LAWRENCE
LIVERMORE
LABORATORY
University of California/Livermore

NUREG/CR-0538
UCRL-52547

WASTE MANAGEMENT TECHNICAL SUPPORT PROJECT
FY 78 END-OF-YEAR REPORT

R. A. Heckman
T. R. Donich

133

Prepared for
U. S. Nuclear Regulatory Commission
by
Lawrence Livermore Laboratory

542 207

7907250299

NOTICE

Available from National Technical Information Service Springfield, Virginia 22161

542 208

NUREG/CR-0538
UCRL-52547

WASTE MANAGEMENT TECHNICAL SUPPORT PROJECT
FY 78 END-OF-YEAR REPORT

R. A. Heckman
T. R. Donich

Manuscript Submitted: December 1978
Date Published: July 1979

Prepared for
Office of Nuclear Material Safety
and Safeguards
U.S. Nuclear Regulatory Commission
Washington, D. C. 20555
Under Interagency Agreement DOE 40-550-75
NRC FIN No. A 0277

by
Lawrence Livermore Laboratory
Livermore, CA 94550

operated by University of California

for the U.S. Department of Energy

512 209

FOREWORD

This report covers the progress during FY 78 by the Waste Management Technical Support Project of the Lawrence Livermore Laboratory in developing a technical base for the Nuclear Regulatory Commission (NRC) to use in formulating guides and regulations for the disposal of nuclear waste in suitable repositories.

This report is intended as an end-of-year progress report as required by SOW FIN A-0277-8.

Any analyses or conclusions contained herein are to be regarded as preliminary and do not reflect NRC positions.

TABLE OF CONTENTS

512 211

512 212

542 213

542 214

LIST OF ILLUSTRATIONS

542 215

542 216

LIST OF TABLES

512 217

512 218

ABSTRACT

This report covers the progress made during FY 78 in the various tasks of the Waste Management Technical Support Project which the Lawrence Livermore Laboratory is conducting for the Nuclear Regulatory Commission (NRC). The project's goal is to develop a technical base to be used by the NRC in developing regulations and regulatory guides for the disposal of radioactive waste in deep geologic repositories. The report covers primarily the ongoing work of Task I: Waste Form Performance and Task II: Site Suitability. Results from these and other tasks are summarized and referenced. All analyses and conclusions are preliminary and do not reflect NRC positions.

542 219

INTRODUCTION

The Lawrence Livermore Laboratory is under contract to the Nuclear Regulatory Commission, Office of Nuclear Material Safety and Safeguards, to provide technical support for the development of regulations and regulatory guides for the disposal of high-level and transuranic wastes in deep geologic repositories. This report summarizes the technical accomplishments during FY 78.

REPORT ORGANIZATION

The report is organized along the lines of the major technical support areas or tasks, as specified in FIN A-0277-8. Task I is concerned with waste performance, Task II with site suitability, Task III with certain thermal considerations for repository design performance, and Task VI with radiological performance objectives. Task VII develops a geoscience data handbook, and Tasks IV, V, and VIII were not funded during FY 78.

The body of the report provides a summary of the important technical issues studied during FY 78 in Tasks I, II, and VI, and only these tasks are discussed in this introduction. To provide the reader with more detailed discussions, appendices on selected topics are included. Detailed technical discussions can also be found through references to original technical reports shown in the reference section. Similarly, references are given at the end of each appendix.

PROJECT OVERVIEW

A hierarchical objective diagram is useful in defining our systems analysis approach to the overall nuclear waste management system problem. This approach is shown in Fig. I1. Consideration of the objectives for the national waste management program through the Federal agency implementation allows us to structure the overall problem into two basic sets of models:

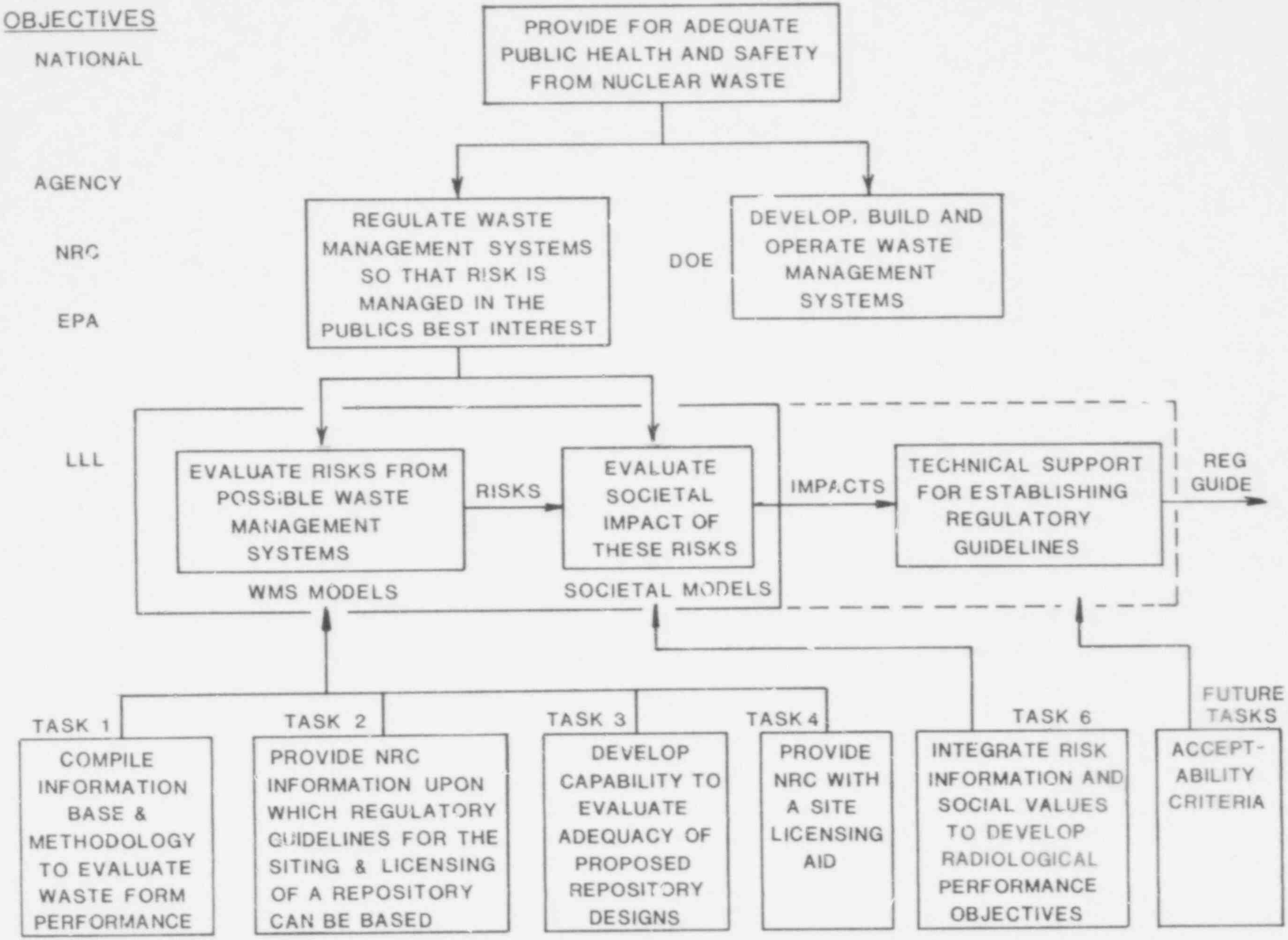

FIG. I1. Overall waste management systems problem.

542 221

waste management system (WMS) models and societal models. The individual tasks develop more detailed models. Specific risks are derived from the WMS models and the evaluation of the societal impact of these risks is made possible through the societal models developed by the radiological performance objectives task. These impacts will be used in future technical support work for establishing regulatory guidelines.

PROJECT REDIRECTION

A major redirection of our effort occurred during FY 78 as a result of the Presidential decision to defer reprocessing of spent fuel. Work on solidified high-level nuclear waste was suspended during the second quarter of FY 78. The storage/disposal of spent fuel in geologic media was emphasized during the remainder of the year.

TASK I

In Task I, three general areas of pre-emplacement radiological risk for solidified high-level waste were studied. Human factors in accidents were investigated at the reprocessing plant, during transportation between reprocessing plant and repository site, and at the repository. Completion of the numerical data needed to quantitatively evaluate the impact of human error on expected radiological dose was not completed due to program redirection, so the analysis to date summarizes the qualitative impacts of human errors on expected doses. Interim storage at the reprocessing plant included two areas: loss of coolant accidents involving loss of coolant circulation and, more importantly, loss of coolant by pool drainage in the event of pool rupture by severe earthquake. Pool drainage accidents can lead to significant radiological releases for closely-spaced storage racks. The seismic analysis reached only a preliminary stage, but produced an outline for a methodology for evaluating seismic risk from a severe earthquake to an interim storage pool.

For the management of spent fuel, work on a number of technical areas was initiated. Potential hazards from the radionuclides in spent fuel as a function of age were calculated. Note was taken of the gaseous radionuclide content of spent fuel, activity that is not present in solidified high-level waste. A three-dimensional thermal analysis was conducted for disposal of spent fuel in a repository. This analysis provided an evaluation of the near-field thermal environment, an important variable in other repository studies. The most sensitive parameter was the spent fuel heat generation rate.

Studies on design concepts for retrievable packages have produced a reference package design, identified the most important failure modes that will require studies to ensure integrity, and allowed development of a risk methodology for container structural integrity.

Work was started on the development of a model to identify and evaluate parameters affecting spent-fuel dissolution, and radionuclide release rate and migration. Preliminary results show significant ground-water effects on solubility for different radionuclides.

Initial work on a survey of corrosion data on candidate container designs in a halite brine solution has identified a number of potential useful materials. A testing program will be necessary to confirm the suitability of these materials.

Modeling studies on brine migration are underway. Results to-date suggest that as much as 40 l of brine could accumulate around each canister during a 10 y period. Thermomechanical effects of this brine accumulation are being modeled. These effects include accelerated creep due to possible pressure solutioning and coupling between the salt and the canister.

The major components of the transuranic (TRU) waste management program include (1) the identification of the inventory and sources of transuranic waste, (2) the development of a functional definition for this waste, (3) the identification of the waste form and package, and (4) the evaluation of the resultant waste management package in a deep geologic waste repository with emphasis on developing radioactivity release functions. Significant progress was made in (1) and (2) with reports being issued. Work in (3) will define both current and future technology with emphasis on final TRU waste characteristics. A methodology was developed for (4), but at present we have only incomplete information.

542 224

TASK II

Fy) saw a significant change in the long-term objective and goals. Whereas earlier work was directed toward regulations, these changes reflect the need to obtain information and tools needed for regulatory guide development. In particular, a licensee in preparation for obtaining a license for a deep geolcgic repository site requires:

- A checklist of qualitative criteria for each host geology
- A list of possible measurements and their uncertainties
- A knowledge of the licensing tools and the methods of translating these measurements into a prediction of site performance.

During FY 78 emphasis was on site characteristics and modeling. Specifically, other site media were investigated, future efforts were scoped, and geophysical measurement uncertainties were analyzed. The modeling effort involved development, revision, and improvement of models. The new goals suggested that the modeling work should first focus on individual barriers to waste migration.

SITE CHARACTERISTICS

Work on site characteristics focused on three general areas: qualitative description, geological/geophysical data, and geological/geophysical measurement techniques and associated errors.

Qualitative description

Simple models were developed to incorporate neccesary assumptions about repository design and operation and to identify the probable ranges in design parameters that might affect long-term repository behavior. Generic settings for repositories sited in basalt, granite, and salt domes were developed. The feasibility of modeling waste repositories located above deep-seated water tables (vadose zone) is being investigated. The characteristics of unsaturated regimes present modeling requirements that are different from those developed for saturated flow conditions. Potential sites in unsaturated

512 225

media may be found at the Nevada Test Site, the Snake River Plain of Idaho, the Columbia Plateau of Washington, and the Colorado Plateau of Arizona.

Geological/geophysical data

This area of investigation encompasses the following:

- Retardation factors used in the mass-transport model.
- The geochemical data base.
- Physical and chemical processes that affect porosity.
- A chemical equilibrium code to define solubility limits for future use in the mass-transport model.
- Sorption characteristics of granites and basalts.

Geological/geophysical Measurements

We investigated the applicability of various measurement techniques for porosity and hydraulic gradient to the evaluations of generic repository sites. Practical applications, limits, and errors associated with geophysical exploration and drilling techniques were evaluated. Uncertainties associated with exploration and exploitation of mineral resources in bedded salt environments were also evaluated with respect to the hazards posed to a repository.

MODELING

Systems models are being developed as the principal instruments of long-term risk assessment. Models for dissolution, recharge/repressurization, local migration, regional migration, and dose were revised. These models, some of which were discussed under Task 1, correspond to various barriers to the migration of waste from the repository to the biosphere. Work was begun in revising these models to accomodate probability distributions as input and output.

Progress has been made in the development of two methods of uncertainty analysis: a probabilistic calculation technique and a Monte Carlo technique. Future-history modeling was begun by developing a preliminary framework for the evaluation of the potential risks associated with loss of administrative control (LOAC). Two modeling approaches, represented by the discrete-events model and the Markov-chain model, were compared. Results indicate that the discrete-events model may be more appropriate and more manageable than the Markov-chain model and would provide information for determining key factors in evaluating post-emplacement risks.

Local waste migration models are being developed on the basis of baseline repository designs to describe the geological barrier presented by the repository environment. Preliminary evaluations of the relative significance of various factors were performed. These factors include non-Darcy flow behavior, steam and thermal convection pathways, and earthquake effects.

The output of the local migration models will be a source term for the regional migration models, for which hydrology and transport models are being developed. Work was done on generic and numeric models for a large sedimentary basin. Preliminary models were developed for each rock type. Work continued on understanding how future climatic changes will affect groundwater flow.

The dose model does not actually represent a real barrier, but is a way to translate release into biological effect. The BIODOSE code uses a reference river system that closely resembles the Columbia River Basin. Analysis of other water systems was begun to investigate the sensitivity of calculated hazards to the choice of reference system.

A simple procedure was developed for approximating the radiation dose release that the systems model calculates. Contours of dose for spent fuel were calculated using a waste release function parametrized by a mean arrival time and pulse width as input to the BIODOSE code. A method of computing net whole-body-equivalent doses was developed to to simplify output and make results more meaningful.

TASK VI

During FY 78, Task VI developed radiological performance objectives (RPOs) that can evaluate a waste management system (WMS) in a way that accounts for the probabilistic nature of its consequences and addresses several value-laden issues that are important to the determination of its acceptability. These issues include temporal risk allocation and occupational vs. non-occupational risk. RPOs are summary performance indices that are specifically designed to provide a defensible and understandable basis for regulations and reg guides.

A Risk Evaluation Index was developed as a basis for generating RPOs. The Risk Evaluation Index is a single number designed to evaluate the overall social risk of a WMS. It provides a quantitative definition of risk, a single scale on which risk can be measured and limited. It also incorporates social values where those values are appropriate: value tradeoffs between different types of risk, and attitudes toward uncertainty. Incorporation of social values is important to the defensibility of any set of regulations regarding nuclear waste management.

The most fundamental conclusions to be drawn from Task VI work, aside from the Risk Evaluation Index and RPOs themselves, are that RPOs incorporating uncertainty can be defined and that a Risk Evaluation Index incorporating uncertainty and based on social values can be developed.

The work of Task VI was scheduled for completion in November 1978. The reader is referred to the end-of-project report (UCRL 52574) for detail beyond that provided in this report.

542 223

TASK I: WASTE FORM PERFORMANCE

INTRODUCTION

The objectives of Task I, Waste Form Performance, are to provide an information base for the NRC to set waste performance criteria, to develop regulatory guides, and to evolve a methodology for licensing. During FY 78 we investigated various aspects of management of solid high-level waste (SHLW), spent fuel, and transuranic (TRU), non high-level, waste. The work conducted is briefly synopsized in this introduction with further details provided in subsequent sections.

For SHLW, we studied the effects of human factors and interim storage accidents on waste management risk during the pre-emplacement period. Preliminary work was also done in interim storage seismic analysis. We are analyzing the transient thermal field resulting from emplacement of SHLW in a deep geologic medium with a highly versatile 3-D finite difference heat transfer code. We have completed a detailed cost comparison of solidification processes for the uranium/plutonium recycle fuel cycle. We began to study other fuel cycle costs when the work direction was changed. Finally, we evaluated the potential hazards of SHLW from different fuel cycles.

In the development of an information base for spent-fuel package performance criteria, we began work on package designs with considerations of disposal and retrieval operations. Studies initiated include spent fuel/package interactions and package/medium interactions. Detailed studies of the following have progressed to varying degrees: repository thermal analysis, package design concepts, corrosion, dissolution, and brine migration. We have also computed the potential hazards from spent fuel radionuclides.

The development of an information base for TRU waste form performance criteria began with the data needed to define the waste management problem. The basic data to be compiled are those on waste inventory and classification systems. We have completed a survey of the data for the former and a report on the latter is being prepared. They provide the basis for determining the waste form and package requirements. Finally, a formal probabilistic radionuclide release function methodology has been developed.

SOLID HIGH-LEVEL WASTE

RISK FROM PRE-EMPLACEMENT ACCIDENTS

During FY 78 three general areas of pre-emplacement risk were investigated as part of Task I. They have been summarized below under the headings of human factors, interim storage, and interim storage seismic analysis.

Human factors in pre-emplacement accidents have been considered at the reprocessing plant, during transportation between the reprocessing plant and the repository, and at the repository. Interim accidents include handling accidents and, more importantly, accidents involving loss of coolant or loss of coolant circulation. The seismic analysis reached only a preliminary stage, but produced an outline for a methodology for evaluating seismic risk.

Human Factors

We made a preliminary effort to define the risks associated with human errors in the HLW management cycle.[1] This effort concentrated on activities at the fuel-reprocessing plant, during transportation, and at the repository prior to emplacement. Quantitative analyses of each operation were not attempted, in part because the necessary data were not available and in part because of the much higher level of effort that would be necessary. Instead, incident reports from the nuclear power industry were reviewed to identify the situations in which human error is most likely during waste management operations.

The analysis indicated that critical operations exist in the design, installation, and maintenance of air filtration systems and in the cask-sealing tasks tha' precede transportation of wastes by train and truck. Filter failure caused by human error is most likely to have serious consequences if the storage pool circulation system also fails, causing pool boiloff and canister failure. Circulation system failure can also be promoted by human error. Human errors during cask-sealing tasks, which occur at a rate of about 10^{-5} per cask shipped, have serious consequences when coupled with truck or train impact accidents. The compilation of numerical data needed to quantitatively evaluate the impact of human error on expected dose would require resources greated than those available.

Interim Storage Accidents

We completed a risk analysis for interim storage accidents,[1] to which refinements in the thermal analysis have recently been added.[2] A summary of the approach taken and of the results appears in the following paragraphs.

Accident Scenarios. Figure 1 shows the event tree that describes the two most important accident scenarios for waste stored in a water-filled pool. These accidents can be described as follows:

- Boiloff accident--loss of coolant circulation in the pool and boiloff of the makeup supply.
- Pool drainage accident--a severe earthquake of Mercalli IX intensity or greater, rupture of the pooling lining, and rupture of the containment building.

Either of these accidents can cause breaching of the canisters as the waste overheats, followed by release of volatile radionuclides. All released volatiles are assumed lost to the biosphere in the drainage accident.

Expected Doses. The expected release during interim storage accidents depends on pool-loading methods and the canister configuration in the pool. The following normal conditions were chosen: batch-loading method, canister

542 231

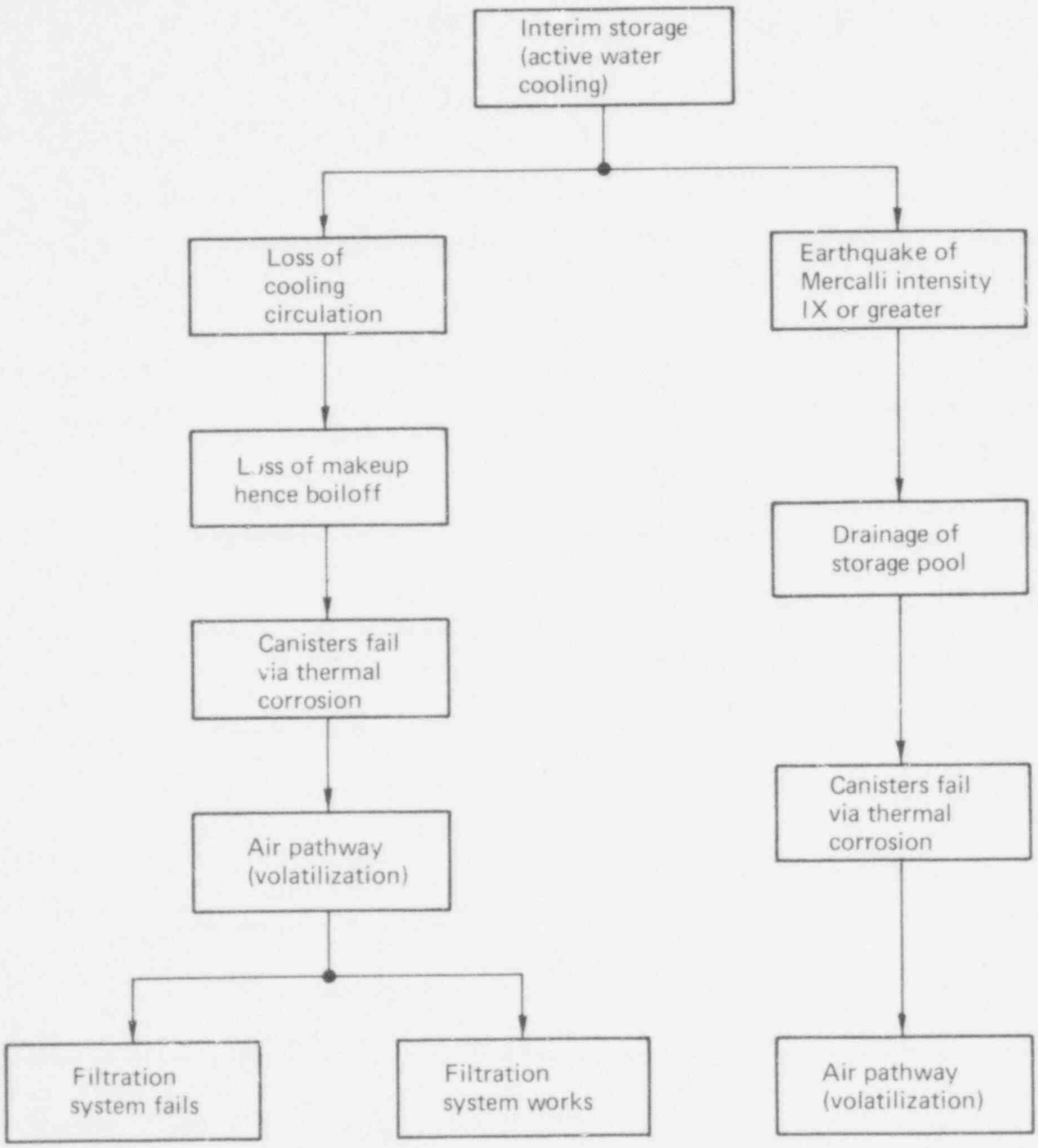

FIG. 1. Event tree for interim storage accidents.

542 232

segregation by age, and 1.5-ft canister spacing. Table 1 summarizes the expected whole-body dose from all pre-emplacement accidents. (The results for transportation accidents have been taken from Ref. 3). The results show that transportation accidents dominate the total risks for spray and fluidized-bed calcine, and that the drainage accident is most important for multibarrier and glass waste forms. The boiloff accident is inconsequential for all four waste forms.

Sensitivity analyses, in which critical parameters were varied to obtain high and low estimates for the expected dose, indicate that the probability of failure for glass and multibarrier wastes can be reduced to zero under the following conditions: continuous loading, aggregation of ages, 2.0-ft canister spacing, and waste ages of at least 1 y.

Interim Storage Seismic Analysis

We developed an outline for the analysis of the risk of failure associated with seismic events. The projected analysis has four major elements, namely, pool description, failure mode identification, seismic risk analysis, and failure analysis. The pool description depends upon the pool site, as well as the pool design. Typical parameters can be chosen for each, (thus describing a generic site), but account should be taken of the characteristics of likely sites. Identifying the failure modes requires study of a three-dimensional, linear model of the storage structure in a simulated seismic environment. The seismic risk analysis produces a probabilistic assessment of critical earthquake parameters, namely, peak acceleration, velocity, and displacement. The final element of the analysis requires nonlinear modeling techniques and produces estimates of the peak acceleration required to cause failure. Combining the results of these last two elements provides an estimate of the risk of failure.

542 233

TABLE 1. Normal expected whole-body doses (in man-rem/MWe-y) from pre-emplacement accidents.

Waste form	Accident Type: Interim storage pool drainage	Accident Type: Truck transportation	Accident Type: Train transportation	Accident Type: Other pre-emplacement	Total: Shipment by truck	Total: Shipment by train
Spray calcine	0	1.2	2.8×10^{-2}	2.0×10^{-13}	1.2	2.8×10^{-2}
Borosilicate glass	0.99	8.0×10^{-3}	4.9×10^{-3}	1.1×10^{-11}	1.0	1.0
Fluidized-bed calcine	0	0.25	7.2×10^{-3}	2×10^{-13}	0.25	7.2×10^{-3}
Multibarrier	1.3	1.3×10^{-5}	1.6×10^{-7}	1.8×10^{-11}	1.3	1.3

A probabilistic approach has been taken to the analysis of seismic risk, the third element in the complete seismic analysis. This approach quantifies the uncertainty in the number, size, and location of future earthquakes, and presents a picture that makes clear the trade-offs between [illegible]y storage-pool designs and the social or economic impact of failure.

The basis of these risk calculations is the total probability theorem

$$P[A] = \iint P\,[A/m \text{ and } r]\; f_M(m) f_R(r)\,dm\,dr$$

Where P[A] is the probability of event A, P[A/m and r] is the probability of A, given events m and r, and f_M and f_R are the independent probabilities that variables M and R will have the values m and r. For example, if A is taken as peak acceleration, M and R would be earthquake magnitude and epicentral distance. Then P[A/m and r] would be a probability distribution (derived from attenuation data) that depends on M and R; $f_M(m)$ would be an actual or postulated frequency distribution; and $f_R(r)$ would be an analytical or numerical distribution of distances.

A computer program has been developed that carries out the integration of probabilities numerically, which makes possible the evaluation of complex site geometries.

REPOSITORY THERMAL ANALYSIS

LLL is performing a thermal analysis of a geologic repository for SHLW. A three-dimensional unit cell model has been developed and the transient near-field temperature distribution is being evaluated using the TRUMP heat transfer computer code.[4] At the close of FY 78, work is continuing on a series of six cases. The first two cases consider ventilated and unventilated salt repositories; the third and fourth cases, ventilated and unventilated repositories in a series of salt and shale layers. The final two cases consider salt repositories ventilated for 5 y and 25 y, then backfilled with salt.

542 235

The initial heat generation rate in the waste canister is assumed to be 3.5 kW. The canister size is 8 ft in length by 1 ft in diameter. Canisters are buried at 18 ft intervals, 10 ft below the floor of the repository room, which has a cross section of 18 ft by 18 ft. The salt support pillars of the room are 70 ft wide. The properties of the salt are temperature dependent.

Since analysis is in progress at the close of FY 78, results are not yet available.

COST COMPARISON OF SOLIDIFICATION PROCESSES

Before environmental impact statements can be prepared for SHLW alternatives, process flowsheets must be developed, capital and operating costs must be estimated, and a cost/risk analysis must be conducted. The results of the cost evaluations that have been completed are summarized here.[5]

Differential cost estimates of the annual operating and maintenance costs and the capital costs for five HLW solidification alternates have been made. To produce these estimates it was necessary to generate conceptual system designs and process flow diagrams for each alternate. Equipment-sizing calculations were performed for the major components based on the processing flow rates and the required equipment capacities.

The annual operating and maintenance cost estimates included the cost of labor, consumables, utilities, shipping casks, shipping, and disposal at a Federal repository. The capital cost included the cost of the component, installation, and building. All estimates are presented in constant 1977 dollars. The differential cost estimates do not include equipment and facilities that are shared with the reprocessing facility or are common among all of the alternates. Table 2 (rail shipments) and Table 3 (truck shipments) present a summary of the total annual cost differential among the five waste form alternates.

542 236

The borosilicate glass alternate has the lowest total annual cost and, therefore, was used as the base for comparison. The other alternates are more costly by $7.4 to $11.9 million per year. The major items in the cost estimates are the disposal costs (in the operating cost estimates) and the HLW storage tanks (in the capital costs estimates). The supercalcine multibarrier alternate ships 180 canisters per year more than the other alternates and consequently has a significantly higher operating cost. Offsetting this, the supercalcine multibarrier alternate does not require HLW storage tanks for decay because of the high heat conductivity of this product. Thus the capital cost for this alternate is significantly lower than the others.

URANIUM-RECYCLE/PLUTONIUM-DISPOSAL FUEL CYCLE

The uranium-recycle/plutonium-disposal fuel cycle is summarized in Fig. 2. The spent fuel is stored at the reactor for 6 mo, then transferred to the reprocessing plant, where plutonium waste is separated from recyclable uranium. The plutonium is solidified with the HLW, then sent with fuel residues and transuranic wastes to a Federal repository.

We developed process flow sheets for this fuel cycle for five waste form alternates: salt cake, spray calcine, fluidized-bed calcine, borosilicate glass, and supercalcine multibarrier.[6] A methodology has also been developed for comparing annual operating and maintenance costs and capital costs for the five alternate waste forms. The underlying assumptions of this cost comparison appear in Ref. 6, but calculations will not be made because work on HLW was terminated during FY 78.

TABLE 2. Cost comparison in 1977 dollars of five waste form alternates (rail shipment).

	Salt cake	Spray calcine	Fluidized-bed calcine	Supercalcine multibarrier	Borosilicate glass
Differential annual operating & main. cost	1,774,000	1,844,000	1,939,000	7,571,000	base
Differential annualized capital cost[a]	9,483,000	9,709,000	9,933,000	(205,000)[b]	base
Total annual cost differential	11,257,000	11,553,000	11,872,000	7,366,000	base

[a]Differential annualized capital costs were computed using a fixed charge rate of 12% per year.

[b]Parentheses ( ) indicate negative differential.

TABLE 3. Cost comparison in 1977 dollars of five waste form alternates (truck shipment).

	Salt cake	Spray calcine	Fluidized-bed calcine	Supercalcine multibarrier	Borosilicate glass
Differential annual operating & main. cost	1,774,000	1,844,000	1,939,000	7,810,000	base
Differential annualized capital cost[a]	9,483,000	9,709,000	9,933,000	(205,000)[b]	base
Total annual cost differential	11,257,000	11,553,000	11,872,000	7,605,000	base

[a]Differential annualized capital costs were computed using a fixed charge rate of 12% per year.

[b]Parentheses ( ) indicate negative differential.

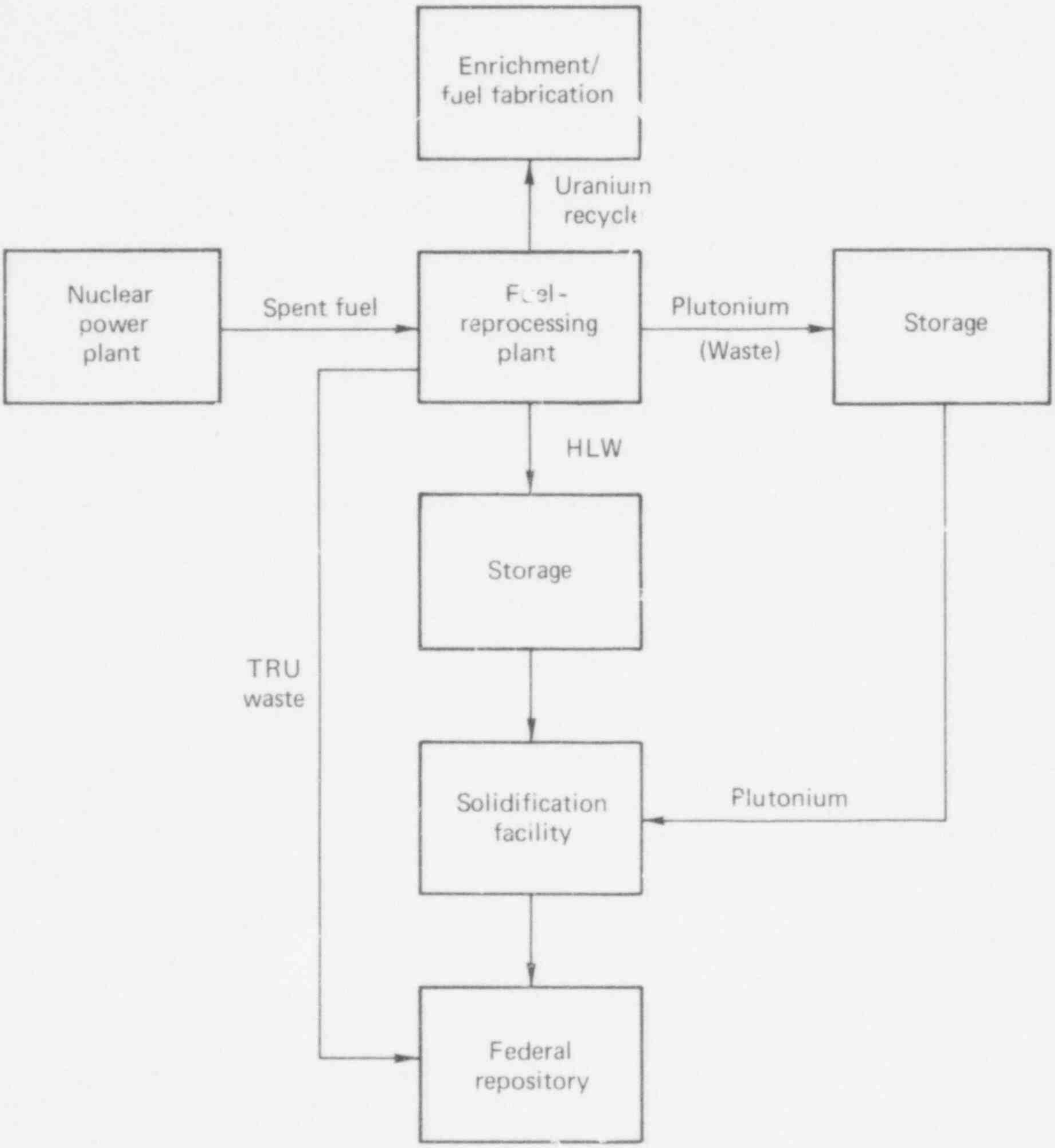

FIG. 2. Flow chart for uranium-recycle/plutonium-disposal fuel cycle.

HAZARDS FROM SHLW FROM PLUTONIUM-DISPOSAL AND PLUTONIUM-STORAGE FUEL CYCLES

We recalculated the radionuclide content of high-level waste from the uranium-recycle/plutonium-storage fuel cycle using the ORIGEN code.[7,8] The same reactor parameters were assumed as in Ref. 1. However, the nuclides ^{135}Cs, ^{59}Ni, and ^{225}Ra have been incorporated into the new calculations, and smaller time steps have been used to reduce interpolation errors. The three nuclides had previously been excluded because consideration had been limited to a list of prescribed nuclides.[1] The potential hazard, in terms of the whole-body-equivalent population dose and the dose to a single individual eating an average diet over 50 y, is illustrated in Figs. 3 and 4 as a function of the waste age. The potential hazards for the HLW from the uranium-recycle/plutonium-disposal fuel cycle are presented in Figs. 5 and 6. Potential hazard is defined as the total dose to the population that would be incurred if 1 MWe-y of waste in soluble form were dumped directly into the river at a given time after removal from the reactor. The population dose is a measure of the accumulated dose to a population over a 50 y period.

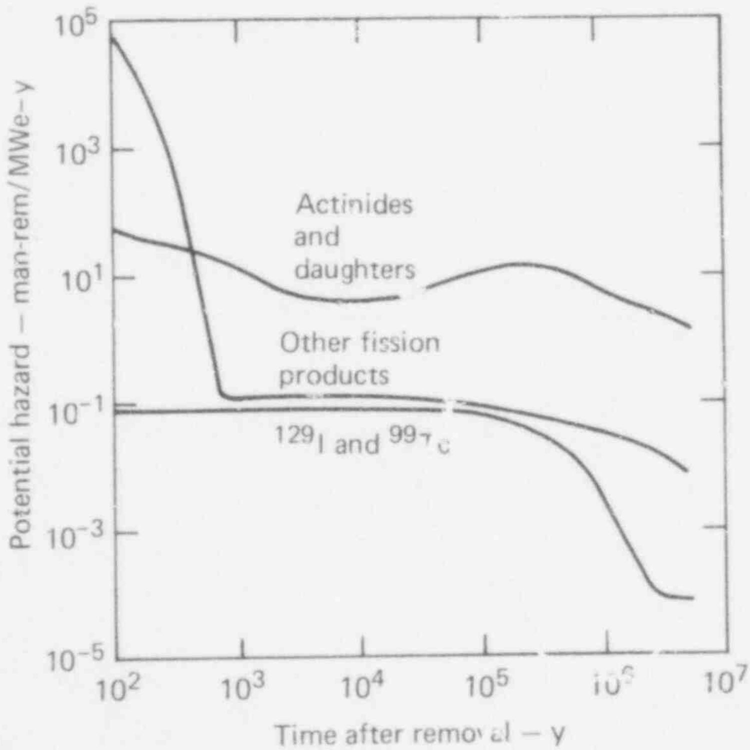

FIG 3. Potential hazard of HLW from the plutonium-storage fuel cycle measured as whole-body-equivalent population dose.

542 240

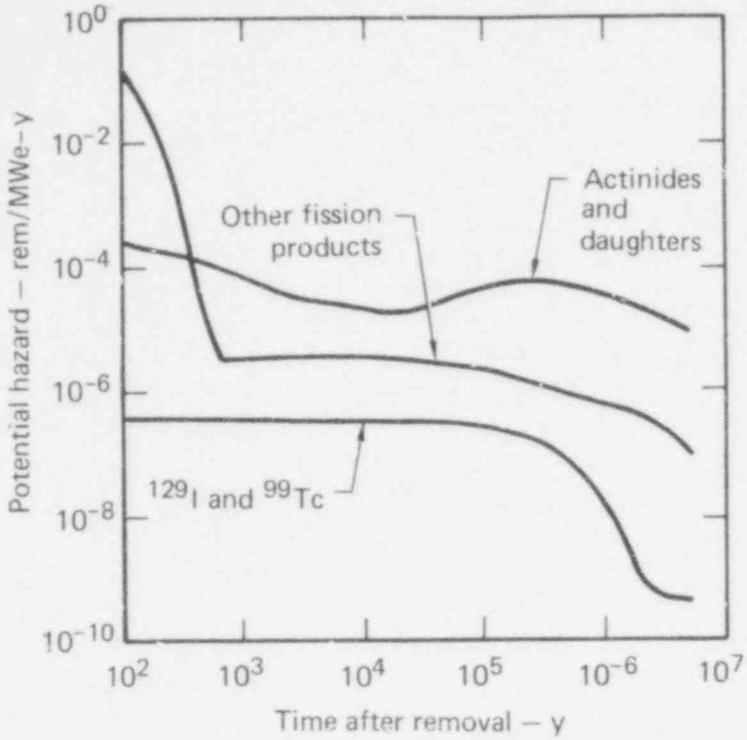

FIG. 4. Potential hazard of HLW from the plutonium-storage fuel cycle measured as 50-y whole-body-equivalent dose to an individual eating an average diet.

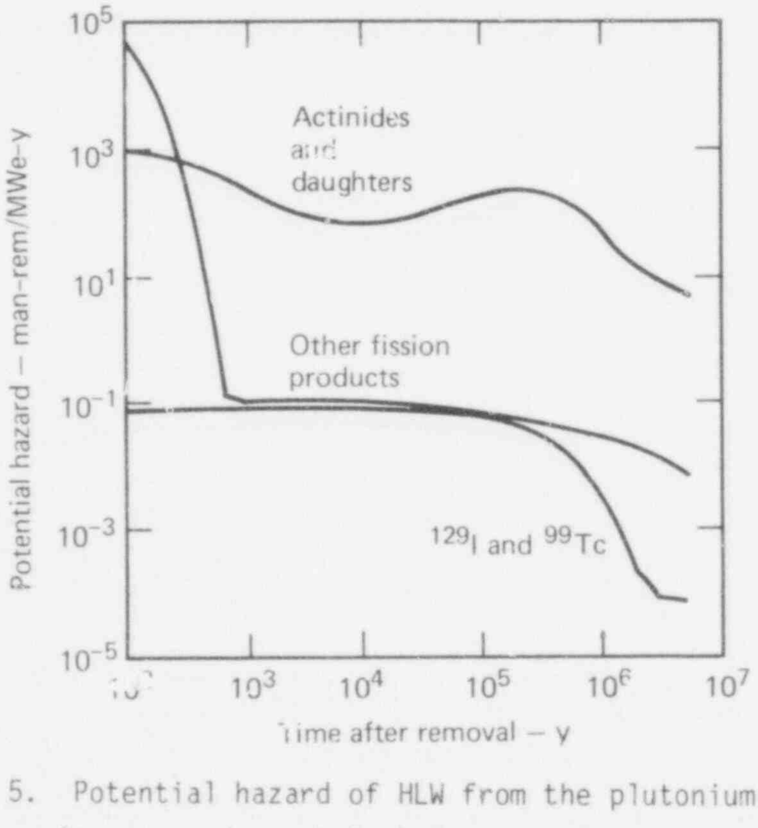

FIG. 5. Potential hazard of HLW from the plutonium-disposal fuel cycle measured as whole-body-equivalent population dose.

542 241

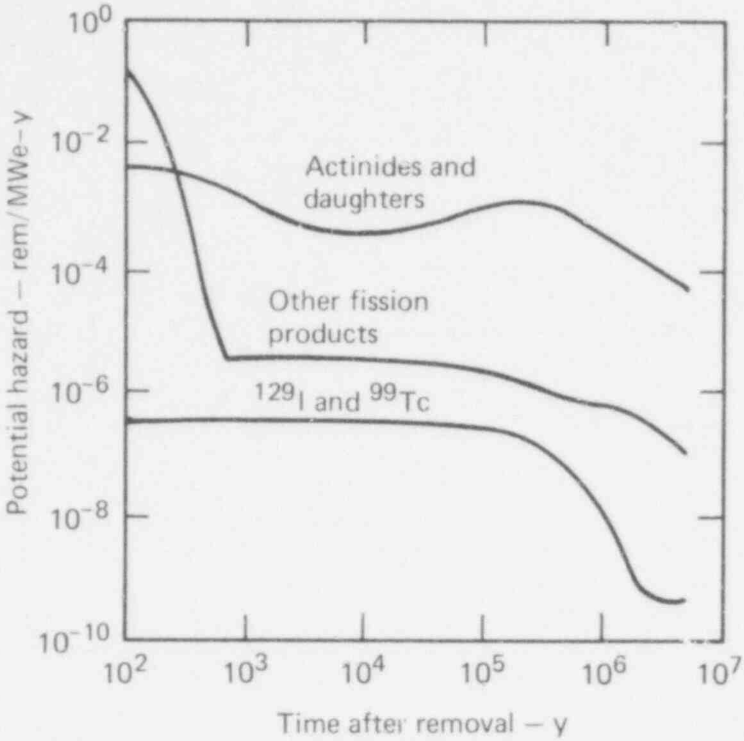

FIG. 6. Potential hazard of HLW from the plutonium-disposal fuel cycle measured as 50-y whole-body-equivalent dose to an individual eating an average diet.

512 242

SPENT-FUEL

HAZARDS FROM SPENT-FUEL RADIONUCLIDES

We calculated the potential hazards from the radionuclides of spent fuel as a function of waste age.[7] The hazards have been calculated for both whole-body-equivalent population dose (Fig. 7) and 50-y whole-body-equivalent dose to an individual eating an average diet (Fig. 8).

We also calculated the gas content of spent fuel using the ORIGEN Code.[8] The results are tabulated in Tables 4 and 5.

If spent fuel is reprocessed, ^{14}C becomes a new gaseous waste. About 4.4×10^{-3} g/MWe-y and 2.0×10^{-2} Ci/MWe-y are produced.[9] Carbon-14 has a half-life of 5.7×10^{3} y. The quantity of ^{222}Rn peaks at about 2×10^{5} y. The peak content is 3.57×10^{-2} Ci/MWe-y for spent fuel and 8.28×10^{-4} Ci/MWe-y for high-level waste from the Pu-storage fuel cycle.

The principal volatiles in spent fuel and HLW are Cs, Te, Ru, and Rh.

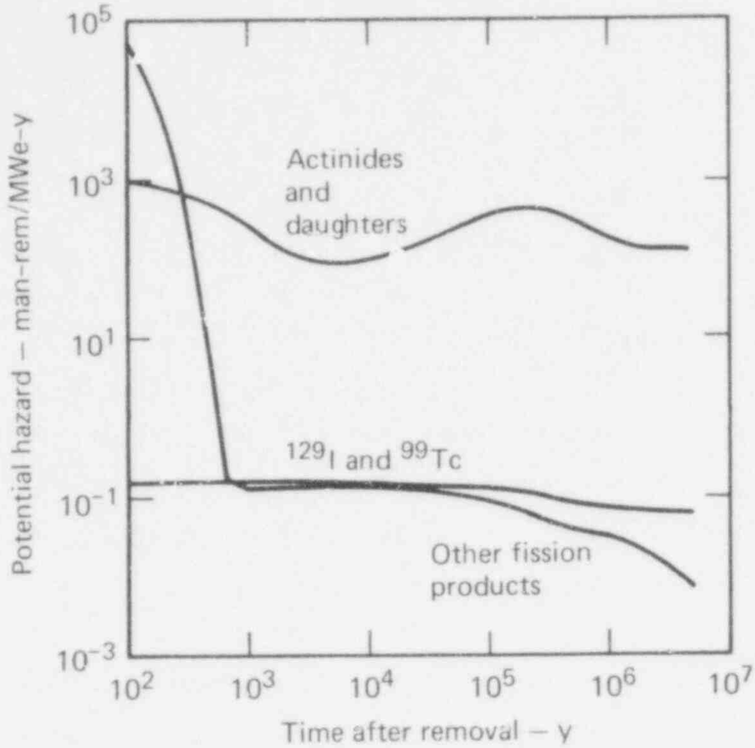

FIG. 7. Potential hazard of spent fuel measured as whole-body-equivalent population dose.

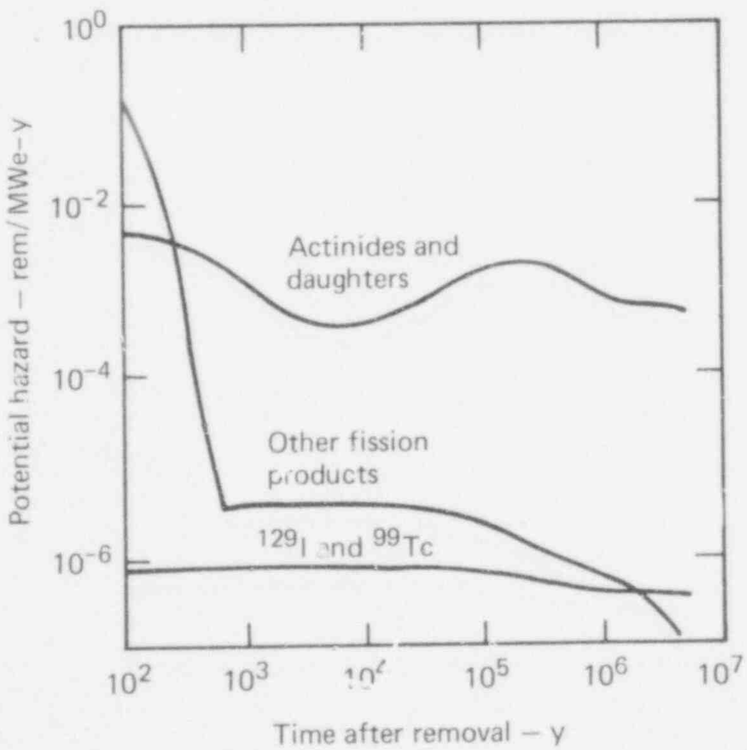

FIG. 8. Potential hazard of spent fuel measured as 50-y whole-body-equivalent dose to an individual eating an average diet.

542 244

TABLE 4. Principal gases in spent fuel after 10 y.

Element and radioisotopes	Half-life, y	g/MWe-y	Ci/MWe-y	Release fraction
Rn (220)	1.8×10^{-6}	5.94×10^{-13}	5.0×10^{-4}	
(222)	0.01			
Kr(85)	10.7	1.13×10^{1}	1.86×10^{2}	30%
Xe(131m)	0.3	1.7×10^{2}	0	10%
(133)	0.01			
I(129)	1.7×10^{7}	8.43	1.17×10^{-3}	2%
(131)	0.02			
H(3)	12.3	1.3×10^{-3}	1.26×10^{1}	
He	stable	2.82×10^{-2}	0	

[a]The release fractions[10] are for waste at a spent-fuel storage facility and do not assume 10-y aging.

TABLE 5. Gases in spent fuel after various lengths of storage.

Element	Mass (g/MWe-y) after storage 100 y	500 y	5×10^{6} y
^{222}Rn	3.83×10^{-12}	1.25×10^{-10}	6.38×10^{-8}
Kr	1.08×10^{1}	1.08×10^{1}	1.08×10^{1}
Xe	1.70×10^{2}	1.70×10^{2}	1.71×10^{2}
I	8.43	8.43	7.11
^{3}H	8.17×10^{-6}	1.33×10^{-15}	--
He	1.62×10^{-1}	5.11×10^{-1}	1.4×10^{1}

5A2 245

REPOSITORY THERMAL ANALYSIS

A three-dimensional thermal analysis of a deep geologic repository for spent fuel is being conducted using the TRUMP computer code.[5,11] The model uses a unit cell consisting of one spent-fuel canister buried in salt beneath a ventilated room in the repository. The purpose of the analysis is to evaluate the near-field thermal environment, which is an important variable in other repository studies, such as analysis of thermal stress, canister corrosion, radionuclide dissolution, and brine migration.

In the current analysis we are considering a baseline design and are conducting parametric studies to determine the effect of changes in the spent-fuel heat generation rate, the convection coefficient for room ventilation, the room air temperature, and the thermal diffusivity of the repository media. The most sensitive parameter found is the heat-generation rate within the unit cell. Fig. 9 shows the radial temperature distribution around the canister at the time of maximum canister temperature for several emplacement times.

DESIGN CONCEPTS FOR RETRIEVABLE PACKAGES

We completed three tasks related to the reference design of retrievable storage canisters for radioactive waste.[12] The three tasks were the reference design itself, the identification of the failure modes most important for studies of structural integrity, and the development of a risk methodology for the structural integrity of the containers.

The reference design is a sealed storage canister based on the waste isolation pilot plant (WIPP) design, with slight modifications that improve resistance to impact failures at either end of the canister. The modifications consist of an alternate lifting yoke arrangement for the top head and a revised bottom-head design for absorbing impact energy (Fig. 10). Welded closures provide the seal at each end. Overpacking is a possibility, but is not included in the preliminary reference design.

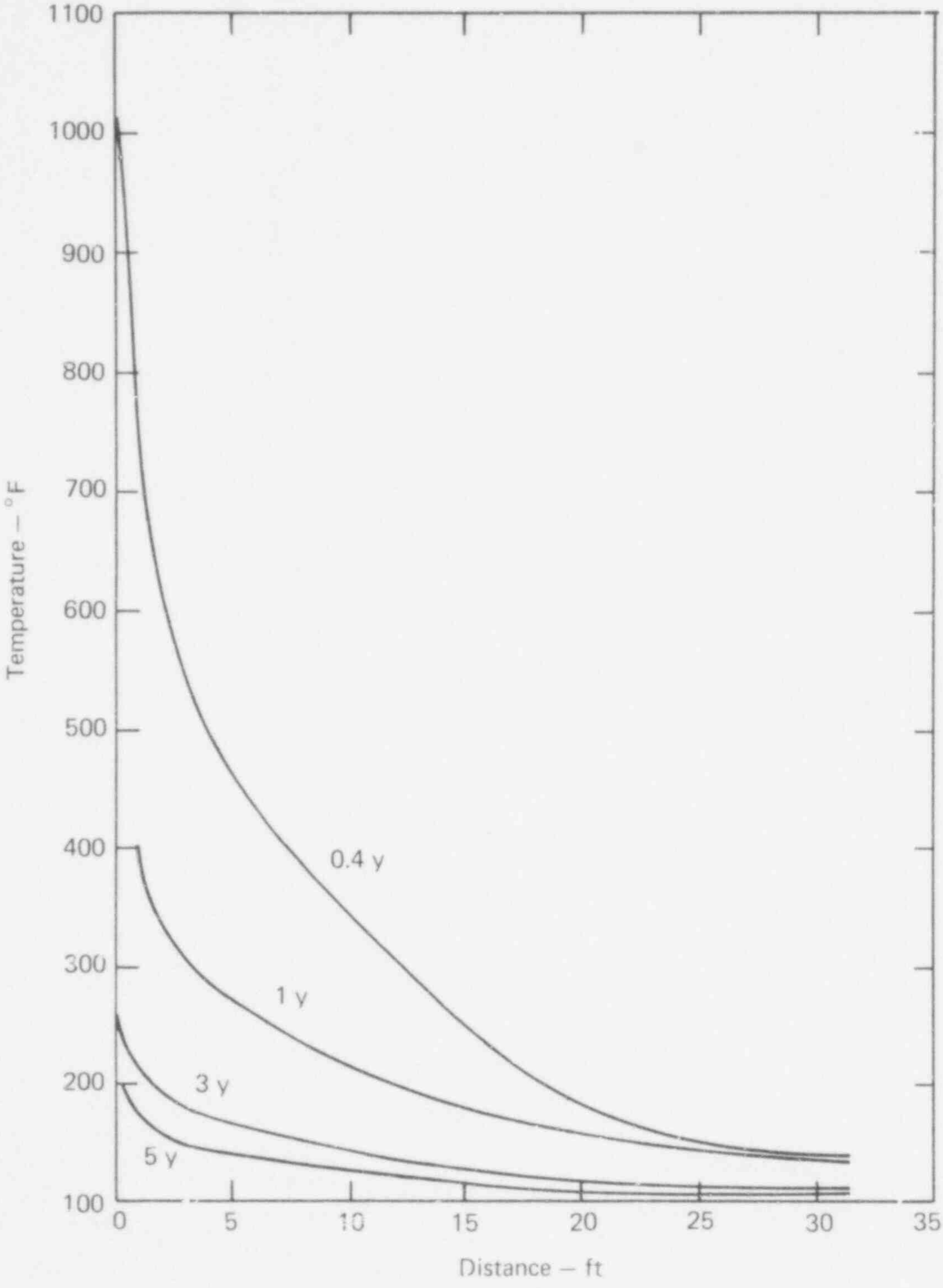

FIG. 9. The radial peak-temperature profile for various emplacement times (following reactor shutdown).

542 247

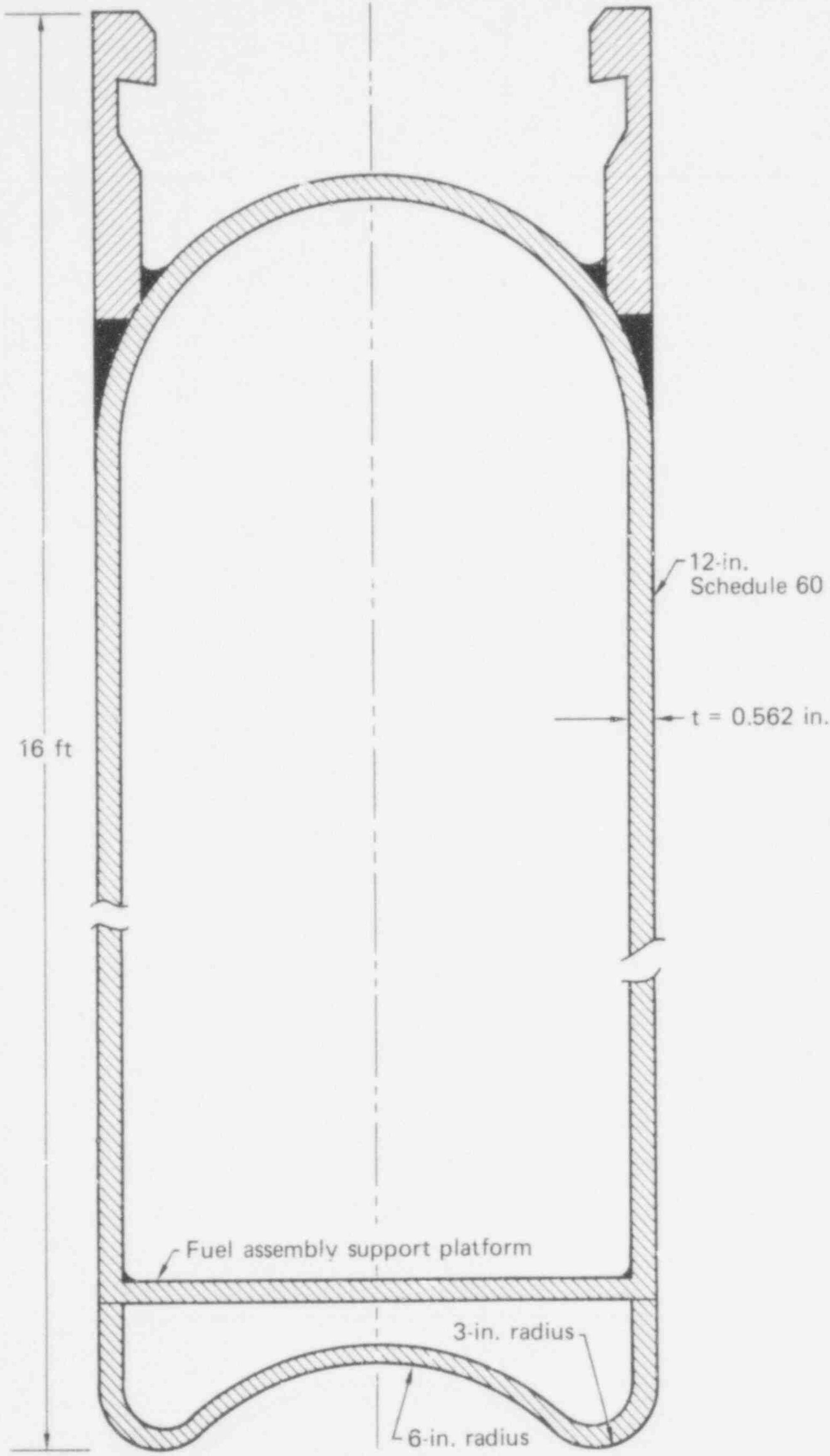

FIG. 10. Reference design for retrievable waste canister.

542 248

The four failure modes that were deemed the most important for the design of the reference canister are: (1) loss of functional capability (2) ductile rupture of the canister (3) buckling of the structural members, and (4) stress-corrosion cracking. Failure scenarios are provided for each of the failure modes.

SPENT-FUEL DISSOLUTION MODELING

A major objective of the dissolution modeling subtask is to develop a computer model to identify and evaluate parameters affecting spent-fuel dissolution, and radionuclide release rate and migration as a result of groundwater intrusion into a deep geologic waste repository.

A methodology was developed, and simplifying assumptions were made to obtain an evaluation procedure that would produce numerical output. Many important chemical and physical properties of the repository and wastes relating to dissolution were identified, and values were assigned during model development. An existing computer program, MINEQL, which calculates the chemical equilibrium composition of aqueous systems, was selected because of its speed, simplicity, and flexibility. Modifications were made to enlarge the data base to include radionuclides existing in spent-fuel wastes, as well as nuclides found in natural waters. Control logic was incorporated so the effects of groundwater composition on waste solubility could be evaluated over a range of system parameters.

It was determined that the groundwater effects on solubility are significant and vary with different radionuclides. Solubilities can be computed for specific nuclides in specific solutions, but it is not reasonable or necessary to assign fixed compositions to any potential intrusive water. To calculate probability values for the concentrations of significant components, statistical analyses could be performed on published data of groundwater in different geologic media, such as salt, shale, granite, and tuff.

See Appendix A for a more complete discussion of spent-fuel dissolution modeling, the assumptions used, and suggestions where additional work seems warranted.

512 249

CORROSION STUDIES

Spent-fuel canisters must isolate the spent fuel from the environment, and must be retrievable during the first 25 to 60 y of storage. When stored in halite deposits (salt beds), however, they will be subject to a severely corrosive environment as hot brine inclusions migrate to the canister. Brines in halite deposits contain not only NaCl, but also high concentrations of the more soluble calcium and magnesium chlorides. These chlorides hydrolyze readily to give acidic brines that are much more corrosive than the neutral NaCl brines. Canister corrosion rates are also accelerated by the elevated temperature resulting from self-heating of the spent fuel and by dissolved oxygen.

No data base exists for the corrosion resistance of materials in halite brines; indeed, these brines have not yet been well characterized. However, other industries have had to cope with concentrated brines, among them geothermal, refrigeration, secondary oil recovery, desalination, and various chemical process industries. For preliminary design purposes we surveyed published literature in these fields to identify the materials and protective measures compatible with hot, acidic brines.[1] These materials and protective methods must, of course, be tested in actual halite brines.

Despite inadequacies in the available data (especially with regard to localized corrosion such as pitting), we found several metals and alloys that appear to be relatively resistant to hot brines. The following exhibited uniform corrosion rates of less than 25 μm/y (1 mil/y) and were resistant to crevice and pitting corrosion.

	Metal or alloy	Composition (major constituents)
Metals:	Ta	Ta
Ti alloys:	Ti Code 12	Ti + 0.3% Mo, 0.8% Ni
	TiPd alloy	Ti + 0.15% Pd
Ni-base alloys:	Inconel 625	Ni + 20%-23% Cr, 8%-10% Mo, 3%-4% Nb/Ta, 1% Co
	Hastelloy C-276	Ni + 15.5% Cr, 16% Mo, 2.5% Co, 3.8% W, 1% Mn, 5.5% Fe, 0.35% V
Fe-base alloy:	29-4	Fe + 29% Cr, 4% Mo

All of these materials are relatively expensive, but cheaper materials, such as carbon steel and low alloy steels, all exhibit excessive corrosion rates.

Materials such as carbon, glass and enamel, fluorocarbons, epoxide-type resins, and polymer cements also showed good resistance to chloride solutions. While these materials are not suitable for constructing the canister, they could be used as a protective coating applied over a base metal.

On the basis of this preliminary survey, we recommend for further study a carbon-steel canister isolated from the brine by a chloride-resistant coating of glass, fluorocarbon, epoxy, or polymer cement. Cathodic protection would probably also be necessary. A testing program is essential to confirm the suitability of these materials and to compare them with uncoated corrosion-resistant metals. From the results of such a program, a methodology may be evolved to predict material performance under various adverse conditions.

BRINE MIGRATION

We investigated processes that can lead to mobilization of brine adjacent to spent-fuel or nuclear waste canisters.[14] Thermal gradients (the Soret effect) are likely to be the dominant cause of such mobilization: the higher the temperatures and the greater the gradient, the faster the migration. Migration rates also depend on the size and composition of the inclusions. Large inclusions should generally move faster than small ones, though the effect of size is relatively unimportant for inclusions 1 mm or more in diameter.

There is considerable uncertainty in the value of some of the independent variables, especially the Soret coefficient, in the equation governing migration. Nevertheless, calculated values for brine accumulation during Project Salt Vault are consistent with observations.[15] Velocities as high as 4×10^{-7} m/s (1.2 m/y) are calculated at the salt-canister boundary. Based on calculated velocity fields, as much as 40 l of brine could accumulate around each canister during a 10-y storage. Future work will concentrate on two thermomechanical effects of this brine accumulation: coupling between the salt and the canister and accelerated creep due to pressure solution.

TRANSURANIC (TRU) WASTE

The major components of the TRU waste program include (1) the identification of the inventory and sources of transuranic waste, (2) the development of a functional definition for this waste, (3) the identification of the waste form and package, and (4) the evaluation of the resultant waste package in a deep geologic waste repository with major emphasis on potential radioactivity release functions.

We conducted a step-by-step program that requires compilation of an extensive amount of information dealing with both commercially generated TRU waste and military waste. The approach has been to develop appropriate working documents that will be used throughout the program. We expect these documents to be revised periodically to reflect an improved data base. The current program has produced a report entitled Inventory and Sources of Transuranic Solid Waste,[16] which is summarized in the following paragraphs. A second report, entitled Review of Potential TRU Waste Classification Systems, will be completed shortly. A third report dealing with various waste forms and waste packages is also being prepared. Further, a TRU data base is being prepared for use in the project to ensure that all participants are using the same information.

INVENTORY AND SOURCES OF TRU SOLID WASTE

We prepared a report to support the development of standards and criteria for TRU-contaminated wastes.[16] Highlights of the report, which presents an overview of TRU-contaminated waste generation, its sources, and past practices dealing with it, are presented below.

Sources of TRU Wastes

Pending numerical definitions being developed by the NRC, the potential sources of radioactive wastes that require geologic repository disposal are as follows:

- HLW and TRU wastes generated by the operation of such Department of Energy (DOE) facilities as Hanford Works, Savannah River Plant, Rocky Flats, the Idaho National Engineering Laboratory (INEL) and referred to as military wastes.
- Reprocessing of fuel discharged from light water power reactors.
- Fabrication of mixed-oxide fuel.
- Unreprocessed fuel discharged from light water power reactors.
- HLW and other wastes generated by the Nuclear Fuel Reprocessing Plant operated by Nuclear Fuel Services, Inc., at West Valley, N. Y.
- Radioactive wastes from decommissioning reactors, independent spent-fuel storage basins, fuel-reprocessing plants, and mixed-oxide fuel-fabrication plants.

DOE Facilities

The TRU waste is currently separated into waste containing greater than 10 nCi/g of TRU alpha radioactivity and those containing less. These TRU wastes are also designated as retrievably or nonretrievably stored. The estimated total volumes of TRU waste buried or stored each fiscal year from 1973 to 1976 at DOE sites show a factor of nearly 2 decrease from 4,695 m^3 to 2,240 m^3 in nonretrievable storage while the average annual volume in retrievable form remained roughly constant at 9,600 m^3, with a volume range of 7,630 m^3 to 11,800 m^3, over the same period.[17] An increase in waste Pu stored or buried rose from a cumulative total of 820 kg in 1972 to 1,030 kg in 1975 and to about 1,135 kg in 1977 (Fig. 11).

Table 6 presents an estimate of future TRU waste burial volumes. These values are somewhat different from the generation rates and reflect the use of volume reduction technology.

512 253

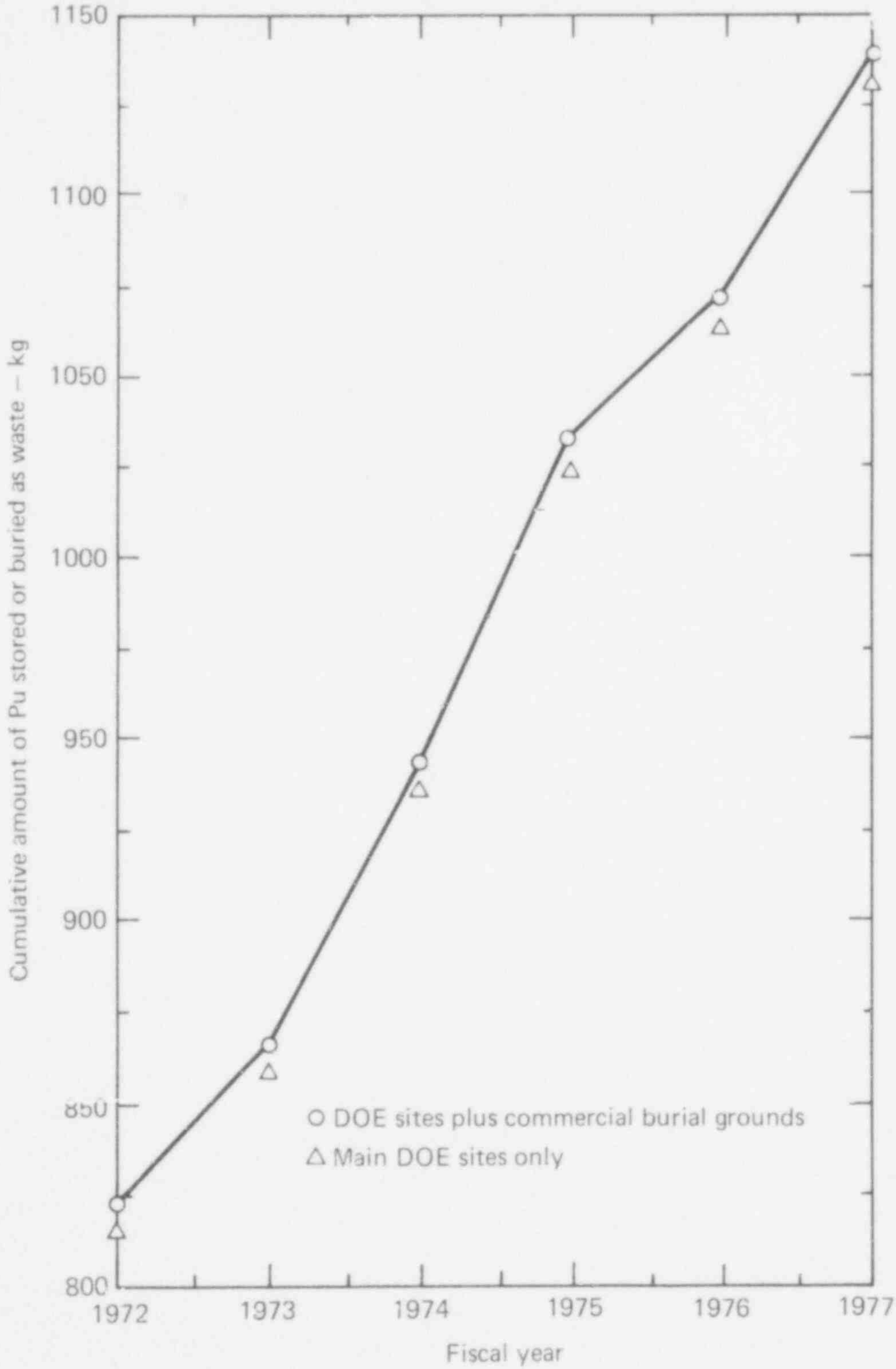

FIG. 11 Cumulative amount of DOE plutonium buried or stored as waste.

542 254

TABLE 6. Estimates of future annual TRU solid waste buried volume from DOE facilities.

Site	TRU waste volume, m^3
Hanford Reservation	1,105
Los Alamos Scientific Laboratory	510
Idaho National Engineering Laboratory[a]	4,700
Oak Ridge National Laboratory	113
Savannah River Plant	170
Other DOE facilities	28
Total	6,626

[a] Includes Rocky Flats.

Commercially Generated TRU Waste

The commercially generated TRU waste includes the waste generated in the nuclear fuel cycle as well as numerous miscellaneous R&D activities and small commercial use of TRU radioisotopes such as ^{241}Am and ^{244}Cm. At present there is no commercial reprocessing of spent fuel. Existing TRU from reprocessing of spent fuel is limited to that generated at the Nuclear Fuel Services, Inc., facility.

A second category of solid wastes, which includes a wide variety of materials such as cladding hulls and miscellaneous wastes from fuel-reprocessing and refabrication steps, represents a problem comparable in importance to that posed by isolation of the actinide component of the HLW. Such wastes can be classified into the following general categories: cladding hulls, intermediate- and low-level waste (ILW/LLW), general trash, equipment, and dry chemicals. Table 7 gives the estimated volumes of TRU wastes for each category.

542 255

TABLE 7. Estimated volumes of solid TRU waste.

Waste type	Volume range, m^3/t[a]	Probable average
Cladding hulls	0.3 to 0.55	0.4
ILW/LLW	0.06 to 0.4	0.2
General trash	0.03 to 10	b
Equipment	0.03 to 0.3	c
Dry chemicals	0.1 to 0.3	0.15

[a]Volume of actual waste. Packaging, shielding, and the irregular shape of packages may increase the space required for storage by a factor of up to 3.
[b]Volume of general trash is independent of processing rate. Assumed to be 1,200 m^3 per y per plant.
[c]Volume of equipment waste category is independent of processing rate. Assumed to be 150 m^3 per y per plant.

The projection of commercial TRU-contaminated waste from fuel cycle activities is typically performed by using such fuel-cycle computer models as NUFUEL, ENFORM, ORSAC, KWIK PLAN, FLYER, and ALPS. These models typically emphasize such items as: material and process flow, isotopic composition, mass, volume and composition of process waste streams, economics, etc. For the purpose of our report, we reviewed a number of projects and presented a projecting basis that most closely reflects the present U.S. nuclear energy policy. It is understood that the NRC is presently developing a modularized, integrated computer model for projecting the quantities, physical characteristics, and associated storage/disposal costs of commercial nuclear wastes generated on a regional basis through the year 2000.

512 256

RELEASE FUNCTION METHODOLOGY

We have developed a release function methodology by which the probability and extent of radioactive release from nuclear waste can be estimated.[18] The methodology takes account of the conditions leading to release, the variation in the fraction of the waste released from the waste form, and the variation in the fraction of the waste that breaches the containment barrier.

In general, we have incomplete information about the values and distribution functions of the parameters in the risk calculations. We are currently making sample calculations to investigate the effects on the calculated risk of having only estimates of expectation values and some general information about the distribution functions.

OTHER TRU WORK

We are now preparing a report entitled Review of Potential TRU Waste Classification Systems to provide a concise overview of the problems of TRU wastes. The purpose of this report is to briefly identify waste classification schemes that have been used in the past and to determine their current applicability. In addition, the report will address work performed by others regarding the development of a more defensible and workable definition of TRU contamination. In general, three levels of TRU contamination or potential contamination will be discussed. The first of these is due to innocuous waste in which TRU levels are effectively zero. At this level, the potential waste would be handled as a totally uncontaminated waste and discarded in a sanitary landfill or by other commercial means. At levels above this innocuous level and below some value of the order of 300 to 400 nCi/g, the material would be considered suitable for disposal in present commercially operated (or equivalent) shallow-land burial sites used for low-level radioactive wastes. At still higher levels, the material would require disposal in a Federal waste repository.

A second report on the waste forms and packaging requirements for TRU is also being prepared. This portion of the program will define the variations in the present TRU wastes as well as future wastes. In addition, new technologies, including such volume reduction techniques as incineration, acid digestion, fluid-bed driers, etc., will be addressed with special emphasis on the final waste characteristics. Furthermore, the report will discuss various solidification techniques that might be employed and will consider the constraints imposed on package design by Department of Transportation regulations and by the tentative repository designs. This information will subsequently be used in the LLL program to determine the potential requirements of the repository design and to assess the performance of the TRU waste package.

TASK II: SITE SUITABILITY

INTRODUCTION

Before FY 78, the Site Suitability Task emphasized the identification of physical processes and parameters important to the success of a bedded salt repository as a container of nuclear waste. A pipe model was constructed to study such repositories and to perform the sensitivity analysis and limited uncertainty analysis required. The model was adequate for this job.

During FY 78, a more comprehensive approach to the problem of Site Suitability was initiated. The goals of Task II were reassessed, which resulted in restructuring of the task. Nevertheless, while this more comprehensive approach was being developed the work left over from FY 77 was continued. FY 78 thus became a year of transition in which both the original and new goals and objectives were pursued.

Because classification and revision of Task II goals was a major effort during FY 78, a summary of that work follows.

LONG-TERM OBJECTIVES/GOALS

The long-term objective of the Site Suitability Task is to provide a sound technical base for the NRC to write Regulatory Guides for the siting of nuclear waste repositories in selected geologic media.

Long-term goals are more easily identified if the point of view of the repository licensee is taken. The licensee must locate a site for a nuclear waste repository that will "protect the public health and safety". The site-selection process will involve two stages: (1) a preliminary selection based on qualitative criteria, and (2) a final selection based on measurements and performance calculations.

542 259

During the preliminary selection process, a list of potential host media with qualitative criteria for each is generated. The qualitative criteria are judgmental statements made by experienced geoscientists and other experts. These statements may deal with such items as groundwater flow, fracturing, topography, etc. In summary, a checklist of features and attributes judged important for the success of the site as a containment medium must be produced for each medium. This checklist of qualitative criteria is used to select a small group of sites for more detailed measurement and assessment.

During the final selection process, measurements followed by detailed calculations are made. "Which measurements are possible?" and "What confidence can be assigned to predictions of site performance based on these measurements?" are key questions that must be answered. By implication, a model that translates site measurements and their uncertainty into a probability distribution for site performance must be created.

What then does the licensee require to select a "suitable site"? He needs:

- A checklist of qualitative criteria for each host geology
- A list of possible measurements and their uncertainties
- A knowledge of the licensing tools and the methods of translating these measurments into a prediction of site performance.

PROGRAM ORGANIZATION

Because of the above requirements, The Site Suitability Task is divided into the three areas: qualitive criteria, measurements and models.

Qualitative Critera

A checklist of site attributes should serve as a guide in the initial site selection process. Parts of the checklist will be unique to the geologic medium considered for repository siting.

Measurements

It is necessary to identify both the measurements pertinent to each medium and to ascertain the types and degrees of the associated uncertainties. The measured data must be related to parameters of the site performance models.

Models

Models are used as the primary site performance tool. There are two basic levels of modeling in the Site Suitability Task. Systems level models are simple probabilistic rapid-running models for assessing the post-sealing site contributions to total risk. More detailed models serve as guides in the construction of systems models and are used to assess the modeling uncertainties in the systems models. The level of detail required in the supporting model is governed by the uncertainties in the input parameters, the level of knowledge of the model system, and the degree of complexity of that system. The systems and detailed models are used to determine the range of measured and design values which may lead to acceptable site performance.

FY 78 EMPHASIS

The emphasis during FY 78 was model support, revision, and improvement. Later in the year a restructuring of the modeling effort took place. The restructuring took the form of focusing on individual barriers to waste migration as study objects. Activities outside of modeling included formulation of long-term site suitability goals (see above), scoping of other siting media, and analyzing geophysical measurements uncertainties.

The materials in this report include summaries of work both completed and still in progress. Much of the work reported here appears in more detailed reports issued during the year. The reader is directed to these reports if more detail is required.

542 261

SITE CHARACTERISTICS

QUALITATIVE DESCRIPTION

Repository Design Models

To properly evaluate site suitability and long-term risks, certain assumptions must be made about the design and operation of the repository. We developed simple design models to identify the probable ranges in design parameters that might affect long-term repository behavior.[19] We also developed generic settings for repositories sited in basalt, granite, and salt domes. These settings are described in detail in Ref. 20. (See also "Generic Models" under "Regional Waste Migration Modeling" below.) Since these data are well documented elsewhere and their results are implicit in the discussions which follow, further elaboration was not considered necessary here.

Vadose Zone Repositories

We are investigating the feasibility of modeling waste repositories located above deep-seated water tables (See Appendix B). The flow of groundwater in unsaturated regimes has several characteristics suggesting that a repository located deep in an unsaturated region will stay dry as long as the water table remains below the repository. While these characteristics may permit a viable waste repository to be constructed, they also require models that are quite different from those developed for saturated flow conditions. The extensions of such models to geologic time scales may require additional research into unsaturated flow phenomena. Potential sites in unsaturated media may be found at the Nevada Test Site, the Snake River Plain of Idaho, the Columbia Plateau of Washington, and possibly the Colorado Plateau of Arizona.

542 262

GEOLOGICAL/GEOPHYSICAL DATA

Geochemistry

The following paragraphs together with Appendix C cover the following:

1. A discussion of the retardation factors used in the mass-transport model
2. An expansion of the geochemical data base incorporating the results of recent research
3. A survey of the physical and chemical processes that affect porosity
4. An evaluation of the uncertainties in the retardation factors
5. The development of a chemical-equilibrium code to define solubility limits for future use in the mass-transport model
6. A brief discussion of the sorption characteristics of granites and basalts.

A brief summary follows, and details can be found in the appendix.

Retardation Factors. In FY 77, a literature survey provided data for estimating retardation factors for the hydrologic model.[21] In the past year, further review of the data has suggested that revisions in the underlying assumptions and minor changes in the retardation factors would improve the model. The changes are discussed in Appendix C. In general, retardation factors have been increased for the salt repository to account for both contamination of the salt layers with layer silicates and the possibility of sorption in the shale layers in the presence of saline groundwater. A small retardation factor has been assigned to the negative species (TcO_4^-, I^-) to account for possible ion filtration in the shale layers.[22]

The relative effects of the various physical and chemical parameters on the porosity, permeability, and sorption characteristics in sedimentary rocks are strongly affected by rock heterogeneity. Local variations in grain size, sorting, mineralogy, mineral alteration, cementation, or dissolution create local changes in retardation. Therefore, the use of a range of parameter values rather than a single value will more accurately describe a site's

512 263

characteristics. Baseline values used in the current hydrologic model must be considered only average values for the flow pathway. A more complete discussion of the variations in porosity and sorption characteristics appears in Appendix C.

We can define three levels of uncertainty in the retardation factors (for actinides and fission products) according to the amount of data available. At each level the uncertainty values are merely estimates based on technical judgment. The uncertainty (U) corresponds to one standard deviation in the logarithmic distribution of the retardation factor. Thus the probable range of a retardation factor K_f can be thought of as extending between K_f/U and K_fU. The three levels of uncertainty correspond to the following types of available data:

1. A literature survey only; that is, no experimental data for a specific site (U = 100)
2. Laboratory measurements for a specific site, but no in situ data available for comparison (this assumes that the laboratory approximates in situ conditions) (U = 10)
3. A combination of laboratory and in situ data (U = 5)

Currently the uncertainty in the retardation factors used in the mass-transport model represents not only the uncertainty in the sorption data, but also our lack of knowledge about how retardation is affected by chemical and physical parameters. These parameters include, in likely order of importance, exchange capacity, solution chemistry, oxidation potential, flow rates, surface area, rock heterogeneity, and thermal effects. Even with accurate and extensive laboratory experiments, but without in situ measurements, the level of uncertainty in the retardation factor may remain unacceptably high. With in situ measurements, we believe that the uncertainties can be reduced to those associated with experimental error, rock heterogeneity, and changes in retardation due to future environmental variations in the system (e.g., changes in the groundwater composition or the extent of fracture flow).

542 264

Solubility Limits. We are currently developing a chemical-equilibrium code designed to identify the major aqueous radionuclide species present in various groundwater systems and to estimate their solubilities. Modifications to an existing code, MINEQL, has produced a code that plots concentration maps of aqueous species for a range of Eh and pH characteristic of natural waters. Preliminary results for uranium[23] show that concentrations are highly dependent on groundwater chemistry. Concentrations varied from 10^{-3}M to 10^{-13}M for total dissolved uranium. Future work will include the investigation of the solubilities of other elements, the development of a dissolution model for spent fuel, and a parameter sensitivity analysis to identify which parameters are important in minimizing solubility and maximizing retardation. This work overlaps with Task I objectives.

Sorption Characteristics of Granites and Basalts.

Granites. As discussed in Appendix C, sorption data for granites are limited. However, recent results indicate that granites may have better sorption properties than was originally thought. In fact, the distribution coefficients for fresh granite compare surprisingly well with those used at LLL to calculate retardation factors for shale. The major concern in modeling a granite repository, however, is estimating retardation factors for fracture flow. Since groundwater flow through granite is almost entirely along fractures, it is important that future experiments be designed to measure sorption in terms of surface area as well as mass.

Basalts. With the excepti of Sr and Cs, sorption data for basalt are nonexistent. However, current geochemical research funded by DOE at Battelle Pacific Northwest Laboratories should give useful results soon. Considering the variation in mineralogy in basalts, we expect that sorption characteristics may be site specific. Retardation factors for a hydrologic model of a basalt repository will depend on the flow regime used to describe the generic site. Groundwater can flow along fractures or layer boundaries, through vesicular layers, or in sedimentary sequences that lie between successive basalt flows. As with granite, we will have difficulty translating experimental distribution coefficients to in situ conditions. A more extensive discussion of basalts appears in Appendix C.

542 265

Data Base Development[23,25,26]

An on-going GEI project is to update parameter values used by the various Task II models to reflect improvements in basic geological data. Part of this work is to provide a data base of geotechnical and mining information needed to develop credible models and reach realistic conclusions. An exhaustive state-of-the-art study was not intended. Rather, the intent was to provide a basic framework of data and, interfacing with the LLL model results, further investigate those areas that appear to affect long-term risk most significantly.

The data base work also included the following:

- A comprehensive listing of geologic, hydrologic, and mining/rock mechanics factors that might affect long-term risk.
- A discussion of the more significant of these factors.
- A literature survey consisting of a compilation of regional geologic and hydrologic data on bedded salt, salt domes, shales, granites, and basalts. This documentary information was supplemented with GAI's professional experience.
- A subsurface natural resource survey related to siting of the repository in areas of minimum existing or potential subsurface mineral resources.
- A discussion of available hydrologic testing techniques in low permeability rock.
- A discussion of rock mechanics information and current mining techniques related to repository construction and performance.

512 266

GEOLOGICAL/GEOPHYSICAL MEASUREMENTS

Geological/geophyscial measurement techniques (and associated errors) that may be applicable to the evaluations of generic repository sites[27] and field tests conducted independently of this program including borehole, surface, and in-mine procedures as well as some applicable laboratory tests are briefly discussed below. Porosity and hydraulic conductivity measurements in the field and in the laboratory, hydraulic gradient, seismic reflection and refraction, radar, electrical resistivity, gravity and magnetic surveys, and drilling are summarized. The practical applications, limits, and measurement errors are described. In addition, uncertainties caused by exploration and exploitation of mineral resources in bedded salt environments are discussed.

Selected Geotechnical Measurements

Porosity. Relatively accurate laboratory measurements of effective porosity can be made by choosing a sufficiently large sample size. These measurements are representative of field conditions only where interstitial porosity is much larger than fracture porosity, such as in a porous sandstone.

Field determinations of porosity can be made with borehole logging devices or by tracer tests. Logging devices sample only a small zone near the borehole, and no quantitative analysis of total measurements uncertainty has been published. Tracer tests can be used only where the hydraulic conductivity is large enough that the tests can be done in a reasonable length of time. No quantitative analysis of test errors has been published. Typically, no test uncertainty is reported with field determinations of porosity.

Hydraulic Gradient. Uncertainties in hydraulic gradient are due mainly to (a) errors associated with the measurement of water levels in wells, (b) spatial variations in gradient due to hydraulic inhomogeneities in the rocks, (c) the time period required for water levels to stabilize in rocks having very low permeability, and (d) the gradient changes due to man's removal of water, oil, and gas that may or may not result in permanent gradient changes.

542 267

Geophysical Exploration and Drilling Techniques

Seismic Reflection and Refraction. To study subsurface characteristics, seismic techniques are often used which include sending an acoustic pulse or wave train into the ground and measuring the resultant reflection or refraction of the pulse. The reflections or refractions are caused by acoustic impedance discontinuities in the ground and they can yield a two-dimensional profile of the subsurface. Measuring seismic reflection gives more detail than measuring refraction, which is generally used for reconnaissance purposes only. Seismic techniques are most useful in locating subsurface structural or stratigraphic anomalies, and such anomalies can then be drilled for more accurate characterization. Seismic techniques do not endanger the integrity of potential repository layers.

Radar. Radar techniques, specifically designed to explore relatively dry rocks such as salt, have successfully detected small water hazards, such as a brine-filled borehole, at distances of 50 to 80 m. One fault was located 400 m from a mining face. Radar probing is most accurately performed from mines. Borehole radar devices have been tested, but their usefulness is limited, and directional control has not yet been developed. Radar probing from the surface is not feasible because the radar beam is severely attenuated by any soil or rock strata containing more than about 0.5% water by volume.

Electrical Resistivity Surveys. Electrical resistivity surveys are made by modeling a best fit between the field data and an assumed subsurface resistivity. The results are characterized by non-unique and commonly oversimplified solutions. Resistivity methods have performed fairly well in detecting near-surface features as large as 700 m across, but they have failed to detect potential hazards such as vertical columns of breccia, at depths of a few hundred meters. A target whose depth is greater than its lateral dimensions cannot be detected. Thus, a spheroidal brine cavity generally cannot be detected from the ground surface.

Gravity Surveys. The resolution of gravity surveys is severely limited by variations in topography. Minor variations, of the order of several meters, obscure the gravity anomalies generated by small targets at any depth.

Therefore, gravity surveys appear to have little value for detecting local repository hazards.

Magnetic Surveys. Magnetic surveys, like resistivity and gravity surveys, are based on a modeling process and yield inherently non-detailed information about the geometry of subsurface structures. Because they respond principally to contrasts in the iron content of rocks, magnetic surveys are most useful for detecting igneous dikes and other intrusive features in sedimentary basins. They are generally not effective in detecting anomalies such as breccia pipes or brine cavities because these features do not create a magnetic contrast with the surrounding rocks.

Drilling Programs. Drilling is not useful for detecting local hazards because the probability of hitting a small target is low for reasonable drilling grid spacings. Drilling is most useful for characterizing potential hazards, once their presence has been indicated by remote survey techniques. Drilling has the inherent limitation that it creates a potential leakage path for radionuclides.

Uncertainties Associated With Exploration and Exploitation of Mineral Resources

Features associated with resource exploitation in bedded salt basins include petroleum exploration and production wells, mineral exploration boreholes, brine wells, solution cavities, mine shafts, and underground excavations. Evaluating the hazards to a repository posed by each kind of feature has two aspects: determining the potential effects of a given feature, and determining the location of existing features or the likelihood of unknown features occurring near the repository site. The former problem can be approached by examining case studies, which can, at least, indicate a probable maximum radius of influence for each feature. The difficulty experienced in locating exploitation features and adequately demonstrating the absence of unknown features depends on several factors: the kind of feature, the local exploitation history, and the state in which the study is being made. The danger of known features can be minimized by placing a repository far outside their maximum observed radius of influence. More danger is posed by unknown features.

542 269

MODELING METHODS

OVERVIEW

The backbone of long-term risk assessment is the systems model, or Long-Term Risk Model (LTRM). It is an assemblage of submodels that correspond to various barriers to the migration of waste from the repository to the biosphere. These submodels are dissolution, recharge/repressurization, local migration, regional migration, and dose. The dose model does not actually represent a real barrier, but is a way to translate release into biological effect.

Systems models are the principal instruments in the definition of that portion of parameter space which leads to acceptable performance. To support and validate systems models, more detailed models are constructed.

Figure 12 illustrates the relationship among the various system models in the LTRM.

During FY 78 revisions in the systems models were planned so that they will use probability distributions as input and output.

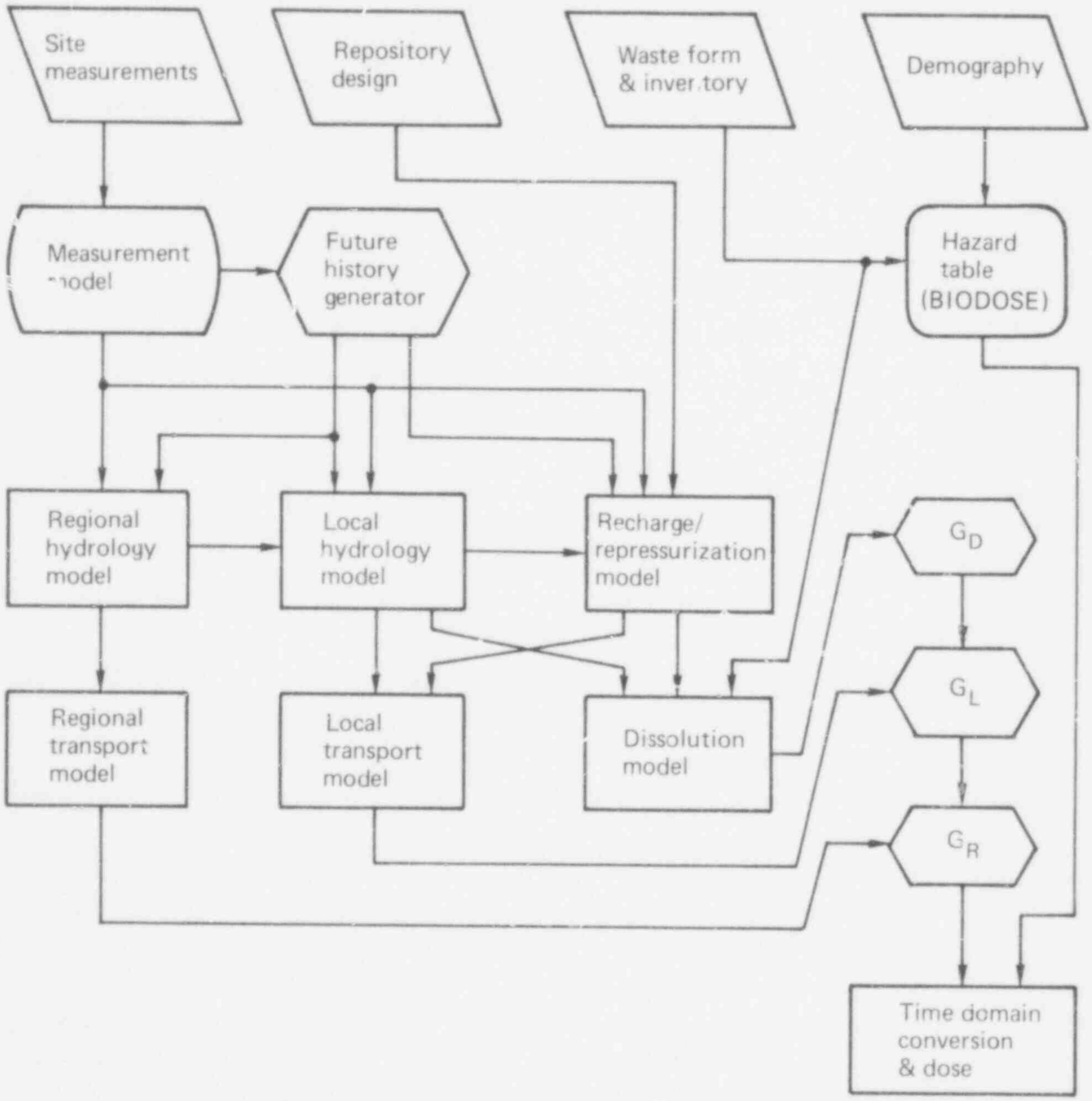

FIG. 12. Proposed organization of Long-Term Risk Model
(Note: G stands for Green's Function).

542 271

UNCERTAINTY ANALYSIS

Development of a Probabilistic Calculation Technique

We worked to bring the overall repository performance modeling activities into sharp focus. Progress is reflected in three documents that are synopsized here.

The first report[28] presents a conceptual framework for the definition and study of long-term risk. A methodology is presented for the propagation of uncertainty through the LTRM. The LTRM is expressed as a convolution of Green's functions.

The second report[29] clarifies the idea of a finite probability distribution (FPD) and details the algorithms required for performing all neccessary operations involving FPD's. An analogy with ordinary arithmetic and calculus operations is pointed out. These algorithms constitute the basic computational methodology required for handling uncertainty in the prediction of performance of waste disposal systems.

The third report[30] presents a blueprint or flow chart (Fig. 13) of the procedures and software modules required to predict repository performance according to the approach outlined in the previous two reports. These three reports are expected to serve as the basis for the development of a new aggregated or macro model during FY 79.

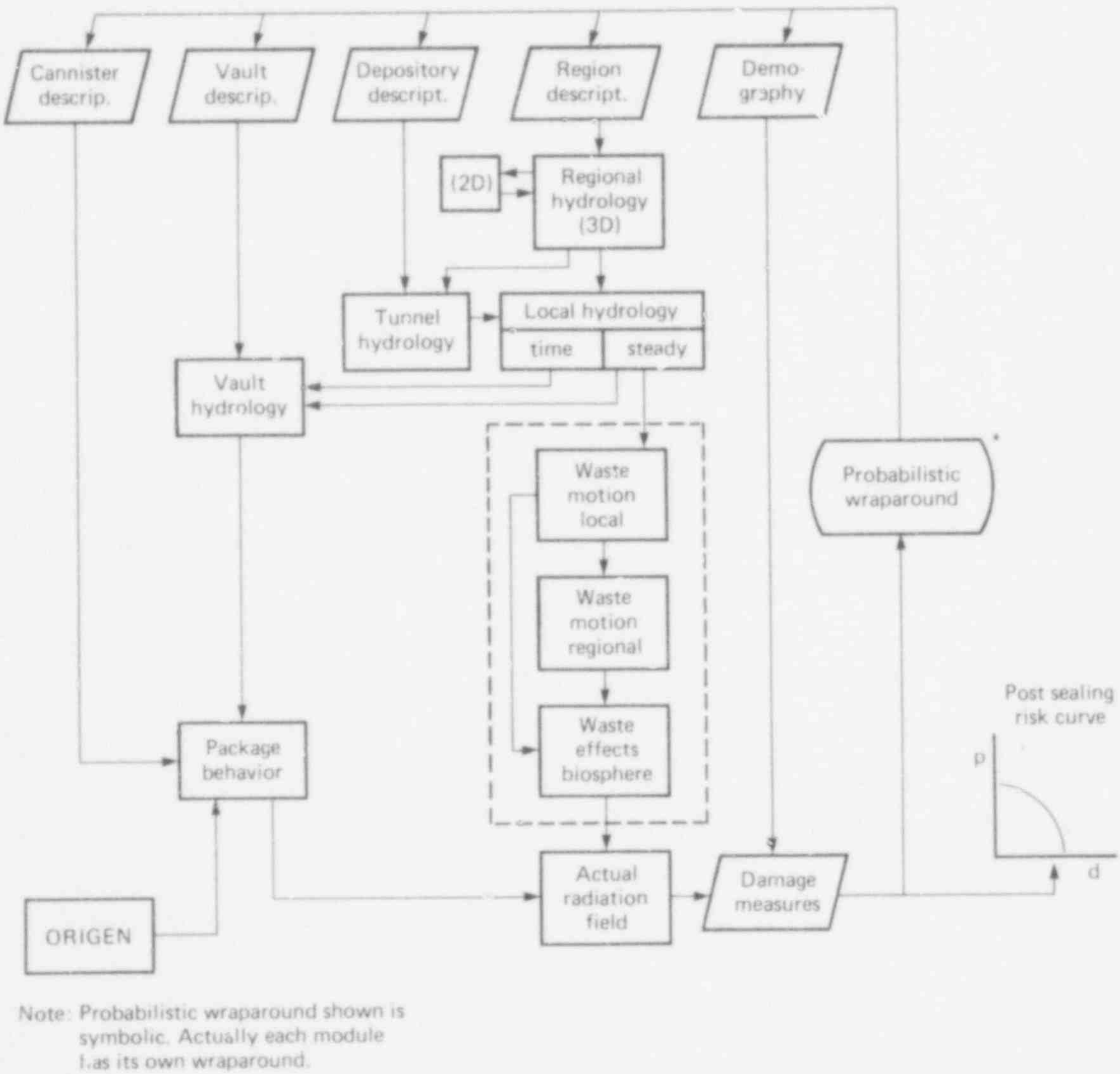

FIG. 13. Computations system flow chart.

542 273

Monte Carlo Analysis

Monte Carlo techniques were used to perform a parametric uncertainty analysis of the NUTRAN simulation system, a long-term risk model.[1,7,31] It is assumed that as part of the repository site selection process approximately 50 boreholes will be drilled in the vicinity of a candidate site to estimate hydrogeologic and geochemical characteristics. Considerable uncertainty in estimates of site parameters can result when the amount of data is limited. Such parameters as effective permeability of the overlying aquifer can be estimated to within a factor of ten at best. Hence, the performance predictions for the repository based on such simulation models as NUTRAN are similarly uncertain. The Monte Carlo analysis evaluates the effect of site parameter uncertainty on the uncertainty of the repository performance predictions.

The repository is characterized both by engineered and natural factors that determine its hydrology and thus the patterns of waste-migration flow. Also required for evaluating repository performance are models for nuclide inventories, dose pathways, and radionuclide intake by human populations. Although all these model components are uncertain, the Monte Carlo analysis during FY 78 considered only the repository site characteristics that were considered to be random variables specified by probability distribution functions (PDF's) that reflect the parameter estimation uncertainty. A computer program, WSRAND, was developed to generate random site characteristic parameters according to specified PDF's. Sets of random site characteristic parameters were then put into the NUTRAN simulation system and a statistical analysis was performed on the ensemble of outputs.

Monte Carlo studies for shale and bedded-salt repositories have provided information about the nature of the performance prediction uncertainty. Figure 14 presents a typical normalized histogram and sample statistics of integrated population dose (IPD) for 50 Monte Carlo trials. The repository is assumed to be an unflawed shale repository with interstitial flow containing 6×10^6 MWe-y of waste.

542 274

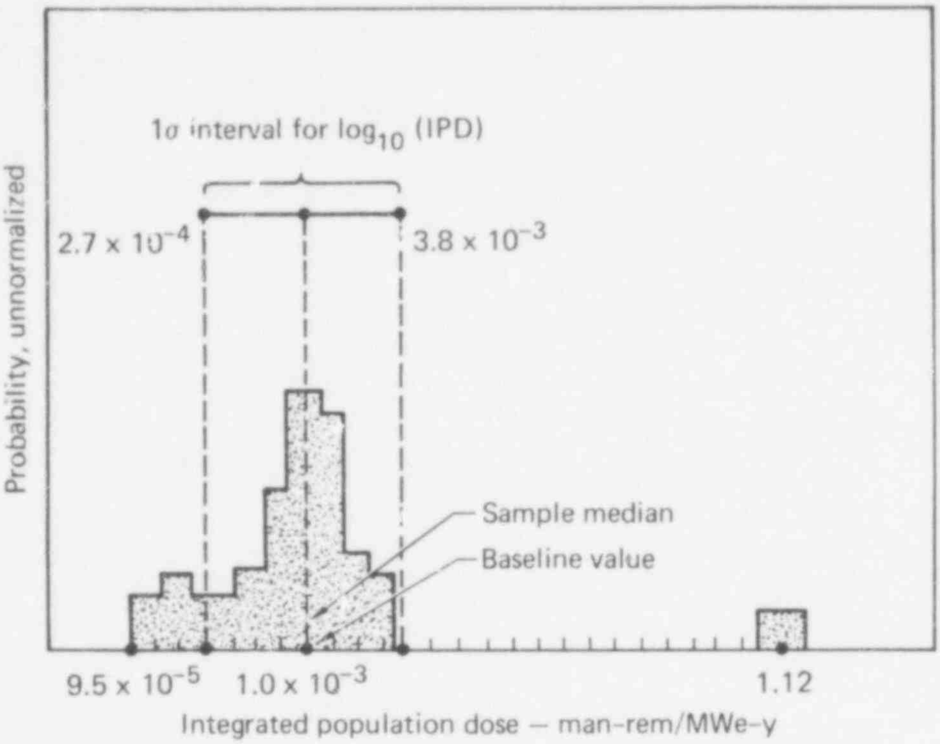

FIG. 14. Sample histogram and statistics of integrated population dose IPD for 50 Monte Carlo trails.

Refinement of the assumed input distributions for the site characteristics will yield improved performance prediction distributions.

542 275

FUTURE-HISTORY MODELING

LOSS OF ADMINISTRATIVE CONTROL (LOAC)

We began future-history modeling by developing a preliminary framework for the evaluation of the potential risks associated with LOAC.[32] A key feature in such models is the way that state changes are handled as required by changes caused by future events.

DISCRETE-EVENTS VS MARKOV-CHAIN MODELING

Workers at the University of California, Riverside, examined the early approch used for mathematical modeling of the post-emplacement risks associated with nuclear waste disposal. The model is intended to produce a probabilistic description of the migration of waste material out of the repository and through the aquifer. Results indicate that the proposed Markov-chain model for estimating expected concentration over time does not adequately describe post-emplacement risks and that the model does not reflect the geologic characteristics of the repository because of the limitations imposed on both the state space and transition probabilities.

A discrete-events model may be more appropriate and more manageable than the Markov chain model, and it may not involve the computational difficulties as originally thought. It would provide more information than just a look at the expected or average concentrations, although this type of information would also be available. It would allow study of specified histories and could therefore aid in determining consequences of a particular order and timing of events, and in determining key factors in evaluating post-emplacement risks.

Appendix D contains the analysis of the two modeling approaches. As currently envisioned, a discrete-events type of model will be used.

LOCAL WASTE MIGRATION MODELING

Local migration models are being developed on the basis of baseline repository designs to describe the geological barrier presented by the repository vicinity. The output of these models will be a source term for the regional migration models. Recharge modeling involves waste wetting times, the initiation time for steady-state flow through the repository, and the assessment of transient waste transport.

SHAFT STUDIES

Three-dimensional analytic modeling of hydrology provided a better understanding of the degree to which the interaction of the shaft, repository, fracture zones and undisturbed rock influence the transport of radionuclides from a repository to an overlying aquifer through shafts, boreholes, and the surrounding rock.[31]

Two modeling approaches have been used. The simpler approach models a vertical shaft and concentric fracture zone in a single layer. To partially account for the influence of other layers on the hydrology of the modeled layer, the flow in these layers was assumed vertical, with the magnitudes of the flow rates computed as if the modeled layer's flow were predominantly vertical as well. In addition, the following assumptions were made in order to define the problem:

- The gradient is purely vertical (far from the shaft).
- The shaft and fracture zone do not continue up into the layer overlying the modeled region. Thus, the vertical component of flow at the interface between these two layers is assumed continuous in the radial direction.
- The effect of confined flow when no horizontal gradient is included in the model is approximated by two layers: one immediately above the

542 277

model layer, the other layer at some lower level, which exhibit no vertical gradient. These regions serve as a water source and a water sink.

The second approach models the entire multi-layer region. Thus it does not assume any vertical predominance of flow. It does assume that the three conditions above still hold. Zero-gradient layers were assumed to be present immediately above and below the modeled region. Anisotropy can be easily incorporated into either of the models.

The solutions for the head take the form of a term linear in z (vertical depth) plus an infinite sum of functions whose z dependence is sinusoidal and whose radial dependence takes the form of Modified Bessel Functions.

COMPARISON OF ONE-AND TWO-DIMENSIONAL MODELS

To evaluate the inability of a one-dimensional groundwater model to interact continously with surrounding hydraulic head gradients, simulations using one-dimensional and two-dimensional groundwater flow models were compared.[33] This approach used two types of models:

- Flow-conserving one-and two-dimensional models.
- A one-dimensional model designed to yield a two-dimensional solution.

The hydraulic conductivities of controlling features were varied and model comparison was based on the travel times of marker particles. The solutions within each of the two model types compare reasonably well.

REPOSITORY RECHARGE MODELING

We developed several simple recharge models to evaluate the components of resaturation and repressurization of the repository after decommissioning. The models and evaluations completed in FY 78 include the following:[25,26,32,34,35]

- An evaluation of recharge through the rock mass assuming an average steady-state flow but considering the effects of entrapped air. The evaluation was validated against a case study that involved flooding of a mine. In general, it was concluded that entrapped air would have little effect in a deep repository.
- An evaluation of the transient recharge and repressurization behavior through the rock mass only. It was found that the time required to re-establish the original gradients (repressurization) after saturating the repository could be significant.
- An evaluation of recharge through horizontal permeable layers only in an otherwise impermeable bedded salt. In general, it was found that the recharge time would be tens of thousands of years for any reasonable set of assumptions.
- An evaluation of recharge through the shaft(s) only. It was found that it may take considerable time, depending on the tunnel backfill properties, for the saturation front to move through the repository.

In previous analyses, the value of repository recharge time was simply assumed to be a baseline value of 100 y. To provide better understanding, a method for relating recharge time to the existence of permeable pathways between the repository and the adjacent aquifers was developed.[7]

In this approach, the amount of water required for recharge is the sum of two terms: (1) the void volume of the repository and (2) the increase of the water content of the adjacent rock due to repressurization. The inflow to the repository at any time is proportional to the pressure difference between it and the aquifers. The pressure in the repository is assumed to be atmospheric until its void space is filled and then to increase linearly with the amount of water absorbed by the rock.

For the baseline repositories[11], with repository layer permeabilities of 10^{-9} cm/sec, recharge times calculated by this method are typically hundreds of years. If, however, the permeability of salt is 10^{-12} cm/sec or lower, recharge times of tens of thousands of years result.

REPFLO MODEL

A computer program, REPFLO, was developed to simulate the groundwater flow and nuclide transport within the repository's storage rooms, corridors, and shafts.[32, 36] In essence, the model consists of a three-dimensional matrix of Darcy-flow pipes that simulates the repository and connects to an upper and a lower aquifer.

At present work is underway to evaluate the REPFLO Model and to recommend improvements. To perform the calculations, it is necessary to have a subroutine to solve a set of linear algebraic equations characterized by real, symmetric, positive-definite banded matrices. This is a proprietary subroutine that was not supplied. Thus a subroutine was developed and tested using a trial problem. It reproduced the earlier result. In the case run, migration is not calculated, but the plan is to continue work to incorporate the migration section of the code and to study the underlying physical assumptions and their internal consistency. Close attention will be paid to the extent to which the migration section can be treated as a module. If necessary, the code structure will be changed to make this modularization possible.

In the course of performing the above work, several questions arose that will be investigated in the near future. These include the following:

- To what extent can the relationship between nodal flow and drawdown head be inferred from two-dimensional calculations applicable to repository units?
- Should the Newton-Raphson solutions be iterated, even when it is assumed that the nodal flows are linear in drawdown?
- Is the Jacobian matrix sufficiently ill-conditioned that rescaling may be advisable?

542 280

- The pipes connecting repository units are assumed to be perfectly impervious in REPFLO. Under what conditions is this assumption justified, and are those conditions satisfied always, sometimes, or never in situations of interest?

We believe that the entire question of dispersion demands much closer study, since this subject is not at all well understood at present. Questions that should be answered include:

- What is the interplay between hydrodynamic dispersion and diffusion in problems involving time scales of centuries or millennia and length scales of many kilometers?
- Is the concept of retardation coefficients meaningful in the present context?
- Are Gaussian reapportionments at intersection nodes reasonable?

PRELIMINARY EVALUATIONS

Preliminary evaluations of various phenomena and factors were performed to determine the relative significance of each and to assess the need for more detailed local waste-migration modeling.[25,32] These evaluations are briefly discussed below.

Non-Darcy Flow Behavior

Groundwater flow in thick, deep geologic formations may not necessarily be related simply to the hydrologic gradient as described by Darcy's law. The actual gradient available to produce flow is complicated by salinity (density) variations, thermal effects, possible osmotic or ionic affects, and the possible existence of a threshold gradient in some rock types. Flow in fractured media, especially sparsely fractured media, may not be adequately modeled using flow models based on Darcy flow. These problems need further study and evaluation.

Steam and Thermal Convection Pathways

Due to the heat generated by the waste, there is a potential for the production of steam and convection groundwater flow. Based on a preliminary evaluation, it appears that steam and thermal convection cells may be

542 281

produced, but this production would probably not result in any significant radiation release. Additional work is needed to confirm this preliminary conclusion.

Earthquake Effects

A preliminary evaluation based on available information and empirical data indicates that there should be minimal effects of earthquake shaking during repository operation. After decommissioning, earthquakes should have virtually no effect unless the event is caused by fault movements at or near the repository.

542 282

REGIONAL WASTE-MIGRATION MODELING

Generic hydrology models are being developed for each siting medium. These models will be capable of describing specific sites by appropriate adjustment of parameters. The results of exercising the detailed generic models will guide revisions of the systems level model. Hydrology models are a necessary basis for studying the transport of radionuclides from the repository vicinity to the biosphere. Transport models will be used to study the variation of regional barrier performance as a function of repository location.

BASIN MODELING

Generic and Numerical Models

A generic model was developed for a large sedimentary basin.[37] The basin's stratigraphy was divided into five vertical hydrologic units. Moving downward from the surface, the units are

- An aquifer, 170 m thick with a hydraulic conductivity, K = 200 m/y.
- An aquitard 136 m thick, K= 20 m/y.
- An aquiclude 170 m thick, K= 1 m/y.
- An aquitard 136 m thick, K= 20 m/y.
- An aquifer 136 m thick, K = 20 m/y.

A numerical model was designed to simulate the hydrologic condition of the model basin. The model contained 1656 nodal points defining 1562 elements. Numerically, the recharge region was treated as a distributed inflow boundary. The groundwater divide, the divergence between basin and regional flow, and the symmetry plane at the discharge region were modeled as zero flow boundaries. The discharge region was simulated by a boundary condition where pressure equals zero. By designating these conditions, groundwater was free to leave the model.

542 283

A steady-state two-dimensional velocity field was calculated for the model. The velocity field, along with other input, was used for computer-code calculations of material transport.[38]

Material Transport

The input data controlling material attenuation are flexible enough to represent a wide range of stratified media. Because of the model geologic basin size, uncertainty is great in those parameters controlling material transport. As a result of the model's generic nature, the transport properties assigned to the geologic media will be required to conform with experimental data as they become available, but such modifications are easily made.

Analyses of Repository Locations

The release of material from a repository was simulated with a constant release point source. The treatment of repositories as point sources was justified because of the large size of the basin relative to the probable size of a repository.

From this study, 10 point sources were positioned within the cross-section of the basin to determine the scope of the movement of material away from the source during the first 10,000 y of simulation.

Future Work

Future work will include selection of reasonable repository locations and simulation of transport for 100,000 y or longer. An S-shaped waste breakthrough curve at the discharge point will be the result of the simulation. The shape of the curve and the breakthrough time are dependent on the repository's position and on transport parameters. Varying the model's input, i.e., dispersion, diffusion, etc., will produce a family of break-through curves for each repository site. After analyzing the break-through curves, statements can be made about the suitability of a particular site under given geohydrologic conditions.

GENERIC MODELS

Baseline Case Generic Geologic and Hydrologic Models of Bedded Salt, Salt Domes, Shale, Granite, and Basalt

Based on the data base information, we built preliminary generic models for each rock type.[20,39] These models incorporate features commonly encountered in the particular geologic setting with appropriate ranges in parameter. The intent is to present models simple enough to be synthesized into a long-term risk model, while also enabling realistic geologic and hydrologic conditions to be evaluated.

Generic Flow Path Models of Basalt, Granite, and Salt Domes

Based on the generic geologic and hydrologic models, we also developed preliminary flow path models. The purpose of these model[illegible] to develop new baseline cases for the current LTRM. Generally only unfl[illegible] flow path models were developed, with appropriate comments on the more sig[illegible]t flows that need to be evaluated. We constructed models for a repository located in basalt.[40] Two baseline cases were established because the large horizontal flow component in the basalt model makes it difficult to predict the distance from the river to the point where the flow path enters the upper aquifer. In one case, the flow path from the repository went directly through the basalt layer into a river. The other flow path included a 16 km upper aquifer before reaching the river. In both cases the repository was assumed to contain spent fuel, and flow through the shaft was neglected.

FUTURE CLIMATE AND GROUNDWATER

Because groundwater movement can have important effects on buried nuclear wastes, hydrologists need to know if future climatic changes will influence the accuracy of groundwater flow calculations. Groundwater recharge (and therefore groundwater flow) depends on surface water balance. (Surface water balance equals precipitation less losses to evaporation, runoff, and storage).

To develop input data for modeling future climatic effects, we have made the following simplifying assumptions:

- Climate (and therefore water balance) will behave in the future very much as it has in the past.
- Groundwater recharge responds linearly to preciptation.
- Future long-term climatic changes can be classified into groups or regimes that are similar to those of the past.

(For a more complete statement of progress and a summary of the methods used to provide the LLL project with climatic input, see Ref. 41).

The problem of hydrology and climate change has recently received more attention[42] and the entire question of climate and its effects on waste management is gaining momentum.[43] The most recent update of work is a proposal by the University of Arizona Laboratory for Tree Ring Research. The laboratory has calibrated a model of ring width and its relationship to various climatic parameters. A history of climatic variation over the past 350 y has been constructed using this model. This period contained an important cool climatic regime (a Little Ice-Age) and a similar regime has a high probability of recurring within the next few hundred years. However, this projection excludes anthropogenic influence.

The Laboratory of Tree Ring Research will provide LLL with seasonal temperature and precipitation means, maxima, and minima for a series of 25 y periods between 1602 and 1900. This material can be used to calculate water balance. We expect that this data will exhibit a higher degree of climatic variability than is usually assumed for this period.

We have selected several particularly interesting geographical regions in the West and Midwest for climatic reconstruction. The data will be used to refine earlier climatic reconstructions and to predict future climate.

RELEASE TO BIOSPHERE

ALTERNATE WATER SYSTEMS

The reference river system used in the BIODOSE code closely resembles the Columbia River Basin (Ref. 1, Appendix G). To understand the sensitivity of the potential hazards calculated from this reference system to the choice of the Columbia as its basis, other water systems are being analyzed:

- An eastern river
- A large fresh-water lake
- A desert river
- A large mid-continental river
- An aquifer discharging directly to the sea

APPROXIMATE METHOD OF RISK CALCULATION

A simple procedure has been developed for approximating the radiation dose release calculated using the more rigorous LTRM model.[32] This method characterizes each flowpath segment by a flow time, or mean passage time of contaminants, and a dispersion time, or contribution to pulse width. For consecutive segments, the flow times are additive and the dispersion times add in quadrature. For a repository in a sedimentary basin, this approach indicated the following:

- The total integrated population dose over all time is essentially constant for all repository performance conditions, provided that the waste escapes between approximately 400 y and 3×10^6 y.
- For short waste release times, the canister dissolution behavior has a significant effect on population dose.

542 287

* If background radiation is considered for comparison, a well-designed repository results in integrated population doses well below the level of background radiation.
* For a host rock dominated by fracture flow and subjected to a vertical gradient, the main containment provided by the host rock is a long resaturation time. Transport time out of the layer would be very brief, except for nuclides with very high retardation.
* An overlying aquifer provides a significant barrier to waste release unless it is used as a groundwater resource.

DOSE OUTPUT FORMAT

Contours

Using a waste release function parameterized by mean arrival time and pulse width, we used the BIODOSE code to generate contours of dose for spent fuel. Figure 15 shows contours of peak 50-y whole-body-equivalent dose to an average individual from waste from the uranium-reprocessing, plutonium-stowaway fuel cycle. The contours are calculated on the assumption that all radionuclides move at the same velocity. Using such contours we can relate the performance of the site and repository as expressed by mean arrival time and pulse width to measures of hazard as expressed by dose.

542 288

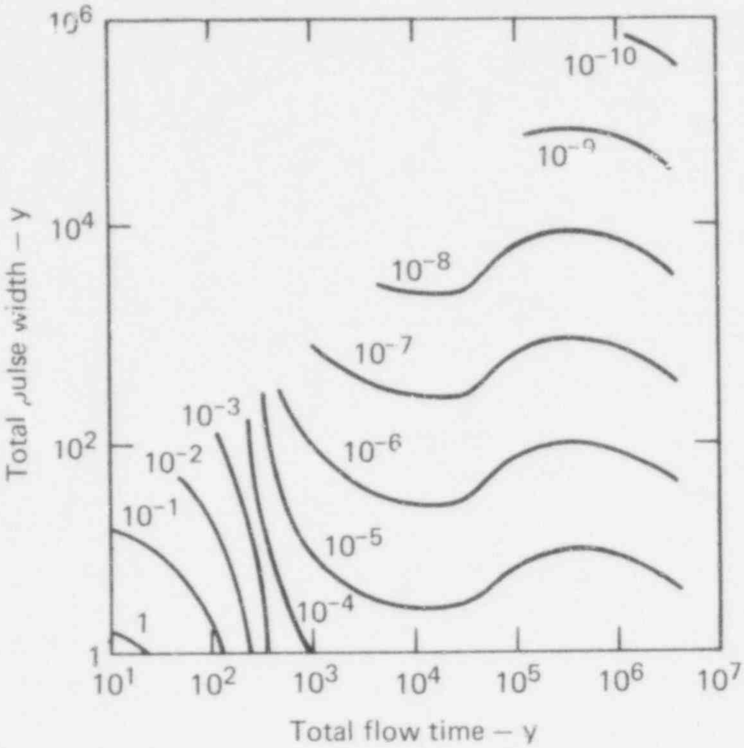

FIG. 15 Contours of equal peak individual dose (rem/MWe-y).

Lumped Parameters

We found that the various geologic parameters affect the movement of radionuclides in the model through two "lumped parameters". Changing one geologic variable is entirely equivalent to changing another if both lumped parameters are unchanged. These lumped parameters are:

$$\bar{z} = z/\alpha$$

$$T = \frac{\alpha B}{V}$$

Physically, T may be interpreted as the time required for the peak of the nuclide pulse to travel the length given by the dispersion constant α, and z is the length of the pipe divided by that dispersion length. Figure 16 shows peak dose to an average individual as a function of T for three repositories, which are described in Ref. 7.

542 289

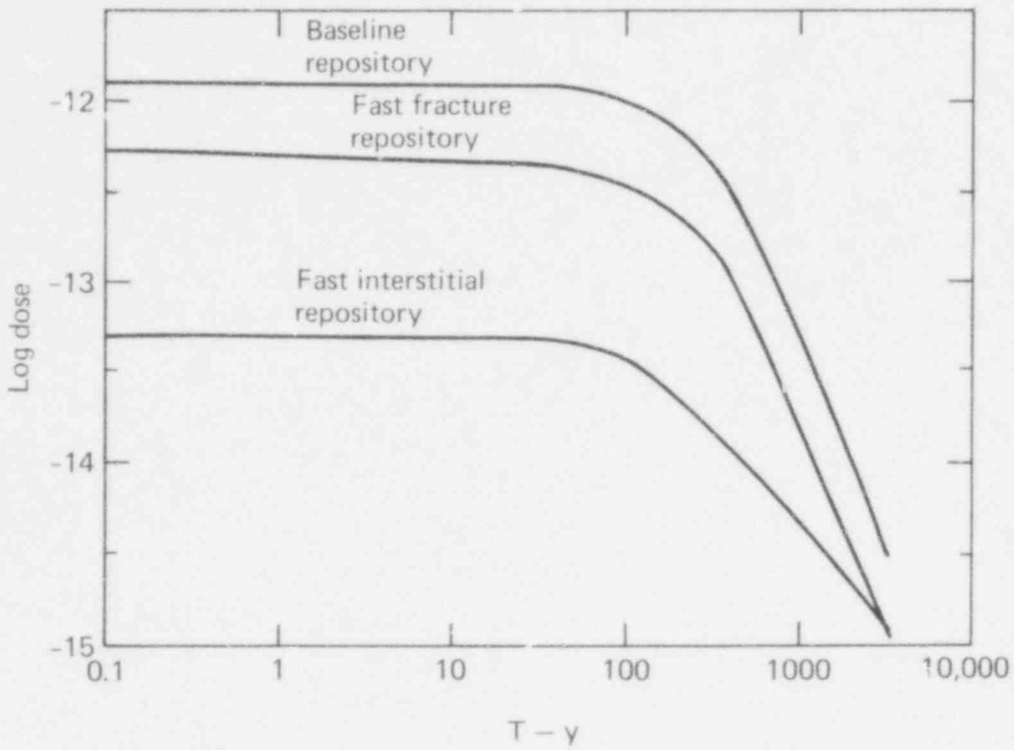

FIG. 16 Peak individual dose vs. lumped parameter, T.

542 290

Weighting of Doses to Organs by Cancer Risk

In order to simplify output and make results more meaningful, a method of computing net whole-body-equivalent doses was developed. These doses are calculated by summing the doses to each individual organ weighted by the cancer risk. The cancer risk factors, R(U), listed in Table 8, were developed from Ref. 44.

TABLE 8. Cancer risk factors, R(U).

U	Organ	R(U)
1	WHOLE BODY	0.451
2	GI-LLI	0.158
3	THYROID	0.034
4	BONE	0.034
5	LIVER	0.014
6	LUNG	0.223
7	KIDNEY	0.086
8	SKIN	0.0

This approach differs from that developed by the International Commission on Radiation Protection (ICRP)[45] in that:

- The ICRP considers both cancer and genetic risks.
- The ICRP calculation places an upper bound on the net whole-body-equivalent of certain organ doses, whereas this method estimates the equivalent for all organs.

542 291

TASK III: Repository Design Performance Criteria

The only part of Task III that was funded during FY 78 was in the area of thermal analysis. See "Spent-fuel repository thermal analysis" in the Task I section of this report.

TASK VI: DEVELOPMENT OF RADIOLOGICAL PERFORMANCE OBJECTIVES

This section is a brief summary of the development of radiological performance objectives (RPOs) for waste management systems (WMSs) by Task VI members and subcontractors during FY 78. Most of the work of the task is scheduled to be completed by November, 1978, at which time the final report for the task (UCRL 52574) will be available. Until then, the reader is referred to reports completed during the project if detail beyond that provided in the following summary is needed.

INTRODUCTION

RPOs are summary performance indices that are specifically designed to provide a defensible and understandable basis for regulations and reg guides. During FY 78 the task developed RPOs that can evaluate a WMS in a way that accounts for the probabilistic nature of its consequences and addresses several value-laden issues that are important to the determination of its acceptability. These issues include temporal risk allocation and occupational vs. non-occupational risk.

A Risk Evaluation Index was developed during FY 78 as a basis for generating RPOs. The Risk Evaluation Index is a single number designed to evaluate the overall social risk of a WMS. The Risk Evaluation Index is necessary for answering the following types of questions:

- Which of two WMSs is safer?
- How much safer is one compared to another?
- Is the risk due to a given WMS acceptable?

In brief, the Risk Evaluation Index provides a quantitative definition of 542 293
risk, a single scale on which risk can be measured and limited. The Risk Evaluation Measure incorporates social values where those values are

appropriate: value tradeoffs between different types of risk, and attitudes toward uncertainty. Incorporation of social values is important to the defensibility of any set of regulations regarding nuclear waste management.

The RPO task was divided into two segments, a short-term effort and a long-term effort. The short-term effort developed RPOs to meet the immediate need for RPOs on a timely basis. The long-term effort is based on Multiple Attribute Decision Analysis (MADA), a methodology identified in previous work[46] as one that could lead to applicable and defensible RPOs. The long-term effort developed a Risk Evaluation Index to be used to evaluate regulations and the resulting WMSs, and is using that Risk Evaluation Index as a basis for generating RPOs.

SHORT-TERM EFFORT

Originally known as the "Strawman RPO" effort, this development of RPOs was based primarily on existing nuclear regulations and radiation standards. This effort stressed an easy-to-understand, straightforward incorporation of probabilities. The work was completed in August, 1978, including two rounds of review and revision on the final report.[47] Among its conclusions are:

- Waste management methods and operations should be designed such that the predicted radiological exposure to a maximum individual is below suitable limits.
- The limits set for potential consequences should scale according to the probability of any event or combination of events that could lead to those consequences (i.e., as predicted consequences become more severe, the events leading to those consequences should become less probable).
- Suggested numerical limits for maximum individuals doses are:

  for annual dose limit

  $D = 0.5\ p^{-0.33}$,

  for lifetime 50 y dose limit

  $D = 25\ p^{-0.2}$,

  where D is in rem and p is the probability of occurrence.

- The numerical limits for average individuals are 1% of these limits. Fig. 17 shows these four limits.
- Where more than one alternative WMS meets the limiting criteria for individual dose, the collective dose should be predicted and an alternative WMS selected on the basis of as low as reasonably achievable (ALARA) principles.
- Doses to future generations should not be discounted (i.e., predicted doses at any time in the future are considered to be as serious as present doses).
- Predicted organ doses should be considered on the basis of equivalent whole body doses as given by ICRP #26.[48]
- Spatial allocation of risk should be done such that the total collective risk is no greater than twice that which would apply locally or to that population receiving the benefit.
- When the hazard potential of the waste repository becomes less than that of a 0.2% uranium-ore deposit, continued concern is not warranted.

Other work completed during FY 78 as part of the short-term effort includes: a review of the goals and objectives developed at NRC and EPA workshops and assessment of their applicability and rationale regarding development of acceptability criteria;[49] and investigation of optimum conversion factors for determination of health effects resulting from exposure to ionizing radiation. The BEIR committee[50] estimate of about 10^{-4} cancers/man-rem was found to be more consistent with observed data than are certain recent higher estimates.

542 295

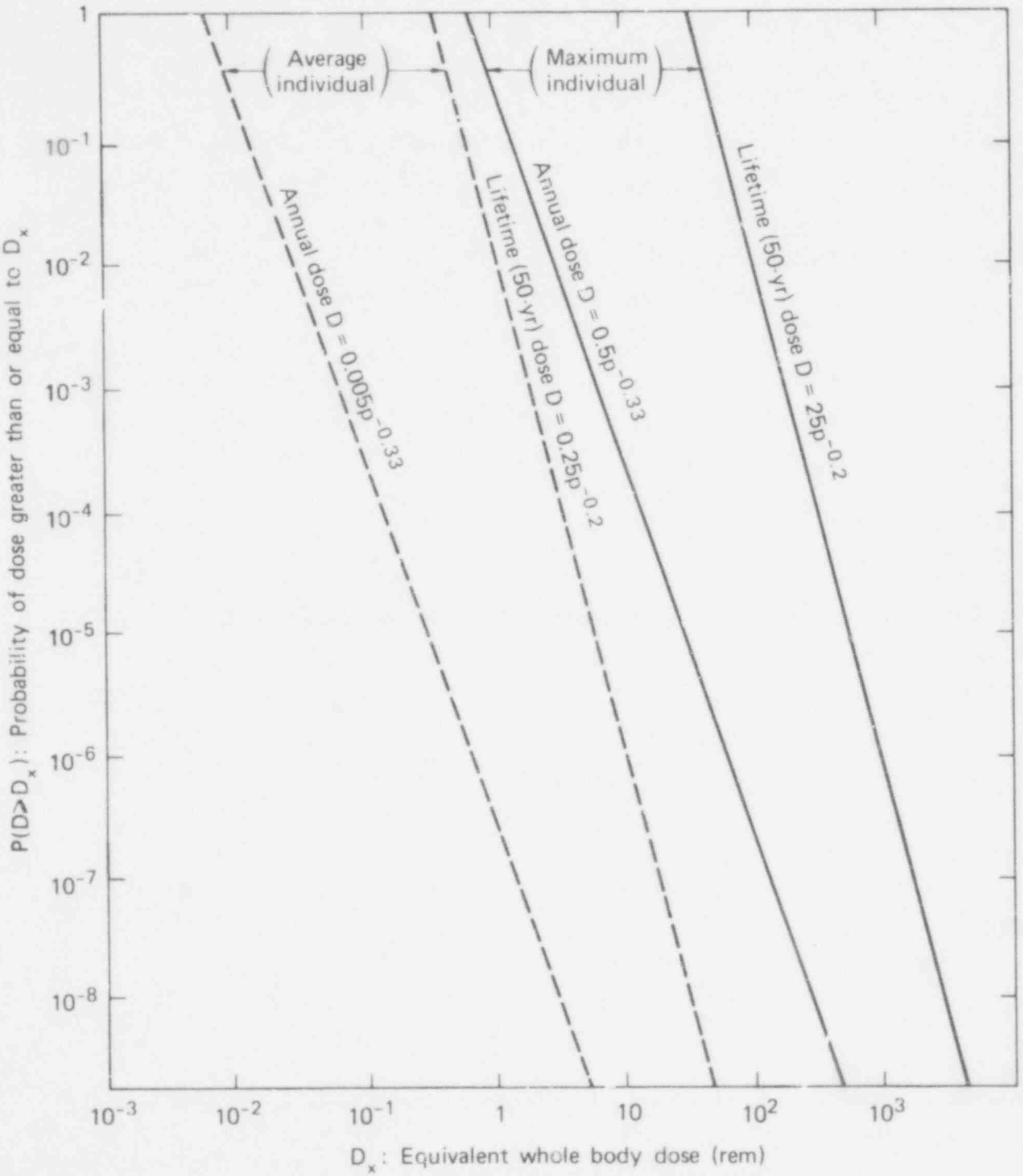

FIG. 17. Radiological performance objectives (individual whole-body dose).

542 296

LONG-TERM EFFORT

The long-term effort is developing both a Risk Evaluation Index and resulting RPOs based on an assessment of social values as elicited from people outside of NRC and LLL. This effort incorporates a more formally complete treatment of probabilities than did the short-term effort, though it is necessarily less easily understood. However, the MADA methodology is simple enough in concept to be described in terms of the following five steps:

STEP 1: DEVELOP THE FORM OF THE RISK EVALUATION INDEX

The Risk Evaluation Index should be expressed in terms of health effects, which people care about and can understand. Literature review and pilot studies indicated that the myriad of types of radiological health effects could most economically be partitioned into the five categories described along the bottom of Fig. 18. In addition, it was found that public values may be sensitive to the type of radiation exposure that caused the health effect. The most economical partitioning of exposure types that still captures the important value sensitivities is described along the bottom of Fig. 19. These two partitions together define a four-row, five-column twenty-element array of health effects. Each element of that array, x_{ij}, is simply the number of health effects of $i^{\underline{th}}$ exposure type, $j^{\underline{th}}$ health effect type.

The mathematical form of the Risk Evaluation Index and its parameters are given in Ref. 51. The Risk Evaluation Index accepts as input the probability distribution over health effect arrays that could result from a WMS and delivers as output a single number that is a measure of the social risk represented by the WMS. That measure can be scaled in terms of equivalent current occupational fatalities, or in terms of any other single dimension.[52,53] It follows that differences in the Risk Evaluation Index between two regulations or their resulting WMSs can be expressed in terms of equivalent lives saved, a very straightforward and meaningful measure.

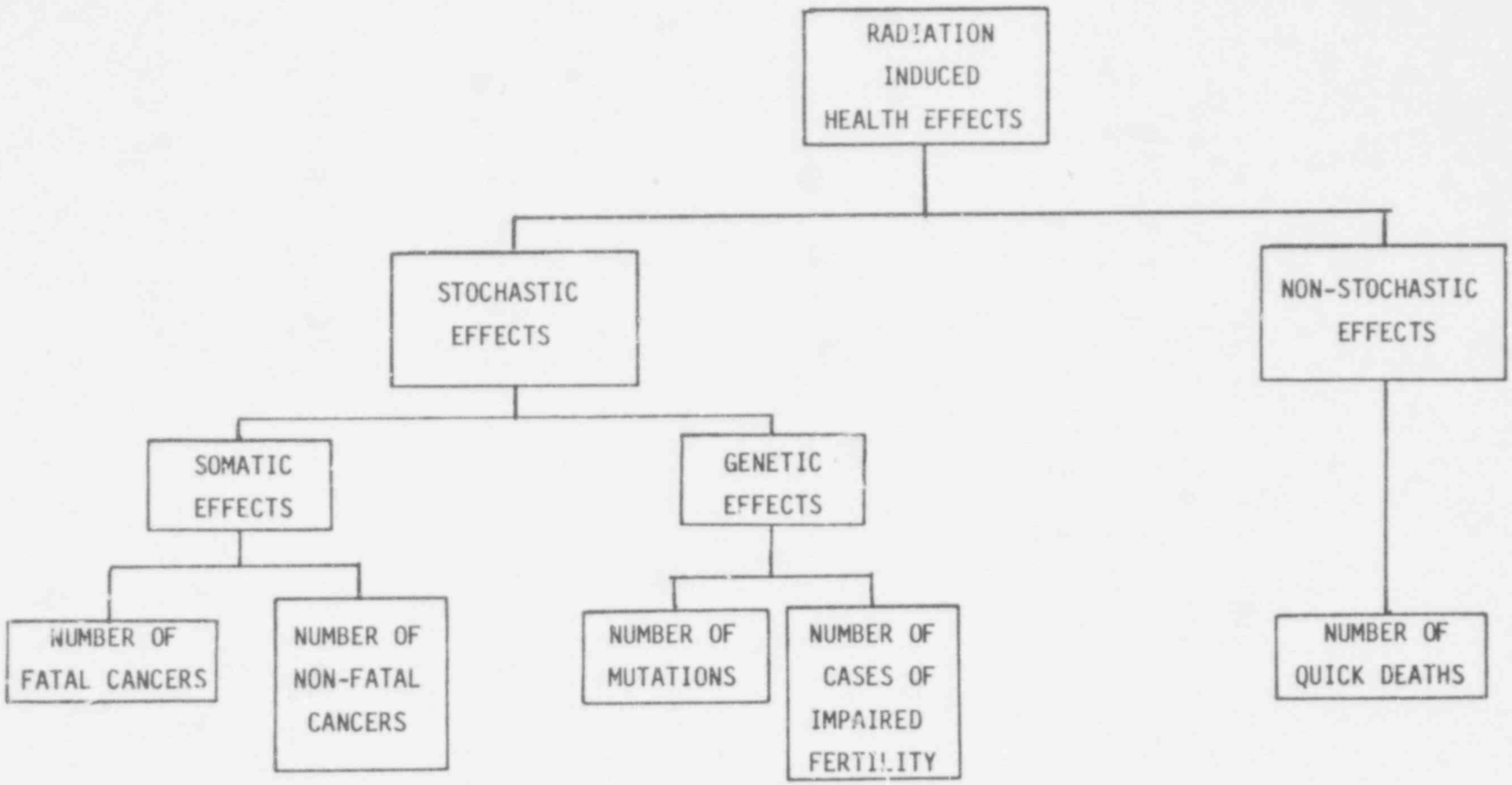

FIG. 18. Categorization of health effects.

542 298

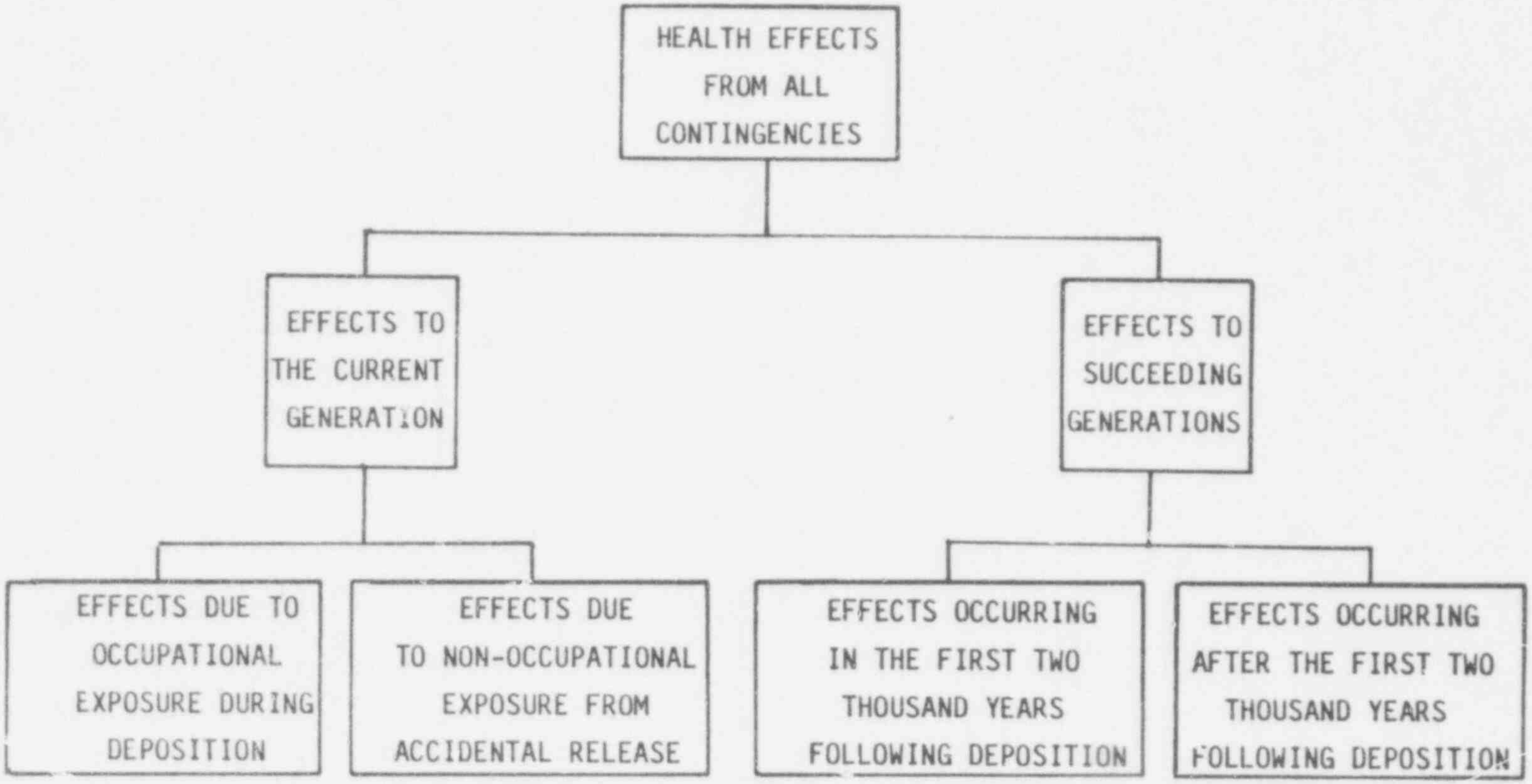

FIG. 19. Categorization of types of radiation exposure.

STEP 2: SELECT THE PANEL OF PEOPLE WHOSE VALUES ARE TO BE REPRESENTED BY THE RISK EVALUATION INDEX

Panel 1: National Advisors

A group of thirteen people was the primary source of values in this study. They were selected because they serve on a nuclear radioactive waste advisory body or are in some other way consulted on such matters by government policy makers.

Three other panels were elicited in an effort to lend some perspective to results obtained from the National Advisors panel:

Panel 2: Generally Active Citizens

This group was composed of 33 citizens who are somewhat abreast of social issues and who have taken identifiable steps to display concern that government actions reflect the general public interest. The sample was drawn from California, New Mexico, and Massachusetts. Absolutely no pretense is made that this sample is in any way representative of the citizens of those states.

Panel 3: Nuclear Power Advocates

This panel was composed of five people who have been identified as taking active stands in favor of further development of nuclear power.

Panel 4: Nuclear Power Opponents

This panel was composed of seven people who have been identified as taking active stands opposing further development of nuclear power, at least until questions of safety have been settled.

542 300

STEP 3: ELICIT THE VALUES OF EACH INDIVIDUAL PANEL MEMBER

The values were elicited during the course of an interview that involved a highly-structured set of questions. A description of those questions is included in Ref. 51.

STEP 4: AGGREGATE THE VALUES OF THE INDIVIDUALS INTO A SINGLE RISK EVALUATION INDEX.

The problem of aggregating individual values is a very difficult one in the field of social choice.[54,55] Although the method of adding utilities (as opposed to one of several other strategies) has definitely been decided upon, the details of calibrating the utility functions across people are still under development. Tentatively, cross-calibration has been achieved by setting equal each person's aversion toward 100 fatal cancers, since the fatal cancer seemed to be the most uniformly understood health effect. The calibration method under development will be included in the final report and may be much more involved than the method described above.

STEP 5: DEVELOP THE RISK EVALUATION INDEX INTO RPOs

The development of RPOs, which will be included in the final report for this task, will consist of bounds on the same sort of axes as shown in Fig. 1, except that the dose will be population dose. There will be three or four RPOs, one for each exposure type described in Fig. 3, with the two post-sealing exposures perhaps governed by the same RPO.

542 301

CONCLUSIONS

The most fundamental conclusions to be drawn from Task VI work, aside from the Risk Evaluation Index and RPOs themselves, are that RPOs incorporating uncertainty can be defined and that a Risk Evaluation Index incorporating uncertainty and based on social values can be developed. The task developed a meaningful single measure of the radiological risk associated with a WMS that is more comprehensive and sensitive to social value considerations than any simple dose-based performance measure. The usefulness of this Risk Evaluation Index lies largely in its structure, as opposed to its parameter values. The tradeoff values elicited from the panels are to be regarded only as reasonable estimates of the social tradeoff values to be included in the Risk Evaluation Index. It is strongly recommended that further elicitation work be done to improve those estimates and enhance their defensibility.

542 302

TASK VII: GEOSCIENCE PARAMETER DATA BASE HANDBOOK

The Geoscience Parameter Data Base Handbook provides the NRC staff with a concise, objective and representative sample of the data useful in assessing risks associated with nuclear waste repositories. Primary, secondary, and postulated data will be presented in sections that cover the parameters that describe the dimensions and geometry, the chemical environment, the hydrogeology, and the physical properties of emplacement media. Three reports were prepared during FY 78.[56,57,58] References 56 and 57 will be combined into a single handbook for the NRC during FY 79.

542 303

ACKNOWLEDGMENT

This report represents a team effort by all members of the Waste Management Technical Support Project at the Lawrence Livermore Laboratory (LLL):

Wendall A. Johnson, Program Manager
John H. O'Brien, Deputy Program Manager

Task I: Waste Form Performance

Henry Cheung, Task Leader
Thomas J. Altenbach
Billy W. Davis
Milton Fuller
Donald D. Jackson
Donald J. Kvam
Harris B. Levy
H. Robert Ralston

Task II: Site Suitability

Alfred M. Kaufman, Task Leader
Terry L. Steinborn, Deputy Task Leader
Dana J. Isherwood
John A. Korver
Thomas G. Naymik
Freda A. Phelps
Gerald L. Potter
Nancy Storch
Lewis D. Thorson
James L. Willows, Jr.

Task III: Repository Design Performance Criteria

Ted F. Harvey, Task Leader

Task VI: Development of Radiological Performance Objectives

John W. Lathrop, Task Leader
Jerry J. Cohen
Stein Weissenberger

Task VII: Geoscience Parameter Database Handbook

Donald F. Towse, Task Leader

Project Office Support:

Carol F. Winton

Margie J. Schmidt

Carol Lynne Naito

We also wish to thank the many subcontractors who provided expertise to the project:

The Analytic Sciences Corp. (TASC), Reading, MA

Decisions and Designs, Inc., McLean, VA

Geotechnical Engineers, Inc. (GEI), Winchester, MA

Golder Associates, Inc. (GAI), Kirkland, WA

Kaplan Associates, Inc., Los Angeles, CA

New Millennium Associates (NMA), Santa Barbara, CA

Pacifica Technology, Del Mar, CA

TERA Corporation, Berkeley, CA

We also wish to thank the Information Services Group at EG&G, San Ramon, CA, for their help in compiling this report.

REFERENCES

1. L. E. Berman, D. A. Ensminger, M. S. Giuffre, C. M. Koplik, S. G. Oston, G. D. Pollak, and B. I. Ross, Analysis of Some Nuclear Waste Management Options, The Analytic Sciences Corp., Reading, MA, TR-1103-1-1 (Jan. 1978).
2. H. Cheung, T. Altenbach, and S. G. Oston, A Thermal Analysis of Nuclear Waste Interim Storage Pools, Lawrence Livermore Laboratory, Livermore, CA, UCRL-52566 (in press, expected to be published in September 1979).
3. R. A. Heckman and T. Holdsworth, A Probabilistic Safety Analysis for Solidified High-Level Nuclear Waste Management Systems: A Status Report, Lawrence Livermore Laboratory, Livermore, CA, UCID-17878, covering work completed in October 1977 (July 1979).
4. A. L. Edwards, TRUMP: A Computer Program for Transient and Steady-State Temperature Distributions in Multidimensional Systems, Lawrence Livermore Laboratory, Livermore, CA, UCRL-14754 (1972).
5. TERA Corporation, Production of Solidified High-Level Wastes: A Cost Comparison of Solidification Processes, Lawrence Livermore Laboratory, Livermore, CA, UCRL-13740 (1977).
6. TERA Corporation, The Uranium-Recycle/Plutonium-Disposal Fuel Cycle, Berkeley, CA, Status report (Feb. 1978).
7. M. E. Hough, C. M. Koplik, G. D. Pollak, and B. I. Ross, Supplementary Analysis of Nuclear Waste Management Options, The Analytic Sciences Corp. Reading, MA TR-1103-1-2 (Mar. 1978).
8. M. T. Bell, ORIGEN--The ORNL Isotope Generation and Depletion Code, Oak Ridge National Laboratory, Oak Ridge, TN, ORNL-4628 (1973).
9. R. A. Brown, "Management of Tritium and ^{14}C," in Proc. Intern. Symp. Management of Waste from the LWR Fuel Cycle (Oak Ridge National Laboratory, Oak Ridge, TN, ORNL CONF-76-0701, July 1976), pp. 364-381.
10. R. English et al., An Analysis of the Technical Status of High-Level Waste and Spent-Fuel Management Systems, Jet Propulsion Laboratory, Pasadena, CA, JPL 5030-40 (Sept. 1977).

11. T. Altenbach, Interim Report on Nuclear Waste Depository Thermal Analysis, Lawrence Livermore Laboratory, Livermore, CA, UCID-17865 (1978).
12. R. E. Nickell, Retrievable Storage Concept Designs, Pacifica Technology, Del Mar, CA, PT-U78-0243 (Aug. 11, 1978).
13. W. Ludemann and R. D. McCright, Corrosion Studies on Retrievable Spent-Fuel Containers: A Progress Report, Lawrence Livermore Laboratory, Livermore, CA, UCID-17917 (December 1978).
14. E. S. Gaffney, Brine Migration, Interim Report, Pacifica Technology, Del Mar, CA, PT-U78-0242 (Aug. 11, 1978).
15. R. L. Bradshaw and W. C. McClain, Project Salt Vault: A Demonstration of High Activity Waste in Underground Salt Mines, Oak Ridge National Laboratory, Oak Ridge, TN, ORNL-4555 (1971).
16. TERA Corporation, Inventory and Sources of Transuranic Solid Waste, Lawrence Livermore Laboratory, Livermore, CA, UCRL-13934 (August 1978).
17. A Brief Survey of Solid Radwaste Management at ERDA Installations, Oak Ridge National Laboratory, Oak Ridge, TN, ORNL/CF-76/129 (1976).
18. H. B. Levy, Methodology for Estimating Accidental Radioactive Releases in Nuclear Waste Management, Lawrence Livermore Laboratory, Livermore, CA, UCRL-81661, Rev. 2 (January 1979).
19. Golder Associates, Inc., Preliminary Generic Repository Design Data for Site Suitability Considerations, Kirkland, WA (April 24, 1978).
20. T. R. Donich, A. M. Kaufman, G. D. Sanier, T. L. Steinborn, D. F. Towse, Additional Media Studies for Site Suitability Criteria, Lawrence Livermore Laboratory, Livermore, CA, UCID 17887 (August 1978).
21. D. Isherwood, Preliminary Report on Retardation Factors and Radionuclide Migration, Lawrence Livermore Laboratory, Livermore, CA, UCID-17551 (1977).
22. D. Isherwood, "Geochemistry and Radionuclide Migration," in Proc. Sym. Waste Management, 1978 (Tucson, Arizona, 1978), pp. 469-482.
23. D. Jackson, Solubility Mapping--A Novel Technique in Chemical Modeling, Lawrence Livermore Laboratory, Livermore, CA, Technical Memo WM-78-684 (August 8, 1978).
24. Golder Associates, Inc., First Report: Development of Site Suitability Criteria for the High-Level Waste Repository, Kirkland, WA (June 1977).

542 307

25. Golder Associates, Inc., Second Report: Development of Site Suitability Criteria for the High-Level Waste Repository, Kirkland, WA (November 1977).

26. Golder Associates, Inc., "Scoping Study of Salt Domes, Basalts, and Granites as Related to Site Suitability for Geologic Disposal of Nuclear Waste, Kirkland, WA (November 1978).

27. Geotechnical Engineers, Inc., Final Report: Uncertainties in the Detection, Measurement, and Analysis of Selected Features Pertinent to Deep Geologic Repositories, Winchester, MA (July 1978).

28. Kaplan Associates, Inc., Notes on the LLL/NRC Waste Disposal Study I--Analytical Framework, Los Angeles, CA, Report KAI-1 (March 1978).

29. Kaplan Associates, Inc., Notes on the LLL/NRC Waste Disposal Study II--Probabilistic Arithmetic, Los Angeles, CA, Report KAI-2 (May 1978).

30. Kaplan Associates, Inc., Notes on the Next Generation Macromodel--A Unified Computations System for Predicting Post-sealing Waste Repository Performance, Los Angeles, CA, Report KAI-4 (August 1978)

31. B. I. Ross, L. E. Berman, D. A. Ensminger, M. E. Hough, S. G. Oston, G. D. Pollak, Aspects of Nuclear Waste Management, The Analytic Sciences Corp., Reading, MA, TR 1309-1 (October 1978).

32. Golder Associates, Inc., Third Report: Development of Site Suitability and Design Performance Data Base for a High-Level Nuclear Waste Repository, Kirkland, WA (May 1978).

33. T. Naymik, An Application of a Method for Comparing One Dimensional and Two Dimensional Models of a Groundwater Flow System, Lawrence Livermore Laboratory, Livermore, CA, UCRL 52541 (August 30, 1978).

34. Golder Associates, Inc. Recharge Behavior, Kirkland, WA (April 24, 1978).

35. Golder Associates, Inc. Interim Report Results of Work Period of April 26 to June 7, Kirkland, WA (June 15, 1978).

36. Golder Associates, Inc. REPFLO Computer Program, Kirkland, WA (April 12, 1978).

37. T. Naymik, L. D. Thorson; Numerical Simulation of Material Transport in a Regional Ground Water Flow System, Lawrence Livermore Laboratory, Livermore, CA, UCRL 52556 (December 31, 1978).

38. T. Naymik, User's Manual for a Two-Dimensional Groundwater Flow Code on the LLL Computer Network, Lawrence Livermore Laboratory, Livermore, CA, UCID 17908 (August 30, 1978).

39. Golder Associates, Inc., General Basin Modeling for Site Suitability Work and Shortcomings in our Modeling Capability, Flow Committee Memos, Kirkland, WA, (June 26, 1978).
40. Letter from A. Sutherland, TASC, to A. M. Kaufman, LLL, Subject: Repository Located in Basalt, dated July 5, 1978.
41. G. L. Potter, Summary of Climatic Input for Waste Management Site Suitability Criteria and State of Progress, Lawrence Livermore Laboratory, Livermore, CA , UCID 17904 (May 3, 1978).
42. J. P. Bruce, "New Directions in Hydrometeorology", Bulletin of the American Meteorological Society 59 (3), 258-266 (1978).
43. F. K. Hare, "Nuclear Wastes And The Hydrologic Cycle", Bulletin of the American Meteorological Society 59 (3), 267-268 (1978).
44. The Effects on Population of Exposure to Low Levels of Ionizing Radiation, NAS-NRC Advisory Committee on the Biological Effect of Ionizing Radiations, National Academy of Science, National Research Council, Washington, D. C. (1972).
45. "Recommendations Of The International Commission On Radiological Protection," Annals of the ICRP, Volume 1, No. 3 (1977).
46. J. J. Cohen, Determination of Acceptable Risk Criteria for Nuclear Waste Management, Lawrence Livermore Laboratory, Livermore, CA, UCRL-52404 (October 21, 1977).
47. J. J. Cohen, ed., Suggested Nuclear Waste Management Radiological Performance Objectives, Lawrence Livermore Laboratory, Livermore, CA, UCID-17880 (August, 1978).
48. International Commission on Radiological Protection, Recommendations of the International Commission on Radiological Protection, ICRP Publ. 26, (Pergamon Press, Oxford, 1977).
49. J. J. Cohen, Review of Issues Relevant to Acceptable Risk Criteria for Nuclear Waste Management, Lawrence Livermore Laboratory, Livermore, CA, UCRL-80793 (February 22, 1978).
50. National Academy of Sciences Advisory Committee on the Biological Effects of Ionizing Radiation (BEIR), The Effects on Populations of Exposure to Low Levels of Ionizing Radiation, National Research Council, Washington, D.C. (Nov. 1972).

542 309

51. S. R. Watson and V. N. Campbell, Radiological Performance Objectives for Radioactive Waste Derived from Public Values, Decisions & Designs, Inc., McLean, VA, Final Report PR 78-10-80 (August, 1978).

52. J. W. Lathrop, Measuring Social Risk and Determining Its Acceptability, Lawrence Livermore Laboratory, Livermore, CA, UCRL-81060 (July, 1978).

53. J. W Lathrop, ed., Development of Radiological Performance Objectives, Interim Results: Trade-Offs in Attitudes Toward Radioactive Waste, Lawrence Livermore Laboratory, Livermore, CA, UCID-17925 (July, 1978).

54. J. C. Harsanyi, Rational Behavior and Bargaining Equilibrium in Games and Social Situations, Cambridge University Press, 1977.

55. H. W. Brock and G. L. Sauer, Procedures for Aggregating Citizen Preferences in the Context of the Nuclear Waste Mangement Problem, Stanford Research Institute, International, Menlo Park, CA (June, 1978).

56. Towse, Donald F., Geoscience Parameter Data Base Handbook: Bedded Salt, Lawrence Livermore Laboratory, Livermore, CA, UCRL 52557 Draft (January 1979).

57. Towse, Donald F., Geologic Factors in Nuclear Waste Disposal, Lawrence Livermore Laboratory, Livermore, CA, UCRL 52522 (July 1978).

RKJ/all

542 310

APPENDIX A:
SPENT-FUEL DISSOLUTION MODELING

Groundwater intrusion into a deep geologic waste repository may result in spent-fuel dissolution and eventual radionuclide release and migration to the biosphere. Evaluation of the rate of such dissolution is a problem in chemical equilibrium and kinetics. It is necessary to identify the parameters that influence the dissolving process, assign probable values to them, and develop a model in which they can be used to quantify the release of potentially hazardous materials.

GENERAL APPROACH TO DISSOLUTION METHODOLOGY

The general approach to dissolution model development is to construct a basic framework of methodology. Simplifying assumptions must be made to create a procedure for evaluating waste dissolution in a format with numerical output. Although many assumptions are unrealistic, they recognize the need to defer consideration of some aspects of the problem. These assumptions, which must be reconsidered later in depth, include:

- The effects of possible interactions of water with repository materials other than spent-fuel waste forms are negligible.
- The rate of release of potentially hazardous elements to intrusive water will be proportional or closely related to the dissolution of the uranium dioxide (UO_2) fuel matrix in which they are dispersed.
- All canisters are identical, and a complete inventory of their contents will be available.
- The rate of water inflow is sufficiently slow that it is at all times fully equilibrated, i.e., saturated with respect to solid uranium oxide/hydroxide phases.
- The prevailing temperature is $298^{o}K$.
- The effects of the radiation field are negligible.

542 311

During waste dissolution model development the following factors will be considered:

- Repository materials, including masses and compositions of wastes, packaging, structural members, and backfill
- Distribution and chemical state of radioactive elements in waste forms
- Chemical composition of groundwater, including elemental stoichiometry, pH, and oxidation potential
- Equilibrium solubilities of materials
- Rates of approach to equilibrium
- Temperature
- Radiation field

COMPUTER MODELING

A complex chemical system with many variables, which must be expressed in terms of probabilities, can be analyzed and evaluated only by computer modeling techniques. An existing computer program [A1] was selected that has three desirable features for this application: speed, simplicity, and flexibility. Modifications were made that greatly increase its usefulness; these are not available in other computer programs. The existing data base was enlarged to include important radionuclides in fuel wastes, as well as elements found in natural waters. Control logic was incorporated, which makes it possible to evaluate the effects of groundwater composition on waste solubility over a wide range of system parameters. The variables whose effects this program can evaluate are average spent-fuel composition and groundwater composition.

For example, it is known that the pH and oxidation potential, or Eh, of an aqueous system have finite limits. In addition, natural waters tend to cluster in patterns in these composition variables, according to the host rocks with which they are associated.[A2] Solubilities for solids can also be computed in terms of pH and Eh;[A3,A4] mapping technique has been devised[A5] for evaluating these parameters, even when the values can be defined only statistically. Figures A1 and A2 illustrate a numerical and visual description of solubility; this technique can be applied to any required system. These exercises were specifically designed to explore the complex relationships between UO_2 solubility, oxidation, pH, and concentrations of chloride and carbonate ions. Figure A2 represents a 99% reduction in available carbonate. The significance of this capability is twofold. We can, for any fuel component for which data exist, identify the critical parameters that determine the maximum release rate (solubility). In addition, we can associate a numerical solubility with any arbitrary solution composition.

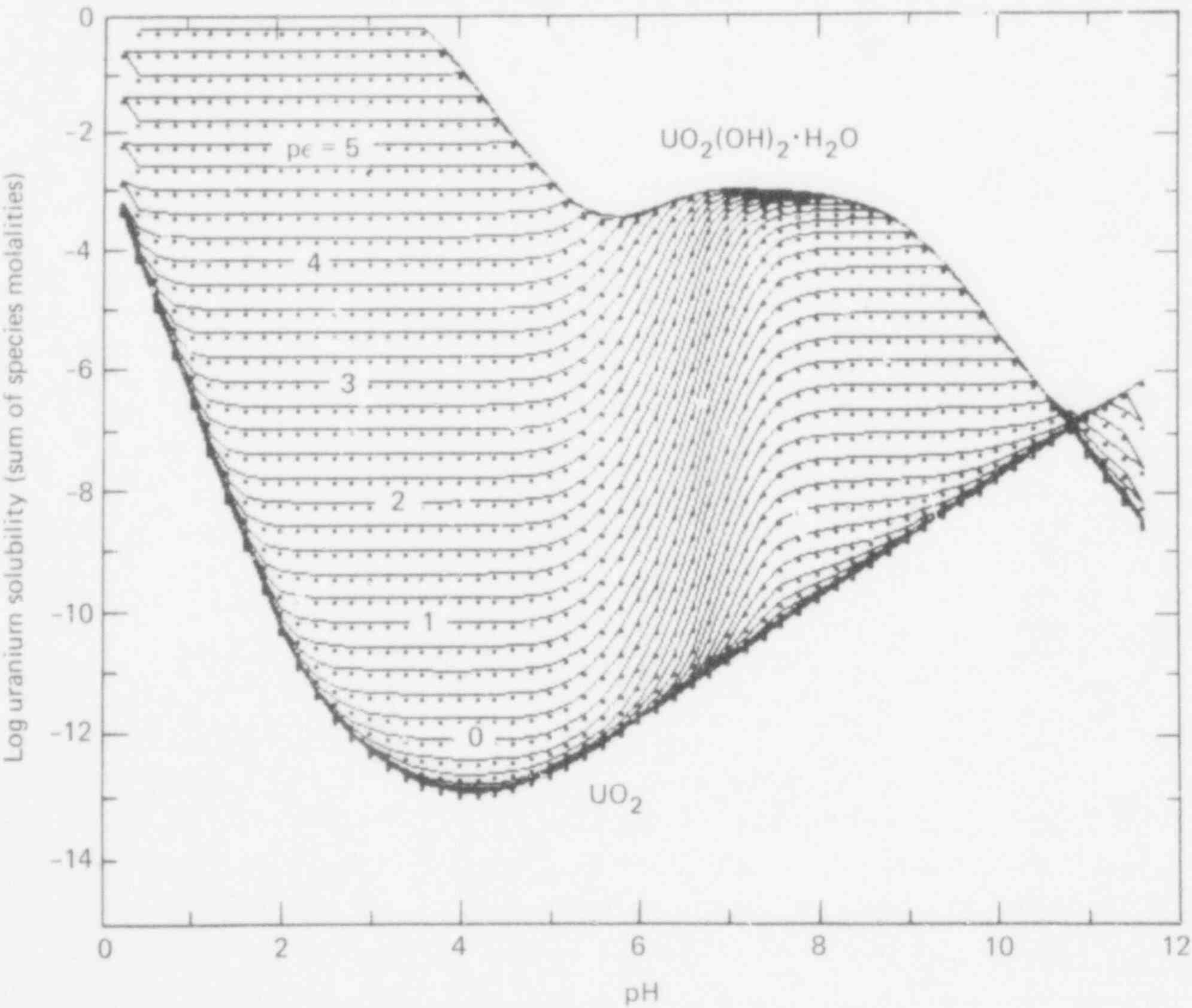

FIG. A1. Total uranium solubility in brine.

542 313

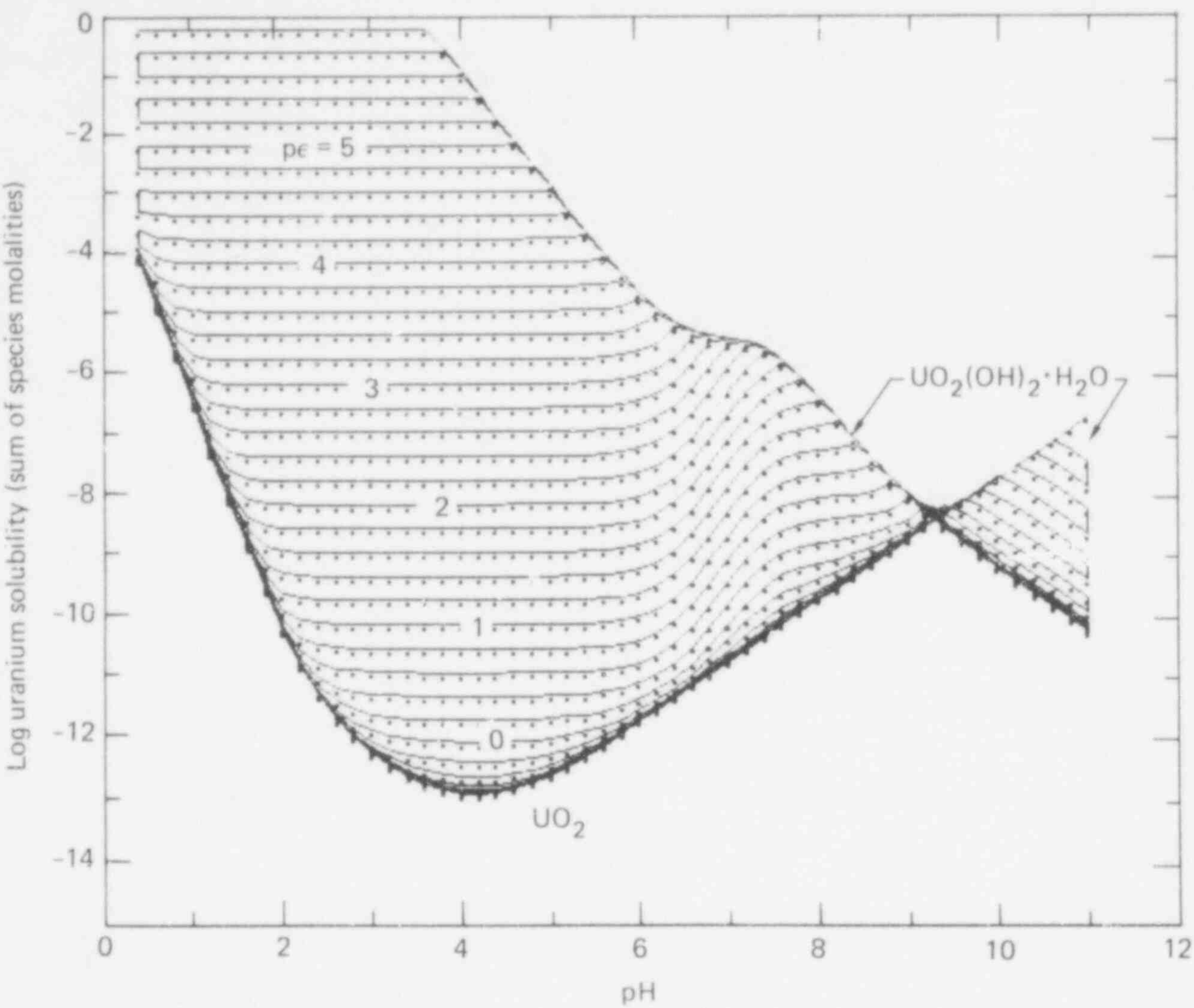

FIG. A2. Total uranium solubility in brine: reduced carbonate concentration.

The effects of groundwater composition on solubility are great, and vary with different radionuclides. Solubilities can be computed for specific elements in specific solutions, but it is not reasonable to assign a fixed composition to any potential intrusive water. It is preferable to calculate probability values for the concentrations of significant components; statistical analyses should be performed on published data for groundwaters in different geologic media such as salt, shale, granite, and tuff.

542 314

CONCLUSIONS AND RECOMMENDATIONS

It is possible to construct a spent-fuel dissolution model to obtain information on radionuclide release and migration resulting from groundwater intrusion into a deep geologic repository. Where physical and chemical data do not exist, a combination of experimentation and extrapolation of existing data will be employed.

There is insufficient useful data on UO_2 dissolution[A6-A9] for comparison with predicted values because groundwaters that have been isolated from the atmosphere for long times are at low oxygen activities. Under these conditions, the UO_2 fuel matrix will have a solubility and dissolving rate that is low and difficult to measure, but nonzero. It is recommended that experimental spent-fuel dissolution studies be performed in which extreme care is given to maintain and accurately measure these conditions.

There is an obvious deficiency in the treatment of equilibria at high ionic strength, an important consideration for groundwaters interacting with salt repositories. To improve the accuracy of chemical computations in highly saline environments, it is recommended that work be expedited on a comprehensive method of predicting activity coefficients for species in concentrated electrolytes.

It is recommended that a mathematical description of the actual physical and chemical states of radionuclides in fuel wastes be developed as these factors will affect the dissolution process.

542 315

FUTURE WORK

Data base expansion will be continued to include species of potentially hazardous radionuclides. A data file will be assembled on temperature reactions of radionuclides.

A survey of solubility behavior for critical radionuclides will be performed to identify parameters affecting dissolution.

Probability functions will be constructed for groundwater compositions in potential repository geologic media such as salt, shale, granite, basalt, and tuff.

A guide will be prepared identifying the specific chemical forms for the aqueous species of critical radionuclides. This information is necessary for developing retardation estimates in migration modeling.

A literature survey and data analysis will be carried out to identify the mechanism(s) of dissolution and oxidation of UO_2, the assumed dominant reactions controlling radionuclide release.

CITED REFERENCES

A1. J. Westall, J. Zachary, and F. Morel, MINEQL, A Computer Program for the Calculation of Chemical Equilibrium Composition of Aqueous Systems, Department of Civil Engineering, Massachusetts Institute of Technology, Cambridge, MA, Tech. Note No. 18 (1976).

A2. L. Becking, I. Kaplan, and D. Moore, "Limits of the Natural Environment in Terms of pH and Oxidation-Reduction Potentials," J. Geol. 68, 243 (1960).

A3. M. Pourbaix, Atlas of Electrochemical Equilibria in Aqueous Solutions (Pergamon Press, New York, 1966).

A4. R. Garrels and C. Christ, Solutions, Minerals and Equilibria (Freeman, Cooper and Co., San Francisco, 1965).

A5. D. Jackson, Solubility Mapping, a Novel Technique in Chemical Modeling, Lawrence Livermore Laboratory, Livermore, CA, Memorandum Wm-78-684 (1978).

A6. P. Laxen, "The Dissolution of UO_2 as an Electron Transfer Reaction," in The Recovery of Uranium, Proc. Symp. Recovery of Uranium From Its Ores and Other Sources, Sao Paulo, Brazil, 1970 (International Atomic Energy Agency, Vienna, 1971).

A7. I. Warren and E. Devuyst, "Leaching of Metal Oxides," in Proc. Intern. Symp. on Hydrometallurgy, D. Evans and R. Shoemaker, Eds., (American Institute of Mining, Metallurgical, and Petroleum Engineers, New York, 1973).

A8. J. Hunt and H. Taube, Tracer Studies on the Oxidation of UO_2 in Acidic Aqueous Solution, U.S.O.N.R. Proj. N.R. 052-415, Final Report-Part II (1963).

A9. T. Newton, The Kinetics of the Oxidation-Reduction Reactions of Uranium, Neptunium, Plutonium and Americium in Aqueous Solutions, U.S. Energy Research and Development Administration, TID-26506 (1975).

542 317

APPENDIX B:
VADOSE ZONE REPOSITORIES

In support of LLL's effort to model nonsalt geologic settings for the terminal storage of radioactive waste, the modeling requirements are being investigated for repositories located above a deep water table in arid regions. As is the case for any hydrologic model, four aspects of the system must be known:

- Hydraulic flow equations
- Hydraulic parameters
- Geometry
- Boundary and initial conditions.

Work during FY 78 on the first two aspects is covered in the section below, which is followed by a section on geometry. The fourth aspect, which depends in part on present and future climatic conditions, is not discussed in this report.

THE HYDRAULICS OF UNSATURATED FLOW

The flow of water in unsaturated soil above the groundwater table exhibits several unique characteristics favorable to the terminal storage of nuclear waste in the unsaturated vadose zone of arid sites. In general, moisture flows slowly in soils with low water content because liquid continuity (common to saturated flow) is limited and water is held tightly to soil particles by adsorption and capillary forces. Some significant features of unsaturated flow are cited below.

- Water pressure in unsaturated soil is subatmospheric; therefore, liquid water does not flow into large cavities where pressure is atmospheric or greater.
- The ease with which water flows through soil (hydraulic conductivity) decreases as the water content decreases (Figs. B1 and B2).

542 318

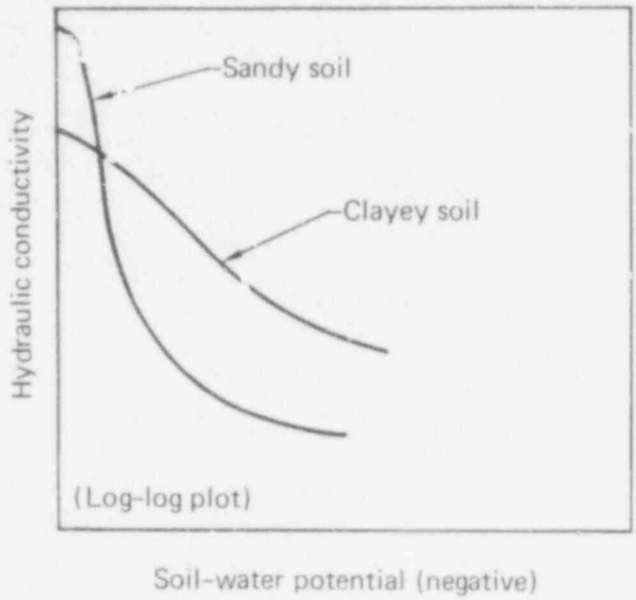

FIG. B1. Hydraulic conductivity vs. negative soil-water potential (suction).[B1]

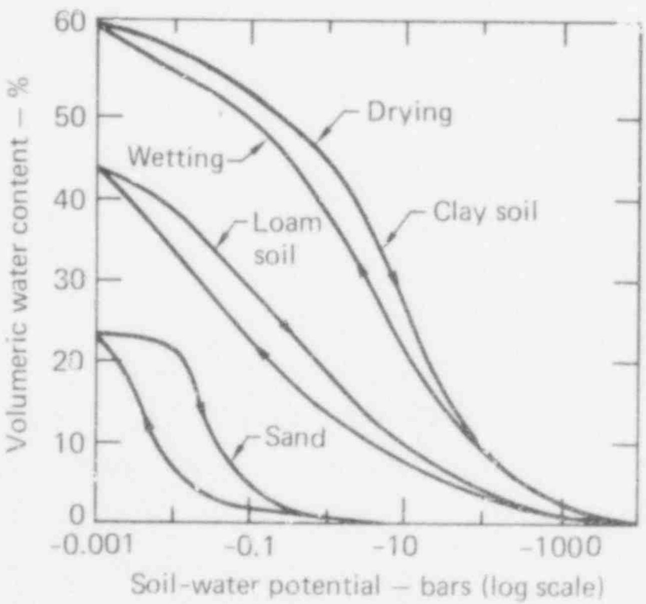

FIG. B2. Representative water retention curves showing hysteresis.[B2]

542 319

- As the water content of a soil decreases, the tension forces (suction) that cause water to remain trapped in the soil increase significantly (Fig. B2).
- The flow of water in wet soils (water contents greater than field capacity* but less than saturation) can be modeled mathematically. Such models have been tested by both field and laboratory experiments.
- Soil is hysteretic in that it behaves differently in wetting than in drying (Fig. B2).
- Water applied to the surface of a soil profile does not completely infiltrate the soil unless it is in sufficient quantity to bring the entire soil profile to field capacity.
- At low water contents (below field capacity), moisture flow is chiefly in the vapor phase, and therefore its ability to transport radionuclides is profoundly reduced.
- As the water content of a soil decreases, the influence of thermal gradients increases. Moisture tends to flow from warm areas to cool areas, and limited data from Hanford suggest that in arid areas the geothermal gradient of $3^{o}C/100$ m may be sufficient to cause a net <u>upward</u> flow of soil moisture through the soil.
- At very low water contents, water movement is a coupled phenomenon including the diffusion of vapor, the exchange of water between liquid and vapor phases (evaporation and condensation), the adsorption of water molecules on solid surfaces, and the flow of heat. Consequently, mathematical simulation of moisture flow at low water contents is in its infancy, and existing theories cannot be applied to field behavior of non-ideal materials.

The above preliminary findings indicate that vadose zones in arid regions may offer locations for nuclear waste repositories because they create areas with no natural pathways for radionuclide migration. Arid areas typically contain soils with very low moisture content and little, if any, net infiltration of water to the soil profile. Without extensive flow of water through the repository, there can be no migration of radionuclides. However, it must be

---

*Field capacity can be defined loosely as the water content that is retained in a free draining soil subject only to the force of gravity.

542 320

emphasized that this suggestion is based on theories that have been tested only for short time periods, in near-surface soils, and on a limited number of soil types. Additional research is required to extend the current theories to longer time periods and to the depths and materials found in arid regions with deep water tables.

CHARACTERIZATION OF ARID REGIONS WITH DEEP WATER TABLES

Modeling the hydrology of unconfined aquifers with deep water tables is important in order to understand potential escape paths for radionuclides deposited in thick, unsaturated zones. Because most hydrologic studies focus on water supplies from shallow water tables, modeling of deep water tables is made difficult by both lack of data and sometimes a fundamental lack of understanding of local conditions. Generally, deep, unconfined aquifers have been studied only at national facilities such as the Nevada Test Site (NTS) and the Idaho National Engineering Laboratory (INEL).

Deep water tables are possible wherever there is a thick, highly transmissive aquifer underlying an area of high topographic relief. At the NTS, the relief is created by basin and range topography. A thick sequence of highly fractured carbonate rocks acts as a regional, highly transmissive aquifer; locally, the water table is as deep as 600 m. Groundwater flow paths from the NTS to discharge points range up to 100 km, flow velocities are relatively slow, and quantities of flow are small. At the INEL, a thick, highly permeable sequence of basalt underlies the sloping Snake River Plain; locally, the water table is deeper than 300 m. At the INEL, flow paths are as long as 150 km, flow velocities are fast, and quantities of flow are large. Thus, at the INEL, radionuclide escape times might be shorter than at the NTS, but dilution would be greater.

Materials in the unsaturated zone at the NTS include soil, thick alluvium, fractured and zeolitized tuff (volcanic rocks), and highly fractured carbonate rocks. At the INEL, there is a relatively thin alluvial cover on top of a highly transmissive sequence of basalts whose permeability is provided by eruption features and cooling fractures.

542 321

Understanding the reasons for and controls on deep water tables is important in predicting possible rises of the water table should rainfall increase during future climatic regimes. At the NTS, large rises in the water table are unlikely because fault-controlled springs may act as safety valves in the event of increased recharge.

At both the NTS and the INEL, the regional aquifer is heterogeneous. However, the long history of groundwater usage and monitoring on the Snake River Plain has led to the development of a fairly good data base, and an initial groundwater model that is now able to predict the effects of both natural and man-made hydrologic changes. The assumption of aquifer homogeneity on a large scale at the INEL has apparently not restricted accurate modeling of the distribution of radionuclides disposed at the site. In contrast, at the NTS, the regional aquifer is largely unused and the data base is poor; assumptions of homogeneity of that aquifer may be invalid, making it difficult to use available data for the purpose of modeling in more than one dimension.

Other areas in the United States have deep water tables, and present research is being expanded to characterize other thick unsaturated sections of rock or soil that are both unsaturated for a depth of at least 500 ft and underlain by a regional aquifer whose hydrology can be easily modeled.

542 322

CITED REFERENCES

B1. D. Hillel, Soil and Water Physical Principles and Processes (Academic Press, New York City, 1971).

B2. R. M. Yong and B. P. Warkentin, Soil Properties and Behavior (Elsevier Scientific Publishing Co., 1975).

542 323

APPENDIX C:
RETARDATION FACTORS

SALT VS SHALE REPOSITORIES

In Cycles I and II, sorption in the salt repository was modeled only in the aquifer. It was assumed that salt had essentially no ion exchange capacity, and that in the barrier bed the high salt content of the groundwater would saturate the exchange sites and severely limit the sorption processes. We also assumed that in the aquifer dilution of the salty groundwater would subsequently nullify the salinity effect on ion exchange, and that sorption could take place as the radionuclides were transported to the biosphere. Further review of the data now suggests that these assumptions be revised. Consequently, the following changes in the retardation factors (K_f) are recommended for the salt repository model (see Table C1).

Salt Layer

The maximum value for the retardation factor has been increased from 1 to 10 for both the fission products and the actinides. This takes into account contamination of the salt by layer silicates and the possible existence of interlayered sedimentary sequences. A small ion exchange capacity is assumed for both the layer silicates and the sedimentary layers.

Shale Barrier Layer

The effect of competing ions on the actinide distribution coefficients (K_d) is not well documented and existing data conflict. As shown in Table C2, large K_d's (5×10^4 and 4×10^2) have been reported for Am in a 90% saturated NaCl solution. Yet, other work[C1] showed a sharp decrease in the values for K_d with 100- to 200-fold increases in Na and Ca ion concentrations for sandy clay samples and no change for sand. The K_d values for Np are already so small that environmental effects are probably not important. 324

TABLE C1. Retardation factors for the Cycle III transport model.

Radionuclide	Retardation factor, K_f[a] minimum	preferred	maximum
Shale Repository			
Iodine and technetium			
Shale layers	1	1	2 (1)
Aquifer	1	1	1
Other fission products	1	10^2	10^3
Actinides	10^2	10^4	10^5
Salt Repository			
Iodine and technetium			
Salt repository layer	1	1	1
Shale barrier layer	1	1	2 (1)
Aquifer	1	1	1
Other fission products			
Salt repository layer	1	1	10 (1)
Shale barrier layer	1	10 (1)	10^2 (1)
Aquifer	1	10^2	10^3
Actinides			
Salt repository layer	1	1	10 (1)
Shale barrier layer	1	10 (1)	10^3 (1)
Aquifer	10 (10^2)	10^3 (10^4)	10^4 (10^5)

[a]Numbers in parentheses are Cycle II values.

542 325

TABLE C2. Distribution coefficients for Am and Np as a function of the concentration of several media.

Experimental medium		Distribution coefficient, K_d	
		Am	Np
Routson et al. (1975)[C1]			
Sandy clay	.002m Ca	67	2.4
	0.02m Ca	1	0.4
Sandy clay	.015m Na	280	3.9
	3m Na	1.6	3.2
Sand	.002m Ca	$>10^3$	2.4
	0.2m Ca	$>10^3$	.4
Van Dalen et al. (1975)[C2]			
illite/kaolinite	90% sat. NaCl	5×10^4	
river sand	90% sat. NaCl	4×10^2	

542 326

Although the data above are meager, they do suggest that some sorption will occur in concentrated solutions. Therefore, a preferred K_f of 10 for both the fission products and the actinides is suggested. Maximum values of 10^2 and 10^3 take into account the generally greater adsorption potential of the actinides.

Aquifer Layer

The actual dilution factor of 70-fold, calculated as the waste enters the aquifer layer, is smaller than originally thought. A saturated NaCl solution diluted by this amount would be about 0.1M, which is still moderately saline. At this concentration the high retardation factor of 10^4 for the actinides would certainly be reduced. Thus the preferred value is now 10^3 with a maximum value of 10^4.

Retardation factors for the shale repository remain unchanged. We have assumed that a shale repository will have fresh groundwater even though groundwaters associated with shales are often slightly saline. The bulk of the fine-grained sediments were deposited in saline environments, and soluble components are likely to be retained as adsorbed ions on clay minerals or in interstitial water that was never completely removed by flushing. Indeed, White et al.[C3] found that one of the outstanding characteristics of shales is the scarcity of waters with less than 10^3 ppm total dissolved solids. The uncertain effect this concentration has on the retardation factor for the shale layer contributes to the total uncertainty.

Iodine and Technetium

Even very small K_d's yield appreciable retardation factors. For example, in shale when $K_d = 0.1$, $K_f = 6$, and when $K_d = 0.01$, $K_f = 1.6$. Experimental errors alone are sufficient to produce what appear to be real (although small) K_d's, thus indicating sorption where in fact none has occurred. The K_f values greater than unity occasionally reported for ^{129}I probably reflect variations in experimental conditions rather than sorption processes. Ion filtration is the only mechanism that has been demonstrated effective in retarding iodine in the absence of organics.[C4] Under ideal conditions, with shale layers acting as semipermeable membranes, the retardation factor for I^- could approach 2. The TcO_{-4} ion may behave in a similar manner. A maximum value of $K_f = 2$ has thus been recommended for both I and Tc in the shale layers.

VARIATIONS IN POROSITY AND SORPTION CHARACTERISTICS

The baseline values for porosity and the distribution coefficients used to estimate the retardation factors must be considered average values for the flow pathway. However, rock formations are not homogeneous, hence a range of parameter values would more accurately describe the rock characteristics than a single value.

Porosity

The physical factors that influence the porosity of sedimentary rocks are the amount of sorting and the grain shape of the original sediments. Sand composed of well-sorted and spherical grains has the greatest porosity. Although porosity is fundamentally not a function of grain size, coarse sands generally have greater porosity than fine sands because of differences in sorting and grain shape. It therefore follows that sedimentary rocks with local differences in sorting, grain shape, or grain size due to changes in the depositional environment will also show local differences in porosity. For example, we can predict a wide variation in porosity values for a sandstone formation with clay lenses and graded bedding (e.g., a pebble layer grading upward to a fine sand). Deformation of original sedimentary structures will also affect porosity. Examples include slumping, which can result in the mixing of clay and sand to form areas of low porosity, and bioturbation, the disturbance of the original structure by animal activity.

The chemical factors that affect porosity in sedimentary rocks are related to changes in pore size during diagenesis. Processes involving cementation, dissolution, recrystallization, and mineral alteration can create or destroy pores. Local and regional variations in porosity can thus be expected in response to changes in groundwater composition, since the relative intensities of these chemical processes are determined by the reactants present in the interstitial water.

Because of these physical and chemical variations, the baseline value of 10% porosity used to model mass transport in sandstone should be understood as the average of a range of values between 0% and 30%. Data to support this conclusion appear in Fig. C1.[C5]

Distribution Coefficients

Many of the physical and chemical processes that affect porosity also influence the distribution coefficients, hence the sorption characteristics, of the rock. Grain size, for example, determines the area available for surface sorption reactions. And since the mineral composition of the rock determines the exchange capacity, mineral alteration, especially the formation of clays, should enhance the exchange capacity. Local variations in grain size, mineral composition, or extent of mineral alteration will, therefore, produce local variations in the sorption characteristics of the rock.

Documentation of the variability we can expect in the sorption characteristics for a rock formation is limited. However, work [C6] has been reported that demonstrates an inverse correlation between grain size and adsorption for an aqueous plutonium-quartz system (Table C3) and that provides data on plutonium adsorption on different minerals (Table C4). Unfortunately, there are no published data for a systematic study of sorption variability in a single formation. Distribution coefficients calculated from experiments using multiple samples from the same rock formation are available;[C7] however, it is not clear whether the reported range of K_d values was a result of the experimental approach or the heterogeneity of the samples. Theoretically, an average K_d value of 500, estimated for plutonium adsorption on sandstone, could vary from 10, for an area composed of a pure quartz sand framework and silica cement, to 10^4, for a clay seam rich in montmorillonite.[C8]

542 329

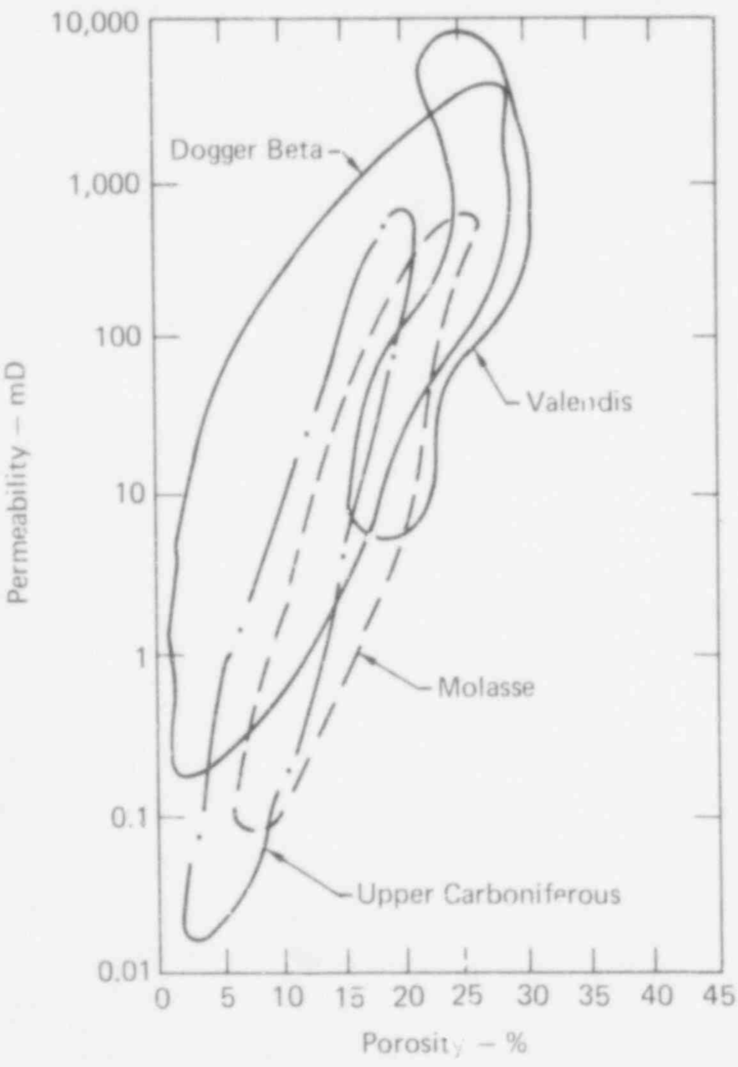

FIG. C1 Variations in the porosity of sandstone formations. The enclosed areas encompass 90% of the data points from the formations indicated. From Ref. C5.

542 330

TABLE C3. Distribution coefficicnts (K_d) of plutonium on quartz particles of various sizes, from a 0.001$\underline{N}$ HNO_3-plutonium solution.

Quartz particle sizes	K_d, ml/g
Coarse sand	5.6
Fine sand	10.0
Coarse silt (20-50 μm)	11.3
Medium silt (5-20 μm)	33.5
Fine silt (2-5 μm)	48.8
Coarse clay (0.2-2 μm)	80.9

TABLE C4. Distribution coefficients (K_d) of plutonium on different minerals of coarse clay size (0.2-2 μm), from a 0.001$\underline{N}$ HNO_3-plutonium solution.

Mineral	K_d, ml/g
Feldspar	170.4
Quartz	82.0
Glauconite	∞
Montmorillonite	157.0
Kaolinite	1091.0

542 331

SORPTION CHARACTERISTICS OF GRANITES AND BASALTS

Granites

The geochemical data base for granites as they relate to the disposal of nuclear waste is limited mostly to sorption studies by the Swedish Government.[C9] An extensive experimental study of granites, basalts, and other rock types is in progress under the guidance of Battelle Pacific Northwest Laboratories, but the data from the participating National Laboratories have not yet been formally released. There are a few K_d values available from other work,[C10,C11] but the supporting information in these studies is poor. Therefore, the following summary on granite geochemistry relies primarily on the Swedish work.

Groundwater Composition. Groundwaters in contact with granite are generally low in dissolved constituents. The dominant ions are usually Na^+ and HCO_3-. Fluoride content is often relatively high while calcium and magnesium are relatively low. Concentrations of SiO_2 can be very high in cold dilute waters. Near-neutral pH/values are common.[C3] These are the chemical characteristics expected of waters in equilibrium with igneous rocks that consist primarily of chemically resistant quartz and sodium-potassium feldspars.

In the Swedish study of the sorption characteristics of granite, a standard groundwater was defined on the basis of available groundwater analyses and the results of equilibrium measurements on clay-rock-water systems. Table C5 shows the average groundwater compositions for two sampling areas north of Malar, Sweden, and one of the standard water compositions (293 mg/l) used in the experiments. A second standard solution of higher ionic strength (1105 mg/l) was also used to account for the possibility of salt water entering the granite. This summary considers primarily the dilute groundwater cases.

542 332

TABLE C5. Average compositions of groundwater from two sampling areas, and composition of experimental standard (pH 8-8.5). From Ref. C9.

Ionic species	Concentration, mg/l		
	Malar-1	Malar-2	standard
Na	17	23	42
K	5	4	5
Mg	11	12	8
Ca	72	41	33
Cl	28	17	93
HCO_3	175	230	94
SO_4	80	20	7
Others	-	-	11
Total	378	347	293

Granite Distribution Coefficients. The Swedish study reported distribution coefficients as a function of temperature (25^o C and 65^o C), water composition (293 mg/l and 1105 mg/l), and radionuclide content (10^{-5}m$\underline{M}$ and 10^{-3}m$\underline{M}$). Table C6 gives selected K_d's for crushed granite in dilute solution (293 mg/l) at the two temperatures and two radionuclide concentrations. In the set of experiments using the more concentrated solution (1105 mg/l), K_d's were lower, often by a factor of two. A test of K_d as a function of the size fraction of the crushed granite produced the results shown in Fig. C2. Cesium shows no correlation, suggesting that Cs sorption is volume dependent rather than surface dependent. For a complete discussion of the experimental technique, see Ref. C9.

542 333

TABLE C6. Distribution coefficients for crushed granite (.063-.105 mm) as a function of temperature and radionuclide concentration, in water with ionic concentration of 293 mg/l, pH 8. From Ref. C9.

		K_d, ml/g	
Radionuclide	Concentration, m*M*	25 ^{o}C	65 ^{o}C
Sr	10^{-5}	8	10
	10^{-2}	10	16
Zr	10^{-5}	1.3×10^3	6.3×10^3
	10^{-2}	2×10^3	3.2×10^3
Tc	10^{-5}	0.5	0
I	10^{-5}	0.8	0
	10^{-3}	0.6	0
Cs	10^{-5}	1.3×10^2	63
	10^{-3}	32	32
Ra	10^{-5}	1×10^2	63
Th	10^{-5}	7.9×10^2	1.3×10^3
	10^{-3}	1.3×10^3	1×10^3
U	10^{-5}	6.3	13
	10^{-3}	5.0	6.3
Np	10^{-5}	40	50
Am	10^{-5}	1.3×10^4	1.3×10^4

512 334

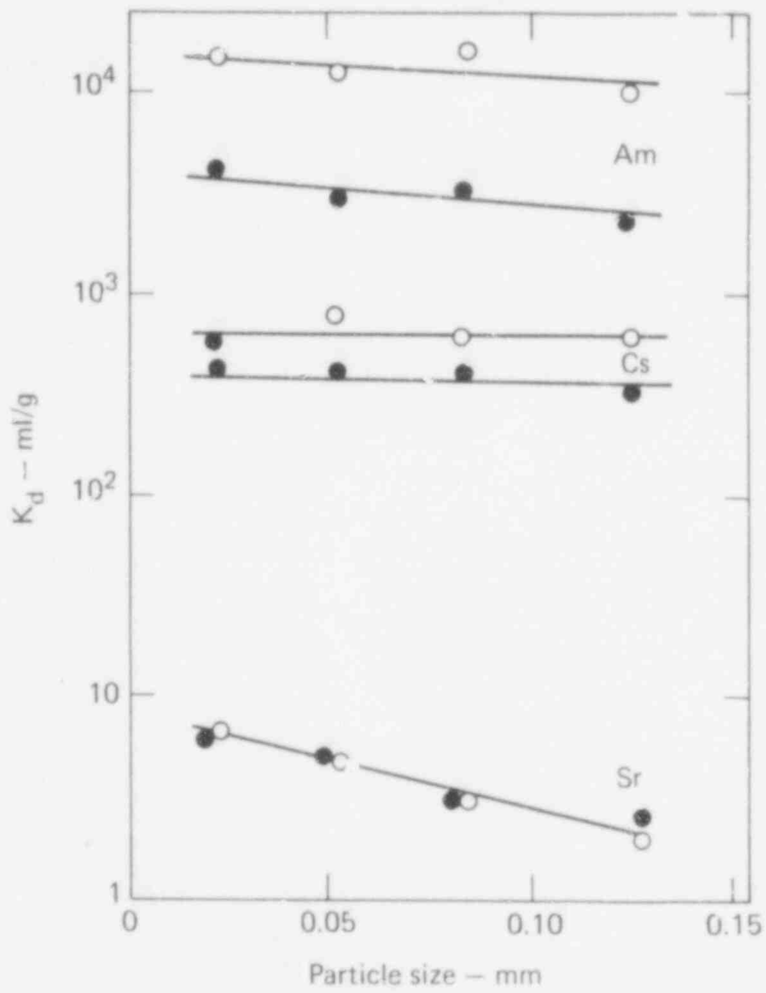

FIG. C2. Distribution coefficients for three radio-nuclides as a function of granite particle size. Closed circles represent measurements made after 6 h; open circles, 7 d. From Ref. C9.

542 335

The Swedish study indicates that the K_d's for freshly crushed granite compare well with those used to calculate retardation factors in our repository model for sedimentary rock:

	Granite	Model
Sr	8-16	10
Cs	31-130	10
Tc, I	0	0
Ra	63-100	300
Th	800-1,300	300
Am	13,000	300
U	5-513	300

Additional data on distribution coefficients would help reduce the uncertainty in the values for granite; however, the major concern in modeling a granite repository is estimating retardation factors for fracture flow.

542 336

Measurements on Rock Surfaces and Fracture Flow. An interesting aspect of the Swedish work is their interest in the sorption characteristics of rock surfaces. They tested both fresh and old rock surfaces, then calculated surface distribution coefficients,

$$K_a = \frac{Ci/m^2 \text{ rock}}{Ci/m^3 \text{ water}}$$

as well as K_d's. However, only a few experiments were made using Sr, Cs, and Am, and the results are probably not significant.

We currently calculate the retardation factor from K_d:

$$K_f = 1 + \frac{\rho}{\theta} K_d$$

However, retardation in rock where fracture flow is the predominate process may not be directly related to K_d. Instead, it may be necessary to calculate K_f from carefully measured surface distribution coefficients and estimates of fracture spacing. In situ studies are currently underway in Sweden on retardation in fractured granites.

Basalts

Groundwater Composition. Groundwaters in contact with basalt have high Ca:Na and Mg:Ca ratios. Minerals common in basalt are less stable and more subject to chemical attack than the minerals of granitic rocks. This chemical instability accounts for relatively high concentrations of SiO_2 in associated groundwaters. The pH, and probably the Eh, are often low; iron and manganese concentrations are often high. As with most rock types, groundwater compositions vary greatly both between and within rock formations.

542 337

Basalt Distribution Coefficients. The few studies on the sorption characteristics of basalt show a wide range of K_d's for Sr and Cs.[C7] Distribution coefficients for other fission products and the actinides are not yet available. Basalt is, however, the only rock type where we actually have *in situ* data for retardation. Measurements for Cs and Sr were made at an Idaho disposal site and the results compared to those predicted by a model.[C12]

Considering the variation in mineralogy common in basalts, sorption characteristics for basalt may be site specific. Thus, these few results do not accurately reflect the complexities of basaltic rock. Groundwater can flow through vesicular layers, along fractures or layer boundaries, and in sedimentary sequences between successive basalt flows. Retardation factors for a basalt repository model will, therefore, depend on which flow regime we use to describe the site. As with granite, we will have difficulty translating experimental K_d's to *in situ* conditions.

Additional data on distribution coefficients should soon be available from the Batelle study mentioned earlier. A DOE study of the Pasco Basin basalts should also provide a vast body of information. How much of this will be related to geochemistry and retardation is unknown.

512 338

CITED REFERENCES

C1. R. Routson, J. Jansen, and A. Robinson, Sorption of ^{99}Tc, ^{237}Np, and ^{241}Am on Two Subsoils from Differing Weathering-Intensity Areas, Battelle Pacific Northwest Laboratories, Richland, WA, BNWL-1189 (1975).

C2. A. Van Dalen, F. de Witte, and J. Wiskstra, "Distribution Coefficients for Some Radionuclides Between Saline Water and Clays, Sandstones, and Other Samples from the Dutch Subsoils," Reactor Centrum Nederland, pp. 75-109 (1975).

C3. D. White, J. Hem, and G. Waring, "Chemical Composition of Sub-Surface Waters," USGS Prof. Paper 440-F (1963).

C4. Y. Kharaka and W. Smalley, "Flow of Water and Solutes Through Compacted Clay," Amer. Assoc. Petrol. Geol. 60, 973-980 (1976).

C5. H. Fuchtbauer, Sediments and Sedimentary Rocks, Vol. 1, Part II (Halstead Press, N.Y., 1974).

C6. E. Evans, Plutonium Retention in Chalk River Soil, Atomic Energy of Canada, Ltd., Chalk River, Ontario, CRNP-660 (1956).

C7. D. Isherwood, Preliminary Report on Retardation Factors and Radionuclide Migration, Lawrence Livermore Laboratory, Livermore, CA, UCID-17551 (1977).

C8. E. Bondietti, S. Reynolds, and M. Shanks, "Interactions of Plutonium with Complexing Substances in Soils and Natural Waters," in Proc. Symp. Transuranium Nuclides in the Environment, San Francisco, Nov. 1976 (IAEA, Vienna, 1976).

C9. B. Allard, H. Kipatsi, and J. Rydberg, "Sorption of Long-Lived Radionuclides in Clay and Rock," KBS 55 (1977) English translation .

C10. W. Beetem, V. Janzer, C. Angelo, and M. Goldberg, "Summary of Distribution Coefficients for Fission Products Between Groundwater and Granite Rocks, Climax Stock," USGS Tech. Letter NTS-13 (Jan. 11, 1962).

C11. W. Nork and P. Fenske, Radioactivity in Water--Project Rulison, U.S. DOE Nevada Operations Office, Las Vegas, NV, NVO-1229-131 (1970).

542 339

C12. J. Robertson and J. Baroclough, "Radioactive- and Chemical-Waste Transport in Groundwater at National Reactor Testing Station, Idaho: 20-Year Case History and Digital Model," in Underground Waste Management and Artificial Recharge, J. Brounstein, Ed. (Amer. Assoc. Petrol. Geol., 1973), pp. 291-322.

542 340

APPENDIX D:
DISCRETE-EVENTS MODELING VS MARKOV-CHAIN MODELING

The purpose of this report is to examine the proposed mathematical modeling of the post-emplacement risks associated with nuclear waste disposal and to make recommendations on ways to improve the modeling. The modeling is intended to describe probabilistically the migration of waste material out of the repository and through the aquifer. This description can then be used in providing guidelines for waste management and in subsequent modeling efforts, such as the Human Usage Model, to assess potential environmental impacts.

GEOLOGIC FACTORS AFFECTING RELEASE FROM REPOSITORY

The diffusion of radioactive waste material over time is affected by the geologic characteristics of both the repository and the aquifer. When waste material is initially placed in the repository, it is assumed that there are no pathways through which leaching of waste can begin and that there are no pressure differentials to cause movement of the small amount of water inherent in the repository medium. The system is assumed to remain in this state until some event or activity results in both (1) a pathway along which nuclides can migrate to surface water and (2) the necessary force gradients to stimulate circulation and diffusion of water near the repository into the aquifer.

The authors concluded after discussion in May, 1978, with geophysicist G. Steiman at the University of California, Riverside, that once a pathway is open, heat generated by the waste material is sufficient to initiate circulation. Therefore, the primary concern in modeling focuses on geologic

542 341

events or activities that can result in the opening of pathways for leaching. Such events include earthquakes, drilling, loss of administrative control, and other such phenomena.

Steiman believed that it was reasonable to disregard any effects that weathering and erosion might have on the proximity of groundwater to the repository. He also felt that it was unnecessary to make distinctions among small, medium, and large earthquakes--that the end results of any earthquakes are approximately the same.

Aquifer characteristics deemed to be of concern in waste transport are path length, water flow rate, and sorption and dispersion: all of these are assumed to be determined by the geologic characteristics of the repository. That is, the state of the repository is assumed to be the same as the state of the aquifer, relative to waste transport. It is thought that changes in the geologic characteristics of the repository result in changes in flow rate, path length, sorption and dispersion, and that these changes can be initiated by events external to the repository, such as earthquakes, drilling, or loss of administrative control. It is further assumed that the flow of groundwater can be described by a network of one-dimensional pipes. Within each pipe, waste transport at a given point x and time t is described by the operator:

$$L^{(g)} = \frac{-V_g}{B_g} \frac{d}{dx} + \frac{\alpha_g V_g}{B_g} \frac{d^2}{dx^2} \qquad \text{(D1)}$$

542 342

where g is the state of the repository at time t, and V_g, B_g, and α_g are respectively the water velocity, sorption retardation, and dispersion coefficient of the aquifer determined by the repository state g.[D1]

DISCUSSION OF THE MARKOV CHAIN MODEL

A Markov chain model for this process has been proposed by TASC as an alternative to the fault tree analysis used in modeling other nuclear waste disposal aspects, such as transportation and handling. Criticisms of the appropriateness of fault tree analysis for the geologic model are justified - a fault tree approach is not very useful in examining post-emplacement risks. However, the Markov model proposed as an alternative has some rather substantial limitations. In putting the Markov model to use, it is necessary to specify a state space. Because of computational restrictions, the number of states must be both finite and limited in number. One possible state space is given by TASC[D2]. The states are actually sequences of events and, as proposed, the state space is quite limited in that it considers only specific sequences of events; recurrences of events are not taken into account. This last limitation may be of some concern in cases in which, for example, several small earthquakes occurring successively can create a combined effect greater than that of a single small earthquake. If the state space proposed in the above-referenced report is used, however, these two different possibilities would be considered as same state and therefore would be treated equivalently in predicting their effect in terms of waste transport. It is doubtful that any state space could be defined in which problems of this nature would not occur, because of both the complexity of the system being modeled and the requirement that the number of states be kept small. While this restriction was made for computational reasons, it may be a severe limitation on th

Markov model proposed uses a continuous time scale, and therefore the t ition probabilities used are necessarily instantaneous transition probabilities. While this time scale poses no theoroetical problems, the actual estimating of these probabilities is more difficult. In the present study, yearly occurrence rates have been used as estimates of the

542 343

instantaneous transition probabilities. Since the yearly occurrence rates are themselves only order-of-magnitude estimates, however, using them for the instantaneous transition probabilities provides only very crude estimates. Further, it is not clear exactly how these estimates can be improved.

The analysis of expected concentration, given in Ref. D1 and designed to eliminate the effects of history, also presents some mathematical problems. This analysis is accomplished by looking at the quantity

$$\varphi_j(x,g,t) = \sum_{h:h(t)=g} c_j(x,h,t)p(h) \tag{D2}$$

where g is the state of the repository at time t, $c_j(x,h,t)$ is the concentration at point x and time t given a particular trajectory h, $p(h)$ is the probability of the trajectory h, and the sum is taken over all trajectories that are in state g at time t. In addition, it is assumed that

$$\frac{d}{dt}\ c_j\ (x,h,t) = L^{(g)}\ c_j(x,h,t) \tag{D3}$$

where $L^{(g)}$ is defined in Equation (D1).

In examining Equation (D2), problems arise immediately as a result of the continuous time base. If time is taken to be continuous, there are uncountably numerous possible trajectories. This multiplicity then results in an uncountable number of terms in the sum. Two problems become apparent: (1) it is not at all clear how P(h) can be defined; and (2) if the number of trajectories that have a nonzero probability is not finite or at most countably infinite, it is not even clear how the sum can be defined. In either case, Equation (D2) does not make sense.

These particular problems could be alleviated if a discrete time Markov chain were used in place of a continuous time chain. In the discrete time case, transisitons occur only at fixed time intervals. Since we are interested in tracking waste movement over such a long period of time, however, there would be difficulty in choosing the time interval. This would necessitate a choice between computational efficiency for the model and its refinement and accuracy.

Once a time interval is chosen, the sum in Equation (D2) then becomes either finite or countable, depending on whether we fixed attention on a finite time span or instead were interested in tracking indefinitely. In either case, the sum in Equation (D2) would then make mathematical sense. However, even though some of the mathematical problems can be corrected by converting to a discrete time base, the development of expected concentration still presents some conceptual difficulties.

TASC's mathematical development of expected concentrations φ_j resulted in the differential equation

$$\frac{d}{dt}\varphi_j(x,g,t) = L^{(g)}\varphi_j(x,g,t)$$

$$+\sum_{g/g'}\lambda_{gg'}\varphi_j(x,g',t) - \sum_{g'/g}\lambda_{g'g}\varphi_j(x,g,t) \qquad \text{(D4)}$$

where $\lambda_{gg'}$ and $\lambda_{g'g}$ are the instantaneous transition probabilities from state g to g' and from state g' to g.

The primary problem arises from the fact that while it is a reasonable assumption that concentration at a given point x is continuous over time, the concentration at point x is not differentiable with respect to time at points of transition from one state to another. For example, suppose that at time t a transition occurs from stage g to g'. Then at least one of the following holds:

$$V_g \neq V_{g'}$$
$$B_g \neq B_{g'}$$
$$\alpha_g \neq \alpha_{g'}$$

(If this were not the case, there would be no difference between state g and g' with respect to waste transport--that is, g and g' would be considered the same state.) What happens then at a point of transition is that the concentration, while remaining continuous, begins to behave in a quite different manner as a result of the restriction

$$\frac{d}{dt}c_j(x,h,t) = -\frac{V_g}{B_g}\frac{d}{dx}c_j(x,h,t) + \frac{\alpha_g V_g}{B_g}\frac{d^2}{dx^2}c_j(x,h,t) \qquad \text{(D5)}$$

542 345

imposed by Equations (D3) and (D1) Therefore, $c_j(x,h,t)$ would not be differentiable at points of transition from one state to another because of the jump-like transitions that occur in the state-dependent constants V_g, B_g, and α_g.

Two critical contradictions are involved. If t <u>is</u> a point of transition, $\frac{d}{dt} c_j(x,h,t)$ does not exist, and the first portion of the analysis of expected concentration, which depends on the differentiability of $c_j(x,h,t)$, is not valid. If, instead, t <u>is not</u> a point of transition, then the second part of the analysis, which attempts to adjust the expected concentration to account for possible transitions both into and out of a given state at time t, is meaningless. In either case, whether t is a point of transition or not, the resulting differential equation (D4) is not correct.

This suggests that it is not possible, at least in the manner proposed, to eliminate the effect of history by looking at the aggregate quantity given by expected concentration over all possible histories. It also raises the question of whether looking only at the expected concentration is necessary for modeling transport of waste over time. TASC gives several reason for tracking expected concentration; the main advantage seen is in the reduction of computational complexity.

THE DISCRETE EVENTS MODEL

In view of the limitations discussed here, it may be beneficial to examine alternative ways of modeling repository state and aquifer characteristics. In particular, since changes in the repository state seem to be initiated by the occurrence of an event, such as an earthquake or drilling that is extraneous to the repository, it is felt that this entire system might be described adequately by a discrete events model.[D3]

The discrete events model is a systematic approach in which events initiate changes from state to state. In a discrete events model, there is a countable number of discrete states defined over a continuous time base. State changes are accomplished through the scheduling and rescheduling of events that effect jump-like transitions. Model updates need to be calculated only at the time an event occurs. Thus, this model does not necessitate extensive computations.

Two assumptions are necessary for this model:

- Only a finite number of events may occur in any finite time interval
- The system operates autonomously between events. This is equivalent to the assumption given in Equation (D3) for the Markov model.

This model handles waste transport, in terms of water velocity, sorption, and dispersion, in the same manner as in the Markov model. Therefore, no major changes in the aquifer model are necessary. There is no problem in using a continuous time base, and the limitations on state space found in the Markov model do not exist.

CONCLUSION

It is felt that the proposed Markov model for estimating expected concentration over time does not adequately describe post-emplacement risks. Also, because of the limitations imposed on both the state space and transition probabilities, the TASC model is not reflective of the geologic characteristics of the repository. We feel that the discrete events model would be more appropriate and more manageable than the Markov chain model, and that it would not involve the computational difficulties foreseen by TASC. It would provide not only the information derived from looking at the expected or average concentrations, but also other information. It would allow an examination of specific histories and could therefore be of aid both in determining consequences of a particular order and timing of events, and in determining key factors in evaluating post-emplacement risks.

542 347

CITED REFERENCES

D1. A. Sutherland and B. Ross, Use of the Markov Chain in Modeling Release of High-Level Waste from an Underground Repository, The Analytic Sciences Corp.(TASC), Reading, Mass., Report BIR-77-02 (26 April 1977).

D2. The Analytic Sciences Corp., Analysis of Nuclear Waste Management, (31 October 1976).

D3. B. P. Zeigler, Theory of Modeling and Simulation, (Wiley, 1976).

RKJ/all

542 348

NRC FORM 335 (7-77) U.S. NUCLEAR REGULATORY COMMISSION **BIBLIOGRAPHIC DATA SHEET**	1. REPORT NUMBER (Assigned by DDC) NUREG/CR-0538 UCRL - 52547
4. TITLE AND SUBTITLE (Add Volume No., if appropriate) Waste Management Technical Support Project FY78 End-of-Year Report	2. (Leave blank)
	3. RECIPIENT'S ACCESSION NO.
7. AUTHOR(S) R. A. Heckman, T. R. Donich	5. DATE REPORT COMPLETED MONTH December 8 YEAR 1978
9. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code) Lawrence Livermore Laboratory Livermore, California 94550	DATE REPORT ISSUED MONTH July YEAR 1979
	6. (Leave blank)
	8. (Leave blank)
12. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code) Office of Nuclear Material Safety and Safeguards U.S. Nuclear Regulatory Commission	10. PROJECT/TASK/WORK UNIT NO.
	11. CONTRACT NO. NRC FIN No. A0277
13. TYPE OF REPORT END OF YEAR	PERIOD COVERED (Inclusive dates) October 1977 to September 1978
15. SUPPLEMENTARY NOTES	14. (Leave blank)

16. ABSTRACT (200 words or less)

This report covers progress made during FY78 in the various tasks of the Waste Management Technical Support Project which Lawrence Livermore Laboratory is conducting for the Nuclear Regulatory Commission. The project's goal is to develop a technical base to be used by the NRC in developing regulations and regulatory guidance for the disposal of radioactive waste in geologic repositories. All analyses and conclusions are preliminary and do not reflect NRC positions.

17. KEY WORDS AND DOCUMENT ANALYSIS	17a. DESCRIPTORS
17b. IDENTIFIERS/OPEN-ENDED TERMS	

18. AVAILABILITY STATEMENT	19. SECURITY CLASS (This report)	21. NO. OF PAGES
Unlimited	20. SECURITY CLASS (This page)	22. PRICE $

NRC FORM 335 (7-77)

542 349

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555

OFFICIAL BUSINESS
PENALTY FOR PRIVATE USE $300

POSTAGE AND FEES PAID
U.S. NUCLEAR REGULATORY
COMMISSION

U.S. MAIL

120555031837 2 AN
US NRC
SECY PUBLIC DOCUMENT ROOM
BRANCH CHIEF
HST LOBBY
WASHINGTON DC 20555

542 350

~~542 207~~