NUCLEAR WASTE MANAGEMENT AND TRANSPORTATION
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FOREWORD

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

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1.0 ADVANCED METHODS FOR MANAGEMENT AND DISPOSAL OF RADIOACTIVE WASTES

K. J. Schneider - Project Manager

The objective of this program is to systematically assess alternative systems for management of radioactive wastes resulting from nuclear fuel cycle alternatives. The information gained from these assessments will be used to develop a rationale and methodology for assistance to the eventual formulation of a comprehensive R&D plan for management and disposal of nuclear fuel cycle wastes.

SUMMARY

Overall plans for the FY75 program are being developed to emphasize overview planning for waste management R&D, development of methodologies for waste management evaluations, and assessments of waste management problems and systems.

Preliminary comparisons of assay data with ORIGEN output show that ORIGEN data are in error by -20 to +80% for transuranic content in high-level waste. Similarly, the use of BWRs and allowance for non-equilibrium nuclear fuel cycles during the next decade will result in some important differences in transuranic content in high-level waste compared to data used in BNWL-1900.

Expansion of overview R&D needs for the disposal concepts studied in report BNWL-1900 (also WASH-1297) has been initiated.

The presence of sodium chloride in water in an underground waste repository can have a detrimental effect in the adsorption of certain radionuclides by the soils in aquifers. However, the extent of this effect in practical cases has not been examined. A preliminary review of safety/risk

studies for ultimate disposal indicates the minimum effort would require modest methodology development for several years followed by significant study effort after selection of the disposal concept.

A preliminary decision analysis model using established procedures with appropriate modifications has been developed to provide for evaluation of waste disposal alternatives.

The instrument for measurement of public attitudes towards waste management factors has been upgraded.

TASK 1: PROGRAM MANAGEMENT

K. J. Schneider

Discussions with AEC staff have resulted in an updated program:

- To accentuate comprehensive R&D planning on all potential disposal concepts
- To develop methodology for public response evaluations, for concept criteria ranking, and for selected safety studies and
- To perform overview studies of selected waste management questions.

The first effort is oriented toward maintaining active cognizance of

all matters influencing waste management throughout the world. This knowledge will be used in developing an advanced comprehensive plan to implement the most promising waste management scheme.

The second effort will provide methodologies for basic evaluations which will eventually be required and for examining public attitudes toward waste management problems.

The third effort is devoted to using available information to evaluate waste management concepts and problems by use of parametric R&D analysis. This process will indicate the bounds that an approach will fall into and will also identify the R&D needed to provide data for the more complete and comprehensive analysis needed for decision making.

The basic assumptions used for the current studies are shown in Table 1. The general relationship of the AWM studies to other waste management studies is shown in Figure 1.

TASK 2: FUEL CYCLE ALTERNATIVES

R. C. Liikala

ASSESSMENT OF HIGH-LEVEL WASTE DATA

R. M. Fleischman, E. T. Merrill,
and C. M. Heeb

The objective of this task in the Advanced Waste Management (AWM) program is to provide data needed to adequately plan and evaluate the effectiveness of advanced strategies for managing nuclear wastes. Waste management strategies will be evaluated on the basis of their ability to manage wastes generated to date in the commercial sector along with wastes expected to be generated by continued

nuclear production of electricity. The responsibility for managing nuclear wastes in terms of assuring public health and safety extends far beyond the time that electricity will be produced by nuclear power plants. Judgments as to the efficacy and efficiency of a given waste management scheme must rest upon its ability to handle all waste generated by this sector of the energy economy, (i.e., from birth to death). Inability to project precisely what the growth of this sector will be does not alleviate the responsibility to project it based upon the best judgment that can be afforded.

Battelle, Pacific Northwest Laboratory (PNL) has been actively involved during the past two years in evaluation of alternative means of disposing of high-level wastes. The results of this work are summarized in WASH-1297⁽¹⁾ and documented in detail in BNWL-1900.⁽²⁾ To provide a consistent basis for analysis, estimates were made of the amount and character of high-level waste expected to be accumulated by the year 2000. These estimates were based upon a combination of assumptions. Since these projections form the source terms that feed directly into planning and analysis of waste management strategies, their periodic review for policy changes and improvements in technical bases is recommended.

Table 2 reviews some of the important technical bases used in BNWL-1900 high-level waste projections. Also listed are suggested refinements and qualitative comments regarding the nature of the waste projection problem as it is affected by

TABLE 1. Basic Assumptions for AWM Study for FY75

1. The study is aimed at all commercially-generated nuclear wastes in the United States.
2. One or more final disposal schemes will commence routine operation by the year 2010.
3. Studies will be made on alternative concepts to the point of having sufficient information for decisions on ultimate disposal.
4. Solidified HLW will begin to be delivered to an AEC Retrievable Surface Storage Facility (RSSF) in 1983. The RSSF would plan to receive waste through the year 2010, although waste receipt/storage for up to 100 years is possible.
5. R&D studies will produce parametric output as much as possible.
6. Topical reports or "white papers" will be issued as frequently as significant work packages are completed.
7. Output from other studies will be used to the maximum extent practicable.
8. The best available resources will be used to perform the studies.
9. Careful review will be made of all key program elements to assure correctness and completeness.

FIGURE 1. AWM and Overall AEC Commercial Waste Disposal Plans

TABLE 2. Review of Waste Projection Technical Bases

BNWL-1900 Technical Bases	Comments and Suggested Refinements
Electrical generating capacity based on 1972 projections of the office of Planning and Analysis	Projected generating capacity is a dynamic, driving function subject to licensing delays and changes in government policy. The assumptions should be updated yearly.
First Pu recycle assumed to begin in LWRs in 1977	Pu recycle has not yet been approved and should be monitored because of the high impact of actinide content in high-level waste.
Spent fuel isotopics discharged from cores prior to reaching equilibrium were based on the simplifying assumption that fuel discharged at a reduced amount of exposure was equivalent to a reduced amount of fuel at goal exposure (e.g. 60 MT at 10,000 MWd/T equivalent to 20 MT at 30,000 MWd/T)	Actinide content is not a linear function of exposure. Non-equilibrium discharge fuel should be characterized separately and input to waste projections.
Reprocessing plant capacity was assumed to be available to reprocess spent fuel without imposed delays.	Failure of the Midwest Reprocessing Facility assures a backlog of spent fuel and places entire reprocessing burden on the Barnwell plant. This will possibly affect Pu recycle timing and time distribution of high-level waste generation.
A single PWR model was assumed adequate to simulate discharge fuel from all LWRs.	BWR discharge fuel is significantly different from PWR fuel so that separate treatment is recommended for adequate waste characterization.
LMFBR fuel management calculations were based on AI and GE follow-on LMFBR designs.	LMFBR designs have changed significantly since the initial design studies. Fuel management calculations should be redone based on current design and advanced fuel concepts (e.g. carbide and nitride fuels) should be included.
ORIGEN calculations were used to generate discharge isotopics for LWRs, LMFBRs, and HTGRs.	The ORIGEN fuel management calculations should be checked against observed assay data for LWRs and detailed fuel management calculations for LMFBRs and HTGRs to determine the accuracy and the conservative/nonconservative nature of the method.

legislative and other socio-economic pressures. The work performed this quarter has been directed mainly at quantifying the effects of suggested improvements in the reactor fuel management models.

LWR High-Level Waste

Nonequilibrium Cases

The spent fuel concentrations listed in BNWL-1900 were based upon the simplifying assumption that fuel discharged at low exposure is equivalent to a reduced amount of fuel at goal exposure. This means, in effect, that isotopic buildup during irradiation is assumed to be linear with exposure. Whereas this may be a reasonable approximation for most fission products, it is incorrect for the transuranic isotopes.

To demonstrate the nonlinear relationship between isotopic concentrations and exposure, measured isotopic concentrations were retrieved from an early PNL analysis of Yankee-Rowe spent fuel.⁽³⁾ Table 3 gives observed actinide concentrations of Yankee-Rowe discharge fuel for exposures ranging from approximately 8,000 to 39,000 MWd/MTM. Data for key isotopes are graphically represented in Figure 2. From this figure it is apparent that low-exposure discharge concentrations of ^{239}Pu and ^{237}Np are underestimated by the linear approximation and concentrations of ^{243}Am and ^{244}Cm are overestimated.

Since low-exposure fuels are discharged during the first two or three years of operation, their importance is enhanced by the industry's growth rate. In each year prior to 1980, over 40% of the operating reactors

are projected to be less than two years old. By 1990, new reactors will still represent 20% of the total, and in the year 2000, 16% of the total.

LWR Representation

A single PWR model was used to represent all LWR discharge fuel in BNWL-1900. Whereas this approximation may be reasonable for estimating the total amount of actinide waste per unit of electricity produced, BWR discharge exposures are less than those of PWRs, and the actinide content per unit mass of waste produced will be significantly different. Variations in the neutron spectra will also cause isotopic concentrations from the two reactor types to be different.

A comparison of BWR and PWR spent fuel concentrations as calculated using the ALTHAEA code⁽⁴⁾ are summarized in Table 4. Comparisons are given for both UO_2 equilibrium and first Pu recycle conditions. PWR and BWR equilibrium discharge exposures were assumed to be 32,000 MWd/MTM and 27,000 MWd/MTM respectively. Percent variations in the spent fuel indicate the magnitude of error introduced by not representing the BWR separately.

Plutonium Recycle

The values listed in Table 4 for first Pu recycle demonstrate the sensitivity of high-level waste concentrations to the assumed starting date for plutonium recycle. The differences in actinide concentrations between the equilibrium UO_2 cycle and first Pu recycle illustrate the net effect of having 1/4 of a batch

TABLE 3. Yankee-Rowe Measured Actinides vs. Exposure⁽³⁾
(0.0001 nuclei per barn cm)

Isotope	Exposure MWD/MTM			
	8077	24,266	31,800	38,961
²³⁴ U	0.0384	0.0337	0.0321	0.0249
²³⁵ U	5.86	3.22	2.625	1.87
²³⁶ U	0.381	0.947	0.908	1.177
²³⁶ Pu	0.013×10^{-6}	0.160×10^{-6}	0.148×10^{-6}	0.274×10^{-6}
²³⁸ Pu	0.0014	0.0225	0.0274	0.0522
²³⁹ Pu	0.8258	1.4045	1.377	1.3685
²⁴⁰ Pu	0.0907	0.4037	0.466	0.5314
²⁴¹ Pu	0.0373	0.2599	0.299	0.3628
²⁴² Pu	0.0020	0.0568	0.084	0.1400
²³⁷ Np	0.0187	0.0910	0.105	--
²⁴¹ Am	0.0003	0.0111	0.010	0.0308
²⁴³ Am	0.096×10^{-2}	0.975×10^{-2}	--	3.901×10^{-2}
²⁴² Cm	0.031×10^{-3}	1.874×10^{-3}	2.7×10^{-3}	5.228×10^{-3}
²⁴⁴ Cm	0.0076×10^{-3}	2.331×10^{-3}	3.65×10^{-3}	11.789×10^{-3}

FIGURE 2. Yankee-Rowe Measured Actinides vs. Exposure

loaded with recycled plutonium. The changes in plutonium, americium, and curium concentrations are significant.

Utilization of plutonium is currently being resisted by certain publics, as indicated by resistance⁽⁵⁾ to plutonium recycle in LWRs (i.e., GESMO)⁽⁶⁾ and statements of the adequacy of the LMFBR impact statement. To our way of thinking, the crux of these issues centers on whether plutonium is a national resource or a waste. As indicated above, the timing of plutonium recycle has a substantial impact on high-level waste generation. Thus the case of having to dispose of all the plutonium generated should be studied to determine what alternatives remain viable should this case ever come to pass.

TABLE 4. (a) Spent Fuel Actinide Concentrations, PWR vs. BWR (g/MTM)

Isotope	Equilibrium UO ₂ Cycle			First Pu Recycle		
	PWR ^(b)	BWR ^(b)	% Variation ^(c)	PWR ^(b)	BWR ^(b)	% Variation ^(c)
²³⁵ U	8738.5	6643.9	27.2	7485.4	7706.7	02.9
²³⁶ Pu	8.77-4	8.91-4	1.6	8.19-4	5.63-4	37.0
²³⁸ Pu	154.7	120.9	24.5	300.8	322.9	7.1
²³⁹ Pu	5023.7	4229.0	17.2	6685.7	5099.2	26.9
²⁴⁰ Pu	2150.1	1991.3	7.7	3818.0	3256.8	15.9
²⁴¹ Pu	1256.0	973.3	25.4	2207.7	1730.1	24.3
²⁴² Pu	444.0	366.1	19.2	1019.6	1586.6	43.5
²⁴¹ Am	40.5	42.0	3.6	119.0	128.2	7.4
²⁴² Am	0.61	0.58	5.0	2.31	2.06	11.4
²⁴³ Am	79.1	58.6	29.8	261.0	430.0	48.9
²⁴² Cm	15.4	13.0	5.6	42.8	42.6	0.5
²⁴³ Cm	0.13	0.11	16.7	0.41	0.54	27.3
²⁴⁴ Cm	18.0	12.0	40.0	105.0	165.3	44.6
Average 17.2			Average 22.9			

- a. In Tables 4 through 7 the values are the results of computer calculations. The number of significant figures does not imply the accuracy of the calculation but are given for the purpose of comparing calculated results.
- b. BWR discharge exposure 27,000 MWd/MTM; PWR discharge exposure 32,000 MWd/MTM.
- c. "% Variation" is the percentage of the absolute difference between the BWR and PWR weights and the average value, i.e., $|200 (PWR-BWR)/(BWR+PWR)|$.

Accuracy of ORIGEN for LWRs

The ORIGEN code⁽⁷⁾ has known theoretical oversimplifications in its treatment of in-reactor fuel management. Sufficient LWR spent fuel assay data exists to form a basis for evaluating the accuracy of the ORIGEN method for calculating spent fuel concentrations.

With the intent of developing an improved ORIGEN cross section library for on-going applications, cross sections in the ORIGEN library were adjusted systematically to give good agreement with observed data. With

two exceptions, ²⁴¹Am and ²⁴²Cm, the adjusted results were normalized successfully to an average of observed data. Cross sections for isotopes not available in the assay data were not adjusted.

The adjusted library was used to regenerate the BNWL-1900 reference PWR, and a comparison is given in Table 5. Errors ranging from approximately -20% to +80% are indicated. It is reasonable to assume that errors of this magnitude are inherent in current high-level waste projections.

TABLE 5. Accuracy of ORIGEN for LWR Discharge Isotopes

Isotope	ORIGEN Reference PWR BNWL-1900	(g/MT)	ORIGEN PWR Normalized to Observed Assay Data	% Error
²³⁴ U	0.603		0.750 ^(a)	-19.6
²³⁵ U	40.2		43.6 ^(a)	-07.8
²³⁶ U	22.8		20.5 ^(a)	11.2
²³⁸ U	4710.		4710 ^(a)	0.0
²³⁷ Np	490		528 ^(a)	-07.2
²³⁶ Pu	2.99×10^{-6}		3.55×10^{-6} ^(a)	-15.8
²³⁸ Pu	0.859		0.728 ^(a)	18.0
²³⁹ Pu	27.0		25.4 ^(a)	6.3
²⁴⁰ Pu	10.9		10.8 ^(a)	0.9
²⁴¹ Pu	5.19		6.28 ^(a)	-17.4
²⁴² Pu	1.78		2.33 ^(a)	-23.6
²⁴⁴ Pu	1.30×10^{-12}		7.14×10^{-13}	82.1
²⁴¹ Am	45.5		56.4 ^(a)	-19.3 ^(b)
²⁴² Am	0.971		1.24	-21.7
²⁴³ Am	96.7		84.6 ^(a)	14.3
²⁴² Cm	5.44		6.64 ^(a)	-18.1 ^(b)
²⁴³ Cm	7.95×10^{-2}		9.51×10^{-2}	-16.4
²⁴⁴ Cm	31.0		19.4 ^(a)	59.8
²⁴⁵ Cm	2.07		1.27	63.0
²⁴⁶ Cm	0.235		0.137	71.5
²⁴⁷ Cm	3.09×10^{-3}		1.76×10^{-3}	75.6
²⁴⁸ Cm	2.12×10^{-4}		1.16×10^{-4}	82.8

a. Results normalized to observed assay data.

b. ²⁴¹Am is 16% low and ²⁴²Cm is 23% low relative to observed data.

LMFBR High Level Wastes

High-level waste projections for LMFBRs have used ORIGEN results based on AI⁽⁸⁾ and GE⁽⁹⁾ follow-on LMFBR designs. During this report quarter, ORIGEN calculations were performed to compare isotopic concentrations with GE follow-on fuel management results. As with the LWRs, discrepancies were found and adjustments were made in the ORIGEN library. In this case, only six of the actinides were available in the GE data. The adjusted library was used to recalculate the BNWL-1900 LMFBR high-level waste concentrations. Table 6 gives a comparison of the BNWL-1900 GE LMFBR with the results from the normalized ORIGEN calculations.

The adjusted results vary significantly from the previous calculations and indicate that adjustments to LMFBR waste concentrations are needed. However, improvements in LMFBR technology since the late 1960s have reflected different LMFBR designs which render the GE results obsolete and reduce the significance of these results. Moreover, recent calculations of the LMFBR breeding ratio with mixed oxide fuel have increased the probability of an early shift to nitride or carbide fuels in the core and/or radial blankets to improve the breeding gain and therefore enhance the economics of the LMFBR fuel cycle.

The high-level waste projections for LMFBRs should be verified by comparison with more recent LMFBR fuel management calculations. These comparisons should address the effects of advanced fuel concepts from the

standpoint of their impact on actinide concentrations in high-level waste.

Summary

Table 7 compares the reference PWR high-level waste from BNWL-1900 to a high-level waste generated by 40% BWR/60% PWR discharge fuel. The observed discrepancies reflect the net effect of including BWRs in the calculation, reducing the PWR goal exposure to 32,000 MWd/MTM, and normalizing the ORIGEN results to agree more closely with observed data. The percent variation ranges from +97% to -20% and demonstrates where the original calculations could be considered conservative. However, judgments on conservatism are dependent upon the point of view of the observer. It is conceivable that grossly underpredicting some of the higher actinide concentrations could have significant impact on one or more of the waste management schemes (i.e., which radionuclides are controlling in which disposal concept). Certainly transmutation concept development is sensitive to errors of this nature.

The results of this quarter's work generally indicate that further refinements in fuel management models for LWRs and LMFBRs would result in significant adjustments in high-level waste concentrations. Moreover, preliminary estimates of the magnitude of suggested refinements have been calculated. These results and other considerations reported above demonstrate the dynamic nature of the source terms that feed into the head

Table 6. LMFBR HLW Concentrations, g/MT

Isotope	ORIGEN BNWL-1900	ORIGEN Normalized to GE Follow-on	GE Follow-on ^(a)
²³⁴ U	1.28	0.111	--
²³⁵ U	6.05	8.50 ^(a)	6.53
²³⁶ U	7.04	1.09	--
²³⁸ U	4380	4360 ^(a)	4369.2
²³⁷ Np	391	244	--
²³⁶ Pu	2.14×10^{-5}	1.15×10^{-5}	--
²³⁸ Pu	3.47	2.68	--
²³⁹ Pu	263	291	290.0
²⁴⁰ Pu	87.7	93.0	93.1
²⁴¹ Pu	16.5	19.1	19.4
²⁴² Pu	12.8	10.7	10.7
²⁴⁴ Pu	1.25×10^{-4}	9.15×10^{-5}	--
²⁴¹ Am	582	685	--
²⁴² Am	21.0	22.1	--
²⁴³ Am	298	225	--
²⁴² Cm	25.5	25.9	--
²⁴³ Cm	3.30	3.13	--
²⁴⁴ Cm	30.2	20.6	--
²⁴⁵ Cm	1.22	0.771	--
²⁴⁶ Cm	5.09×10^{-2}	2.93×10^{-2}	--
²⁴⁷ Cm	1.12×10^{-3}	6.00×10^{-4}	--
²⁴⁸ Cm	2.81×10^{-5}	1.41×10^{-5}	--

a. 0.3% of ²³⁸U charged to the core assumed to be ²³⁵U

TABLE 7. Equilibrium Cycle Discharge HLW Concentrations, g/MT

Isotope	Reference PWR ^(c)	PWR/BWR Mix ^(a)	% Variation
²³⁴ U	0.603	1.05	-54.1
²³⁵ U	40.2	39.5	+ 1.8
²³⁶ U	22.8	20.8	+ 9.2
²³⁸ U	4710	4730	- 0.4
²³⁷ Np	490	528	- 7.5
²³⁶ Pu	2.99×10^{-6}	3.41×10^{-6}	-13.1
²³⁸ Pu	0.859	0.735	+15.6
²³⁹ Pu	27.0	23.8	+12.6
²⁴⁰ Pu	10.9	10.5	+ 3.7
²⁴¹ Pu	5.19	5.6	- 7.6
²⁴² Pu	1.78	2.07	-15.1
²⁴⁴ Pu	1.30×10^{-12}	4.63×10^{-13}	+95.0
²⁴¹ Am	45.5	54.5 ^(b)	-18.0
²⁴² Am _m	0.971	1.17	- 9.3
²⁴³ Am	96.7	71.0	+30.6
²⁴² Cm	5.44	5.95 ^(b)	- 9.0
²⁴³ Cm	7.95×10^{-2}	8.86×10^{-2}	-10.8
²⁴⁴ Cm	31.0	15.4	+67.2
²⁴⁵ Cm	2.07	0.929	+76.1
²⁴⁶ Cm	0.235	9.91×10^{-2}	+81.4
²⁴⁷ Cm	3.09×10^{-3}	1.18×10^{-3}	+89.5
²⁴⁸ Cm	2.12×10^{-4}	7.36×10^{-5}	+96.9

a. 40% BWR to goal of 27,000 MWd/T and 60% PWR to goal of 32,000 MWd/T, assuming a waste loss of 0.5%. Normalized to observed data.

b. ²⁴¹Am is 16% low and ²⁴²Cm is 23% low relative to observed data.

c. Goal exposure of 33,000 MWd/T as reported in BNWL-1900.

end of developing waste management strategies. They are reported here to document the rationale for an ongoing program to assess the validity of waste projection methodology and determine the effects of uncertainties on the various programs to develop storage, conversion and disposal concepts.

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TASK 3: WASTE MANAGEMENT OPTIONS

M. R. Kreiter

WASTE DISPOSAL RESEARCH AND DEVELOPMENT SCENARIOS -

M. R. Kreiter

Preliminary research and development requirements were expanded from those in BNWL-1900 for radioactive waste management schemes incorporating terrestrial and extraterrestrial disposal, and transmutation. Included in the terrestrial schemes were geologic, seabed, and ice sheet locations. For each general category, a single disposal concept was chosen around which a preliminary hypothetical scenario identifying research and development needs was prepared. The

scenarios are directed toward the first five years of a program to develop a viable disposal concept with the ultimate end point being commercial facilities some time later. Costs have not yet been estimated.

GEOLOGIC DISPOSAL - M. R. Kreiter

The concept chosen for R&D investigations of geologic disposal of solid radioactive waste was a mined cavity with no waste melting. The scenario includes tasks covering chemical-physical evaluations, geological and biological considerations, site selection, borehole sealing, and emplacement engineering, as discussed below.

Chemical-Physical Evaluations

This task would be directed toward obtaining quantitative values for certain mechanical, chemical, and thermal properties of candidate geologic media basic to waste disposal site design.

Key physical and chemical information on candidate geologic environments would be developed. Available information would be assembled and missing data would be obtained from laboratory experiments related to the transport of critical radionuclides through selected candidate disposal media (e.g., salt, quartzite, granite, shale, soils, etc.). Physical property information would be developed for utilization in risk analysis studies and for predicting transport times of those radionuclides which are basic to long-term waste disposal considerations. The properties would be determined for candidate disposal media and their surroundings, and

would include but not be limited to porosity, permeability, ion exchange capacity, and chemical interaction with waste forms.

A study would be instituted to obtain additional information pertinent to mechanical and thermal properties of potential geological host rock materials. Properties such as creep, tensile strength, and thermal expansion would be regarded from rock mechanics and containment considerations while the thermal properties such as thermal conductivity and specific heat would be obtained to predict the effects of an imposed heat source, represented by the emplaced radioactive material, upon the geologic formation.

Certain knowledge basic to calculating transport of critical radionuclides through the soil would be used in other programs to estimate the health hazards associated with disposing of radioactive waste in geologic formations.

Geologic Analysis

This task is primarily associated with the analysis of historic geologic events relevant to radioactive waste disposal in geologic formations as a basis for predicting future events.

A variety of geologic events could conceivably impair containment integrity in abnormal, catastrophic occurrences or perhaps under normal long-term situations. These events would be the subject of R&D studies performed in this task. An obvious event to be considered is seismic activity. Movement, folding, or faulting of the geologic environment would

be considered. The possibility of erosion and denudation and the possibility of leaching by groundwater would also be considered for geologic formations of interest. Various geologies would be evaluated and ordered as to preference for application.

Biological Effects

This task objective is to ascertain the biological effects of disposing of radioactive waste in geologic formations.

Studies would be instituted to determine the extent to which heat and radiation affect biological systems. Investigations to better define the existing knowledge of transport of radionuclides in the food chain will be undertaken. Many of the contemplated studies could be covered under programs funded by other sources.

Site Selection

The purpose of this task is to identify specific geologic environments and sites with the potential to meet criteria for long-lived radioactive waste disposal.

The study would initially define broad criteria for geologic disposal siting based on input from the other tasks. It would then identify broad regional areas of conterminous United States which have potential for meeting the criteria as waste disposal sites. The general areas defined would then be investigated in more detail and compared with geologic siting criteria developed to permit selecting disposal sites on the basis of optimum geologic settings. The siting criteria developed would include general requirements on rock

types and properties, relationship to groundwater, seismicity, faulting, flood potential, population distribution, land use, resource potential, and other considerations. Criteria would be examined on the basis of safety/risk analyses.

New survey techniques would be investigated for their application to defining surface and subsurface characteristics of importance to the siting of a waste disposal area. These techniques should include infrared imagery, satellite photography and side-looking radar.

Borehole Sealing

The objective of this task is to establish acceptable long-term sealing techniques for use in host rocks. A study to develop sealing techniques which will maintain their integrity for the time periods being considered for radioactive waste disposal would be implemented. The adequacy of present and potential grouts, cements and advanced techniques for sealing geologic media which hold potential for disposal of radioactive waste would be determined. If necessary, new, acceptable techniques would be developed (e.g., sealing with material identical to that removed).

Emplacement Engineering

This task would be directed toward investigation and assessment of techniques for emplacing radioactive waste in a geologic formation. Various means of emplacement would be developed to the point of commercial application. These means would include shaft and hole forming, retrievability, emplacement equipment

development, material compatibility, interim monitoring, etc. Emplacement techniques and concept acceptability would be confirmed in a pilot plant evaluation study prior to commercial application.

SEABED DISPOSAL - R. W. Wallace

Emplacement of solidified radioactive waste in deep ocean basement rock was chosen as the seabed disposal concept for R&D examination. Included in this scenario are tasks to obtain geologic, geophysical, oceanographic, meteorological, and biological data. Tasks to investigate engineering emplacement techniques and to consider international policies are also included.

Chemical-Physical Evaluations

This task would provide information on the basic properties of material of the seabed. A program would be undertaken to sample both the relatively unconsolidated sediments and the more dense bedrock presently considered to be the disposal medium. Mineral and chemical composition, particle size, porosity, permeability, water content, thermal conductivity, shear strength, compressibility and other physical properties would be investigated. The data are needed to develop emplacement methods, to predict or model the behavior of the material during and after emplacement of the waste, and to determine the effects of heat and radiation on the seabed material.

Geophysical Analysis

This task would consist of obtaining detailed maps of the site areas

showing depth of water, sea floor topography, thickness of sedimentary material and configuration of the bedrock underlying the sediments. The data would be obtained by a combination of precision depth recording and seismic methods of profiling and sub-bottom profiling. Geophysical investigation of site areas would measure heat flow at the sea floor and would monitor for seismic/microseismic activity.

Oceanographic Analysis

This task would provide data on ocean currents, particularly deep currents and deep tides in the disposal site areas. These data would be collected over a period of at least several years to assure that the results would distinguish between short-term and seasonal variations and the main current/tidal effects. This task would also include studies of temperature, density and salinity stratification, and of the general stability of the water mass at the site. These studies would yield information necessary to predict the transport and dispersion of any material accidentally released into the water mass at the site and prediction of possible future activities.

Meteorological Analysis

The purpose of this task would be to aid in determining the type and design of surface equipment needed for disposal operations, the number of days in a given time period when this equipment could be expected operable, and prediction of potential future climate on seabed disposal. This

task would investigate the effects of climatic conditions in the site areas. Data would be collected on winds, sea conditions, storm frequency and severity, storm paths, visibility and surface currents.

Biological Analysis

This task would investigate the plant and animal populations of the site areas to determine the types of individuals in the various communities, the density and distribution of the types and their interrelationships, their effects in ecologic chains and in the transport of radionuclides. This information is necessary to evaluate the effects of the waste disposal operations on the biota and to assess their role in concentrating and/or transporting any radioactive material that might accidentally get into the water mass.

Emplacement Engineering

This task would investigate and assess the various engineering techniques for waste emplacement in the seabed and transport to the disposal site. A technique would be selected as the most suitable, preliminary estimates of the specifications and needed equipment made, and the emplacement system described. This task would also include investigation of different materials to find which would give the best performance for waste canisters emplaced in the seabed. Radiation and temperature effects would be considered as well as the effects of seawater, pressure, and the physical properties of the seabed material.

International Policy

The purpose of this task would be to consider existing treaties, policies and agreements regulating use of the oceans with the objective of defining technical bases for attempting to reach an international agreement for use of the seabed for waste disposal. A number of policies at present affect use of the seas for transport and disposal of radioactive waste. To modify these policies would require international agreement but could conceivably lead to an international waste repository. This task would provide the technical bases for potential modification of international policies.

ICE SHEET DISPOSAL - R. W. Wallace

Anchored emplacement of solidified radioactive waste serves as the reference concept for the ice sheet disposal scenario. Proposed R&D tasks are directed toward obtaining geological and geophysical data, performing melt and climatological studies, investigating engineering emplacement techniques, and considering international policies.

Geological/Geophysical

This task would provide basic information on the ice and ice sheets necessary to evaluate a scheme of waste disposal on the ice sheet. It would include measurement and evaluation of such data as flow rates, particle flow paths, particle residence times, direction of flow lines, discontinuities within the ice such as density and temperature zones, and physical properties.

Melt Studies

This task would investigate the melting effects of the heat in the waste on the ice at the disposal site. Included in this task would be studies of the melting rate; the shape, size and propagation of the melt cavity; and the design and testing of ice anchors used on the surface of the ice for the anchored emplacement concept.

Climatological Studies

This task would consist of a thorough study of the evidence of ice sheet changes in the past as a result of climatic variations. The purpose would be to describe the response of the ice sheet to climatic changes and to predict as well as possible the future of the ice sheet based on extrapolated world climatic trends. This task would require the collection, assimilation and correlation of a large amount of data not only from the polar areas but also involving global patterns and trends.

Emplacement Engineering

This task would investigate the various techniques for emplacing waste canisters in the ice sheet. Because the waste would be essentially self-emplacing, most of the effort in this task would be directed toward evaluation of the various over-sea and over-ice transport and handling methods, and design and specifications for the ice anchors and supporting cable system. Canister material will be specified that will give the best performance for relatively shallow emplacement within the ice sheet, and the total emplacement system will be described.

International Policy

This task would investigate the possibility of establishing by treaty or agreement an international waste repository in Antarctica to provide the technical bases for pertinent policy changes. The purpose of this task is to review the existing policies, treaties, and agreements that would affect the use of the ice sheets for waste disposal. Greenland is not currently considered because it is Danish territory, and waste disposal is prohibited in Antarctica by international treaty.

TRANSMUTATION - R. C. Liikala

The principal task for transmuting long-lived constituents in radioactive waste to shorter-lived or nonradioactive constituents is to evaluate the efficiency of actinide transmutation in fission reactors. If the results of the efficiency evaluation is positive and it is decided there is need for transmutation, a task to develop and demonstrate actinide recycle in fission reactors will be undertaken.

Evaluate Efficiency of Actinide Transmutation in Fission Reactors

The objective of this task is to confirm the success of transmutation of actinides in fission reactors and provide quantitative data for a basis to evaluate whether the need is compelling relative to other waste management alternatives. The optimum nuclear fuel cycle for transmutation will be identified. The efforts involve three major activities. The

first activity is to evaluate actinide transmutation, considering:

- The use of AEC production reactors
- Each of the various reactor types currently operational or expected to be commercially viable as central power stations between now and the early 1990s
- A special-purpose actinide burner to determine the efficiency of each in reducing actinide inventory of high-level waste.

The reduction of toxicity in actinide recycle is directly related to the ability to separate actinides from the high-level waste stream. Therefore, R&D efforts are needed to determine the level of partitioning technically achievable and the feasibility of other means of separation (e.g., laser isotope separation). These efforts comprise the second major activity in this phase of the program.

The third activity is to evaluate the impact of recycling actinides in these reactors by assessing the incremental risks at the affected process stages in these nuclear fuel cycles. This information is needed to assure that advantages accruing to waste management objectives via destruction of actinides in the reactor are not negated by increases of actinides in processing waste streams such as in fabrication and partitioning.

The data developed in the activities will be used to determine the viability of transmutation for each of these nuclear fuel cycles in order to identify the most likely fission transmutation cycle.

Development and Demonstration of Actinide Recycle in Fission Reactors

The objective of this phase of the program is to demonstrate the viability of the concept of recycling actinides in the nuclear fuel cycle(s) identified as most likely in Task 1. The time required for accomplishing this objective is estimated at somewhere between 10 and 15 years and the cost about in the range of 75 to 100 million dollars. The efforts of this phase could represent a large-scale cooperative program involving government and industry in the event that actinide recycle in commercial facilities represents the most likely strategy to develop.

EXTRATERRESTRIAL DISPOSAL -

K. Drumheller

Of several possible destinations, this scenario considered only solar impact for the disposal of radioactive waste. The R&D scenario comprises the following seven tasks.

Waste Content

At present the principal code used to establish isotopic content of different wastes is ORIGEN. The accuracy of this code is suitable for the type of investigative work done to date. However, if extraterrestrial disposal is to be used for the actinide or transuranic wastes, a more accurate picture of true isotopic content is desirable than that obtainable in present codes.

Because of uncertainty and physics effects for extended exposure in different types of reactors, existing data on the isotopic content of spent

fuel is limited. A program to analyze spent fuel from several different fuel cycles is required.

Disposal Priority

The purpose of this task is to put into more clear perspective the safety incentives and problems for space disposal of certain radioisotopes. With such information available, choices can be made regarding their disposal needs. Though a number of analyses of the relative potential hazards of different isotopes exist, additional information would be advantageous for the analysis of space disposal. Past comparisons of the potential hazards in space of one isotope, such as ^{238}Pu with another such as ^{147}Pm , have not covered the total range of variables for full evaluation of the incentives for space disposal.

Partitioning

Partitioning is a fundamental prerequisite for space disposal. Therefore the timing and successful completion of partitioning process development is critical to space disposal.

It appears that separating the transuranic and fission product stream could provide a waste fraction in reasonable form for disposal in space. Nonetheless, it may prove desirable to remove the curium from this product to minimize heat problems for space disposal. It might also be advantageous to extract the neptunium for conversion to ^{238}Pu for other purposes. If long-lived fission products such as ^{129}I are proved

to have long-term toxicity, it might be beneficial to remove the iodine from the fission product stream and ship it to space with the transuranics.

Capsule Design

It appears possible today to design a capsule which would satisfy stringent technical and safety criteria. However, in a number of areas additional data and refinement of data and methods could improve the safety and reduce the overall cost of disposal. Typical of these are:

- Criticality and Radiation Neutronics Calculations

Additional code and experimental studies are required to refine criticality data and the radiation dose rates.

- Heat Transfer Analysis

Heat transfer under a wide variety of circumstances is one of the more critical aspects of design for space disposal. The capsule must be able to withstand such diverse circumstances as deep burial in sand, re-entry, impact in space vacuum, and explosion of the rocket on the launch pad. Optimization of materials, configuration, and environment will require much refinement.

- Refined Structural Analysis

Some data exist on impact resistance of large capsules, but significantly more needs to be done to make this a practical system.

- Impact Testing

Testing for structural considerations, shielding, burial corrosion resistance, and any other possible circumstances will be required.

Encapsulation Process Development

Encapsulation processes are expected to be similar to those existing for space applications of radionuclide heat sources. Some of the key steps requiring development are:

- Particle Preparation

Processes exist for preparation and micro-encapsulation of particles. In general, however, these processes are applicable to small scale production. Developmental work is required to provide production-type processes.

- Compacting of Matrix

The development of processes is required for compacting the matrix surrounding the fuel established in the capsule design.

- Containment

The containment shell surrounding the capsule will also require some process development studies. Extension of existing fabrication techniques could likely be used, particularly if the shell is stainless steel.

Because of the increased size, quantity, and complexity of transuranic waste capsules and the need for greater integrity, more extensive process development by factors of 5 to 10 than that required for smaller capsules is expected to be required for nuclear waste capsules.

Space Flight

Developmental work in this area is likely to be all under NASA direction. Some of the primary studies expected are:

- Disposal Trajectory

The determination of a disposal trajectory with no significant unknowns is one of the major technical

needs of this total program. Development in this area might involve propelling several waste containers into the sun and monitoring for detectable transuranics which might return during the next 5 to 30 years.

- Vehicle Development

NASA analysis to date has been based on existing concepts. Some development would undoubtedly be required to adapt and optimize present concept vehicles to waste disposal.

- Advanced Systems

Advanced systems such as particle acceleration and solar sails have been examined only briefly.

If space disposal is considered practical with current systems, it is probable that significant economic gains could be achieved through development of advanced systems. While potential safety improvements are not apparent, there is some probability that additional safety could also be achieved through utilization of advanced systems.

It is therefore expected that a much more detailed analysis of advanced systems would be pursued before space disposal can be implemented. If advanced systems show significant promise, they could warrant a complete and extensive developmental program comparable to that for conventional vehicles described above.

Safety Analysis

The principal constituent of safety analysis is the execution of the space flight including potential accident considerations. Some other studies are required for the additional processing which must be done to put material in the form for flight.

The safety aspects of flight will include the effect of earthly accidents such as explosion on the launch pad, failure to achieve orbit, or re-entry from a partial orbit. Space flight effects will include establishing realistic probabilities and consequences of malfunctions or accidents.

Safety studies will also include estimation of effects of space disposal on our environment.

POTENTIAL GEOLOGIC DISPOSAL AREAS - R. W. Wallace

The computerized program for identifying potential disposal sites that meet specific geologic criteria is now functional. It includes for the continental United States the following geologic information:

- 1) Seismic risk zones 0, 1, 2, 3.
- 2) Areas inundated by a 60-meter and a 150-meter sea level rise.
- 3) Southern extent of the last stage of continental glaciation.

The program is designed to select areas within the United States possessing the specified combinations of the geologic parameters stored in the data file. For example, if one requested a map of the United States showing the area of zero to minor historical earthquake damage (Seismic Risk Zone 0 and 1) and above sea level should a 60-meter rise occur, the output would be as shown in Figure 3.

If the output requested were for the same Seismic Risk Zones but to include a 150-meter sea level rise, the map for these conditions would be as shown in Figure 4.

FIGURE 3. Map of Conterminous U.S. upon Which Areas Located Above a Sea Level Rise of 60 Meters and in Seismic Risk Zones 0 and 1 Are Identified (Stippled Portions)

FIGURE 5. Map of Conterminous U.S. upon Which Areas Located Above a Sea Level Rise of 150 Meters and in Seismic Risk Zones 0, 1, and 2 Are Identified (Stippled Portions)

FIGURE 4. Map of Conterminous U.S. upon Which Areas Located Above a Sea Level Rise of 150 Meters and in Seismic Risk Zones 0 and 1 Are Identified (Stippled Portions)

Using a similar procedure, a map showing the area of moderate historical earthquake damage (including the areas of zero and minor damage) or Seismic Risk Zones 0, 1 and 2, combined with the area above sea level if a 150-meter sea level rise should occur, is shown in Figure 5.

The data file is being expanded to include more geologic information and

some meteorological data that bear onsite selection. Examples of the information to be added is depth to bedrock, possibly type of bedrock, location of the larger sedimentary basins, and perhaps mean annual precipitation, run-off and depth to groundwater.

TASK 4: SAFETY EVALUATIONS

G. Jansen

SORPTION OF RADIONUCLIDES ON SOIL -

R. C. Routson

Previous measurements of sorption of the radionuclides technetium and neptunium on western desert soil and southeastern U.S. sub-soil as a function of salt concentration are now being incorporated into pathways analyses of geologic repositories. The rapid movement rate of neptunium in both soils prompted additional investigation of americium. During this quarter, measurements were made on americium adsorption from sodium-containing solutions ranging from 3M

to 0.015M, calcium-containing solutions ranging from 0.2M to 0.002M on the western soil, and with calcium-containing solutions on the south-eastern soil. No results are available pending completion of counting of the samples. With completion of the americium measurements, attempts will be made to coordinate future needed measurements, such as for Cm, U, and Th through other programs.

PATHWAYS ANALYSES - G. Jansen and H. C. Burkholder

Partial results were obtained in the analysis of the effect of salt on migration of radionuclides from a repository in a salt formation after a hypothetical contact by water. The applicability of these results to such a repository is still unknown, because the effect of the difference between the line source of salt and the actual widespread geometry of a repository is unknown. Furthermore, the frequency of saltwater injection rates into the aquifer must be described before application. In risk analysis the saltwater injection rate represents the severity of consequences of the system failure. The probability of occurrence of each saltwater injection rate must eventually be estimated and multiplied by the consequences to obtain the risk associated with the water leach pathway.

One factor determining the isolation provided by underground waste disposal is the presence of salt at the disposal site. If waste containment is breached, water flowing past

the salt formation may transport radionuclides to man's environment faster and at higher concentrations than if salt were not present. The objective of this work is to make a quantitative estimate of the salt effect using a simple mathematical model.

Model Development

The mathematical model assumes transient flow through a one-dimensional soil bed one meter thick with decay on the soil and in solution. An impulse release of nuclides at the repository is assumed. The differential equation and boundary conditions for this model (in nondimensional form) are presented in Equations 1, 2, and 3.

$$K(\eta) \frac{\partial N}{\partial \theta} + \frac{\partial N}{\partial \eta} + RK(\eta)N = 0 \quad (1)$$

$$N = 0 \quad @ \quad \theta = 0; \quad \eta > 0 \quad (2)$$

$$N = N_0 \frac{V}{L} \delta(\theta) \quad @ \quad \eta = 0; \quad \theta > 0 \quad (3)$$

where

N = radionuclide discharge rate (Ci/yr)

N_0 = radionuclide inventory at the time of release (Ci)

$\theta = Vt/L$ = dimensionless time

$\eta = Z/L$ = dimensionless distance

$R = \lambda L/V$ = decay number

$K(\eta) = 1 + \frac{\rho}{\epsilon} K_D(\eta)$ = water velocity/nuclide velocity

$\delta(\theta)$ = unit impulse function

V = water velocity (km/yr)

t = time (yr)

L = path length (km)

K_D = distribution coefficient (ml/g)

ρ = soil density (g/ml)

ϵ = soil porosity
 $\lambda = 0.693/t_{1/2}$ = decay constant
 $t_{1/2}$ = nuclide half life
 Z = distance from repository, km

The distribution coefficient has been determined experimentally for a number of nuclides and has the form shown in Equation (4).⁽¹⁾

$$K_D = \exp \left[C_1 - C_2 \ln (C_{Na}) \right] \quad (4)$$

where

C_1 and C_2 = empirical constants
 $C_{Na}(\eta)$ = salt concentration
 (g-mole/liter)

The salt concentration in a planar aquifer flowing from a point source in a bedded salt formation is described by Equation (5).⁽²⁾

$$C_{Na}(\eta) = \frac{C_{Na,o} Q}{2 \sqrt{\pi D_t} LV/\epsilon} (\eta)^{-1/2} \quad (5)$$

where

$C_{Na,o}$ = salt concentration at saturation, g-mole/liter
 Q = salt injection rate liter/meter of aquifer thickness/day
 D_t = transverse dispersion coefficient, cm^2/min .

After combining Equations 1, 4, and 5, noting that $K(\eta) \approx \frac{\rho}{\epsilon} K_D(\eta)$ for strongly adsorbed nuclides, solving by using a Laplace transformation and integration, and applying the boundary conditions (Equations 2 and 3), Equation 6 is obtained.

$$N = N_0 \frac{V}{L} \delta \left(\theta - \frac{J}{H+1} \eta^H + 1 \right) \exp(-R\theta) \quad (6)$$

where

$$J = \frac{\rho}{\epsilon} \exp \left[C_1 - C_2 \ln \left(\frac{C_{Na,o} Q}{2 \sqrt{\pi D_t} LV/\epsilon} \right) \right]$$

$$H = C_2/2.$$

The $J \eta^H + 1/H + 1$ term physically represents the delay of the released radionuclides relative to water at any given position along the migration path. At the migration path outlet ($\eta = 1$), $J/H + 1$ represents the "effective" water velocity/nuclide velocity ratio over the migration path or in other words an "effective" nuclide distribution coefficient between the liquid and the soil for the migration path. Thus, the nuclide discharge time after year 2000, t_D , is calculated by Equation 7 and the nuclide rate at the time of discharge, N_D , is calculated by Equation 8.

$$t_D = \frac{L}{V} \frac{J}{(H+1)} + t_R \quad (7)$$

$$N_D = N_i \exp(-\lambda t_D) \quad (8)$$

where

t_R = number of years after year 2000 that release occurs.
 N_i = radionuclide concentration at year 2000.

Results and Discussion

Calculations were made for the radionuclides of 11 elements (the

only ones for which K_D vs. C_{Na} data exists)^(3,4) released from an underground high-level waste repository containing all waste from the nuclear power economy through the year 2000. The release (leach) incident was assumed to occur in the year 2100. The leach rate was 0.3%/yr; the path length was 10 miles; and the water velocity was 1 ft/day. The soil density was 1.5 g/ml; the soil porosity was 0.33; and the transverse salt dispersion coefficient was $0.001/\text{cm}^2/\text{min}$. Saltwater injection rates ranged from 0 to 10^4 liters/meter/day. Radionuclide concentrations at the year 2000 were obtained from ORIGEN runs.

The results of these calculations are shown in Figures 6 and 7. Figure 6 shows the effect of saltwater injection rate on nuclide discharge time. Hydrogen, technetium, and iodine do not adsorb on the soil and migrate at the same velocity as the water. Neptunium weakly adsorbs on the soil, and the adsorption is virtually independent of salt concentration. The adsorption of strontium and cesium is very strongly dependent on salt concentration, and both have the same migration time as water at a saltwater injection rate of 10^4 liters/meter/day. This represents a reduction of almost three orders of magnitude in the discharge time relative to the no-salt case. The nuclide discharge times for europium, promethium, samarium, and holmium are reduced by two orders of magnitude, and the discharge time for plutonium is reduced by one order of magnitude at a saltwater injection rate of 10^4 liters/meter/day.

FIGURE 6. Saltwater Injection Rate (Liters/Meter/Day)

Figure 7 shows the effect of the saltwater injection rate on the nuclide concentration at the time of discharge. Strontium-90, ^{137}Cs , ^{239}Pu , and ^{240}Pu discharge before decaying off at saltwater injection rates of 10^4 liters/meter/day. The ^{242}Pu concentration increases slightly as saltwater injection rate increases. The ^3H , ^{99}Tc , ^{129}I , ^{135}Cs , and ^{237}Np discharge concentrations are independent of salt concentration. Promethium-147, ^{151}Sm , ^{152}Eu , ^{154}Eu , ^{155}Eu , $^{166\text{m}}\text{Ho}$, ^{238}Pu , and ^{241}Pu decay off before coming out in the range of the saltwater injection rates shown on Figure 7.

FIGURE 7. Saltwater Injection Rate (Liters/Meter/Day)

The very strong effect of salt on the discharge concentration for some of the nuclides shown here points out the need for experimental determination of the effect of salt on the distribution coefficients for the other actinides and fission products present in the waste. Only in this way can the effect of salt on dose be estimated for geologic disposal of high-level waste. Likewise, the magnitude of saltwater injection rates and frequency of occurrence needs to be estimated for bedded or domed salt formations in order to determine the risk associated with waste burial in salt formations.

Conclusions

The presence of salt (sodium chloride) at an underground waste repository could decrease the radionuclide migration time and could increase the radionuclide concentrations at the time of discharge into a surface water body if the salt content is high enough. As the salt injection rate reaches the range 10^3 to 10^4 liters/meter/day, the peak quantities of ^{90}Sr and ^{137}Cs in the aquifer effluent rise rapidly because of ion exchange competition from sodium. These nuclides are adsorbed to the point that they decay off completely before discharging when no salt is present but the presence of a saturated salt solution along the entire path length would result in negligible adsorption. Axial dispersion (not included in the present calculations) will decrease the discharge rates from those presented here but will be less effective in reducing discharge rates as the saltwater injection rate increases. The results point out the need for:

- (1) experimental measurements of the effect of salt concentration on nuclide adsorption for the other actinides and fission products and
- (2) an estimation of the magnitude of salt injection rates and their frequency of occurrence for bedded salt formations.

The present mathematical analysis overestimates the discharge rates because axial dispersion was neglected in the mathematical model. However, axial dispersion is less effective in reducing discharge rates in the salt case than in the non-salt case. This is true because the salt

causes the nuclide distribution coefficients to increase with increasing distance from the repository. Hence, as the nuclide disperses forward from the migrating nuclide peak, it is more strongly adsorbed on the soil which slows its migration; as the nuclide disperses backward, it is less strongly adsorbed, which speeds its migration. Thus the presence of salt causes the dispersion to be self-eliminating, with the result that the migrating nuclide peak is less disperse (sharper) than if no salt were present. Furthermore, since the average gradient of salt concentration with distance increases with increasing salt injection rate, axial dispersion should be less effective in reducing discharge rate as salt injection rate increases. Hence the present analysis underestimates the salt effect. Future work will include axial dispersion in the mathematical model and provide a more realistic estimation of the salt effect.

SAFETY/RISK RESEARCH AND DEVELOPMENT NEEDS - G. Jansen

Two preliminary alternative scenarios were developed for safety/risk evaluation needs of ultimate disposal. In the first case, methodology and its proof development plus preliminary safety evaluations would be performed for each of the five generic disposal concepts (geologic, seabed, ice sheet, transmutation, space disposal) to provide the minimum safety/risk input for the decision on the

concept(s) to be developed for application. This is estimated to require about five years of significant safety studies. Following this, detailed evaluation (in the same range as the Rasmussen study) would be required for the final safety/risk analysis of the disposal concept selected, including input to siting criteria and system optimization.

In the second case, safety methodology plus methodology proof would be performed over a period of about five years at a modest effort. At that point, the preliminary analysis discussed above could be provided over the following few years period to obtain the minimum safety/risk information for decision making; or alternatively, the latter could be deleted if a decision were made without the preliminary safety/risk information. Once the decision is made the detailed analysis requirements would be comparable to that in the first case.

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TASK 5: WASTE MANAGEMENT STRATEGIES

D. E. Deonigi and J. B. Burnham

WASTE MANAGEMENT DECISION ANALYSIS -

J. W. Litchfield, J. B. Burnham

Efforts during the quarter focused on four areas:

- Work has advanced in developing the decision analysis model to be used to predict the preferences of various groups on factors important to waste disposal. This decision modeling concept has been documented, and a preliminary draft of an article for submission to a professional trade journal has been written.
- The needed computer software requirements have been defined. Computer programs have not yet been developed; however, we do expect to begin this effort in the next quarter.
- Initial technical data on each waste management concept have been collected from technical task leaders on waste management studies here at Battelle. Further data collection will be necessary to validate this and additional information. It is planned that this data collection effort will be completed in the next quarter.

The Churchman-Ackoff⁽¹⁾ approximate measure of value method of evaluating multiattributed alternatives has provided the basis for a number of decision analysis applications reported in the literature. Recent accounts have been reported of more complex decision models which attempt to improve upon the

Churchman-Ackoff approach. For research and development the authors have not found these models entirely suitable. An alternative model is described which is directly applicable to research and development decision analysis.

Decision making in the R&D environment transcends a wide range of complexity. Effective decision analysis is especially important because of the large resource commitment typically involved, the uncertainty of the outcomes, and the multiple attributes which are relevant. Moreover, when R&D decisions affect highly sensitive areas--such as development of technology for handling nuclear wastes--where decision makers may be called upon to support their decisions before governmental and public groups, it is critical that logical and consistent methods be applied. These methods should serve to ensure that the larger problem has been separated into simpler problems which facilitate direct analysis and can be logically combined into an overall evaluation model.

Fundamentally the problem of the decision maker is to choose a rational course of action in an environment which is uncertain, complex and constantly undergoing change.

Decision analysis is generally considered to involve two broad dimensions. The first of these may be termed goals, or objectives, which are sought. In any given decision analysis, several competing goals may be relevant. For example, in an R&D context, one decision goal might be to maintain R&D costs at existing

levels while another might be to increase the number of R&D specialty areas. It is likely that these goals may conflict in that one implies limiting resource expenditure while the other implies additional resource expenditure. It has been found useful to express these goals in terms of their attributes X_i , i.e., R&D costs and R&D specialty areas, and to assign weight w_i to them indicating their importance relative to one another.

A second major dimension contains the alternative courses of action A_j which may be taken and the efficiency e_{ij} with which they service each attribute. A sketch of this model is shown below:

		Attributes			
		w_1	w_2		w_n
		X_1	X_2	...	X_n
Alternatives	A_1				
	A_2				
	.				
	.				
	A_n				

Let w_i = estimated value of i^{th} factor

and e_{ij} = the efficiency of the j^{th} alternative in achieving the i^{th} factor.

Then $\sum e_{ij}w_i$ = expected utility, or worth, of the j^{th} alternative.

This model assumes, roughly, that the utility of any alternative is equal to the sum of the utility of

its parts. Put another way, the worth of any attribute w_i should not be affected by the level to which any other attribute is achieved. This model was first delineated by Churchman and Ackoff (C-A)⁽¹⁾ and has formed the basis for several applications reported in the literature.⁽²⁻⁴⁾

The C-A type model does not take into account the uncertainty of an alternative's attribute level. The e_{ij} elements are treated as if they were known with certainty. This can be a severe limitation where uncertainty is a significant factor--a condition characteristic of most decision analysis situations, particularly in R&D. It has been argued that this limitation is offset by the fact that the C-A method is consequently easier to administer.⁽³⁾ However, experience at Battelle suggests that those managers and scientists from whom the basic inputs are elicited are much more comfortable thinking in terms of a range of probable values than with point estimates. Situations frequently occur where eliciting point estimates would not only be unreasonable but could discredit the entire analysis.

An alternative model utilizing probability distribution functions has been reported by Keeney.⁽⁵⁾ Several methods have been suggested in the literature for eliciting probability density functions. Most of these have had difficulty for the person supplying judgments if 1) he is not accustomed to thinking of probability distributions in terms of fractiles and/or 2) the probability distributions take on irregular

shapes. In eliciting probability distributions from engineers and scientists, at least, it has been generally more effective to pursue

questioning of the following nature (these are typical questions and responses):

We assume that we are eliciting information about a single attribute which we have identified as R&D cost X_i and some R&D alternative A_j .

Analyst: Since considerable uncertainty is associated with how well the alternative A_j services the attribute R&D cost X_i , I would first of all like to ask what you perceive as being the practical maximum R&D cost which you would expect.

Decision Maker: Given my understanding of the alternative, I would estimate that the maximum cost would not exceed \$10 million.

Analyst: And the minimum?

Decision Maker: Well, I know that a good deal of research is still required in order to even determine whether the underlying assumptions are valid. So I would expect that \$5 million is a practical minimum.

Analyst: Okay, you are doing great. I have marked these points on graph paper as you can see. Within this range what value or what range of values are most likely?

Decision Maker: A particular value is pretty hard for me to support; I am more comfortable about a likely range of values--probably \$7 to \$9 million.

Analyst: Do you envision equal likelihood for each value over that range?

Decision Maker: Yes, I think so.

Analyst: Then I'll draw a flat curve over that range. How about the shape of the curve between \$9 and \$12 million and between \$5 and \$7 million?

Decision Maker: My feeling is that the likelihood of costs being above \$7 million is much greater than the likelihood of their falling below \$7 million.

Analyst: Would you draw in your perception of what those two segments of the curve would look like?

Decision Maker: (sees this on graph)

This process is repeated for each alternative/attribute combination. Most subjects, at least in the R&D area, seem able to relate well to this type of approach. The fractile method has thus far proven difficult to apply, even in simple test situations. The most frequent objection may be roughly translated as: "Look, I feel that I know something about the probability distribution for this attribute, but you are forcing me to think in terms which make me uncertain as to whether I am correctly translating this information."

The method we used allows complete freedom in defining the shape of each probability density function. This greatly improved the ability of decision makers to estimate subjective probabilities. It is a simple procedure to normalize each probability density function estimate so that the area under the function equals one. This normalized probability density function is then used as a probabilistic estimate of the efficiency of each alternative.

Utility theory is fundamental to decision analysis since it is concerned with judgments of preferability or worth among any set of similar alternatives. A definitive discussion of utility theory is contained in Reference (6).

In this study we are primarily concerned with two aspects of utility theory. The first of these is the expression of the relative worth of the various attributes under consideration, w_i . Methods of determining relative worth have been widely discussed in the literature.^(1,2,4-9) Most methods attempt

not only to express attribute worth in the ordinal form, $X_k > X_l$, but also to scale these preferences in terms of relative degree of worth, termed cardinal ranking. The primary problem we have encountered in developing cardinal rankings (which is treated only briefly in the literature) is that of combining the judgments of several decision makers.

The approach we prefer is that of carrying a group's distribution of cardinal rankings through the analysis, developing scores by means of Monte Carlo simulation. This methodology is discussed in more detail later.

A second concern relates to the possibility of relative utility being affected by the magnitude of the attribute level. For example, the C-A model assumes a constant level of utility over all levels of attribute performance. That is, the attribute X_k may be assigned a value w_k without regard to the level of X_k which is achieved. This is a necessary condition where quantitative measures have not been developed for each evaluation factor. See, for example, References 3, 4 and 10. Alternatively, Miller⁽⁹⁾ and Ellis and Keeney⁽⁸⁾ have recognized the usefulness of separating attributes to a level where quantitative performance measures can be applied. While not every attribute lends itself to this process, a good many do--given some additional analysis. At Battelle we have found that R&D decision makers can deal in more concrete ways with attributes which are defined by some performance measurement.

Separation to measurable levels helps to further clarify the decision analysis, although it raises the issue of the adequacy of a given performance measure to represent attribute levels. Nevertheless, most decision analysis is aided, not hindered, by a clear statement of how well attributes are being serviced.⁽⁹⁾

As a result of separating attributes to a quantitatively measurable level, utility curves can be defined. An approach to defining utility curves has been outlined by Schlaifer⁽¹⁰⁾ and applied by Keeney.^(5,11) This procedure develops certain equivalent values given a gamble concerning two extreme values. The subject is always presented with two mutually exclusive choices. One will involve a 50-50 chance between two possible outcomes; the second will involve a certain outcome. The user specifies the minimum and maximum values which may occur. A more direct assessment technique has been necessary in our work at Battelle. The general approach is a visual one. On graph paper utility is scaled along the vertical axis from 0 to 1, marked off at intervals of 0.1. The range of attribute levels is scaled along the horizontal axis. The best attainable value is set to a utility of 1.0. The decision maker is then asked to evaluate several discrete values over that range in terms of their utility, relative to the best attainable value (1.0). For example, if the best (least) expected R&D cost is, say, \$5 million and the

worst expected cost is \$12 million, \$5 million is assigned a maximum normalized utility of 1.0. The decision maker is asked to indicate his relative utilities for \$7 million, \$9 million, \$11 million and \$12 million. A curve is then fit through these points. This graphic approach has proven beneficial by providing an immediate approximation of the decision maker's utility curve, facilitating exploration of his underlying assumptions and rationale during the same interview.

With these improvements a decision analysis methodology was formulated. It was then further defined into a decision model which will assist in determining the preferences of each public group for factors affecting waste disposal.

A Model for R&D Decision Making

In light of some of the limitations discussed, we outlined a decision model which will be more useful for decision analysis in R&D. An illustration of this model is shown in Figure 8 and is described in the following sections. It is assumed that the attributes and their performance measures have been determined as shown in Figure 10.

Probabilistic Statement of Attribute Levels

The first input to the analysis is an estimate of the degree to which each AWM disposal alternative serves each attribute. Most attributes have been assigned a quantitative performance measure. This estimate will take the form of a

FIGURE 8. Schematic of Model for R&D Decision Analysis

probabilistic statement of the chances of realizing various attribute levels, as was previously discussed. Figure 8, Diagram A, illustrates how estimates of attribute levels might look over several alternatives.

As a result of this initial information, a "relevant range" is defined. The relevant range is that range of performance levels for each attribute

which contains all possible outcomes of all of the alternative AWM concepts.

Utility on Attribute Levels

Secondly, input is gathered concerning utility of various attribute levels over the relevant range. These value assessments are elicited by the following general procedure:

Analyst: Consider the ideal attribute level profile presented below. This has been determined from the relevant range data by setting each attribute's level at the "best" end of the relevant range.

Ideal AWM Profile

Emplacement Cost	XX
R&D Costs	XX
Time Required for Development	XX
Protective Reaction	XX
Detectability	XX
Retrievability	XX
Emplacement Risk	XX
Long Term Risk	XX

We now want to consider various levels of each attribute because of uncertainty as to the actual level achievable. This will be accomplished by holding all but one attribute constant and setting that one attribute at four levels other than ideal.

On the graph paper provided (Figure 9), we have assigned the ideal attribute attainment level a value of 1.0. We would like to have you indicate your perception of the relative value of four less desirable attribute levels specified by the vertical lines.

This is repeated for each attribute, with a piecewise linear curve being fit through the respective points. The result of this exercise is a series of utility functions as sketched in Figure 8, Diagram B.

FIGURE 9. Attribute Level Utility: R&D Costs

Weighting of Attributes

At the same time that utility information is gathered, a weighting of the attributes is elicited. This is accomplished with a methodology similar to that described by Miller,⁽⁹⁾ including an attribute hierarchy as shown in Figure 10. The respondent assigns a weight to each level of the hierarchy. The weight for each lowest-level attribute is determined by multiplying together all relative attribute weights for each higher level attribute to which it is subordinate. For example, the overall

weighting assigned to protective reaction would be the product of (the relative weight assigned to risk of system failure) x (the relative weight assigned to internal risk) x (the relative weight assigned to protective reaction).

Since a number of individual decision makers are involved in making a group decision, the results can generally be represented by a frequency histogram as shown in Figure 8, Diagram C.

Analysis

A computerized simulation technique is used to assimilate the three basic data inputs gathered from the decision makers. This simulation predicts preferences based on the group's estimates of attribute levels, utility functions, and their evaluation of the relative importance of the attributes. The outputs are in the form shown in Figure 11, indicating the chances of various amounts of utility, or worth, each alternative has for the decision making group.

FIGURE 10. Attribute Hierarchy

FIGURE 11. Example Results of Total Utility of Two Alternatives

To simulate several decision makers' preferences, the three basic inputs shown in Figure 8 are combined in a unique fashion so that most group information is now included in the

final output shown in Figure 11. As shown in Figure 12, the probability density functions (pdf) on attribute levels (a probabilistic statement of the chances of an alternative achieving various attribute levels) and the group utility functions are combined to produce a transformed pdf on utility u_{ij} . These pdf's on utility are still not scaled to include the relative importance of each factor. This scaling and the aggregation of all attributes in an alternative is then the final step in the simulation. Monte Carlo simulation techniques (12-18) are used to perform the evaluation of the total utility for an alternative.

FIGURE 12. Transformation of Attribute Level Relationships

Let u_{ij} = pdf on utility of attribute i in alternative j ;
 w_i = pdf on cardinal ranking for attribute i ;
 U_j = total utility for alternative j .

Then the simulation will estimate total utility by the following objective function: $U_j = \sum_i u_{ij} w_i$. This analysis will be performed for each alternative for each group. The simulation will entail the generation of two random variables, one from the pdf on utility and the other from the pdf on cardinal ranking.

The transformation of the pdf on attribute levels through the utility

function can be accomplished using the following relationship from Hahn and Shapiro:⁽¹⁹⁾

$f(x)$ - density function of the random variable x ;
 $w = h(x)$ - monotonic function of x .

The transformed pdf resulting from the transformation of $f(x)$ through the function $h(x)$ is as follows:

$$p(w) = f(x(w)) \left| \frac{dx}{dw} \right|.$$

This transformation is well known for continuous density functions. However, for computational ease we use utility functions which are piecewise linear and density functions which are discrete. The same theoretical transformation of Hahn and Shapiro is achievable in the discrete case as follows:

Let $f(x_i)$ = probability of an occurrence in interval i ;

x_{i-1} = the beginning of the interval x_i ;

x_{i+1} = the end of the interval x_i ;

$u(x)$ = the piecewise linear segment of the utility function in the region of x_{i-1} and x_{i+1} .

Assume $u(x) = mx + b$.

Then the interval x_i can be transformed through the utility function:

$$u_{i-1} = u(x_{i-1}) = m(x_{i-1}) + b;$$

$$u_{i+1} = u(x_{i+1}) = m(x_{i+1}) + b.$$

The width of the interval u_i is now:

$$W_i = |u_{i+1} - u_{i-1}|.$$

Since $f(x_i)$ is the probability of an occurrence of the random variable x in the interval x_i and $f(u_i)$

is the probability of an occurrence of the transformed random variable u in the interval $u_{i+1} \rightarrow u_{i-1}$, then the following must be true:

$$f(u_i) = f(x_i),$$

and therefore the height $h(u_i)$ of the histogram in the i interval on the utility scale is:

$$h(u_i) = f(x_i) | W_i.$$

In this manner all pdf's on factor attainment can be transformed through the appropriate utility function for that factor.

Conclusion

In developing decision analysis in an R&D setting, it has been found necessary to modify and expand upon the methodologies reported in the literature. The product of this development is theoretically less rigorous in some ways than a few of the reported methods; but it is more rigorous in several key respects. Most important of these is the capability of this model to maintain the maximum degree of information flow from the decision makers through the analysis. This allows for communication of the uncertainty in each alternative which may be more important than the expected utility of these alternatives.

The model represents an attempt to use theoretically sound procedures modified in ways necessary for application in the decision making of an R&D organization.

PUBLIC RESPONSE MEASUREMENTS TOWARD NUCLEAR WASTE MANAGEMENT -

William S. Maynard (HARC) and
J. B. Burnham

Work during this quarter was directed toward 1) translating methods

identified during the previous quarter into questionnaire formats and 2) developing a package of information to precede the questionnaire and to acquaint respondents with both the nature of the nuclear waste problem and the specific factors on which they will make judgments.

The general methodology will differ from the FY74 approach in that the factors themselves (e.g., detectability, retrievability, costs, etc.) will be stressed rather than the generic disposal concepts. Both the information package and the questionnaire instrument will emphasize the factors; generic disposal concepts will be used primarily to illustrate how the factors could vary, e.g., high versus low detection of leakage, etc.

The first part of the questionnaire will be a direct ranking and magnitude estimation procedure which will yield estimates of the relative importance respondents attach to each factor. An example of the ranking and magnitude estimating procedure is presented in Exhibit A.

The rationale for emphasizing the factors rather than the generic concepts is that the estimates of importance should be as independent as possible of any given disposal procedure.

A second part of the questionnaire will consist of procedures to obtain utility functions from respondents for each factor. The objective here is to estimate the usefulness or psychological value of varying levels of each factor. To obtain informed judgments, each factor must be scaled into units which are meaningful to respondents. Exhibit A-3 shows an example of the ways in which a factor

could be scaled along several dimensions for inclusion in the survey instrument.

A final section of the questionnaire will consist of questions designed to measure attitudes about the problem of nuclear wastes and related issues. Responses to these questions will be useful in 1) characterizing the orientation of each sample of respondents and 2) as potentially useful normative data about the public's

perception of the nuclear waste problem. Examples of questionnaire items are shown in Exhibit C.

The information package consists of a script and list of required visual aids. These materials will be used to produce a film which describes the history and scope of the waste problem, defines the factors of interest for this study, and illustrates these.

EXHIBIT A

FACTORS FOR EVALUATING WASTE DISPOSAL METHODS^(a)

Retrievability - ease with which waste materials could be recovered for emergency procedures or for further processing; time required to retrieve wastes.

Emplacement operations - reliability and safety of procedures used in transporting and disposing of wastes.

Long-term stability - length of time the wastes would remain in their disposal location; resistance to acts of nature and acts of man.

Detectability - whether or not detection of waste leaks would be possible; length of time before leaks could be detected.

Protective reaction - how well population could be protected if leaks did occur.

Costs - dollar costs to dispose of wastes.

Distance from populated areas - remoteness or isolation from populations.

a. Further evaluation of these factors is continuing. It may well be that the decision methodology will require a consolidation of some of them.

EXHIBIT A-2

PART I RANKING: PART A

The seven factors for evaluating nuclear waste management procedures are shown on a separate page. Read over this list and then indicate below the order of importance which you think should be attached to them.

On the top line, write the name of the criterion or factor which you think is "most important." On the second line write the criterion which is next in importance. Continue until you have placed the factor on the bottom line that you feel is least important when compared to the other six.

- (most important) 1. _____
2. _____
3. _____
4. _____
5. _____
6. _____
- (least important) 7. _____

Next, we want to get a more precise estimate of how important each criterion is in comparison to the others. To do this, we want you to go down the list of criteria as they are ranked above and compare them two at a time. In this manner, you will compare #1 with #2, #2 with #3, #3 with #4, etc.

EXHIBIT A-3

RANKING: PART B

On the lines below, write the names of the factors as you arranged them on Page 2. On lines "1" and "2," write the names of the factors which you ranked as being first and second in importance. Then on lines "2" and "3," write the factors which you ranked as second and third in importance. Continue until on lines "6" and "7" you have written the factors which you earlier ranked as sixth and seventh in importance.

1. _____	____	4. _____	____
2. _____	____	5. _____	____
2. _____	____	5. _____	____
3. _____	____	6. _____	____
3. _____	____	6. _____	____
4. _____	____	7. _____	____

Now, consider the two factors in each pair, and indicate how important each is when compared with the other. In the box to the right of each factor, we want you to write numbers which describe how important the second factor of each pair is when compared with the first factor of each pair.

To illustrate: Say you ranked Detectability as #1, Costs as #2, and Distance as #3 in importance. You would first write factors as shown here:

1. <u>Detectability</u>	1,000
2. <u>Costs</u>	975
2. <u>Costs</u>	1,000
3. <u>Distance</u>	300

EXHIBIT A-4

Now, pick any number you wish and put it in the box to the right of Detectability. In the box to the right of Costs, you would then write a number that describes how important Costs are when compared with Detectability. For example, if you assigned Detectability the number 1,000 and you think Costs are almost as important as Detectability, you might write a number like 950 or 975 beside Costs, as shown in the example above. On the other hand, if you feel that Costs are much less important, you would put a much smaller number, perhaps 300 or 100.

Then for the second pair, you would write a number in the box beside Costs. In the box beside Distance, you would write a number which describes how important Distance is when compared to Costs. In the example above, Distance was thought to be much less important than Costs.

Return to the pairs of factors as you have written them and assign numbers to each pair of factors as described in the example. Remember that you can choose any number you wish to put beside the first factor in each pair. For the second factor in each pair, write a number that shows how important you feel it is compared to the first factor.

EXHIBIT B

PROTECTIVE REACTION

If nuclear waste escaped into man's environment, how effectively could the population be protected? How difficult would it be to protect humanity by either 1) evacuating specific areas or 2) containing the dispersal of leaked wastes?

	0	25	50	75	100
<u>Extent of Dispersal (Radius of miles affected)</u>	Worldwide	State-wide	5 miles	500 yards	No dispersal beyond container
<u>Population Exposed</u>	Worldwide	Major population center	Communities in immediate area	Maintenance personnel only	No one
<u>Difficulty in Containing Dispersed Wastes</u>	Impossible	Very difficult	Moderately difficult	Slightly difficult	Very easy, using existing technology
<u>Extent of Containment of Leaked Wastes</u>	Total dispersion	Slight containment	Moderate ability to contain	Almost complete containment	Total containment
<u>Examples</u>	Ice disposal, Ocean disposal, Earth orbit		Geologic disposal	Surface disposal	

EXHIBIT B-2

LONG-TERM STABILITY

After the waste material is safely stored or disposed, how likely are future leaks of the wastes? How long could we expect the wastes to remain in their storage area? How susceptible are the stored/disposed wastes to "Acts of God" such as volcanoes, geologic upheavals, tornados, and meteorites, or to manmade acts such as sabotage, war, or indifference on the part of future generations?

	0	25	50	75	100
<u>Length of time which wastes remain in place</u>	0-100 years	500 years	10,000 years	500,000 years	over 1,000,000 years
<u>Susceptibility of method to natural phenomena</u>	Very susceptible	Fairly susceptible	Somewhat susceptible	Only slightly susceptible	Not at all susceptible
<u>Susceptibility of method to man made actions</u>	Very susceptible	Fairly susceptible	Somewhat susceptible	Only slightly susceptible	Not at all susceptible
<u>Examples</u>		Surface storage	Geologic disposal	Ocean disposal Earth orbit Ice cap disposal	

EXHIBIT C

This section contains some general questions about nuclear wastes and other issues related to nuclear power.

DIRECTIONS. Read each statement and indicate whether you agree or disagree with it. Circle one of the five choices after each question.

	Strongly Agree	Agree	Partly Agree/ Partly Disagree	Disagree	Strongly Disagree
1. Before I took this questionnaire, I hadn't thought much about the problem of nuclear wastes.	SA	A	AD	D	SD
2. The Atomic Energy Commission is doing a good job of keeping the public informed about the advantages and disadvantages of nuclear power.	SA	A	AD	D	SD
3. Nuclear power is the most promising source of electrical energy for the next 30 years.	SA	A	AD	D	SD
4. Other problems facing the world today such as overpopulation and the economy are much more serious than deciding what to do with nuclear wastes.	SA	A	AD	D	SD
5. The best solution to the problem of radioactive wastes is to put them where no one can ever get to them.	SA	A	AD	D	SD
6. The Atomic Energy Commission is doing a good job of insuring that nuclear power is acceptably safe.	SA	A	AD	D	SD
7. Based on past operating experience, nuclear energy has proved itself to be a clean and safe source of electrical power.	SA	A	AD	D	SD
8. Much of the money now being spent on nuclear power should be spent instead on developing other sources of energy such as geothermal and solar power.	SA	A	AD	D	SD
9. The leakage of nuclear wastes is one of the most serious threats to mankind.	SA	A	AD	D	SD
10. Nuclear waste material should be temporarily stored rather than permanently disposed of.	SA	A	AD	D	SD
11. No more nuclear power plants should be built until we have a safe solution to the problem of nuclear wastes.	SA	A	AD	D	SD
12. Considering the need for energy, our society should be willing to tolerate the risks from nuclear power.	SA	A	AD	D	SD

Sex: M F Education (years of school or highest degree) _____

Age: _____ Occupation: _____

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TASK 6: WASTE MANAGEMENT PLAN

K. J. Schneider

Potential formats for the overall waste management plan have been investigated. One key part of the format for the plan is expected to include a matrix of plans for basic categories of activities for each of the waste disposal concepts. The plans will include graphic presentations, key milestones, and decision points. Currently the plans are aiming at selection of one or two final disposal concepts about 1980 and routine operation of the concepts about 2010.

A significant need is to expand the effort so that disposal of all

nuclear wastes is included in the planning function. Then the planning studies must identify areas where short-term R&D is needed in order to have adequate information to make a decision between waste management alternatives.

The planning is aimed at bringing the state of knowledge about each

concept to a sufficient state for decision making. When this condition is reached, the information will be used in conjunction with the decision model for the AEC to recommend decisions. At this point, the R&D plan would be followed through to the implementation stage.

2.0 SEPARATION OF ACTINIDE ELEMENTS FROM RADIOACTIVE WASTE

E. J. Wheelwright - Project Manager

The objective is to develop the technology for removing the biologically significant long-lived radionuclides (principally actinides) from commercial radioactive wastes or during fuel reprocessing so that the bulk of the waste would become radioactively nontoxic in less than 1,000 years. The FY75 objective is to determine the technical feasibility.

SUMMARY

Waste partitioning at PNL in FY74 was limited to a study of the feasibility of applying the concept to Hanford Purex Acid Waste. No laboratory work was performed. There is no waste of this kind at the present time, and such waste will be generated only if N Reactor fuels are processed at the Hanford Purex plant. A final report will be issued during October 1974.

The scope has been increased and the directions altered for FY75. The funding level has been set at \$400,000, with half of the effort directed toward partitioning of waste solution or solidified waste and the other half of the effort directed toward identifying alternative processing flowsheets which would accomplish the partitioning objectives during normal fuel reprocessing. No attention is to be given to AEC waste in FY75.

Work assignments have been made for Tasks 1, 2, and 4, defined in the FY75 program, and work has been started. In addition to these three defined work areas, work has been started to define the scope, objectives, and methodology of a study of

the incentives for waste partitioning. When these work assignments have been completed, we should have answers to the questions of both technical feasibility and desirability of partitioning.

TECHNICAL PROGRESS

PNL assisted the AEC-RL in hosting the Second Multi-Site Waste Partitioning Meeting on September 4 and 5, 1974. Both FY74 and FY75 programs were discussed. ORNL and PNL will work on commercial waste while Allied Chemical will work on AEC waste. The new thrust is to consider partitioning as part of the chemical reprocessing plant operation.

Ray Walton, AEC Headquarters, assigned these persons as coordinators in establishing guidelines for all AEC contractors working on partitioning:

Target No. 1 - Required degree of actinide decontamination of fission-product fraction

Assigned to ORNL - Rex Leuze

Target No. 2 - Acceptable storage of decontaminated waste salts (fission-product fraction)

Assigned to PNL - George Jansen/
Earl Wheelwright

Target No. 3 - Acceptable storage of actinide fraction

Assigned to PNL - George Jansen/
Earl Wheelwright

Target No. 4 - Required degree of fission-product decontamination of the actinide fraction

Assigned to ORNL - Rex Leuze

Target No. 5 - Disposal of the actinide fraction

Assigned to ORNL - Rex Leuze

Target No. 6 - Analytical procedures

Assigned to AAC - Bill Maeck

Offsite attendees were:

R. D. Walton	AEC-HQ
E. H. Hardison	AEC-OR
W. H. Hale	DuPont
R. E. Leuze	ORNL
J. O. Blomeke	ORNL
W. D. Bond	ORNL
W. J. Maeck	Allied Chemical Corp.
L. D. McIsaac	Allied Chemical Corp.

TASK 1: ROLE OF SOLIDS IN WASTE

PARTITIONING - F. P. Roberts

The solids present in high-level radioactive waste have three sources:

- (1) Residues resulting from incomplete dissolution of the fuel--primarily difficultly soluble fission products, but may include undissolved PuO_2 .
- (2) Cladding fines from the mechanical head-end steps.
- (3) Solids formed during the chemical processing of the fuel and subsequent waste treatment steps such as denitration and concentration.

Experimental studies are planned to characterize the solids and evaluate the effects of such solids upon processes used to extract the actinides from reprocessing plant streams or from the waste. A small amount of irradiated Yankee Reactor fuel has

been obtained, and work is in progress to dissolve this fuel under conditions which will yield a typical dissolver solution. This will then be processed through tributylphosphate (TBP) cycles simulating the Purex process to yield a typical high-level waste. Solids present at each stage will be characterized. However, equipment is being procured, and the fuel dissolution and simulated reprocessing is scheduled for the next quarter.

TASK 2: PARTITIONING OF SOLIDIFIED WASTE - J. A. Partridge

The initial literature search has been completed, and the laboratory work has been started. The work in this task will be directed toward calcine as the form of solidified waste to be used as a starting material for a partitioning process. Initial laboratory studies will be performed with calcine prepared from a synthetic liquid waste corresponding in composition to the waste resulting from the AGNS flowsheet. Work is now under way to prepare some of this synthetic calcine.

Plans are also under way to prepare a calcine from typical high-level waste in the 325 Building High Level Radiation Facility. Tests with this calcine will be used to verify the results obtained with the synthetic calcine. The waste to be used for preparation of this calcine will be that obtained from processing irradiated Yankee reactor fuel through

TBP cycles simulating the Purex process in conjunction with the work in Task 1.

It is anticipated that complete dissolution of the calcine prior to partitioning will be difficult, and that such steps as molten salt fusions may be required to achieve complete dissolution. Initial dissolution studies will be with nitric acid.

TASK 3: PARTITIONING NON-HIGH-LEVEL WASTE (not yet assigned)

No progress to report.

TASK 4: LOW-ADDITIVES PARTITIONING FLOWSHEET - J. L. Swanson

Literature data on the extraction of americium by phosphonates, phosphinates, phosphine oxides, and bidentate phosphonates have been examined. Of these classes of compounds, only phosphine oxides show little promise for the extraction of americium from Purex process first-cycle waste (likely $\sim 3M$ HNO_3). We have begun experimental testing of dibutylbutylphosphonate (DBBP). We have requested that some butyldibutylphosphinate be synthesized and have initially contacted the Wateree Chemical Company regarding availability of bidentate phosphonates. These data indicate that Am could be extracted from first-cycle waste solution by DBBP and that the Am could be kept in the organic phase or stripped into the aqueous phase in subsequent columns, depending on the aqueous composition.

Extraction of Am by 100% DBBP has been studied at HNO_3 concentrations of 10 to 16M to supplement the data available in the literature for HNO_3 concentrations up to 7M. The americium distribution coefficient was found to be 0.8 at 10M HNO_3 , 1.0 at 12M HNO_3 , and 1.9 at 16M HNO_3 . Data from the literature include values of 2.2 at 3M HNO_3 , and 0.8 at 7M HNO_3 , and flow rate employed. Hopefully, partial separation from lanthanides (and other fission products) can be achieved in sequential operations under varying conditions. This anticipation is based on the fact that literature data for extraction of selected lanthanides (La, Pr, and Eu) by 58% DBBP-kerosene show variations in relatively high and low-acid extractability as well as in extractability itself.

Extraction of HNO_3 by 100% DBBP has also been determined at high concentrations to supplement available literature data. Distribution coefficients were found to be 0.56 at 5.5M HNO_3 , 0.45 at 8.5M HNO_3 , 0.36 at 12.4M HNO_3 , and 0.43 at 15.7M HNO_3 . This substantial extraction of HNO_3 will likely complicate development of a suitable flowsheet.

Tests with 100% DBBP in our single-stage miniature centrifugal contactor have shown good phase separation when the aqueous phase contains about 3M HNO_3 , but attempts to operate with only about 1.5M HNO_3 in the aqueous phase were unsuccessful.

3.0 DECONTAMINATION AND DENSIFICATION OF CHOP-LEACH CLADDING RESIDUES

R. L. Dillon and R. S. Kemper - Project Managers

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 feasibility and 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

Analysis of a piece of irradiated Zircaloy-4 fuel cladding has been started to determine the distribution of transuranic contamination in the cladding. Initial results confirm the low level of activity expected. Precision analysis is currently in process to identify and quantify the radioactive species present in the cladding metal.

Development of a preliminary decontamination and densification method requires 1) a process for removal of zirconium dioxide from metallic Zircaloy cladding, 2) melting and casting of Zircaloy-stainless steel-Inconel alloy ingots, and 3) removal from the molten alloys of the last

traces of transuranic contamination. Experimental work modifying zirconium oxide corrosion films in anhydrous hydrogen fluoride at 550°C to 620°C followed by aqueous removal of the modified zirconium dioxide film has shown promise as a potential process step. Zircaloy-2 autoclaved in 360°C water to 35 mg/dm² weight gain was easily stripped to bare metal with gross metal loss of less than 20 microns. Zircaloy samples with heavy oxide corrosion layers were more difficult to strip. It will require process optimization to produce satisfactory results.

Six experimental 8.2 kg (18 pound) ingots have been vacuum induction melted in graphite crucibles and cast

into graphite molds. The compositions that were melted are as follows:

90 w/o Zircaloy-2 - 6.7 wt% 304L -
3.3 w/o Inconel 718
85 w/o Zircaloy-2 - 10.0 wt% 304L -
5.0 w/o Inconel 718
80 w/o Zircaloy-2 - 13.4 wt% 304L -
6.6 w/o Inconel 718

It appears that the 85 wt% Zircaloy-2 alloy can be poured at a temperature lower than can the other two compositions and that the resulting crucible reaction is at a minimum. Rockwell C hardness measurements of this composition are R_c 33-36. This composition has been sectioned on a power hacksaw using a high-speed steel blade.

TECHNICAL PROGRESS

ACTINIDE DISTRIBUTION IN CHOP-LEACH FUEL HULLS - B. Griggs

Design of surface decontamination and subsequent molten metal decontamination processes for fuel hulls requires an accurate knowledge of the distribution, chemical state and type of transuranic contamination of the hull material. An initial sample for analytical determination of these parameters was obtained from a fuel rod (F-2 [HQ]) exposed in Saxton Reactor about six years ago. Fuel composition was 6.6% PuO_2 in UO_2 . A 0.6 cm piece of F-2(HQ) cladding was lightly surface abraded to remove corrosion product oxide and etched 30 seconds total in two nitric-hydrofluoric acid etch solutions. These decontaminated pieces were moved to an uncontaminated area and etched 20 seconds more in two etches and briefly leached in concentrated nitric acid for complete surface decontamination. Less than 25 microns

were removed from the surfaces in the total treatment sequence. The sample was then dissolved in etch solution in two approximately equal steps. One sample contained the dissolved outer and inner 0.14 cm surface layer and the other sample the residual 0.28 cm center. Radiation readings with survey instruments after decontamination showed 20 mR at contact and no detectable α . Total α counts and γ scans of the two samples are nearly identical, indicating the completeness of surface decontamination. The two samples are now being analyzed by emission mass spectrometry, α energy analyses and chemical separation of important species for accurate determination of transuranic contamination resulting from uranium and thorium impurity in the original Zircaloy.

PRELIMINARY DECONTAMINATION AND DENSIFICATION PROCESS - R. G. Nelson and B. Griggs

Review and selection of potential processes for decontamination and melting of chop-leach cladding residues have been completed and discussed in previous publications.^(1,2) Briefly stated, the process will develop: 1) a gas phase treatment of cladding hulls by a fluoride to render the corrosion product oxide soluble in dilute aqueous fluoride solutions, 2) oxide removal and light etching in dilute aqueous fluoride solutions (nitric-hydrofluoric acid or oxalic-hydrofluoric acid-hydrogen peroxide solutions), 3) melting of the hulls into an eutectic stainless steel-Zircaloy alloy by induction heating in graphite crucibles with

and without a flux or solid adsorbent. If the metal is to be reused for reactor applications, a more conventional cold wall crucible and arc melting of the segregated stainless steel, Inconel, and zirconium alloys would be required.

The gas phase fluoride treatment of the autoclaved Zircaloy to make the corrosion product films readily removable by aqueous decontaminants and the decontamination of the treated zirconium alloys are process steps requiring immediate development. The oxidized metal surfaces on which the gas phase conditioning process will be demonstrated are being produced by autoclaving Zircaloy-2 tubing, Inconel 718, and 304 stainless steel strip at 360°C in high-purity water. Samples of Zircaloy-2 removed after 1546 and 3446 hours of autoclaving with average weight gains of 35 and 47 mg/dm² respectively have been used for experimentation.

Samples of corroded Zircaloy have been treated in gaseous hydrogen fluoride (HF)-argon mixtures at 530°C to 620°C in the apparatus shown in Figure 13. Treatment times from 5 minutes to over one hour have been explored in 5, 50, and 100% HF in argon. Thin films (35 mg/dm² weight gain) have been modified sufficiently in 5% HF at 580°C in 20 minutes to effect their complete removal in the subsequent aqueous etch. The somewhat heavier film of 47 mg/dm² was resistant to the same treatment but was satisfactorily modified in 15 minutes at 590°C in 100% HF to produce a nearly complete film removal in subsequent aqueous treatment.

FIGURE 13. Gas or Molten Salt Process Equipment

Weight gains during HF treatment range between a few and several hundred mg/dm². In general only those samples showing several hundred mg/dm² weight gain can be subsequently stripped. However, this will also be a function of the original oxide thickness, and samples with thicker oxides may require weight gains of more than 1000 mg/dm² for complete modification. An X-ray diffraction pattern from a modified oxide corresponded to ASTM card number 20-1471 (ZrO_{0.44}F_{3.12}). Studies of the various parameters of the reaction including the use of other gases such as CF₄ and BF₃ are being planned in order to develop the best oxide modification process.

The aqueous removal process has been successfully accomplished with a decontaminant developed for gas reactor decontamination.⁽³⁾ In general, if the reagent reacts too rapidly

with Zircaloy metal, it will severely attack the metal before completely removing the modified film. The reagent found useful for stripping the modified film consists of 0.4 M ammonium oxalate - 0.16 M ammonium citrate - 0.1 M ammonium fluoride - 0.3 hydrogen peroxide used at 90°C for periods from 20 minutes to over an hour. It is planned to modify this solution soon to optimize its stripping of the modified oxide film. The corrosion rate of bare Zircaloy in the solution at pH 5 is roughly 8 microns/hour. The treatment of fluoride modified films in the solution causes the films to slough off before dissolving, indicating a complex mechanism in either the fluoride modification process or the

aqueous removal process. The decontaminating efficiency of the process on contaminated tubing has not yet been explored.

Zircaloy-stainless steel-Inconel alloys have been melted to determine both optimum melting practice and alloy properties. Table 8 is a summary of the heats made during this reporting period. Figures 14, 15, and 16 are photographs of the crucibles and ingots of the first three heats listed in Table 8. These ingots are all of the same composition, 85 wt% Zircaloy-2 - 10 wt% 304L stainless steel - 5 wt% Inconel 718. The longitudinal crack in Heat no. 8-8-74 can be attributed to the poor vacuum during melting and pouring and/or the off composition resulting

TABLE 8. Summary of 8.2 kg (18 Pound) Heats Melted in Graphite

Heat Number	Composition, Wt%			Maximum Temp °C	Pour Temp °C	Relative Crucible Reaction	Leak Rate Microns Per Min. 5 Min. Test	Remarks
8-8-74	85	10	5	1350	1295	Mild	10.0	0.85 kg (1.88 pounds) not melted, left in crucible. Highly discolored. Longitudinal crack length of ingot; no discoloration on ingot.
8-29-74	85	10	5	1365	1315	Mild: Crucible Reusable	0.1	0.08 kg (0.18 pounds) left in crucible; 7.25 kg (15.98 pounds) ingot weight; hardness of splattered material Rockwell C-35-37; no discoloration on ingot.
9-5-74	85	10	5	1307	1291	Very Mild: Crucible Reusable	0.12-0.14	Excellent pour; 8.13 kg (17.92 pounds) ingot weight. Amount left in crucible not measurable. Ingot is being sectioned for testing and evaluation. No discoloration on ingot.
9-12-74	90	6.7	3.3	1415	1415	Severe	0.12	Very poor casting. Consists of blobs of metal fused together. Mush pour. Higher pour temperatures required for good casting practice. 2.18 kg (4.81 pounds) left in crucible. No ingot discoloration.
9-18-74	80	13.4	6.6	1355	1315	Mild: Crucible Reusable	0.24	Good fluidity, good pour. Ingot is full of cracks.
9-26-74	80	13.4	6.6	1300	not applicable	Severe: Crucible had to be broken to remove ingot	0.16	Ingot was allowed to freeze in the crucible, providing an extremely slow cool down. No visible cracks in ingot.

Negative No. 745588-1

FIGURE 14. 85 wt% Zircaloy 2 - 10 wt% 304L Stainless Steel - 5 wt% Inconel 718

Negative No. 745588-3

FIGURE 15. 85 wt% Zircaloy 2 - 10 wt% 304L Stainless Steel - 5 wt% Inconel 718

Negative No. 745588-4

FIGURE 16. 85 wt% Zircaloy 2 - 10 wt% 304L Stainless Steel - 5 wt% Inconel 718

from incomplete melting. Heat no. 9-5-74 was sectioned using a power hacksaw with a high-speed steel blade. Hardness reading of sectioned samples were Rockwell C-33-36. Prior to hacksawing, this ingot was placed in a three jaw chuck, which caused several cracks to appear in the ingot.

Metallographic techniques are currently being developed for these alloys. Photomicrographs of the as-polished as-cast composition 85 wt% Zircaloy-2 - 10 wt% 304L stainless steel - 5 wt% Inconel 718 are shown in Figure 17. This preliminary work

shows three distinct micro constituents. The edge specimen shows the chilling effect of the mold wall.

Heat No. 9-12-74 a 90 wt% Zircaloy 2-6.7 wt% 304L Stainless Steel-3.3 wt% Inconel 718 composition was poured at approximately 1415°C. The pour was mushy and the ingot consisted of blobs of metal fused together. Even at the ~1415°C pouring temperature, crucible reaction was severe. This coupled with the higher pouring temperatures required for good ingot practice makes this alloy a poor candidate for graphite melting.

Negative No. 4J1249A

Negative No. 4J1250A

FIGURE 17. As Cast 85 wt% Zircaloy 2 - 10 wt% 304L Stainless Steel - 5 wt% Inconel 718 Heat No. 9-5-74 as Polished

Heat No. 9-18-74 an 80 wt% Zircaloy 2-13.4 wt% 304L Stainless Steel-6.6 wt% Inconel 718 exhibited good fluidity and good pouring characteristics with a mild crucible reaction at a pouring temperature of 1315°C. The ingot, however, was full of cracks.

Heat No. 9-26-74 was an attempt to produce slow cooling of the 80 wt% Zircaloy 2 alloy by allowing it to cool in the crucible from 1300°C. This resulted in what appears to be a crack-free ingot, however, crucible reaction was severe, and the crucible had to be broken to remove the ingot.

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

Melting is performed in a 50 kW, 3000 cycle vacuum induction melting furnace originally designed for uranium melting. The crucible and coil assembly are of the tilt type. A graphite susceptor heats an outer graphite crucible which in turn heats the inner or melting crucible. The vacuum pumping system consists of a 10,000 liter per second oil diffusion pump Leybold type-EG-A5 mechanical Booster pump, and a Leybold 200S rotary gas ballast backing and roughing pump.

Melting stock is sheet chopped into 2.5 cm squares having thicknesses of Zircaloy 1.79 mm (0.070 inch), 304L Stainless Steel 0.94 mm (0.037 inch), and Inconel 718 0.58 mm (0.023 inch). The 14.0 cm (5.5 inch)

crucible is charged with layers of the 304L - Inconel mixture between layers of Zircaloy-2.

The furnace is charged in the afternoon and pumped overnight. The following morning chamber pressure is less than one micron. A five-minute rate of rise test is made on the furnace chamber prior to starting the heat. During the last five heats this has been maintained at less than 0.25 microns per minute. The heat is started with a power setting of 5 kW and gradually increased to 10 to 11 kW. The pouring temperature is reached in approximately 2.5 to 3 hours and held for 30 minutes, and the metal then is poured. The temperature is measured with a chromel-alumel thermocouple located on the outside of the melting crucible. Also optical pyrometer readings are taken and are compared with the thermocouple readings. At about 1250 to 1300°C the chromel-alumel thermocouple is destroyed and the temperatures are then calculated from the optical pyrometer readings. The temperature at which to pour is determined by the temperature uniformity of the liquid surface, the absence of unmelted material, and the operator's observation of its fluidity. The metal is poured into a 8.9 cm (3.5 inch) diameter x 30.5 cm (12 inch) deep split graphite mold. Five of the six 8.2 kg (18 pound) heats made to date have been poured into the same mold and no mold reaction has been observed.

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4.0 WASTE TREATMENT AND HANDLING PROCESSES

L. K. Mudge - Project Manager

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

The principal program objective is to develop a pyrolysis-incineration process for safe and economical reduction of the volume of alpha wastes by 10- to 50-fold as a precursor to long-term storage and/or disposal. The technical and economic merits of this process will be defined to allow its comparison to other processes being considered for volume reduction of alpha wastes. The program objectives 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 non-contaminated simulated wastes and later with actual wastes from Hanford operations.

SUMMARY

During July, August and September efforts were made to identify potential problems in mechanical design, select components for an experimental facility, define a revised scope for the research program, and initiate detailed design of the pyrolysis-incineration experimental facility. An annual report for FY74, BNWL-1861, was completed.

TECHNICAL PROGRESS

The design of a pyrolysis-incineration system to handle low-level radioactive solid wastes, begun in March 1974, is continuing at this time. Current work has concentrated on gathering information essential to the design. Various similar processes were studied, including incineration, pyrolysis, and coal gasification systems, to identify the equipment used and the success or

failure of this equipment. Based on these studies, information from equipment manufacturers, and the unique process requirements of our system, preliminary equipment selections were made. In a visit to equipment suppliers and other project sites, several contacts were made to identify current related work and potential problem areas. Several observations were made from the study of the related processes and equipment for an alpha waste processing plant.

- Incoming solid wastes require close inspection to remove all possible undesirable material, such as glass bottles and metal, before further processing.
- For our application, the most desirable type of shredding apparatus for the solid waste appears to be the counter-rotating system with cam-type blades and an automatic reversing feature to prevent jamming.

- The average size of waste fed into a reactor can be 2 to 4 in. for a batch feed input but should be around 1 in. for a continuous feed process.
- Horsepower requirements for shredding of the solid wastes will be in the range of 0.8 to 1.5 horsepower-minute per pound.
- The feed hopper used for transfer of shredded solid wastes to the reactor will most likely be sealed at its inlet to prevent air leakage into the process.
- Screw feeding of waste into the reactor seems most promising of all techniques investigated at this point. The screw feeder will be a straight barrel, constant-pitch device, since the use of both a tapered screw or a variable pitch screw has been somewhat unsuccessful.
- A negative pressure air flow system is preferable to a positive pressure system when considering the feeding of radioactive-bearing solid wastes.
- A modified version of the grate used in the Wellman-Galusha[®] gasifier looks satisfactory to our needs. The most durable materials of construction for the grate system for ash removal are Inconel, Hastelloy C, and Hastelloy X.
- Careful design of the valving used at the ash removal area of the reactor will be necessary to prevent excessive leakage and wear. Numerous processes have experienced problems in this area.
- Allowance for expansion of the pyrolysis-incineration reactor due to thermal cycling is essential.
- A cyclone separator can be used to remove the majority of particulates in the off-gas stream. A fairly constant gas flow rate will be required to insure efficient particulate removal.
- Water scrubbers used to date for particulate removal have been, on the whole, unsatisfactory. It would be desirable to eliminate this component from our system if at all possible. The water scrubber creates an off-gas stream saturated with water, a major cause of premature high efficiency particulate air (HEPA) filter failure.
- For the areas in contact with the off-gas stream, corrosion resistant materials of construction appear to be the higher stainless steels in the 300 series (314, 316) and Hastelloy C.
- The off-gas cleanup system is a critical item in a facility of this type. A system which eliminates the tars very early and runs completely dry seems most desirable. Methods other than aqueous scrubbing for chloride removal should be examined.
- The flow rate of the off-gas stream should be kept to a minimum. Methods other than excess air addition should be considered for the cooling of the off-gas stream.
- The HEPA filters currently in use in most installations of this type

have a relatively short life and handle relatively low flow rates. A new filter has been recommended by General Electric Company personnel: a Flanders Superflow® filter capable of handling a larger flow rate over a longer time period before pressure drop becomes excessive.

- A parallel bank of HEPA filters is advantageous to avoid blowback through the system if one bank of filters is blinded.

Based upon these observations, developed design criteria, and the decision to construct a reduced-capacity experimental facility, detailed design has begun. A schematic of the pyrolysis-incineration reactor to be used is shown in Figure 18. The reactor will have an inside diameter of about 30 cm and a height of about 200 cm. It will be equipped with a mechanical grate and distributor as shown. No gas cleanup or feed shredding equipment will be included in the facility. This facility will be used primarily to establish process

feasibility, to study problems of air channeling in the pyrolysis-incineration reactor, and to identify the composition and amount of tars contained in the off-gas stream. Detailed designs of the reactor, the ash removal system, and the bed compaction-distribution mechanism are currently under way. The designs are expected to be complete by mid-October.

FIGURE 18. Pyrolysis Incinerator Reactor

51,060

63-77

BNWL-1876

5.0 TRITIUM SEPARATION AND FIXATION

H. H. Van Tuyl - Project Manager
L. L. Burger - Principal Investigator

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

SUMMARY

Reexamination and evaluation of the solvent extraction and vapor pressure data of aqueous solutions have confirmed earlier conclusions that the isotopic effect on extraction of water by organic compounds is very small. Similarly, the effect of solutes on the vapor pressure isotopic ratio is extremely small regardless of their effect on the ordering or "structure" of liquid water.

Several approaches to fixing tritiated water into organic polymers have been considered. A method involving the formation and polymerization of tritiated acetaldehyde was chosen for initial study. The reaction steps have been studied and samples prepared.

It was decided that the initial experiment for isotope separation by molecular excitation should involve a two-level process: simultaneous pumping of a combination IR band by laser radiation and electronic excitation from the higher IR band. The choice of this method over multilevel vibrational excitation was due to the absence of fundamental vibration-rotation data for excited states of

the water molecule. The frequencies to be used have been chosen, and the experiment is being designed.

TECHNICAL PROGRESS

WATER EXTRACTION STUDIES -

R. D. Scheele, L. L. Burger

During the past year a number of experiments were carried out to examine isotopic separation by extraction of water by organic solvents. Occasionally separation factors differing from unity were observed, but invariably these were shown to be the result of experimental errors. It may be instructive to summarize some of these observations.

Water containing heavy hydrogen isotopes possesses more structure (more and stronger hydrogen bonds) than does light water. Any separation by solvent extraction would be due to distinctive differences in hydrogen bonding with the solvent molecules. The latter do exist and are demonstrated by differences in a wide variety of physical properties, (1-9) such as:

- IR Spectral Shifts and Intensities
- NMR Shifts
- Solubility

- Thermodynamics of Solutions, e.g., ΔH and ΔG of Solution
- Volume Effects, Expansibility, Temperature of Maximum Density
- EMF Measurements
- Viscosity
- Dielectric Relaxation
- Acoustical Absorption

The ranges of these physical properties are appreciable as various classes of solvents, or solvents containing specific functional groups, are dissolved. For example, the IR frequency shifts (OH stretch) for DOH in various solvents are shown in Table 9.

TABLE 9. OH Stretching Frequencies of DOH Solutions

Solvent	ν , cm^{-1}
Vapor	3707
CCl_4	3662
$\text{CH}_2\text{Cl}-\text{CH}_2\text{Cl}$	3635
Et_2O	3550
Tetrahydrofuran	3524
Acetone	3520
Triethylamine	3515
Diisopropyl phenyl phosphonate	3480
Tributyl phosphate, TBP	3470
Dibutyl butylphosphonate	3470
$\text{D}_2\text{O}(\ell)$	3407
Pyridine	3405
$\text{D}_2\text{O}(\text{s})$	3300

Another interesting "indicator" is the difference in solubility of light water and heavy water in organic liquids. Table 10 lists a few examples. The data are from Rabinovich⁽⁴⁾ except for TBP. This compound has

been extensively studied in our laboratory. The theoretical value assumes the only energy important is that required to break the bonds between water molecules for which $\Delta H_{\text{D}_2\text{O}} - \Delta H_{\text{H}_2\text{O}} = 330 \text{ cal/mole}$.

TABLE 10. Solubility of H_2O and D_2O in Organics

Compound	$(N_{\text{H}} - N_{\text{D}})/N_{\text{H}}^{(a)}$
Triethylamine	0.30
CS_2	0.21
Benzene	0.17
CHCl_3	0.15
Furfural	0.11
Et_2O	0.07
Sec-BuOH	0.04
TBP	0.02
"Theory"	0.15

a. N_{H} = mole fraction H_2O in solvent saturated with pure H_2O , etc.

Using the various physical properties as indicators of the degree of structure, the effects of different organic and inorganic solutes can be examined. From such studies it has been shown that some solutes tend to promote the degree of order or structure of water while others tend to destroy it. (4, 10-12) In the former category are small cations, most alcohols and miscellaneous organic compounds such as tetrabutyl ammonium complexes. The latter or structure breaking solutes include most cations and many large anions such as perchlorate. It seemed reasonable to expect that if solvent extraction isotope effect existed, then it could be

enhanced by either promoting or destroying the water structure by addition of solutes. Furthermore, the latter could be tested independently by measuring the vapor pressure isotopic ratio of the solutions.

Studies were carried out with both tritiated water and deuterated water using ethers, ketones, organic phosphates and phosphonates, and amines. Some of these, such as the phosphonates and amines form very strong hydrogen bonds and might be expected to show the maximum effect. A few experiments were carried out on the extraction of HNO_3 and HNO_2 both of which form very strong complexes with solvents like tributylphosphate and ion pair complexes with amines.

With some of the solvents, dilutions with nonpolar diluents were also tried. Several auxiliary investigations were carried out on the equilibrated organic phase, such as precipitation of a portion of the water by dilution with CCl_4 and distillation of water from the organic.

There was some indication that dibutyl butylphosphonate gave a small preferential extraction of HTO , but the analytical measurements were not precise enough to obtain a firm value. No solvent showed a separation factor different from unity by more than 1 or 2%.

The solutes tested are listed in Table 11 along with the separation factors, $(\text{H}_2\text{O}/\text{HTO})$ vapor/ $\text{H}_2\text{O}/\text{HTO}$ liquid, measured.

It is concluded that water isotopic enrichment by solvent extraction is very small. Likewise, addition of solutes to change the degree of order

or structure of the liquid water has a very small effect on the vapor pressure isotopic ratio.

The small solvent extraction effect is not surprising since the differences in bond strengths between water molecules and between solvent and water are small. Thus, although the isotopic effects on the physical properties of one phase are real and significant, compensation occurs between two phases in equilibrium. We could not reproduce the reported⁽¹⁴⁾ separation of ~ 1.4 using alkyl amines and are inclined to believe the value of 1.01 found by Skripov and Rusinov⁽¹⁵⁾ to be more reliable. The compensation explanation is supported in a thermodynamic argument by Linderstrom-Lang.⁽¹⁶⁾

TRITIUM FIXATION - E. C. Martin,
D. C. Elliott, J. Franz

Among the numerous approaches proposed to limit the spread of tritium into the environs has been the incorporation of tritium into organic polymers.⁽¹⁷⁾ Implicit in the formulation of such a polymer is the requirement that the tritium atom be situated in such a manner that exchange with hydrogen (from atmospheric moisture, etc.) be minimized. Thus all polymeric materials in which tritium would be in a labile condition, i.e., alcoholic, carboxylic, ammoniacal groups, etc., must be ruled out. Since hydrogen exchange with carbon-hydrogen skeletal functions, on the other hand, would be minimal, polymers have been sought that would meet this condition. The ideal solution to this problem would

TABLE 11. Effect of Solutes on Vapor Pressure Isotopic Ratio of Tritiated Water

Compound	Concentration	°C	$\alpha(H/T) \pm 0.03$
H ₂ O (a)	--	~25	1.09
KNO ₂	3M	~50	1.08
KI	3M	~50	1.06
Na ₂ Cr ₂ O ₇ (pH8.2)	3M	~50	1.04
Na ₂ Cr ₂ O ₇ (pH4)	3M	~50	1.06
AlCl ₃	3M	~50	1.04
LaNO ₃	3M	35	1.05
LaNO ₃	3M	80	1.05
CsCl	3M	35	1.10
NaF	2M	40	1.09
(CH ₃ C ₂ H ₄) ₄ NOH	10%	40	1.09
HO(CH ₂) ₅ OH	~6:1 W/Org	40	1.05
(HOC ₂ H ₄ OC ₂ H ₄) ₂ O	~3:1 W/Org	35	1.05
HOC ₂ H ₄ OH	1.5:1 W/Org	35	1.05
NaNO ₂	3M	50	1.11
NaNO ₂	6M	~50	1.08
NaOH	6M	~50	1.04
NaClO ₄	3M	~50	1.08
LiCl	3M	~50	1.09
CsOH	3M	40	1.06
(C ₄ H ₉) ₄ NBr	1M	40	1.08
(C ₄ H ₉)NCI	1M	35	1.10
H ₂ O (b)	--	35	1.08
NaNO ₂ (b)	3M	35	1.07

a. Van Hook's values for pure water are 1.095 at 25°, 1.065 at 50°, and 1.030 at 100°C. (13)

b. Using deuterated H₂O, 3%, in place of HTO.

be to react tritiated water or tritium gas with a monomeric material which can be readily polymerized. One obvious approach is the reaction of water with calcium carbide to form ditritiated acetylene:

Polymerization reactions of acetylene can then be initiated to "fix" tritium in a relatively permanent form.

Brookhaven National Laboratories have explored this possibility using gamma radiation as the polymerizing technique: (18)

Crosslinkages would give a three dimensional saturated product.

We have considered the reaction in which acetylene is hydrated using a

sulfuric acid solution of mercuric sulfate as the reaction medium. The resultant product is acetaldehyde, which in the extreme situation could contain as many as four tritium atoms.

or normal acetylene could be reacted with tritiated water to give:

When acetaldehyde is reacted with phenol, a product is formed that is analogous to Bakelite (phenol-formaldehyde polymer).

Production of tritiated acetylene requires calcination of the calcium hydroxide and recycle of the tritiated water. Thus it appears that manufactured acetylene could more efficiently be substituted in the process of making tritiated acetaldehyde. The lowered tritium content of the polymer may be more than compensated for by elimination of the calcium carbide step.

Hydration of Acetylene to Acetaldehyde

The hydration of acetylene to acetaldehyde has already been developed as a feasible manufacturing scheme by the Germans during World War II. The reported conversion of acetylene per

pass is about 55%, and the yield of acetaldehyde is 93 to 95% of theoretical.⁽¹⁹⁾ The product is recovered as a 7% aqueous solution of acetaldehyde which can be purified by distillation of 99.9% purity. Byproducts are acetone, diacetyl, crotonaldehyde and acetic acid.

Our laboratory experiments gave a conversion of only a few percent. This low yield probably resulted from insufficient mixing of the heterogeneous solution, resulting in poor contact time between acetylene and the mercuric sulfate catalyst. Some equipment modifications will be made so that yields may be increased, but because satisfactory industrial scale synthesis of acetaldehyde has already been reported, only a limited effort will be made in this direction. Product identification was made by preparation of the 2,4-dinitrophenylhydrazone derivative and by comparison of the infrared spectrum with that of an authentic sample of acetaldehyde. To date, the synthesis of acetaldehyde has been confined to water or deuterated water. Preparations are now under way for the synthesis of tritiated acetaldehyde.

Preparation of the Acetaldehyde-Phenol Resin

Initially attempts were made to prepare the polymer using the same procedure as though the reactants were formaldehyde and phenol rather than acetaldehyde and phenol. Although eventually a tough, water insoluble polymer was formed, considerable time was required for the curing, and significant losses of materials resulted. A modified procedure was

adopted which results in a polymer with the desired properties within three or four hours. It was found that substitution of resorcinol for phenol accelerates the reaction rate significantly. Even so, unless the molar concentration of acetaldehyde relative to the resorcinol is reduced from about 2.2 to 1, the uptake of acetaldehyde becomes slower as the reaction proceeds, until finally a fixed quantity of acetaldehyde continues to reflux at the cold finger. When the reduction of acetaldehyde indicated above was made, the reaction proceeded more completely so that after a reaction time of about an hour only negligible refluxing was observed at the cold finger. A mole of formaldehyde added to the reaction mixture at this point causes an instantaneous solidification of the polymer. The polymer is cured by heating it to 80°C under a vacuum of about 10 to 15 Torr. The off-gases can be collected in a dry ice-acetone trap and examined by gas chromatography. Acetaldehyde recovered from the cold trap amounts to about 3 to 4%. The product is hard, brittle, and water insoluble.

Other Polymers

Urea-acetaldehyde polymers have been briefly examined as a possible candidate for fixing tritium. Since the product formed showed appreciable water solubility, no further investigations were made.

Investigations will be made of the possibility of preparing other monomers from acetylene. It is possible, for example, to convert acetylene to

ethylene by suitable hydrogenation conditions. Thus, a very simple monomer could be prepared which would contain tritium.⁽²⁰⁾

The tritiated ethylene could then be converted to polyethylene.⁽²¹⁾

Another monomer that appears suitable for tritium fixation is acrylonitrile. German and American patents⁽²²⁾ have been filed on processes of reacting acetylene with hydrocyanic acid in either the vapor phase or the liquid phase to the desired material.

Acrylonitrile can be readily converted to a polymer using anionic polymerization techniques.⁽²³⁾

SEPARATION OF TRITIUM ISOTOPES USING LASER-INDUCED MOLECULAR EXCITATION AND REACTION - J. R. Morrey and L. L. Burger

Previous reports have discussed the possible schemes for tritium separation involving laser excitation. Initial calculations were directed at multivibrational excitation within the ground electronic state followed

by reaction with an added molecular species. There seems little doubt that this can be a highly efficient use of laser energy, and calculations show that combinations exist which permit pumping a molecule to a high vibrational level with a single frequency. Figure 19 schematically illustrates the ways in which H_2O may be excited to high vibrational states, using one or two frequencies of the laser. The reader is cautioned that detailed molecular spectra for excited states are not available at the precision necessary to pick out the exact frequencies required and that a basic research program is required to obtain them.

FIGURE 19. Schemes for Exciting H_2O

However, the figure serves as an illustration of possible techniques for excitation. In A, two laser frequencies are used: the first to pump to the 100 level, the second to pump from that to the 120 level, and then the first, again, to pump from the

120 level to the 032 level. This scheme requires some sophistication and synchronization of the laser pulses. In B, only one laser frequency is used, but associated with the pumping to the 021 vibrational level must be a vibrational relaxation to the 120 level which will decrease the efficiency of the overall process. Schemes C and D are more desirable in that no molecular relaxation processes would be needed to complete the pumping scheme. Notice that pumping requires vibrational combination transitions, most of which have very low conventional absorption cross-sections. These are offset by the fact that the true cross-section generally can be expected to be a hundred-fold stronger than that traditionally measured by spectrophotometry, and, too, by the very much increased intensity of a laser over any other light source.

Because the detailed molecular spectra for excited states required in the above described scheme are unavailable, our initial experiments involve another less demanding spectroscopic scheme to excite HDO to the vibrational 110 level, followed by excitation to the first excited unstable electronic state. This scheme is illustrated in Figure 20. The absorption coefficient of water vapor in this unstable state is given in Figure 21.⁽²⁴⁾ Free radicals

$H + DO + D + HO$ are expected to be generated and these radicals will be gettered by a free radical scavenger. The source for the vibrational excitation will be a Chromatix laser. The schematic of the experiment is shown in Figure 22. The flash lamp around

FIGURE 20. Electronic Excitation Scheme for HDO

FIGURE 21. Absorption Coefficient of H_2O Vapor

FIGURE 22. Experimental Arrangement

the reaction cell is not shown. Although the tunable laser, which is tuned by changing the temperature of the parametric oscillator, is very low power, our calculations tell us that sufficient pumping will be possible to induce reactions sufficient to detect separation.

The flash lamps and control units for the experiment have been identified but cannot yet be purchased until capital money is released, presumably in October. The

Chromatix laser and other accessory gear is in place.

The major technical problems associated with this experiment are expected to be wavelength filtering of the flash lamp, low laser energy, separation of reaction products, and detection of the low yields. The first experiments will involve $\text{HDO}/\text{H}_2\text{O}$ separation, and, if successful or encouraging, will be followed by separation of HTO which will require additional spectral data.

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6.0 CHARACTERIZATION OF 300 AREA AND WYE BURIAL GROUNDS

J. R. Eliason - Project Manager

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

SUMMARY

Funding for the 300 Area and Wye burial grounds study was authorized during August. The original 189 is being evaluated, and several changes are being made at the request of the AEC. This study will include a preliminary review of all 300 Area burial ground. Based on this review, critical areas will be identified and a program initiated to conduct studies directed toward these problems during FY75.

Principal investigators for each major task were identified and are developing their detailed task work plans.

Preliminary estimates of funding have been outlined for FY75 for each major task, which may have to be modified as task work plans are evaluated and formalized.

A search for disposal records, documents and maps identifying and delineating the disposal sites was begun, and historical records are being summarized.

TECHNICAL PROGRESS

The following tasks have been identified:

- Review of historical records
- Waste detection and mapping
- Sampling and analyses of sediments
- Instrument wells and characterization of sediments
- Evaluation of moisture movement mechanisms and modeling
- Evaluation of biological transport mechanisms
- Evaluation and reporting

TASK DESCRIPTIONS

- Review of Historical Records

Principal Investigators:

A. E. Reisenauer and K. L. Kipp

This review has been initiated, and preliminary descriptions of the burial grounds are now being compiled and will be provided to all principal investigators. These preliminary data will provide the information required to develop a detailed task work plan for FY75 and subsequent fiscal years.

A complete analysis of historical records including reporting of historical radionuclide and materials inventories for each burial site is scheduled to be completed in FY75.

- Waste Detection and Mapping

Principal Investigator:

G. A. Sandness

The detection and mapping program will require conducting a comprehensive evaluation of geophysical techniques to design the proper mapping program based on the complex character of the waste materials. Instrumentation is to be acquired and preliminary field mapping conducted during FY75. The detailed task work plan now being prepared should provide a better estimate of time and cost for this task.

- Sampling and Analyses of Sediments

Principal Investigators:

V. L. McGhan and L. L. Ames

Samples of sediments are to be collected from the burial sites to determine the present contamination patterns and migration rates. These studies will include designing appropriate drilling and sampling techniques, designing optimum sampling patterns for each site, analyzing the samples, mapping contamination patterns, and calculating migration rates. During FY75 these studies will be limited to design and testing of techniques. The detailed task work plan will provide estimates of time and cost for this task in subsequent fiscal years.

- Instrument Wells and Characterization of Sediments

Principal Investigator:

S. J. Phillips

Soils will be characterized, and

instruments will be installed for measuring soil moisture movement both for field and laboratory testing.

These studies will be limited during FY75 to planning, design, and field testing of instrumentation. The detailed task work plan now being prepared will provide an initial estimate of time and costs.

- Evaluation of Moisture

Movement Mechanisms and Modeling

Principal Investigator:

H. P. Foote

Identification and theoretical evaluation of soil moisture transport mechanisms will be conducted. Models are to be developed and verified with field and laboratory testing. These models will be used to predict the effects of long-term environmental changes on waste migration rates. The detailed task work plan now being prepared will provide an initial estimate of time and costs.

- Evaluation of Biological

Transport Mechanisms

Principal Investigator:

W. H. Rickard

Preliminary investigations of each site including identification of present plant and animal life and analyses of selected samples will be conducted during FY75. This program will include detailed evaluation of the biological transport which may develop with long-term environmental changes. The detailed task work plan now being prepared will provide an estimate of time and costs.

- Evaluation and Reporting

Principal Investigators:

A. E. Reisenauer and J. R. Eliason

Reporting required by the program will be prepared, and data provided will be analyzed.

Detailed task work plans are now being finalized, and initial field studies are under way. Each site is being photographed, surface samples of vegetation collected, and surface dose rates measured.

Historical records that have been located are being summarized for each site to provide data needed for a preliminary review. These data and preliminary field and photographic reconnaissance of each site are being collected.

An initial review of historical data and preliminary field studies is now being conducted. This review is designed to identify sites which may be analyzed during this fiscal year.

1. The first part of the document discusses the importance of maintaining accurate records of all transactions. It emphasizes that proper record-keeping is essential for the integrity of the financial system and for the ability to detect and prevent fraud.

2. The second part of the document outlines the specific procedures for recording transactions. It details the steps involved in the accounting cycle, from identifying the transaction to posting it to the appropriate ledger account.

3. The third part of the document discusses the role of the auditor in verifying the accuracy of the records. It describes the various techniques used by auditors to test the reliability of the accounting system and to ensure that the financial statements are true and fair.

4. The fourth part of the document discusses the importance of internal controls in preventing errors and fraud. It describes the various types of controls that can be implemented, such as segregation of duties and the use of physical safeguards.

5. The fifth part of the document discusses the importance of the audit trail in providing evidence for the audit. It describes the various types of evidence that can be obtained, such as documents and physical assets, and the importance of maintaining a clear and complete trail of all transactions.

7.0 DISPOSITION OF RETIRED, CONTAMINATED (FUTURE WMT) FACILITIES AT HANFORD

K. M. Harmon - Project Manager

The program has the following objectives for FY75-76: to establish methods, costs and priorities for the disposition of retired (excessed) Hanford facilities which will be transferred to WMT; to formulate plans and specifications for a project to accomplish permanent disposal of a specific Hanford facility; and to identify required facility disposition R&D.

SUMMARY

The program has been revised, a work plan prepared, and six retired Hanford facilities have been selected as candidates for demonstration disposition projects. Evaluation of the six candidates was started to permit ranking them for the first project.

A "disposition resource book" was planned to summarize the information collected in pursuit of the first program objective (establish methods, costs and priorities...). Published information related to retired liquid radioactive waste disposal sites in the 200 Areas was collected, and preparation of facility summaries on these sites was initiated for incorporation in the appendix of the resource book.

A literature search relating to the disposition of contaminated nuclear facilities was completed and work begun on setting up an information center on decontaminating, decommissioning and disposing of nuclear facilities.

TECHNICAL PROGRESS

TASK 1: PLANNING AND EVALUATION

K. M. Harmon

A work plan for FY75-76 was prepared based on the following activities:

1. Preparation of a loose-leaf, easily updated "Disposition Resource Book." The resource book is to summarize the results of BNW studies of methods, costs and priorities for the decontamination and dismantling (D&D) of different categories of retired Hanford facilities to alternative levels of residual contamination: for unrestricted use; for conditional use for non-nuclear activities; and for nuclear activities. The alternative in which the site or facility is simply to be sealed in some way and kept under surveillance is also to be considered. The resource book is also to include an appendix which will provide a brief disposition history for each facility.

2. Preparation of detailed plans, including procedures and specifications, environmental assessment and

safety studies, and cost estimates for the disposition of a specific facility.

3. Identification of new technology needed for disposition activities. An overall schedule is shown in Figure 23.

In work on selecting a facility for the first demonstration project, meetings with representatives of the other Hanford contractors have led to selection of the following as representatives of the various types of Hanford facilities ultimately requiring disposition action:

1. 107H, a reactor cooling water retention basin.
2. 105H, an old production reactor.

3. 233-S, the plant used for concentration of plutonium product from the Redox Plant.

4. 201-C, the pilot plant used for hot demonstration of the Purex process and for strontium processing.

5. 216-T-6, a small 200 Area crib used for disposal of sump waste from one of the bismuth phosphate separations plants.

6. 300-Wye burial ground, used for burial of laboratory wastes. Using the criteria that the first demonstration project should have a high priority for disposition, should be relatively simple, should offer the chance to learn much, and should be ready for actual operations to start in FY76, our present ranking is as

	FY 1975					FY 1976				
	J-S	O-D	J-M	A-J		J-S	O-D	J-M	A-J	J-S
PREPARE PROJECT PLAN - DEMO A	1	2	3,4	5,6	7	8	9	10		
ESTABLISH METHODS, COSTS, PRIORITIES FOR WMT FACILITIES AT HANFORD			3	2,4	11		3*	2*	4*	11*
IDENTIFY R&D NEEDED	12	12*				12*	12*			

MILESTONES:

1. SELECT FACILITY
2. SELECT PREFERRED DISPOSITION STRATEGY
3. DEFINE DISPOSITION CRITERIA
4. SELECT PREFERRED D&D PROCEDURES
5. COMPLETE SITE CHARACTERIZATION
6. ISSUE PROJECT PLAN (SCOPING DOCUMENT)

* DENOTES UPDATE OF PRELIMINARY DOCUMENT

7. ISSUE ACTIVITY SPECIFICATIONS (INCLUDING COST ESTIMATES)
8. ISSUE ENVIRONMENTAL ASSESSMENT
9. TRANSMIT TECHNICAL SPECIFICATIONS TO FACILITY LANDLORD FOR IMPLEMENTATION
10. START PROJECT OPERATIONS
11. ISSUE DISPOSITION RESOURCE BOOK
12. ISSUE "R&D NEEDS" DOCUMENT

FIGURE 23. Disposition of Retired Contaminated Facilities at Hanford

follows: 1. 233-S; 2. 216-T-6; 3. 107H; and 4-6. 201-C, 105H and 300 Wye.

TASK 2: SITE CHARACTERIZATION

B. W. Mercer

Published information related to retired liquid radioactive waste disposal sites in the 200 Areas was collected, and preparation of facility summaries on these sites was initiated for incorporation in the appendix of the resource book. The facility summaries will contain information pertinent to the disposition of the facilities, including location, description, site characterization, incentives for disposition, disposition alternatives, and disposition history. The completed summaries will document the comprehensive information required for disposition of these retired facilities.

In an effort supporting the selection of a site for the first demonstration project, three liquid disposal cribs in the 200 West Area were identified as potential candidates for demonstrating decontamination and decommissioning techniques for a subsurface soil zone contaminated with actinides and fission products sorbed from aqueous radioactive wastewaters. The three cribs selected for investigation were 216-S-1 and 2, 216-S-7, and 216-T-6. These cribs are potentially hazardous to personnel in the area due to possible cave-ins resulting from failure of timbered structures supporting the soil over the liquid discharge caverns.

However, the 216-S-1 and 2 cribs and the 216-S-7 crib were eliminated from consideration as demonstration

sites because of the large amount of radioactivity discharged to these cribs and the deep penetration of radioactivity into the soil beneath the cribs. A field evaluation of the 216-S-1 and 2 cribs in 1966 (ISO-SA-62) indicated significant penetration of strontium and cesium below the 100-ft level. A gamma scintillation probe of a well near the 216-S-7 crib also showed significant penetration of gross gamma activity below the 100-ft level. It was concluded that any attempt to remove or immobilize a large fraction of the contaminated soil beneath these cribs would entail considerable costs.

The 216-T-6 crib was selected as the best candidate of the three because of the smaller quantity of radioactivity disposed to this crib and the small depth of penetration of the activity. Gamma scintillation probes of wells around this crib show activity only to the 20-ft level. If this activity is assumed to be ^{137}Cs , approximately 80,000 cu ft of soil would be contaminated with this isotope. It was conservatively estimated that ^{90}Sr penetration down the soil columns might be three times, and the lateral dispersion twice, that of the ^{137}Cs . This yielded a very rough estimate of 10^6 cu ft of contaminated soil, which would also require a large expenditure of funds for excavation or immobilization, but much less than that for 216-S-1 and 2 or 216-S-7. The 216-T-6 crib also has 14 monitoring wells around the crib that could be used for further characterization work utilizing improved well monitoring devices.

Excavation or immobilization of a large fraction of contaminated soil beneath a crib represents a decontamination and decommissioning activity that might allow either unrestricted or minimal restricted use of the land at the crib site. As an alternative to this approach, excavation of only the top layer of soil (containing sorbed actinides) could be accomplished at a much lower cost. This would alleviate the long-term hazards of leaving an actinide such as ^{239}Pu in the soil. The cost of site characterization might be limited to that involving only one core sample taken from the center or low spot of the crib.

TASK 3: PROCEDURES AND TECHNIQUES

H. T. Fullman and M. S. Hansen

Work was initiated on setting up an information center on disposition of nuclear facilities. The literature search has been completed and all pertinent references have been ordered. To date, approximately 450 references have been ordered and about half have been received. We are now in the process of cataloging the references and setting up a file system. The various references will be indexed by report number, author and key words using a computer print-out.

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

C. L. Brown - Project Manager

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

The objectives of this program are:

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

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

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

SUMMARY

The compounds $\text{Pu}(\text{NO}_3)_4 \cdot 3\text{H}_2\text{O}$, $\text{K}_2\text{Pu}(\text{NO}_3)_6$, and $(\text{NH}_4)_2\text{Pu}(\text{NO}_3)_6$ have all been prepared using 85% ^{238}Pu and vacuum evaporation techniques. The salts were then used to determine gas-generation rates and the composition of the gases formed as a result of radiolytic decomposition.

Thermal gravimetric analysis was also performed on each of the three salts to determine thermal stability and formation of intermediates before final conversion to plutonium oxide.

The compound $(\text{NH}_4)_2\text{Pu}(\text{NO}_3)_6$ has been eliminated from consideration as a shipping form because of excess gas evolution, and the compound $\text{K}_2\text{Pu}(\text{NO}_3)_6$ may be because of the intro-

duction of potassium into a process stream.

TECHNICAL PROGRESS

SOLID PLUTONIUM SHIPPING FORMS

G. H. Bryan, Principal Investigator

Radiolytic gas-generation studies of $\text{Pu}(\text{NO}_3)_4 \cdot 3\text{H}_2\text{O}$, $\text{K}_2\text{Pu}(\text{NO}_3)_6$, and $(\text{NH}_4)_2\text{Pu}(\text{NO}_3)_6$ have been completed. Results indicate the compound $\text{K}_2\text{Pu}(\text{NO}_3)_6$ evolves the least amount of gas per unit weight of Pu with a maximum G_T value of 0.18, $\text{Pu}(\text{NO}_3)_4 \cdot 3\text{H}_2\text{O}$ evolves slightly more gas with a maximum G_T value of 0.19, and the compound $(\text{NH}_4)_2\text{Pu}(\text{NO}_3)_6$ evolves about four times as much gas as the other two salts with a maximum G_T value of 1.0. The ammonium salt therefore has been eliminated from consideration as

a shipping form. (See Figure 24 for G_T values for each compound as a function of accumulated alpha dose.)

FIGURE 24. G Values as a Function of Accumulated Radiation Dose

The composition of the radiolytic decomposition gases evolved from each compound was determined by gas mass-spectrometer analysis and are shown

in Table 12 as a function of accumulated alpha dose for each of the three compounds under consideration.

Each of the three salts was prepared for this study by vacuum evaporation techniques (using a slight stoichiometric excess of potassium or ammonium nitrate in the case of $K_2Pu(NO_3)_6$ and $(NH_4)_2Pu(NO_3)_6$ from plutonium which was 85% ^{238}Pu to accelerate the acquisition of experimental results).

Thermogravimetric analysis of each of the three salts indicates $Pu(NO_3)_4 \cdot 3H_2O$ and $K_2Pu(NO_3)_6$ have intermediates formed before final conversion to the oxide (Figures 25 and 26). The compound $(NH_4)_2Pu(NO_3)_6$ does not appear to have any intermediates (Figure 27). Each of the compounds used for this experiment was prepared in the same manner as described for the gas evolution study; however, ^{239}Pu (<1% ^{238}Pu) was used for purposes of radiolytic stability. $K_2Pu(NO_3)_6$ would appear to have a slight advantage from a radiolytic-gas evolution standpoint.

TABLE 12. Composition of Radiolytic Gases Evolved From $Pu(NO_3)_4 \cdot 3H_2O$, $(NH_4)_2Pu(NO_3)_6$, and $K_2Pu(NO_3)_6$ as a Function of Accumulated Alpha Dose (a)

		Dose					
		1 Mev x $10^{17}/g$ Pu			3 Mev x $10^{17}/g$ Pu		
		$Pu(NO_3)_4 \cdot 3H_2O$	$(NH_4)_2Pu(NO_3)_6$	$K_2Pu(NO_3)_6$	$Pu(NO_3)_4 \cdot 3H_2O$	$(NH_4)_2Pu(NO_3)_6$	$K_2Pu(NO_3)_6$
O_2	%	86	0.1	24	85	0.1	28
N_2	%	8	66	6	5	66	7
H_2	%	0.4	0.5	---	0.5	0.3	---
NO_x	%	4	18	55	7	23	53
N_2O	%	---	14	14	---	9	6
CO_2	%	3	---	---	3	1	6

a. Typical reactor grade plutonium might undergo a dose of about 1.3×10^{17} Mev/g per month whereas the dose from the material used to compile this table was 7.8×10^{18} Mev/g per month

But it may be eliminated from consideration as a shipping form because of the introduction of an extraneous

cation (K) into a process stream upon dissolution in nitric acid, thereby creating waste disposal problems.

FIGURE 25. Thermogravimetric Curve for Plutonium Nitrate

FIGURE 26. Thermogravimetric Curve for Ammonium Hexanitratoplutonate

FIGURE 27. Thermogravimetric Curve for Potassium Hexanitratoplutonate

9.0 TRANSPORTATION SAFETY STUDIES

L. D. Williams - Project Manager

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

SUMMARY

Risk, as used in this study, is the product of the probability and the consequences of an accident. To date, essentially all effort has been in analyzing the risk of shipping plutonium either as the liquid nitrate or the solid oxide powder. Concentrating on plutonium shipments has permitted completion of an evaluation, thus demonstrating the workability of the methodology. For the analysis, the model (still under development) has necessarily been computerized. This means that subsequent analyses can build on the methodology created for plutonium shipments. The preliminary findings of the plutonium shipping risk study are presently available as a preliminary draft,⁽¹⁾ planned to be finalized as a report by February 1975.

TECHNICAL PROGRESSRisk Model Development

The risk model being developed has been summarized in a paper submitted at the Fourth International Symposium on Packaging and Transportation of Radioactive Materials.⁽²⁾

In the computerized model, the code, named HEAD, starts with a fault tree showing the various ways the barriers preventing release can be

breached either by accidental forces or improper packaging. Failure probabilities and the fraction of material which will pass through the barrier upon failure are input into the code. The code then forms a list of failure sequences, each one of which will release material to the environment.

The code next examines the failure sequence under various conditions of weather and population. It ranks the sequences according to their risk level. The total risk for a shipment of material is given and the number of people affected.

The code presents the user with a list of the failure events which cause the greatest risk. In addition, the code can calculate either the risk associated with transport through a specific region of the country or can consider the risk for shipments through several regions.

The code has been used with success for plutonium associated shipments and now will be expanded to evaluate the risks of other radioactive material and toxic chemicals.

Risk of Shipping Plutonium

Sponsored by the Transportation Branch of the Atomic Energy Commission's Division of Waste Management and Transportation, Battelle, Pacific

Northwest Laboratories conducted a research and development program to investigate the safety considerations of shipping plutonium by truck. The objective of this study was to develop a methodology for determining the risk of transporting radioactive material and to apply it to current and future plutonium shipping practices. The results, documented in a preliminary draft, have been distributed to the Commission for comment.

This report describes a risk assessment of shipping plutonium oxide and liquid plutonium nitrate by truck. Experience has shown that current plutonium shipping practices are good. There has been no detected release of plutonium to the environment during shipment. Our study indicated that even with the increased flow of plutonium across the U.S. in 1980, the risks of plutonium shipment are small relative to other societal risks.

The essence of this study was prepared for the Risk Assessment II Session of the Fourth International Symposium on Packaging and Transportation of Radioactive Material in Miami Beach, Florida, September 22-27, 1974.

The study results have been related to a future point--the year 1980--when plutonium shipments are expected to be more frequent. To conduct this risk assessment, certain assumptions had to be made about the nuclear economy and the transportation system in 1980. The assumptions used for the comparison are:

- A total of 13 metric tons of plutonium is shipped by truck (exclusive carrier).

- Shipping systems and regulations are the same as 1974.
- PuO_2 is shipped in 6M containers and liquid $\text{Pu}(\text{NO}_3)_4$ in L-10 containers.
- Shipments are across central and southeastern U.S.
- No evacuation of people occurs following accidental releases.

The relative risk of the two shipping forms of plutonium are not influenced by the assumptions on plutonium flow and shipping route.

The results of the risk assessment are graphically shown in Figure 28. There they are compared to the risk of operating 100 nuclear reactors and to other societal events taken from the Rasmussen Study (WASH-1400 Draft). It can be seen that shipment of liquid $\text{Pu}(\text{NO}_3)_4$ for the most part entails less risk than the 100 nuclear reactors and that PuO_2 shipments have about three orders of magnitude less risk than nitrate shipments. A curve has been added to indicate the importance of liquid $\text{Pu}(\text{NO}_3)_4$ releases during a fire when the thermal insulation in the L-10 containers has not been compromised. The apparent conclusion is that if the integrity of the thermal insulation can be assured and maintained during accident conditions, then the lower curve will better represent the risk of shipping liquid $\text{Pu}(\text{NO}_3)_4$.

An interesting comparison can be made using the Rasmussen 100 reactor curves and the liquid $\text{Pu}(\text{NO}_3)_4$ curve: their risk spectrum is almost identical to that of getting killed by meteors.

FIGURE 28. Risk Spectrum of Plutonium Shipments in 1980

To permit comparisons such as those made on the following graph, the estimated population doses for the postulated release sequences have been related to cancer deaths exceeding those normally occurring. These conversion factors have been derived from the BEIR Report.⁽³⁾ It is worthwhile noting that in the 30 years since plutonium was first produced no evidence has conclusively linked or even suggested cancer development in humans exposed to plutonium. This evidence involves hundreds of workers in both the Manhattan Project and the Atomic Energy Program. It includes some 26 young men who were exposed in their work during World War II to plutonium in sufficiently high air concentrations that the most recent

estimates of their body burdens range from 1/10 to 10 times the present maximum permissible body burden of 0.04 μCi as recommended by the ICRP.

Risk Evaluations of Shipping Other Materials

Attention is being directed at evaluating the risk of shipping other materials. A major effort is now being made to summarize relevant information which can be used as a basis for assessment of UF_6 , fresh fuel, spent fuel, and waste shipments. This summary, nearly complete at this time, will appear in the next quarterly report.

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