is April 15. Bids for the major components of the facility were reviewed during the quarter. All equipment components are now in-hand.

Ash residue from the pyrolysis-incineration of simulated waste in laboratory studies was converted to glass by John Mendel. Glasses containing 20, 25, 30, 35, and 40 weight percent ash were prepared by melting at 1000°C for 3 hr and at 1200°C for 3 hr. Results from accelerated leach tests showed a significant reduction in leach rate for glass prepared at 1200°C compared to glass prepared at 1000°C.

TECHNICAL PROGRESS

PILOT PLANT STUDIES - R. A. Walter, L. K. Mudge, and R. G. Sullivan

All components have been obtained for the pyrolysis-incineration equipment shown schematically in the previous quarterly report. Assembly of the grate, reactor shaft, distributor, screw feeder, and lock hopper on the support frame is currently under way. An installation schedule for the equipment is shown in Figure 11.

Approximately one month of operation should be available before the end of the fiscal year. The composition of the simulated waste to be used is shown in Table 13. The waste components are now being obtained and run plans are being prepared. A throughput rate of 20 lb/hr should be easily obtainable with the equipment, and rates as high as 100 lb/hr may be possible.

FIGURE 11. Experimental Alpha Waste Facility Installation Schedule

TABLE 13. Estimated Waste Composition To Be Used in Study

Component	Weight Percent
Paper	15
Rags	15
Wood	10
Latex Rubber	5
Hypalon Rubber	10
Neoprene Rubber	10
PVC	20
Polyethylene	15

Data that will be obtained during operation of the equipment include off-gas compositions, particulate concentration in the off-gas, flow rates of all streams, tar production rate, and temperatures of all streams and the reactor. Particulates will be

determined by isokinetic sampling of the gas stream. Part of the particulate load will be tars. Particulates and tars will be characterized as to inorganic ash content. Analysis of the data will allow assessment of channeling and gas cleanup requirements.

ASH FIXATION AND PROCESS COMBINATION STUDIES - J. E. Mendel, H. Hollis and L. K. Mudge

The feed of composition shown in Table 14 was processed by pyrolysis-incineration in laboratory scale studies. Ash obtained in the studies was characterized and converted to a glass.

The ash as received was grayish tan, relatively soft material, with particles ranging from about 1/2 in.

TABLE 14. Feed Composition

Component	Weight Percent
Rubber Tubing	15
Neoprene	15
PVC	20
Tygon	10
Polyethylene	10
Tissue Paper	15
Rags	15

down to fluffy fines estimated at less than 100 mesh. The bulk density was determined:

As received = 0.211 g/cc

After pulverizing to
pass 6 mesh screen = 0.273 g/cc

Tap density of
pulverized material = 0.409 g/cc

Composition of the ash as determined by chemical analysis is shown in Table 15. It is interesting to note that 4% chloride still remains in the ash. The ash composition appears much like the composition of ash from tissue paper as shown by an emission spectrograph analysis of tissue paper ash.

Five glass samples were prepared in which the glass frit:ash ratio was varied systematically from 80:20 to 60:40, typical of the frit:waste ratios being studied in HLW fixation.

The zinc borosilicate glass frit, used 73-1, was the glass frit that has been studied most extensively for HLW fixation. The first melts were prepared by premixing the glass frit and pulverized ash and melting in platinum crucibles for 3 hr at 1000°C followed by furnace cooling. The cooled melts ranged from cream color

TABLE 15. Chemical Analysis of Ash from Pyrolysis-Incineration Process(a)

Ash Constituents	Weight Percent
H ₂ O ^(b)	4.6
SiO ₂	34.2
TiO ₂	16.6
Al ₂ O ₃	17.0
CaO	5.5
CaCl ₂ ^(c)	3.6
CaSO ₄ ^(c)	1.6
NaCl ^(c)	1.8
KCl ^(c)	1.3
BaO, Cr ₂ O ₃ , Fe ₂ O ₃ , La ₂ O ₃ , MgO, NiO, ZnO	0.02 - 2.0 each
B ₂ O ₃ , Co ₃ O ₄ , CuO, MnO ₂ , MoO ₃ , SnO ₂ , ZnO ₂	Trace amounts

a. Run details were given in BNWL-1861.

b. Adsorbed water released at 110°C.

c. Arbitrary assignments of 4.0% Cl and 0.4% S were found by analysis.

to a mottled tan as the ash content increased. The melts were not true glasses but contained large quantities of crystalline residue; however, they were coherent and nonporous, which is a major goal. Determination of such properties as viscosity will have to wait until more ash sample is available.

The samples were leach tested by the standard accelerated procedure, the Soxhlet test. Leachability was not a function of ash content, as shown in Table 16. For all 5 melts, the leach rate was about a factor of 10 higher than is obtained with the reference HLW glass. The glasses were remelted for 3 hr at 1200°C and

TABLE 16. Results of Accelerated Leach Tests

Frit:Ash Ratio	Soxhlet Leach Test, Wt% Loss	
	After Melting at 1000°C, 3 hr	After Melting at 1200°C, 3 hr
	After 72 hr	After 72 hr
80:20	5.2	1.2
75:25	7.0	2.0
70:30	6.9	1.8
65:35	6.0	1.7
60:40	5.6	2.7

furnace cooled. Results indicate that a considerably reduced leachability was achieved by using the higher melting temperature (see Table 16).

Volume reduction factors were calculated using the density of the glasses and taking the density of the initial ash to be 0.4 g/cc. Results are shown in Table 17.

TABLE 17. Ash:Glass Volume Reduction Factors

Frit:Ash Ratio	Glass Density, g/cc	Volume Reduction Ash to Glass
80:20	3.0242	1.51
75:25	3.1856	1.99
70:30	3.2318	2.43
65:35	3.3166	2.90
60:40	3.5098	3.51

The preliminary experiments on ash fixation indicate that large amounts of typical ash can be incorporated in a monolithic "glass" form; however, much more development will be required. For instance, it is important that the actual leach rate of plutonium from the glasses be determined before evaluation is carried much further. A basic problem is the chloride content of the ashes. No reference examples of chloride-containing (above fractions of 1%) glasses are known. Thus, the behavior of chloride in glasses will have to be studied in much more depth.

It may be possible to operate the pyrolysis-incineration unit as a slagging gasifier such that, by addition of the glass frit with the waste feed, glass may be obtained directly from the unit. A need for a separate glass making plant could thus be eliminated since the glass plant and incinerator would be combined. No significant incentive for other processing combinations has been identified to date.

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

Three types of tritiated polymers have been prepared and examined. Leach tests show that the tritium is fixed in the polymer structure. One of these is prepared by direct hydrogenation using HT, the others via the HTO route. Several other concepts for polymer formation are discussed. Rough cost data are provided, and a comparison is made with other storage methods on the basis of kg's of material required per kg of water.

An experimental problem which may have prevented selective isotopic excitation by the infrared laser was discovered. Radio-frequency noise from the flash triggering system was interfering with synchronization of the laser pulse such that maximum energy overlap was not occurring. This has been remedied.

The contract with General Electric (San Jose) for the water recycle study has been signed, and work is under way.

TECHNICAL PROGRESS

TRITIUM FIXATION - J. A. Franz and L. L. Burger

Polymer Preparation

Fixation of tritiated water into a bakelite polymer was described in a previous report.⁽¹⁾ Equations (1) to (4) depict the sequence involved.

Step 1 is optional and, if used, the process requires the calcining of the calcium hydroxide to recover the tritium. The conversion of acetylene to acetaldehyde provides a monomer more readily polymerized than acetylene itself. Requirements for the purity of acetaldehyde are low, and the degree of conversion to polymer is

high (97%). Although the conversion of acetylene to acetaldehyde requires a carefully monitored catalyst medium, the industrial process is well established and proceeds in nearly quantitative yields.⁽²⁾

The copper catalyzed condensation of acetylene and HCN produces acrylonitrile.⁽³⁾ The radical-induced emulsion polymerization of acrylonitrile produces polyacrylonitrile.⁽⁴⁾ Equations (5) to (7) depict this process.

This process was also discussed in the previous progress report.⁽¹⁾

Again, a possible option (with some disadvantages) exists of adding the tritium via HCN rather than by step (5).

A disadvantage of this industrial process is the requirement of relatively pure acrylonitrile monomer to achieve high yields in polymerization.⁽³⁾ Specifically, an impurity (divinylacetylene) produced in step (6) must be removed by an extraction sequence. This purification step may introduce an unacceptable level of complexity to the process. In any case, pilot demonstration will be necessary to determine practical efficiencies of conversion of acetylene to acrylonitrile, which vary from 60 to 95% in literature descriptions,⁽³⁾ and to accurately determine costs. We have prepared tritiated acrylonitrile in our laboratory using

a Nieuwland catalyst mixture resembling the intended industrial procedure and have polymerized it under conditions of emulsion polymerization for leach testing in this study.

One of the simplest procedures for incorporating tritium in a polymer is the electrolysis of tritiated water and the catalytic hydrogenation of polystyrene to form polyvinylcyclohexane. Equations (8) and (9).

The simplicity of the one-step process is particularly attractive because of the relative ease of containment of the tritium and the absence of by-products and waste streams. These advantages probably outweigh the apparent disadvantage of the high-pressure hydrogenation process. In our laboratory we have carried out the hydrogenation of polystyrene in the presence of a nickel-aluminum alloy catalyst at 160° under 60 atm hydrogen pressure.⁽⁵⁾

Leach Tests

Bakelite was tested by soaking the crude polymer exhibiting $2,874 \text{ d g}^{-1} \text{ min}^{-1}$ in water. An immediate loss of tritium to the wash solution due to surface contamination of the polymer occurred, but no further increase occurred on soaking for a longer time. When a sample of the bakelite was first rinsed, extended soaking in a rinse solution resulted in no detectable tritium loss to the rinse solution after 6 weeks of rinse. Combustion of the polymer and analysis of

water confirmed the tritium level of the polymer. Similarly, polyacrylonitrile gave no initial loss of tritium due to surface contamination. However, following a rinse no loss of activity to a rinse solution over an extended period of time (6 weeks) was detectable. Hydrogenated polystyrene did not exhibit an initial release and did not undergo detect-

able leaching of tritium. The tritium content of polyacrylonitrile and hydrogenated polystyrene was also determined by analysis of water from combustion of the polymers.

Table 18 summarizes results of leach tests of tritiated polymers. These results clearly indicate that all three polymer types adequately fix tritium.

TABLE 18. Leaching Data

Polymer	T Content ^(a)	Total T Content Exposed per ml Rinse Solution	Average Rate of Increase of T Content of Rinse Solutions ^(b)
Bakelite	2874 d min ⁻¹ g ⁻¹	1430 d min ⁻¹	0.0 ± 1.2
Poly acrylonitrile	7650 d min ⁻¹ g ⁻¹	5130 d min ⁻¹	0.0 ± 2.0
Hydrogenated Polystyrene	1000 d min ⁻¹ g ⁻¹	1200 d min ⁻¹ ^(c)	0.0 ± 2.0 ^(c)

a. Determined from combustion analysis of polymer and scintillation counting of resulting water.

b. Average of the sum of the change in tritium detected between tests divided by elapsed time between tests. $[\Delta(\text{d min}^{-1} \text{g}^{-1})/\Delta t(\text{days})]$

c. Leach tests of hydrogenated polystyrene are still under way.

Cost Estimates for Polymers

Table 19 presents cost estimates for the polymers. The figures are based on minimum approximate material costs taken from Chemical Marketing Reporter.⁽⁶⁾ They assume approximately 100% conversion of calcium carbide to polymer in the first two cases. For polyacrylonitrile, costs of synthesis and purification are more difficult to estimate. We note that bulk quantities of acrylonitrile are available for \$0.23/lb commercially. The figures are probably accurate to within a factor of 2. More accurate figures would require

pilot plant tests of the processes. The cost cited for the polystyrene process is probably closest to the real process cost.

TABLE 19. Cost Estimates for Polymers

Medium	Cost Per Lb Water Disposed	Wt. of Polymer Per Lb Water Disposed
Bakelite	\$1.00 ^(a) 0.60 ^(b)	2.80
Polyacrylonitrile	0.74	2.94
Hydrogenated Polystyrene	1.25 ^(c)	3.8

a. Using resorcinol.

b. Using phenol.

c. Includes cost of electrolysis of water.

Future Polymer Work

Several other unique approaches are being considered. Highly attractive are the mild reductions carried out with tris(triphenylphosphine) rhodium chloride as a catalyst. The scheme below shows the tritiation of inexpensive⁽⁶⁾ styrene using tritium from the electrolysis of water, and a rhodium catalyst, followed by polymerization of the tritiated ethyl benzene with paraformaldehyde to form polymer 3 for storage. The reaction of terminal alkenes such as styrene proceeds

rapidly at ambient or near-ambient conditions of temperature and pressure. Procedures for the preparation and use of rhodium catalysts for terminal alkenes are well established.⁽⁷⁾ Reduction of styrene has also been done under ambient conditions using $\text{Co}(\text{CN})_5^{3-}$.⁽⁸⁾ Conditions for the polymerization of substituted benzenes with paraformaldehyde are well established.⁽⁹⁾ The availability of highly efficient homogeneous reduction systems suggests that rubberlike polymers such as polyisopropene and polybutadiene possessing olefin linkages could be hydrogenated directly.

Among several systems for efficient hydrogen gettinger not yet explored is the easy reduction of methyl pyridinium salts under 1 atm H_2 and ambient temperature.⁽¹⁰⁾ Thus, synthesis

of monomer 4 provides a substrate which could be mixed with palladium catalyst

in a solid or viscous phase or in solution. Exposure to tritium would give compound 5, which could be separated from the catalyst and polymerized with a bifunctional amine (1,6-diaminohexane) or disposed of directly. The latter methods will necessarily have higher material costs than those previously studied.

Comparison of Fixation Methods

Many different fixation schemes have been proposed. They include hydrate formation, sorption by clays, molecular sieves, and other solids, cements, storage of liquid H_2O in cylinders, organic polymers, metal hydrides, and storage of hydrogen gas. Each has its own set of problems which may range from the requirement of high integrity containment for water, to high exchange rates

with concrete, to limited high temperature stability for organic compounds and metal hydrides. A discussion of these points and a comparison with other disposal schemes, such as deep well injection, will not be made here. However, one aspect of storage, the volume or weight of material required to fix a kg of water is important, and rough estimates can easily be made. These are listed in Table 20 for several storage schemes. No containment is assumed for the metal hydrides or the organic compounds. The additional weight would be minimal. The containment for water assumes the critical volume is required and a container to withstand the critical pressure (3226 psi, 218.3 atm). Specified storage or transportation limitations thus determine the complexity of the storage system.

TABLE 20. Tritium Storage Efficiency

Form	G Atoms/G	G/G H ₂ O	Comments
H ₂ O	0.11	1.00-	3.5 g/g for 18 kg in type 1H cylinder
H ₂	1.00	0.11	28 g/g C 2200 psi in type 1A cylinder
UH ₃	0.014	7.9	Common experimental storage method
LiH	0.13	0.85	Compound very reactive
ZrH ₂	0.021	5.29	{ Zr and Ti hydrides are somewhat less reactive than parent metals. }
ZrH	0.011	10.6	
TiH ₂	0.036	3.08	
LaNi ₅ H ₄	0.0087	12.8	Typical intermetallic compound
Portland cement	0.02	5.0	Assuming 20% H ₂ O
Molecular sieve	0.02	5.0	Assuming 20% H ₂ O
Bakelite	0.040	2.8	
Polyacrylonitrile	0.038	2.9	
Hydrogenated polystyrene	0.029	3.8	
Phenyl propargyl ether, 20% on support	0.0060	18.6	
Polymerized acetylene	0.076	1.4	

LASER EXCITATION EXPERIMENTS -

T. J. Whitaker and J. R. Morrey

Early experiments had indicated that no HD, D₂, or H₂ was formed when HDO vapor (from a 50% D₂O/50% H₂O solution) was bombarded with infrared radiation from a tunable Nd:YAG laser and simultaneous ultraviolet radiation from two xenon flash-lamps. It was felt that the UV radiation from the flash-lamps was not intense enough to give measurable results at the high frequency required to pump HDO molecules from the 110 excited vibrational level to the dissociation band. The wavelength of the radiation from the laser was changed to excite ground state HDO molecules to the higher energy 011 transition. The lower frequency radiation required from the flash-lamps to pump HDO molecules from this excited state to the dissociation band would be in a region of higher flash-lamp intensity.

However, experiments using the 011 transition still did not show any HD, D₂, or H₂ formation. An analysis of the initial experimental conditions revealed sizable quantities of oxygen from air in the sample cell. The computer kinetics model developed earlier was used with a new initial oxygen concentration. The result indicated that high concentrations of oxygen in the initial reaction mixture would suppress formation of HD, D₂, and H₂. To avoid this problem, the sample cell was loaded with HDO

vapor in a vacuum system. New experiments using this loading technique and only the flash-lamps were performed. These did show HD formation and even more H_2 formation. This indicated that the flash-lamps do have enough intensity to dissociate H_2O , D_2O , and HDO excited molecules.

Only a few of the lower vibrational excited states populated to any significant extent at room temperature. The relative populations of these states and the fact that HD was formed in detectable quantities imply that the flash-lamps currently being used can pump HDO molecules from the 110 vibrational state to the dissociation band. Thus, because of the larger absorbance of HDO at this wavelength ($\sim 2.37 \mu m$) and because the 110 excited state has much of the excitation energy in the O-D stretch, the laser has been set to excite HDO molecules to this 110 vibrational state. However, the experiments using the vacuum loading technique have revealed a problem. Because some of the lower vibrational states of H_2O , D_2O , and HDO are appreciably populated at 370°K, the enrichment factor could suffer. This "hot band" problem is being considered, and experiments at room temperature are planned to lessen the effect. The kinetics model is being revised for room temperature HDO dissociation and to account for diffusion in the sample cell.

An experimental problem was discovered recently. In the process of

synchronizing the flash-lamps with the laser pulse, radiofrequency noise from the flash-lamp triggering mechanism was adversely affecting the Q-switch on the laser. This prevented proper overlap of the flash-lamps with the laser. The intensity peaks had appeared to overlap due to the stretching of the peaks by the detectors being used. The problem has been remedied by changing the grounding and by the use of pulse transformers in the triggering cable. Photodiodes are now used as detectors for the synchronization of intensities.

WATER RECYCLE STUDIES - L. L. Burger

A contract with General Electric Company at San Jose has been signed under which they will study the feasibility of increased water recycle in a reprocessing plant. The study scope will include:

1. The definition of the probable distribution of tritium within a reprocessing plant of the scale required for viable operation in the late 1980s.
2. The assessment of the probable effectiveness of tritium retention for the range of steady-state and transient operating conditions that could be anticipated.
3. The evaluation of the impact of the tritiated water recycle on the design and operational requirements for such a plant.
4. The preparation of a report summarizing the results and conclusions of the above-mentioned tasks.

REFERENCES

1. Nuclear Waste Management and Transportation Quarterly Progress Report October-December 1974, BNWL-1899, Battelle, Pacific Northwest Laboratories, Richland, WA, March 1975.
2. J. W. Copenhaver and M. H. Bigelow, Acetylene and Carbon Monoxide Chemistry, Reinhold Publishing Corp., New York, NY.
3. M. A. Dalin, I. K. Kolchin and B. R. Serebryakov, "Acrylonitrile," Progress in Materials Science Series, Volume VI, Technomic Publishing Company, Westport, CT, pp. 18-48, 1971.
4. W. R. Sorenson and T. W. Campbell, Preparative Methods of Polymer Chemistry, Interscience Publishers, New York, NY, p. 236, 1968.
5. Alternative conditions for the hydrogenation of polystyrene have been reported: Ger. Patent 1,131,885, Chem. Abstr., vol. 57, p. 12736d, 1962.
6. Chemical Marketing Reporter, vol. 207, no. 4, Schnell Publishing Co., New York, NY, Feb. 3, 1975.
7. E. L. Muettert, Ed., Transition Metal Hydrides. The Hydrogen Series, Marcel Dekker, New York, NY, 1971. For leading references see also P. S. Hallman, D. Evans, J. A. Osborn, and G. Wilkinson, Chem. Comm., no. 7, p. 305, 1967; P. S. Hallman, B. R. McGarvey, and G. Wilkinson, J. Chem. Soc., A, 3143 1968; D. Rose, J. D. Gilbert, R. P. Richardson, and G. Wilkinson, J. Chem. Soc., A, p. 2610, 1969; J. Ogata, R. Iwata, and Y. Ikeda, Tetrahedron Letters, no. 34, p. 3071, 1970; L. Horner, H. Siegel, and H. Büthe, Angew. Chem. Intern. Ed. Engl., vol. 7, p. 942, 1968.
8. A. Zakhariev, Ivanova, V., Shapov, D., Dokl. Bolg. Akad. Nauk, vol. 2412, pp. 211-14, 1971.
9. R. Wegler, Angew. Chem., vol. 60, p. 88, 1948.
10. E. Wenkert et al., J. Org. Chem., vol. 33, p. 1287, 1968.

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

Development and evaluation have continued on geophysical techniques for detection and mapping of waste materials. Various survey techniques have accurately located buried materials in and adjacent to disposal trenches. Metal detector, thermal infrared, magnetometer, acoustic transmission, acoustic refraction, and acoustic reflection techniques have been field-tested and evaluated. Ground-penetrating radar systems have been identified as a promising technique.

Numerous drilling procedures, equipment, and techniques have been evaluated for use in drilling of contaminated burial ground facilities. Currently, a small-diameter drill system is being used to collect sediment samples from various locations adjacent to the 300 North Burial Ground.

Two instrumented wells have been installed at the 300 North Burial Ground. These wells will be used for neutron moisture and gamma density probe access. Sediment samples are currently being characterized as to various physical parameters concerning fluid transport. Computer codes have been written for analysis of

physical parameters delineated by various laboratory techniques.

Computer models for partially-saturated flow have been evaluated which will predict nonisothermal fluid migration variables. Meteorological and physical data collected under other tasks will supply necessary input and boundary conditions for these models.

Burrowing rodent populations in and adjacent to burial ground facilities are being evaluated. Burial ground and control populations are being evaluated as to accessibility to contaminated waste materials.

TECHNICAL PROGRESS

TASK 1 - REVIEW OF HISTORICAL

RECORDS - A. E. Reisenauer

Draft completed second quarter.

TASK 2 - WASTE DETECTION AND

MAPPING - G. A. Sandness

In the past quarter, geophysical techniques have been developed and evaluated for detecting and mapping buried wastes and waste containers. In addition, a metal detector survey of the 300 North Burial Ground has been completed. The results show significant errors in the present map of that burial ground.

Current work is focused on the refinement of acoustic and magnetic techniques and on the testing of ground-penetrating radar.

The detection and mapping techniques studied are described below:

Metal Detector

A commercial metal detector was acquired, tested, and used in a survey of the 300 North Burial Ground. A grid was established in the burial ground before the survey was conducted. The line spacing in the grid is 50 ft, and the intersections are marked by wooden stakes.

The results of the metal detector survey are shown in Figure 12. This figure was made by overlaying the available map of the 300 North Burial Ground (AEC drawing No. H-E-9982) on an aerial photograph of the burial ground. The white lines were painted on the ground to delineate buried metal objects observed by the metal detector. The black lines were taken from the map and show the trench boundaries, storage drum locations, and the perimeter fence. Several discrepancies are obvious in comparing the survey results with the existing map. The most striking examples are found at the north and south ends of the burial ground where large masses of buried metal extend well outside the trench boundaries as shown on the map.

This survey shows that there are significant errors in the existing map of this burial ground.

The metal detector was particularly effective in locating the buried oil drums and pipes in the

area's southwest corner. It is also effective in showing the presence of large metal objects or masses in trenches. However, other techniques are needed to delineate trench boundaries.

Magnetometer

Magnetic field measurements can potentially be used to locate buried ferrous objects such as tools, steel containers, steel scrap, etc. However, detailed data interpretation is difficult, particularly in areas of large concentrations of these objects. Preliminary field experiments indicate that magnetometer surveys may be useful as a reconnaissance method.

In addition, a simple computer modeling technique has been devised for estimating the size, location and depth of isolated ferrous objects from magnetometer survey data. These results would supplement those obtained with a metal detector.

Thermal Infrared Imagery

Under certain conditions it is possible to identify features such as filled trenches in thermal IR imagery. Distinctive temperature, thermal emissivity or thermal inertia patterns may exist in excavated areas due to changes in soil property and moisture content.

To test this technique an aerial thermal IR scanner survey was conducted over the 300 North Burial Ground. Analysis of the imagery is still incomplete. But preliminary results indicate that vegetation may be a dominant factor in producing observed thermal patterns.

FIGURE 12. An Overlay of an Aerial Photo of the 300N Burial Ground and the Existing Map of that Burial Ground. The white lines were painted on the ground to show the outlines of metallic objects found with a metal detector. The black lines show the mapped trench boundaries, storage drum locations, and the perimeter fence.

Acoustics

Acoustic techniques offer the possibility of directly observing trench boundaries and buried material within the trenches. Variations in acoustic properties due to differences in material, sediment porosity, particle orientation, and moisture content are observable, in principle, as reflective interfaces, changes in acoustic velocity, and variations in mechanical impedance. But the development of practical techniques for utilizing

these effects to detect and map buried wastes are at or beyond the state-of-the-art. This is because the sand and gravel comprising the ground material in the burial grounds exhibit highly unfavorable energy transmission and coupling characteristics. Combined with unusually short acoustic travel times and a need for relatively high resolution, these characteristics cause significant difficulties. Nevertheless, we believe that an acoustic technique can

be developed which will be a powerful tool for the detection and detailed mapping of buried wastes.

Acoustic transmission, refraction, and reflection have been measured in the 300 North Burial Ground to determine acoustic velocities, to identify reflecting and refracting layers, to test sound sources and receivers, and to develop survey and data processing techniques. Initial results have shown the existence of an acoustic interface, possibly related to the vertical soil moisture distribution, at a depth of approximately 5 ft. The compressional acoustic velocity above this interface varies between 400 fps and 650 fps, depending on location within the burial ground. The compressional velocity below the interface is typically 1250 fps.

Figure 13 is an acoustic reflection profile along a 50-ft-long traverse line within the 300 North Burial Ground. The data represent acoustic signals received from reflecting interfaces in the ground. These interfaces and a trench wall are shown by the solid and dashed lines, respectively. The area to the left of the dashed line is believed to be a trench containing waste material. This figure is an example of results obtained with an initial version of an acoustic reflection profiling technique.

Ground Penetrating Radar

Extensive discussions with radar manufacturers and research laboratories indicate that specially designed systems may be effectively

used to locate and map buried materials. It is well known that electromagnetic waves are reflected by electrical conductivity contrasts or interfaces within the ground. Such interfaces can correspond to soil moisture or soil density variations as well as to material changes in the ground. A ground-penetrating radar system with appropriate display capabilities could provide an effective means for detecting and mapping buried wastes. The major advantage of a radar system over an acoustic system is that radar requires no physical coupling to the ground. A radar system could, therefore, be truck-mounted for a highly efficient and rapid survey capability.

A radar of the required type is, again, a state-of-the-art device. Specialized, short-pulse systems are needed for short-range object detection and for adequate ground penetration. The performance of such a system under the conditions existing in the Hanford burial grounds is difficult to predict.

For this reason, discussions are currently under way with two radar research laboratories to arrange a field test of a prototype ground penetrating radar system.

The current studies of magnetic and thermal IR survey techniques will be completed. However, further work will focus primarily on development and testing of acoustic and radar systems.

Pending the results of the forthcoming field tests of a radar system, a major immediate effort will be made

FIGURE 13. An Acoustic Reflection Profile in the 300N Burial Ground. The dots represent acoustic signals received from reflecting interfaces in the ground. The solid and dashed lines are interpretations of the data--the solid lines showing interfaces and the dashed line showing one wall of the trench. The area to the left of the dashed line is assumed to be a filled trench. A painted circle (metallic object) is crossed at the 30 ft point on the traverse.

to complete needed measurements of the acoustic properties of the burial grounds and to develop an effective technique for acoustic imaging. Acoustic holography is potentially such a technique.

Completion of this subtask will include the specification, acquiring and testing of an operational hardware system for waste detection and mapping.

TASK 3 - DRILL SAMPLING WELLS AND ANALYZE SAMPLES - V. L. McGhan

Drilling operations adjacent to burial grounds continue in order to

define stratigraphic and fluid transport parameters. Continuous sampling with split tube core barrel drilling techniques has acquired more than 64 individual sediment cores. Continuous logging of each drilled hole has indicated a relatively moist zone <15% H₂O by volume to approximately 20 ft and a drier zone <5% by volume from 20 ft to greater than 38 ft.

Sediment in the general area of the 300 North Burial Ground facility is composed of deep, very dark grey coarse sands. The texture ranges from sand to loamy sand with <5% clay fraction.

These sediments occur on gently to moderately sloping terrain. The topography is hummocky and occurs in large longitudinal dunes within terrace deposits of glacial origin.

Specialized drilling techniques have been developed for use in unconfined sediments. A split tube core barrel, drill rod, and small-diameter test well casing are extensively used. Drilling small-diameter test wells using a falling mass technique has proven effective and reliable for small-scale portable research operations.

TASK 4 - INSTRUMENTED WELLS AND SOIL CHARACTERIZATION - S. J. Phillips

The migration of meteoric waters and contaminated radionuclide wastes through the vadose zone in burial grounds is being evaluated. Two instrumented wells have been installed to depths of 32 ft. These wells allow access of neutron and gamma probes used in monitoring soil moisture and density, respectively. In addition, thermocouple psychrometer and thermocouple sensors have been fabricated for in situ installation in access wells. These sensors will monitor the relationships of matrix potential and temperature to the various depths. Laboratory characterization of sediment samples collected from access wells is continuing for determination of 0.10, 0.33, 0.66, 1.0, 5.0, 10.0, 15.0 -bar matrix potential, particle density, bulk density, relative hydraulic conductivity, and fluid content variables. A computer data display and analysis pro-

gram has been written for fluid retentivity, hydraulic conductivity, air entry pressure, fluid content, and analytically determined physical parameters. This program uses an unsaturated hydraulic conductivity (fluid desorption) function represented by

$$\psi_e = \psi_e(\theta/\theta_s)^{-b}$$

where θ is the analytically determined saturated fluid content, ψ_e is the air entry pressure, and b is an empirically determined constant. The hydraulic conductivity function at any value of saturation is then

$$k = k_s(\theta/\theta_s)^{2b+3}$$

where k_s is the saturated hydraulic conductivity.

TASK 5 - EVALUATION OF MOISTURE MOVEMENT MECHANISMS AND MODELING -

H. P. Foote

Computer programs for partially-saturated flow have been evaluated. These programs simulate long-term environmental changes of waste migration rates. They will use field meteorological data and laboratory-determined physical parameters delineated through other tasks.

We are continuing to modify existing computer codes to include nonisothermal flow. In situ nonisothermal conditions found in burial ground facilities have required meteorological, topographic, and temperature gradient data input to accurately simulate fluid transport.

TASK 6 - EVALUATION OF BIOLOGICAL
TRANSPORT MECHANISMS - W. H. Rickard

Burrowing rodent populations are being studied in and adjacent to burial grounds. This analysis will allow quantitative determinations of rodents inhabiting underground burrows potentially contaminated with radioactive waste products. Rodents

are captured within the confines of burial grounds during the nocturnal cycle using developed trapping techniques. In addition, a control population is captured at a specific distance from the burial grounds. The rodent populations of different burial grounds may reflect the accessibility of contamination relative to species, age, and mode of habitat.

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

Progress on the D&D at Hanford study is summarized in Table 21. The contaminated liquid disposal sites have been classified on the basis of radionuclide content; data sheets summarizing available information for these sites have nearly been completed; general disposition alternatives have been identified; preliminary release criteria have been established; and a methodology for setting D&D priorities has been defined.

The D&D Information Center now includes approximately 600 documents and books. The computer printout of the catalogued references has been updated with the receipt of additional material.

In planning for disposition of the 233-S Building as a D&D demonstration, outlines have been prepared for the six activity specifications required, and volumes of waste materials from the project have been estimated. Relative risk and cost estimates were prepared for the five 233-S disposition alternatives.

TECHNICAL PROGRESS

TASK 1 - DEVELOP METHODS, COSTS, AND PRIORITIES OF D&D AT HANFORD

SUBTASK 2 - SITE CHARACTERIZATION - B. W. Mercer

Work continued on compiling characterization data and preparing descriptive information for D&D at Hanford. The scope of this work has been expanded to include active and standby contaminated facilities. Preliminary drafts of generic descriptions of the Retention Basin Systems, Contaminated Liquid Disposal Sites, and Transuranic Facilities have been completed.

Contaminated liquid disposal sites including cribs, trenches, ponds, ditches, and unplanned releases were classified according to their radionuclide content, to simplify considerations of disposition alternatives. Cribs will be classed as transuranic sites if discharge of $>0.04 \text{ g Pu/m}^2$ of crib bottom area has occurred. This limit was set on the basis of a) the regulation requiring retrievable storage for soil contaminated

TABLE 21. Status: Study of D&D at Hanford

Element of Study	Transuranic Facilities	Retention Basin Systems	Liquid Disposal Sites	Fuel Storage Basins	Solids Disposal Sites	Reactor Buildings	Gas & Exhaust Air Systs.	Fuel Reproc'g Facilities	Uranium Facilities	Waste Management Facilities	Laboratories	Miscellaneous
Facility Description (BW Mercer)	C	C	C									
Site Characterization (BW Mercer)	C	C	C									
Disposition Alternatives (MR Kreiter, GE Koester)	C	C	C	C	C							
Procedures & Techniques (HT Fullam)	C	C										
Waste Disposal (HT Fullam, MR Kreiter)												
Evaluation of Alternatives	Administrative Controls Required (CE Jenkins)											
	Safety Analysis (CE Jenkins)											
	Cost Estimate (JC King)											

Note: "C" denotes completion this quarter. "CX" denotes completion in prior quarters.

with plutonium at levels >10 nCi/g of soil, and b) the assumption that the bulk of the plutonium added to the crib is in the top 6 in. of soil. Classed as fission product sites are cribs into which sufficient beta activity was discharged to leave >0.04 Ci/m² (as of December 31, 1973). This limit was set on the basis that above it consideration must be given to: 1) radiation exposure if excavation is planned to remove transuranic contaminated soil and 2) preventing dispersal of the radioactivity by plants, animals, or natural erosion. The site classifications are as follows:

- I. Transuranic Sites (>0.04 g Pu/m², <0.04 Beta Ci/m²)
 - A. Acidic High-Salt Process Wastes
 - B. Neutral-Basic Wastes
- II. Transuranic-Fission Product Sites (>0.04 g Pu/m², >0.04 Beta Ci/m²)
 - A. High-Salt Wastes
 - B. Low-Salt Wastes
- III. Fission Produce Sites (<0.04 g Pu/m², <0.04 Beta Ci/m²)
 - A. High-Salt Wastes
 - B. Low-Salt Wastes
- IV. Low-Activity Sites (<0.04 g Pu/m², <0.04 Beta Ci/m²)
- V. Reactor Area Sites

- A. Rupture Debris Cribs and Trenches
- B. Laboratory and Decontamination Wastes

VI. Ponds and Ditches

The above classifications were picked on the following assumptions concerning eventual requirements for disposition of this type of facility:

- a. Soil containing more than 10 nCi Pu/g may have to be mined and placed in retrievable storage or processed for ultimate disposal. Class I facilities, with little beta-gamma activity, could be handled in the near future while it may be advisable to delay significant effort with Class II sites until the fission products have decayed.
- b. Disposition of 200 Area Class III sites containing gross quantities of fission products but less than 10 nCi Pu/g may require nothing more than stabilization of the site in some manner to prevent transport into the environment until the fission products have decayed. It is anticipated that low-activity sites (Class IV) will require no treatment other than maintenance to avoid industrial hazards and that Class VI sites (ponds, etc.) will require only application of a protective soil cover to prevent dispersal of surface contamination.
- c. The 100 Area sites (Class V) may require special treatment, depending on the results of further characterization studies.

These assumptions have not yet been evaluated and have been used thus far only in selecting site classifications.

SUBTASK 3 - DISPOSITION -

M. R. Kreiter

Disposition Alternatives -

M. R. Kreiter and G. E. Koester

The various classes of Hanford contaminated facilities and sites have been reviewed and alternative disposition strategies and activities identified. In general, the alternatives being considered for the various classes of facilities are variations of those defined as follows for the 100 Area retention basin systems:

- a. Prepare the system and its surroundings for interim (up to 20 years) maintenance and surveillance. Objective: delay any extensive D&D until ultimate waste policy has been established for the Hanford reservation.
- b. Immobilize the contamination in place. Objective: prepare the system and immediate surroundings for long-term unrestricted access but restricted use.
- c. Remove all "Hazardous" contamination. Objective: clear the site of transuranics and any other radionuclides found by risk analysis to pose hazards to the public, in order to allow unrestricted use of the site as soon as the short-lived isotopes ($t_{1/2} \leq 30$ years) have decayed to insignificant levels.
- d. Remove all detectable contamination. Objective: allow unrestricted use of the site.

Criteria Development - J. M. Selby and E. E. Oscarson

A detailed evaluation of existing and proposed limits was undertaken with the goal of suggesting reasonable release criteria for Hanford D&D projects. A literature search was completed to identify studies and proposed guidance which might be useful in developing criteria. About 30 references were studied in this effort.

Figure 14 graphically shows the evaluated criteria for surface contamination. Considering the way the criteria are distributed, the values of the U.S. NRC Division of Materials and Licensing⁽¹⁾ appear to be as appropriate as any other suggested limits for release of contaminated surfaces in D&D activities at Hanford. For plutonium contamination the limits would be 500 dpm alpha/100 cm² average, and 2,500 dpm alpha/100 cm² maximum for fixed contamination, and 100 dpm alpha/100 cm² for removable contamination.

To date, little has been written on release limits for contaminated solids or liquids. Figure 15 contains a compilation of the proposed criteria we were able to find. As is apparent, there are no readily acceptable criteria which directly apply to both solid and liquid contamination. Consequently, it is suggested that the NCRP-ICRP nonoccupational water standard of 5 pCi/g ($\approx$ 10 dpm α /g) for soluble plutonium be applied to all transuranics in liquids. Because the pathways from soil to man are usually less critical than those from water to man, we are proposing that solid contamination release criteria for

FIGURE 14. Release Criteria for Contamination of Surfaces

FIGURE 15. Release Criteria for Contamination of Liquids and Solids

transuranics be the same as originally proposed in BNWL-1697,⁽¹⁰⁾ i.e., 10 pCi/g.

Considering the smaller relative hazards generally associated with beta-gamma emitting isotopes, release criteria tenfold higher than for plutonium appear to be in order. These limits would be 50 pCi/g liquid and 100 pCi/g solid. Table 22 is a detailed compilation of these criteria.

TABLE 22. Proposed Radioactivity Concentration Levels for Unrestricted Release

Isotope	Solid	Liquid
Transuranic Isotopes	10 pCi/g (≈ 20 dpm α /g)	5 pCi/g (≈ 10 dpm α /g)
Beta-Gamma Isotopes	100 pCi/g (≈ 200 dpm/g)	50 pCi/g (≈ 100 dpm/g)

There are problems associated with this broad approach to release since it does not attempt to distinguish among nuclides. For example, such an approach would suggest that on this basis criteria for ^{90}Sr contaminated liquids would be 50 pCi/ml--considerably higher than the Nonoccupational Concentration Guide of 0.3 pCi/ml.

To assure acceptability of use of the broad criteria, exposure pathway analyses must be performed and must indicate that all suggested limits would result in negligible exposure to population regardless of subsequent land use. Assumptions as to particulate size and chemical state, as well as any other assumptions on usage patterns, agricultural practices, etc., should also be factored into the evaluation.

Priority Development - J. C. King and J. W. Litchfield

A decision methodology permitting use of multiple probabilistic measures of performance is being adapted for assigning priorities for disposition of retired Hanford facilities. This methodology was developed at Battelle-Northwest for use in selecting ultimate waste disposal alternatives for the Advanced Waste Management Program.

Implementing the decision methodology requires development of a comprehensive, mutually independent set of decision criteria. All important criteria necessary to evaluate the attributes of a given alternative should be included, providing there is little or no interaction between the criteria. A metric is developed for each criterion so as to facilitate quantitative estimation of the performance of each alternative in achieving each criterion. Table 23 describes the tentative set of criteria developed to assign priorities for retired Hanford facilities.

Figure 16 is a diagram of the methodology being used for assigning priorities. Following development of decision criteria, three additional types of information are required to implement the establishment of priorities:

- a. Persons knowledgeable of the characteristics of the alternatives will estimate the performance of each alternative (e.g., each facility class) regarding each decision criterion. Note that the concept of "performance" used in this analysis is a broad one. Future use restriction, technical

TABLE 23. Tentative Performance Criteria

Future Use of the Site

The extent to which an existing facility restricts the desired future use of the facility site.

Technical Feasibility

The capability of achieving desired site or facility conditions using existing or proposed technology.

Economy

The cost of placing a facility in a desired condition, including cost of disposal or restoration, and the cost of predisposal and postdisposal surveillance and maintenance.

Research and Development Spinoff

The opportunities presented by a facility to demonstrate or refine new decontamination or disposal processes.

Environmental Impact

The hazards presented by the facility to workers, public, and nonhuman biota. Other environmental impacts perhaps of significance include effects of chemical toxins, physical impact of the facility on the natural environment, and social impacts.

Security

The extent to which a facility presents problems of security of nuclear materials.

FIGURE 16. Sequence of Operations for Prioritization Methodology

feasibility, cost, R&D spinoff, environmental impact, and security are all elements of performance.

Figure 17 shows an example of an estimate of performance for a hypothetical Facility Class A. The estimate is probabilistic in form, more accurately reflecting uncertainties in knowledge of future conditions than single-value estimates.

- b. A utility or importance function will be compiled for each decision criterion. The utility function describes the importance or "utility" of any level of performance through the expected range of performance levels for each criterion. Importance is evaluated on an arbitrary scale of 0 to 1 with "0" representing a level of performance having least

FIGURE 17. Estimated Cost for Disposition of Facility Class A

importance and "1" representing a level of performance having maximum importance to the decision maker. Figure 18 illustrates a utility function of risk. In this example there is some low level of risk (R_1) below which further improvement is not of significant importance. Likewise, there may be some high level of risk (R_2) above which all levels of risk are equally important. Intermediate levels of risk vary in importance as shown. Utility or importance curves for each decision criterion will be solicited from the cognizant decision makers.

- c. Weights will be assigned to each decision criterion, reflecting the relative importance of the criteria to the decision makers.

The decision methodology will provide an estimate of the overall importance of each facility class, using the information described above, as depicted in Figure 16. The probabilistic estimates of performance are combined with the appropriate utility

FIGURE 18. Utility Function of Risk

functions to produce an estimate of importance of each performance criterion for each facility class. These estimates are then scaled to reflect relative importance of each criterion using the criteria weights. Resulting importance levels of each criterion are added together to produce an estimate of overall importance for each facility class. This estimate will be in the form of a probability density function since original probability estimates of performance were carried through the analysis. Resulting probability density functions (PDF) of overall importance can be displayed on a common scale showing the relative performance of alternative facility classes (Figure 19). An alternative method of display is by summing the area under each PDF to generate curves of cumulative total worth (Figure 20). This latter method of display may clearly separate preferred alternatives (for example, Facility Classes F, E and C in Figure 20).

FIGURE 19. Probability Density Functions of Total Importance for Three Facility Classes

FIGURE 20. Curves of Cumulative Total Importance for Seven Hypothetical Facility Classes

Plans for Next Quarter. The phases of this subtask now authorized, including adaptation of a suitable prioritization methodology and development of decision criteria, are expected to be completed during next quarter.

SUBTASK 5 - PROCEDURES AND TECHNIQUES - H. T. Fullam and W. E. Sande

Work continues on assembling the information center. To date approximately 600 documents and books have

been obtained and about 100 more have been ordered. The computer printout of the catalogued references is updated periodically as additional references are received. Copies of the computer printout are available from H. T. Fullam. The collected documents are stored in the 300 Area Library (Room 209 of the 3760 Building) and are available to anyone wanting to use them.

The first draft of the annotated bibliography will be finished by the end of March. After review, the bibliography will be issued as a BNWL report.

Work continues on investigating and determining disposition activities, techniques and procedures, and waste volumes for transuranic facilities, retention basins, and process liquid waste disposal sites at Hanford. Studies on fuel storage basins and gas and exhaust air systems will be completed by the end of March. The work activities required for each alternative disposition strategy are defined, and the special techniques and procedures required to accomplish the various options are described. The types and volumes of contaminated wastes generated by the various disposition strategies are also identified. The objective in these sections of the handbook is to provide sufficient information on the disposition strategies to allow meaningful risk assessment and cost estimates to be made.

TASK 2 - FORMULATE PLANS AND SPECIFICATIONS FOR A D&D DEMONSTRATION

SUBTASK 1 - PLANNING AND EVALUATION -

J. R. Carrell and K. M. Harmon

The alternatives for disposition (D&D) of the 233-S (Redox Plutonium Concentration) Building were evaluated and the decision made to plan for ultimate razing of the building, while retaining the option to stop the program with the building intact and available for other use. The program plan will emphasize the testing of advanced technology and the development of project planning and control methodology which will be useful in the D&D of similar facilities.

SUBTASK 2 - SITE CHARACTERIZATIONS -

B. W. Mercer and R. L. Brodzinski

Neutron flux measurements around the 233-S process hood were completed, and the metal discs used in determining the neutron fluxes were calibrated. Based on the most recent calibration data, the plutonium inventory in the 233-S process hood is estimated to be ≤ 1.6 kg.

SUBTASK 5 - PROCEDURES AND TECHNIQUES -

C. H. Allen and

P. L. Koehmstedt

Preparation and Activity Specifications

A conceptual decontamination and sequential dismantling procedure was outlined for the 233-S Building. It was based on the philosophy of 1) no manned entry into the grossly contaminated process hood and 2) maximum R&D in situ. Dismantling, handling, and packaging are to be done by installation of glove ports, hoist, and extended-reach tooling.

Development of activity specifications is proceeding, based on the following six major activities:

Activity I. Detailed Characterization of Facility

- a. Define contamination levels.
- b. Identify available utilities (electrical, plumbing, ventilation, etc.).
- c. Verify disconnect and plugging of all REDOX transfer lines.
- d. Identify any required criticality control procedures.
- e. Survey process equipment for deadlegs.
- f. Identify any special dismantle/removal problems.

Activity II. Site Preparation

- a. Conduct preliminary facility contamination cleanup (vacuum, paint, etc.).
- b. Install glove ports in process cell.
- c. Install glove port load-out air lock with hoist and ventilation system.
- d. Install cell hoist.
- e. Install extended-reach tools in hood.

Activity III. Process Equipment Removal

- a. Drain all deadlegs and remove fluid in critical-safe manner.
- b. Start dismantling in upper south end of cell.
- c. Continue dismantling downward.
- d. Remove process equipment to load-out cell.
- e. Repeat b, c, d as required.
- f. Clean cell interior to low level.

Activity IV. In-Cell Research Program

- a. Decontaminate lengthy pipe interiors, exteriors.

- b. Decontaminate painted steel surfaces.
- c. Decontaminate painted and bare horizontal and vertical concrete surfaces.
- d. Decontaminate rigid plastic surfaces.
- e. Test fire-retardant Pu fixation materials.

Activity V. Final Decontamination

- a. Remove all research items and equipment from the cell.
- b. Reclean cell interior surfaces as required.
- c. Erect "greenhouse" surrounding viewing windows if required.
- d. Install staging as required.
- e. Remove rigid plastic windows and framing.
- f. Remove and transfer vertical concrete surfaces and ceiling.
- g. Remove and transfer staging.
- h. Remove and transfer process hood bottom.
- i. Remove and transfer greenhouse.
- j. Isolate building into sections for decontamination starting with area most remote from load-out area.
- k. Remove contaminated surfaces within isolated section and transfer to load-out cell.
- l. Repeat k, leaving all decontaminated sections isolated until building decontamination is complete.
- m. Remove and transfer all doors, gratings, railing brackets, conduit, etc.
- n. Remove and transfer ventilation equipment and remaining utilities.

Activity VI. Final Demolition

- a. Demolish and remove building shell, foundation, etc.
- b. Grade to desired level.

Note: Radiation surveys and monitoring will be conducted as required.

Estimation of Material Volumes in 233-S Building

A preliminary estimate of the total volume of materials in the 233-S Building has been completed for planning purposes. The material volume was categorized in four sections: building structure, process piping, service piping, and process vessels. The volumes of these materials, calculated as uncompacted or unnested in order to conservatively estimate disposal requirements, are:

Building structure (concrete, steel, metal partitions, etc.)	476 yd ³
Process piping (1/8 in. to 4 in. diam)	40 ft ³
Service piping (1/2 in. to 4 in. diam)	15 ft ³
Process vessels (to 7 in. diam)	165 ft ³

SUBTASKS 6 - SAFETY- AND 7 - COST

ESTIMATING - C. E. Jenkins and

J. C. King

Preliminary cost estimates and relative (qualitative) risk assessments were made for the five disposition alternatives identified for 233-S. The results are summarized in Table 24.

TABLE 24. Relative Risk and Cost Estimates for 233-S Disposition Alternatives

Alternative Strategy	Relative Risk (a)			Cost (b) Estimate
	Project Hazards	Short-Term (<100 years)	Long-Term (>100 years)	
1. Perform minimum maintenance and leave in present status	5	1	1	200 k
2. Enclose and seal the structure	4	2	1	1200 k
3. Entomb (encase and fill with concrete)	3	4	3	1700 k
4. Strip interior and decontaminate	1	3	4	2400 k
5. Strip and decontaminate; demolish structure	1	5	5	3500 k

- a. On the scale used here, 1 indicates the highest risk and 5 indicates the lowest.
- b. Cost estimates include: design & engineering; project operations; packaging for 20-year retrievable storage; and transportation to onsite storage area. They do not include costs of future maintenance and surveillance or burial/storage costs.

REFERENCES

- U. S. Atomic Energy Commission, Guidelines for Decontamination of Facilities and Equipment Prior to Release for Unrestricted Use or Termination of Licenses for By-product, Source, or Special Nuclear Material, Nuclear Regulatory Commission, Division of Materials Licensing, April 22, 1970.
- Letter, J. F. Sommers to Jacob Shapiro, Som-93-74, "Comments on Proposed Standard - N328," dated December 13, 1974.
- American National Standards Institute, Proposed American National Standard - Control of Radioactive Surface Contamination of Materials, Equipment and Facilities to be Released for Uncontrolled Use, ANSI N13-3A, September 1973.
- H. J. Dunster, "Surface Contamination Measurements as an Index of Control of Radioactive Materials," Health Physics, vol. 8, pp. 353-356, 1962.
- T. Ishihara and Y. Yoshida, "Monitoring of the Working Environment," SM-114/31, Proceedings of the Regional Seminar on Radiation Monitoring, Bombay, IAEA, Vienna, 1969.
- International Commission on Radiological Protection, "British Codes of Practice," Handling and Disposal of Radioactive Materials in Hospitals and Medical Research Establishments, ICRP 5, p. 20, 1964.
- ICRP, "Atomic Energy of Canada, Ltd.," ICRP 5, p. 20, 1964.
- ICRP, "France CEA," ICRP 5, p. 20, 1964.
- ICRP, "Oak Ridge National Laboratory," ICRP 5, p. 21, 1964.
- M. J. Szulinski, Compiler, Preliminary Problem Definition Decommissioning the Hanford Site, ARH-2164, Richland, WA, April 1972.
- J. M. Selby et al., Considerations in the Assessment of the Consequences of Effluents from Mixed Oxide Fuel Fabrication Plants, BNWL-1697, Battelle Pacific Northwest Laboratories, Richland, WA, June 1973.

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

H. H. Van Tuyl - Project Manager

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 of the 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 $\text{Pu}(\text{NO}_3) \cdot \text{XH}_2\text{O}$, $\text{K}_2\text{Pu}(\text{NO}_3)_6$, and $(\text{NH}_4)_2\text{Pu}(\text{NO}_3)_6$.

SUMMARY

$\text{Pu}(\text{NO}_3)_4 \cdot \text{XH}_2\text{O}$ has been produced on a semi-pilot plant scale in a vessel simulating dimensionally a scaled-down shipping container to determine production parameters.

Equipment has been designed and fabricated to produce $\text{Pu}(\text{NO}_3)_4 \cdot \text{XH}_2\text{O}$ on a kilogram scale. Actual production is expected in late March.

TECHNICAL PROGRESS

SOLID PLUTONIUM SHIPPING FORMS -

G. H. Bryan

$\text{Pu}(\text{NO}_3)_4 \cdot \text{XH}_2\text{O}$ has been produced on a semi-pilot plant scale using 175 ml of 325 g/l Pu in 7.7 M HNO_3 . The

evaporation vessel simulated a scaled-down shipping container in its dimensions (25 cm long by 4 cm diam). Five hours at 1 mm Hg and approximately 60°C were required to form the solid $\text{Pu}(\text{NO}_3)_4 \cdot \text{XH}_2\text{O}$. (See Figure 21). This solid was heated in a vacuum for an additional 2 hr at about 80°C to insure uniform composition. Previous experimental results have indicated that the dry solid $\text{Pu}(\text{NO}_3)_4 \cdot 3\text{H}_2\text{O}$ is first obtained; however, further heating resulted in a weight-stable compound with two waters of hydration.

The evaporation vessel to be used during the pilot plant production of $\text{Pu}(\text{NO}_3)_4 \cdot \text{XH}_2\text{O}$ will be as shown in

FIGURE 21. Evaporation of Plutonium Nitrate Solution at Raised Temperatures

Figure 22. The design permits easy access to the interior to allow sampling for product analysis after a demonstration run. The equipment arrangement, as shown in Figure 23, will be sufficiently versatile to allow selection of optimum operation parameters for production.

The total plutonium content of the initial plutonium nitrate feed will be about 1 kg containing 4 to 7%

^{240}Pu in 5M HNO₃. The plutonium content of the final product will be determined by weighing before and after calcining.

The first pilot plant demonstration run should be completed during late March. All material has been received that is required to complete the facility; however, a shortage of maintenance personnel has caused some delay.

FIGURE 22. Evaporation Vessel

FIGURE 23. Equipment Arrangement

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

Final revision and editing of BNWL-1846, The Risk of Transporting Plutonium Oxide and Liquid Plutonium Nitrate by Truck, has begun. Comments received on a draft of the report are being considered and will be incorporated wherever possible. It is expected that the report will be issued during the next quarter.

System descriptions and accident environment information for plutonium oxide powder and plutonium nitrate solution shipments by rail are being developed or being obtained. Fault trees have been developed for indicating the possible sequences of events that could result in a release of plutonium during a rail shipment. A preliminary risk assessment of shipping PuO_2 and $\text{Pu}(\text{NO}_3)_4$ by rail will be made during the next quarter.

Information needed for the risk assessment of shipping UF_6 and of shipping low- and intermediate-level waste is being developed. A preliminary set of survey questions was developed from the UF_6 fault tree for use in generating a data base on the conditions of UF_6 cylinders in transit. The survey questionnaire was tested during a visit to the UF_6 facility at Holifield National Laboratory (HNL).

The release of certain radioactive materials (e.g., UF_6) in some circumstances can constitute more of a chemical hazard than a radioactive hazard. In anticipation of this, the risk analysis code HEAD has been modified to also analyze the risk from chemically hazardous materials.

TECHNICAL PROGRESS

The comments were received from offsite on BNWL-1846, The Risk of Transporting Plutonium Oxide and Liquid Plutonium Nitrate by Truck. The comments were edited and condensed into a usable form. The comments in general were very constructive. Several comments indicate that the report is a step in a new direction in risk analysis. Most of the comments are not critical of the analytical method used or of the document's technical content.

The risk analysis of shipping PuO_2 in 6M's and $\text{Pu}(\text{NO}_3)_4$ in L-10's by rail has been proceeding. The fault trees have been drawn describing the barriers which must be breached to allow a release of material. The trees were used to determine what information is required to complete the risk analysis. This information has been and will be obtained from Sandia and from other literature sources.

We are awaiting additional information before proceeding further in the analysis. We are gathering this information into a rough draft which will parallel the risk analysis done for the truck transporting of PuO_2 and $\text{Pu}(\text{NO}_3)_4$.

Additional information for the UF_6 and non-high-level waste transporting system has been gathered to aid in producing the risk analyses for these systems.

Additional information for the waste system was found by literature searches and by viewing the waste handling practices at the Atlantic Richfield Hanford Company and Holifield National Laboratory. The two sites visited helped in showing the complex mixture of materials that make up the waste. Both locations package their waste for onsite storage or burial so that their procedures reflect their own needs for packaging waste rather than those requirements that would be enforced if the waste were to be shipped offsite. There is presently no standard way to package waste. There is also no standard way to define and segregate the waste into types. Each location has its own method.

We will study waste packaging further before selecting a reference waste composition or a reference waste packaging method.

Additional information for the UF_6 system was obtained from literature and from a visit to HNL's K-25 Toll Facility. At the Toll Facility the methods of transporting UF_6 were discussed. The UF_6 industry is consistent in the way that the vessels are

shipped. There have been no incidents with the transporting of UF_6 since the current method of tiedown began.

The response from the survey questionnaire at HNL showed the exactness used in checking the shipments for defects in the structure of the vessel and for contaminants in the UF_6 .

The possible ways by which the UF_6 can be released from the UF_6 vessel have been identified as:

1. Breach of vessel with no fire present,
2. Breach of vessel with fire present,
3. Breach of the vessel with the vessel submerged in water, and
4. Breach of the vessel because of internal pressure caused by a fire.

The different release modes for the UF_6 allow different release rates. The first one would allow only a small amount of material to escape, and such a release could easily be stopped. The second and third could release all the inventory, depending on the length of time exposed to either the fire or the water. The fourth could release essentially the total inventory of the vessel within about ten minutes.

The UF_6 forms a protective barrier of UO_2F_2 over its surface when exposed to the normal air environment. This barrier limits the amount of UF_6 released. The UF_6 will self-seal a small hole in the vessel with UO_2F_2 . However, when exposed to high temperature or to water, the UF_6 will continue to be released.

The hazard from UF_6 with a ^{235}U enrichment below 10% is chemical in nature. The UF_6 converts quickly into HF and UO_2F_2 upon contact with moisture. It has been found that UO_2F_2 is more chemically toxic than radiotoxic if the ^{235}U enrichment is less than 10%.

The HF produced is both acutely and chronically toxic. The very corrosive nature of the HF causes damage upon contact. The chronic effect is that a fluorine concentrate buildup in the body occurs, causing fluorine poisoning.

The risk analysis code, HEAD, has been initially adapted to accept chemically toxic materials. The code uses given concentration levels and calculates how many people are affected by the release, depending on windspeed, atmospheric stability, and the amount of material released during a 30-min interval. The code also calculated the probability of receiving that concentration level. The adaptation takes into account the windspeed in a more complex manner than that used previously. This new system will be useful in dealing with chemically toxic materials.

DISTRIBUTIONNo. of
CopiesNo. of
CopiesOFFSITEUNITED STATES

<p>1 ERDA Chicago Patent Group
9800 South Cass Avenue
Argonne, IL 60439
A. A. Churm</p> <p>2 NRC Directorate of Licensing
for Fuels and Materials
4915 St. Elmo Ave.
Bethesda, MD 20014

Deputy Director for
Fuels and Materials
S. H. Smiley

Chief, Technical Support Branch
for Fuels and Materials
R. B. Chitwood</p> <p>1 ERDA Division of Biomedical and
Environmental Research
Earth Sciences Branch
Washington, DC 20545
W. G. Belter</p> <p>1 ERDA Division of Production and
Materials Management
Washington, DC 20545
F. P. Baranowski</p> <p>2 ERDA Division of Reactor
Research and Development
Washington, DC 20545

Assistant Director for
Reactor Technology
E. E. Sinclair

Chief, Fuel Recycle Branch
W. H. McVey</p> <p>9 ERDA Division of Waste Manage-
ment and Transportation
Washington, DC 20545

G. H. Daley
J. C. Dempsey
W. K. Eister
O. P. Gormley
A. F. Perge
F. K. Pittman
R. W. Ramsey
R. D. Walton
R. A. Wolfe</p> | <p>1 ERDA Idaho Operations Office
P.O. Box 2108
Idaho Falls, ID 83401
K. K. Kennedy</p> <p>2 ERDA Oak Ridge Operations Office
P.O. Box X
Oak Ridge, TN 37830

E. H. Hardison
O. T. Turmelle</p> <p>1 ERDA Savannah River Operations
Office
P.O. Box A
Aiken, SC 29801
P. D. Fairchild</p> <p>194 ERDA Technical Information
Center</p> <p>3 Allied Chemical Corporation
550 - 2nd Street
Idaho Falls, ID 83401

J. A. Buckham
B. C. Musgrave
C. M. Slansky</p> <p>1 Allied-Gulf Nuclear Services
P.O. Box 847
Barnwell, SC 29812
A. Schneider</p> <p>2 Argonne National Laboratory
9700 South Cass Ave.
Argonne, IL 60439

N. M. Levitz
M. J. Steindler/
L. E. Trevorow</p> <p>1 Battelle Memorial Institute
505 King Avenue
Columbus, OH 43201
E. S. Bartlett</p> <p>2 Brookhaven National Laboratory
Research Library,
Reference Section
Information Division
Upton, Long Island, NY 11973

J. J. Reilly
R. H. Wiswall</p> |
|--|--|

No. of
Copies

- 1 Brookhaven National Laboratory
Energy Systems Library
Building 902
Upton, NY 11973
Chaim Braun
- 1 Combustion Division
Combustion Engineering, Inc.
Windsor, CT 06095
Jack Parry
- 2 Dow Chemical Company (ERDA)
Rocky Flats Division
P.O. Box 888
Golden, CO 80401
M. A. Thompson
D. L. Ziegler
- 2 duPont Company, Aiken (ERDA)
E. I. duPont DeNemours and Co.
Savannah River Laboratory
Aiken, SC 29801
C. H. Ice
A. S. Jennings
- 1 duPont Company, Wilmington (ERDA)
E. I. duPont DeNemours and Co.
Wilmington, DE 19898
A. A. Johnson
- 2 Electric Power Research
Institute
3412 Hillview Avenue
P.O. Box 10412
Palo Alto, CA 94304
M. Levenson
E. Zebroski
- 1 Environmental Protection Agency
Technology Assessment Div. (AW-559)
Office of Radiation Programs
U.S. Environmental Protection
Agency
Washington, DC 20460
G. L. Meyer
- 1 Environmental Protection Agency
P.O. Box 15027
Las Vegas, NV 89114
B. Mann

No. of
Copies

- 1 Environmental Protection Agency
5555 Ridge Avenue
Cincinnati, OH 45213
R. E. Landreth
- 1 General Electric Company
175 Curtner Avenue
(M/C 160)
San Jose, CA 95125
R. G. Barnes
- 1 General Electric Company
Midwest Fuel Recovery Plant
Route 1, Box 219-B
Morris, IL 60450
E. E. Voiland
- 1 General Electric Company
Vallecitos Nuclear Center
Vallecitos Road
Pleasanton, CA 94566
W. H. Reas
- 1 Gulf Energy and Environmental
Systems
P.O. Box 81608
San Diego, CA 92138
L. H. Brooks
- 4 Holifield National
Laboratory (ERDA)
Central Research Library
Document Reference Section
Central Research Library, ORNL
Laboratory Records Dept., ORNL
Laboratory Records Dept.,
ORNL-RC
- 2 Los Alamos Scientific
Laboratory (ERDA)
P.O. Box 1663
Los Alamos, NM 87544
C. W. Christenson
H. S. Jordan
- 2 Monsanto Research Corporation
Mound Laboratory
P.O. Box 32
Miamisburg, OH 45342
C. J. Kershner
P. Lamberger

No. of
Copies

- 1 National Lead Company
111 Broadway
New York, NY 10006

Stephen Brown
- 1 National Academy of Sciences
Committee of Radioactive Waste
Management
National Research Council
2101 Constitution Avenue
Washington, DC 20418

D. Cyrus Klingsberg,
Technical Secretary
- 1 Natural Resources Defense
Council, Inc.
664 Hamilton Avenue
Palo Alto, CA 94301

T. Lash
- 2 Nuclear Fuel Services, Inc.
P.O. Box 124
West Valley, NY 14171

J. P. Duckworth,
Plant Manager

6000 Executive Blvd., Suite 600
Rockville, MD 20852

E. D. North, Director of
Technical Administration
- 1 NYS Atomic and Space Develop-
ment Authority
230 Park Ave., Rm. 2425
New York, NY 10017

J. G. Cline, General Manager
- 3 Sandia Laboratories
Albuquerque, NM 87107

W. P. Bishop
R. L. Hagengruber
A. C. Zuppero
- 5 Union Carbide Corporation (HNL)
Chemical Technology Division
P.O. Box Y
Oak Ridge, TN 37830

J. O. Blomeke
D. E. Ferguson
E. H. Kobisk
K. H. Lin
W. C. McClain

No. of
Copies

- 1 University of New Mexico
Albuquerque, NM 87131

S. E. Logan
- 1 U.S. Department of Interior
Bureau of Mines
Albany Research Center
1450 W. Queen Avenue
Albany, OR 97321

R. A. Beall

FOREIGN

- 1 EURATOM
Health Physics Division
29, Rue Aldringer
Luxembourg, BELGIUM

Rene Amavis
- 1 Atomic Energy Research Establish-
ment
Harwell, Didcot,
Berks, ENGLAND

K. D. B. Johnson
- 1 United Kingdom Atomic Energy
Authority
Risley, ENGLAND

D. W. Clelland
- 1 Centre de Marcoule
B. P. 106
30 - Bagnols S/Ceze
FRANCE

R. Bonniaud
- 3 Centre d'Etudes Nucleaires
de Fontenay-aux-Roses
Boite Postale 6
92 - Fontenay-aux-Roses
FRANCE

P. J. Regnaut
J. Sauteron
Y. J. Sousselier

No. of
Copies

- 3 Institut für Chemische Technologie
Kernforschungsanlage
Jülich GmbH
D517 Jülich
Postfach 365
GERMANY
- E. R. Merz
K. H. Rattay
Dr. W. Heimerl
(Postfach 1913)
- 1 MGH/GWK/Wiederanfertigung -
Sanlage/WAK
Karlsruhe, GERMANY
- W. A. Issel
- 1 Nuclear Research Center
Waste Management Dept.
D75 Karlsruhe
Weberstr. 5
GERMANY
- H. Krause
- 2 CNEN
Direttore di Programma
Eurex Del CNEN
Saluggia "Vercelli", ITALY
- Giacomo Calleri
- Laboratorio di Ingegneria
Sanitaria
Via Anguillarese km 1 + 300
Roma, ITALY
- Willy Bocola
- 2 Bhabha Atomic Research Centre
Waste Treatment Division
Trombay, Bombay, INDIA
- N. S. Sunder Rajan
K. T. Thomas

No. of
Copies

- 2 International Atomic Energy Agency
Kärtner Ring 11
P.O. Box 590
A-1011, Vienna, AUSTRIA
- 1 Dr. H. F. Ramdohr
c/o Friedrich Uhde GmbH
46 Durtmund
Degglingstr. 10-12
GERMANY

ONSITE

- 6 ERDA Richland Operations Office
- O. J. Elgert
R. B. Goranson
N. T. Karagianes
R. D. Fogerson
D. J. Squires
F. R. Standerfer
- 7 Atlantic Richfield Hanford Company
- D. R. Gustafson
R. E. Isaacson
D. C. Nelson
A. E. Smith/M. J. Szulinski
J. H. Warren
D. D. Wodrich
File Copy
- 1 Exxon
Richland, WA 99352
- S. J. Beard
- 2 United Nuclear Industries, Inc.
- 2 Westinghouse Hanford Company
- A. G. Blasewitz
C. R. Cooley

No. of
Copies.

38 Battelle-Northwest

E. L. Alpen
J. W. Bartlett
R. J. Bashor
C. L. Brown
L. L. Burger
H. C. Burkholder
N. E. Carter
M. O. Cloninger
G. J. Dau
R. L. Dillon
J. R. Eliason
J. W. Finnigan
J. C. Fox
J. J. Fuquay
B. Griggs
G. Jansen
R. S. Kemper
M. R. Kreiter
K. M. Harmon
J. L. McElroy
L. K. Mudge
R. G. Nelson
R. E. Nightingale
D. E. Olesen
A. M. Platt
J. A. Powell
K. J. Schneider
P. C. Walkup
E. J. Wheelwright
L. D. Williams
W. K. Winegardner
H. H. Van Tuyl
Technical Information (5)
Technical Publications