298
1-12-76
4-70 plus
Special Duties

Dr 1889

MASTER

BNWL-1952
UC-70
Waste Management

Nuclear Waste Management and Transportation Quarterly Progress Report July Through September 1975

November 1975

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

Battelle
Pacific Northwest Laboratories

BNWL-1952

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, 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. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
Under Contract E(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 \$5.45; Microfiche \$2.25

BNWL-1952
UC-70
Waste Management

NUCLEAR WASTE MANAGEMENT AND
TRANSPORTATION QUARTERLY PROGRESS
REPORT JULY THROUGH SEPTEMBER 1975

Compiled by

A. M. Platt
Nuclear Waste Technology Program

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, 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.

November 1975

Battelle
Pacific Northwest Laboratories
Richland, Washington 99352

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

FOREWORD

This document is one of a series of technical progress reports designed to report on all ERDA nuclear waste management and transportation programs at Battelle, Pacific Northwest Laboratories, except for the Commercial Radio-active 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 April through June 1975, BNWL-1936.

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

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
FISSION BY-PRODUCT MANAGEMENT	2
MAJOR TASKS	3
1. Partitioning and Fuel Reprocessing	5
2. Candidate By-Products	6
3. By-Product Disposition Technology	10
4. Infrastructural Logistics	10
5. Systems Model	10
6. National Programs	10
TASK 2 - DEVELOPMENT OF EVALUATION METHODOLOGY	11
RADIONUCLIDE MIGRATION FROM NONSALT FORMATIONS	11
PUBLIC ATTITUDES TOWARD NUCLEAR WASTE MANAGEMENT	13
TASK 3 - SPECIAL EVALUATIONS	15
WASTE MANAGEMENT FOR ¹²⁹ I and ⁹⁹ Tc	15
2.0 DECONTAMINATION AND DENSIFICATION OF CHOP-LEACH CLADDING RESIDUES**	19
TRANSURANIC ELEMENT DISTRIBUTION IN CHOP-LEACH CLADDING RESIDUES	20
DECONTAMINATION FACILITY	20
DENSIFICATION PROCESS	22
3.0 PYROLYSIS-INCINERATION OF COMBUSTIBLE ALPHA WASTES**	31
PILOT PLANT STUDIES	31
ASH FIXATION STUDIES	32
4.0 TRITIUM SEPARATION AND FIXATION**	35
TECHNICAL ALTERNATIVES DOCUMENT	35
POLYMER TRITIUM FIXATION	35
METAL HYDRIDE FIXATION	36

* SPONSOR: Environmental Control Technology Division

** SPONSOR: Nuclear Fuel Cycle and Production Division

5.0	DISPOSITION (D&D) OF RETIRED CONTAMINATED FACILITIES AT HANFORD*	39
TASK 1 -	ESTABLISH METHODS, COSTS AND PRIORITIES FOR D&D AT HANFORD	39
	D&D Costs and Priorities	41
TASK 2 -	FORMULATE PLANS AND SPECIFICATIONS FOR A D&D DEMONSTRATION	41
TASK 3 -	EXPLORATORY R&D	42
	Demonstration of Techniques for Stabilization of a Waste Burial Ground	42
	Decontamination of Stainless Steel	42
6.0	TRANSPORTATION SAFETY STUDIES*	43
	Risk Assessment of Plutonium Transport by Truck	43
	Risk Assessment of Plutonium Transport by Rail	43
	Risk Assessment of Non-High-Level Waste Shipments	43
	Risk Analysis Computer Code Development	44
	DISTRIBUTION	47

* SPONSOR: Environmental Control Technology Division
 ** SPONSOR: Nuclear Fuel Cycle and Production Division

LIST OF FIGURES

1	Fission By-Product Management Alternatives	4
2	Comparison of Trend Projections and Forecasts for Primary and Fission By-Product Rhodium	8
3	Comparison of Trend Projections and Forecasts for Primary and Fission By-Product Palladium	9
4	Waste Management Control Surface for Incremental Background Dose with No Partitioning	12
5	Flow Schematic of the Decontamination Facility	22
6	ID Surface Imperfections on Zircaloy-4 Refabricated Clad Tubing	23
7	Longitudinal Section of 4-In. Diam Inductoslag Ingot (Heat Number 460)	25
8	Longitudinal Section of 4-In. Diam Inductoslag Ingot (Heat Number 460) of Pure Inconel 718	26
9	Transverse Section of 4-In. Diam Inductoslag Ingot (Heat Number 460) of CF3 (Casting Grade 304 SS)	27
10	Transverse Section of 4-In. Diam Inductoslag Ingot (Heat Number 460) of 67 wt% CF3 (Casting Grade 304 SS) - 33 wt% Inconel 718	28
11	Transverse Section of 4-In. Diam Inductoslag Ingot (Heat Number 460) of Pure Inconel 718	29
12	Ash Obtained from Pyrolysis-Incineration of Simulated Alpha Waste	32
13	Top of Reactor Opened at End of Run 4	33
14	Distributor at the End of Run 4	34
15	Zirconium-Hydrogen Phase Diagram	37

LIST OF TABLES

1	Potentially Useful By-Product Elements Recoverable from Spent Nuclear Power Fuels Identification, Properties, Availability, Recovery Processes, Production Level Achieved, and Potentially Major Uses	7
2	Clustering of Respondent Groups Based on Discriminant Analysis	14
3	Analyses of a High-Exposure PWR Cladding Hull	21
4	Scenarios for D&D at Hanford	40
5	Beta-Gamma Waste System Description	45
6	Alpha Waste System Description	46
7	Alpha-Beta-Gamma Waste System Description	46

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.

The structure and objectives of some of the work in this program are expected to change as a result of reorganization of waste management within ERDA. The transition is not yet complete; however, it is expected that emphasis will be on the impact of new nuclear fuel cycles (HTGR, CTR, etc.) on waste management and the needs for control of effluents to the environment.

This quarter's report deals primarily with studies started prior to the ERDA reorganization.

SUMMARY

A conceptual plan was developed for management of high-level waste for purposes of recovery and use of economic values.

Work on radionuclide migration from nonsalt formations was completed. The report, soon to be issued, concludes that there are achievable conditions of final geologic storage for which incentives to partition transuranics and other nuclides from the waste are negligible.

Analysis of data from the survey of public attitudes toward nuclear waste management is near completion. Differences in respondents' attitudes were found to be real and not an artifact of the measurement technique.

The study of alternatives for management of iodine and technetium was completed.

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 PNL.

Revision of the draft PNL plan for R&D on disposal of high-level and transuranic waste has been suspended as a result of revision of ERDA priorities. This work is expected to be reactivated in the context of comprehensive planning for waste management systems.

As a step in this comprehensive planning, A. S. Kubo (of the U.S. Military Academy) working at PNL for the summer did a study of alternatives for management of high-level waste. Portions of his report follow:

FISSION BY-PRODUCT MANAGEMENT -

A. S. Kubo

Recent events and those of the past clearly show that issues of the back-end of the fuel cycle are primarily problems of political and social decision making rather than of technical or economic feasibility. Moreover, delays have shown that socio-political factors of decision making have caused abrupt shifts in Federal policies. These abrupt changes have made solution seriatim the norm when it is inappropriate and highly detrimental to national goals.

There is no reason to presume that public climate will change in the near future. Although present public sentiment generally favors science, disenchantment with any proposed policy for the back-end of the fuel cycle might occur in numerous ways.⁽¹⁾ First, there is adequate indication that important sectors of the knowledgeable public assess current Federal nuclear waste management policies as being poor.

Second, public institutions in general face a credibility problem stemming from escalated stakes rather than libelous performance. Having worked for years to create an informed constituency, decision makers must live with the consequences. Candor, informativeness, and comprehensive explanation of public policies will characterize this pluralism. Finally,

events demonstrate that public opinion will become more sophisticated with time, making it imperative that public policy be based on a very broad constituency.

The public climate has made the task of introducing new technologies all the more difficult and costly. It has always been risky and time-consuming to launch a new technology, requiring nearly two decades on the average. Regulatory action in certain new technologies has further lengthened the innovative process, e.g., the extension of FDA actions on new drugs, six months in 1962, forty months in 1969, to a highly variable duration in 1972.⁽²⁾ These delays onerously affect the risk of investment and exacerbate the usual temporal mismatch between innovation and management.⁽³⁻⁵⁾ This risk coupled with the extremely volatile money markets has caused an upward spiralling capital cost, making it even more difficult for private industry to demonstrate the back-end of the fuel cycle.

Concomitantly, the world community verges on a resource crisis. These shortages, whether fundamental or contrived, will impact greatly on the nation's fundamental programs. As the twenty-first century approaches, greater impetus will focus on resource recovery systems, recycling, reuse, and recovery of many forms of materials now classified as wastes.⁽⁶⁻¹¹⁾

Use of selected fission by-products could well reduce the rate of depletion of certain natural resources. A concept for a program to recover and use selected fission products is described below.

The Fission By-Product Management Program focuses on a systematic analysis of the alternative strategies for managing the back-end of the nuclear fuel cycle. This program aims at assessing the technological, economic, and political environment of the intermediate future to:

1. Ascertain the potentially acceptable management strategies.
2. Reduce disruptive surprises that delay program development.
3. Initiate the demonstration of the back-end of the nuclear fuel cycle.

Programmatically, the developed Federal By-Product Management System will:

1. Control the movement of Fission by-products.
2. Establish a Fission By-Product Repository that can:
 - a. Reprocess selected valuable by-products.
 - b. Store potentially valuable by-products.
 - c. Prepare for disposal by-products of poor potential value.
 - d. Dispose of by-products as required.
3. Control the return of the valuable fission by-products to the fuel cycle and other operational systems
4. Control the by-products prepared for disposal.

The major alternatives to be considered in the Fission By-Product Management Program are depicted in Figure 1 and grouped generically as:

1. Alternatives for Spent Fuel management such as immediate reprocessing, indeterminate storage, and use of heat generation.

2. Alternatives for fuel reprocessing that consider:

- a. Should plutonium be recycled and if so who should do the processing?
- b. Should multi-value extractions be made that add to the operational risk of fuel reprocessing?
- c. If multi-valued extractions occur, could the currently unprofitable and sensitive extractions occur at a Federal By-Product Repository?
- d. How will large regional nuclear energy centers or Integrated (Dispersed) Fuel Cycle Facilities impact the By-Product Management Program?

3. Alternatives for Clad Storage/ Disposal such as advanced technologies to use the voluminous material as waste containers or potential resource materials.
4. Alternatives for waste disposal to insure compatibility with the overall By-Product Management Program.
5. Assess the future environment to discern potentially disruptive trends, anticipate new technologies, and forecast fuel cycle variables.

MAJOR TASKS

To support this assessment of the back-end of the nuclear fuel cycle and develop a viable fission by-product management system, five major tasks have been outlined. Clearly many have been analyzed previously but many have not. More importantly, the individual analyses must be integrated to identify potentially favorable near-, intermediate-, and long-term fission by-product management stratagems.

FIGURE 1. Fission By-Product Management Alternatives

1. Partitioning and Fuel Reprocessing

The range of fuel partitioning options can be described by a four-dimensional array of time, space, responsibility, and chemistry.

- a. Time: The impact of reprocessing has historically been viewed to reduce fuel cycle costs. The question of indeterminately deferred reprocessing or the impact of sequential reprocessing should be assessed.
- b. Space: Present plans consider a single reprocessing facility where one-time value-waste separations are made. Conceivably this function can be apportioned between two or more geographically separate facilities to permit closer control, economies of scale, extraction of potentially valuable or sensitive materials, etc. Conversely, colocation of many fuel logistics functions to form an integrated fuel cycle facility would enhance security, permit economies of scale, and reduce internal transportation at the risk of over centralization.
- c. Responsibility: Present concepts focus on commercial reprocessing plants supported by Federal waste repositories. Alternatively, totally Federal reprocessing facilities and repositories are a distinct option, as is commercial reprocessing supported by Federal by-product repositories. The Federal By-Product Repository could perform a reprocessing and storage function for potentially valuable and sensitive materials.

- d. Chemistry: The range of chemical options should consider the complete span of alternatives from extraction of all actinides and lanthanides, through all the actinides, selected actinides, to selected fission products. For apparent reasons some fission by-products are immediately valuable, e.g., uranium and plutonium. Others have future value either by their presence as material resources or by their absence from the ultimate wastes. Previous studies have focused on extracting plutonium, uranium, all the actinides, and a few selected fission products such as strontium and cesium. The technology for plutonium-uranium separation is well established and the raffinates well defined. But the universal extraction of the actinides is hampered by the homologous lanthanides. Future strategies may not require "clean" actinides. For example, storage for future use can tolerate considerable contamination, as would transmutation recycle in fast neutron flux facilities (fast breeders or fusion devices). The selective extractions of fission by-products have heretofore used weapons production wastes aimed at sole product extractions. These limited extractions have been from wastes of special character causing little impact on by-product streams. Opportunities definitely exist for the integrated recovery of more than one isotope or element from a

given process configuration. However, rudimentary process chemistry flowsheets should be developed to anticipate the impact these options would have on fission by-product management, and the management of the more dilute and voluminous low-level and transuranium (TRU) contaminated wastes.

2. Candidate By-Products (See Table 1)

a. Assess candidate by-products for possible extraction:

- 1) Heat: Of the heat-producing fission by-product radioisotopes, the fission products ^{90}Sr , ^{137}Cs , ^{144}Ce , and ^{147}Pm and the actinides ^{238}Pu , ^{242}Cm , ^{244}Cm , and ^{241}Am appear as the most likely candidates. (12,13)

Preliminary assessments, however, indicate that the anticipated annual quantities of the fission products will far exceed potential annual needs. (15,16) Thus saving these products now appears unnecessary. But a similar conclusion is not clear for the actinides, especially ^{238}Pu (and its progenitor ^{237}Np). Of the fissile isotopes, ^{233}U , ^{235}U , ^{239}Pu , and ^{241}Pu will be the most abundant and valuable. Present assessments, primarily economic, justify their immediate capture and reuse, notwithstanding the safeguards issue regarding plutonium.

- 2) Radioisotopic Irradiators:

Irradiator values are based on isotopes emitting either neutron, gamma, or beta radiation.

Neutron emitters have historically been based on naturally occurring alpha emitters that utilize α -n reactions such as ^{210}Po -Be and ^{226}Ra -Be sources.

With the development of nuclear fission reactors, artificially produced highly intense alpha-emitting actinides (^{238}Pu , ^{239}Pu , ^{241}Am , and ^{242}Cm) have gradually replaced the natural alpha sources where size and intensity were important. Also the availability of the transplutonium elements permits production of ^{252}Cf that spontaneously fissions, yielding an intense neutron source. The industrial applications of neutron technology have grown in recent years, but their future remains unclear. The commercial use of gamma and beta irradiators is even more universal, but again previous assessments indicate an overabundance of these values for the foreseeable future.

- 3) Radioisotopic Tracers: Radioactive tracers are utilized throughout industry and extensively in research and health care. The use of tracers is usually characterized by minute quantities of high-purity isotopes. These characteristics generally make potential recovery schemes uneconomic for independent nuclear fuel reprocessors, and has led to special purpose commercial isotope generators or federal isotope production facilities.

TABLE 1. Potentially Useful By-Product Elements Recoverable from Spent Nuclear Power Fuels Identification, Properties, Availability, Recovery Processes, Production Level Achieved, and Potentially Major Uses (Compiled from Reference 14)

Element	Principal Isotopes of Interest	Est. % of Isotope in Element	Half Life Years	Curies Per Gram Isotope (100%)	Watts (Thermal) Per Gram Isotope (100%)	Type and Energy of Principal Radiation MEV	Concentration in Power Reactor Fuels Grams/Ton*	Suggested or Potential Major Use	Recovery Processes Used	Approximate Level of Isotope Production Achieved in U.S.**	Where Done***
Krypton	⁸⁵ Kr	6.5	10.74	397	0.623	80.67	17	Phosphors, Special Light or Power Source	Cryogenic	50,000 Ci	NRTS, HNL
Strontium	⁹⁰ Sr	50	28.9	142	0.96	82.26	411	Power Source	Precipitation Solvent Extraction Ion Exchange	Multikilograms	RL, PNL, HNL
Technetium	⁹⁹ Tc	100	2.13 x 10 ⁵	0.017	----	80.29	628	Chemical Process Catalyst, Alloying Agent	Ion Exchange Precipitation	Kilograms	RL, PNL, HNL
Rhodium	¹⁰³ Rh	100	Stable	Stable	----	----	337	Chem. Process Catalyst, Alloying Agent, Corrosion Resistant Material	Ion Exchange Precipitation	A Few Grams	RL, PNL
Ruthenium	¹⁰⁶ Ru	3.3	1.0	3375	33.1	83.35	1708 ^(a)	Chem. Process Catalyst, Alloying Agent, Radiation Source (8)	Volatility Precipitation	100 Curies	PNL
Palladium	¹⁰⁷ Pd (7 x 10 ⁶ yr)	16	"Stable"	"Stable"	----	80.035	875 ^(a)	Chem. Process Catalyst, Alloying Agent, Corrosion Resistant Material	Ion Exchange Precipitation	A Few Grams	PNL, RL
Xenon	All Are Stable	100	Stable	Stable	----	----	3990	Special Inert Gas, Electronic Uses Human Anesthesia	Cryogenic	Kilograms	NRTS, HNL
Cesium	¹³⁷ Cs (& ¹³⁴ Cs)	35	30.2	87	0.42	γ0.67 β1.17	950	Radiation Source	Ion Exchange Precipitation	Multikilograms	RL, PNL, HNL
Cerium	¹⁴⁴ Ce	4.5	0.78	3190	25.6	β2.98 γ2.18	120	Power Source, Radiation Source (8)	Precipitation Solvent Extraction	Multigrams	RL, PNL, HNL
Promethium	¹⁴⁷ Pm	100	2.62	929	0.33	80.23	66	Power Source, β Source, Phosphors	Solvent Extraction & Chromatographic Ion Exchange	Kilograms	HNL, PNL
Neptunium	²³⁷ Np	100	2.14 x 10 ⁶	0.0007	----	α	407	Target for ²³⁸ Pu	Solvent Extraction Ion Exchange	Multikilograms	PNL, RL SRL, SRP
Americium	²⁴¹ Am	50	433	3.25	0.11	α, γ0.06	47	γ and α Source,	Solvent Extraction Chromatographic Ion Exchange	Kilograms	PNL, SRL SRP
	²⁴³ Am	50	7370	0.2	----	α, γ0.075	53.3	Target for ²⁴⁴ Cm			
Curium	²⁴⁴ Cm	100	17.85	82.1	2.84	α, γ0.04	10.7	α Source, Power Source	Same as for Americium	Kilograms	PNL, HNL SRL, SRP

* At exposure of 25,000 MWD/Ton (metric) in typical light water reactors.

(a) Grams of total element.

** All of this production (except for Np) has been on an intermittent basis--no prolonged routine or continuous production. Np production has been routine for many years.

*** HNL - Holifield National Laboratory, Oak Ridge, Tennessee
 NRTS - National Reactor Test Station, Idaho Falls, Idaho
 RL - Richland, Washington (Hanford USAEC, by General Electric, Isochem, and Atlantic Richfield Hanford Cos.)
 PNL - Pacific Northwest Laboratory, Richland, Washington
 SRL - Savannah River Laboratory, Aiken, South Carolina
 SRP - Savannah River Plant, Aiken, South Carolina

4) Mineral Resources: A study of critical mineral resources indicates a few fission by-products of potential value: The Platinum Metals Group (rhodium and palladium), chromium (in stainless steels), zirconium (in cladding materials), and the noble gas xenon.

a. Platinum metals and chromium are especially critical since foreign sources provide most of the annual U.S. needs. (6) Studies indicate that the fission by-products rhodium and palladium are especially promising candidates based on their scarcity, high value and low radioactivity. (17,18) But these studies indicate that some years of aging are necessary to attain this low level of radioactivity. (Fission by-product palladium will always contain ^{107}Pd , a 7×10^6 year half life, beta emitter.) Current estimates of annual values in fission by-product rhodium and palladium and their historic consumption are depicted on Figures 2 and 3. At the currently predicted consumption pattern, 40 to 50% of the known palladium and rhodium reserves will be consumed by the end of this century. (17)

b. The voluminous cladding hull wastes resulting from reprocessed nuclear fuels contain significant chromium and zirconium values. Although

A. Trend projections and forecasts for primary palladium taken from Mineral Facts and Problems, 1970 Edition, Bureau of Mines, U.S. Department of the Interior, Superintendent of Documents, Washington, D.C. 20402.

B. Forecasts for fission by-product palladium taken from L. L. Clark et al., A Market Analysis of the Fission Products Rh, Pd, Ru, and Tc, BNWL-1690, Battelle, Pacific Northwest Laboratories, Richland, WA 99352, February 1973.

FIGURE 2. Comparison of Trend Projections and Forecasts for Primary and Fission By-Product Rhodium

carry-over and induced radioactivity limit applications for these resources, very restricted utilization might be possible, e.g., solidification and storage containers for the fission by-products. (19)

c. Fission by-product xenon can be purified with current technology to yield a product that is insignificantly radioactive after a year's decay. This gas has two potential applications: (20,21)

A. Trend projections and forecasts for primary rhodium taken from Mineral Facts and Problems, 1970 Edition, Bureau of Mines, U.S. Department of the Interior, Superintendent of Documents, Washington, D.C. 20402.

B. Forecasts for fission by-product rhodium taken from L. L. Clark, et al., A Market Analysis of the Fission Products Rh, Pd, Ru, and Tc BNWL-1690, Battelle, Pacific Northwest Laboratories, Richland, WA 99352, February 1973.

FIGURE 3. Comparison of Trend Projections and Forecasts for Primary and Fission By-Product Palladium

to replace krypton in specialty incandescent lamps and to use as a human anesthesia.

- d. The numerous other fission by-products do not represent significant potential value either because of their radioactivity or the substantial natural resources that exist for the projected future.

5) Research Materials: The transuranium actinides as a group and technetium are elements not normally found in

nature and thus represent research values in the fission by-products. To date, this need has not been clearly assessed.

6) Toxic Materials: The long-term risk of waste disposal can be reduced if certain materials are removed from the wastes. For example, preliminary studies in isotope migration induced by underground water indicate reduced concentrations of technetium and neptunium can lower doses to future populations in event of a repository failure.⁽²²⁾ Curiously, potential exploitable values have been identified for both elements.

- b. Assess the market potential of the candidate by-products. For some the present market will support their recovery. However, for most, extraction at present would be clearly unprofitable. For the latter, where potential justifies, recoverable by-product storage would be appropriate. This would permit future recovery, charging the cost to the user, while rightfully charging the producer the storage fee.
- c. Assess the supply of the candidate by-products. For some, the present extraction and long-term storage may be the only possible strategy to have a usable resource at a future time. For others, the annual supply may so overwhelm potential demand that preparation for ultimate disposal is clearly indicated.

3. By-Product Disposition Technology

Previous back-end analyses have considered a narrow range of alternatives. When the full range of alternatives of Figure 1 is considered, the current disposition technology should be reassessed to consider the expansion of these functions:

a. Spent Fuel Handling

- 1) Interim storage awaiting fuel reprocessing
- 2) Indeterminate storage.

b. Material Conditioning

- 1) By-product solidification and/or concentration for potential recovery or disposal
- 2) Storage containers for potential recovery or disposal
- 3) Cladding and hardware processing technology.

c. Storage

- 1) Interim liquid storage facilities
- 2) Retrievable storage facilities for interim by-product control awaiting potential value recovery.
- 3) By-Product repositories for interim by-product control awaiting potential value recovery.

d. Disposal: Irrespective of the near-term strategy for fission by-product management and utilization, ultimate disposal must be planned for all conceivable alternatives. (The range of options implied follows BNWL-1900.)

1) Terrestrial Options

- a. Geologic Concepts
- b. Ice Sheet Concepts
- c. Seabed Concepts

2) Extraterrestrial Options

- a. Solar and Earth Orbits
- b. Solar Impacts
- c. Solar Escape

3) Elimination Option - Transmutation

4. Infrastructural Logistics Consideration of the back-end of the nuclear fuel cycle must be integrated with the total fuel cycle, and the alternative strategy impacts must be assessed as they require support from Federal Licensing and Administrative requirements, the commercial nuclear industry, the need for capital investments, the impact on money markets, the added requirement for technical and supportive manpower, the use of natural resources, and the impact on the transportation industry.

5. Systems Model There is a fundamental need to develop a systems model that can support inventory control and safeguard requirements, identify the interlinks between the head-end, utilization, and back-end of the fuel cycle, predict economic impacts, and assess risks and benefits of the myriad of alternatives suggested by the fission by-product management system. Use of fission by-products will most likely necessitate sophisticated operations that will increase both costs and risks. But, unfortunately, to date costs and benefits have not been quantitatively assessed.

6. National Programs An overview of national programs in medicine, space, energy, transportation, environment, education, national security, human and mineral resources, and fundamental research concurrently with the

assessment of the total fuel cycle is mandatory in order to prevent unintentional suboptimization and long-term program inflexibility caused by short-sighted, near-term decision making.

TASK 2 - DEVELOPMENT OF EVALUATION

METHODOLOGY

RADIONUCLIDE MIGRATION FROM NONSALT FORMATIONS - H. C. Burkholder and M. O. Cloninger

Work on this subtask has been reported in Section 2.0, "Separation of Actinide Elements from Radioactive Waste," in previous quarterly reports. This is the final report of work on the evaluation of partitioning incentives for high-level waste. A topical report is to be issued in October which includes all work performed for the evaluation of partitioning incentives task (H. C. Burkholder, M. O. Cloninger, D. A. Baker, and G. Jansen, Incentives for Partitioning High-Level Waste, BNWL-1927).

The incentives for separating and eliminating various elements (but particularly the transuranics) from radioactive waste prior to final geologic storage were investigated. Exposure pathways to man were defined, and potential radiation doses to an individual living within the region of influence of the underground storage site were calculated. The accumulated high-level waste (i.e., the fission product waste produced by reprocessing spent fuel) from the U.S. nuclear power economy through the Year 2000 (including all tritium, carbon, and iodine from spent fuel) was

the assumed radionuclide source, and western U.S. desert subsoil was the assumed geologic medium.

The results of the study showed that for reasonable storage conditions the potential radiation dose was of the same order, or less than, routine doses from natural sources. The study therefore concluded that for the situations investigated the incentives for special effort to remove any elements, including the transuranics, from high-level waste are negligible. The study results also showed that incentives exist for converting high-level calcine into glass.

The study required numerous assumptions concerned with the transport of radioactivity to man and the effects of radioactivity on man. The assumptions used, on the whole, maximized the estimated potential radiation doses. Thus, incentives for removing elements from the waste tended to be maximized. Incentives were also maximized by assuming that elements removed from the waste could be eliminated from the earth without risk.

The conclusion that partitioning incentives are negligible--despite assumptions tending to maximize them--is based on comparison of predicted potential radiation doses with routine doses from natural sources. This comparison is made using three-dimensional plots such as Figure 4. Plotted on the axes of Figure 4 are three parameters (path length, leach time, and time of initial release) over which man has some degree of selection or control when designing

FIGURE 4. Waste Management Control Surface for Incremental Background Dose with No Partitioning

a waste management system. An increase in the magnitude of any of these parameters results in greater isolation of the waste from man's environment. The surface located in Figure 4 corresponds to an assumed incremental dose level of "background." Combinations of path length, leach time, and time of initial release which define points located inside or under the surface (i.e., between

the surface and the origin) have incremental doses above background; combinations located outside or over have incremental doses below "background."

If the final storage site is separated from the biosphere by at least 5 miles of western U.S. desert soil or equivalent, the waste form has a leach time of at least 15,000 years (1 ft diam x 10 ft long monolithic

borosilicate glass waste form with a surface flux of slightly less than 10^{-5} g/cm²/day), and the waste container fails in the Year 2000 or later, the waste management system is characterized by a point located just above the surface in Figure 4 where the ²²⁶Ra ridge and ¹⁴C ledge meet.

Similar surfaces could be drawn on Figure 4 which correspond to other assumed dose levels. The analysis of such situations shows that there are feasible conditions of geologic emplacement where the predicted incremental dose to man is calculated to be as low as one one-hundredth of "background." Although "background" varies with geographic location and consensus standards of allowable incremental dose have not yet been fully established, we concluded that high-level waste in glass form can safely be placed in selected geologic media.

The methods used in the study can be extended to evaluate any combination of waste type and geologic medium at sites that are candidates for final geologic storage.

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

Major activities during this quarter have included preparation of the final report of FY-1975 research, validation of measurement methods used in the data collection instrument, and data reduction analysis--aggregating both at the level of questionnaire items and at the level of respondent subgroups.

The first two chapters of the final report have been completed and have undergone two initial revisions.

Much of the questionnaire content was aimed at assessing, via multiple measurement methods, the relative importance which respondents attached to four issues associated with nuclear waste disposal: short-term safety, long-term safety, cost, and accident detection and recovery.

Analyses now completed help substantiate the validity of the attitude measuring procedures used in the study. A pattern of correlation was obtained indicating that 1) the four factors used in the study were of psychological relevance to the respondents and 2) differences in the relative importance of the factors is not due to measurement artifact, but instead reflects actual differences as perceived by the study respondents.

A total of 22 groups of respondents were surveyed during the fourth quarter of FY-1975. Recent analyses have been directed toward using empirical procedures to aggregate these groups into a smaller number of subgroups. Discriminant function analyses applied to a large set of questionnaire items have resulted in the empirical reduction of the 22 groups into five basic respondent groups. The results of the clustering procedure are shown in Table 2. Questionnaire data, such as presented briefly in the July 1975 quarterly report will be presented and interpreted in the final report using these clusters.

The most immediate project objective is the submission of the draft

TABLE 2. Clustering of Respondent Groups
Based on Discriminant Analysis

I. Environmentalists - N = 69

Local Environmental Organization (West)
Women's University Organization (Northwest)
Public Interest Lobbyists (Midwest)
National Environmental Organization (Midwest)
National Environmental Organization (Northeast)
National Environmental Organization (South)

II. Research Engineers - N = 51

Public Officials (Northwest)
Corporation Engineers (Northwest)

III. Church Members - N = 54

Church Members (Northeast)
Church Members (Northwest)

IV. University Students/Civic Organizations - N = 209

Undergraduate Civil Engineering Class (Northwest)
Undergraduate Physics Class (West)
Undergraduate Civil Engineering Class (South)
Undergraduate Psychology Class (Northeast)
Continuing Education Class (Northeast)
Recreationists (Midwest)
Civic Organization (Midwest)
Civic Organization (West)
Public Utility Employees (South)

V. Jr. High and High School Students - N = 82

Jr. High School Students (Northwest)
High School Physics Class (Midwest)
High School Environmental Science Class (South)

final report. Based on the analyses now on hand, October 15 is the anticipated target data for completion of this report. Follow-on activities include preparation of three topical

reports for publication in social science journals. These topical reports were referred to in the July quarterly report.

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.

Results of all studies started in FY-1975 have previously been summarized in these reports except for the study on management of iodine and technetium and the study on deferred reprocessing. The work on deferred reprocessing has been incorporated in ERDA's ongoing comprehensive assessment of waste management alternatives; it will be reported elsewhere in the future.

WASTE MANAGEMENT FOR ^{129}I and ^{99}Tc -

H. G. Rieck and J. S. Fruchter

Various methods have been proposed for managing the radioactive wastes produced by the nuclear power cycle. Chemical mobility, biological activity, long half-life and low decay energy are properties of ^{99}Tc and ^{129}I which must be considered in selecting a method for managing these isotopes. The level and type of management effort will be determined by their predicted risk to man. Further research is required to properly evaluate this risk.

Separation of ^{99}Tc and ^{129}I from the nuclear fuels reprocessing stream appears feasible by either ion exchange or solvent extraction processes. Several processes have been successfully used to recover iodine; decontamination factors of 10^6 have

been obtained. Although studies have been conducted on technetium recovery, a preferred method has not been found.

Geologic disposal concepts can be used for ^{99}Tc management if a stable compound can be found. Deep injection into a stable geologic formation would be a preferred method. Disposal of ^{129}I in a bedded salt formation is feasible if isotopic dilution following a release can be verified.

If a stable compound or matrix can be found for iodine and technetium and if a suitable container can be developed, seabed disposal is a possible technique. The consequence of a container failure and subsequent release of the waste would be minimized by dilution processes, and in the case of ^{129}I isotopic dilution would be a beneficial consequence.

Extraterrestrial ejection and transmutation processes are concepts which could be used to eliminate ^{99}Tc and ^{129}I . Present space technology could be used to develop the mechanics necessary for a space waste disposal mission; however, development of space tugs and shuttles as well as a satisfactory waste container for the isotope would be required. A disadvantage of space disposal is the permanent loss of metals required in construction of the expendable space vehicles.

Present day reactor technology does not provide an economical system for transmutation processes. With the advent of fission reactors, the neutronics will be favorable for successful transmutation management of ^{99}Tc and ^{129}I . Problem areas will be the transmutation target container design and isotopic purification of the target.

Isotopic dilution is a technique that could be used for management of ^{129}I . The feasibility of this technique depends on the economics of obtaining large required amounts of ^{129}I . A problem area will be the disposal of the diluted mixture.

REFERENCES

1. C. B. Kaufman, "The Future Social Climate for R&D," Research Management, pp. 29-32, January 1975.
2. A. E. Rupp, "Radioisotopes: 1980," Isotopes and Radiation Technology, vol. 5, no. 2, pp. 89-91, Winter 1967-1968.
3. R. C. Dean, Jr., "The Temporal Mismatch - Innovation's Pace vs. Management's Time Horizon," Research Management, pp. 12-15, May 1974.
4. S. M. Greenfield, "Incentives and Disincentives of EPA Regulations," Research Management, pp. 11-14, March 1974.
5. The Fuel Cycle Task Force, Nuclear Fuel Cycle, ERDA-33, U.S. Energy Research and Development Administration, Washington, D.C., March 1975.
6. Bureau of Mines, Mineral Facts and Problems, 1970 Edition, U.S. Department of the Interior, Superintendent of Documents, Washington, D.C., 1970.
7. E. N. Cameron, Ed., The Mineral Position of the United States, 1975-2000, The University of Wisconsin Press, Wisconsin, 1973.
8. Committee on Resources and Man, Resources and Man, National Academy of Sciences - National Research Council, W. H. Freeman and Co., San Francisco, CA, 1969.
9. H. H. Landsberg, "Policy Elements of U.S. Resource Supply Problems," Resource Policy, pp. 104-114, December 1974.
10. R. W. McKee, Technical, Economic and Policy Considerations Affecting Future Production, Marketing, and Use of Cesium-137 and Strontium-90, BNWL-1686, Battelle-Northwest, Richland, WA, December 1972.
11. Metal Statistics 1973, 66th Annual Edition, Fairchild Publications, Inc., New York, NY, 1973.
12. R. P. Perret, "Power from Radioisotopes," Aspetti Scientifici E Tecnici Delle Applicazioni Industriali Dei Radioisotopi, XI Congresso Nucleare di Roma, pp. 43-60, 23-24 Maggio 1966.
13. C. A. Rohrmann, Personal Communication (Interview on Fission By-Product Values and Waste Management Alternatives), Battelle-Northwest, Richland, WA, June 24, 1975.
14. C. A. Rohrmann, "The Availability and Applications of Radioactive and Stable By-Products from the Chemical Processing of Spent Nuclear Power Fuels," Presented at the 10th Japan Conference on Radioisotopes, Tokyo, Japan, November 16-18, 1971, BNWL-SA-4108, Battelle-Northwest, Richland, WA.
15. A. S. Kubo, Technology Assessment of High-Level Nuclear Waste Management, Unpublished Ph.D. Thesis, Massachusetts Institute of Technology, May 1973.
16. T. S. Weissman, "Report on the Fission Product Conversion and Encapsulation Facility," Aspetti Scientifici E Tecnici Delle Applicazioni Industriali Dei Radioisotopi, XI Congresso Nucleare di Roma, pp. 189-210, 23-24 Maggio 1966.
17. L. L. Clark, F. P. Roberts, J. C. Sheppard, and J. D. Kaser, A Market Analysis of the Fission Products Rh, Pd, Ru, and Tc, BNWL-1690, Battelle-Northwest, Richland, WA, February 1973.

18. C. A. Rohrmann, "Large-Scale Exploitation of Fission-Product Rhodium and Palladium," Isotopes and Radiation Technology, vol. 6, no. 4, pp. 352-358, Summer 1969.
19. A. M. Platt, Personal Communication (Discussion of Possible Uses of Spent Fuel Cladding Hulls), Battelle-Northwest, Richland, WA, June 25, 1975.
20. G. A. Cook, Ed., Argon, Helium and the Rare Gases, 2 vols., Interscience Publishers, New York, 1961.
21. C. A. Rohrmann, "Fission-Product Xenon and Krypton - An Opportunity for Large-Scale Utilization," Isotopes and Radiation Technology, vol. 8, no. 3, pp. 253-260, Spring 1971.
22. D. H. Lester, G. Jansen, and H. C. Burkholder, "Migration of Radionuclide Chains Through an Adsorbing Medium," presented at the National Meeting of the American Institute of Chemical Engineers, March 16-20, 1975, Houston, Texas, BNWL-SA-5079, Battelle, Pacific Northwest Laboratories, Richland, WA.

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

2.0 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

The analysis of high-exposure PWR fuel cladding has confirmed previous estimates that much of the transuranic activity can be leached from the fuel clad, a second substantial portion is confined to the corrosion product oxide, and only about 1/100 of 1% is in the cladding metal.

Chopped Zircaloy-4 (unirradiated) cladding that had been melted by the Inductoslag process (Heat Number 431) has been refabricated into cladding tube by a commercial cladding tube manufacturer. Spherical particles of zirconium or zirconia partially imbedded on the ID of the finished tubing resulted in a poor ID surface. Ingot chemistry, hardness, tubing

performance during fabrication and the lack of any other observable defects indicate that the quality of this material is suitable for any fabrication process. Standard autoclave testing of samples from the tubing showed normal weight gains and acceptable black lustrous films.

Macroscopic examination of Inductoslag melted pure 304 stainless steel, an alloy of 67 wt% 304 - 33 wt% Inconel 718 and pure Inconel 718, shows good ingot quality in the pure materials, while the alloy retains some unmelted stainless steel. Ingot quality is suitable for storage; however, additional melting development is required to achieve complete homogeneity.

TECHNICAL PROGRESS

TRANSURANIC ELEMENT DISTRIBUTION IN CHOP-LEACH CLADDING RESIDUES -

B. Griggs

Initial treatment of a section of high-exposure PWR fuel to get samples for analysis was previously described.⁽¹⁾ Briefly, 2.5-cm long pieces of a PWR fuel rod were leached twice in 8 M HNO_3 for a total period of 24 hr with temperatures as high as 99°C, which removed most of the fuel comparable to a dissolver. The leached Zircaloy cladding was further leached in an ammonium oxalate-citrate-fluoride-peroxide solution for 12 hr at 95°C, which removed 56% of the alpha activity on the piece. After chemical and/or mechanical removal of the oxide corrosion product layer plus a small (~ 0.02 mm) amount of underlying metal, the base metal was dissolved for analyses. Dissolution of a comparable piece with the leached surface oxide still in place allows the calculation of base metal, surface oxide, and leachable contamination in the Zircaloy hull. The analyses of this cladding are shown in Table 3.

Examination of Table 3 shows that a substantial fraction of the residual TRU elements can be leached from the cladding surface. The isotopic analysis of the current uranium in the base metal compares with the fuel burnup, assuming it was originally natural uranium. The fuel isotopic was originally 5.84% ^{235}U . No diffusion of TRU elements into the cladding base metal is observable. The low original uranium contamination of the base metal at 0.3 ppm and very low thorium contamination (see low

^{233}U) resulted in more than 10 nci/g of TRU elements even if one converts the ^{241}Pu to ^{241}Am . The analytical results have not yet been correlated with the ORIGEN computer calculation.

DECONTAMINATION FACILITY -

R. G. Moles

On October 2 and 3, a visit was made to Rayco Machine Company, Portland, Oregon, to inspect the quality of welding being performed on the decontamination vessels, condenser/gas separator, and HF storage vessel.

An inline heater assembly for use of preheating the argon carrier gas and the hydrogen fluoride has been built and tested. This heater consists of a 30-in. length of insulated 3/4-in. Inconel Schedule 40 pipe with nichrome wire wrap. Using an argon gas flow at 100 cc/min, the heatup time to 600°C is about 10 min. This is considerably less heatup time than required for a conventional tube type furnace. The cool down time will be shortened many times over that of a conventional furnace.

As reported previously, we have collected the instrumentation required for the prototype hot cell decontamination facility at 1706KEL, 100-K Area. Assembly of this instrumentation is about 50% complete.

An electrical power failure has caused a shutdown of the 1705KEL hot cell and building exhaust system. Due to this failure, a review of possible redundant ventilating system and emergency electrical power is being reviewed. A small backup exhaust fan for the hot cell or an auxiliary backup electrical power or both will have to be installed to

TABLE 3. Analyses of a High-Exposure PWR Cladding Hull

Analyses	Surface ^(b) Leach	Surface Oxide ^(c) Plus < 25 μ m of Metal	Base Metal ^(d)	Base Metal ^(e) Outside 0.12 mm
Total Alpha ^(f)	$8.7 \cdot 10^3$ nci/cm ^{2(g)}	$6.7 \cdot 10^3$ nci/cm ²	8.2 nci/cm ² 25 nci/g	29 nci/g
Total U	$1.04 \cdot 10^5$ ng/cm ²	$8.52 \cdot 10^4$ ng/cm ²	106 ng/cm ² 318 ng/g	(a)
²³³ U	$1.5 \cdot 10^{-4}$ atom%	$4.1 \cdot 10^{-4}$ atom%	$1.99 \cdot 10^{-1}$ atom%	(a)
²³⁴ U	$1.88 \cdot 10^{-2}$	$1.91 \cdot 10^{-2}$	$3.82 \cdot 10^{-2}$	(a)
²³⁵ U	$9.52 \cdot 10^{-1}$	$9.69 \cdot 10^{-1}$	$1.45 \cdot 10^{-1}$	(a)
²³⁶ U	$9.79 \cdot 10^{-1}$	$9.69 \cdot 10^{-1}$	$1.16 \cdot 10^{-1}$	(a)
²³⁸ U	$9.80 \cdot 10^1$	$9.80 \cdot 10^1$	$9.95 \cdot 10^1$	(a)
Total Pu ^(h)	$3.9 \cdot 10^3$ ng/cm ² $6.3 \cdot 10^4$ nci/cm ²	$3.4 \cdot 10^3$ ng/cm ² $5.5 \cdot 10^4$ nci/cm ²	$8.7 \cdot 10^1$ nci/cm ² $2.6 \cdot 10^2$ nci/g	$1.4 \cdot 10^1$ ng/g $2.4 \cdot 10^2$ nci/g
²³⁸ Pu	$1.2 \cdot 10^3$ nci/cm ²	$1.1 \cdot 10^3$ nci/cm ²	2.6 nci/g	2.7 nci/g
²³⁹ Pu	$1.2 \cdot 10^2$	$1.0 \cdot 10^2$	0.49	0.44
²⁴⁰ Pu	$2.0 \cdot 10^2$	$1.7 \cdot 10^2$	0.70	0.68
²⁴¹ Pu	$6.1 \cdot 10^4$	$5.4 \cdot 10^4$	256	240
²⁴² Pu	1.7	1.5	$6.5 \cdot 10^{-3}$	$7.3 \cdot 10^3$
²⁴¹ Am	$2.7 \cdot 10^2$	$2.1 \cdot 10^2$	1.11	0.97
²⁴³ Am	$2.6 \cdot 10^1$	$1.6 \cdot 10^1$	0.08	0.09
²⁴² Cm	$2.5 \cdot 10^3$	$2.0 \cdot 10^3$	9.5	9.7
²⁴⁴ Cm	$2.7 \cdot 10^3$	$2.1 \cdot 10^3$	9.8	12.0
¹²⁵ Sb ⁽ⁱ⁾	--	$2.0 \cdot 10^5$	ND	$1.89 \cdot 10^6$
⁹⁵ Zr	--	$2.6 \cdot 10^4$	ND	$1.98 \cdot 10^5$
⁹⁵ Nb	--	$5.3 \cdot 10^4$	ND	$4.38 \cdot 10^5$
¹³⁷ Cs	$1.6 \cdot 10^6$	$6.4 \cdot 10^5$	ND	$1.55 \cdot 10^2$
¹³⁴ Cs	$1.6 \cdot 10^6$	$5.7 \cdot 10^5$	ND	$1.16 \cdot 10^2$
¹¹³ Sn	--	$1.1 \cdot 10^4$	ND	$9.05 \cdot 10^5$
⁶⁰ Co	--	$2.7 \cdot 10^4$	ND	$4.67 \cdot 10^4$
⁵⁴ Mn	--	$7.8 \cdot 10^3$	ND	$4.67 \cdot 10^4$
¹⁰⁶ Ru	$2.7 \cdot 10^5$	$7.6 \cdot 10^5$	ND	
¹⁴⁴ Ce	$7.6 \cdot 10^4$	$2.8 \cdot 10^5$	ND	

a. The analytical sample for this determination was lightly contaminated with natural uranium during analysis, making the uranium analyses invalid.

b. The surface leach was conducted by treating a 2.5-cm piece of cladding in five successive aliquots of a 0.4 M ammonium oxalate - 0.16 M ammonium citrate - 0.1 M ammonium fluoride 0.3 M hydrogen peroxide addition at 95°C for 12 hr.

c. Analysis of the surface oxide plus less than 25 μ m of metal content was determined by dissolving a piece of leached cladding and correcting for the radioisotopes in the base metal.

d. Base metal concentrations were determined on a piece of cladding with less than 0.05 mm removed from the surface.

e. Base metal (outside 0.12 mm) was determined by preferentially dissolving the inside surface of a clad sample (with all surface oxide removed) until only 0.12 mm remained. This was then dissolved for analyses.

f. Total alpha measurements are affected by self shielding and are only an approximation to insure that all alpha emitters have been included in separate analyses.

g. Cm² always refers to the area of the fuel-clad interface.

h. The Pu isotopic analysis for the residual surface fuel was 1.86 atom% ²³⁸Pu, 49.4 atom% ²³⁹Pu, 22.0 atom% ²⁴⁰Pu, 15.6 atom% ²⁴¹Pu, and 11.2 atom% ²⁴²Pu. The Pu isotopic analysis from the residual uranium irradiated in base metal was 1.01 atom% ²³⁸Pu, 51.9 atom% ²³⁹Pu, 19.7 atom% ²⁴⁰Pu, 16.5 atom% ²⁴¹Pu, and 10.9 atom% ²⁴²Pu.

i. Decay-corrected to June 1975.

insure radiological control within the hot cell. The study of these systems will be complete within 10 days.

During the exhaust system study, fabrication and installation of the prototype decontamination will continue. Figure 5 is a flow schematic of the decontamination facility to be installed in the 100K Area hot cell.

DENSIFICATION PROCESS - R. G. Nelson

The melting of chopped Zircaloy-4 (unirradiated) cladding hulls (Inductoslag Heat Number 431) and subsequent extrusion has been described in a previous report.⁽²⁾ The extruded

3.8 cm OD x 2.5 cm ID tube blank was reduced by a commercial cladding tube manufacturer to a finished size of 11.18 mm OD x 0.74 mm wall.

Ultrasonic testing showed that this tubing would normally be rejected for cladding use. Metallographic and SEM examination (Figure 6) of the rejectable portions of the tubing showed small spherical particles were partially imbedded in the ID of the tubing. Microprobe analysis identified these as zirconium or zirconia particles. Ingot chemistry, hardness, tubing performance during fabrication, and the lack of any

FIGURE 5. Flow Schematic of the Decontamination Facility

Neg B-10332

1000X

SEM

Neg 4K1257B

250X

"B" Etch

Neg 4K1256A

100X

As Polished

FIGURE 6. ID Surface Imperfections on Zircaloy-4
Refabricated Clad Tubing

other observable defects indicate that improper billet inspection and cleaning were the probable causes for the poor ID surface.

Samples of the etched recycled Zircaloy tubing were tested in a standard 400°C - 1500 psi autoclave test for 72 hr. The four samples gained weight between 16.2 and 17.2 mg/dm² (16.5 mg/dm² average), which is a normal weight gain. All surfaces were black and lustrous.

A 34-kg heat (Number 460) consisting of three sections, (a) about 7 kg of alloy CF3 (304 stainless steel casting grade), (b) 14 kg of a 2 to 1 by weight mixture of CF3 and Inconel 718, and (c) 14 kg of pure Inconel 718 has been melted by the U.S. Bureau of Mines Inductoslag process.⁽³⁾ Figures 7 through 11 show sections of this ingot. The pure CF3 (casting Grade 304) and the Inconel 718 are of adequate quality for storage or refabrication. The 67 wt% CF3 - 33 wt% Inconel 718 material in Figures 7 and 9 are suitable for

storage; however, improved melting techniques will be required for complete homogeneity. The shrink cavity at the top of the pure Inconel 718 material (Figure 10) is the result of a sudden power outage during melting. This cavity can be minimized by a scheduled power cutback at the end of melting.

REFERENCES

1. A. M. Platt, Compiler, Nuclear Waste Management and Transportation Progress Report April through June 1975, BNWL-1936, Battelle, Pacific Northwest Laboratories, Richland, WA, pp. 41-43, September 1975.
2. A. M. Platt, Compiler, Nuclear Waste Management and Transportation Progress Report January through March 1975, BNWL-1913, Battelle, Pacific Northwest Laboratories, Richland, WA, pp. 28-33, June 1975.
3. A. M. Platt, Compiler, Nuclear Waste Management and Transportation Progress Report April through June 1975, BNWL-1936, Battelle, Pacific Northwest Laboratories, Richland, WA, pp. 47-48, September 1975.

Neg 756895-5

FIGURE 7. Longitudinal Section of 4-In. Diam Inductoslag Ingot (Heat Number 460)

Neg 756895-4

FIGURE 8. Longitudinal Section of 4-In. Diam Inductoslag Ingot (Heat Number 460) of Pure Inconel 718

Neg 756895-2

FIGURE 9. Transverse Section of 4-In. Diam Inductoslag Ingot
(Heat Number 460) of CF3 (Casting Grade 304 SS)

Neg 756895-3

FIGURE 10. Transverse Section of 4-In. Diam Inductoslag Ingot (Heat Number 460) of 67 wt% CF3 (Casting Grade 304 SS) - 33 wt% Inconel 718

Neg 756895-1

FIGURE 11. Transverse Section of 4-In. Diam Inductoslag
Ingot (Heat Number 460) of Pure Inconel 718

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

3.0 PYROLYSIS-INCINERATION OF COMBUSTIBLE ALPHA WASTES

L. K. Mudge and P. C. Walkup - 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 ultimate 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

Modification of the demonstration unit was completed as required to overcome problems that occurred during operation of the unit. Changes include relocation of the off-gas line outlet, installation of an air flow controller, installation of steam addition lines, reinforcement of the distributor support, and re-design of the distributor plate.

Analysis of ash collected during the run indicates a tap density of 0.76 g/cm³ and a carbon content of 13 wt%. This represents a 99.5% burnup of the simulated waste, which had an ash content of 3.6 wt%. The volume reduction factor for the simulated waste of bulk density 0.13 g/cc is calculated to be 140.

TECHNICAL PROGRESS

PILOT PLANT STUDIES - L. K. Mudge, G. L. Roberts, G. F. Schiefelbein, G. E. Stegen, R. G. Sullivan, and R. A. Walter

Ash collected during the end of run 4 reported in the last quarterly

report appeared to be residues from the simulated waste. The ash, shown in Figure 12, was dark gray in color and crystalline. Carbon content of the ash, determined by firing a sample at 950°C for 2 hr, was 13%. This amount of carbon in the ash represents 1/2% of the combustible portion of the original simulated waste, which contained 3.6% ash, determined as follows:

$$\frac{\text{carbon in ash}}{\text{carbon in waste}} \cdot 100 = \frac{0.13 \text{ g C}}{0.87 \text{ g ash}} \times \frac{0.036 \text{ g ash}}{\text{g waste}} \times 100 = 0.5.$$

The tap density of the ash was 0.76 g/cc. For simulated waste of bulk density 0.13 g/cc and ash content of 3.6%, the volume reduction factor is 140 determined as follows:

$$\text{Volume Reduction Factor} = \frac{\text{volume residue}}{\text{volume waste}} = \frac{0.76 \text{ g residue}}{\text{cc residue}} \times \frac{0.87 \text{ g ash}}{1 \text{ g residue}} \times \frac{\text{g waste}}{0.036 \text{ g ash}} \times \frac{\text{cc waste}}{0.13 \text{ g waste}} = 141.$$

A plugging problem in the off-gas line was encountered during run 4 and is shown in the photograph of the inside of the top of the reactor after run 4 in Figure 13. The line is

FIGURE 12. Ash Obtained from Pyrolysis-Incineration of Simulated Alpha Waste

plugged mainly with rags from feed that is added to the reactor at a point close to the off-gas line. The distributor plate, usually left rotating in this region of the reactor during feed addition, sweeps the feed past the gas outlet where the light components, such as rags, are swept into the line. A further problem stems from the design of the distributor plate, shown in Figure 13 in the condition after run 4, which allows material to accumulate on top of the plate and be lifted to the top of the reactor where the scrapers remove the accumulated material. The material then falls in close proximity to the off-gas line, and again the light components are swept into the line.

Modifications were made to relocate the off-gas line to the top cover so that material could not fall

past the gas outlet. Subsequent testing of the system showed that plugging still occurred because of material on top of the distributor plate. The distributor plate, Figure 14 was, therefore, replaced by three 1-in. x 1/2-in. arms located around the shaft at 120° angles, slanted to provide for some compaction during rotation on the waste bed and tapered to push the material to the edge of the reactor. Installation of the new distributor design will be completed in the second week of October.

The distributor shaft and seals have been replaced because of damage by scoring that occurred during operation. The distributor support has been reinforced to reduce damage to the new shaft and stabilize operation of the distributor.

Difficulty in maintaining constant air flow was encountered during operation of the unit. An automatic control valve was installed in the air inlet line to correct this difficulty.

Temperatures above 1200°C were noted in the combustion zone of the reactor. The capability for steam addition to the reactor was installed to test the effectiveness of steam addition for control of the combustion zone temperature.

ASH FIXATION STUDIES - J. E. Mendel and H. Hollis

No further ash fixation studies were conducted during the quarter. Results of previous studies were included in a paper presented at the 80th National American Institute of Chemical Engineers Meeting in Boston, September 7-10, 1975, entitled, "Glass Forms for Alpha Waste Management."

FIGURE 13. Top of Reactor Cpened at Erc of Run 4

FIGURE 14. Distributor at the End of Run 4

4.0 TRITIUM SEPARATION AND FIXATION

L. L. Burger - Project Manager

The goals of the program are to evaluate the tritium problem in the nuclear fuel cycle and to examine methods of fixing tritiated water for permanent storage.

SUMMARY

The apparatus for studying the formation of zirconium hydride as a tritium storage method is ready for testing. Some of the experimental factors are discussed.

Three procedures for the fixation of tritium in organic medium were conceived and examined. Catalytic tritiation of styrene monomer and of polystyrene, and the reaction of water with an isocyanate to form a urea were found to be highly efficient and simple methods for tritium fixation amenable to the special problems of handling of radioactive material.

TECHNICAL PROGRESS

TECHNICAL ALTERNATIVES DOCUMENT -

L. L. Burger

The comprehensive evaluation of the alternatives in radioactive waste management in the nuclear fuel cycle is divided into a number of tasks. The one on gases has been assigned to Allied Chemical, Idaho Falls, who in turn have requested that we contribute to the tritium section. A meeting was held in Denver to coordinate the work. We have prepared an outline of the tritium section and writing will commence in October.

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

In previous work we developed several procedures⁽¹⁾ for the fixation of tritium in organic media. This work led us to search for simpler and more efficient processes more amenable to the handling of radioactive materials. Accordingly, several processes were conceived that allow single-step fixation of tritium (in the form of water or hydrogen) and which eliminated the disadvantages of previous methods, i.e., incomplete reaction of radioactive monomers during polymer formation, complex processes involving the transfer and reactions of gaseous radioactive precursors (such as tritiated acetylene).

The first method is the reaction of tritium gas directly with polystyrene in solution in the presence of a rhodium-on-alumina catalyst.⁽²⁾ The reduction of some of the phenyl rings to cyclohexane rings proceeds at room temperature and atmospheric pressure. Evaporation of the solvent after filtration to remove the catalyst gives crystalline polystyrene-polyvinylcyclohexane. The method has the advantage of simplicity (one-step fixation) in a polymer whose phenyl rings impart stability to radiation.

Furthermore, the procedure allows mild conditions of hydrogenation which eliminate the problem of loss of tritium known to occur through the walls of vessels under high pressures and temperatures.

The second process examined this quarter is the direct incorporation of THO in a substituted urea by reaction with an isocyanate. Treatment of a solution of phenylisocyanate in dioxane with a slight excess of water results, after about 1/2 hr of stirring, in the precipitation of 1,3-diphenylurea in >98% yield. Equation 1-3 depicts the reaction sequence leading from phenylisocyanate to 1,3-diphenylurea.⁽³⁾ The powdered urea is compacted into a suitable

container which is encased in polymethylmethacrylate or polystyrene. This procedure has been tested without tritium, and is now being repeated with tritiated water for leach testing and testing of structural integrity of the containment material. The procedure is a rapid, quantitative and simple procedure for the fixation of water. The cost of the organic medium limits its use to highly concentrated tritiated water. A further disadvantage may be the relatively low boiling point of the product urea (262°). It may be desirable to prepare derivatives of the urea with polar substituents to render it less volatile.

A final method now being tested is the incorporation of tritium in an organic monomer by catalytic tritiation using a homogeneous catalyst. Tests are now under way to determine the lifetime of the catalyst, to recover and regenerate the catalyst, and to find optimal conditions for the conversion of the tritiated product into a polymer. Preliminary results will be reported later.

All of these procedures provide highly efficient and simple techniques for the fixation of tritium.

METAL HYDRIDE FIXATION -

R. D. Scheele, L. L. Burger

The experimental apparatus to be used for the preparation of the zirconium hydride and tritide is a modified version of the Sievert's apparatus described by Blackledge.⁽⁴⁾ To provide maximum safety during the hydriding operation, a metal system was prepared. A Lindburg tube resistance furnace is being used to heat the zirconium sample between 400° to 1000°C. The system is now complete and ready for testing.

Zirconium samples will first be degassed at elevated temperatures and high vacuums.⁽⁴⁻⁷⁾ Following this step, hydrogen and/or tritium is reacted with the zirconium⁽⁴⁻⁸⁾ at various partial pressures of hydrogen 100 to 900 torr. The amount of hydrogen absorbed by the zirconium will be monitored by the amount of hydrogen added to the system and the subsequent pressure drop, and confirmed by the sample's gain in mass.

Study of the phase diagram (Figure 15) for zirconium hydride^(5,9)

FIGURE 15. Zirconium-Hydrogen Phase Diagram⁽¹²⁾

shows five phases. The alpha phase is hydrogen saturated metal,⁽¹⁾ the beta phase is the hydrogen saturated high-temperature phase,⁽⁴⁾ the delta phase is the hydride with H/Zr ratio of 1.4 to 1.6,⁽⁵⁾ epsilon is ZrH_{1.8} to ZrH₂; the last phase, gamma, is a metastable hydride coexisting with alpha and delta phases. The stability of the gamma phase is controversial.^(5,10)

The delta and epsilon phases are the only two phases which exist from 900°C to room temperature and are therefore of primary interest as a storage form for tritium. The delta phase has been shown to provide a more monolithic and uniform massive hydride than does the epsilon phase.

High concentrations of hydrogen cause cracking and thereby undermine the product's integrity and usefulness as a storage material.

Delta phase zirconium hydride will be prepared initially by degassing 0.50-in. lengths of 0.25- and 0.75-in. diam zircaloy-2 and 0.50-in. diam zircaloy-4 rods, at 600°C or higher for 2 to 4 hr. The hydrogen will then be introduced until a partial pressure of 100 torr is reached. The pressure will be maintained at this point until a H/Zr ratio of 1.4 to 1.6 is achieved.⁽⁸⁾

Potential problem areas include the effect of impurities, hydriding rates, pyrophorocity, and general physical and chemical stability especially after long storage (and decay) times, tritium leaching, and process design. The economic attractiveness of metal hydrides for tritium storage depends on the use of cladding hulls as the source of metal. Study of the treatment of cladding hulls is already under way in another PNL program.

REFERENCES

1. J. A. Franz and L. L. Burger, Polymeric Media for Tritium Fixation, BNWL-430, Battelle, Pacific Northwest Laboratories, Richland, WA, May 1975.
2. For examples of extremely mild reductions of aromatic rings, see leading references in Modern Synthetic Reactions, H. O. House, W. A. Benjamin, Inc., Menlo Park, CA, 1972, p. 13.
3. Phenylisocyanate finds limited applications as a dehydrating agent in organic reactions. See Reagents for Organic Synthesis, vol. 1, L. F. Fieser and M. Fieser, John Wiley and Sons, Inc., New York, NY, 1967, p. 842.

4. H. M. Mueller, J. P. Blackledge, and G. G. Libowitz, Metal Hydrides, Academic Press, 1968.
5. P. T. Gilbert, Jr., Zirconium Hydride, A Compendium on the Systems Zirconium-Hydrogen and Hafnium-Hydrogen and Related Topics, Atomics International, 1956.
6. M. J. Trzeciak, D. F. Dilthey, M. W. Mallet, Study of Hydrides, BMI-1112, Battelle Memorial Institute, Columbus, OH, 1956.
7. M. Steinberg, R. Wiswall, et al., Tritium Concentration and Fixation, Progress Report, January-March 1974, BNL 18879, Brookhaven National Laboratory, Upton, NY.
8. M. Steinberg, R. Wiswall, et al., Tritium Concentration and Fixation, Progress Report, April-June 1974, BNL 19303, Brookhaven National Laboratory, Upton, NY.
9. R. L. Beck, "Zirconium-Hydrogen Phase System," Trans. American Society for Metals, vol. 55, 1962.
10. K. G. Barraclough, C. J. Beevers, The Nature of the γ Phase in Zirconium-Hydrogen Alloys. I. Less Common Metals, vol. 35, p. 177, 1973.
11. R. L. Beck and W. M. Mueller, "Zirconium Hydrides and Hafnium Hydrides," in W. M. Mueller, J. P. Blackledge and G. C. Libowitz, Metal Hydrides, Academic Press, 1968.

5.0 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; 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

Development of a long-range plan for the D&D of retired ERDA facilities at Hanford was continued with the identification of several alternative cases and commencement of work on evaluation of those cases.

Activity specifications for D&D of the 233-S Building were completed and approved by PNL and ARHCO. Work has begun on installation of an experimental burial trends, for demonstration of stabilization techniques.

Further studies were made of chemical and electrochemical decontamination of stainless steel.

TECHNICAL PROGRESS

TASK 1 - ESTABLISH METHODS, COSTS AND PRIORITIES FOR D&D AT HANFORD -

K. M. Harmon, B. W. Mercer, and H. T. Fullam

Because of the large number and variety of the retired, contaminated sites and facilities at Hanford, preparing a long-range plan for D&D at Hanford is proving to be highly complex. To provide a basis for selection among a variety of D&D alternatives, several cases were defined and work started on their evaluation.

The options, described in more detail in Table 4 include:

1. Dedication of the entire reservation for nuclear purposes.
2. Cleanup of the areas near the Columbia River (100 and 300 Areas) and dedication of the 200 Areas for indefinite use for nuclear activities, including the storage or burial of wastes.
3. Cleanup of all retired ERDA facilities and sites on the Hanford Reservation
 - a) As quickly as manpower and equipment permit
 - b) On a schedule compatible with the goal of minimizing costs and total radiation exposure. This option assumes that maximum advantage will be taken of delays in disposition action until short-lived isotopes have decayed to innocuous levels.

Evaluation of the cases is proceeding as follows:

1. A "typical" site or facility has been selected for each facility class.
2. Disposition alternatives for each class of facility are being reviewed. Where more than one op-

TABLE 4. Scenarios for D&D at Hanford

CASE 1	CASE 2	CASE 3
DEDICATION OF THE RESERVATION FOR NUCLEAR SERVICE	UNRESTRICTED RELEASE OF 100 AND 300 AREA SITES; DEDICATION OF THE 200 AREAS FOR NUCLEAR SERVICE	UNRESTRICTED RELEASE OF THE HANFORD RESERVATION
<p>OBJECTIVE. Prepare the Hanford Reservation for dedication for indefinite-length service as a site for continued nuclear activities, including the storage and/or disposal of nuclear wastes.</p>	<p>OBJECTIVE. Except for the 200 Areas, prepare all retired contaminated ERDA facilities and sites on the Hanford Reservation for unrestricted public access and use. Prepare the 200 Areas for indefinite-length service as a site for continued nuclear activities, including the storage and/or burial of nuclear wastes; reduce the number and extent of contaminated sites on the 200 Areas plateau, as far as practical.</p>	<p>OBJECTIVE. Prepare all retired contaminated ERDA facilities and sites on the Hanford Reservation for unrestricted public access and site use.</p>
SCENARIOS	SCENARIOS	SCENARIOS
<p>1.1 <u>All Sites and Facilities: Layaway</u></p> <p>Leave the contamination in place and provide normal maintenance/surveillance as long as required.</p>	<p>2.1 <u>100/300/600 (Selected) Area Sites and Facilities: Decontaminate or Dismantle</u></p> <p>Apply one of the three scenarios identified under Case 3. Relocate all contaminated wastes produced by D&D activities to the 200 Areas.</p>	<p>3.1 <u>All Sites and Facilities: Decontaminate or Dismantle as Soon as Possible</u></p> <p>Proceed with disposition of all facilities and sites as rapidly as possible with available manpower and equipment. Package all wastes for transport offsite.</p>
<p>1.2 <u>All Sites and Facilities: Protective Storage or Entombment</u></p> <p>Prepare the sites and facilities to remain in place with minimal additional maintenance and surveillance costs, until the activity has decayed to innocuous levels or until disposition is undertaken. This may require decontamination or removal of selected structures and hardening of ground disposal or storage sites.</p>	<p><u>200 Areas: Layaway</u></p> <p>Leave the contamination in place and provide normal maintenance/surveillance as long as required. (Same as Case 1.1)</p> <p>2.2 <u>100/300/600 (Selected) Area Sites and Facilities: Decontaminate or Dismantle (Same as 2.1)</u></p> <p><u>200 Areas: Protective Storage or Entombment</u></p> <p>Prepare the sites and facilities to remain in place with minimal additional maintenance and surveillance costs, until the activity has decayed to innocuous levels or until disposition is undertaken. This may require decontamination or removal of selected structures and hardening of ground disposal or storage sites. (Same as Case 1.2)</p>	<p>3.2 <u>All Sites and Facilities: Use Selective Delay in Decontamination or Dismantling to Reduce Costs</u></p> <p>Proceed with disposition of all facilities and sites contaminated with long-lived radionuclides (Pu, ¹⁴C, ¹²⁹I) as rapidly as possible. Facilities contaminated only with short-lived isotopes ($T_{1/2} < 31$ years) may be prepared for protective storage, entombed, or disposed of on a schedule designed to minimize total costs (interim maintenance plus final disposition costs) and radiation exposure (occupational plus public). Package all wastes contaminated with long-lived nuclides for transport offsite.</p> <p>3.3 <u>All Sites and Facilities: Use Selective Delay in Decontamination or Dismantling to Minimize Costs</u></p> <p>Proceed with D&D at a pace which will minimize total costs and total radiation exposure. All long-lived radionuclides are to be removed ultimately, but disposition may be delayed until short-lived nuclides ($T_{1/2} < 31$ years) have decayed to innocuous levels. Treat facilities and sites contaminated only with short-lived nuclides as in Case 3.2. Package all wastes containing long-lived nuclides for transport offsite.</p>

- tion exists for a given D&D case, an alternative will be selected. Major activities required for each case will be defined in enough detail to permit cost estimating.
3. Release criteria corresponding to various potential site uses or waste disposal modes will be established, D&D cost estimates made, and hazards evaluated for each of the "typical" sites and each of the pertinent D&D-at-Hanford cases.
 4. The results of the above studies will be used in setting priorities for D&D actions at Hanford.

D&D Costs and Priorities - J. C. King, J. W. Litchfield, and C. L. McDonald

Work started on preparing a computer-based information system for the contaminated facilities at Hanford, to simplify future prioritization studies. The ALADIN language of the INFONET System was selected, being among the most flexible and dependable and requiring the least amount of program development. The system can be operated in either interactive or batch mode, using a wide variety of input or output devices, including the portable "Execuport" terminal. Simple, one-word commands are used to sort the data base using one or multiple criteria, to print lists and to update or modify the data base. In addition, an easy-to-use, flexible report generator is available. Computational subroutines (facility prioritization and cost estimating subroutines, for example) can easily access, as well as store computational results in the data base.

Identification of desirable information to store within the system has been completed, totalling 76 data fields for each facility. Contaminated Liquid Disposal Site information will be entered into the system initially, with 45 fields of data for each facility having been coded and keypunched at present. The programs for file creation also have been written.

A plan was formed for estimating cost for the various options to be studied. To cope with the large number of facilities involved, "unit cost" estimating will be employed. Detailed cost estimates will be prepared for representative facilities within each facility class, using cost accounting categories applicable to remaining facilities within the class. For each accounting category, costs will be reduced to an appropriate unit basis. Costs will then be scaled to remaining facilities within the facility class.

Development of a D&D information system subroutine to support the Churchman-Ackoff prioritization methodology was considered. This approach was found feasible and will be employed for prioritization.

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

J. R. Carrell, C. A. Allen, D. E. Blahnik, and P. L. Koehmstedt

Activity Specifications for D&D of the 233-S Building were completed and approved by PNL and ARHCO.

Milestone charts representing the activities so far identified were prepared independently by PNL and ARHCO

and reconciled. The Activity Specifications and schedules will be included in the Program Plan now planned for release in October.

TASK 3 - EXPLORATORY R&D -

K. M. Harmon

Demonstration of Techniques for Stabilization of a Waste Burial Ground - E. L. Klepper

Work has begun on installing the experimental burial trench outside of the 200 West Area. The work is being done by J. A. Jones in conjunction with an experimental set of plots for ARHCO. A work order to Jones for \$15K was written for this project. Work should be completed by October 1 so that the surface can be planted to cheat grass. Installation of the fence has been delayed until next spring.

Decontamination of Stainless Steel - J. A. Partridge

Improved Pu decontamination conditions with stainless steel were achieved this month using two different approaches. The first approach was to vary the composition of the Meservey-type solution (ammonium oxalate, citrate, hydrofluoric acid, hydrogen peroxide). In tests in which the HF concentration was 0.5M, Pu DFs of 400 $\geq$ 1700 were achieved, compared to DFs of 20 to 100 observed previously in the same type of tests at 0.1M HF.

Hydrogen peroxide concentration has an effect, but it appears that our previous tests at 0.01 to 0.02M peroxide were in the optimum range. A test this month at 0.05M peroxide gave a Pu DF of only 9, and a test in which there was no peroxide resulted in a Pu DF of 10.

The pH of the Meservey solution in the range of pH 2 to 4 appeared to have little effect. No definite trend of Pu DF as a function of pH was obtained from the test results.

Even though the Pu decontamination factors achieved with the Meservey-type solutions at 0.5M HF are significantly higher than those achieved earlier at 0.1 to 0.2M HF, this solution appears to have no advantage (with respect to Pu DF) over nitric-hydrofluoric acid for decontaminating the stainless steel surfaces we have been testing.

The second experimental approach which resulted in good Pu DF was to use an electropolishing technique. Tests were made using five different aqueous solutions: 1) a Meservey solution, 2) 80% H_3PO_4 , 3) 55% H_3PO_4 -15% H_2SO_4 , 4) 8M HNO_3 , and 5) 8M HNO_3 -0.1M HF. Solutions 2) and 3) are being used by ARHCO personnel to decontaminate tools electrochemically. The test solutions were maintained at 50 to 60°C during the 20-min tests. The current was maintained at 1 amp (current density of approximately 100 amps/ft²) on the 1/2-in. pieces of stainless steel tubing. The best results were obtained with solutions 4) and 5), which gave Pu DFs of 6000 and 2600, respectively. The final Pu count on each decontaminated piece was near background level. Thus caution needs to be exercised in the use of the actual magnitude of these DF values since they could be in error by $\pm$ 50 to 100%. However, the results are very encouraging because they demonstrate a very high Pu DF with an aqueous solution (nitric acid) that is relatively simple to dispose of.

6.0 TRANSPORTATION SAFETY STUDIES

L. D. Williams - Project Manager

The objective of the Transportation Safety Studies Program is to develop and use a model for calculating the risks associated with the shipping of radioactive materials.

SUMMARY

Report BNWL-1846, Assessment of the Risk of Transporting Plutonium Oxide and Liquid Plutonium Nitrate by Truck, is being printed and will be distributed in mid-October.

A preliminary assessment of the risk in the shipment of plutonium oxide and liquid plutonium nitrate by rail has been completed and is currently undergoing second party review.

System descriptions have been completed for use in assessing the risks in the shipment of non-high-level wastes. Three types of waste (β - γ , α , and α - β - γ) are being considered in the assessment.

The risk analysis code HEAD has been modified to increase its utility.

TECHNICAL PROGRESS

Risk Assessment of Plutonium Transport by Truck

The report Assessment of the Risk of Transporting Plutonium Oxide and Liquid Plutonium Nitrate by Truck (BNWL-1846) is currently being printed and will be distributed in mid-October. Numerous revisions were made in the report in response to comments of internal and external reviewers of an earlier draft. The changes improved the clarity of presentation and resulted in increased

rigor; however, they did not significantly affect the results of the analysis.

Risk Assessment of Plutonium Transport by Rail

A preliminary assessment of the risk of transporting plutonium oxide and liquid plutonium nitrate by rail was reported in the last Quarterly Report. The risk assessment is currently undergoing internal quality assurance review. Event probabilities, failure thresholds, and fault trees are being second-party reviewed for correctness and completeness. Reviews completed to date indicate that some event descriptions in the fault trees are hard to understand. This problem results from the complexity of some events together with the limited number of descriptive words permitted by the fault tree drawing program, ACORN. Event descriptions are being revised to improve clarity.

Risk Assessment of Non-High-Level Waste Shipments

System descriptions for use in the non-high-level waste shipment analysis have been completed. The waste quantities which will be used in the analysis correspond to those that will be shipped annually from 100

nuclear power plants. For The analysis three types of non-high-level waste are considered: β - γ waste, α waste, and α - β - γ waste.

The β - γ waste is produced at fuel fabrication facilities, nuclear power plants, and fuel reprocessing facilities. The waste contains less than 10 $\mu\text{Ci/kg}$ of alpha decaying isotopes. The level of β - γ radiation is low enough that, in general, special shielding and containers are not required. The assumed β - γ systems description is shown in Table 5. The table gives the amounts of material shipped and the system characteristics.

The α -waste is produced at fuel fabrication facilities and fuel reprocessing plants. The alpha activity level of the waste is greater than 10 $\mu\text{Ci/kg}$, with the β - γ level low enough to allow packaging in unshielded drums. The alpha waste system description is given in Table 6. The packaged α waste is assumed to be shipped in an ATMX-600 railcar. These specially designed railcars act as an additional barrier to the release of the waste.

The α - β - γ waste is produced at fuel reprocessing plants. It contains greater than 10 $\mu\text{Ci/kg}$ of alpha activity and has a β - γ activity level that requires shielding. It is shipped in special containers similar to irradiated fuel shipping casks. The quantities and other information about the α - β - γ waste system are found in Table 7.

The three tables will be used as bases for risk assessments of non-high-level waste shipments.

Risk Analysis Computer Code Development

In the course of using the risk analysis computer code HEAD, several obstacles to efficient use of its output were discovered. These included output format, designation of units for numerical values, and the need to interpolate between output values. Several modifications to HEAD were made to alleviate these problems.

TABLE 5. Beta-Gamma Waste System Description

	Beta-Gamma		
	Fabrication	Power Plants	Reprocessor
Characteristics	Paper and rags 39.8% Plastic 20.6% Rubber 18.6% Metal 10.3% Glass 3.6% Misc. 6.8%	Same as Fab. plus resins sludges filters	Same as Fab.
Activity	0.001 Ci β - γ /ft ³	1 Ci β - γ /ft ³ or 0.5 Ci/drum	0.001 Ci/ft ³
Major Isotopes	¹⁰⁶ Ru, ⁹⁵ Zr, ⁹⁵ Nb, ¹³⁷ Cs, ¹⁴⁴ Ce, ⁹⁹ Tc, etc.	⁵⁴ Mn, ⁵⁵ Fe, ⁵⁸ Co, ⁶⁰ Co ¹³⁴ Cs, ¹³⁷ Cs	¹³⁴ Cs, ¹³⁷ Cs
Container Used	55 gallon, 17C	55 gallon, 17C	55 gallon, 17C
Mode of Transport	Truck	Truck	Truck
Destination	Burial Site	Burial Site	Burial Site
Waste Prepared	Compacted 5X	39% Compact 5X, 49% fixed in cement Vol. increase by 4X(a) 12% increase by 2.5(b)	Compacted 5X
Amount Untreated	802,893 ft ³	400,000 ft ³	107,107 ft ³
Amount after Treatment	160,579 ft ³	934,178 ft ³	21,421 ft ³
No. of 55 Drums at 7.35 ft ³ /drum	21,848	127,099 (4237 at 0.5 Ci/drum)	2,915
No. of Shipments at 50/shipment	437	2,542	59
	500 lb/drum	4,237 barrels at 500 lb 16,580 bar. at 782 lb 106,282 bar. at 908 lb Typical Shipment Load 3.3% 500 lb drums 13.0% 782 lb drums 83.6% 908 lb drums	500 lb/drum

a. Volume increase due to mixing waste with cement 1:3 ratio.

b. Volume increase due to placing material inside concrete annulus in drum, which decreases the amount of waste each drum can hold.

TABLE 6. Alpha Waste System Description

	Alpha	
	Fabrication	Reprocessor
Characteristics	Same as β - γ	Same as β - γ
Activity	$>10 \mu\text{Ci/kg}$	Same as Fab.
Major Isotopes	0.25 g/ft^3 Pu or U	Same as Fab.
Container	Polyethylene lined 17C 55-gal. drum placed in ATMX railcar	Same as Fab.
Mode of Transport	Rail	Rail
Destination	Federal Repository	Federal Repository
Waste Prepared	Compaction 5X	Compaction 5X
Amount Untreated	$302,734 \text{ ft}^3$	$40,385 \text{ ft}^3$
Amount after Treatment	$60,546.8 \text{ ft}^3$	$8,077 \text{ ft}^3$
No. of 55 drums at $7.35 \text{ ft}^3/\text{drum}$	8,238	1,099
No. of Shipments at 140/shipment	59	8
	500 lb/drum	500 lb/drum

TABLE 7. Alpha-Beta-Gamma Waste System Description

	Alpha-Beta-Gamma
	Reprocessor
Characteristics	Same as α
Activity	$>10 \mu\text{Ci } \alpha/\text{kg}$ $10\text{-}1000 \text{ mrem/hr}$ on surface of container
Isotopes	0.025 g/ft^3 Pu or U mixed fission products
Container	Steel container 2 ft OD 10 ft length 3 containers/cask
Mode of Transport	Truck
Destination	Federal Repository
Waste Prepared	Compaction 5X
Amount Untreated	140,000
Amount after Treatment	28,000
No. of Containers at $25 \text{ ft}^3/\text{container}$	1,120
No. of Shipments at 3 containers/shipment	374
	55,000 lb/3 containers and 1 cask

DISTRIBUTIONNo. of
CopiesOFFSITEUNITED STATES

- 1 ERDA Chicago Patent Group
9800 South Cass Avenue
Argonne, IL 60439

A. A. Churm

- 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

- 1 NRC Division of Materials and
Fuel Cycle Facility Licensing
Washington, DC 20555

Chief, Waste Management Branch

W. P. Bishop

- 1 ERDA Division of Biomedical and
Environmental Research
Earth Sciences Branch
Washington, DC 20545

W. G. Belter

- 4 ERDA Division of Environmental
Control Technology
Washington, DC 20545

H. Glauberman

H. Hollister

M. S. Kearney

W. E. Mott

- 8 ERDA Division of Nuclear Fuel
Cycle and Production
Washington, DC 20545

F. P. Baranowski

G. H. Daly (2)

O. P. Gormley

A. F. Perge

R. W. Ramsey (3)

No. of
Copies

- 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

- 2 ERDA Idaho Operations Office
P.O. Box 2108
Idaho Falls, ID 83401

K. K. Kennedy

W. F. Hendrickson

- 2 ERDA Oak Ridge Operations Office
P.O. Box X
Oak Ridge, TN 37830

E. H. Hardison

O. T. Turmelle

- 1 ERDA Savannah River Operations
Office
P.O. Box A
Aiken, SC 29801

R. L. Chandler

- 200 ERDA Technical Information
Center

- 3 Allied Chemical Corporation
550 - 2nd Street
Idaho Falls, ID 83401

J. A. Buckham

B. C. Musgrave

C. M. Slansky

- 1 Allied-Gulf Nuclear Services
P.O. Box 847
Barnwell, SC 29812

A. Schneider

- 2 Argonne National Laboratory
9700 South Cass Ave.
Argonne, IL 60439

N. M. Levitz

M. J. Steindler/

L. E. Trevorrow

No. of
Copies

- 1 Battelle Memorial Institute
505 King Ave.
Columbus, OH 43201

E. S. Bartlett
- 2 Brookhaven National Laboratory
Research Library,
Reference Section
Information Division
Upton, Long Island, NY 11973

J. J. Reilly
R. H. Wiswall
- 1 Brookhaven National Laboratory
Energy Systems Library
Building 902
Upton, NY 11973

Chaim Braun
- 1 Combustion Division
Combustion Engineering, Inc.
Windsor, CT 06095
- 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 Ave.
P.O. Box 10412
Palo Alto, CA 94304

L. Henning
M. Levenson
E. Zebroski

No. of
Copies

- 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
- 1 Environmental Protection Agency
5555 Ridge Ave.
Cincinnati, OH 45213

R. E. Landreth
- 1 General Electric Company
175 Curtner Ave.
(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

No. of
Copies

- 3 Los Alamos Scientific Laboratory (ERDA)
P.O. Box 1663
Los Alamos, NM 87544
- C. W. Christenson
L. J. Johnson
H. S. Jordan
- 2 Monsanto Research Corporation
Mound Laboratory
P.O. Box 32
Miamisburg, OH 45342
- C. J. Kershner
P. Lamberger
- 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

No. of
Copies

- 1 Nuclear Water and Waste Technology, Inc.
P.O. Box 6406
San Jose, CA 95150
- W. L. Pearl
- 1 NYS Atomic and Space Development Authority
230 Park Avenue, Rm. 2425
New York, NY 10017
- J. G. Cline, General Manager
- 1 Princeton University Center for Environmental Studies, Engineering School
Princeton, NJ 08540
- Frank von Hippel
- 2 Sandia Laboratories
Albuquerque, NM 87107
- J. Sivinski
W. Weart
- 1 Transnuclear, Inc.
One North Broadway
White Plains, NY 10601
- B. R. Teer
- 6 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
W. C. Ulrich
- 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

No. of
Copies

No. of
Copies

FOREIGN

<p>1 <u>EURATOM</u>
Health Physics Division
29, Rue Aldringer
Luxembourg, BELGIUM</p> <p>Rene Amavis</p> <p>1 <u>Atomic Energy Research Establishment</u>
Harwell, Didcot,
Berks, ENGLAND</p> <p>K. D. B. Johnson</p> <p>1 <u>United Kingdom Atomic Energy Authority</u>
Risley, ENGLAND</p> <p>D. W. Clelland</p> <p>1 <u>Centre de Marcoule</u>
B. P. 106
30 - Bagnols S/Ceze
FRANCE</p> <p>R. Bonniaud</p> <p>3 <u>Centre d'Etudes Nucleaires de Fontenay-aux-Roses</u>
Boite Postale 6
92 - Fontenay-aux-Roses
FRANCE</p> <p>P. J. Regnaut
J. Sauteron
Y. J. Sousselier</p> <p>3 <u>Institut für Chemische Technologie</u>
Kernforschungsanlage
Julich GmbH
D517 Julich
Postfach 365
GERMANY</p> <p>E. R. Merz
K. H. Rattay
W. Heimerl
(Postfach 1913)</p> <p>1 <u>KEWA Kernbrennstoff-Wiederaufarbeitungs-Gesellschaft m.b.H.</u>
Projektleitung
7501 Leopoldshafen
Postfach 60
GERMANY</p> <p>W. A. Issel</p> | <p>1 <u>Nuclear Research Center</u>
Waste Management Dept.
D75 Karlsruhe
Weberstr. 5
GERMANY</p> <p>H. Krause</p> <p>1 <u>NUKEM GmbH</u>
645 Hanau 11
Postfach 110080
WEST GERMANY</p> <p>H. Witte</p> <p>2 <u>CNEN</u>
Direttore di Programma
Eurex Del CNEN
Saluggia "Vercelli", ITALY</p> <p>Giacomo Calleri</p> <p>Laboratorio di Ingegneria
Sanitaria
Via Anguillarese km 1 + 300
Roma, ITALY</p> <p>Willy Bocola</p> <p>1 <u>Laboratorio Rifiuti Radioattivi</u>
C.N.E.N. - C.S.N. Casaccia
Casella Postale 2400
00100 Rome, ITALY</p> <p>Ferruccio Gera</p> <p>2 <u>Bhabha Atomic Research Centre</u>
Waste Treatment Division
Trombay, Bombay, INDIA</p> <p>N. S. Sunder Rajan
K. T. Thomas</p> <p>1 <u>C.W.M.F. Project</u>
P.O. Kalpakkam
Chingleput Dist.
Tamil Nadu, INDIA</p> <p>R. V. Amalraj</p> <p>2 <u>International Atomic Energy Agency</u>
Kärtner Ring 11
P.O. Box 590
A-1011, Vienna, AUSTRIA</p> <p>1 H. F. Ramdohr
c/o Friedrich Uhde GmbH
46 Durtmund
Deggingstr. 10-12
GERMANY</p> |
|---|--|

No. of
Copies

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

1 Joint Center for Graduate Study

J. Cooper

3 United Nuclear Industries, Inc.

A. P. Larrick

2 Westinghouse Hanford Company

A. G. Blasewitz
C. R. Cooley

No. of
Copies

40 Battelle-Northwest

T. W. Ambrose
J. W. Bartlett
R. J. Bashor
C. L. Brown
L. L. Burger
H. C. Burkholder
R. E. Burns
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
J. H. Jarrett
R. S. Kemper
M. R. Kreiter
K. M. Harmon
R. C. Liikala
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