UCRL-53188-81
Distribution Category UC-70

UCRL--53188-81

DE82 020575

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Nuclear Waste-Form Risk Assessment for U.S. Defense Waste at Savannah River Plant Annual Report FY 1981

**H. Cheung
L. L. Edwards
T. F. Harvey
D. D. Jackson
M. A. Revelli**

Manuscript Date: December 1981

LAWRENCE LIVERMORE LABORATORY
University of California • Livermore, California • 94550

Available from: National Technical Information Service • U.S. Department of Commerce
5285 Port Royal Road • Springfield, VA 22161 • \$8.00 per copy • (Microfiche \$3.50)

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

leg

Nuclear Waste-Form Risk Assessment for U.S. Defense Waste at Savannah River Plant Annual Report FY 1981

Executive Summary

1.0 Introduction

Savannah River Plant (SRP) has been supporting the Lawrence Livermore National Laboratory (LLNL) in its present effort to perform risk assessments of alternative waste forms for defense waste. This effort relates to choosing a suitable combination of solid form and geologic medium on the basis of risk of exposure to future generations; therefore, the focus is on post-closure considerations of deep geologic repositories. The waste forms being investigated include borosilicate glass, SYNROC, and others. Geologic media under consideration are bedded salt, basalt, and tuff. The results of our work during FY 1981 are presented in this, our second annual report. The two complementary tasks that comprise our program, analysis of waste-form dissolution and risk assessment, are described below.

2.0 Analysis of Waste-Form Dissolution

2.1 Introduction

In our FY 1980 studies of the chemistry of nuclear waste-form dissolution, we considered the thermodynamics of host phases and began to examine leaching reactions in groundwater. Our three main tasks in the current (FY 1981) phase of waste-form chemistry were (1) to continue to examine individual mineral-phase dissolution processes, (2) to study multiphase systems, where more complex interactions are possible, and (3) to establish a closer relationship between experimental waste-form leaching data and the long-term performance of a waste form in a geologic repository.

We examined the dissolution processes of the host phases by computing their thermochemical stabilities with respect to dissolution under given conditions of pH, temperature, and composition. Available, estimated, or assumed thermochemical data were used in these calculations by computer simulation of chemical equilibria.

In analyzing the long-term risks of a repository, we need to interpret experimentally determined leach rates in terms of the rates at which radionuclides are released from the waste form as a function of time. To facilitate this interpretation, we developed a computer code (MSC1, Multiple Shrinking Cylinder) to extrapolate, convert, and analyze dissolution data.

2.2 Dissolution of SYNROC Phases

Using computer simulations, we explored the dissolution chemistry of four individual SYNROC phases—zirconolite, barium hollandite, pollucite, and nepheline. The effects of deviations from the estimated free energies of formation and of variations in water pH were examined. We also investigated the nature of the resulting equilibrium solid phases and the concentrations of significant aqueous species.

We found the chemistry of the zirconolite-water system to be complex: three major phases, TiO_2 , $\text{Zr}(\text{OH})_4$, and $\text{CaO} \cdot \text{ZrO}_2 \cdot 2\text{TiO}_2$, appear in various combinations. It was found that phase transformations and solubilities can be sensitive functions of solid stability and solution pH. Major aqueous species were identified, and their concentrations were determined. Chloride complex species were not found to be significant, so only minor effects of chlorides from salt-repository groundwaters would be expected.

We used the previously estimated free energy of formation for barium hollandite to determine phases formed at equilibrium and aqueous species in the hollandite-water system as a function of pH. The interval $6 < \text{pH} < 12$ was examined. The preferential leaching of barium caused TiO_2 to be present at all

times. In neutral and acid solutions, $\text{Al}_2\text{O}_3 \cdot \text{TiO}_2$ is the major solid phase as barium is released to solution. Above $\text{pH}=7$, barium hollandite was stable; by $\text{pH}=8$, 98% of the barium had precipitated into this solid. Hollandites should, therefore, perform better in alkaline groundwaters.

Nepheline ($\text{NaAlSi}_3\text{O}_8$), the SYNROC D phase designed to absorb the 2.2 to 5.6% sodium present in defense waste sludges, is structurally important to the composite because it is a continuous phase. The results of computations of reactions leading to the precipitation of crystalline nepheline in aqueous solution showed that the rate of conversion, rather than thermodynamic stability, is the chief barrier to release from this phase.

We have begun a preliminary study of multiphase equilibria, i.e., more than one SYNROC phase dissolving simultaneously, and have found that the SYNROC phases often dissolve incongruently. Under certain conditions, the interaction between phases may drastically affect their stability. Our preliminary results showed that the instability of perovskite can, in turn, destabilize barium hollandite; that is, calcium released from perovskite competes successfully for the aluminum from hollandite. However, the effects of many other parameters remain to be investigated, including temperature, water composition, silicate formation, radionuclide element loading, and changes in the activities of components due to solid solution formation. We have established the direction not only for further analytical investigations, but have pointed the way for experimental studies.

2.3 Conversion of Experimental Leach Rates to Release Estimates

In the course of examining waste-form leaching data, we found a continuing need for a method to interpret such data in terms of the release of material from a waste form in storage in a geologic repository. We have developed a fast, general procedure to convert dissolution rates into release functions for input to risk-analysis codes. Our procedure extrapolates experimental dissolution rates to very long times, converts these rates into release functions for risk analysis, and analyzes the effects of the many other physical and chemical parameters of a waste-form/repository system on the release of waste elements.

Calculations based on the assumption of a uniformly shrinking waste-form cylinder are carried out by computer code (MSC1). This code computes mass losses in a series of time increments whose sizes can be varied as required. Our method considers the differences in dissolution rates in the several phases of a waste form, dissolution rate as functions of time, temperature, water composition, etc., repository water flow rate, and saturation.

3.0 Risk Assessment

3.1 Introduction

The most probable mechanism for release of significant quantities of radionuclides to the biosphere from a closed, deep geologic nuclear repository is groundwater transport. The process involves events that allow groundwater to contact the waste, leach the radionuclides, and, subsequently, transport them to the biosphere. Predicting doses to individuals who live near a nuclear waste repository requires careful modeling of the repository excavation, its backfill, and its effects on surrounding hydrologic conditions. Further, to provide decision-makers with technically complete information, evaluating the hazards associated with the disposal of high-level radioactive wastes in deep geologic strata demands that we acknowledge the uncertainties implicit in our predictions.

The systems analysis subtask of the Nuclear Waste Form Risk Assessment Project is to investigate the post-closure consequences associated with mined geologic repositories, with the objective of assessing the impact of candidate waste forms and geologic siting media on the overall performance of the waste-isolation system.

The highly detailed and time-dependent, three-dimensional computer code required to properly couple all the processes (e.g., hydrology, meteorological recharge, heat conduction, waste-canister stress and corrosion, waste-package leaching, water chemistry, repository refill and repressurization, nuclide transport, sorption, etc.) is not only complex, but will be too costly to run for very many simulations. Furthermore, in some cases, e.g., leaching or sorption, the processes may not be understood well enough to warrant detailed models.

We recognize that such a computer model is highly desirable (perhaps necessary) for best-estimate simulations of "real-world" measurements. However, a great deal of understanding can be gained by

using simpler models to do bounding calculations and sensitivity and uncertainty analyses. While these simpler models may be only qualitatively correct, relative measures of risk may be readily ascertained. Therefore, we have developed the MISER network-hydrology and nuclide-transport code and the XCOMP parametric comparison code on the basis of simple models.

3.2 The MISER Code

The MISER computer code was written to provide a computationally efficient analysis tool for studies of radionuclide release from closed geologic nuclear waste repositories. Because the development of MISER has drawn heavily on previous modeling efforts at LLNL and elsewhere, it is similar to the previous models. By the same token, it has several significant differences.

Our model is based on the following assumptions. Of these, at least the first are common to the other models referenced.

1. Three-dimensional single-phase flow in a porous medium may be adequately approximated by a network of one-dimensional stream tubes.
2. Thermal energy, heat conduction, waste-canister stress, water chemistry and saturation, diffusion, and lateral dispersion may be neglected.
3. Flow is slow enough for instantaneous ion-exchange equilibrium to result.
4. The repository scale is small compared to the regional hydrology; thus, a steady-state hydrology may be employed to decouple the hydrology and the nuclide transport.
5. The retardation factor is the same for all nuclides in the same decay chain.
6. The dissolution rate of the waste form is the same for all nuclides in the same decay chain.
7. The nuclides are, on the basis of retardations, dissolutions, and decay chains placed in three groups: (1) anions (e.g. ^{99}Tc , ^{129}I , ^{14}C), (2) cations (other fission products), and (3) actinides.
8. The radionuclide transport path is assumed to be semi-infinite for setting boundary conditions.

The MISER code traces nuclide flows through the network of stream tubes (tree branches) from nuclide sources to risk-sensitive observation points so that each branch is a series of sequential network paths. The appropriate source-strength fraction is computed on the basis of water-flow fractions. The approximate Green's function is then convoluted with the source function and resulting flux accumulated on a discretized time line. Radioactive decay is not used explicitly in our transport model. We compute a flux of nuclide groups as a function of time and then use tabulated results from the ORIGEN isotope generation and depletion code as multipliers in our dose/hazard calculations.

The development of MISER also represents a departure from prior modeling efforts at LLNL and elsewhere. By coupling the flow-path approach to an efficient algorithm, we have achieved in MISER a tool capable of analyzing a repository in levels of detail heretofore not possible. For example, one can consider the breaching time of a single canister among thousands of others. On the other hand, the code can also perform comparative analyses of many repositories and/or scenarios at less resolution of detail. This capability also allows us to account for uncertainties with Monte Carlo techniques where appropriate.

Further work needs to be done in the validation of transport models, particularly with respect to the near-field effects of repository design, placement of waste, and waste dissolution and release.

3.3 Applications of MISER

3.3.1 Introduction. In our initial application, we studied the well-release scenario. Our intent was to examine a performance feature sensitive to engineered features and waste-form characteristics. We considered an individual who drinks 2.2 litres of contaminated water per day from a well near the repository shaft. We determined the conservatism and weaknesses of the point-source assumption, compared the point-source and the distributed-source repositories, and studied the effects of some potential engineered features of repositories.

3.3.2 The Point-Source Repository. Results showing the calculated relative peak annual dose for a number of point-source repositories indicate that point-source models can lead to very misleading predictions of individual doses. An apparently conservative strategy might not lead to a conservative result. For example, models that appear heuristically conservative do not predict a dose that is as high as the best estimate. Alternatively, ultraconservative models can produce results that are actually two or three orders of magnitude too high. Besides being substantially too high or too low, point-source models can predict the wrong radionuclides as the main cause of peak dose.

We conclude that the choices in a point-source model are quite arbitrary and can lead to poor or misleading predictions of near-field individual doses. Predictions from point-source models of curies released to the near field must also be considered very questionable. To adequately predict repository releases will require more sophisticated models of the repository than point-source models.

3.3.3 Point-and Distributed-Source Comparisons. We show results generated to test the effect of smearing-out an identical amount of waste over different fractional areas of a repository. The amount of waste smeared-out totals a single canister for each case. The observed reduction of calculated dose (the more reduction, the greater the spatial separation of the canisters) will tend to reduce the significance of lower leach-rate waste forms. We see that the difference between a conservative point-source model and a detailed 1000-canister model is about two orders of magnitude. Besides these differences, the radionuclide causing the peak dose also changes from one case to the other.

3.3.4 Engineering Features of Repositories. We analyzed the impacts of some simple engineering of the hydraulic parameters of a repository on the dose to future individuals living close to the repository. Some of the simple engineered considerations show potential for reducing near-field doses. The cases we have analyzed are referred to as base case, upstream-shaft, off-tunnel storage, and hydraulic bypass. For our base case, we chose a downstream shaft with a repository storage area that is uniformly filled over its entire area.

Calculations show that engineered features could be integrated into a repository design that would lead to substantial reductions in forecasts of near-field doses to individuals. We suspect that a reduction of over four orders of magnitude can be obtained by combining the features examined here for a specific site. Searching for optimum results and expanding the features could further improve the near-field individual dose. Such site-specific numerical studies would be relatively inexpensive and certainly help attain an ALARA-designed repository.

3.4 Parametric Analysis

We conducted parametric analyses to study the impact of generic waste-form properties under a range of geologic conditions on overall waste-isolation performance. The results of this study will help us select the alternate media-siting scenarios to be studied in detail during the final phase of this project. The analyses were designed: (1) to simplify modeling of the overall system, (2) to span the range of credible performance measures, and (3) to provide a framework for evaluating scenario-specific analyses. Waste-form dissolution and radionuclide geotransport are two processes common to almost all release schemes for a deep geologic repository. The algorithms describing these processes require only a few input parameters, such as dissolution time, canister breach time, transit time, and geologic dispersivity, to characterize their outcome despite the host of physical, geological, and chemical variables that may give rise to a particular performance measure.

The XCOMP computer code was developed to study the effects produced by varying these parameters of the release function and the dose function without regard to details of flow-path network description. The code was written to vary these parameters systematically over specified ranges so that scenarios in which waste-form performance contributes significantly to dose may be readily determined. Parametric analyses covering wide ranges of canister breach time, waste-dissolution time, transit time, and geologic dispersivity have been completed, and the ranges and combinations of parameters that adversely affect dose have been identified.

1.0 Introduction

1.1 Background

Significant quantities of high-level nuclear waste have been generated in defense programs at the Savannah River Plant (SRP). The waste has been stored in aboveground storage tanks and will be incorporated into a solid host form suitable for permanent disposal in a deep geologic repository. The Department of Energy (DOE) has been sponsoring research and development on borosilicate glass, the reference waste form for SRP waste, and on potential alternative hosts such as SYNROC, coated particles, and others. The possible geologic media in which to construct the final repository might be bedded salt, basalt, or tuff. The choice of a suitable combination of solid form and geologic medium depends, in part, on the resulting risk of exposure to present and future generations. Savannah River Laboratory has been supporting the present work at the Lawrence Livermore National Laboratory (LLNL) to perform an assessment of risk for the storage of borosilicate glass and alternative waste forms containing SRP waste in deep geologic repositories. This, the second annual report, presents a discourse of our work during FY 1981. The program consists of the two complementary tasks described below.

1.2 Chemical Modeling

During FY 1980,¹ we studied and analyzed existing information on the processes, mechanisms, and modeling of dissolution of borosilicate glass and SYNROC. We reviewed and analyzed experimental data on the dissolution of SYNROC. We also began to study the thermochemistry of dissolution of SYNROC phases in water. Thermochemical data for SYNROC phases, associated phases, and aqueous species were compiled. The solubilities of rutile and perovskite were computed over a range of pH values. During FY 1981, we extended our equilibrium modeling to zirconolite, barium hollandite, and nepheline. In addition, we began to investigate multiphase equilibria.

Equilibria between zirconolite and one-molal NaCl aqueous solutions were computed for pH values of 6 to 12 and over a range of assumed free

energy of formation from -3531 to -3588 kJ/mole (-844 to -860 kcal/mole). We found that zirconolite dissolves incongruently. Calcium is released to the water phase, and TiO_2 and $\text{Zr}(\text{OH})_4$ are formed. Dissolution is sensitive to pH, but no significant chloride complexes were observed.

The solubility of barium hollandite in one-molal NaCl was computed for pH values of 6 to 12 and free energy of formation from -6703 to -6770 kJ/mole (-1602 to -1618 kcal/mole). The principal aqueous species was found to be Ba^{++} . Concentration measurements could clarify the hollandite stability problem with a relatively limited amount of experimental work.

Nepheline is expected to absorb the sodium present in defense-waste sludges. Calculations of reactions leading to its formation were carried out to determine its stability in aqueous solutions in the pH range 4 to 12. We found that nepheline in contact with such solutions would be converted to $\text{Al}_6\text{SiO}_{13}$ (mullite). Thus only its rate of conversion, not its chemical stability in water, could be relied on as the barrier to sodium release.

Thermochemical stability was estimated for pollucite, a major host phase for cesium. We carried out calculations similar to those for nepheline, but could find no conditions under which it was stable in aqueous systems.

A number of simulations were carried out to study possible consequences of chemical interactions among the SYNROC phases, perovskite, zirconolite, and barium hollandite during their simultaneous dissolution in water at pH 8.5. The free energies of formation of the latter two phases were varied over the ranges given above. Some significant interaction effects were observed. In the region of free energy of formation above about -3566 kJ/mole (-852 kcal/mole), zirconolite is unstable. At no point in the region mapped was either perovskite or barium hollandite stable. These and other effects observed lead to a preliminary interpretation that instability of perovskite can destabilize barium hollandite. It is apparent that both analytical simulation of multiphase interactions and experimental studies should be carried further to clarify the performance of various assemblages of SYNROC phases.

1.3 Risk Assessment

Our work on risk assessment deals specifically with the effects of waste form on repository performance. It involves the collection and interpretation of geological, geochemical, and hydrological data from site characterization, waste-media interactions, *in situ* testing, testing to obtain data for the simulation models, coupling individual models into a system of models for performance assessment, and, finally, verification of individual models and system components. This is a formidable task, and many aspects such as detailed systems modeling and a comprehensive treatment of disruptive events are beyond the scope and funding level of the current project. However, qualitative, relative performance predictions are possible.

Prediction of the transport of radionuclides from a nuclear waste repository in a deep geologic medium requires careful description of the geology, the hydrology, and chemical interactions over long periods of time. We must deal not only with the complexity of the phenomena, but with the uncertainties inherent in measurements and in the extrapolation of data over extended periods (10^6 years and longer). Despite the fact that it, too, may introduce additional uncertainties, computer modeling seems to be the most appropriate means of analysis. At the outset of the program, we reviewed the available computer models. They range in degree of complexity from simple one-dimensional, single flow-path models to those involving highly detailed, three-dimensional, finite-difference and finite-element representations.*

Because of convenience and availability, we initially selected the MACRO² codes developed at LLNL. These codes are based on the concept of a general model-management tool that systematically samples finite-probability representations of parameters to propagate uncertainty to the final results. The initial version, MACRO1, did not readily allow physical constraints and correlations, while the language features of its successor were much more general than we required. Further, its usage of conditional probability arrays in

some cases of highly constrained and correlated systems could lead to an inordinate number of computations.

Recognizing the need for a physically consistent yet versatile model, we have developed the MISER code. It has been written to provide a computationally efficient analysis tool to study near-field and repository design features in a probabilistic framework for post-closure nuclide transport and release phenomena. Basically, the code is written in a modular fashion to allow rapid implementation of new or improved models. It is user/analyst oriented, and extensive graphic output is implemented. MISER solves a general network of stream-tube pathways for a consistent, steady-state hydrology, traces "tree branches" of nuclide transport from numerous sources using a "propagator" approach, and computes hazard and/or dose to man at risk-sensitive observation points. Monte Carlo techniques are applied, where appropriate, to account for parameter uncertainties and correlations.

The MISER analyses reported here concern the conservatism of the point-source model, point- and extended-source comparisons, and some engineering features of repositories. These deterministic analyses are viewed as necessary precursors to analyses of waste-form effects. Some probabilistic analyses dealing with waste release have been computed with MISER; however, interpretations and discussions had not been completed when this report was written.

The potential dose to man from a repository may be approximated by using both a release function and a dose function. The former depends on transit time, dispersion, corrosion, and dissolution rates, while the latter depends on inventory, toxicity, population, water use, and other factors. Scenarios in which waste-form performance has any significant effect may be determined by parametric analyses. So that such analyses can be carried out efficiently, the XCOMP code was developed to systematically vary the parameters of both the release and dose functions. XCOMP does not take into account any network of flow paths, as in the case of MISER. It merely considers mean transit time and dispersion for a single radionuclide release pulse. Extensive calculations have been performed to study the consequences of variations in canister breach time, waste-dissolution time, mean overall transit time, and geologic dispersivity for a reference canister of Savannah River waste.

*No single, highly detailed code exists to couple all hydrological, chemical, transport, canister stress, corrosion; and other phenomena using either finite-difference or finite-element methodology.

2.0 Analysis of Waste-Form Dissolution

2.1 Introduction

In previous studies of nuclear waste-form dissolution, we considered the thermochemistry of host phases and began to examine their leaching reactions in groundwater. The current phase of waste-form chemistry had three main tasks: (1) to continue examination of individual mineral-phase dissolution processes, (2) to extend them to multi-phase systems, where more complex interactions are possible, and (3) to establish a closer relationship between experimental waste-form leaching data and the long-term performance of a waste form in a geologic repository.

The objective in studying the leaching of individual host phases is to determine what new product phases could be formed and how the relative and absolute concentrations of aqueous species would be affected. The dissolution processes of these phases were examined by computer simulations, using values for their thermochemical stabilities. When these values were not available, we chose to estimate them and then to explore the sensitivities of the results by duplicate calculations using a range of stabilities. Such an approach is feasible only by computer simulation.

The relationship between leach data and the analysis of long-term risks was approached by the development of a performance model that can help interpret experimental evidence in terms of the potential dissolution of material from a waste form. Such a model can perform several useful functions: extrapolate experimental dissolution rates to long times, convert them into release functions from waste forms in a repository, and analyze the effects of other physical and chemical parameters of a waste-form/repository system.

2.2 Thermochemistry and SYNROC Dissolution

2.2.1 Zirconolite

Zirconolite is a major phase in the various SYNROC compositions under development as candidates for the immobilization of radioactive wastes. Its principal function is to incorporate and retain the quadrivalent actinides of waste sludges. We feel that assessment of the long-term leachability of zirconolite, and the post-closure performance of waste forms in general, will require a

fairly detailed understanding of the chemical mechanisms involved. This means we must go beyond empirical leaching studies to validated numerical models that can evaluate all the parameters that might affect dissolving processes.

The thermodynamic stability of zirconolite has apparently not been measured, although we have estimated that the free energy of formation at 298-K, ΔG_{298}° , is about -3556 kJ/mole (-850 kcal/mole). Even with this minimal information, we can explore dissolution phenomena and gain considerable information on the response of zirconolite to leaching by natural waters; that is, the solid phases and maximum concentrations of aqueous species that can be formed. It is possible to use the stability of a solid phase, in this case zirconolite, as a parameter in examining the changes in the solid-water system.

Calculations of equilibria between zirconolite and aqueous solutions were first carried out using the free-energy estimate mentioned above. The total masses of calcium and zirconium in the system were assumed to be 0.01 mole/kg water; that of titanium was assumed to be 0.02 mole/kg water. Since no variations in the oxidation states of the component elements were to be expected, the pE (equilibrium with atmospheric oxygen) was fixed at 12.5 throughout. To qualitatively check for the possible effects of chloride from groundwater in a salt repository, the solution was one molal in NaCl. Chloride complexes for the elements considered were less stable than hydroxy complexes, and no significant effects were found.

Successive equilibrium calculations were performed at pH increments of 0.2 between pH=6.0 and 12.0. The calculated concentrations of the various species formed are shown in Fig. 1. We found that zirconolite dissolved incongruently, releasing primarily calcium to the water phase, with the formation of TiO_2 and $\text{Zr}(\text{OH})_4$. At pH values below about 8.6, the total 0.01 mole of calcium was leached out, leaving a solid residue of $\text{Zr}(\text{OH})_4$ and TiO_2 (the constant calcium molality below pH 8.6 in Fig. 1 is an artifact of the restricted amount of calcium input to this calculation). Titanium and zirconium species in the pH range 6.0 to 8.6 were all below 10^{-6} molal. The solid-phase assemblage changed with increasing pH: zirconolite appeared above pH=8.6, and $\text{Zr}(\text{OH})_4$ disappeared above pH=10.0. The calculated distributions of calcium, zirconium, and titanium among the various solid

Figure 1. Solids and species in the zirconolite-water system, $\Delta G_{f,298}^\circ = -850$ kcal/mole.

phases and aqueous species (for the specific masses of elements considered in the system) are shown in Figs. 2-4.

Similar computations were made for alternate values of zirconolite stability between -3531 and -3598 kJ/mole (-844 to -860 kcal/mole). The results, in terms of significant species and solid phases, are shown in Fig. 5 for $\Delta G = -3598$ kJ/mole. In the region where the pH is below about 7, the principal reaction would probably correspond to

At higher pH values, an amount (moles) of zirconium equal to the calcium would be released, as in Eq. (2):

Figure 2. Distribution of calcium in the zirconolite-water system, $\Delta G_{f,298}^\circ = -850$ kcal/mole.

Figure 3. Distribution of zirconium in the zirconolite-water system, $\Delta G_{f,298}^{\circ} = -850$ kcal/mole.

Figure 4. Distribution of titanium in the zirconolite-water system, $\Delta G_{f,298}^{\circ} = -850$ kcal/mole.

Figure 5. Solids and species in the zirconolite-water system, $\Delta G_{f,298}^{\circ} = -850$ kcal/mole.

That these reactions, as written, consume protons implies that dissolution would be sensitive to pH.

Figure 6 shows the dissolved calcium molality as functions of zirconolite stability and pH. If the free energy of formation at 298 K is -3556 kJ/mole (-850 kcal/mole) as previously estimated, the calcium, zirconium, and titanium distributions (Figs. 2-4) are most sensitive to pH in the range 8.5 to 9.5. This means that reaction with groundwaters, which might well fall within this range, would be correspondingly sensitive to pH and other pH-modifying reactions. However, note that this range shifts about one pH unit for every 1.6-kJ/mole (0.4-kcal/mole) change in the free en-

ergy of formation of zirconolite, so we cannot define it precisely at present.

2.2.2 Barium Hollandite

We previously estimated the free energy of formation of barium hollandite at approximately -6736 kJ/mole (-1610 kcal/mol).¹ Using this value, we have now carried out calculations for the dissolution of barium hollandite in one-molal NaCl. Because of the uncertainties in the data, we have used, in the present calculations, a series of alternate values from $\Delta G_{298}^\circ = -6703$ to -6770 kJ/mole (-1602 to -1618 kcal/mole) and have varied the pH in increments of 0.2 from 6 to 12.

Figure 6. Calcium release from zirconolite, with variable stability and pH.

The solubility data that were computed are summarized in Fig. 7. The total molalities of dissolved species are shown as a function of pH and solid-phase free energy. The upper limit at total molality of 0.01 is due merely to the total masses of elements input to the problem in this pH range.

Each point on Fig. 7 represents solubility at a fixed pH and free energy. The dissolved barium concentrations reach a minimum, defined by the lower boundary in the figure. There is another set of curves, increasing from this minimum, in which barium hollandite begins to redissolve in more alkaline solutions, with the formation of predominantly hydroxy complex species. Since the effect is very small, the set is not shown in Fig. 7. (One example is given in Fig. 8.)

Note that the estimate of -6736 kJ/mole (-1610 kcal/mole) would allow very high equilibrium concentrations of barium in water that is neutral to moderately alkaline. Therefore, the estimate may be too high when we consider the low leachabilities that have been observed experimentally.

Figures 9-16 show distributions of barium, aluminum, and titanium as a function of pH and solid-phase free energy. Simulations of more complex systems with arbitrary solid and solution masses and chemical compositions could be generated, but do not seem warranted at this time. The relations shown in the figures would be distorted somewhat by reactions with other ligands and phases. Since the principal aqueous species is always Ba^{++} , we have not done the calculations for dissolution in pure water. However, the results would not be very different.

Figure 7 may be helpful in designing dissolution experiments. Inspection shows that, at constant pH, errors of 15% in measurements of total barium concentrations should correspond to deviations of about 100 cal/mole in ΔG_{298} for barium hollandite. However, the converse also seems to be true; accurate values for solid stability are required for good prediction of solubilities. It is evident from Fig. 7 (as well as from Fig. 8) that experimental measurements should not be taken at high pH for two reasons: solubilities can be much lower, and there is a region where they are very insensitive to pH.

Normally, dissolution experiments with leach-resistant materials would be carried out at elevated temperatures because of higher rates and solubilities. However, the thermodynamic functions for BaCl^+ , BaOH^+ , and barium hollandite are not known, so the interpretation of results at

higher temperatures would be difficult. Note that every dissolution measurement in which pH, water volume, and barium concentrations are measured provides some information on stability. If the pH is allowed to vary (Fig. 7), the total aqueous barium concentration will follow some curved path. In time, it will approach some pH-molality line asymptotically and stop at equilibrium at the true free energy. We conclude that a relatively small amount of good experimental leaching data could greatly clarify the hollandite stability problem.

2.2.3 Nepheline

Nepheline, $\text{NaAlSi}_3\text{O}_8$, is the SYNROC D phase designed to absorb the 2.2 to 5.6% sodium present in defense-waste sludges. Silica, and aluminum if necessary, is added in formulation to ensure the formation of nepheline. This phase is important in two other respects: it contains cesium and strontium in solid solution, and it is host to some cesium as dispersed particles of pollucite, $\text{CsAlSi}_2\text{O}_6$. It is also structurally important to the composite in that it is a continuous phase and can form as much as 25 wt% of the waste form. Depending on the conditions of its formation, it may be partly crystalline.

We have carried out computations of reactions that might lead to the precipitation of crystalline nepheline in aqueous solution. However, we found no combinations of composition (Na, Al, Si) and pH in the range 4 to 12 in which this phase could be formed. Sodium remained in solution, and the solid phase produced an aluminosilicate $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ (3:2 mullite). The addition of NaCl up to a concentration of one molal had no effect, and the addition of silica had no effect up to quartz saturation. Increasing alumina to tenfold excess only precipitated gibbsite, $\text{Al}(\text{OH})_3$. The rates of leaching and solid-state conversion, then, are only barriers to release from this phase. The structure, porosity, and cohesiveness of the residual material are of considerable importance for the release of its radionuclide constituents, for the mechanical integrity of the waste form, and for the protection of other adjoining phases from access to water.

2.2.4 Pollucite

Cesium is known to form three aluminosilicates. One of these, pollucite, is, potentially, of considerable importance as a host phase for cesium in crystalline ceramic nuclear waste forms. These phases, together with their stoichiometric

Figure 7. Computed solubility functions for barium hollandite.

Figure 8. Distribution of barium species in equilibrium with barium hollandite, $\Delta G_{f,298}^{\circ} = -1612$ kcal/mole.

equivalents, are listed below in terms of component oxides.

CsAlSiO_4	$\text{Cs}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$	Pollucite
$\text{CsAlSi}_2\text{O}_6$	$\text{Cs}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 4\text{SiO}_2$	
$\text{CsAlSi}_5\text{O}_{12}$	$\text{Cs}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 10\text{SiO}_2$	

Pollucite is observed in SYNROC composites as crystallites in nepheline. If sodium from the nepheline host phase enters pollucite, solid solutions that cover most of the composition range from pollucite to analcime, $\text{NaAlSi}_2\text{O}_6 \cdot \text{H}_2\text{O}$ can be formed.³ Because of differences in ionic size, a sodium ion plus water molecule can replace a cesium ion at approximately a 1:1 ratio.

It is interesting to note that the removal of cesium from pollucite at high temperatures leads to the formation of mullite.⁴ This is analogous to the conversion of nepheline to mullite, as predicted above. In the formation of mullite from pollucite, the excess silica goes into the silica-rich

phase $\text{CsAlSi}_5\text{O}_{12}$, while that in the formation of mullite from nepheline goes into solution.

These phases have long been known and can be prepared either hydrothermally^{5,6} or by high-temperature dry synthesis⁴; however, their thermochemical stabilities have apparently not been measured. We have examined the possibility of estimating the free energy of formation of pollucite, with the objective of calculating its dissolution reactions with water.

Various techniques have been proposed for estimating the stabilities of silicates.⁷⁻¹² For several reasons, we cannot make use of these correlation schemes. Most important is the nonexistence of cesium silicates with known properties. Therefore, as an alternative, we have examined Group-1A phases for periodic relationships that could be used. Two analog framework silicates with known stabilities exist—leucite (KAlSi_2O_6) and dehydrated analcime ($\text{NaAlSi}_2\text{O}_6$). Rubidium analcime is known, but no data are available. Although data

Figure 9. Equilibrium distribution of barium in the dissolution of barium hollandite, $\Delta G_{f,298}^{\circ} = -1609$ kcal/mole.

Figure 10. Equilibrium distribution of barium in the dissolution of barium hollandite, $\Delta G_{f,298}^{\circ} = -1610$ kcal/mole.

Figure 11. Equilibrium distribution of aluminum in the dissolution of barium hollandite, $\Delta G_{f,298}^{\circ} = -1610$ kcal/mole.

Figure 12. Equilibrium distribution of titanium in the dissolution of barium hollandite, $\Delta G_{f,298}^{\circ} = -1610$ kcal/mole.

Figure 13. Equilibrium distribution of barium in the dissolution of barium hollandite, $\Delta G_{f,298}^{\circ} = -1612$ kcal/mole.

Figure 14. Equilibrium distribution of barium in the dissolution of barium hollandite, $\Delta G_{f,298}^{\circ} = -1614$ kcal/mole.

Figure 15. Equilibrium distribution of barium in the dissolution of barium hollandite, $\Delta G_{f,298}^{\circ} = -1616$ kcal/mole.

Figure 16. Equilibrium distribution of barium in the dissolution of barium hollandite, $\Delta G_{f,298}^{\circ} = -1618$ kcal/mole.

on cesium phases are also relatively sparse, we have assembled in Table 1¹³⁻¹⁸ and plotted in Fig. 17 the stabilities of seven phase types where all values for the sodium, potassium, rubidium, and cesium members are known.

Although there is a great diversity in the absolute values of free energy, it is clear from Fig. 1 that there is also considerable regularity in trends between members of Group 1A. The lowest curve is for the aluminosilicates, with an extrapolation to rubidium and cesium. We believe that the correlation is sufficiently good to place the free energy of formation of pollucite at about -2853 kJ/mole (-683 kcal/mole).

We made use of this value in computations of equilibria in water at 298 K. Pollucite was found to react in the same way as nepheline. That is, it lost cesium to the solution with the formation of mullite. The parameters investigated were a solid/solution ratio from 10⁻⁵ to 12, pH=4-12, and pollucite free energy of formation from -683 kcal/mole down to -800 kcal/mole.

How, then, is it possible to prepare pollucite at all? Computer analysis indicates that the principal dissolution reaction is

for which $\Delta G_{r,298}^\circ = -3154$ kJ/mole, or -526 kcal/mole of $\text{CsAlSi}_2\text{O}_6$. However, this reaction requires a large molar excess of water. Preparations of pollucite⁴⁻⁶ have been carried out by the formation and dehydration of gels, followed either by crystallization at moderate temperatures in autoclaves or by firing at high temperatures.

This means, essentially, that pollucite can only be formed by solid-state reactions equivalent to

for which $\Delta G_{r,298}^\circ = -51$ kcal/mole of $\text{CsAlSi}_2\text{O}_6$. The small amount of water is used only as a device to obtain intimate mixing of the reactants. It is either driven off entirely in firing or, in hydrothermal synthesis, it is present at temperature in insufficient volume to dissolve much cesium and silica.

We note in passing that Gallagher⁴ suggests the gaseous species CsAlO_2 to explain an apparent equimolar loss of cesium and aluminum in the firing of pollucite preparations. This is a bizarre species and would be of interest to high-temperature chemists. O-Al-OCs would be a previously unknown analog of O-Al-OH for which data exist.¹⁹ Its properties could be estimated if the loss of cesium during fabrication proved to be a significant problem.^{20,21}

2.2.5 Multiphase Equilibria

We have assessed the possible consequences of chemical interactions between SYNROC phases during their dissolution in water. The objective of this assessment was to determine what new product phases could be formed and how the relative and absolute concentrations of aqueous species would be affected. It should be useful to compare such estimates with experimental data. In a number of simulations, an attempt was made to precipitate simultaneously the three major SYNROC phases, perovskite, zirconolite, and barium hollandite. Because of uncertainties in the stabilities of the last two phases, this was a parametric study in which both stabilities were varied.

Table 1. Stabilities of alkali metal phases, $\Delta G_{f,298}^\circ = -1612$ kcal/mole.

Element	M ₂ SO ₄	Ref.	MCl	Ref.	MC10 ₄	Ref.	MAISi ₂ O ₆	Ref.
Na	-303.39	13	-91.842	13	-65.16	14	-674.989	15
K	-315.407	16	-97.736	15	-72.46	13	-687.354	16
Rb	-312.8	14	-97.47	13	-73.54	13		
Cs	-311.365	14	-99.100	17	-73.28	18		
Element	M ₂ SO	Ref.	MOH	Ref.	MNO ₃	Ref.	M ₂ CO ₃	Ref.
Na	-86.4	13	-90.739	16	-87.73	13	-250.5	13
K	-86.707	19	-90.567	16	-94.298	16	-254.444	19
Rb	-80.6	14	-87.1	14	-93.3	14	-249.3	14
Cs	-77.6	14	-88.60	16	-93.82	14	-243.6	14

Figure 17. Free energies of formation of Group-IA phases.

The region where ΔG_f° for zirconolite and barium hollandite was, respectively, -3558 to -3600 kJ/mole (-850 to -860 kcal/mole) and -6739 to -6865 kJ/mole (-1610 to -1640 kcal/mole) was examined.

Some preliminary results of the study are shown in Fig. 18. The figure is a solid-phase-product stability map for dissolution of a simple SYNROC-like composite in which only the components needed to form the three end-member phases were input. The pH was fixed at 8.5 as a reasonable value for groundwater, and the temperature was fixed at 298 K. Some significant effects of interaction were found: there were differences from the reactions of the individual phases. No $\text{Al}_2\text{O}_3 \cdot \text{TiO}_2$ was formed, and zirconolite was no longer stable above about -3566 kJ/mole (-852 kcal/mole). Instead, four phases developed in a fairly complex set of combinations that were a function of the free-energy ratio for zirconolite/hollandite (Fig. 18). At no point in the region

mapped was either perovskite or barium hollandite precipitated.

Our preliminary interpretation is that the instability of perovskite can, in turn, destabilize barium hollandite. That is, calcium released from perovskite competes successfully for the aluminum from hollandite. However, keep in mind that the effects of many other parameters must still be investigated, including temperature, water composition, silicate formation, radionuclide element loading, and changes in the activities of components as a result of solid-solution formation.

2.3 Conversion of Experimental Leach Rates to Release Estimates

In the course of examining waste-form leaching, we found a continuing need for a performance model to help interpret such data in terms of the release of material from a waste form stored

Figure 18. Computed equilibrium solid phases formed in the dissolution of perovskite plus zirconolite barium hollandite.

in a geologic repository. The following paragraphs describe a fast, general-purpose interface code we have developed to convert dissolution rates into release functions for input to risk-analysis codes.

Such a performance model has three functions: to extrapolate experimental dissolution rates to very long times, to convert these rates into release functions for risk analysis, and to analyze the effects of the many other physical and chemical parameters of a waste-form/repository system on the release of waste elements. As-measured rates are not necessarily dominant factors in release. What any single rate means in terms of long-term leaching by groundwater is not obvious.

In extrapolating experimental data, we must recognize that there is some ambiguity in leach rates derived from the concentrations of aqueous solutes. These data can be interpreted in several ways as evidence of the leachability of:

1. Actually determined elements.
2. All elements occupying equivalent lattice sites.
3. All elements other than those originating from the host lattice.
4. The entire host phase.
5. The entire waste form.

We consider the first interpretation to be the most reasonable—that concentrations of leached elements primarily reflect their own properties, and that extension to other elements is a questionable expedient. In a completely self-consistent numerical performance model, it is necessary to be specific about the exact meanings of measured rates and to account for all material changes in the waste form. For example, we should make an explicit distinction between true mass fluxes and those that have been computed with the assumption of congruent dissolution. If congruency is not established or cannot reasonably be assumed, such computed releases cannot be assigned any physical significance. Experimentally, the most common dissolution process for complex mineral phases is the preferential removal or replacement of one or more elements so that a change in solid composition takes place. If a dissolved element appears in more than one solid phase, its origin may be difficult to identify.

The model we have constructed is substantially physical and empirical at this point and, therefore, circumvents some problems of interpretation. For example, no distinction has yet been made between the dissolution of a phase and a component element. Any type of leaching data available can be accommodated without forcing

consistency. That is, as a practical matter, chemical interactions between components that are not yet well understood can be omitted. We can still examine the effects of many parameters without a perfect understanding of all the processes that take place, and it is useful to do so. As more chemical detail is added, any inconsistencies that exist will, no doubt, become apparent.

The model considers a single waste-form cylinder. Uniform shrinking is assumed, i.e., the cylindrical or outer surface of a phase recedes everywhere at the same specified rate. We have also made some other initial assumptions:

- The waste form maintains its physical integrity without collapsing or spalling.
- Component phases dissolve independently and congruently, are physically continuous, and are exposed to leaching in proportion to their volume fractions.
- Multiple phases can be considered, as in a SYNROC composite.
- Each phase initially occupies the entire volume of the original waste form, at an effective density determined by its true density and volume fraction. It maintains this effective density as it leaches (at its cylindrical or outer surface) and, therefore, occupies a decreasing volume that is determined by its leach rate.

The waste-form phases, then, shrink independently, resulting in a composite cylinder of inhomogeneous composition. Calculations for the model are carried out by the computer program MSC1, which computes mass losses in a series of time increments whose sizes can be varied as required.

A surface that recedes at a constant velocity is appropriate for the linear leach rates often observed experimentally. However, other processes in a leaching system, such as diffusion and surface depletion at early times or the approach to saturation at later times, may cause deviations from simple linear behavior. Therefore, we have provided the capability for variable leach rates. Equation (5) is convenient for this purpose and has often been used to fit other empirical data, such as heat capacities,

$$M = C_1 + C_2T + C_3T^2 + C_4T^3 + C_5T^{-2}, \quad (5)$$

where M is mass flux (mass per unit area per unit time), T is time, and C_1 to C_5 are constants determined by least-squares fit to experimental data. The mass flux is computed for each time increment and is used to determine the release.

The first chemical constraint examined was that on the release of silica. The maximum amount of silica that can be removed from a waste form that is releasing silica is computed from the water volume and the saturation concentration. The volume of water that flows during each time increment is computed from the input linear flow rate and the connected porosity of the formation. The saturation concentration in brines, C_s , is computed from the Kelvin temperature T and Eq. (6),

$$C_s = 10 \exp -(1.673 + 626.2/T) \text{ g SiO}_2/\text{g H}_2\text{O}. \quad (6)$$

The mass released in saturated flow can be compared with that computed using Eq. (5) and the silica content of the waste form. If necessary, a saturation constraint can then be applied. In this model, an initial aqueous concentration of silica in the groundwater should act to suppress dissolution even though we have not yet provided for the input and use of a water composition.

An example of the input for a three-phase waste form is shown in Tables 2 and 3. Table 4 shows the various release parameters computed for the progressive leaching of such a composite. We have given the major component the nominal leach rate, mass, and geometry of a cylinder of SRP 21-35-15 borosilicate glass,²² reacting with brine at 363 K (90°C). The resulting cumulative fractional releases are shown in Fig. 19. To illustrate the operation of the model, we have added smaller masses of phases X and Y, which leach at lower rates. These phases seem to be completely removed by about 6000 and 13,000 years, respectively. From Table 4, we see that, at the assumed water flow rate and at a silica content of 42.2% for the glass, all the silica released can just be accommodated by the water: a solution is formed that is slightly undersaturated with respect to amorphous silica. For other sets of repository conditions, this will not necessarily be true.

2.4 Summary and Conclusions

The dissolution chemistry of four individual SYNROC phases—zirconolite, barium hollandite, nepheline, and pollucite—was explored by computer simulations. We also examined the effects of deviations from the estimated free energies of formation and of variations in water pH. In addition, we investigated the nature of the resulting equilibrium solid phases and the concentrations of significant aqueous species.

The chemistry of the zirconolite-water system was found to be complex: three major phases appear in various combinations. Phase transformations and solubilities were found to be sensitive functions of solid stability and solution pH. Major aqueous species were identified and their concentrations determined. Chloride complex species were not found to be significant, so that only minor effects of chlorides from salt-repository groundwaters would be expected.

We made the following conclusions. Under most likely conditions, calcium would be lost preferentially. The order of total dissolved concentrations of elements at equilibrium would be $\text{Ca} > \text{Zr} > \text{Ti}$ or $(\text{Ca}=\text{Zr}) > \text{Ti}$. The calcium release is a sensitive function of pH and especially of zirconolite stability, so it could not reasonably be predicted from estimated values of free energy. However, we believe that the converse is true: experimental measurements of calcium released from pure single-phase zirconolite at constant pH could be used to determine its stability. If its free energy of formation at 298 K is -3556 kJ/mole (-850 kcal/mole) as previously estimated, the calcium, zirconium, and titanium distributions are most sensitive to pH in the pH range 8.5 to 9.5. This means that reaction with groundwaters, which might well fall within this range, would be sensitive to other pH-modifying reactions. In SYNROC development work, zirconolite is always leached in the presence of perovskite. In

Table 2. Waste-form parameters.

1.4821E+06	WMASS	INITIAL TOTAL MASS, G
2.5000E+00	WDENS	SOLID DENSITY, G / ML
7.8260E+00	RATIO	L / R, CYLINDER LENGTH / RADIUS
1.0000E-01	DINCR	INITIAL TIME INCREMENT, DAYS
3.6300E+02	TEMPK	KELVIN TEMPERATURE
5.0000E-01	VELOC	LINEAR WATER FLOW VELOCITY, CM/YEAR
1.2000E+00	BDENS	WATER DENSITY, G/ML
1.0000E-02	POROS	FRACTIONAL CONNECTED POROSITY OF HOST FORM
6.0960E+02	SPACE	CANISTER SPACING, CM
3	NPSES	NO. OF REACTING COMPONENTS

Table 3. Component dissolution parameters.

P 1 GL		TYPE, I.D. NO., AND SYMBOL OF MATERIAL LEACHED
1.4810E+06	PMASS	TOTAL INITIAL GRAMS PHASE IN CYLINDER
2.5000E+00	PDENS	PHASE DENSITY, G/ML
1.2000E-07	PCONS(PIDNO,1)	CONSTANT C1 IN RATE FUNCTION
0.	PCONS(PIDNO,2)	C2
0.	PCONS(PIDNO,3)	C3
0.	PCONS(PIDNO,4)	C4
0.	PCONS(PIDNO,5)	C5
P 2 XX		
1.0000E+03	PMASS	
2.5000E+00	PDENS	
1.0000E-08	PCONS(PIDNO,1)	
0.	PCONS(PIDNO,2)	
0.	PCONS(PIDNO,3)	
0.	PCONS(PIDNO,4)	
0.	PCONS(PIDNO,5)	
P 3 YY		
1.0000E+02	PMASS	
2.5000E+00	PDENS	
2.4000E-09	PCONS(PIDNO,1)	
0.	PCONS(PIDNO,2)	
0.	PCONS(PIDNO,3)	
0.	PCONS(PIDNO,4)	
0.	PCONS(PIDNO,5)	

such experiments, calcium from zirconolite is asked by a more rapid release from perovskite.

That zirconolite would leach in this way was not unexpected because of the relative solubilities of the component oxides, CaO, ZrO₂, and TiO₂. The main question is how tightly the calcium is bonded in this phase or, in other words, how much calcium must be released before equilibrium is attained, if at all.

The previously estimated free energy of formation for barium hollandite was used to determine phases formed at equilibrium and aqueous species in the hollandite-water system as a function of pH. The interval 6 < pH < 12 was examined. The preferential leaching of barium caused TiO₂ to be present at all times. In neutral and acid solutions, Al₂O₃·TiO₂ is the major solid phase as barium is released to solution. Above pH=7, barium hollandite was stable; by pH=8, 98% of the barium had precipitated into this solid. Hollandites should, therefore, perform better in alkaline groundwaters.

The parameters whose possible effects on leaching have been surveyed are free energy of formation of the solid phase (from -6703 to -6770 kJ/mole, including our previous estimate of -6736 kJ/mole) and pH in the range 6 to 12. The aqueous phase was one-molal NaCl. The dis-

tributions of barium between various solid and aqueous species were estimated as a function of these parameters. Barium should, by far, be the major element leached and Ba⁺⁺ the principal species. The sensitivity of solubility to solid free energy was estimated. If the free energy of formation at 298 K is -6736 kJ/mole (-1610 kcal/mole) as previously estimated, solubility would not be a useful constraint on leaching by groundwaters.

We estimated the stability of the cesium silicate, pollucite. No equilibrium solid/aqueous system was found in which either nepheline or pollucite was stable.

It appears, from the various SYNROC phases examined to date, that the release of alkali and alkaline earth elements, or their substituents, is a general phenomenon and that, except for calcium, these elements do not, themselves, have hydrous oxide phases with solubilities comparable in magnitude to those of aluminum, zirconium, or titanium.

Our preliminary examination of the interaction effects in a system where the three major phases were leached simultaneously shows that dissolution is, to some extent, system-specific. Important interactions are to be expected and, in some cases, lead to different solid phases and solubilities. However, the mechanisms are best

Table 4. Computed leaching functions.

TIME, YEARS	I.D.	MASS CORROSION RATE, G/Y	INCREMENT TO FRACTION RELEASED	CUMULATIVE FRACTION RELEASED	CUMULATIVE MASS RELEASED, G	MASS REMAINING, G	LENGTH, CM	RADIUS, CM	CUMULATIVE WATER FLOW, CUBIC METERS	CUMULATIVE SILICA REMOVABLE AT SATURATION, G
2.738E-04	P 1 GL	2.029E+00	3.751E-10	3.751E-10	5.555E-04	1.481E+06	2.260E+02	2.889E+01	5.087E-07	2.441E-04
	P 2 XX	1.691E-01	4.629E-08	4.629E-08	4.629E-05	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.058E-02	1.111E-07	1.111E-07	1.111E-05	1.000E+02	2.260E+02	2.889E+01		
6.297E-04	P 1 GL	2.028E+00	4.873E-10	8.624E-10	1.277E-03	1.481E+06	2.260E+02	2.889E+01	1.170E-06	5.614E-04
	P 2 XX	1.690E-01	6.015E-08	1.064E-07	1.064E-04	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	1.444E-07	2.554E-07	2.554E-05	1.000E+02	2.260E+02	2.889E+01		
1.092E-03	P 1 GL	2.028E+00	6.335E-10	1.496E-09	2.216E-03	1.481E+06	2.260E+02	2.889E+01	2.030E-06	9.740E-04
	P 2 XX	1.690E-01	7.819E-08	1.846E-07	1.846E-04	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	1.877E-07	4.431E-07	4.431E-05	1.000E+02	2.260E+02	2.889E+01		
1.694E-03	P 1 GL	2.028E+00	8.236E-10	2.320E-09	3.435E-03	1.481E+06	2.260E+02	2.889E+01	3.147E-06	1.510E-03
	P 2 XX	1.690E-01	1.016E-07	2.863E-07	2.863E-04	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	2.440E-07	6.871E-07	6.871E-05	1.000E+02	2.260E+02	2.889E+01		
2.476E-03	P 1 GL	2.028E+00	1.071E-09	3.390E-09	5.021E-03	1.481E+06	2.260E+02	2.889E+01	4.600E-06	2.207E-03
	P 2 XX	1.690E-01	1.321E-07	4.184E-07	4.184E-04	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	3.171E-07	1.004E-06	1.004E-04	1.000E+02	2.260E+02	2.889E+01		
3.492E-03	P 1 GL	2.028E+00	1.392E-09	4.782E-09	7.092E-03	1.481E+06	2.260E+02	2.889E+01	6.489E-06	3.114E-03
	P 2 XX	1.690E-01	1.718E-07	5.902E-07	5.902E-04	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	4.123E-07	1.416E-06	1.416E-04	1.000E+02	2.260E+02	2.889E+01		
4.814E-03	P 1 GL	2.028E+00	1.809E-09	6.592E-09	9.762E-03	1.481E+06	2.260E+02	2.889E+01	8.945E-06	4.292E-03
	P 2 XX	1.690E-01	2.233E-07	8.135E-07	8.135E-04	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	5.360E-07	1.952E-06	1.952E-04	1.000E+02	2.260E+02	2.889E+01		
6.532E-03	P 1 GL	2.028E+00	2.352E-09	8.944E-09	1.325E-02	1.481E+06	2.260E+02	2.889E+01	1.214E-05	5.824E-03
	P 2 XX	1.690E-01	2.903E-07	1.104E-06	1.104E-03	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	6.968E-07	2.649E-06	2.649E-04	1.000E+02	2.260E+02	2.889E+01		
8.765E-03	P 1 GL	2.028E+00	3.058E-09	1.200E-08	1.777E-02	1.481E+06	2.260E+02	2.889E+01	1.629E-05	7.815E-03
	P 2 XX	1.690E-01	3.774E-07	1.481E-06	1.481E-03	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	9.058E-07	3.555E-06	3.555E-04	1.000E+02	2.260E+02	2.889E+01		
1.167E-02	P 1 GL	2.028E+00	3.975E-09	1.598E-08	2.366E-02	1.481E+06	2.260E+02	2.889E+01	2.168E-05	1.040E-02
	P 2 XX	1.690E-01	4.906E-07	1.972E-06	1.972E-03	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	1.178E-06	4.732E-06	4.732E-04	1.000E+02	2.260E+02	2.889E+01		
1.544E-02	P 1 GL	2.028E+00	5.168E-09	2.115E-08	3.132E-02	1.481E+06	2.260E+02	2.889E+01	2.869E-05	1.377E-02
	P 2 XX	1.690E-01	6.378E-07	2.610E-06	2.610E-03	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	1.531E-06	6.263E-06	6.263E-04	1.000E+02	2.260E+02	2.889E+01		
2.035E-02	P 1 GL	2.028E+00	6.718E-09	2.786E-08	4.127E-02	1.481E+06	2.260E+02	2.889E+01	3.781E-05	1.814E-02
	P 2 XX	1.690E-01	8.292E-07	3.439E-06	3.439E-03	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	1.990E-06	8.253E-06	8.253E-04	1.000E+02	2.260E+02	2.889E+01		
2.673E-02	P 1 GL	2.028E+00	8.734E-09	3.660E-08	5.420E-02	1.481E+06	2.260E+02	2.889E+01	4.966E-05	2.383E-02
	P 2 XX	1.690E-01	1.078E-06	4.517E-06	4.517E-03	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	2.587E-06	1.084E-05	1.084E-03	1.000E+02	2.260E+02	2.889E+01		

Table 4. (Continued)

TIME, YEARS	I.D.	MASS CORROSION RATE, G/Y	INCREMENT TO FRACTION RELEASED	CUMULATIVE FRACTION RELEASED	CUMULATIVE MASS RELEASED, G	MASS REMAINING, G	LENGTH, CM	RADIUS, CM	CUMULATIVE WATER FLOW, CUBIC METERS	CUMULATIVE SILICA REMOVABLE AT SATURATION, G
3.502E-02	P 1 GL	2.028E+00	1.135E-08	4.795E-08	7.102E-02	1.481E+06	2.260E+02	2.889E+01	6.507E-05	3.122E-02
	P 2 XX	1.690E-01	1.401E-06	5.918E-06	5.918E-03	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	3.363E-06	1.420E-05	1.420E-03	1.000E+02	2.260E+02	2.889E+01		
4.580E-02	P 1 GL	2.028E+00	1.476E-08	6.271E-08	9.288E-02	1.481E+06	2.260E+02	2.889E+01	8.510E-05	4.084E-02
	P 2 XX	1.690E-01	1.822E-06	7.740E-06	7.740E-03	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	4.372E-06	1.858E-05	1.858E-03	1.000E+02	2.260E+02	2.889E+01		
5.981E-02	P 1 GL	2.028E+00	1.919E-08	8.190E-08	1.213E-01	1.481E+06	2.260E+02	2.889E+01	1.111E-04	5.333E-02
	P 2 XX	1.690E-01	2.368E-06	1.011E-05	1.011E-02	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	5.683E-06	2.426E-05	2.426E-03	1.000E+02	2.260E+02	2.889E+01		
7.803E-02	P 1 GL	2.028E+00	2.495E-08	1.068E-07	1.582E-01	1.481E+06	2.260E+02	2.889E+01	1.450E-04	6.957E-02
	P 2 XX	1.690E-01	3.079E-06	1.319E-05	1.319E-02	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	7.388E-06	3.165E-05	3.165E-03	1.000E+02	2.260E+02	2.889E+01		
1.017E-01	P 1 GL	2.028E+00	3.243E-08	1.393E-07	2.063E-01	1.481E+06	2.260E+02	2.889E+01	1.890E-04	9.069E-02
	P 2 XX	1.690E-01	4.002E-06	1.719E-05	1.719E-02	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.056E-02	9.605E-06	4.125E-05	4.125E-03	1.000E+02	2.260E+02	2.889E+01		
1.375E-01	P 1 GL	2.028E+00	4.216E-08	1.814E-07	2.687E-01	1.481E+06	2.260E+02	2.889E+01	2.462E-04	1.181E-01
	P 2 XX	1.690E-01	5.203E-06	2.239E-05	2.239E-02	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.055E-02	1.249E-05	5.374E-05	5.374E-03	9.999E+01	2.260E+02	2.889E+01		
1.725E-01	P 1 GL	2.028E+00	5.480E-08	2.362E-07	3.499E-01	1.481E+06	2.260E+02	2.889E+01	3.206E-04	1.538E-01
	P 2 XX	1.690E-01	6.764E-06	2.915E-05	2.915E-02	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.055E-02	1.623E-05	6.997E-05	6.997E-03	9.999E+01	2.260E+02	2.889E+01		
2.246E-01	P 1 GL	2.028E+00	7.125E-08	3.075E-07	4.554E-01	1.481E+06	2.260E+02	2.889E+01	4.173E-04	2.002E-01
	P 2 XX	1.690E-01	8.793E-06	3.795E-05	3.795E-02	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.055E-02	2.110E-05	9.107E-05	9.107E-03	9.999E+01	2.260E+02	2.889E+01		
2.922E-01	P 1 GL	2.028E+00	9.262E-08	4.001E-07	5.926E-01	1.481E+06	2.260E+02	2.889E+01	5.429E-04	2.605E-01
	P 2 XX	1.690E-01	1.143E-05	4.938E-05	4.938E-02	1.000E+03	2.260E+02	2.889E+01		
	P 3 YY	4.055E-02	2.743E-05	1.185E-04	1.185E-02	9.999E+01	2.260E+02	2.889E+01		
3.801E-01	P 1 GL	2.028E+00	1.204E-07	5.205E-07	7.709E-01	1.481E+06	2.260E+02	2.889E+01	7.063E-04	3.389E-01
	P 2 XX	1.690E-01	1.486E-05	6.424E-05	6.424E-02	9.999E+02	2.260E+02	2.889E+01		
	P 3 YY	4.055E-02	3.566E-05	1.542E-04	1.542E-02	9.998E+01	2.260E+02	2.889E+01		
4.945E-01	P 1 GL	2.028E+00	1.565E-07	6.770E-07	1.003E+00	1.481E+06	2.260E+02	2.889E+01	9.187E-04	4.409E-01
	P 2 XX	1.690E-01	1.932E-05	8.355E-05	8.355E-02	9.999E+02	2.260E+02	2.889E+01		
	P 3 YY	4.055E-02	4.635E-05	2.005E-04	2.005E-02	9.998E+01	2.260E+02	2.889E+01		
6.431E-01	P 1 GL	2.028E+00	2.035E-07	8.805E-07	1.304E+00	1.481E+06	2.260E+02	2.889E+01	1.195E-03	5.734E-01
	P 2 XX	1.690E-01	2.511E-05	1.087E-04	1.087E-01	9.999E+02	2.260E+02	2.889E+01		
	P 3 YY	4.054E-02	6.025E-05	2.606E-04	2.606E-02	9.997E+01	2.260E+02	2.889E+01		
8.363E-01	P 1 GL	2.028E+00	2.645E-07	1.145E-06	1.696E+00	1.481E+06	2.260E+02	2.889E+01	1.554E-03	7.456E-01
	P 2 XX	1.690E-01	3.264E-05	1.413E-04	1.413E-01	9.999E+02	2.260E+02	2.889E+01		
	P 3 YY	4.054E-02	7.832E-05	3.391E-04	3.391E-02	9.997E+01	2.260E+02	2.888E+01		

Table 4. (Continued)

TIME, YEARS	I.D.	MASS CORROSION RATE, G/Y	INCREMENT TO FRACTION RELEASED	CUMULATIVE FRACTION RELEASED	CUMULATIVE MASS RELEASED, G	MASS REMAINING, G	LENGTH, CM	RADIUS, CM	CUMULATIVE WATER FLOW, CUBIC METERS	CUMULATIVE SILICA REMOVABLE AT SATURATION, G
1.007E+00	P 1 GL	2.028E+00	3.439E-07	1.489E-06	2.205E+00	1.481E+06	2.260E+02	2.889E+01	2.020E-03	9.695E-01
	P 2 XX	1.690E-01	4.243E-05	1.837E-04	1.837E-01	9.998E+02	2.260E+02	2.889E+01		
	P 3 YY	4.054E-02	1.018E-04	4.409E-04	4.409E-02	9.996E+01	2.260E+02	2.888E+01		
1.414E+00	P 1 GL	2.028E+00	4.471E-07	1.936E-06	2.867E+00	1.481E+06	2.260E+02	2.889E+01	2.627E-03	1.261E+00
	P 2 XX	1.689E-01	5.516E-05	2.389E-04	2.389E-01	9.998E+02	2.260E+02	2.889E+01		
	P 3 YY	4.053E-02	1.323E-04	5.732E-04	5.732E-02	9.994E+01	2.260E+02	2.888E+01		
1.838E+00	P 1 GL	2.028E+00	5.812E-07	2.517E-06	3.728E+00	1.481E+06	2.260E+02	2.889E+01	3.416E-03	1.639E+00
	P 2 XX	1.689E-01	7.170E-05	3.106E-04	3.106E-01	9.997E+02	2.260E+02	2.889E+01		
	P 3 YY	4.052E-02	1.720E-04	7.452E-04	7.452E-02	9.993E+01	2.260E+02	2.888E+01		
2.390E+00	P 1 GL	2.028E+00	7.555E-07	3.273E-06	4.847E+00	1.481E+06	2.260E+02	2.889E+01	4.441E-03	2.131E+00
	P 2 XX	1.689E-01	9.320E-05	4.038E-04	4.038E-01	9.996E+02	2.260E+02	2.888E+01		
	P 3 YY	4.051E-02	2.235E-04	9.687E-04	9.687E-02	9.990E+01	2.260E+02	2.888E+01		
3.107E+00	P 1 GL	2.028E+00	9.822E-07	4.255E-06	6.301E+00	1.481E+06	2.260E+02	2.889E+01	5.774E-03	2.771E+00
	P 2 XX	1.689E-01	1.211E-04	5.249E-04	5.249E-01	9.995E+02	2.260E+02	2.888E+01		
	P 3 YY	4.050E-02	2.905E-04	1.259E-03	1.259E-01	9.987E+01	2.260E+02	2.887E+01		
4.040E+00	P 1 GL	2.028E+00	1.277E-06	5.532E-06	8.192E+00	1.481E+06	2.260E+02	2.889E+01	7.507E-03	3.602E+00
	P 2 XX	1.689E-01	1.575E-04	6.824E-04	6.824E-01	9.993E+02	2.260E+02	2.888E+01		
	P 3 YY	4.048E-02	3.775E-04	1.637E-03	1.637E-01	9.984E+01	2.260E+02	2.887E+01		
5.252E+00	P 1 GL	2.028E+00	1.660E-06	7.192E-06	1.065E+01	1.481E+06	2.260E+02	2.889E+01	9.759E-03	4.683E+00
	P 2 XX	1.688E-01	2.046E-04	8.870E-04	8.870E-01	9.991E+02	2.260E+02	2.888E+01		
	P 3 YY	4.045E-02	4.904E-04	2.127E-03	2.127E-01	9.979E+01	2.259E+02	2.886E+01		
6.828E+00	P 1 GL	2.028E+00	2.158E-06	9.349E-06	1.385E+01	1.481E+06	2.260E+02	2.889E+01	1.269E-02	6.088E+00
	P 2 XX	1.688E-01	2.659E-04	1.153E-03	1.153E+00	9.988E+02	2.260E+02	2.887E+01		
	P 3 YY	4.042E-02	6.370E-04	2.764E-03	2.764E-01	9.972E+01	2.259E+02	2.885E+01		
8.877E+00	P 1 GL	2.028E+00	2.805E-06	1.215E-05	1.800E+01	1.481E+06	2.260E+02	2.889E+01	1.649E-02	7.915E+00
	P 2 XX	1.687E-01	3.456E-04	1.499E-03	1.499E+00	9.985E+02	2.260E+02	2.887E+01		
	P 3 YY	4.038E-02	8.273E-04	3.591E-03	3.591E-01	9.964E+01	2.259E+02	2.884E+01		
1.154E+01	P 1 GL	2.028E+00	3.647E-06	1.580E-05	2.340E+01	1.481E+06	2.260E+02	2.889E+01	2.144E-02	1.029E+01
	P 2 XX	1.686E-01	4.490E-04	1.948E-03	1.948E+00	9.981E+02	2.260E+02	2.886E+01		
	P 3 YY	4.033E-02	1.074E-03	4.666E-03	4.666E-01	9.953E+01	2.259E+02	2.883E+01		
1.500E+01	P 1 GL	2.028E+00	4.741E-06	2.054E-05	3.042E+01	1.481E+06	2.260E+02	2.889E+01	2.788E-02	1.338E+01
	P 2 XX	1.685E-01	5.833E-04	2.531E-03	2.531E+00	9.975E+02	2.259E+02	2.886E+01		
	P 3 YY	4.026E-02	1.394E-03	6.060E-03	6.060E-01	9.939E+01	2.258E+02	2.881E+01		
1.950E+01	P 1 GL	2.028E+00	6.163E-06	2.671E-05	3.955E+01	1.481E+06	2.260E+02	2.889E+01	3.624E-02	1.739E+01
	P 2 XX	1.683E-01	7.576E-04	3.289E-03	3.289E+00	9.967E+02	2.259E+02	2.885E+01		
	P 3 YY	4.017E-02	1.808E-03	7.868E-03	7.868E-01	9.921E+01	2.258E+02	2.879E+01		
2.536E+01	P 1 GL	2.028E+00	8.012E-06	3.472E-05	5.142E+01	1.481E+06	2.260E+02	2.889E+01	4.711E-02	2.261E+01
	P 2 XX	1.681E-01	9.838E-04	4.272E-03	4.272E+00	9.957E+02	2.259E+02	2.883E+01		
	P 3 YY	4.006E-02	2.344E-03	1.021E-02	1.021E+00	9.898E+01	2.257E+02	2.876E+01		

Table 4. (Continued)

TIME, YEARS	I.D.	MASS CORROSION RATE, G/Y	INCREMENT TO FRACTION RELEASED	CUMULATIVE FRACTION RELEASED	CUMULATIVE MASS RELEASED, G	MASS REMAINING, G	LENGTH, CM	RADIUS, CM	CUMULATIVE WATER FLOW, CUBIC METERS	CUMULATIVE REMOVABLE SILICA AT SATURATION, G
3.296E+01	P 1 GL	2.028E+00	1.042E-05	4.513E-05	6.684E+01	1.481E+06	2.260E+02	2.889E+01	6.124E-02	2.939E+01
	P 2 XX	1.679E-01	1.277E-03	5.549E-03	5.549E+00	9.945E+02	2.259E+02	2.882E+01		
	P 3 YY	3.991E-02	3.036E-03	1.325E-02	1.325E+00	9.868E+01	2.257E+02	2.872E+01		
4.285E+01	P 1 GL	2.028E+00	1.354E-05	5.867E-05	8.689E+01	1.481E+06	2.260E+02	2.889E+01	7.962E-02	3.821E+01
	P 2 XX	1.675E-01	1.657E-03	7.206E-03	7.206E+00	9.928E+02	2.258E+02	2.880E+01		
	P 3 YY	3.972E-02	3.928E-03	1.718E-02	1.718E+00	9.828E+01	2.256E+02	2.867E+01		
5.571E+01	P 1 GL	2.028E+00	1.760E-05	7.627E-05	1.130E+02	1.481E+06	2.260E+02	2.889E+01	1.035E-01	4.967E+01
	P 2 XX	1.671E-01	2.148E-03	9.354E-03	9.354E+00	9.906E+02	2.258E+02	2.877E+01		
	P 3 YY	3.948E-02	5.075E-03	2.225E-02	2.225E+00	9.777E+01	2.254E+02	2.860E+01		
7.242E+01	P 1 GL	2.028E+00	2.288E-05	9.916E-05	1.469E+02	1.481E+06	2.260E+02	2.889E+01	1.346E-01	6.457E+01
	P 2 XX	1.665E-01	2.783E-03	1.214E-02	1.214E+01	9.879E+02	2.257E+02	2.873E+01		
	P 3 YY	3.916E-02	6.544E-03	2.880E-02	2.880E+00	9.712E+01	2.252E+02	2.851E+01		
9.414E+01	P 1 GL	2.028E+00	2.975E-05	1.289E-04	1.909E+02	1.481E+06	2.260E+02	2.889E+01	1.749E-01	8.394E+01
	P 2 XX	1.658E-01	3.602E-03	1.574E-02	1.574E+01	9.843E+02	2.256E+02	2.869E+01		
	P 3 YY	3.875E-02	8.419E-03	3.721E-02	3.721E+00	9.628E+01	2.250E+02	2.840E+01		
1.224E+02	P 1 GL	2.028E+00	3.867E-05	1.676E-04	2.482E+02	1.481E+06	2.260E+02	2.889E+01	2.274E-01	1.091E+02
	P 2 XX	1.649E-01	4.657E-03	2.040E-02	2.040E+01	9.796E+02	2.255E+02	2.862E+01		
	P 3 YY	3.822E-02	1.080E-02	4.801E-02	4.801E+00	9.520E+01	2.247E+02	2.825E+01		
1.591E+02	P 1 GL	2.028E+00	5.027E-05	2.178E-04	3.226E+02	1.481E+06	2.260E+02	2.889E+01	2.956E-01	1.419E+02
	P 2 XX	1.637E-01	6.009E-03	2.641E-02	2.641E+01	9.736E+02	2.253E+02	2.854E+01		
	P 3 YY	3.756E-02	1.379E-02	6.180E-02	6.180E+00	9.382E+01	2.243E+02	2.806E+01		
2.068E+02	P 1 GL	2.028E+00	6.535E-05	2.832E-04	4.194E+02	1.481E+06	2.260E+02	2.889E+01	3.843E-01	1.844E+02
	P 2 XX	1.621E-01	7.737E-03	3.414E-02	3.414E+01	9.659E+02	2.251E+02	2.844E+01		
	P 3 YY	3.671E-02	1.752E-02	7.932E-02	7.932E+00	9.207E+01	2.239E+02	2.781E+01		
2.689E+02	P 1 GL	2.028E+00	8.495E-05	3.681E-04	5.452E+02	1.480E+06	2.260E+02	2.888E+01	4.996E-01	2.397E+02
	P 2 XX	1.601E-01	9.934E-03	4.408E-02	4.408E+01	9.559E+02	2.248E+02	2.831E+01		
	P 3 YY	3.563E-02	2.211E-02	1.014E-01	1.014E+01	8.986E+01	2.232E+02	2.749E+01		
3.496E+02	P 1 GL	2.028E+00	1.104E-04	4.786E-04	7.088E+02	1.480E+06	2.260E+02	2.888E+01	6.495E-01	3.117E+02
	P 2 XX	1.575E-01	1.271E-02	5.678E-02	5.678E+01	9.432E+02	2.245E+02	2.813E+01		
	P 3 YY	3.429E-02	2.766E-02	1.291E-01	1.291E+01	8.709E+01	2.224E+02	2.707E+01		
4.544E+02	P 1 GL	2.027E+00	1.436E-04	6.221E-04	9.214E+02	1.480E+06	2.260E+02	2.888E+01	8.443E-01	4.052E+02
	P 2 XX	1.543E-01	1.618E-02	7.296E-02	7.296E+01	9.270E+02	2.240E+02	2.791E+01		
	P 3 YY	3.263E-02	3.422E-02	1.633E-01	1.633E+01	8.367E+01	2.213E+02	2.653E+01		
5.907E+02	P 1 GL	2.027E+00	1.866E-04	8.087E-04	1.198E+03	1.480E+06	2.260E+02	2.888E+01	1.098E+00	5.267E+02
	P 2 XX	1.501E-01	2.047E-02	9.343E-02	9.343E+01	9.066E+02	2.234E+02	2.761E+01		
	P 3 YY	3.062E-02	4.174E-02	2.051E-01	2.051E+01	7.949E+01	2.199E+02	2.582E+01		
7.680E+02	P 1 GL	2.027E+00	2.426E-04	1.051E-03	1.557E+03	1.479E+06	2.260E+02	2.888E+01	1.427E+00	6.847E+02
	P 2 XX	1.450E-01	2.569E-02	1.191E-01	1.191E+02	8.809E+02	2.227E+02	2.723E+01		
	P 3 YY	2.821E-02	4.999E-02	2.550E-01	2.550E+01	7.450E+01	2.180E+02	2.490E+01		

Table 4. (Continued)

TIME, YEARS	I.D.	MASS CORROSION RATE, G/Y	INCREMENT TO FRACTION RELEASED	CUMULATIVE FRACTION RELEASED	CUMULATIVE MASS RELEASED, G	MASS REMAINING, G	LENGTH, CM	RADIUS, CM	CUMULATIVE WATER FLOW, CUBIC METERS	CUMULATIVE SILICA REMOVABLE AT SATURATION, G
9.984E+02	P 1 GL	2.027E+00	3.153E-04	1.367E-03	2.024E+03	1.479E+06	2.260E+02	2.887E+01	1.855E+00	8.901E+02
	P 2 XX	1.385E-01	3.192E-02	1.510E-01	1.510E+02	8.490E+02	2.217E+02	2.673E+01		
	P 3 YY	2.539E-02	5.850E-02	3.135E-01	3.135E+01	6.865E+01	2.156E+02	2.370E+01		
1.298E+03	P 1 GL	2.027E+00	4.098E-04	1.776E-03	2.631E+03	1.478E+06	2.260E+02	2.887E+01	2.412E+00	1.157E+03
	P 2 XX	1.307E-01	3.914E-02	1.902E-01	1.902E+02	8.098E+02	2.204E+02	2.608E+01		
	P 3 YY	2.221E-02	6.652E-02	3.801E-01	3.801E+01	6.199E+01	2.125E+02	2.214E+01		
1.687E+03	P 1 GL	2.026E+00	5.327E-04	2.309E-03	3.420E+03	1.478E+06	2.259E+02	2.886E+01	3.135E+00	1.504E+03
	P 2 XX	1.212E-01	4.720E-02	2.374E-01	2.374E+02	7.626E+02	2.187E+02	2.524E+01		
	P 3 YY	1.875E-02	7.301E-02	4.531E-01	4.531E+01	5.469E+01	2.085E+02	2.012E+01		
2.193E+03	P 1 GL	2.026E+00	6.923E-04	3.001E-03	4.445E+03	1.477E+06	2.259E+02	2.885E+01	4.075E+00	1.956E+03
	P 2 XX	1.101E-01	5.571E-02	2.931E-01	2.931E+02	7.069E+02	2.165E+02	2.414E+01		
	P 3 YY	1.517E-02	7.679E-02	5.299E-01	5.299E+01	4.701E+01	2.032E+02	1.749E+01		
2.851E+03	P 1 GL	2.025E+00	8.997E-04	3.901E-03	5.778E+03	1.475E+06	2.259E+02	2.884E+01	5.298E+00	2.542E+03
	P 2 XX	9.731E-02	6.403E-02	3.571E-01	3.571E+02	6.429E+02	2.137E+02	2.271E+01		
	P 3 YY	1.171E-02	7.706E-02	6.069E-01	6.069E+01	3.931E+01	1.964E+02	1.407E+01		
3.707E+03	P 1 GL	2.024E+00	1.169E-03	5.070E-03	7.509E+03	1.473E+06	2.259E+02	2.882E+01	6.888E+00	3.305E+03
	P 2 XX	8.324E-02	7.120E-02	4.283E-01	4.283E+02	5.717E+02	2.099E+02	2.086E+01		
	P 3 YY	8.731E-03	7.468E-02	6.816E-01	6.816E+01	3.184E+01	1.875E+02	9.625E+00		
4.819E+03	P 1 GL	2.023E+00	1.519E-03	6.589E-03	9.758E+03	1.471E+06	2.258E+02	2.880E+01	8.954E+00	4.297E+03
	P 2 XX	6.839E-02	7.606E-02	5.044E-01	5.044E+02	4.956E+02	2.051E+02	1.845E+01		
	P 3 YY	7.538E-03	8.382E-02	7.654E-01	7.654E+01	2.346E+01	1.759E+02	3.846E+00		
6.265E+03	P 1 GL	2.021E+00	1.973E-03	8.562E-03	1.268E+04	1.468E+06	2.258E+02	2.878E+01	1.164E+01	5.585E+03
	P 2 XX	5.366E-02	7.758E-02	5.820E-01	5.820E+02	4.180E+02	1.989E+02	1.532E+01		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
8.144E+03	P 1 GL	2.019E+00	2.562E-03	1.112E-02	1.647E+04	1.465E+06	2.257E+02	2.875E+01	1.513E+01	7.261E+03
	P 2 XX	4.032E-02	7.577E-02	6.577E-01	6.577E+02	3.423E+02	1.907E+02	1.125E+01		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
1.059E+04	P 1 GL	2.016E+00	3.326E-03	1.445E-02	2.140E+04	1.460E+06	2.256E+02	2.870E+01	1.967E+01	9.439E+03
	P 2 XX	3.118E-02	7.617E-02	7.339E-01	7.339E+02	2.661E+02	1.802E+02	5.964E+00		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
1.376E+04	P 1 GL	2.013E+00	4.316E-03	1.877E-02	2.779E+04	1.453E+06	2.255E+02	2.865E+01	2.557E+01	1.227E+04
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
1.789E+04	P 1 GL	2.008E+00	5.598E-03	2.436E-02	3.608E+04	1.445E+06	2.254E+02	2.858E+01	3.324E+01	1.595E+04
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
2.326E+04	P 1 GL	2.002E+00	7.254E-03	3.162E-02	4.683E+04	1.434E+06	2.252E+02	2.848E+01	4.322E+01	2.074E+04
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		

Table 4. (Continued)

TIME, YEARS	I.D.	MASS CORROSION RATE, G/Y	INCREMENT TO FRACTION RELEASED	CUMULATIVE FRACTION RELEASED	CUMULATIVE MASS RELEASED, G	MASS REMAINING, G	LENGTH, CM	RADIUS, CM	CUMULATIVE WATER FLOW, CUBIC METERS	CUMULATIVE SILICA REMOVABLE AT SATURATION, G
3.024E+04	P 1 GL	1.993E+00	9.391E-03	4.101E-02	6.074E+04	1.420E+06	2.249E+02	2.836E+01	5.618E+01	2.696E+04
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
3.931E+04	P 1 GL	1.982E+00	1.214E-02	5.315E-02	7.872E+04	1.402E+06	2.246E+02	2.820E+01	7.304E+01	3.505E+04
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
5.110E+04	P 1 GL	1.968E+00	1.567E-02	6.883E-02	1.019E+05	1.379E+06	2.242E+02	2.799E+01	9.495E+01	4.556E+04
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
6.643E+04	P 1 GL	1.950E+00	2.019E-02	8.901E-02	1.318E+05	1.349E+06	2.237E+02	2.772E+01	1.234E+02	5.923E+04
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
8.636E+04	P 1 GL	1.927E+00	2.593E-02	1.149E-01	1.702E+05	1.311E+06	2.230E+02	2.737E+01	1.605E+02	7.700E+04
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
1.073E+05	P 1 GL	1.895E+00	2.678E-02	1.417E-01	2.099E+05	1.271E+06	2.222E+02	2.701E+01	1.993E+02	9.566E+04
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
1.293E+05	P 1 GL	1.862E+00	2.762E-02	1.693E-01	2.508E+05	1.230E+06	2.215E+02	2.662E+01	2.402E+02	1.152E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
1.523E+05	P 1 GL	1.827E+00	2.846E-02	1.978E-01	2.929E+05	1.188E+06	2.207E+02	2.622E+01	2.830E+02	1.358E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
1.766E+05	P 1 GL	1.791E+00	2.930E-02	2.271E-01	3.363E+05	1.145E+06	2.198E+02	2.579E+01	3.281E+02	1.574E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
2.020E+05	P 1 GL	1.753E+00	3.012E-02	2.572E-01	3.809E+05	1.100E+06	2.189E+02	2.535E+01	3.753E+02	1.801E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
2.287E+05	P 1 GL	1.714E+00	3.092E-02	2.881E-01	4.267E+05	1.054E+06	2.180E+02	2.488E+01	4.249E+02	2.039E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
2.567E+05	P 1 GL	1.674E+00	3.169E-02	3.190E-01	4.737E+05	1.007E+06	2.170E+02	2.438E+01	4.770E+02	2.289E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
2.862E+05	P 1 GL	1.631E+00	3.243E-02	3.523E-01	5.217E+05	9.593E+05	2.160E+02	2.387E+01	5.318E+02	2.552E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.

Table 4. (Continued)

TIME, YEARS	I.D.	MASS CORROSION RATE, G/Y	INCREMENT TO FRACTION RELEASED	CUMULATIVE FRACTION RELEASED	CUMULATIVE MASS RELEASED, G	MASS REMAINING, G	LENGTH, CM	RADIUS, CM	CUMULATIVE WATER FLOW, CUBIC METERS	CUMULATIVE REMOVABLE SILICA AT SATURATION, G
3.171E+05	P 1 GL	1.587E+00	3.313E-02	3.854E-01	5.700E+05	9.102E+05	2.149E+02	2.333E+01	5.892E+02	2.827E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
3.496E+05	P 1 GL	1.541E+00	3.378E-02	4.192E-01	6.208E+05	8.602E+05	2.137E+02	2.276E+01	6.495E+02	3.117E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
3.837E+05	P 1 GL	1.494E+00	3.438E-02	4.535E-01	6.717E+05	8.093E+05	2.125E+02	2.216E+01	7.129E+02	3.421E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
4.194E+05	P 1 GL	1.444E+00	3.490E-02	4.884E-01	7.234E+05	7.576E+05	2.113E+02	2.153E+01	7.794E+02	3.740E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
4.570E+05	P 1 GL	1.393E+00	3.535E-02	5.238E-01	7.757E+05	7.053E+05	2.100E+02	2.087E+01	8.492E+02	4.075E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
4.965E+05	P 1 GL	1.340E+00	3.570E-02	5.595E-01	8.286E+05	6.524E+05	2.086E+02	2.018E+01	9.225E+02	4.427E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
5.379E+05	P 1 GL	1.285E+00	3.594E-02	5.954E-01	8.818E+05	5.992E+05	2.071E+02	1.945E+01	9.995E+02	4.796E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
5.814E+05	P 1 GL	1.228E+00	3.607E-02	6.315E-01	9.353E+05	5.457E+05	2.056E+02	1.869E+01	1.080E+03	5.184E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
6.271E+05	P 1 GL	1.169E+00	3.606E-02	6.676E-01	9.887E+05	4.923E+05	2.040E+02	1.789E+01	1.165E+03	5.591E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
6.751E+05	P 1 GL	1.108E+00	3.589E-02	7.034E-01	1.042E+06	4.392E+05	2.023E+02	1.705E+01	1.254E+03	6.019E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
7.254E+05	P 1 GL	1.045E+00	3.555E-02	7.390E-01	1.094E+06	3.866E+05	2.005E+02	1.616E+01	1.348E+03	6.468E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
7.783E+05	P 1 GL	9.805E-01	3.501E-02	7.740E-01	1.146E+06	3.347E+05	1.987E+02	1.523E+01	1.446E+03	6.939E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.
8.338E+05	P 1 GL	9.136E-01	3.425E-02	8.082E-01	1.197E+06	2.840E+05	1.967E+02	1.426E+01	1.549E+03	7.434E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.	0.	0.
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.	0.	0.

Table 4. (Continued)

TIME, YEARS	I.D.	MASS CORROSION RATE, G/Y	INCREMENT TO FRACTION RELEASED	CUMULATIVE FRACTION RELEASED	CUMULATIVE MASS RELEASED, G	MASS REMAINING, G	LENGTH, CM	RADIUS, CM	CUMULATIVE WATER FLOW, CUBIC METERS	CUMULATIVE REMOVABLE SILICA AT SATURATION, G
8.921E+05	P 1 GL	8.447E-01	3.325E-02	8.415E-01	1.246E+06	2.347E+05	1.947E+02	1.324E+01	1.658E+03	7.954E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
9.533E+05	P 1 GL	7.735E-01	3.197E-02	8.735E-01	1.294E+06	1.874E+05	1.925E+02	1.216E+01	1.771E+03	8.500E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
1.018E+06	P 1 GL	6.998E-01	3.037E-02	9.038E-01	1.339E+06	1.424E+05	1.903E+02	1.103E+01	1.891E+03	9.073E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
1.085E+06	P 1 GL	6.228E-01	2.838E-02	9.322E-01	1.381E+06	1.004E+05	1.879E+02	9.851E+00	2.016E+03	9.675E+05
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
1.156E+06	P 1 GL	5.408E-01	2.588E-02	9.581E-01	1.419E+06	6.206E+04	1.854E+02	8.608E+00	2.148E+03	1.031E+06
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
1.230E+06	P 1 GL	4.478E-01	2.250E-02	9.806E-01	1.452E+06	2.874E+04	1.828E+02	7.302E+00	2.286E+03	1.097E+06
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
1.309E+06	P 1 GL	3.157E-01	1.665E-02	9.973E-01	1.477E+06	4.072E+03	1.801E+02	5.931E+00	2.431E+03	1.167E+06
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.		
1.391E+06	P 1 GL	0.	0.	1.000E+00	1.481E+06	0.	1.772E+02	4.492E+00	2.584E+03	1.240E+06
	P 2 XX	0.	0.	1.000E+00	1.000E+03	0.	0.	0.		
	P 3 YY	0.	0.	1.000E+00	1.000E+02	0.	0.	0.

Figure 19. Computed cumulative releases of mass from phases of a composite waste form.

explored by determining the stabilities of single-phase materials. It is evident that some careful experimental work is needed to provide a satisfactory basis for the interpretation and extrapolation of leach behavior. While it appears possible to design static leaching experiments that could greatly clarify dissolution processes, they should examine relatively simple systems with the objective of determining thermodynamic stabilities. Accurate values thus obtained can be used to resolve a variety of dissolution questions in diverse systems.

Eventually, it will also be necessary to examine how stabilities toward water leaching are affected by changes in solid-phase compositions; that is, how the substitution of other elements in the solid solutions (of which these are only end members) affect the stabilities and what the dissolved concentrations of such substituents would be at equilibrium. The dissolution of these phases with stoichiometries that can be formed with various waste compositions and loadings will, no doubt, turn out to be a complex problem. Some

variability in the composition of the waste stream during processing is to be expected. It is important to determine that such variability does not lead to unacceptable changes in release rates.

Calculations like those we have carried out provide no information on absolute leach rates or on the rate of approach to equilibrium. They suggest what data to look for experimentally, how difficult they may be to obtain, and what can be done with them.

We believe that the performance model we have developed would be a useful attachment to risk-analysis codes and would provide release functions that are more closely and continuously coupled to experimental data. We recognize that many of the initial assumptions invoked in this model are oversimplifications. The chemistry of silica, for example, is much more complex than indicated. Its activity in the solid is likely to be considerably lower than that in a pure silica phase, resulting in a lower aqueous concentration at saturation. However, we believe that the model

can be readily revised to provide improved release estimates as more chemical details become available.

The logical extension of this model is to add thermochemical and experimental kinetic data for other waste-form phases. The simple, generalized leach-rate functions we have used as a data base can be replaced by any theoretical or empirical relations that may be discovered, of whatever degree of complexity. In addition, such relationships will almost certainly contain parameters known to MSC1, like groundwater flow rate and volume, system geometry, and temperature. Also, the mass fluxes from the various reactions cause chemical changes in the leaching system. Such changes, especially those in solution, can be evaluated by chemical equilibrium codes. It appears feasible to couple MSC1 to such a code. This would give us the capability for system-specific calculations, allowing experimentally based leaching data to be applied more directly to repository release problems.

3.0 Risk Assessment

3.1 Introduction

The most probable mechanism for release of significant quantities of radionuclides to the biosphere from a closed deep geologic nuclear repository is groundwater transport. The process involves events that allow groundwater to contact the waste, leach the radionuclides, and, subsequently, transport them to the biosphere.

Predicting doses to individuals who live near a nuclear waste repository requires careful modeling of the repository excavation, its backfill, and its effects on surrounding hydrologic conditions. The availability of waste to individuals always starts with a radionuclide release from one of many spatially separated canisters in a repository. The transport of waste from each canister to an individual can occur by many pathways. If an observer were able to follow the migration of the many waste pulses, he would see a system of pulses moving through various pathways at different times and, finally, arriving at the biosphere release point.

"To provide decision makers with technically complete information, evaluating the hazards associated with the disposal of high level radioactive wastes in deep geologic strata demands that we acknowledge the uncertainties implicit in our predictions. These uncertainties by no means invalidate the findings of waste-disposal studies, but only by properly accounting for the uncertainties can we assure that conclusions and forecasts will stand up to criticism."²³

Not only are we faced with those uncertainties inherent in predicting the future and those associated with measurements of the present, but with the complexity of radionuclide transport in geologic media over long periods of time (at least 10^6 years). This complexity is such that simulation of the processes by computer modeling seems to be the most desirable means of analysis. However, mathematical computer models are still another source of uncertainty. How well do they actually model the processes? Answers to such questions are ascertained by exercising and validating the models.

The highly detailed, time-dependent, three-dimensional computer code required to properly couple all the processes—e.g., hydrology, meteorological recharge, heat conduction, waste-

canister stress and corrosion, waste-package leaching, water chemistry, repository refill and repressurization, nuclide transport, sorption, etc.—is not only complex, but will be too costly to run for very many simulations. Furthermore, in some cases (e.g., leaching or sorption), the processes may not be understood well enough to warrant detailed models.

The development of a performance-assessment technology should occur in three stages:

1. The development of individual models.
2. The development of coupled models.
3. The integration of coupled models into a modeling system.

The U.S. Geological Survey recently proposed a policy of model development.

"The procedure for developing and verifying individual models includes: (1) collecting laboratory and field observations of physical phenomena; (2) developing an empirical or theoretical working hypothesis; and (3) testing and verifying the working hypothesis. Details of the verification process vary greatly depending on the subject matter of the model. Far-field scenario analysis involves natural phenomena over long time periods; because such models are impossible to verify by direct observation, general agreement by peer review is necessary. In contrast, some models that are part of the near-field scenario analysis and the near-field consequence analysis can be verified by appropriate *in situ* tests. These tests may demonstrate the need for model modification and further observations of the physical system under different conditions from those used for model development.

"The process of observation and model modification to predict these observations will continue until a reasonable demonstration that predictions and observations are in agreement has been achieved. Peer review of the various steps used in model verification, from the basic physics used in model formulation to the comparison of predicted and measured test results, should lead to

model acceptance. In a few cases, it will be possible to verify radionuclide transport models by comparison of simulated results with field data from natural ore bodies.

"The general procedure for developing and verifying coupled models follows a similar process. However, the verification of the coupled models is more difficult because conducting the tests and interpreting the experimental results are more difficult.

"The integration of the coupled models into a modeling system involves the development of an "executive" model which manages the interactions of the coupled models. The isolation system performance model cannot be verified because system behavior cannot be observed or tested over the time period of

interest; however, data collected during the period of repository operation (approximately 40 years) will provide verification for much of the near-field analysis system."²⁴

We recognize that such a computer model is highly desirable (perhaps necessary) to do best-estimate simulations of "real-world" measurements. However, we can obtain a great deal of understanding about bounding calculations, uncertainty, and sensitivity by using simpler models.^{2,25-27} While these simpler models may be only qualitatively correct, relative measures of risk may be readily ascertained.

The systems analysis subtask of the Nuclear Waste Form Risk Assessment Project is to investigate the post-closure consequences associated with mined geologic repositories. Figure 20 shows the many subsystems that contribute to post-closure performance. Individually, these

Figure 20. Systems-level models link the various elements that contribute to overall systems performance.

subsystems can be modeled at several levels of detail (i.e., from detailed codes that reproduce experimental data to codes that model only dominant subsystem features). Each approach has relative merits for particular applications, and each should be pursued to ensure that the resulting analyses are, ultimately, in agreement.

The three-part LLNL approach has been: (1) to separate the system into physically distinct subsystems—waste-form package, repository site, regional aquifer, etc., (2) to identify and model the key features within each subsystem, and (3) to combine this understanding into an overall systems-level model to evaluate the performance of alternate waste forms in terms of radiological risk to man. The focus of the task was to calculate and assess the sensitivity of results to both individual input parameters and their uncertainties and then to quantify the resulting uncertainty associated with the consequences.

Sections 3.2 and 3.3, which follow, describe the codes, their applications to risk assessment, and the results obtained.

3.2 Probabilistic Systems Assessment of Deep Geologic Repositories

3.2.1 Introduction and General Description of MISER Network Hydrology and Radionuclide Transport Code

The MISER computer code was written to provide a computationally efficient analysis tool for studies of radionuclide release from closed geologic nuclear waste repositories. Because its development drew heavily on previous modeling efforts at LLNL and elsewhere,^{2,27} it is similar to previous models. By the same token, it has several significant differences.*

All of the referenced models have three basic assumptions:

1. Three-dimensional single-phase flow in a porous medium may be adequately approximated by a network of one-dimensional stream tubes.

2. Thermal energy, heat conduction, waste-canister stress, water chemistry and saturation, diffusion, and lateral dispersion may be neglected.

3. Flow is slow enough for instantaneous ion-exchange equilibrium to occur.

If one further assumes that the repository recharge is sufficiently fast, then a steady-state hydrology may be employed to decouple the hydrology and the nuclide transport.

The concept underlying the MISER code is that the user/analyst describes a general network of hydrologic flow paths with heads specified at boundary conditions (sources and sinks for flow). The user also specifies the radionuclide sources (beginnings of paths) and the risk-sensitive, nuclide-release observation points (ends of paths). Then the code traces "tree branches" of water flow from sources to sinks through all possible pathways before a subset of branches for nuclide transport is extracted from the full set.

The dimensions of the array are set by parameters, so the number of paths, branches, nuclide sources, and release points are, in theory, limited only by the size of the computer and the time available for computations.

Where appropriate, uncertainties are accounted for by using Monte Carlo techniques. A random-number generator selects the set of hydrologic, release, and transport parameters to be used at each trial on the basis of specified distribution types and ranges. Choices that do not satisfy the specified constraints are thrown out, and new values are chosen. Parameters specified as functional forms due to correlations are computed, the resulting equations are solved for the prescribed hazard/dose measurement, and the peak and time of peak of the measurement are added to a discretized output space. Regression techniques may be employed to estimate low-probability tails and confidence levels.

To increase computational efficiency, the Monte Carlo is nested in four levels of increasing numbers of computational steps. The hydrology requires a matrix solution of conservation and D'Arcy equations and is the outermost loop. Effective porosity and longitudinal dispersivity enter the next loop for computing water mean transit times and a unit impulse dispersion. Nuclide retardations are in the third loop and, finally, the waste-release mechanisms are in the innermost loop.

Details of the modules and their models appear in Sections 3.2.2 through 3.2.4. Sample applications are described in Section 3.2.5.

*MACRO,² for example, systematically samples all values of finite-probability distributions representing problem parameters and variables, which may lead to an inordinate number of calculations in a physically consistent system of constraints and correlations. MISER uses Monte Carlo techniques where appropriate. The modeling of MISER transport phenomena differs from that for either NUTRAN²⁵ or NWFT.²⁶

3.2.2 Hydrology Model

Under the simplifying assumptions of the preceding section, we solve the steady-state hydrology using D'Arcy's laws and conservation of water relationships. We assume a network of N paths, $N \geq 2$, and at least two boundary conditions.

The network of stream tubes are described by assuming that all flows are positive from the source(s) to sink(s) at which hydrologic heads are specified. Then, by interactive input, flows from each path to subsequent paths or boundary conditions are flagged in a "connector" matrix. The code then traces all "tree branches" from sources to sinks, producing a set of D'Arcy law equations.

$$\left\{ \begin{array}{l} \sum Q_n Z_n / K_n A_n = H_R^m - H_E^m : m=1,2,\dots,NB \\ \{n : \text{path } n \text{ is in branch } m\} \end{array} \right\} \quad (7)$$

where

- NB = number of tree branches in the network
- Q_n = D'Arcy flux
- K_n = permeability
- A_n = cross-sectional area
- Z_n = path length
- H_E^m = boundary head at exit of branch m
- H_R^m = boundary head at root of branch m
- n = path number.

The conservation of water at path junctions also produces a set of equations

$$\left\{ \begin{array}{l} \sum Q_i \\ \{i : \text{path } i \text{ flows to junction } m\} \end{array} \right\} = \sum Q_j \quad , \quad (8)$$

$\{j : \text{path } j \text{ flows from junction } m\}$

where $m=1, 2, \dots, NJ$; NJ = number of path junctions in the network. In general, $NB + NJ \geq N$, where N is the number of paths (equal number of unknowns), but a linearly independent set of N equations can be chosen. We use the LLNL MATHLIB²⁸ routines to obtain a rapid solution of the $NB + NJ$ equations for the unknowns $Q_1, Q_2, \dots, Q_N$. If any Q_n is computed to be negative, the code redefines the connector matrix and tree branches so all resulting flows are positive. An example will clarify the hydrology solution methodology. Suppose we wish to solve the 16-path network of Fig. 21. We assume boundary-condition heads so that $H_R^1 > H_E^1$, $H_R^2 > H_E^2$, $H_R^1 < H_R^2$, and $H_E^1 < H_E^2$, i.e., flow is generally upward and to the right, with the exception of path 8, which flows to the left.

Figure 21. Sample hydrology network of stream-tube flow paths. Arrows indicate assumed direction of flow. Radionuclide sources are at S_1 and S_2 and risk-sensitive observation points are at O_1 and O_2 .

The connector matrix is defined by the user input of

path 1 flow to 2
path 2 flow to 3
path 3 flow to 4
path 4 flow to B.C.

· ·
· ·
· ·

path 10 flow to 8,6,9

· ·
· ·
· ·

path 16 flow to B.C.

The code then traces the branches

1 → 2 → 3 → 4
14 → 12 → 10 → 8 → 5 → 2 → 3 → 4
14 → 12 → 10 → 6 → 3 → 4
14 → 12 → 10 → 9 → 7 → 4
14 → 15 → 13 → 11 → 7 → 4
14 → 15 → 16

for a total of six branches, i.e., NB=6.

There are 10 junctions, NJ=10, in the network, resulting in the conservation equations

$$\begin{aligned} Q_1 + Q_5 &= Q_2 \\ Q_2 + Q_6 &= Q_3 \\ Q_3 + Q_7 &= Q_4 \\ Q_8 &= Q_5 \\ Q_{10} &= Q_8 + Q_6 + Q_9 \\ Q_9 + Q_{11} &= Q_7 \\ Q_{12} &= Q_{10} \\ Q_{13} &= Q_{11} \\ Q_{14} &= Q_{12} + Q_{15} \\ Q_{15} &= Q_{13} + Q_{16} \end{aligned}$$

The user supplies parameters for Z_n , K_n , and A_n , which may be uncertain; in that case, they are selected using the aforementioned Monte Carlo techniques. Then, letting $x_n = Z_n/K_n A_n$, the codes sets up the matrix equation

$$\begin{bmatrix} X_1 & X_2 & X_3 & X_4 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & X_2 & X_3 & X_4 & X_5 & 0 & 0 & X_8 & 0 & X_{10} & 0 & X_{12} & 0 & X_{14} & 0 & 0 \\ 0 & 0 & X_3 & X_4 & 0 & X_6 & 0 & 0 & 0 & X_{10} & 0 & X_{12} & 0 & X_{14} & 0 & 0 \\ 0 & 0 & 0 & X_4 & 0 & 0 & X_7 & 0 & X_9 & X_{10} & 0 & X_{12} & 0 & X_{14} & 0 & 0 \\ 0 & 0 & 0 & X_4 & 0 & 0 & X_7 & 0 & 0 & 0 & X_{11} & 0 & X_{13} & X_{14} & X_{15} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & X_{14} & X_{15} & X_{16} \\ 1 & -1 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & -1 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & -1 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -1 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -1 & 0 & -1 & -1 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -1 & 0 & 1 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -1 & 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -1 & 0 & 1 & -1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -1 & 0 & 1 & -1 & 0 \end{bmatrix} \begin{bmatrix} Q_1 \\ Q_2 \\ Q_3 \\ Q_4 \\ Q_5 \\ Q_6 \\ Q_7 \\ Q_8 \\ Q_9 \\ Q_{10} \\ Q_{11} \\ Q_{12} \\ Q_{13} \\ Q_{14} \\ Q_{15} \\ Q_{16} \end{bmatrix} = \begin{bmatrix} H_R^1 - H_E^1 \\ H_R^1 - H_E^2 \\ H_R^1 - H_E^3 \\ H_R^1 - H_E^4 \\ H_R^1 - H_E^5 \\ H_R^2 - H_E^2 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix} \quad (9)$$

which is solved for Q_n , $n=1, 16$.

Suppose that the permeabilities were such that, in path 9, the resulting flow were negative. Then, the connector matrix and tree branches would be altered to reflect that the branch

14 → 12 → 10 → 9 → 7 → 4

becomes

14 → 15 → 13 → 11 → 9 → 6 → 3 → 4 and

14 → 15 → 13 → 11 → 9 → 8 → 5 → 2 → 3 → 4.

The conservation laws are

$$Q_{11} = Q_9 + Q_7 \text{ (not } Q_9 + Q_{11} = Q_7 \text{)}$$

and

$$Q_9 + Q_{10} = Q_6 + Q_8 \text{ (not } Q_{10} = Q_6 + Q_8 + Q_9 \text{)}.$$

This gives us 17 equations and 16 unknowns. However, if we subtract row 3 from row 2 of the matrix, we get just the difference of the last two D'Arcy equations. Thus, a dependent set and one equation may be eliminated to produce a solution.*

The conservation laws are used to compute the water-split fractions, f_m , at each junction to complete the steady-state hydrology solution.

The validity of this stream-tube approximation to a real three-dimensional hydrology should be checked using results from finite-element or finite-difference codes, as well as experimental data. Obviously, if the stream-tubes chosen are far from reality, the approximation will be poor. On the other hand, the size and expense of many computer runs almost precludes consideration of the three-dimensional codes for sensitivity and uncertainty analyses. Further, the uncertainties associated with the measurement and chemical properties of nuclide transport may overwhelm the hydrology model uncertainty.

3.2.3 Nuclide Transport Model

General Procedure. Our general procedure for transport of radionuclides is to extract from the set of hydrology tree branches that subset that leads from all nuclide sources to all risk-sensitive ob-

servation points. Then we apply the results of the waste-pipe calculus^{29,30} to each series of paths in a branch to approximate the waste flux at the release points. The flux pulses are accumulated on a discretized time line, then converted to hazard or dose measures. The waste is assumed to split in the same fractions as the water at each junction in the network.

Radioactive decay is not used explicitly in our transport model. We first compute a flux of nuclide groups as a function of time. Then we use tabulated results from the ORIGEN isotope and depletion code³¹ as multipliers in the dose/hazard calculations described in Section 3.2.4.

Using our example from the preceding section and assuming nuclide sources at S_1 and S_2 , with observation points at O_1 and O_2 , we select the branches

$$\left. \begin{array}{l} 10 \rightarrow 8 \rightarrow 5 \\ 10 \rightarrow 8 \rightarrow 5 \rightarrow 2 \rightarrow 3 \rightarrow 4 \\ 10 \rightarrow 6 \rightarrow 3 \rightarrow 4 \\ 10 \rightarrow 9 \rightarrow 7 \rightarrow 4 \\ 11 \rightarrow 7 \rightarrow 4 \end{array} \right\} \begin{array}{l} \text{for } S_1 \text{ to } O_1 \\ \text{for } S_1 \text{ to } O_2 \\ \text{for } S_1 \text{ to } O_2 \\ \text{for } S_1 \text{ to } O_2 \\ \text{for } S_2 \text{ to } O_2. \end{array}$$

Now an observer located at O_1 , as a function of time, would see as many waste pulses as the number, N_1 , of canisters with different release times or durations located at S_1 . At O_2 , he would see $3 \times N_1 + N_2$ pulses, where N_2 is the number of canisters at S_2 . As the number of overlapping (in time) pulses increases, the exact form of each individual pulse becomes decreasingly important.

Transport Green's Function. For completeness, and to delineate the assumptions and approximations, we extract from Refs. 29, 30, and 32 the derivation of our transport models.

One of the fundamental equations governing the transport and dispersion of ions in one-dimensional flow through porous media is

$$\begin{aligned} \frac{\partial m_r}{\partial t} + \frac{\partial C_r}{\partial t} + v \frac{\partial C_r}{\partial x} &= \alpha v \frac{\partial^2 C_r}{\partial x^2} \\ &- \lambda_r C_r + \sum_{s \neq r} \lambda'_s C_s \\ &- \lambda_r m_r + \sum_{s \neq r} \lambda'_s m_s, \quad (10) \end{aligned}$$

*MISER only eliminates equations, if necessary, for storage purposes. We actually solve m equations for n unknowns, $m \geq n$, using the LLNL-MATHLIB²⁸ routines.

where

λ_r = radioactive decay constant of nuclide r
(= $\ln 2$ /half-life)

λ_s^r = production rate of nuclide r from decay of nuclide s

v = interstitial water flow velocity (= $Q/A\eta$, η is effective porosity)

α = longitudinal dispersion constant

m_r = amount of ion r sorbed on the solid per unit volume

C_r = concentration of nuclide r in the interstitial water

x = spatial coordinate

t = time.

We note that $v(\partial C_r/\partial x)$ is simple advection and that $\alpha v(\partial^2 C_r/\partial x^2)$ is a hydrodynamic dispersion term. Diffusion and lateral dispersion have been neglected.

Assume that flow is slow enough for instantaneous ion-exchange equilibrium to result and that the ion-exchange absorption isotherm is linear. Then,

$$m_r = \frac{\rho}{\eta} K_d C_r, \quad (11)$$

where

ρ = bulk density

η = effective porosity

K_d = distribution coefficient.

Assume that the retardation factor, B_r , is the same for all nuclides in the same decay chain.*

Then .

$$B_r = 1 + \frac{\rho}{\eta} K_d$$

and

$$m_r = \frac{\rho}{\eta} K_d C_r = (B_r - 1)C_r. \quad (12)$$

Substitution simplifies Eq. (10) to

$$B_r \frac{\partial C_r}{\partial t} + v \frac{\partial C_r}{\partial x} = \alpha v \frac{\partial^2 C_r}{\partial x^2} - B_r \lambda_r C_r + \sum_{s \neq r} B_s \lambda_s^r C_s^r. \quad (13)$$

Assume that the dissolution rate for the waste form is the same for all nuclides in the same decay chain. Let $I_r(t)$ be the total amount of nuclide r at time t that was originally emplaced in the repository. Then the decay equation for I_r is given by:

$$\frac{\partial I_r}{\partial t} = -\lambda_r I_r + \sum_{s \neq r} \lambda_s^r I_s. \quad (14)$$

Now let $c_r(x,t) = C_r(x,t)/I_r(t)$ be the concentration of nuclide r per unit inventory of waste, I_r . Then

$$\begin{aligned} \frac{\partial C_r}{\partial t} &= I_r \frac{\partial c_r}{\partial t} + c_r \frac{\partial I_r}{\partial t} \\ &= I_r \frac{\partial c_r}{\partial t} - c_r \left(\lambda_r I_r - \sum_{s \neq r} \lambda_s^r I_s \right) \end{aligned}$$

and

$$\frac{\partial C_r}{\partial x} = I_r \frac{\partial c_r}{\partial x}, \quad \frac{\partial^2 C_r}{\partial x^2} = I_r \frac{\partial^2 c_r}{\partial x^2},$$

which, when substituted in Eq. (13), gives

$$B_r \frac{\partial c_r}{\partial t} + v \frac{\partial c_r}{\partial x} = \alpha v \frac{\partial^2 c_r}{\partial x^2} + \frac{1}{I_r} \sum_s \lambda_s^r I_s (B_s c_s - B_r c_r). \quad (15)$$

We have assumed that $B_r = B_s$ in the same decay chain. Thus, for nuclides in the same chain, we may factor B_r from the last term, apply uniqueness, and arrive at the fundamental equation

$$\frac{\partial c_r}{\partial t} + \frac{v}{B_r} \frac{\partial c_r}{\partial x} - \alpha \frac{v}{B_r} \frac{\partial^2 c_r}{\partial x^2} = 0, \quad (16)$$

where $c_r = c_r(x,t)$ is the concentration of nuclide r at spatial point x and time t per unit inventory of r at t .

A reasonable assumption is to place the nuclides in three groups based on retardations, dissolutions, and decay chains. They are

1. Anions (e.g., ^{99}Tc , ^{129}I , and ^{14}C).
2. Cations (other fission products).
3. Actinides.

To model the one-dimensional nuclide transport, we must solve (or approximate) Eq. (16) for reasonable boundary conditions over a series of paths within each tree branch.

*As we learn more about retardation factors, we recognize that this assumption needs to be reexamined.

Since boundary conditions are often more easily set with mass flux than with concentration, we note that if $J(x,t)$ is the mass flux, then

$$J(x,t) = V(x,t) \left(c(x,t) - \alpha \frac{\partial c(x,t)}{\partial x} \right), \quad (17)$$

where V is the water volumetric flow rate. Differentiation and substitution in Eq. (16) gives the identical equation

$$\frac{\partial J_r}{\partial t} + \frac{v}{B_r} \frac{\partial J_r}{\partial x} - \alpha \frac{v}{B_r} \frac{\partial^2 J_r}{\partial x^2} = 0, \quad (18)$$

in terms of nuclide mass flux per unit inventory.

We will drop the subscript r for the rest of this discussion, keeping in mind that the equations apply to nuclides in the same decay chain or (approximately) in the same retardation group.

Let us assume the initial and boundary conditions

$J(x,0) = 0$, no nuclides are present initially.

$J(0,t) = \delta(t)$, unit impulse input at one end.

$J(x,t) \rightarrow 0$ as $x \rightarrow \infty$ (semi-infinite path).

The semi-infinite path assumption has been investigated by Oston.³³ Generally, results obtained by the semi-infinite path assumption fall between those of his "slug-flow" and "diffusion" boundary condition assumptions. In reality, we have none of the above. However, as the path length z becomes large compared to the dispersion constant α , results coalesce. If $z/\alpha > 10$, the results are almost indistinguishable.

The solution for unit impulse is a Green's function and has the form

$$J(x,t) = G(x,t) = \frac{x}{\left(\frac{4\pi v \alpha t^3}{B} \right)^{1/2}} \exp \left[- \frac{\left(x - \frac{v}{B} t \right)^2}{4\alpha \frac{v}{B} t} \right]. \quad (19)$$

For a path length of z , let us write

$$\tau = \frac{zB}{v} \text{ as the mean transit time for a pulse}$$

and

$$\sigma = \left(\frac{2\alpha}{z} \right)^{1/2} \tau \text{ as the dispersive spread of the pulse}$$

about the mean.

Then the Green's function becomes

$$G(\tau, \sigma; t) = \frac{1}{(2\pi)^{1/2} \sigma} \left(\frac{\tau}{t} \right)^{3/2} \exp \left[- \frac{(t-\tau)^2 \tau}{2\sigma^2 t} \right]. \quad (20)$$

If the path has flux input at $x=0$, given by $S(t)$, then, by the convolution theorem, the output flux, $J(\sigma, \tau; t)$, at $x=z$ is

$$J(\sigma, \tau; t) = \int_0^t S(t') G(t, \sigma; t-t') dt' \quad (21)$$

In Ref. 29, the Laplace transform theory is applied to show that for a series sequence of paths, $n=1, 2, \dots, N$, the transform of $J(\sigma, \tau; t)$ is a product of transforms, e.g.,

$$\overline{J(\sigma, \tau; s)} = \prod_{n=1}^N \overline{G(\sigma_n, \tau_n; s)} \overline{S(s)}, \quad (22)$$

where

$$\overline{G(\sigma_n, \tau_n; s)} = \int_0^\infty e^{-st} G(\sigma_n, \tau_n; t) dt,$$

and

$$\overline{S(s)} = \int_0^\infty e^{-st} S(t) dt,$$

where $S(t)$ is the initial source function, and the mean time and width parameters add as follows:

$$\left. \begin{aligned} \tau &= \sum_{n=1}^N \tau_n + \tau_0 \\ \sigma^2 &= \sum_{n=1}^N \sigma_n^2 + \sigma_0^2 \end{aligned} \right\}. \quad (23)$$

τ_0 and σ_0 are the mean and variance of the source function.

We are interested in the final mass transfer function $J(\sigma, \tau; t)$ at the end of a series of paths that is either the inverse Laplace transform of Eq. (22) or a convolution integral in time, e.g., for two paths,

$$J(\sigma, \tau; t) = \int_0^t dt'' \int_0^{t''} G(\sigma_2, \tau_2; t-t'') \times G(\sigma_1, \tau_1; t''-t') S(t') dt'.$$

Reference 29 suggests that the inverse transform may be obtained by the method of steepest descent. In either case, computation time will be costly; therefore, we chose to use approximate methods.

O'Connell's extensions to the waste-pipe calculus³⁰ shows that if we assume (1) that the flow-path network is a single-series path and (2) that the source term is narrow in width and drops off more quickly for high times t , compared to the overall (convoluted) Green's function, then

$$J(\sigma, \tau; t) \approx G(\sigma, \tau; t),$$

which is Eq. (20), with σ and τ as defined by Eq. (23).

As indicated in Ref. 30, this approximation has the same first and second moments as the exact function within our assumptions.* We note that the approximation is still valid if assumption (2) does not hold, but we must perform the convolution integral.

The MISER code traces nuclide flows through the tree branches from nuclide sources to risk-sensitive observation points so that each branch is a series of sequential network paths. The appropriate fraction is computed based on water-flow fractions. The approximate Green's function is then convoluted with the source function and resulting flux accumulated on a discretized time line,

$$g(\sigma, \tau; t) = \frac{1}{(2\pi\sigma)^{1/2}} \exp\left[-\frac{(t - \tau)^2}{2\sigma^2}\right].$$

Further work needs to be done to validate transport models, particularly with respect to the near-field effects of repository design, placement of waste, and waste dissolution and release.

Waste Release and Convolution Models. It is obvious from the discussions of chemical modeling (Section 2) that waste dissolution and release is a major area of modeling uncertainty. MISER currently models the release as a step or Gaussian (or Gaussian-like) function; however, these may be limiting cases with reality somewhere between.

*One might observe that the Gaussian function also has the same first and second moments. In a system of many overlapping pulses, the exact form of the pulse is washed out and either of the above may also produce reasonable approximations. Since we are interested particularly in near-field scenarios, further studies in this area are necessary, particularly in the cases of $z/a < 10$.

For step-function release, we assume that, for each nuclide group r , the waste has a constant dissolution rate beginning at time TC and lasting for a period TD_r ; that is,

$$S_r(t) = \begin{cases} 0 & \text{for } t < TC \text{ and } t > (TC + TD_r) \\ \frac{\bar{I}_r}{TD_r} & \text{for } TC \leq t \leq (TC + TD_r) \end{cases} \quad (24)$$

where $\bar{I}_r$ is the path-fractional reduced inventory of group r and TC is a time that reflects the age of the waste at time of emplacement added to the time at which the canister is assumed to have breached because of corrosion.

We use the approximate Green's function of Eq. (20) and evaluate the convolution integral of Eq. (21). In this case, an analytical solution exists in terms of error functions. Further, if t is very large compared to the dissolution time, the step function may be approximated as a delta function. Then

$$J_r(\sigma_r, \tau_r; t) = \begin{cases} 0 & \text{if } t \leq TC \\ \frac{\bar{I}_r \tau_r}{TD_r} \varphi(\beta, \gamma) & \text{if } TC \leq t < (TC + TD_r) \\ \frac{\bar{I}_r \tau_r}{TD_r} \left\{ \varphi(\beta, \gamma) - \varphi\left(\beta, \gamma - \frac{TD_r}{\tau_r}\right) \right\} & \text{if } (TC + TD_r) < t < 10^3 TD_r \\ \bar{I}_r G(\tau_r, \sigma_r; t) & \text{if } 10^3 TD_r \leq t \end{cases},$$

where $\gamma = t/\tau_r$, $\beta = \sigma_r/\tau_r$, and

$$\varphi(\beta, \gamma) = \frac{1}{2} \left\{ \exp\left(\frac{2}{\beta^2}\right) \operatorname{erfc}\left[\frac{\gamma^2 + 1}{(2\beta)^{1/2} \gamma^{3/2}}\right] - \operatorname{erfc}\left[\frac{\gamma^2 - 1}{(2\beta)^{1/2} \gamma^{3/2}}\right] \right\}, \quad (25)$$

and erfc is the complementary error function.

Since we are going to accumulate the sums of many waste pulses on a discretized time line, we must produce, in a finite number of steps, a reasonable approximation of the flux for the MISER computer model. Further, the dose/hazard curves may vary rapidly in time so that the peak dose does not necessarily occur at the time of peak flux.

If we differentiate the Green's function, we find that the peak occurs at time

$$\hat{t}_r = \frac{\tau_r}{2} - 3\left(\frac{\sigma_r}{\tau_r}\right)^2 + \left\{ 9\left(\frac{\sigma_r}{\tau_r}\right)^4 + 4 \right\}^{1/2}.$$

Let $J_r = J_r(\sigma_r, \tau_r; t_i)$. Then we determine truncation limits by successive evaluations, using $0.9^m \hat{t}_r$ and $1.1^m \hat{t}_r$, $m=1, 2, \dots$ to find $\underline{m}$ and $\bar{m}$ so that

$$J_r(\sigma_r, \tau_r; 0.9^{\underline{m}} \hat{t}_r) < \epsilon \hat{J}_r < J_r(\sigma_r, \tau_r; 0.9^{\bar{m}-1} \hat{t}_r)$$

and

$$J_r(\sigma_r, \tau_r; 1.1^{\bar{m}} \hat{t}_r) < \epsilon \hat{J}_r < J_r(\sigma_r, \tau_r; 1.1^{\bar{m}+1} \hat{t}_r) ,$$

where ϵ is an arbitrary parameter (e.g., $\epsilon \approx 10^{-10}$).

The time range of interest is then set to

$$\underline{t} = 0.9^{\underline{m}} \hat{t}_r + TC$$

$$\bar{t} = 1.1^{\bar{m}} \hat{t}_r + TC + TD_r .$$

The leading portion is evaluated over $\underline{M}$ (≈ 20) time steps with $\Delta t = (\bar{t} - \underline{t})/(\underline{M} - 1)$, $t_1 = \underline{t}$, $t_m = t_{m-1} + \Delta t$, $m=2, 3, \dots, \underline{M}$. The remaining time $\bar{t}$ to $\bar{t}$ is subdivided using the algorithms

$$N = \max \left[\left(\left\lceil \frac{TD_r}{\sigma_r} \right\rceil + 1 \right) \underline{M}, 1000 - \underline{M} \right] ,$$

where $[TD_r/\sigma_r]$ is the integer part of TD_r/σ_r , and $\underline{N}$ (≈ 4) is a parameter.

If $N \Delta t \geq \bar{t} - \underline{t}$, set $\delta t = (\bar{t} - \underline{t})/(N - 1)$ and $R=1$; otherwise, set $\delta t = \Delta t$ and let $R=1.01^j$, where $j = \min \{1, 2, 3, \dots\}$ such that

$$\bar{t} - \underline{t} \leq \Delta t (R^{N-1} - 1)/(R - 1) ,$$

i.e., R is the ratio that is a minimum integral power of 1.01 to produce a geometric progression of time steps to span the time of interest in N steps. Then we compute $J_r(\sigma_r, \tau_r; t_i)$ by Eq. (25) for

$$\begin{aligned} t_1 &= \underline{t} \\ t_j &= t_{j-1} + \Delta t \text{ for } j=2, \dots, \underline{M}, \text{ and} \\ t_j &= t_{j-1} + \delta t R^{j-1}, j = \underline{M}+1, \dots, \underline{M}+N. \end{aligned}$$

Finally, if the discretized output time line is given by T_i , $i=1, 2, \dots, I$, we integral average the

$J_r(\sigma_r, \tau_r; t_i)$ to produce values to add to the existing waste flux.* That is,

$$J_r^k(T_i) = \sum J_r^k(\sigma_r, \tau_r; T_i) \quad (26)$$

{all branches leading to observation point k }

is the resulting flux of nuclide group r at observation point k .

If we assume that the waste release mechanism is "Gaussian-like," we could model the release as the Green's function, Eq. (20). In this case, we set $\sigma_r = TD_r/4$ and $\tau_r = TC + (TD_r/2)$ and merely add the terms as though the release were an additional path in the branch. We truncate $J_k(\sigma_r, \tau_r; t)$ at $\tau_r \pm 2 \sigma_r$ and compute over $\underline{M}$ (≈ 20) time steps. Finally, we sum these results as in Eq. (26).

The second model is much more computationally efficient because the tabular values for the error function do not enter. That is, the convolution integral need not be evaluated. However, we must remember that, in both cases, we are using only the approximate Green's function, which may not be valid if $\tau/\sigma < 10$.

3.2.4 Dose and Hazard Models

In general, the calculation of potential hazard or dose to man at any time, t , after release may be approximated by two pieces of information: (1) The value of the release function $J_r(t)$ in appropriate units of mass flow per year for nuclide type r , and (2) a hazard or dose function, $H_r(t)$, based on percent of inventory, toxicity, population, water use, and other human-related factors for nuclide type r .

The potential hazard or dose rate, $D(t)$, is calculated by

$$D(t) = \sum_r J_r(t) H_r(t) \quad (27)$$

*An option exists in MISER to move the time of peak flux, $\hat{t}$, to the nearest discretized output time T_i . In this case, the t_j are all shifted by the amount $T_i - \hat{t}$. Since we are primarily interested in peak doses, this shift helps compensate for sharp pulses that might otherwise be lost in the discretized output time line.

in appropriate units, e.g., in curies, rems, or relative units per year. The peak dose is then given by

$$\hat{D} = \max_t |D(t)| ,$$

occurring at time $\hat{t}$, and the total dose is given by

$$\underline{D} = \int_0^\infty D(t)dt .$$

In the preceding section, we showed the calculation of the release functions $J_r^k(T_i)$ for nuclide group r at observation point k on the discretized output time line, T_i .

In MISER, the discretized time line is user defined by

$$T_i = T_1 10^{(i-1)/m}, i=1, 2, \dots, I ,$$

where T_1 is the user-specified minimum time of interest. If $\bar{T}$ is the user-specified maximum time of interest, then I is computed so that $T_{I-1} \leq \bar{T} < T_I$. The number of points per decade, m , is also user specified.

In the case of Monte Carlo calculations, the hazard/dose measures are discretized in the same manner, e.g., we have

$$\hat{D}_j = \hat{D}_1 10^{(j-1)/m}, j=1, 2, \dots, I .$$

The outcome of each trial results in D and t for peak dose and time of peak. Then the counter for the bin (i,j) is incremented when

$$\hat{D}_{j-1/2} \leq \hat{D} < \hat{D}_{j+1/2}$$

and

$$T_{i-1/2} \leq t < T_{i+1/2} ,$$

where $x_{i+1/2} = x_1 10^{(2i-1)/2m}$ for $x = D$ or T . Results that fall outside the range of interest are also counted.

MISER currently allows for estimation of potential hazard or dose in the three models discussed in the following sections.

Curies/Year Potential Hazard Model. In this model, we do not consider any human use factors. A potential hazard is computed in curies per year or curies at each risk-sensitive observation point. The values for $H_r(t)$ are obtained by use of the ORIGEN code (ORNL Isotope Generation and Depletion Code).³¹

The ORIGEN code has many uses. However, it is used mainly to document the generation and depletion of nuclides in the operation of a nuclear reactor and in the subsequent decay of radioactivity after the fuel assembly has been removed. Provision is made for the reprocessing of spent fuel. The decay of spent fuel or reprocessing waste may be followed for an indefinite time.

The ORIGEN code uses the matrix exponential method to solve the linear differential equations for nuclear transmutation and decay that can be expressed as:

$$\dot{X} = A X(t) , \quad (28)$$

where

$$A = (B - I)\Lambda + (T - I)\Sigma\phi$$

B = decay transition matrix with b_{ij} = fraction of decays of nuclide j that produce nuclide i

I = identity matrix

T = transmutation matrix with t_{ij} = fraction of neutron captures with j to produce i

Λ and Σ = diagonal matrices whose i th elements are λ_i and σ_i , respectively with

λ_i = decay constant (s^{-1}) and

σ_i = capture cross sections ($cm^2/atom$)

$\dot{X}(t) = dx/dt$

ϕ = scalar flux (n/cm^2-s).

The solution is given by

$$X(t) = X(0) \exp(-At) , \quad (29)$$

using numerical techniques presented by Bell.³¹

From the ORIGEN output, we obtain a table of activity (curies) for each nuclide either in the original inventory or produced by decay as a function of time at selected time points. Table 5 is an example based on Savannah River Defense Waste Inventory. For three nuclide groups based on retardation factors, MISER sums the activities,

$$\left. \begin{aligned} H_1(t_i) &= a_{129}(t_i) + a_{99Tc}(t_i) + a_{14C}(t_i) \\ H_2(t_i) &= \sum_k a_k(t_i) \\ &\quad \{k: \text{other fission products}\} \\ H_3(t_i) &= \sum_k a_k(t_i) \\ &\quad \{k: \text{actinides}\} \end{aligned} \right\} \quad (30)$$

Table 5. Savannah River Laboratory defense waste (curies/canister). The first set is the 14 timesteps for the calculations. Remaining sets are the activity of the particular nuclide as a function of the 14 timesteps.

87	14				
sr1 defense waste-87 nuclides(5/23/88 memo-table 5)ci/canister					
0.000000e+00	1.000000e+03	3.000000e+00	1.000000e+01	3.000000e+01	
1.000000e+02	3.000000e+02	1.000000e+03	3.000000e+03	1.000000e+04	
3.000000e+04	1.000000e+05	3.000000e+05	1.000000e+06		
am243					
5.9228300e-03	5.9222700e-03	5.9211600e-03	5.9172600e-03	5.9061400e-03	
5.8673800e-03	5.7580400e-03	5.3911700e-03	4.4667400e-03	2.3124300e-03	
3.5249000e-04	4.8743900e-07	3.3013200e-15	8.4414900e-44		
am241					
3.5983500e+01	3.5981400e+01	3.5969600e+01	3.5860900e+01	3.5180300e+01	
3.1703900e+01	2.3010700e+01	7.4860100e+00	3.0260900e-01	4.0187300e-06	
4.6818700e-20	7.9893400e-69	0.	0.		
pu240					
4.5919600e+00	4.5914800e+00	4.5905000e+00	4.5871100e+00	4.5774100e+00	
4.5436300e+00	4.4484700e+00	4.1309500e+00	3.3428800e+00	1.5937000e+00	
1.9196500e-01	1.6432500e-04	7.4852300e-14	5.0433500e-46		
pu239					
7.3071300e+00	7.3069200e+00	7.3065000e+00	7.3050200e+00	7.3008200e+00	
7.2861100e+00	7.2442600e+00	7.0996600e+00	6.7022200e+00	5.4784100e+00	
3.0794200e+00	4.1003600e-01	1.2911300e-03	2.2620100e-12		
pu238					
4.4161100e+02	4.3813600e+02	4.3126900e+02	4.0807000e+02	3.4843600e+02	
2.0044100e+02	4.1294400e+01	1.6405300e-01	4.6420600e-08	3.2681400e-22	
0.0221000e-62	0.	0.	0.		
np237					
9.3872800e-03	9.3908300e-03	9.3979600e-03	9.4232000e-03	9.4963500e-03	
9.7432900e-03	1.0318000e-02	1.1341900e-02	1.1809200e-02	1.1802500e-02	
1.1726200e-02	1.1463400e-02	1.0744300e-02	8.5646200e-03		
u230					
3.0095900e-04	3.0095900e-04	3.0095900e-04	3.0095900e-04	3.0095900e-04	
3.0095900e-04	3.0095900e-04	3.0095900e-04	3.0095800e-04	3.0095800e-04	
3.0095700e-04	3.0095400e-04	3.0094400e-04	3.0091200e-04		
u235					
5.4637000e-05	5.4637000e-05	5.4637000e-05	5.4637000e-05	5.4637000e-05	
5.4637000e-05	5.4637000e-05	5.4637000e-05	5.4636900e-05	5.4636500e-05	
5.4635500e-05	5.4631700e-05	5.4620900e-05	5.4583300e-05		
u234					
1.3714400e-01	1.3839000e-01	1.4085300e-01	1.4917300e-01	1.7055700e-01	
2.2361100e-01	2.8055800e-01	2.9473400e-01	2.9313000e-01	2.8738400e-01	
2.7158200e-01	2.2282400e-01	1.2668000e-01	1.7945800e-02		
u233					
2.0271100e-06	2.0679900e-06	2.1497900e-06	2.4365700e-06	3.2601900e-06	
6.1916500e-06	1.4931700e-05	4.8135300e-05	1.4904600e-04	4.9925600e-04	
1.4388000e-03	4.1053600e-03	8.1423200e-03	9.0884600e-03		
pa231					
5.7956300e-08	5.9118700e-08	6.1443700e-08	6.9586400e-08	9.2865800e-08	
1.7483500e-07	4.1239500e-07	1.2830400e-06	4.0887500e-06	1.6992100e-05	
6.8166200e-05	2.2355600e-04	3.0315800e-04	3.0457000e-04		
th232					
2.9432400e-12	3.0021100e-12	3.1198400e-12	3.5318900e-12	4.7091900e-12	
8.8297200e-12	2.0602600e-11	6.1807200e-11	1.7953000e-10	5.9150500e-10	
1.7681000e-09	5.8807000e-09	1.7584100e-08	5.8003600e-08		
th230					
4.3675500e-05	4.5068700e-05	4.7487300e-05	5.6280900e-05	8.4017900e-05	
2.0493200e-04	6.5351500e-04	2.4176000e-03	7.4256000e-03	2.4065800e-02	
6.4649800e-02	1.4615900e-01	1.5519800e-01	2.6025200e-02		
th229					
4.7659800e-09	4.9569700e-09	5.3466200e-09	6.8583500e-09	1.2217300e-08	
4.3243700e-08	2.3953000e-07	2.2372600e-06	1.9056200e-05	1.7634000e-04	

Table 5. (Continued)

9.7363800e-04	3.7478000e-03	8.0147500e-03	9.1123600e-03	
ac227				
3.2817000e-08	3.3869200e-08	3.5998300e-08	4.3684800e-08	6.7158400e-08
1.5691900e-07	4.2511200e-07	1.4095200e-06	4.5836100e-06	1.9196700e-05
7.7215100e-05	2.5350600e-04	3.4384000e-04	3.4544300e-04	
ra228				
4.9132900e-12	5.03309900e-12	5.2665200e-12	6.0917700e-12	8.4531600e-12
1.6743700e-11	4.0612400e-11	1.2617300e-10	3.8628900e-10	1.4287900e-09
4.8773800e-09	1.7548700e-08	5.3679300e-08	1.7846200e-07	
ra226				
4.1271000e-07	4.3179600e-07	4.7149900e-07	6.2708100e-07	1.2244700e-06
5.4337800e-06	4.0233000e-05	4.4207600e-04	3.2510400e-03	1.8736600e-02
6.0391900e-02	1.4456700e-01	1.5576600e-01	2.6193500e-02	
pc210				
1.3419400e-07	1.4406300e-07	1.6192900e-07	2.3359200e-07	5.2852100e-07
2.5747200e-06	1.9152700e-05	1.9164500e-04	1.5453700e-03	6.8491100e-03
2.0437600e-02	3.5603200e-02	2.6098200e-02	3.6423100e-04	
pb210				
1.4211400e-07	1.5068600e-07	1.6880800e-07	2.4332300e-07	5.6372800e-07
3.3282500e-06	3.1331600e-05	4.0726100e-04	3.1916800e-03	1.8655000e-02
6.0308000e-02	1.4447500e-01	1.5564100e-01	2.5946200e-02	
eu154				
3.3868400e+00	3.1244000e+00	2.6589600e+00	1.5118900e+00	3.0128100e-01
1.0642800e-03	1.0509400e-10	3.1797300e-35	0.	0.
0.	0.	0.	0.	0.
sm151				
1.4267800e+02	1.4161700e+02	1.3952100e+02	1.3242400e+02	1.1407300e+02
6.7678700e+01	1.5228100e+01	8.2284200e-02	2.7363100e-08	5.8072500e-31
0.	0.	0.	0.	0.
cs137				
6.5723000e+03	6.4226900e+03	6.1335900e+03	5.2204500e+03	3.2937300e+03
6.5708200e+02	6.5678300e+00	6.5573400e-07	6.5277000e-27	0.
0.	0.	0.	0.	0.
cs135				
6.1875500e-02	6.1875500e-02	6.1875400e-02	6.1875300e-02	6.1874900e-02
6.1873600e-02	6.1869900e-02	6.1856800e-02	6.1819500e-02	6.1689200e-02
6.1318200e-02	6.0037400e-02	5.6523200e-02	4.5766400e-02	
1129				
2.0505100e-21	2.0505100e-21	2.0505100e-21	2.0505100e-21	2.0505000e-21
2.0505000e-21	2.0504800e-21	2.0504200e-21	2.0502400e-21	2.0496100e-21
2.0478200e-21	2.0415800e-21	2.0238500e-21	1.9629800e-21	
sn126				
1.5608200e-02	1.5608200e-02	1.5608200e-02	1.5608100e-02	1.5607900e-02
1.5607100e-02	1.5605000e-02	1.5597400e-02	1.5575800e-02	1.5506300e-02
1.5286800e-02	1.4562300e-02	1.2676000e-02	7.8004000e-03	
pd107				
9.4534000e-03	9.4533900e-03	9.4533900e-03	9.4533900e-03	9.4533700e-03
9.4532900e-03	9.4531000e-03	9.4523900e-03	9.4503700e-03	9.4433100e-03
9.4231800e-03	9.3530500e-03	9.1555500e-03	8.4945600e-03	
tc99				
2.5454500e+00	2.5454400e+00	2.5454200e+00	2.5453600e+00	2.5452000e+00
2.5446200e+00	2.5429600e+00	2.5371800e+00	2.5207100e+00	2.4639400e+00
2.3086900e+00	1.8383800e+00	9.5891100e-01	9.8282900e-02	
nb93m				
0.	0.	0.	0.	0.
0.	0.	0.	0.	0.
0.	0.	0.	0.	0.
zr93				
1.8786100e+00	1.8786100e+00	1.8786000e+00	1.8786000e+00	1.8785700e+00
1.8784700e+00	1.8782000e+00	1.8772400e+00	1.8745000e+00	1.8649400e+00

Table 5. (Continued)

1.8379100e+00	1.7463300e+00	1.5090700e+00	9.0519800e-01	
sr90				
5.6130500e+03	5.4762900e+03	5.2126800e+03	4.3860300e+03	2.6780300e+03
4.7634400e+02	3.4305200e+00	1.0874200e-07	4.0813500e-29	0.
0.	0.	0.	0.	
se79				
1.4059500e-01	1.4059400e-01	1.4059100e-01	1.4058000e-01	1.4055000e-01
1.4044500e-01	1.4014600e-01	1.3910300e-01	1.3616500e-01	1.2636500e-01
1.0207900e-01	4.8363900e-02	5.7229700e-03	3.2619500e-06	
ni59				
0.	0.	0.	0.	0.
0.	0.	0.	0.	0.
0.	0.	0.	0.	
c14				
0.	0.	0.	0.	0.
0.	0.	0.	0.	0.
0.	0.	0.	0.	
np239				
0.	0.	0.	0.	0.
0.	0.	0.	0.	0.
0.	0.	0.	0.	
ac225				
4.7472900e-09	4.9382400e-09	5.3275100e-09	6.8358800e-09	1.2187300e-08
4.3102400e-08	2.3770000e-07	2.1933500e-06	1.8838100e-05	1.7619900e-04
9.7363400e-04	3.7478000e-03	8.0147400e-03	9.1123600e-03	
ra225				
4.7548200e-09	4.9457900e-09	5.3352100e-09	6.8449400e-09	1.2199400e-08
4.3208600e-08	2.3944700e-07	2.2370000e-06	1.9055500e-05	1.7633800e-04
9.7363600e-04	3.7478000e-03	8.0147500e-03	9.1123600e-03	
ra223				
5.2701700e-08	3.3753000e-08	3.5880300e-08	4.3561400e-08	6.7019100e-08
1.5660300e-07	4.2274900e-07	1.3760600e-06	4.3070100e-06	1.6185800e-05
6.0833200e-05	1.9984700e-04	3.3051800e-04	3.4539900e-04	
sb126				
2.1851500e-03	2.1851500e-03	2.1851400e-03	2.1851300e-03	2.1851000e-03
2.1850000e-03	2.1846900e-03	2.1836300e-03	2.1806100e-03	2.1700400e-03
2.1401500e-03	2.0387200e-03	1.7746400e-03	1.0920600e-03	
y90				
5.6145200e+03	5.4777200e+03	5.2140400e+03	4.3871700e+03	2.6787300e+03
4.7646800e+02	3.4314100e+00	1.0877000e-07	4.0824100e-29	0.
0.	0.	0.	0.	
cm244				
1.2162000e-02	1.1705300e-02	1.0842700e-02	8.2942500e-03	3.8576600e-03
2.6469100e-04	1.2537400e-07	2.8996600e-19	1.6483700e-52	0.
0.	0.	0.	0.	
cm242				
0.8179000e-03	0.7777800e-03	0.6980900e-03	0.4248100e-03	7.6904200e-03
5.5887500e-03	2.2449900e-03	9.2227800e-05	1.0089200e-08	1.3814300e-22
3.3909200e-62	0.	0.	0.	
pu241				
3.5496900e+01	3.3862000e+01	3.0814500e+01	2.2151600e+01	8.6265000e+00
3.1793600e-01	2.5505600e-05	1.1794100e-19	1.3021000e-60	0.
0.	0.	0.	0.	
pm147				
6.2224600e-03	4.7774500e-03	2.8162100e-03	4.4290100e-04	2.2438700e-06
2.0769400e-14	2.3138200e-37	0.	0.	0.
0.	0.	0.	0.	
cel44				
4.8190400e-17	1.9785800e-17	3.3353300e-18	6.5597800e-21	1.2154900e-28
1.0526800e-55	0.	0.	0.	0.

Table 5. (Continued)

θ .	θ .	θ .	θ .	θ .
cs134				
2.1732100e-05	1.5522900e-05	7.9197200e-06	7.5126500e-07	8.9778900e-10
5.2969100e-20	3.1465600e-49	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
ru106				
2.0095600e-13	1.0118800e-13	2.5656000e-14	2.1056800e-16	2.3119600e-22
3.2067300e-43	θ .	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
nb95				
θ .	θ .	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
zr95				
θ .	θ .	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
y91				
θ .	θ .	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
sr89				
θ .	θ .	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
am242m				
5.0858300e-05	5.0626900e-05	5.0167300e-05	4.8591100e-05	4.4355400e-05
3.2233800e-05	1.2948300e-05	5.3193500e-07	5.8190700e-11	7.9675700e-25
1.9557500e-64	θ .	θ .	θ .	θ .
pu242				
6.1650200e-03	6.1650200e-03	6.1650100e-03	6.1649500e-03	6.1647900e-03
6.1641800e-03	6.1622500e-03	6.1547100e-03	6.1327100e-03	6.0562900e-03
5.8431800e-03	5.1546400e-03	3.6026600e-03	1.0282800e-03	θ .
u236				
1.1949400e-03	1.1950700e-03	1.1953500e-03	1.1963000e-03	1.1990100e-03
1.2084600e-03	1.2350700e-03	1.3238900e-03	1.5442300e-03	2.0331700e-03
2.4239300e-03	2.4724800e-03	2.4579200e-03	2.4075100e-03	θ .
co60				
5.0180400e-02	4.3931000e-02	3.3785200e-02	1.3422700e-02	9.6040400e-04
9.4103300e-08	3.3093100e-19	2.6989500e-59	θ .	θ .
θ .	θ .	θ .	θ .	θ .
rb87				
9.3369700e-06	9.3369700e-06	9.3369700e-06	9.3369700e-06	9.3369700e-06
9.3369700e-06	9.3369700e-06	9.3369700e-06	9.3369700e-06	9.3369700e-06
9.3369700e-06	9.3369500e-06	9.3369300e-06	9.3368300e-06	θ .
ag110				
2.0165100e-22	7.3839700e-23	9.9007700e-24	8.7396900e-27	1.6416800e-35
4.7163300e-66	θ .	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
fn115				
2.3181100e-25	2.3181100e-26	2.3181100e-26	2.3181100e-26	2.3181100e-26
2.3181100e-26	2.3181100e-26	2.3181100e-26	2.3181100e-26	2.3181100e-26
2.3181100e-26	2.3181100e-26	2.3181100e-26	2.3181100e-26	2.3181100e-26
sn121m				
1.2620200e-02	1.2454200e-02	1.2113400e-02	1.0992400e-02	8.3291200e-03
3.1540500e-03	1.9675500e-04	1.1929400e-08	1.0645900e-20	7.1470200e-63
θ .	θ .	θ .	θ .	θ .
sn123				
6.6644500e-45	9.3639800e-46	1.8486400e-47	1.9985900e-53	θ .
θ .	θ .	θ .	θ .	θ .

Table 5. (Continued)

θ .	θ .	θ .	θ .	θ .
sb129				
3.7291888e-04	2.8929288e-04	1.7418188e-04	2.9439488e-05	1.8347788e-07
3.5865888e-15	3.1882288e-37	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
sb126m				
1.5688288e-02	1.5688288e-02	1.5688288e-02	1.5688188e-02	1.5687988e-02
1.5687188e-02	1.5685888e-02	1.3413888e-02	1.3395288e-02	1.3338388e-02
1.3146688e-02	1.2523688e-02	1.8981488e-02	6.7883488e-03	
tel25m				
9.1866488e-05	7.8646588e-05	4.2516388e-05	7.1892588e-06	4.4885988e-08
8.5638288e-16	7.5788788e-38	θ .	θ .	θ .
θ .	θ .	θ .	θ .	θ .
cel42				
9.8588888e-06	9.8588888e-06	9.8588888e-06	9.8588888e-06	9.8588888e-06
9.8588888e-06	9.8588888e-06	9.8588888e-06	9.8588888e-06	9.8588888e-06
9.8588888e-06	9.8588888e-06	9.8588888e-06	9.8588888e-06	9.8588888e-06
nd144				
5.8869888e-18	5.8869888e-18	5.8869888e-18	5.8869888e-18	5.8869888e-18
5.8869888e-18	5.8869888e-18	5.8869888e-18	5.8869888e-18	5.8869888e-18
5.8869888e-18	5.8869888e-18	5.8869888e-18	5.8869888e-18	5.8869888e-18
sm147				
2.5868788e-06	2.5868788e-06	2.5868788e-06	2.5868788e-06	2.5868788e-06
2.5868788e-06	2.5868788e-06	2.5868788e-06	2.5868788e-06	2.5868788e-06
2.5868788e-06	2.5868788e-06	2.5868788e-06	2.5868788e-06	2.5868788e-06
sm143				
5.8187388e-12	5.8187388e-12	5.8187388e-12	5.8187388e-12	5.8187388e-12
5.8187388e-12	5.8187388e-12	5.8187388e-12	5.8187388e-12	5.8187388e-12
5.8187388e-12	5.8187388e-12	5.8187388e-12	5.8187388e-12	5.8187388e-12
sm149				
1.8811688e-12	1.8811688e-12	1.8811688e-12	1.8811688e-12	1.8811688e-12
1.8811688e-12	1.8811688e-12	1.8811688e-12	1.8811688e-12	1.8811688e-12
1.8811688e-12	1.8811688e-12	1.8811688e-12	1.8811688e-12	1.8811688e-12
eu152				
1.8839488e-01	1.8276288e-01	9.2361588e-02	6.3575188e-02	2.1878888e-02
5.2217888e-04	1.2118288e-08	7.2963888e-25	θ .	θ .
θ .	θ .	θ .	θ .	θ .
eu155				
7.2241788e-02	6.2521688e-02	4.6828888e-02	1.7829588e-02	9.4629988e-04
3.8276188e-08	1.8744788e-28	1.2592988e-64	θ .	θ .
θ .	θ .	θ .	θ .	θ .
b1218				
1.4852988e-07	1.4988888e-07	1.6693688e-07	2.4865688e-07	5.5769288e-07
3.2938888e-06	3.1814888e-05	4.8317388e-04	3.1597388e-03	1.8468488e-02
5.9784988e-02	1.4383888e-01	1.5488588e-01	2.5686888e-02	
b1218m				
4.8281288e-15	4.3529388e-15	5.8775688e-15	8.3324488e-15	2.5819288e-14
2.6824988e-13	5.3835588e-12	1.9141988e-18	5.8781888e-09	1.8814988e-07
1.8922888e-06	8.8537888e-06	3.9754388e-05	7.8285688e-05	
rn222				
4.1242688e-07	4.3158488e-07	4.7119288e-07	6.2671788e-07	1.2239388e-06
5.4324788e-06	4.8229888e-05	4.4286388e-04	3.2518188e-03	1.8736488e-02
6.8398888e-02	1.4455488e-01	1.5569488e-01	2.5954888e-02	
ra224				
4.1854188e-03	4.8659188e-03	3.9888688e-03	3.7271188e-03	3.8718688e-03
1.5613688e-03	2.2583788e-04	2.6168888e-07	1.9428988e-09	2.8359888e-09
5.9589488e-09	1.8268788e-08	5.3948288e-08	1.7846688e-07	
th227				
2.8756188e-08	2.9679788e-08	3.1548888e-08	3.8296888e-08	5.8988288e-08
1.3774188e-07	3.7388888e-07	1.2379388e-06	4.8258188e-06	1.6868988e-05

Table 5. (Continued)

6.7320000e-05	2.2266200e-04	3.0200500e-04	3.0341300e-04	
th228				
4.1048300e-03	4.0653400e-03	3.9875000e-03	3.7265900e-03	3.0714300e-03
1.5611400e-03	2.2580500e-04	2.6157300e-07	1.9532800e-09	2.9309500e-09
6.2088400e-09	1.8421600e-08	5.3940500e-08	1.7846600e-07	
th231				
5.4997100e-05	5.5004300e-05	5.5018700e-05	5.5069100e-05	5.5212900e-05
5.5715700e-05	5.7146800e-05	6.2091000e-05	7.5686200e-05	1.1752500e-04
1.9955000e-04	2.9080500e-04	3.0472200e-04	3.0455600e-04	
th234				
3.0095900e-04	3.0095900e-04	3.0095900e-04	3.0095900e-04	3.0095900e-04
3.0095900e-04	3.0095900e-04	3.0095900e-04	3.0096100e-04	3.0096700e-04
3.0098500e-04	3.0104100e-04	3.0116600e-04	3.0135700e-04	
pa233				
9.3872600e-03	9.3908100e-03	9.3979400e-03	9.4231700e-03	9.4963300e-03
9.7432800e-03	1.0318000e-02	1.1341900e-02	1.1809200e-02	1.1802500e-02
1.1726200e-02	1.1463400e-02	1.0744300e-02	8.5646200e-03	
u232				
3.9953100e-03	3.9560800e-03	3.8811100e-03	3.6271500e-03	2.9894700e-03
1.5194900e-03	2.1978000e-04	2.5451400e-07	1.5673400e-09	1.5025800e-09
1.3319400e-09	8.7346100e-10	2.6163700e-10	3.8486000e-12	
np236				
1.7920200e-08	1.7920100e-08	1.7919900e-08	1.7919100e-08	1.7917000e-08
1.7909400e-08	1.7887000e-08	1.7812500e-08	1.7599100e-08	1.6872000e-08
1.4955900e-08	9.8078000e-09	2.9378400e-09	4.3214600e-11	
pu236				
4.9966500e-08	3.9526500e-08	2.4919900e-08	5.8478100e-09	1.6275300e-09
1.5939300e-09	1.5920600e-09	1.5853500e-09	1.5663600e-09	1.5016400e-09
1.3311100e-09	8.7291600e-10	2.6147400e-10	3.8462000e-12	
am242				
1.0631200e-02	1.0582900e-02	1.0486800e-02	1.0157300e-02	9.2718900e-03
6.7380300e-03	2.7066500e-03	1.1119400e-04	1.2164000e-08	1.6655100e-22
4.0882300e-62	0.	0.	0.	
cm243				
1.0461400e-03	1.0210100e-03	9.7253000e-04	8.2028400e-04	5.0432500e-04
9.1901700e-05	7.0922900e-07	2.8635500e-14	2.1455900e-35	0.
0.	0.	0.	0.	
cm245				
6.8313900e-06	6.8302400e-06	6.8297200e-06	6.8258100e-06	6.8146500e-06
6.7757500e-06	6.6558100e-06	6.2949000e-06	5.3450000e-06	3.0151200e-06
5.8734800e-07	1.9162800e-09	1.5078200e-16	2.0606200e-41	
cm246				
5.4274100e-07	5.4266200e-07	5.4250300e-07	5.4194600e-07	5.4036000e-07
5.3484500e-07	5.1939700e-07	4.6875800e-07	3.4967100e-07	1.2535800e-07
6.6875800e-09	2.3451900e-13	4.3785000e-26	1.2314400e-70	
cm247				
6.7431800e-13	6.7431800e-13	6.7431800e-13	6.7431800e-13	6.7431700e-13
6.7431500e-13	6.7431000e-13	6.7428800e-13	6.7422900e-13	6.7401900e-13
6.7342000e-13	6.7132900e-13	6.6538900e-13	6.4501200e-13	
cm248				
7.0408500e-13	7.0408300e-13	7.0408000e-13	7.0407000e-13	7.0404100e-13
7.0394100e-13	7.0365400e-13	7.0264900e-13	6.9978800e-13	6.8986300e-13
6.6227700e-13	5.7412500e-13	3.8174200e-13	9.1504500e-14	

to produce the potential hazard functions for Eq. (27).

Figures 22 through 25 are plots of $H_1(t_i)$, $H_2(t_i)$, $H_3(t_i)$ and their summation for the nuclides of Table 5.

Since the t_i of the hazard tables need not agree with the T_i of our discretized time line, we linearly interpolate the t_i to obtain values of $H_n(T_i)$. If the time of interest extends beyond the 10^6 years covered by our ORIGEN tables, new tables must be obtained. To use more than three release/transport groups, one need only change the summation algorithms and provide storage for more than three groups. We note that this model produces only potential hazard in terms of activity; no human-use factors are considered.

BIODOSE Dose-To-Man Model. Human doses will depend on the movement of waste to wells or surface waters, the concentration in the ecosystem, human diets and living habits, and the biochemistry of the waste nuclides in the human body. These factors are modeled in the BIODOSE code^{25,32,34} to produce a set of tables $H_n(t_i)$ for use in Eq. (27) in the manner described previously. In this case, the results are in rem or man-rem. The following discussion is essentially "lifted" from Ref. 25.

Environmental assessment of future releases requires consideration of the cycling of long-lived radionuclides in the biosphere and the possibility of local buildup and concentration. Because of vast uncertainty about future conditions, the basis

Figure 22. Group-I activity as a function of time for Savannah River defense waste.

Figure 24. Group-III activity as a function of time for Savannah River defense waste.

Figure 23. Group-II activity as a function of time for Savannah River defense waste.

Figure 25. Total activity as a function of time for Savannah River defense waste.

of model predictions for times far into the future must be defined carefully. The basic philosophy of BIODOSE is to provide tools that can be used to evaluate the consequences of radionuclide release under conditions not too far removed from those experienced today.

Transport in surface water systems is calculated using a compartment or box model. Because the release of waste is expected to occur over long time periods, the surface water system is assumed to be in dynamic equilibrium with the input flow of the waste. This simplification allows one to calculate the doses per unit input flow of waste independent of the waste release function.

The transport model computes the concentrations of each radionuclide in soil, sediment, and water systems. The model is composed of compartments for each system and assumes uniform mixing within each compartment. The following dynamic processes, which affect concentration, are considered: dilution by mixing, diffusion between sediment and water, sedimentation, man's removal of water and food products, ion exchange, biogeochemical processes, and radioactive decay. The potential for accumulating radio-

nuclides in the topsoil because of the irrigation of crop lands is also modeled. The complexity of this model is limited by the accuracy with which the transfer coefficients between compartments can be modeled.

The concentrations calculated for the sediment, topsoil, and water subsystems are used with a simple ecosystem model to give radionuclide dose rates to man. Doses are assumed to be received by ingestion and external exposure. The ingestion pathways modeled are drinking water, aquatic foods, irrigated crops, and farm animals. External exposure to both the water subsystems and the topsoil is considered. The pathways considered are summarized in Fig. 26.

Doses for individuals and populations are computed as a function of living habits and usage. Although estimates for the local demography are not possible far into the future, reasonable limits to *potential* population exposure can be developed. For example, the population dose from the aquatic food pathway depends on the net harvest, not on the size of the local population. Similarly, the vegetable and animal pathways depend primarily on the irrigation rate. Since these pathways

Figure 26. Pathways modeled in BIODOSE.

are dominant, the potential population dose from a river system is relatively independent of the local demography. Population doses were, therefore, based on estimates of the usage rates of the water system rather than on population.

Tables 6 through 8 show dose values, based on the Columbia River system of usage, for the 53 significant nuclides per canister of Savannah

River defense waste. Figures 27 through 30 are time-dependent plots of dose to the liver of an individual using surface water.

Well-Release Scenario Dose-To-Man Model. The dose-to-man model is intended to display a performance measure sensitive to engineered features (including waste-form performance) in high-level nuclear waste repositories. This obviously

Table 6. Columbia River base-case calculation of potential dose to a worst-situated individual with an average diet and lifestyle (rem) for ^{129}I and ^{99}Tc (Group 1): SRP glass waste (curies/canister).³⁵

time	body equiv	body	gl - 111	thyroid	bone	liver	lung	kidney	skin
1.0	1.616e-06	9.218e-08	9.541e-06	5.989e-13	2.249e-07	3.045e-07	5.989e-13	5.989e-13	1.182e-12
1.0	1.684e-06	9.553e-08	1.036e-05	6.119e-13	2.134e-07	3.172e-07	6.119e-13	6.119e-13	1.224e-12
3.0	1.684e-06	9.553e-08	1.036e-05	6.119e-13	2.134e-07	3.172e-07	6.119e-13	6.119e-13	1.224e-12
10.0	1.693e-06	9.553e-08	1.036e-05	6.119e-13	2.134e-07	3.172e-07	6.119e-13	6.119e-13	1.224e-12
30.0	1.683e-06	9.553e-08	1.036e-05	6.118e-13	2.134e-07	3.172e-07	6.118e-13	6.118e-13	1.224e-12
1.00e+02	1.693e-06	9.551e-08	1.035e-05	6.117e-13	2.134e-07	3.171e-07	6.117e-13	6.117e-13	1.223e-12
3.00e+02	1.662e-06	9.545e-08	1.035e-05	6.113e-13	2.132e-07	3.169e-07	6.113e-13	6.113e-13	1.223e-12
1.00e+03	1.678e-06	9.526e-08	1.032e-05	6.099e-13	2.127e-07	3.162e-07	6.099e-13	6.099e-13	1.220e-12
3.00e+03	1.657e-06	9.470e-08	1.026e-05	6.059e-13	2.114e-07	3.141e-07	6.059e-13	6.059e-13	1.212e-12
1.00e+04	1.630e-06	9.280e-08	1.003e-05	5.923e-13	2.066e-07	3.070e-07	5.923e-13	5.923e-13	1.185e-12
3.00e+04	1.527e-06	7.758e-08	9.394e-06	5.550e-13	1.936e-07	2.877e-07	5.550e-13	5.550e-13	1.118e-12
1.00e+05	1.216e-06	6.177e-08	7.480e-06	4.419e-13	1.541e-07	2.291e-07	4.419e-13	4.419e-13	8.638e-13
3.00e+05	1.242e-07	3.222e-08	3.992e-06	2.305e-13	8.241e-08	1.195e-07	2.305e-13	2.305e-13	4.613e-13
1.00e+06	5.580e-08	3.383e-09	3.999e-07	2.363e-14	8.241e-09	1.225e-08	2.363e-14	2.363e-14	4.725e-14

Table 7. Columbia River base-case calculation of potential dose to a worst-situated individual with an average diet and lifestyle (rem) for fission products inventory type (Group II): SRP glass waste (curies/canister).³⁵

time	body equiv	body	gl - 111	thyroid	bone	liver	lung	kidney	skin
1.0	2.187e+00	3.591e+00	5.688e-01	2.139e-03	1.243e+01	1.147e+00	2.142e-03	4.861e-01	2.512e-03
1.0	2.415e+00	3.956e+00	6.341e-01	7.859e-03	1.391e+01	1.128e+00	7.853e-03	4.825e-01	9.256e-03
3.0	2.691e+00	4.396e+00	7.267e-01	1.593e-02	1.572e+01	1.898e+00	1.593e-02	3.948e-01	1.861e-02
10.0	2.762e+00	4.490e+00	7.757e-01	3.837e-02	1.628e+01	9.624e-01	3.837e-02	3.643e-01	4.481e-02
30.0	1.812e+00	2.925e+00	5.326e-01	5.775e-02	1.851e+01	6.539e-01	5.775e-02	2.688e-01	6.747e-02
1.00e+02	3.390e-01	5.412e-01	1.861e-01	2.113e-02	1.894e+00	1.438e-01	2.113e-02	6.441e-02	2.478e-02
3.00e+02	2.672e-03	4.289e-03	9.262e-04	2.526e-04	1.480e-02	1.494e-03	2.561e-04	6.937e-04	3.417e-04
1.00e+03	3.614e-06	2.191e-06	5.897e-06	1.824e-06	4.876e-06	5.387e-06	4.595e-06	5.885e-06	1.485e-06
3.00e+03	3.466e-06	2.895e-06	4.699e-06	9.388e-07	4.782e-06	5.168e-06	4.585e-06	5.630e-06	1.831e-06
1.00e+04	3.489e-06	2.861e-06	4.646e-06	9.334e-07	4.764e-06	4.991e-06	4.486e-06	5.333e-06	1.826e-06
3.00e+04	3.264e-06	1.973e-06	4.587e-06	9.286e-07	4.713e-06	4.549e-06	4.435e-06	4.586e-06	1.812e-06
1.00e+05	2.985e-06	1.758e-06	4.135e-06	8.769e-07	4.541e-06	3.543e-06	4.268e-06	2.948e-06	9.643e-07
3.00e+05	2.421e-06	1.475e-06	3.518e-06	7.633e-07	4.886e-06	2.598e-06	3.881e-06	1.552e-06	8.394e-07
1.00e+06	1.636e-06	1.826e-06	2.264e-06	4.697e-07	2.857e-06	1.973e-06	2.577e-06	1.847e-06	5.165e-07

Table 8. Columbia River base-case calculation of potential dose to a worst-situated individual with an average diet and lifestyle (rem) for actinide inventory type (Group III): SRP glass waste (curies/canister).³⁵

time	body equiv	body	gl - 111	thyroid	bone	liver	lung	kidney	skin
1.0	1.444e-03	5.171e-04	2.693e-03	4.599e-06	1.486e-02	3.209e-03	4.599e-06	2.989e-03	1.315e-05
1.0	1.438e-03	5.158e-04	2.678e-03	5.153e-06	1.398e-02	3.279e-03	5.153e-06	2.981e-03	1.418e-05
3.0	1.425e-03	5.132e-04	2.649e-03	6.252e-06	1.383e-02	3.258e-03	6.252e-06	2.966e-03	1.680e-05
10.0	1.383e-03	5.839e-04	2.549e-03	1.882e-05	1.332e-02	3.185e-03	1.882e-05	2.911e-03	2.242e-05
30.0	1.268e-03	4.775e-04	2.285e-03	2.882e-05	1.194e-02	2.977e-03	2.882e-05	2.758e-03	3.988e-05
1.00e+02	9.574e-04	4.811e-04	1.596e-03	4.655e-05	8.337e-03	2.369e-03	4.655e-05	2.259e-03	8.886e-05
3.00e+02	5.484e-04	2.868e-04	7.539e-04	7.565e-05	3.985e-03	1.457e-03	7.565e-05	1.473e-03	1.198e-04
1.00e+03	2.224e-04	1.574e-04	2.583e-04	4.357e-05	1.415e-03	4.987e-04	4.357e-05	5.286e-04	7.478e-05
3.00e+03	2.848e-04	3.158e-04	7.655e-05	8.384e-06	1.822e-03	8.838e-05	8.384e-06	9.559e-05	2.239e-05
1.00e+04	9.231e-04	1.598e-03	8.278e-05	2.866e-05	3.868e-03	1.786e-04	2.866e-05	8.225e-05	3.556e-05
3.00e+04	2.888e-03	5.396e-03	1.348e-04	5.541e-05	1.147e-02	4.773e-04	5.541e-05	1.842e-04	7.465e-05
1.00e+05	6.841e-03	1.288e-02	2.586e-04	1.263e-04	2.697e-02	1.822e-03	1.263e-04	1.685e-04	1.544e-04
3.00e+05	7.368e-03	1.387e-02	2.785e-04	1.372e-04	2.918e-02	1.826e-03	1.372e-04	1.572e-04	1.633e-04
1.00e+06	1.266e-03	2.353e-03	7.652e-05	2.928e-05	5.223e-03	1.686e-04	2.928e-05	3.376e-05	3.459e-05

Figure 27. Group-I dose to liver (from Table 6).

Figure 29. Group-III dose to liver (from Table 8).

Figure 28. Group-II dose to liver (from Table 7).

Figure 30. Summation of all group doses to liver (from Tables 6-8).

precludes a long transport scenario that masks the effects of design factors.

We would also like to choose a performance measure that has been used traditionally in the regulation of the nuclear fuel cycle, i.e., individual dose. Acceptable population doses have been achieved by regulating the maximum dose to the public and to individuals in the nuclear work force.

Our measure of repository design performance is the standard man who drinks 2.2 litres of water per day from a well located just downstream from the shaft of our repository. Depend-

ing on the placement of the well and shaft relative to the repository, the standard man will receive dose from the flow of radioactivity from the shaft or through the barrier media, or both. The shaft radioactivity will be diluted during the flow from the shaft to the well. The amount of dilution depends on the distance of travel, as well as on the relative amounts of water flow in the shaft and aquifer.

We consider a "sampling well" whose diameter is smaller than that of the shaft and assume that the withdrawal rate is slow enough to ensure that the magnitude and direction of aquifer water

velocity are unaffected by well operation. Piezometric heads in the aquifer also remain unchanged, and we assume that the well samples the aquifer water uniformly with respect to depths. Further, to study the "worst-case" dose to man, we assume that both the well and shaft are placed on the centerline of the repository.

Appendix F of Ref. 32 shows that, on the centerline, the concentration in the aquifer, from a continuous source f , can be approximated by

$$C \approx \frac{f}{4\eta_a b q_a [\pi(\alpha - 2\mu)x]^{1/2}} \quad (31)$$

where

- η_a is the effective porosity
- b is the thickness of the aquifer
- q_a is the actual water velocity
- $\alpha - 2\mu$ is the lateral dispersion constant

and x is the distance from the source, f . Clearly, we must restrict x so that the denominator is greater than or equal to 1.

Let x be the distance from the shaft to the well. Then, if

$$F_S = 4\eta_a q_a b [\pi(\alpha - 2\mu)x]^{1/2} < 1 \quad (32)$$

we consider an approximation based on Fig. 31. Here we assume that there are no effects from

lateral dispersion and that a well will mix the water in the vertical direction.

We assume a circular shaft for which we have input the cross-sectional area A_s so that its diameter is

$$d_s = 2(A_s/\pi)^{1/2}$$

Then, from the figure, we compute

$$F_{NS} = Q_S \min \left[1, \frac{(d_s + x) v_s}{b v_a} \right] \quad (33)$$

where $v = q\eta$ is the D'Arcy velocity in the shaft or aquifer and Q_S is the D'Arcy flux in the shaft.

For flow through the media to the aquifer, we compute the dilution factor by

$$F_M = a b q_a \eta_a \quad (34)$$

where

- a is the width of the repository
- b is the thickness of the aquifer
- $q_a \eta_a$ is the water velocity in the aquifer.

In each case, F_S , F_{NS} , and F_m are in cubic metres per year.

In our transport and release models, we have computed the release function at observation

Figure 31. Schematic for close-to-shaft well-concentration calculation, with $b_w = (x + d_s) \tan \theta$.

point k to give $J_r^k(\sigma_r, \tau_r; T)$ in units of initial inventory per year, e.g., canisters/year or MWe-year/year, at the discretized time points T_i for nuclide-retardation group r. We then note that

$$C_r^k \equiv \frac{J_r^k}{F_m}$$

for $m = S, NS$, or M gives us concentration of a nuclide group r, e.g., canisters per cubic metre.

We assume that our standard man consumes all his water from the well at the rate of 2.2 litres per day. Then, for example,

$$C_r^k \frac{\text{canisters}}{\text{m}^3} \times \frac{2.2 \text{ litres}}{\text{day}} \times \frac{365 \text{ days}}{\text{year}} \times \frac{0.001 \text{ m}^3}{\text{litre}}$$

gives

$$\bar{C}_r^k = 0.803 J_r^k / F_m \frac{\text{canisters}}{\text{year}} \quad (35)$$

of nuclide group r at observation point k with well scenario $m = S, NS$, or M .

If we use the potential hazard tables produced by the ORIGEN code in the same manner as we did in the "Curies/Year Potential Hazard Model" section of 3.2.4, then

$$\begin{aligned} \bar{D}_r^k(T_i) &= H_r(T_i) \bar{C}_r^k(T_i) \\ &= 0.803 \frac{J_r^k(T_i)}{F} H_r(T_i) \frac{C_i}{\text{year}} \quad (36) \end{aligned}$$

is a potential hazard measure.

The equivalent whole-body dose, 50-year commitment discussed in Ref. 36, provides a rational method for combining doses to different organs from different radionuclides into a single dose measure. This dose measure can then be used to analyze repository-engineered features.

The basic quantity used for such a combination is the "annual limit of intake" (ALI) for each radionuclide. If an ALI for nuclide n is ingested, then the 50-year dose commitment is 5 rem. Table 9 is a listing of ALI for numerous nuclides in becquerels (Bq); we note that one curie = 3.7×10^{10} Bq.

Under our assumptions, we may write the effective annual whole-body dose from nuclide n by

$$D_n^k(T_i) = 3.7 \times 10^{10} \bar{D}_r^k(T_i) / \text{ALI}_n \frac{\text{Sv}}{\text{year}} \quad (37)$$

where $D_r^k(T_i)$ is from Eq. (36), with r chosen so that nuclide n is a member of retardation group r and ALI_n (in Bq), the annual limit of intake, is from Table 9.

Then, the final dose is obtained by summation so that

$$D^k(T_i) = \sum_n D_n^k(T_i) \frac{\text{Sv}}{\text{year}} \quad (38)$$

3.2.5 Selected Analyses Using MISER

Introduction. In May 1980, the Nuclear Regulatory Commission (NRC) published a proposed rule³⁷ that, among other considerations, developed a set of technical standards pertaining to the near-field performance of a nuclear waste

TABLE 9. Annual limits of intake for nuclear industry workers.

Nuclide	ALI (Bq)	Nuclide	ALI (Bq)
²⁴³ Am	8×10^4	^{93m} Nb	5×10^8
²⁴¹ Am	8×10^4	⁹³ Zr	2×10^8
²⁴⁰ Pu	4×10^5	⁹⁰ Sr	2×10^6
²³⁹ Pu	4×10^5	⁷⁹ Se	7×10^7
²³⁸ Pu	5×10^5	⁵⁹ Ni	1×10^9
²³⁷ Np	5×10^3	¹²⁶ Sn	2×10^7
²³⁸ U	7×10^5	²³⁹ Np	5×10^7
²³⁵ U	6×10^5	²²⁵ Ac	2×10^6
²³⁴ U	6×10^5	²²⁵ Ra	5×10^5
²³³ U	6×10^5	²²³ Ra	3×10^5
²³¹ Pa	1×10^3	¹²⁶ Sb	2×10^7
²³² Th	7×10^4	⁹⁰ Y	2×10^7
²³⁰ Th	4×10^5	²⁴⁴ Cm	2×10^5
²²⁹ Th	5×10^4	²⁴² Cm	3×10^6
²²⁷ Ac	1×10^5	²⁴¹ Pu	2×10^7
²²⁸ Ra	2×10^5	¹⁴⁷ Pm	2×10^8
²²⁶ Ra	1×10^5	¹⁴⁴ Ce	9×10^6
²¹⁰ Po	1×10^5	¹³⁴ Cs	3×10^6
²¹⁰ Pb	5×10^4	¹⁰⁶ Ru	9×10^6
¹⁵⁴ Eu	3×10^7	⁹⁵ Nb	8×10^7
¹⁵¹ Sm	6×10^8	⁹⁵ Zr	5×10^7
¹³⁷ Cs	4×10^6	⁹¹ Y	2×10^7
¹³⁵ Cs	3×10^7	⁸⁹ Sr	2×10^7
¹²⁹ I	5×10^5	^{242m} Am	7×10^4
¹⁰⁷ Pd	1×10^9	²⁴² Pu	5×10^5
⁹⁹ Tc	2×10^8	²³⁶ U	6×10^5

repository. Two excerpts from this proposed rule specifically concern near-field effects:

1. "The department shall design the underground facility to provide reasonable assurance of containment of all radionuclides for the first one thousand years after decommissioning . . . assuming . . . that some of the waste dissolves soon after decommissioning.
2. "Starting one thousand years after decommissioning . . . the radionuclides . . . will be released from the underground facility at an annual rate that is as low as reasonably achievable."

To attain these goals, the proposed rules provide for three sequential but independent barriers: the engineered waste package, the engineered underground facility, and the natural rock features associated with near- and far-field flow. Each barrier is considered a major factor of high-level nuclear waste containment,^{37,38} and standards have been proposed for each.

In this section, we focus on the underground facility barrier as a prelude to detailed analyses of waste-form characteristics. This barrier is of primary importance in minimizing individual dose in the near-field environment if the waste package fails. Under such conditions, it will have the largest effect on keeping local individual doses to a minimum.

Traditionally, the regulation of dose for the operating period of nuclear facilities has been accomplished mainly by specifying a maximum dose that an individual can receive.³⁹ The operating period for a nuclear-waste disposal facility is the period the waste must be contained, or at least not released in quantities high enough to produce a dose above some policy-dictated level. The waste-containment period of major interest here is that period after the repository has been sealed, during which time natural forces are operating on it and remedial actions are the most difficult to accomplish.

Because some of the most uncertain parameters used in the prediction of far-future doses are those associated with the far-future population (their location, number, and eating habits), the dose for the limiting individual is, technically, the most predictable far-future effect. By limiting individual dose, we can protect the far-future population in the same manner as today's population, which is routinely subjected to small amounts of

radionuclides released to the biosphere under controlled conditions. These routine releases result in individual doses far below natural background radiation.⁴⁰

Population doses can be well approximated by a point-source repository model for regional flow basins if one assumes a specific set of population characteristics (usually similar to today's population parameters) and a sufficiently long geosphere transport to the biosphere. On the other hand, the limiting individual's dose cannot, in many cases, be well approximated with a point-source model (see below) because the dose is strongly dependent on details of repository design that are not included in a point-source model.

It would appear that we could obtain a realistic prediction of a repository's far-future performance if we (1) use a population dose standard and establish by fiat what the far-future population will be like, or (2) pick a limiting-individual dose standard and improve our modeling of the near-field region around the repository. We contend that the second alternative will lead to more satisfying technical answers when predicting far-future doses, assuming that we use a reasonable definition of limiting individual. Using a limiting individual as a licensing tool to compare different sites, mine designs, and waste-package configurations appears to be more technically defensible than using the more uncertain population dose.

Predicting doses to individuals who live near a nuclear waste repository requires careful modeling of the repository excavation, its backfill, and its effect on surrounding hydrologic conditions. The availability of waste to individuals always starts with a radionuclide release from one of many spatially separated canisters. The transport of waste from a canister to an individual can take many pathways. If an observer were able to follow the migration of the many waste pulses to a biosphere release point, he would see a system of pulses moving through various pathways, at different times, before it finally impacts the biosphere release point.

Point-source repository (PSR) models, which often have only one sequence of pathways to a release point, ignore the network detail, spatial separation, and branching that occur within a real repository.^{26,27,41} Since the spatial extent of a repository leads to initial dilution of the radioactivity within the repository, which is important for calculating individual doses, we needed a model to study network effects on dilution. MISER was developed for that purpose.

In our initial applications of MISER, we consider an individual who drinks contaminated water from a well near the repository shaft. To do this, we must carefully calculate the trajectory of the release plume from each canister. Our model helps us sort the relative effects that might be obtained by designing and modifying hydrologic networks and their physical attributes so as to reduce individual doses.

In constructing MISER, as it is applied to this work, we linked several numerical tools at Lawrence Livermore National Laboratory to calculate deterministically the approximate magnitude of conservation inherent in the PSR assumption. Our intent was to build a model that allows individual dose to be sensitive to an extended source repository (ESR). By varying the network of an ESR model from a point-source model to a detailed network model with multiple source terms, multiple boundary conditions, and multiple pathways, we can obtain a relative measure of the conservatism of the PSR model. To establish the unmasked effects of different mine designs, we have chosen appropriate but not necessarily realistic parameters for the waste package and high hydraulic gradients.

Choice of System Parameters. We had two motives in mind when we chose the system parameters for this study: to define a system that could calculate dose to the "limiting individual," and to choose a system that could test underground facility (mine) design. The fact that we chose limiting-individual dose as an appropriate performance measure is actually consistent with the second motive, to test the design. Near-field dose is most sensitive to mine design. Far-field population doses are less sensitive to mine-design details and are essentially, if integrated over time, only sensitive to the biosphere emergence time of the radionuclides. Thus, if we want to measure the effectiveness of mine designs as barriers, we must concentrate primarily on near-field effects, which excludes population dose as an effective performance measure.

That waste-package parameters can mask the effects of mine design as a migration barrier is obvious. For example, if the waste package never fails, i.e., the breach time is infinite or the leach rate is zero, the limiting individual dose is always zero. Consequently, mine-design changes will not lead to changes in the calculated dose for this type of scenario. Another set of parameters that can lead to zero results is hydraulic gradients. If the

gradients in the system are zero and remain zero, there will be no radionuclide migration from the system.

To test the sensitivity of individual dose to mine design, we have selected a model that is sensitive to mine-design parameters. In effect, we have chosen waste-package parameters and hydraulic gradients that lead to mine-design sensitivity in the predictions, making the mine-design barrier the only effective barrier in the model. To do this, we have chosen 1000 years as the point at which to initiate the transport so as not to influence the results unduly with very short-lived fission products. At 1000 years the canisters are considered breached, and the waste is leached out at an average leach rate of 1% per year. Under these circumstances, the major effect reducing the dose is the mine layout. Finally, we have chosen hydraulic gradients that are large enough to cause fairly high flows. The vertical gradient is 0.1 m of water head per metre. The resulting high flow rates tend to enhance the effect of the distributed source on individual dose.

Figure 32 is a side view of the general qualitative features of our model. Before the underground facility is built, the regional pressure gradient has a hydraulic gradient from right to left and from bottom to top. Arrows approximate the directions of flow. Basically, water flows into the lower aquifer from the right. Some of this water migrates upward into the lower aquitard and traverses it, finally merging with the water in the upper aquitard.

When the mine is excavated, the hydraulic pressure field around the excavation evolves from the one associated with the undisturbed media to a new one consistent with the excavation features, causing local perturbations to the regional flow field. Depending on the permeability and area of the tunnel, for example, more or less of the water will move along the tunnel and up the shaft. Thus, although the boundary conditions for the system remain essentially the same, local flow conditions in the repository can vary greatly because of mine design. Maximum variations from the virginal flow conditions will occur when the repository storage area, tunnel, and shaft are highly permeable compared to the aquitard. This could be the case for deteriorated backfill or large fracture zones caused by the excavation of drifts. To demonstrate these effects, we chose a scenario in which the backfill is deteriorated at 1000 years, the canisters have been breached, and the hydrology is steady state.

Figure 32. Side view of the near-field features of our high-level waste-repository excavation model.

As indicated in Table 10, the aquifers are considered to be sandstone with a typical sandstone permeability and effective porosity.⁴² We have assumed that the deteriorated backfill will have the same permeability as the aquifers, but that its effective porosity will be an order of magnitude smaller. The effect is high volumetric flow rates and flow speeds for water in the tunnel and shaft.

One can see from Fig. 32 that our limiting individual has, unfortunately, placed his well just downstream of the shaft. The shaft seal is assumed to have failed completely by 1000 years. Thus, water reaching the top of the shaft becomes a dispersing plume in the upper aquifer. This plume intersects the lower portion of the limiting individual's low-volume well. The well mixes water drawn from the aquifer and shaft thoroughly, diluting the contaminated water from the shaft plume with purer water from the aquifer. If the limiting individual drinks 2.2 litres of water per day from his well, he receives toxic radionuclides from three transport groups: (1) anion fission products, with no retardation, (2) cation fission products, which move 10 times slower than the anions, and (3) actinides, which move 100 times slower than the cations.

In all scenarios, we have distributed one canister of typical defense waste.⁴³ If a point-source

model is used, all the waste is in one canister at a designated location. If a distributed source of 25 canisters is used, for example, each model canister has 1/25th of the waste that is in one canister of defense waste. Thus, each scenario is normalized to the same number of initial curies of waste. The radioactivity of the defense waste is defined as a function of time and contains 53 radionuclides. The dose to the limiting individual is determined by considering the equivalent whole-body dose for each nuclide ingested in the form of a 50-year dose commitment.³⁶ The total equivalent whole-body dose is then obtained by taking a weighted sum over all the ingested radionuclides and all the canisters within the system.

Description of Repository. Figure 33 is a perspective drawing showing the qualitative features of a numerical model that uses 25 flow cells to model the repository storage area. The shaft and tunnel are on the repository's line of symmetry. The horizontal boundary conditions for the base-case scenario are head gradients that cause a regional flow from right to left, parallel to the tunnel. The shaft is downstream of the repository storage area. This produces a situation where part of the waste from every canister reaches the top of the shaft.

Table 10. Repository and media parameters at 1000 years.

Upper and lower aquifers	
● Depth of upper aquifer = 100 m	● Length = 3.14 km
● Depth to lower aquifer = 900 m	● Thickness = 200 m
● Rock type = sandstone	● Effective porosity = 0.1
● Hydraulic conductivity = 10^{-4} cm/s	
Rock barrier	
● Depth to aquitard = 300 m	● Length = 3.14 km
● Vertical pressure gradient = 0.1 m/m	● Thickness = 600 m
● Rock type = shale	● Effective porosity = 0.05
● Hydraulic conductivity = 10^{-8} cm/s	
Repository	
● Depth = 600 m	● Shaft
● Storage area	Length = 300 m
Length = 2.5 km	Area = 64 m^2
Width = 2.0 km	● Thickness = 6 m
● Tunnel	● Backfill
Length = 440 m	Hydraulic conductivity = 10^{-4} cm/s
Area = 150 m^2	Effective porosity = 0.01
Comments	
● Steady-state hydrology	● Regional hydraulic pressure gradient = 0.005 m/m
● Deteriorated backfill and shaft seals

Figure 33. Perspective drawing, showing qualitative features of one numerical model that uses 25 flow cells to model the repository storage area.

To display the typical flow cell (shown by the cutaway in Fig. 33 and located near its geometric center), we have depicted the upper aquitard as being transparent. The typical flow cell is shown to run from the top of the lower aquifer through the repository storage area to the bottom of the upper aquifer. The hatched strata are the upper and lower aquifers, and shaded areas 1, 2, and 3 are, respectively, the bottom of the lower aquitard, the top of the storage area, and the top of the upper aquitard.

For the typical cell, uncontaminated water flows from the right of Fig. 33 through the lower aquifer to the cell. It is pushed by the vertical head gradient through area 1 and up into the lower aquitard. Then it moves through the lower aquitard into the repository storage area where, if the canister has been breached, the leaching waste mixes with the passing water. The contaminated water is then split into three flow tubes that move along orthogonal flow directions. The amount of water diverted into each flow tube depends on the

calculated pressure gradients in the repository. Part of the contaminated water moves parallel to the tunnel into the adjacent downstream flow cell. Another part moves toward the tunnel. Most of the water, however, moves into the upper aquitard barrier where, because of the barrier's permeability, the flow is very slow.

Schematic to Describe MISER Runs. To help differentiate between our underground facility models, we have developed a schematic shorthand. Schematics of two cases are shown in Fig. 34. The left schematic, consisting of both a top and a side view, shows a one-cell hydraulic flow model of a repository storage area. The schematic at right shows a 25-cell flow model of the same storage area. The flow cells for each are depicted by rectangular segments, as seen in the top views.

The "X" in each schematic shows where the waste is emplaced originally, and "O" shows the location of the limiting individual ("observer"). The dot, seen in the side view, is where the tunnel meets the storage area.

Note that the top and side views are not drawn to scale. Thus the schematic features, unlike those in a mechanical drafting diagram, do not line up. The main purpose of the top view is not alignment, but to show where the waste is

located in the storage area. The side view is meant to show the flow paths that waste must travel to reach the observer.

One-Cell Model. The one-cell case depicted in Fig. 34 shows a single series pathway from the waste to the observer. For this case, all waste first migrates from the center of the storage area where it was originally emplaced to the edge of the storage area where it enters the tunnel. Because of the large cross-sectional area in the storage area, compared to that of the tunnel, there is a convergence of streamlines at the tunnel entrance. Thus, there is a large increase in flow rate as the waste enters the tunnel. The time required for the waste to transit the tunnel and shaft to the observer is very short compared to the transition time in the storage area. The schematic shows that the waste goes through the storage area to the tunnel, through the tunnel to the shaft, and then up the shaft to the observation point. This is a series pathway. When it gets to "O," our limiting individual ingests 2.2 litres of contaminated water per day.

25-Cell Model. Our more-realistic hydraulic model of the repository storage area (shown at right in Fig. 34) has seven parallel paths

Figure 34. Two examples of the schematic devised to help describe computer scenarios.

by which waste traverses to the observer. Where two or more downstream flow tubes leave a node in the schematic, the radioactive waste splits into two or more pulses.

For the displayed 25-cell case, we have placed the radioactive waste on the centerline cell furthest upstream from the observer. Because of branching, this cell produces more pulses than any other centerline cell. Off-centerline cells could produce even more pulses.

By using centerline symmetry in the model, the hydrology of the 25-cell storage area is actually modeled with 88 flow cells. Without symmetry, the system would require 141 flow cells. The only flow cells shown in the schematic are those on the centerline that also have waste flowing through them. The upstream flow cells, which contain uncontaminated water, and the off-centerline flow cells are not shown.

A detailed discussion of the flow in the 25-cell case should prove enlightening. The leftmost vertical line, depicting the shaft in the 25-cell repository, is 300 m long and 64 m² in area. The next vertical line to the right is a schematic of the flow path taken by the waste that leaks out of the tunnel and then flows through the aquitard barrier to the upper aquifer and from there to the observer. The vertical lines to the right of the dot—25 in all—are flow pathways from the storage area to the upper aquifer. The five lines shown represent the five centerline cells. Each of the 25 cells is 0.2 km² in cross-sectional area. Because of the large area of these vertical cells, compared to the horizontal flow cells, most of the waste flows upward through the aquitard barrier even though its permeability is much less than the deteriorated backfill permeability of the storage area and tunnel.

If we follow the waste from its source to the observer, as indicated in the side view of the 25-cell repository, we see it starts its journey by splitting into two parts. At the initial node, most of the waste moves upward through the rightmost flow path of the aquitard to the upper aquifer. Once in the upper aquifer the waste does not split again, but proceeds in its entirety to disperse, advect, and mix with other waste as it moves along the upper aquifer until it reaches "O." The remaining portion of waste at "X" starts its journey by moving left along the centerline of the storage area, splitting (leaking) within each of the four flow cells it must traverse as it moves toward the connecting tunnel. Each time it crosses a cell, it leaks radioactivity to the surrounding rock. The leaked waste slowly moves vertically through the barrier

rock to the upper aquifer. The leaked waste that finally reaches the tunnel moves into the tunnel entrance and through it to the shaft. Even as it moves along the tunnel, the waste can leak into the rock barrier. The waste left at the end of the tunnel moves up the shaft to the observer. Generally, the shaft flow produces the peak dose to the limiting individual.

Conservatism of the Point-Source Model. In this section we analyze different point-source repository models to establish how their assumptions affect predictions of individual dose. Figure 35 shows 10 separate point-source models, each of which simulates the same physical system. The 10 cases differ in that they use different modeling strategies. By comparing schematics, one can see that Cases 1, 2, 3, 5, and 8 are one-cell storage-area hydrology models. The other five cases are 25-cell storage-area hydrology models.

The two peak annual dose curves shown in Fig. 35 are represented by a thick line and a thin line. The thick line is the peak dose calculated for 10⁶ years, and the thin line is the peak dose that occurs before 10⁴ years. Where they merge, the peak dose for 10⁶ years occurs before 10⁴ years. This occurs for Cases 5–10. For Cases 1–4, the retarded fission products and actinides move through the system quickly enough to be counted before they decay away or before 10⁶ years have passed. In Cases 1–3, all the waste is at the boundary of the tunnel and the storage area. For Case 4, it is barely inside the storage area. Thus, these cases do not have enough flow within the storage area to detain realistically the radionuclides that can be retarded; therefore, these radionuclides cause the peak dose after 10⁴ years. In Cases 5–10, the unretarded anion fission products cause the peak dose.

Canister Breaching. All cases except Case 2 have initial release of the waste pulse at 1000 years. Case 2, however, has 25 canisters of waste that are initially released at times randomly chosen between 1000 and 2000 years. Each of the 25 canisters is assumed to have 1/25 of the waste of a real canister, which normalizes Case 2 to the same amount of waste as the other 9 cases. Although, for Case 2, there is no effect on the peak dose for 10⁶ years, there is a factor of several reduction in the 10⁴-year dose. Real repositories will have canisters breaching at different times. Depending on breach-time parameter values, they can have a major effect on near-field individual doses.

Waste-Pulse Branching. In Fig. 35, we have arranged the results in descending order of

Figure 35. Results, arranged in descending order of peak 10^6 -year dose, showing discrepancies between various point-source models of a physical waste repository.

peak 10^6 -year dose. Cases 3 and 5 are single-pathway models. As shown in their schematics, they have only one path from "X" to "O." Case 1, on the other hand, has two pathways along which waste migrates to the observer. Thus, for every pulse of waste released from "X" in Case 1, there will be two pulses reaching "O." For Cases 3 and 5, there is only one pulse reaching "O" for each pulse released from "X." For Case 10, there are seven pulses reaching "O" for each pulse released.

Branching occurs at each node in the model. Water flows both up through the vertical flow cells and through the horizontal flow cells, flowing without mixing in a flow cell until it reaches a node. The steady-state speeds and directions of flow in each flow cell are determined numerically for the entire system by solving a set of hydrology equations derived from D'Arcy's Law and the

continuity equation. Boundary conditions for the system are determined by using regional pressure gradients. Water is conserved at each node. The assumption used for waste branching is that the waste pulse is thoroughly mixed with the water. The waste then splits with the same fraction as the water leaving a node. The water fractions are obtained when the hydrology equations are solved. Thus, for each of the nodes on the centerline in the schematic of Case 10, the pulse splits into two parts. Most of the waste flows into the vertical path following the major portion of the water; the remaining waste and water flow down the centerline of the storage area toward the tunnel. The result is that seven pulses, each moving along a different path, arrive at "O" for Case 10.

Comparison With Best Estimate. Additional results (thick and thin horizontal lines) plotted on the figure are from our most complete

modeling effort (best estimate). We compare the results from point-source models with these results. For our best-estimate model, we see that the 10^6 -year dose is about a factor of 4 greater than the 10^4 -year dose. This is not predicted by point-source models.

The more-conservative point-source cases predict a value that is about a factor of 400 greater than our best estimate. If we use a point-source strategy that minimizes the peak-dose prediction, the results for the 10^6 -year dose are about a factor of 100 smaller than our best estimate.

The peak doses from the point-source model for Cases 5-10 are due to unretarded ^{99}Tc . For the best-estimate case, the peak dose is due to retarded actinides that reach the observer at about 75,000 years, not from ^{99}Tc . Thus, even the radionuclides that cause the peak dose can be predicted incorrectly by the point-source models.

The difference in the predicted radionuclides that cause the peak dose is due mainly to the fact that our best-estimate model is a 1000-canister model, each canister of which has a different initial release time. They breach between 1000 and 2000 years. The point-source models, however, have only one canister that breaches at 1000 years. This leads to a single pulse from the nearest storage cell of relatively nondiluted ^{99}Tc , which causes the peak dose. In the best-estimate model, the ^{99}Tc is diluted by the many canisters releasing at different times and locations. Thus, the peak dose for the best estimate is due to retarded cations, not to ^{99}Tc , which migrates as an anion.

In summary, point-source models can lead to very misleading predictions of individual dose. An apparently conservative strategy might not lead to a conservative result. For example, Cases 7 and 8, which appear heuristically conservative, do not predict a dose as high as the best estimate. Alternatively, ultraconservative models (i.e., Case 1) can produce results that are actually two or three orders of magnitude too high. Besides being substantially too high or too low, point-source models can predict the wrong radionuclides as the main cause of peak dose.

We conclude that the choices in a point-source model are quite arbitrary and can lead to poor or misleading predictions of near-field individual doses. Also, predictions from point-source models of curies released to the near field must be considered questionable. To adequately predict repository releases (to be compatible with 10CFR60 licensing requirements, for example) will require more sophisticated models of the repository than point-source models.

Point-Source and Extended-Source Comparisons. Figure 36 shows the results generated to test the effect of smearing-out the same amount of waste over different fractional areas of a repository. In each case, the amount of smeared-out waste totals a single canister. The observed reduction of calculated dose (the more reduction, the greater the spatial separation of the canisters) will tend to reduce the significance of lower leach-rate waste forms. In fact, where the permeabilities are quite low, the individual's dose could be very insensitive to the choice of waste form.

The relative peak annual dose to a limiting individual is shown on the ordinate of the figure. On the abscissa, we have labeled the cases by the areal fraction of the repository that has been used to store the waste. The top view of the schematic shows the location and areal coverage of the waste in the repository.

The 0/25 case is a point-source repository. Other cases, shown to the right of the 0/25 case, have more detail and spatial coverage of the repository. To its right is a model that covers 1/25 of the repository area. The next four cases cover 2/25, 4/25, 9/25, and 25/25 of the area of the repository. Each of these cases has one canister per cell. The rightmost case is a model of a 1000-canister repository.

We have plotted two curves in Fig. 36. The first curve, depicted by a thick line, shows the peak dose to the limiting individual, which occurs in the first million years. The second curve, the thin line, is the maximum dose reached in the first ten thousand years. For these cases, we notice that the peak dose always occurs after ten thousand years. This is especially true of the point-source repository case, which has an order of magnitude difference between the thick and the thin line. The difference is caused by retarded actinides contributing to the dose before one million years because they do not have to traverse the slow flow portions of the repository.

A large difference between the thick and thin lines results for the 1000-canister case, i.e., the 25/25* case. Here initial leach time occurs between 1000 and 2000 years. In all other cases, all canisters leach simultaneously at 1000 years. This produces a large effect on the anion pulses because they have no retardation. Its effect on the retarded fission products and actinides is insignificant because their release into the upper aquifer covers tens of thousands of years.

Some Engineering Features of Repositories. Some simple engineering of the hydraulic parameters of a repository can reduce the dose to

*40 canisters/cell

Figure 36. Results generated to test the effect of smearing-out the same amount of waste over different fractional areas of a repository, showing dose sensitivity to spatial distribution of the source term.

future individuals living close to a repository. In this section, we examine some simple engineered considerations that show potential for reducing near-field doses. To save computer costs, a 9-cell repository model was used instead of a 25-cell one. Comparison shows that results from the 9-cell model are, consistently, about 15% below the 25-cell model. Since the range of doses for the engineered-features cases we have examined is two orders of magnitude (see Fig. 37), the 9-cell model approximation appears adequate for a relative comparison of repositories with different design features.

The curve shown in Fig. 37 is peak individual dose in the first 10⁶ years. The cases we have analyzed are referred to as base case, upstream-shaft, off-tunnel storage, and hydraulic bypass, each of which is discussed below.

Base Case. For our base case, we chose a downstream shaft with a repository storage area that is uniformly filled over its entire area. This is similar to Case 25/25 in Fig. 36. The limiting individual's peak annual dose is 3.0×10^{-5} relative units. Given enough time, all base-case radioactivity from each of the nine cells of the repository

Figure 37. Peak individual dose in the first 10^6 years, showing dose sensitivity to various design features.

migrates to the observer. This is shown for center-line cells by the side view of the schematic.

Upstream-Shaft. If we specify flow from the left by changing our model boundary conditions, the observer finds himself upstream with respect to the horizontal regional gradients. The solution of the hydrology equations for the upstream-shaft system, however, shows water flows in the tunnel to the left, actually against the regional horizontal gradient. This is driven by the stronger vertical gradient. Because the vertical gradient is large and the permeability of the tunnel and shaft is much greater than the aquitard barrier, some radioactivity in the repository migrates from the repository, through the tunnel, and up the shaft to the observer. This is the path of least resistance for a portion of the repository waste. The migration through the tunnel is shown in the Case 2 schematic for waste nearest the tunnel. The side-view schematic for Case 2 shows that two-thirds of the radioactivity on the centerline moves up through the aquitard barrier and down the aquifer to the right. This waste is never seen by our observer at "O." Arrows show the direction of waste flow when the direction is other than toward the observation point.

The upstream-shaft dose to the limiting individual is reduced by about a factor of 2 over that for the base case. The conclusion, however, is sensitive to the hydrology parameters chosen for this study. More horizontal gradient and less vertical gradient could result in zero observed dose at the observation point, which means that the limiting dose would be at a different point in the system.

Off-Tunnel Storage. Returning to the downstream-shaft case (our base case), we design a system so that there is a waste-exclusion area along the centerline of the repository. We model this area as no waste in the three centerline cells, shown in the Case 3 schematic. Unlike Case 2, waste from all the cells ultimately migrates to the downstream observation point. The fact that it is delayed by the exclusion zone, however, reduces the limiting individual's dose about a factor of 10 below the base-case value.

Hydraulic Bypass. The final case is for a design with an hydraulic bypass. The engineered bypass encircles the repository by 50 m. The dashed lines in the schematic indicate portions of the bypass where water flows but waste does not, while the solid lines show where waste flows from its storage location to the observation point.

The 10-m-high bypass runs below the 2-km width of the repository, and its permeability is the

same as that of the aquifer. Thus, the bypass can be conceptualized as another horizon to the repository, with 10-m-high ceilings and a 2-km width. It is filled with very permeable material. This 20,000-m² hydraulic conduit runs under the tunnel and shaft and connects into vertical conduits. These vertical conduits have the same permeability as the aquifer and 20,000-m² area. They are located at both ends of the repository.

Our limiting individual's dose for this repository design is reduced by two orders of magnitude from the base-case value. One might ask what dose the limiting individual would receive if he moved his well 50 m to the left, where it would then be downstream of the bypass discharge into the upper aquifer. Since the dilution in the bypass is much greater than that in the tunnel and shaft, the limiting individual's dose should still be below our base-case results. In other words, the hydraulic bypass can effectively reduce the limiting individual's dose whether he is at the well or at the bypass discharge.

We conclude that engineered features could be integrated into a repository design that would lead to substantial reductions in forecasts of near-field doses to individuals. We suspect that a reduction of over four orders of magnitude can be obtained by combining the features examined here for a specific site. Searching for optimum results and expanding the features could improve the near-field individual dose even further. Such site-specific numerical studies would be relatively inexpensive and certainly help attain an ALARA-designed repository.

3.3 Parametric Analysis

3.3.1 Introduction

Parametric analyses were initiated to study the impact of generic waste-form properties under a range of geologic conditions on overall waste-isolation performance. The results of this study can address many hypothetical questions associated with the Bedded Salt Base Case Analysis¹ completed last year and will provide valuable guidance in selecting the alternate media-siting scenarios to be studied in detail during the final phase of this project. These analyses were designed with three thoughts in mind: (1) to simplify modeling the overall system, (2) to span the range of credible performance measures, and (3) to provide a framework for evaluating scenario-specific analyses. The XCOMP computer code described

in Section 3.3.2 was developed and used to perform the calculations.

Numerous failure scenarios can be proposed to characterize the release of nuclear waste from a deep geologic repository into the biosphere; however, waste-form dissolution and radionuclide geotransport are the two processes common to most release schemes. The algorithms describing these processes (see Section 3.2.3) require only a few input parameters to characterize their outcome despite the host of physical, geological, and chemical variables that may give rise to a particular performance measure. For a point-source[†], step-release pulse that travels along a single flow path to the biosphere, these two processes can be characterized by four fundamental parameters:

- Canister breach time.
- Waste dissolution time.
- Transit time from the repository to the biosphere.
- Waste-pulse width due to geologic dispersivity.

From these four systems characteristics, XCOMP calculates a peak-normalized flux and time-of-peak normalized flux. The normalized flux differs from the activity flux in that the normalized flux does not take nuclide decay into account and is calculated as a fraction of the emplaced inventory. For a given inventory (e.g., SRP Defense Waste), performance measures can be calculated that represent the nuclide activities released to the biosphere. Throughout these analyses, overall performance measures have been keyed to activity released. Peak curie flux (curies per canister per year) is reported to indicate individual cancer risk and time-of-peak arrival to describe the post-closure immediacy of this risk. The activity levels reported were based on ORIGEN inventory data for a reference canister of SRP waste.³⁵ A dose-to-man performance measure is also possible within the methodology of these analyses. We should emphasize, however, that these dose results require a number of additional assumptions (e.g., water-use systems, bio-uptake factors, population trends, etc.).

The parameter ranges selected for these analyses are not associated with any site-specific or generic class of siting scenarios. Each parameter range is intended to bracket only the physically credible values that the system characteristics might assume, and no attempt is made to correlate

these inputs with the numerous combinations of geologic, chemical, and physical variables that might give rise to the value of the parameter. By spanning the range of possible input parameters, parametric analyses illustrate the values and variations of the resulting (both expected and abnormal) scenario performance measures.

These analyses examined the impact of the following systems characteristics on overall performance measures. Note that the range associated with each systems characteristic encompasses (where applicable) the NRC-proposed performance standards.⁴⁴

Breach time (T_b): Includes repository-operation-period refill and repressurization times and canister corrosion time.
Minimum: 1.0 year
Maximum: 1.0×10^3 years

Waste dissolution time (T_d): The period of time, starting at T_b , over which a homogeneous waste matrix dissolves at a uniform constant rate.
Minimum: 1.0 year
Maximum: 1.0×10^6 years

Transit time (T_t): Mean overall transit time of waste pulse from release point to biosphere; retardation effects are included by selecting a T_t appropriate to the nuclide group.
Minimum: 1.0 year
Maximum: 1.0×10^6 years

Pulse width (S): Spreading effect on initial release pulse because of hydrologic dispersivity.
Minimum: 10% of transit time
Maximum: 100% of transit time

Inventory (I): Reference canister of SRP radioactive waste,³⁵ nuclides grouped by retardation properties. See Figs. 22-24.

Group I—Tc, I (no retardation)
Minimum activity (at 10^6 years): 9.8×10^{-2} Ci/canister. Maximum activity (at 1 year): 2.5 Ci/canister

[†]Note that an extended discussion of the limitations of point-source assumptions is presented in Section 3.2.5.

Group II—other fission products (moderate retardation) Minimum activity (at 10^6 years): 9.8×10^{-1} Ci/canister. Maximum activity (at 1 year): 1.8×10^4 Ci/canister

Group III—actinides and daughters (strong retardation) Minimum activity (at 10^6 years): 2.1×10^{-1} Ci/canister. Maximum activity (at 1 year): 5.2×10^2 Ci/canister

The ranges and combinations of system characteristics that strongly impact the overall results can be identified by varying each parameter in turn over its range of values, with all others held constant during analysis.

Most of the emphasis in previous consequence analyses has been on specific waste-isolation scenarios and calculated performance measures appropriate to such a hypothetical setting. In addition to the results of scenario-specific analyses, however, one must also consider the likelihood of such a scenario or, at least, how the results compare to "expected" system performance. To date, there has been little agreement in defining either scenario probabilities or expected performance. In the absence of such information, the parametric analyses that follow can provide a useful framework in which to view the results of specific analyses.

3.3.2 Parametric Analysis Code

In Section 3.2, we developed transport, release, and dose models based on the previously described parameters: T_b , T_d , T_v , S , and I . The XCOMP computer code is written to systematically vary these parameters over analyst-specified ranges and a finite number of steps. The ranges are equally subdivided logarithmically. Graphics output is computer drawn as a function of any specific parameter. The analyst may compare two different waste forms or the same waste form with differing choices of parameters. He may further choose to compare different transport and/or convolution with release models. There is also an option to set dispersion as a ratio of mean transit time.

Note that the XCOMP code does not take into account any network of flow paths. Rather, it assumes only some mean transit time for a single waste pulse.

3.3.3 Parametric Studies

The analyses described below consider the impact of each system characteristic on overall systems performance. Because a given systems performance is not unique to a single set of input parameters, however, the analyses must be considered as a whole and the possibility of dominant parameter tradeoffs must be recognized. To simplify interpretation of the complete analysis, it is helpful to first identify the more-general performance trends so that specific cases may be interpreted in light of the findings.

Pulse Width. An increase in geologic dispersivity causes the pulse width (S) to increase. The impact of this pulse-spreading is generally assumed to reduce the peak curie flux. To investigate this assumption, we calculated system performance as a function of transit time for ratios of $S/T_t = 0.1$ (low geologic dispersivity) to $S/t_t = 1.0$ (high geologic dispersivity); the studies were carried out for several waste-dissolution times with an assumed negligible breach time ($T_b = 1.0$ year). Figures 38(a) and 38(b) illustrate the impact of pulse width on peak normalized flux for the representative cases where $T_d = 10^2$ and 10^4 years, respectively. As expected, the larger T_d results in a lower initial peak flux that falls off approximately linearly when $T_t \geq T_d$. In agreement with general expectation, the peak flux for the case of $S/T_t = 1.0$ is less than or equal to that for the case of $S/T_t = 0.1$ in all instances. When nuclide group decay is included (i.e., peak activity flux), however, this expectation must be reexamined. Figure 39(a) shows the peak activity-flux performance measure for Group-II nuclides, plotted as a function of T_b for $T_d = 1.0 \times 10^4$ years. Because of the almost step-function decrease in fission-product nuclides (see Fig. 22) over the 10,000-year time scale, the wider (more geologically dispersed) waste pulse may not yield the minimum hazard over these times. Increased dispersivity does reduce the hazard, as expected, for the Group-I and Group-III nuclides, as shown in Figs. 39(b) and 39(c).

The trend in peak-flux arrival times can be summarized as follows:

- For relatively short T_d ($\sim 10^2$ years), the wider ($S/T_t = 1.0$ pulse) exhibits an earlier peak-arrival time (about one order of magnitude for all but the shortest transit times). See Fig. 40(a).
- For relatively long T_d ($\sim 10^6$ years), the narrower ($S/T_t = 0.1$ pulse) exhibits an earlier peak-arrival time (about one order of magnitude for all transit times). See Fig. 40(b).

Figure 38. Impact of pulse width on peak normalized flux.

- For intermediate T_d 's ($\sim 10^4$ years), the wider pulse arrives earlier for long transit times while the narrower pulse arrives earlier for short transit times [see Fig. 40(c)].

Because of their different nuclear decay characteristics, peak activity-flux arrival times for the three nuclide groups differ slightly from the above results.

Breach Time. Throughout the FY 1980 analyses, no credit was taken for leach protection by the waste canister after emplacement, i.e., $T_b = 0.0$ years. A preliminary study² examined the impact of this assumption and found it to produce conservative (i.e., worst-case) results for the FY 1980 base-case siting scenario. The current parametric analyses expanded the scope of this study to encompass the previously defined parameter ranges in order to better understand how and under what conditions breach time impacted overall performance.

Figure 39. Impact of pulse width on peak activity flux—(a) Group-II, (b) Group-I, and (c) Group-III nuclides.

The general behavior of breach time on the peak activity flux can be demonstrated by the Group-II nuclides, shown in Figs. 41(a) and 41(b), as a function of waste-dissolution time for $S/T_t = 0.5$ and transit times of 10^2 and 10^4 years, respectively. For transit times less than the breach time [see Fig. 41(a)], longer T_b reduces hazard due to the additional time available for nuclide decay

Figure 40: Impact of pulse width on time of peak flux arrival.

before the onset of initial release. This effect is observed for all waste-dissolution times and only for those nuclide groups whose activity does, in fact, decay during breach time (unlike Group I). The hazard curves for transit times longer than breach time [see Fig. 41(b)] are identical for both long and short breach times because any effect of T_b is masked by the impact of the longer transit time. This observation is supported by Figs. 42(a)

Figure 41. Impact of breach time on peak activity flux as a function of waste-dissolution time—Group-II nuclides.

and 42(b), which plot the peak activity flux of Group-II nuclides for one- and one-thousand-year breach times vs transit times for $T_d = 10^2$ and 10^4 years, respectively. The hazard curves in both figures are separated initially by about four orders of magnitude and coalesce near transit times of 10^3 years, at which time the initial radioactivity of the Group-II nuclides has decayed by four orders of magnitude. The performance results presented in Figs. 41(a) through 42(b) are consistent internally. Differences in the shapes and magnitudes of the hazard curves in Figs. 41(a) and 41(b) are due to the differences in transit time and will be discussed below.

The impact of breach time on the time of peak activity flux for Group-I nuclides is illustrated in Fig. 43 and is essentially identical for all nuclide groups and waste-dissolution times. As expected, the time of peak arrival is delayed until the breach time for $T_i \leq T_b$ and T_b has negligible impact on arrival times where $T_i \gg T_b$.

Figure 42. Impact of breach time on peak activity flux as a function of transit time—Group-II nuclides.

Transit Time and Waste-Dissolution Time. We designed a series of analyses to systematically examine the impacts of transit time and waste-dissolution time on the overall performance of a waste-isolation system. The peak activity flux (curies per canister per year) and time-of-peak activity flux (years) for each nuclide group have been calculated first as a function of transit time (1.0 to 1.0×10^6 years) for waste-dissolution times of 1.0 , 1.0×10^4 , and 1.0×10^6 years and second as a function of waste-dissolution time (1.0 to 1.0×10^6 years) for transit times of 1.0 , 1.0×10^2 , 1.0×10^4 , and 1.0×10^6 years. To simplify the interpretation of these analyses, S/T_t and T_b were fixed at 0.2 and 1.0 year, respectively, except where otherwise noted. The performance impact trends, discussed in the preceding sections for pulse width and breach time, respectively, provide the data necessary to interpolate these results to other scenario conditions.

Figure 43. Impact of breach time on peak activity flux as a function of transit time—Group-I nuclides.

The peak activity flux (curies per canister per year) of each nuclide group is plotted as a function of T_t for several T_d values in Figs. 44(a), (b), and (c). In most cases, the peak activity flux released to the biosphere from each nuclide group is less than 10^{-1} curie per canister per year. Only release scenarios with overall transit times of less than 10^3 years were identified as possible exceptions. Actual activity fluxes of more than 10^{-1} curie per canister per year were predicted, however, only when the impact of short transit time was compounded by short waste-dissolution time; even these exceptions can be mitigated by a longer breach time or an increased pulse width. Compare, for example, Fig. 44(b) ($T_b = 1.0$ year) with Fig. 45 ($T_b = 10^3$ year); note that this impact is as described in the preceding section on breach time.

Figures 46(a), (b), and (c) show the times of peak activity flux for each of the nuclide groups as a function of T_t for several T_d values. Peak flux-arrival times could never be less than breach times, as illustrated by a comparison of Fig. 46(b) ($T_b = 1.0$ year) with Fig. 47 ($T_b = 10^3$ years), but otherwise appear essentially linear on the log-log plot. Several orders of magnitude variation in waste-dissolution time produced only factor-of-2 changes in peak flux-arrival times, and even this effect was only evident for $T_d < T_t$.

The peak activity flux for each nuclide group is again plotted in Figs. 48(a), (b), and (c), but as a function of waste-dissolution time for several T_t values. For short transit times, the peak activity flux is essentially linear on a log-log plot against waste-dissolution time. The Group-I curves [see

Figure 44. Impact of waste-dissolution time on peak activity flux – (a) Group-I, (b) Group-II, and (c) Group-III nuclides.

Figure 45. Impact of waste-dissolution time on peak activity flux – Group-II nuclides.

Figure 46. Impact of waste-dissolution time on time of peak activity flux—(a) Group-I, (b) Group-II, and (c) Group-III nuclides.

Figure 47. Impact of waste-dissolution time on time of peak activity flux—Group-I nuclides.

Figure 48. Impact of transit time on peak activity flux—(a) Group-I, (b) Group-II, and (c) Group-III nuclides.

Fig. 48(a)] clearly illustrate the impact of waste-pulse width and the tradeoff between pulse width due to the fixed S/T_t ratio or to the dissolution time. Because there is essentially no nuclear decay for times less than 10^5 years, the only reduction in Group-I peak flux comes about by increasing the width of the waste pulse. The $T_t = 10^6$ -year dose curve does result in a lower peak activity flux as a result of Group-I decay, as do all longer ($T_t > 1$ year) transit times for Groups II and III.

Figure 49. Impact of transit time on time of peak activity flux—(a) Group I, (b) Group-II, and (c) Group-III nuclides.

Finally, the time of peak activity flux is plotted in Figs. 49(a), (b), and (c) for each nuclide group as a function of waste-dissolution time for several T_t values. It is obvious from these curves that T_t dominates the time of peak activity-flux arrival; increasing the breach time uniformly shifts these plots to correspondingly later times.

REFERENCES

1. *Nuclear Waste Form Risk Assessment for U.S. Defense Waste at Savannah River Plant, Annual Report Fiscal Year 1980*, Lawrence Livermore National Laboratory, Livermore, Calif., UCRL-53188-80 (1981).
2. L. L. Edwards, *MACRO1: A Code to Test a Methodology for Analyzing Nuclear Waste Management Systems*, Lawrence Livermore National Laboratory, Livermore, Calif., UCRL-52736 (1979).
3. P. Cerny, "The Present Status of the Analcime-Pollucite Series," *Canadian Min.* **12**, 334-341 (1974).
4. S. A. Gallagher and G. J. McCarthy, "Preparation and X-ray Characterization of Pollucite ($\text{CsAlSi}_2\text{O}_6$)," *J. Inorg. Nucl. Chem.*, **43** 1773-1777 (1981).
5. R. M. Barrer and N. McCallum, "Hydrothermal Chemistry of Silicates, Part 4, Rubidium and Cesium Aluminosilicates," *J. Chem. Soc. (London)* **53**, 4029-4035 (1953).
6. R. M. Barrer, J. W. Baynham, and N. McCallum, "Hydrothermal Chemistry of Silicates, Part 5, Compounds Structurally Related to Analcite," *J. Chem. Soc. (London)* **53**, 4035-4041 (1953).
7. S. A. Kashik, I. K. Karpov, and G. V. Kozlova, "An Empirical Method of Calculating Free Energies for Layer Silicates," *Geochem. Intl.* **7**, 70-74 (1978).
8. Y. Tardy and R. M. Garrels, "Prediction of Gibbs Energies of Formation of Compounds from the Elements—2. Monovalent and Divalent Metal Silicates," *Geochimica Et Cosmochimica Acta* **41**, 87-92 (1977).
9. Y. Tardy and R. M. Garrels, "Prediction of Gibbs Energies of Formation of Layer Silicates," *Geochimica Et Cosmochimica Acta* **38**, 1101-1116 (1974).
10. I. K. Karpov and S. A. Kashik, "Computer Calculation of Standard Isobaric-Isothermal Potentials of Silicates by Multiple Regression from a Crystallo-chemical Classification," *Geochem. Intl.* **5**, 706-713 (1968).
11. J. O. Nriagu, "Thermochemical Approximations for Clay Minerals," *Am. Min.* **60**, 834-839 (1975).
12. C. H. Chen, "A Method of Estimation of Standard Free Energies of Formation of Silicate Minerals at 298.15 K," *J. Am. Sci.* **275**, 801-817 (1975).
13. D. D. Wagman, W. H. Evans, V. B. Parker, and R. H. Chumm, *Chemical Thermodynamic Properties of Compounds from Sodium, Potassium, and Rubidium: An Interim Tabulation of Selected Values*, National Bureau of Standards, NBSIR 1034 (1976).
14. W. M. Latimer, *The Oxidation States of the Elements and Their Potentials in Aqueous Solutions* (Prentice-Hall, New York), 2nd ed. (1952).
15. H. C. Helgeson, H. C. Delany, J. M. Nesbitt, H. Wayne, and D. Bird, "Summary and Critique of the Thermodynamic Properties of Rock-Forming Minerals," *J. Am. Sci.* **276-A** (1978).
16. R. A. Robie, B. S. Hemingway, and J. R. Fisher, *Thermodynamic Properties of Minerals and Related Substances at 298.15 K and 1 bar (10 exp 5 pascals) Pressure and at Higher Temperatures*, U.S. Geological Survey Bulletin 1452 (1978).
17. G. B. Naumov, B. N. Ryzhenko, and I. L. Khodarovsky, *Handbook of Thermodynamic Data*, I. Barnes and V. Speltz, Eds., U.S. Geological Survey, USGSWRD-74-001 (1974). (English translation by G. Soleimani, 1971.)
18. F. D. Rossini, D. D. Wagman, W. H. Evans, S. Levine, and I. Jaffe, *Selected Values of Chemical Thermodynamic Properties*, National Bureau of Standards Circular 500 (1952).
19. *JANAF Thermochemical Tables*, Dow Chemical Company, Midland, Michigan.
20. D. D. Jackson, *Thermodynamics of the Gaseous Hydroxides*, Lawrence Livermore National Laboratory, Livermore, Calif., UCRL-51137 (1971).
21. R. Bedford and D. D. Jackson, *Volatilities of the Fission Product and Uranium Oxides*, Lawrence Livermore National Laboratory, Livermore, Calif., UCRL-12314 (1965).
22. J. R. Wiley, "Leach Rates of High Activity Waste from Borosilicate Glass," *Nucl. Tech.*, **43** 268-272 (1979).
23. T. F. Harvey, *Suggested Technical Scheme to Help Resolve Regulatory Issues*, Lawrence Livermore National Laboratory, Livermore, Calif., UCID-17858 (1978).
24. *Earth Science Technical Plan for Disposal of Radioactive Waste in a Mined Repository* (draft), Office of Nuclear Waste Management, U.S. Department of Energy and Geological Survey, DOE/TIC-4033 (1980).
25. B. I. Ross et al., *NUTRAN: A Computer Model of Long-Term Hazards from Waste Repositories*, The Analytical Sciences Corporation, Reading, Mass. (1979).

26. J. E. Campbell, P. C. Kaestner, B. S. Langkopf, and R. B. Lantz, *Risk Methodology for Geologic Disposal of Radioactive Waste: The Network Flow and Transport (NWFT) Model*, Sandia National Laboratory, Albuquerque, N.M., NUREG/CR-1190, SAND 79-1920 (1980).
27. A. M. Kaufman, L. L. Edwards, and W. F. O'Connell, "A Repository Post-Selling Risk Analysis Using MACRO," in *Proc. Symp. Waste Mgmt., Tuscon, Arizona, 1980*, 109.
28. P. F. DuBois and R. C. Basinger, *MATHLIB*, Lawrence Livermore National Laboratory, Livermore, Calif., LCSD-442, Rev. 0 (1980).
29. A. M. Kaufman, *Waste Pipe Calculus*, Lawrence Livermore National Laboratory, Livermore, Calif., UCID-17953 (1978).
30. W. J. O'Connell, *Waste Pipe Calculus Extensions*, Lawrence Livermore National Laboratory, Livermore, Calif., UCID-18136 (1979).
31. M. J. Bell, *ORIGEN—The ORNL Isotope Generation and Depletion Code*, Oak Ridge National Laboratory, Oak Ridge, Tenn., ORNL-4628 (1973).
32. L. E. Berman et al., *Analysis of Some Nuclear Waste Management Options*, Lawrence Livermore National Laboratory, Livermore, Calif., UCRL-13917, Vols. I and II (1978).
33. S. G. Oston, *Finite Medium Green's Functions Solutions to Nuclide Transport in Porous Media*, The Analytical Sciences Corporation, Reading, Mass., TR-1485-2 (1979).
34. N. A. Bonner and Y. C. Ng, *BIODOSE: A Code for Predicting the Dose to Man from Radionuclides Released from Underground Nuclear Waste Repositories*, Lawrence Livermore National Laboratory, Livermore, Calif., UCID-18652 (1980).
35. T. H. Gould, Savannah River Laboratory, Aiken, South Carolina, private communication (May 23, 1980).
36. Committee 2, International Commission on Radiological Protection, "Limits for Intakes of Radionuclides by Workers," *Radiation Protection*, ICRP 30, Part 1 (Pergamon Press, N.Y., 1978).
37. U.S. Nuclear Regulatory Commission, "Technical Criteria for Regulating Geologic Disposal High-Level Radioactive Waste—Advance Notice of Proposed Rulemaking," *Fed. Reg.* 45(94), (1980).
38. U.S. Department of Energy, *Statement of Position of the U.S. Department of Energy, Waste Confidence Rulemaking*, DOE/NE-0007, PR-50, 51 (44FR61372), (1980).
39. International Commission on Radiological Protection, *Recommendations of the International Commission on Radiological Protection*, ICRP 9 (1965).
40. U.S. Atomic Energy Commission, *Reactor Safety Study—An Assessment of Accident Risks in U.S. Commercial Nuclear Power Plants*, Wash-1400, NUREG-75/014 (1975).
41. H. C. Burkholder, M. O. Cloninger, D. A. Baker, and G. Jansen, "Incentives for Partitioning High Level Waste," *Nucl. Tech.* 31, 202 (1976).
42. R. Heckman, D. Towse, D. Isherwood, T. Harvey, and T. Holdsworth, *High Level Waste Repository Site Suitability Study—Status Report*, Lawrence Livermore National Laboratory, Livermore, Calif., UCRL-52633, NUREG/CR-0578 (1979).
43. J. Chandler, Savannah River Laboratory, unpublished data (1981).
44. U.S. Nuclear Regulatory Commission, "Disposal of High Level Radioactive Wastes in Geologic Repositories," *Fed. Reg.* 48 (130), (1981). Also see 10 CFR60.