NUREG/CR-6505, Vol. 1
ORNL/TM-13323/V1

The Potential for Criticality Following Disposal of Uranium at Low-Level Waste Facilities

Uranium Blended with Soil

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Prepared for
U. S. Nuclear Regulatory Commission

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Manuscript Completed: January 1997
Date Published: June 1997

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U.S. Nuclear Regulatory Commission
Washington, DC 20555-0001
NRC Job Code L1376

ABSTRACT

The purpose of this study was to evaluate whether or not fissile uranium in low-level-waste (LLW) facilities can be concentrated by hydrogeochemical processes to permit nuclear criticality. A team of experts in hydrology, geology, geochemistry, soil chemistry, and criticality safety was formed to develop achievable scenarios for hydrogeochemical increases in concentration of special nuclear material (SNM), and to use these scenarios to aid in evaluating the potential for nuclear criticality. The team's approach was to perform simultaneous hydrogeochemical and nuclear criticality studies to (1) identify some achievable scenarios for uranium migration and concentration increase at LLW disposal facilities, (2) model groundwater transport and subsequent concentration increase via sorption or precipitation of uranium, and (3) evaluate the potential for nuclear criticality resulting from potential increases in uranium concentration over disposal limits. The analysis of SNM was restricted to ^{235}U in the present scope of work. The outcome of the work indicates that criticality is possible given established regulatory limits on SNM disposal. However, a review based on actual disposal records of an existing site operation indicates that the potential for criticality is not a concern under current burial practices.

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EXECUTIVE SUMMARY

The purpose of this study was to evaluate whether or not fissile uranium in low-level-waste (LLW) facilities can be concentrated by hydrogeochemical processes to permit nuclear criticality. This investigation presents the first attempt to jointly study the potential for nuclear criticality at LLW facilities using both quantitative hydrogeochemical processes and nuclear criticality safety calculations.

The study was initiated because of a rule change petition (Federal Register, 1993) requesting a predisposal concentration limit for special nuclear material (SNM) in soil to amend the present mass limit specified by Title 10, *Code of Federal Regulations*, Part 150 (10 CFRP. 150). Preliminary analysis by the U.S. Nuclear Regulatory Commission (NRC) suggested that concentrations of SNM in geometries that constitute nuclear criticality safety concerns are plausible. The NRC staff subsequently determined that further analysis was needed to evaluate the hydrogeochemical processes that might lead to an increase in concentration of SNM, and to evaluate the potential for nuclear criticality under conditions representative of LLW disposal facilities. The NRC asked Oak Ridge National Laboratory (ORNL) to form a team of experts in hydrology, geology, geochemistry, soil chemistry, and criticality safety to develop possible scenarios for hydrogeochemical increases in concentrations of SNM and to use these scenarios to aid in evaluating the potential for nuclear criticality.

The team's approach was to perform simultaneous hydrogeochemical and nuclear criticality studies to (1) identify some realistic scenarios for uranium migration and concentration increase at LLW disposal facilities, (2) model groundwater transport and subsequent concentration increase via sorption or precipitation of uranium, (3) evaluate the potential for nuclear criticality resulting from potential increases in uranium concentration over disposal limits, and (4) estimate potential radiation exposures to personnel resulting from criticality consequences. The analysis of SNM was restricted to ^{235}U in the present scope of work. Three outcomes of uranium concentration are possible:

1. Uranium concentration is increased to levels that *do* pose a criticality safety concern.

2. Uranium concentration is increased, but levels *do not* pose a criticality safety concern.

3. Uranium concentration does not increase

There are numerous combinations of variables that may lead to or support nuclear criticality in a waste matrix, herein referred to as soil. These variables include

- the composition of the soil (e.g., SiO_2, concrete debris, contaminated combustibles, iron scrap);

- the enrichment of ^{235}U mass relative to the total uranium mass (e.g., less than 5 wt % ^{235}U from commercial power reactor fuel fabrication processes, ~93 wt % ^{235}U from research reactor fuel fabrication processes, 0.7 wt % from natural uranium processes, and less than about 0.2 wt % from enrichment process tails);

- the density of the soil (e.g., grams of compacted debris per cubic centimeter, tons of compacted debris per cubic yard);

- the density of the ^{235}U within the soil (e.g., grams of ^{235}U per cubic centimeter);

- the degree of neutron moderation in the soil (e.g., typically grams of H_2O per cubic centimeter, grams of H_2O per gram of soil, and hydrogen-to-^{235}U atom ratio);

- the degree of neutron reflection (e.g., thickness and density of reflecting materials such as concrete, soil, moist or saturated soil);

- the geometry or distribution of the ^{235}U in the soil (e.g., neutronically infinite deposits, finite geometries typically characterized as spheres, infinite-length cylinders, and finite thickness slabs of infinite length and width); and

Because of the numerous combinations of parameters that could be considered in nuclear criticality evaluations, bounding and simplifying assumptions were used. Nuclear criticality evaluations were performed for simple finite-media geometries and infinite media assuming various densities of the ^{235}U isotope and water for two generic soil media (both having a dry and uncontaminated density of 1.6 g of soil media per cubic centimeter): "SiO_2 soil" (the most conservative media because pure SiO_2 is the least likely soil composition to absorb neutrons, thereby enhancing the potential for criticality) and a "nominal soil" composed of minerals and secondary phases representative of a world-average soil composition. The infinite-media neutron-multiplication constant, k_∞, was determined for mixtures of nominal soil/^{235}U/water and SiO_2 soil/^{235}U/water. Additionally, sphere diameters, infinitely long cylinder diameters, and thicknesses of slabs (infinite in two dimensions) were determined for finite-media geometry neutron-multiplication constants, k_{eff}, equal to a fiducial "critical" value of 0.95. The value of 0.95, rather than 1.0, was selected as a critical value to conservatively account for methods and data uncertainty. The finite-media geometry calculations included a 2-m-thick "neutron reflector" on the surfaces of each type geometry. The neutron reflector consisted of uncontaminated soil having a water content identical to the contaminated soil. The calculations were performed with a one-dimensional (1-D) discrete-ordinates neutron transport theory code in SCALE (1995) using two sets of realistic geologic parameters for various densities of ^{235}U. Calculated critical densities of ^{235}U for various finite-media geometries were translated into concentration factors (CF) (i.e., the ratio of the required ^{235}U density to sustain criticality divided by the maximum authorized ^{235}U density allowed for burial at Envirocare of Utah, Inc.). These CFs were then compared with results from the hydrogeochemical modeling to determine if criticality is plausible for various uranium-concentrating hydrogeochemical scenarios.

Potential, direct radiation exposures were estimated for two postulated simple types of criticalities based upon the hydrogeochemical concentration of fully enriched uranium disposed of within an assumed SiO_2-soil waste matrix. The locations of the estimated personnel radiation exposures were for positions centered over the concentrated deposit 1 m directly above the disposal-site grade and 1 m above the disposal-site grade but displaced about 90 m from the assumed critical concentrated disposals. The concentrated disposals were assumed to have disklike cylindrical geometries with vertical axes. Also, the disklike deposits were assumed to be centered between the disposal trench floor and the disposal-site grade (i.e., about 5 m below the surface of the disposal-site grade). The selected disklike deposits had uranium concentrations consistent with uranium CFs of 3.6 and 10.5 observed from the hydrogeochemical scenarios. Since the postulated critical events are sustained by optimum neutron moderation from water in the soil, the potential fission yields of each event were estimated from the thermal fission energy required to vaporize a sufficient quantity of soil moisture and to "shut down" the criticality. The estimated fission yield estimates were 2.8×10^{21} and 4.1×10^{21} fissions for CFs of 3.6 and 10.5, respectively. The estimated integrated radiation exposures for the two postulated criticalities (2.8×10^{21} and 4.1×10^{21} fissions) ranged from about 150 to 56 rem at 1 m above the disposal-site grade and about 57 to 165 mrem at 1 m above the disposal-site grade but displaced 90 m from the cylindrical axes of the postulated concentrated disposals. The seemingly anomalous estimates of higher radiation exposures for smaller fission yields are the result of the lower density disposal (CF of 3.6) being nearer to the surface of the disposal-site grade than the higher density disposal (CF of 10.5).

Likewise, simplifying assumptions were used for the hydrogeochemical simulations because of the numerous combinations of parameters that can affect the mobilization of ^{235}U. The hydrogeochemical scenarios that were considered were generalized representations of plausible conditions at LLW disposal sites. However, several site-specific concerns were incorporated. The study was directed at permissible disposal practices by the Utah Department of Environmental Quality, Division of Radiation Control, Radioactive Material License as applied to Envirocare of Utah, Inc. Disposal practices at this site were examined to suggest transport distances, likely chemical agents, and where possible, initial concentrations. Two approaches to bounding the initial concentration of ^{235}U were used. First, it was assumed that disposal was at the licensed maximum permissible concentration of ^{235}U (770 pCi/g of soil or 0.0006 g of $^{235}U/cm^3$ of soil assuming 1.6 g of soil/cm^3). Consideration of the maximum permissible levels allows the worst-case concentrations to be evaluated and can also be used as a basis for evaluating a possible rule change to increase permissible concentrations. Second, actual disposed of quantities, estimated from disposal records, were used. This analysis provided a realistic envelope and assessment of present conditions.

Because the spectrum of geochemical and hydrologic processes and conditions that could be relevant to increasing uranium concentration is extremely broad, examining all resulting potential scenarios in detail is not feasible. Therefore, to constrain the range of geologically reasonable scenarios, uranium ore and soil formation processes were reviewed to identify realistic processes, geometry, and time frames that might be expected in LLW disposal cells. The size and shape of ore bodies, or mineral concentrations, in soils placed bounds on realistic geometries for both the first stage of criticality calculations and on scenarios developed with steady-state, equilibrium modeling.

Hydrogeochemical scenarios were developed in conjunction with NRC staff. Hydrogeochemical modeling considered two processes: (1) mobilization of the uranium presently sorbed in the soil and (2) immobilization and increase in concentration of the uranium at a new location. These processes were modeled in a simple 1-D flow system. The simulated column was 10 m long and represented a waste cell plus sorption zone with vertical infiltration of water. Water flow was modeled as a slug of water that entered through the cell cap. The simulated column contained one or several zones of different chemistry that could concentrate uranium. These zones included (1) a single zone at the bottom of the column with a greater abundance of adsorption sites than the rest of the soil column, (2) multiple zones of higher sorption distributed at different levels within the column, or (3) a reducing zone at the bottom of the column.

Simulation of uranium transport and fate in LLW sites requires a computer code that includes multiple uranium species and sorption or precipitation that vary in time and space along flow paths. Thus, a multispecies reactive transport code was used to examine the behavior of uranium [Parallel Aquifer and Reservoir Simulator, or PARSim (Arbogast et al., 1994)]. In addition, the U.S. Geological Survey code PHREEQE (Parkhurst, Thorstenson, and Plummer, 1982) was used to calculate chemical equilibria (without transport) for initial conditions and for reducing conditions.

The mobilizing agents, competing complexes, and mineral and sorption equilibria used in the analysis are only a subset of conditions that could exist at a site. They were selected as a first test case but do not represent actual conditions at a particular site. As such, further consideration of site conditions is needed to evaluate whether the modeling represents a worst-case scenario. A sensitivity analysis was performed to examine the effect of varying the values of uranium and total inorganic carbon concentrations, pH, velocity, density of sorption sites and number of sorption zones.

The key assumptions in the study were as follows:

- Saturated flow occurs through a 1-D column with uniform porosity under equilibrium conditions [*Transport times will be much slower in unsaturated soil, and enhanced concentration of uranium caused by lateral transport—that is, two-dimensional (2-D) or three-dimensional (3-D) flow—is ignored*.]

- Selection of "most likely" complexes and mineral phases is limited. *(Other mobilization and demobilization agents should be modeled to evaluate their importance*.)

- Water content and ^{235}U concentration for the criticality assessment span a broad range of values.

- Simplistic deposit geometries, having no density gradients, were used in the criticality assessment. *(Smaller quantities of fissile material within equivalent volumes may be required to reach criticality for certain density gradients.)*

The criticality concern at the Envirocare site, as currently used, is judged to be vanishingly small because of the historic burial practices (tamping of materials to eliminate significant voids), recorded inventories and inferred low uranium enrichments (evidenced from provided records), and the CFs required for criticality concern. Although the Envirocare site was not explicitly modeled, the maximum disposal concentrations of ^{235}U that are licensed by Utah were enveloped within the criticality evaluations. The average weight percent concentration of ^{235}U in the LLW inventory at Envirocare over the years 1988–95 was estimated to be 0.42%. This average enrichment is below the minimum enrichment of 1% ^{235}U required to achieve nuclear criticality in a water-moderated, homogeneous media. Even if slightly higher enrichments occurred locally, large uranium CFs would be required to reach nuclear criticality.

The results of comparing the hydrogeochemical modeling simulations and criticality safety evaluations indicated that the increase of uranium concentration to levels of concern is not expected but is theoretically possible for current regulatory limits. This result is not unexpected since it has been observed that uranium ores occur in nature, and soil-forming processes concentrate other elements. What is useful about this work is that the sensitivity analysis identified factors that lead to increases in uranium concentration, the methods to evaluate other conditions have been tested, and the limitations of this type of work are now better known. For example, to concentrate uranium, a mobilizing agent must be sufficiently concentrated to transport uranium but not so high that immobilization cannot occur.

Nevertheless, the current and proposed regulatory limits do not exclude the possibility of concentrating uranium to critical densities. The recommendation from this work is that the NRC extend the approach described here to help formulate regulatory positions (regulations, guidelines, operating criteria, etc.) that will limit the potential for criticality within LLW disposal cells. Uranium concentration increase and criticality must be explored over a broader range of LLW disposal-site environments. Parameter ranges, and possibly key mobilization and immobilization mechanisms, relevant to a dry, carbonate-rich disposal cell in Clive, Utah, may be very different from those relevant to a disposal cell in a humid, highly weathered soil environment. Furthermore, uncertainties in hydrogeochemical model outcomes must be reduced, both within the scope of work presented here and in any future work exploring other hydrogeochemical settings. This reduction will require an iterative process that first examines a broad range of parameter values, followed by further analyses within a more refined range of parameters based on conditions that appear to raise the greatest safety concerns.

Specific operational recommendations to prevent reconfiguration of uranium are also suggested from this preliminary work. Inhibiting water infiltration would inhibit mobility and lengthen travel times by orders of magnitude. Soil caps, if not properly designed and maintained, may not provide an adequate barrier during storm events. Avoiding redox zonation would minimize the potential for concentration increase because redox fronts

can cause changes in uranium mobility and concentration. For example, avoiding disposal of reducing material in the unsaturated zone could help maintain oxidizing conditions. Maintaining a reducing zone and precipitating uranium would not be expected to occur in this setting. However, under oxidizing conditions, uranium could be immobilized by other minerals or mobilized and dispersed outside the disposal cell. Reducing the areal density of fissile material within a waste cell reduces the possible density within a concentrated slab. Areal density can be reduced by limiting disposal thicknesses or disposal concentrations. Nuclear criticality safety calculations can provide the maximum areal densities for fissile material that are acceptably subcritical. Avoiding disposal of highly enriched waste similarly reduces the fissile mass available to concentrate uranium to levels of concern.

SUMMARY OF CONCLUSIONS

- A small number of unexpected, but theoretically possible, scenarios indicate that criticality at a LLW facility is possible based on licensing disposal limits at Envirocare of Utah, Inc.

- Configurations evaluated are geologically reasonable but tend toward worst-case scenarios. The evaluation indicates that a somewhat narrow range of conditions is required for criticality to be of concern. Further characterization of site-specific parameters would better define the range of conditions.

- Under unsaturated conditions, long times would be required to reconfigure uranium (on the order of thousands of years).

- Conditions presenting criticality safety concerns do not currently exist at the Envirocare site based on disposal records that imply dispersal and dilution of the ^{235}U isotope with very large quantities of natural (0.7 wt % ^{235}U) and depleted (~0.2 wt % ^{235}U) uranium.

SUMMARY OF RECOMMENDATIONS

- The analysis presented here suggests that rule changes to provide for concentration limits on SNM should consider the potential for concentration increases of SNM by hydrogeochemical processes.

- Further analysis of scenarios, site-specific data, and recommendations to prevent reconfiguration of SNM should be made for existing and proposed rules.

- Reconfiguration of SNM should be prevented by inhibiting water infiltration, maintaining uniform redox conditions, and adding demobilizing agents. Areal density limits for disposal of ^{235}U and uranium enrichment limits, based upon possible hydrogeochemical uranium-concentrating processes, should be considered.

ACKNOWLEDGMENTS

This work was supported by the NRC under Task 14, Reconcentration of SNM in Low-Level Disposal Facilities, of JCN L1376, Technical Support for Design, Construction, Operation, and Performance Reviews for Low-Level Waste. V. Colten-Bradley of the NRC provided technical direction and contributed substantially to the work throughout the duration of this project.

The authors would like to acknowledge the helpful assistance provided by Patricia B. Fox and Lester M. Petrie, Jr., for the modeling of numerous neutronic calculations and graphic representations of the computational results and Robert L. Childs for the modeling and interpretation of the radiation transport computations. Assistance with hydrogeochemical modeling and visualization of results was provided by Lianfa Song. Soil chemistry data were provided by Baohua Gu, detailed description of soil genesis was provided by David Lietzke, and technical advice on modeling was provided by Mark Elless, Phil Jardine, and Fredrik Saaf. We also acknowledge thoughtful reviews of this manuscript by Gary Jacobs, Liyuan Liang, and Ellen Smith.

1 INTRODUCTION

This task was undertaken to evaluate the potential for nuclear criticality at low-level waste (LLW) disposal facilities. Simultaneous studies of hydrogeochemical simulations and of nuclear criticality parametric evaluations were performed to determine the parametric phase space (hydrogeochemical and neutronic) that would mutually indicate nuclear criticality in two different "soil" matrices.

1.1 PURPOSE

The purpose of this study was to evaluate the potential for hydrogeochemical processes to mobilize and concentrate special nuclear material (SNM) contained in soil and construction debris disposed of in LLW facilities into configurations of sufficient density and geometry to permit nuclear criticality. The evaluation of increases in SNM concentration is restricted to criticality safety concerns associated with the ^{235}U isotope of uranium. Consideration of fissile radioisotopes other than ^{235}U are beyond the scope of this study.

1.2 RATIONALE FOR STUDY

A rule change petition (Federal Register, 1993) requested that the U.S. Nuclear Regulatory Commission (NRC) develop a concentration limit for SNM in soil to amend the present mass limit specified by Title 10, *Code of Federal Regulations*, Part 150 (10 CFR P. 150). Under the proposed rule, a licensee would be authorized to possess an unlimited quantity of SNM if the mass ratio of soil to SNM is greater than specified values. The Division of Fuel Cycle Safety and Safeguards has proposed the following SNM limits:

- 11500 g soil per 1 g ^{239}Pu,
- 5000 g soil per 1 g ^{235}U (100% enriched), and
- 250 g soil per 1 g ^{235}U (10% enriched).

These limits ensure subcritical conditions, provided the concentrations do not change. The following assumptions were used to obtain the limits:

1. uniform distribution of SNM in soil,
2. silicon dioxide (SiO_2) as soil model, and
3. spherical geometry and optimal water content for nuclear criticality.

In determining these limits, no consideration was made for increases in concentration of SNM resulting from solution transport to produce new chemical compounds and geometry.

To support this rule-making activity that would add concentration limits of SNM in soil to the possession limits set by 10 CFR P. 150, the NRC performed a preliminary conservative analysis to evaluate the potential for SNM at a LLW disposal facility to be dissolved, transported in solution, and concentrated as a precipitate. The impetus for this analysis is the potential for such concentrated SNM to result in a nuclear criticality. The results of the preliminary analysis performed by the NRC staff suggest that, under the assumptions of the postulated scenarios, concentration of SNM in geometries that constitute nuclear criticality safety concerns is plausible.

Subsequent to the preliminary evaluation, the NRC staff determined that further analysis was needed to assess the criticality safety concern. Specifically, the NRC judged that additional studies are required to address the potential of SNM disposed of at an LLW facility to become sufficiently concentrated by hydrogeochemical processes to form a critical mass. Thus, Oak Ridge National Laboratory (ORNL) was asked to develop plausible scenarios for changes to SNM concentration and configuration at an LLW site and to evaluate the potential for nuclear criticality on the basis of these scenarios.

1.3 OBJECTIVES

The objectives of this project are to (a) identify achievable scenarios for hydrogeochemical migration accompanied by increases in the concentration of SNM disposed of at LLW disposal facilities, (b) quantitatively model the geochemical scenarios for increasing the concentration of SNM by solute transport in groundwater and subsequent sorption or precipitation of uranium from solution, and (c) evaluate the potential for nuclear criticality under the hydrogeochemical scenarios modeled by using chemical and physical conditions representative of those existing at an LLW disposal site.

1.4 SITES CONSIDERED

This study focuses on two specific circumstances at the LLW facility operated by Envirocare of Utah, Inc., in Clive, Utah. First, the study was directed at disposal practices that are permissible under the Utah Department of Environmental Quality, Division of Radiation Control, Radioactive Material License as applied to Envirocare (i.e., construction of waste cell liners, fill, and caps; the physical form of waste; limitations on compactible and noncompactible debris; and specifications for compaction). Second, the study was limited to maximum ^{235}U concentrations authorized by this license. The maximum average concentration of ^{235}U permitted in disposed waste is 770 pCi/g (Envirocare of Utah, Inc., license UT 2300249), which equates to ~0.6 g of ^{235}U per liter of soil or 1.6 kg of soil with an assumed reference density of 1.6 g/cm^3 based upon Sposito (1989) and Shacklette and Boerngen (1984). The average reported ^{235}U concentrations disposed of at the site were also considered based on Envirocare disposal manifest records. No other specific LLW sites were evaluated. However, the same analysis methods can be used to evaluate other site-specific conditions.

Because information was not available to verify that concentration processes are active at the Envirocare site, and because concentration of uranium may require long periods of time, natural analogs were also considered to provide a broader perspective. Consideration of analogs provides bounds on the sizes and shapes likely to be assumed by the concentrated uranium and on the time scales required to produce these naturally formed concentrations. Analogs included soil genesis and ore deposition.

1.5 GENERAL APPROACH

To approach this problem, a series of reasonable hydrogeochemical scenarios for increasing uranium concentration were developed for which three outcomes are possible (Fig. 1.5-1):

1. uranium concentration is increased to levels that *do* pose a safety concern;

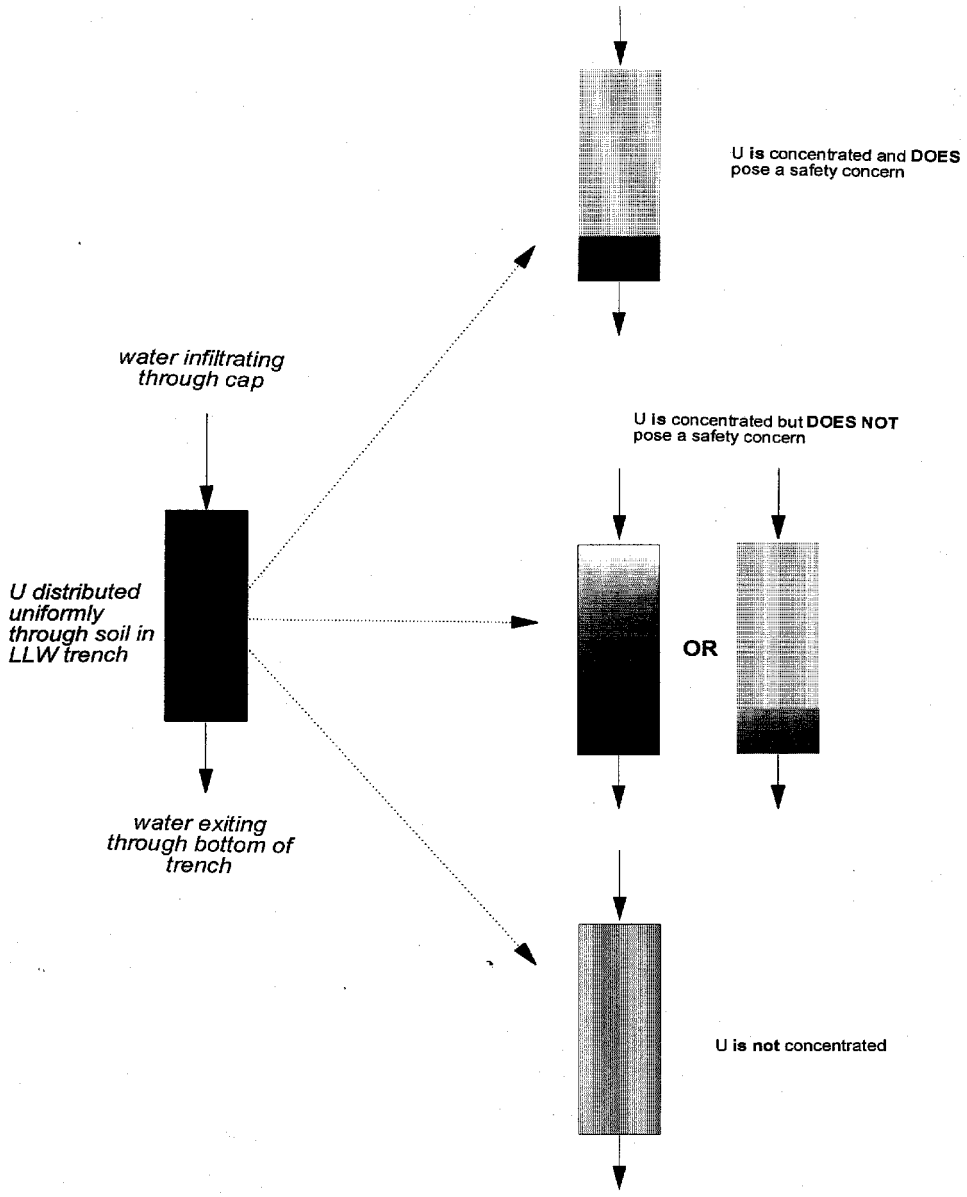


Figure 1.5-1 Possible outcomes for uranium transport in hypothetical columns. Degree of shading indicates relative concentration of uranium. When uranium is concentrated but does not pose a criticality concern, it may be because of (a) insufficient concentration during mobilization or (b) insufficient initial concentration

2. uranium concentration is increased, but levels *do not* raise a safety concern; or

3. uranium concentration is not increased.

Specific task elements followed in this work included (a) criticality safety evaluations that simulated geologically realistic soil mineral compositions, uranium concentrations, and physical configurations; (b) simulation of realistic, albeit simplified, scenarios of interacting hydrologic and geochemical processes that could mobilize and concentrate uranium; and (c) integration of the criticality and hydrogeochemical results to interpret the likelihood of nuclear criticality over a range of uranium concentrations and configurations bounded by hydrogeochemical modeling or by comparison with geological analogs.

1.5.1 Hydrogeochemical Reactive Transport Modeling

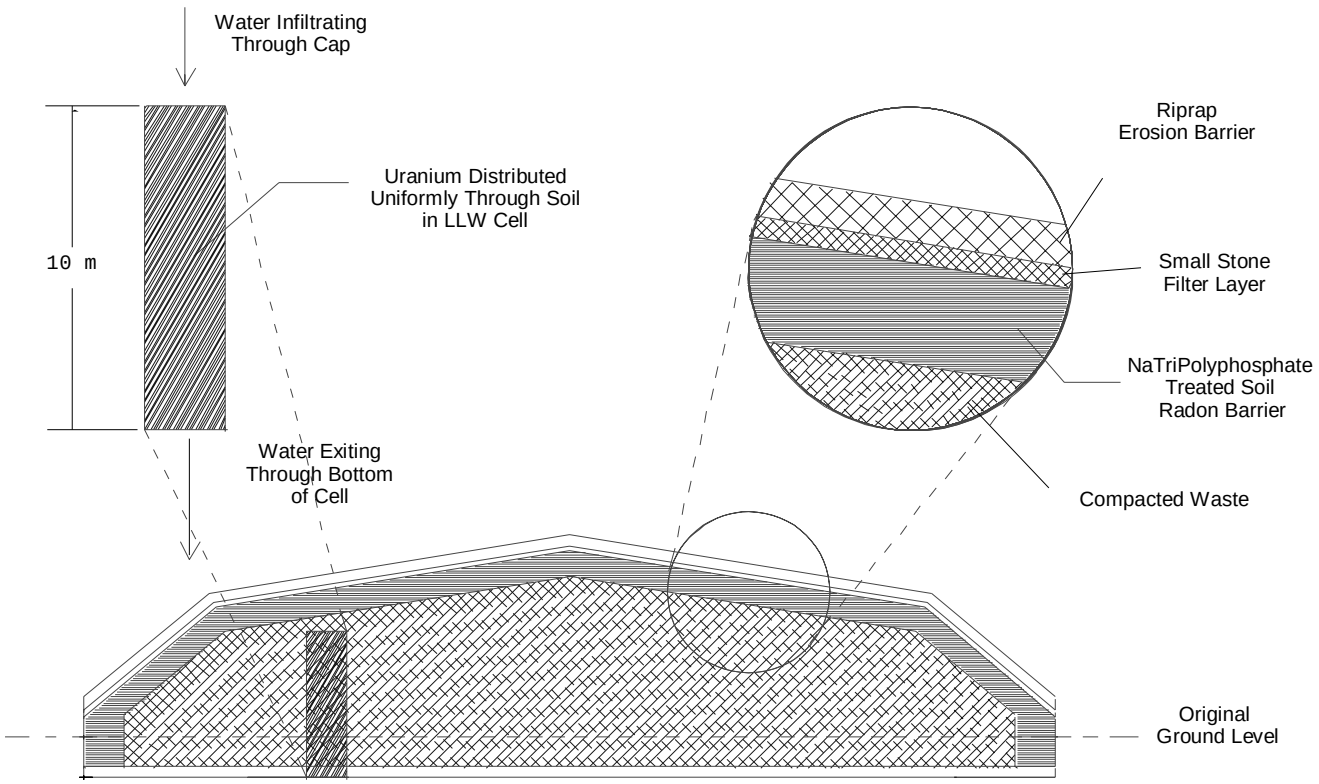
Hydrogeochemical scenarios that can reasonably permit increasing the concentration of uranium involve (1) mobilization of uranium in the disposed of soil followed by (2) subsequent immobilization of the migrating uranium into a localized zone. Mobilization processes include uranium ligand dissolution, desorption, and complexation, especially as carbonate or phosphate complexes. Immobilization mechanisms include evaporation, colloid coagulation, adsorption, and precipitation. Precipitation is particularly relevant under reducing conditions because uranium species have low solubility at low redox potential. The problem was conceptualized as the one-dimensional (1-D) vertical transport of uranium through a 10-m-long column, which represented the uranium-contaminated soil in a disposal cell (Fig. 1.5-2). Water, which represented the composition of rainwater leachate infiltrating the 2-m-thick cap of the cell, entered the top of the 10-m column.

Immobilization was conceptualized as (1) a single zone with a greater abundance of adsorption sites at the bottom of the column, (2) multiple zones of higher sorption distributed at different levels within the column, or (3) a reducing zone at the bottom of the column. The effect of varying the leachate composition on uranium mobilization and concentration under oxidizing conditions was examined by using a formal sensitivity analysis (described in Sect. 7.1.3).

Simulation of the complex geochemical processes that can lead to mobilization and increases in concentration of uranium requires a treatment that deals explicitly with the presence of multiple uranium species, each capable of undergoing adsorption or precipitation processes that vary in time and space along hydrologic flow paths. A multispecies reaction code, designed for parallel supercomputers [PARSim (Arbogast, Dawson, and Wheeler, 1994)], was used to examine the behavior of uranium. All model runs assumed saturated flow, and the number of pore volumes needed to mobilize and concentrate uranium provided a temporal tracing of the processes. Because unsaturated conditions would result in a lower permeability and longer travel times (see Sects. 7 and 8 for additional discussion of the implications of modeling saturated conditions), consideration of saturated conditions for the disposal environment represents a worst-case scenario.

1.5.2 Nuclear Criticality Evaluation

To provide a relevant exchange and coupling of technical information between the nuclear criticality safety and hydrogeochemical analyses, the criticality safety calculations were performed in two stages. The first stage began with hydrogeochemical estimates of geometry and physicochemical parameters (e.g., mineral chemistry, soil density, and soil moisture) as input for the initial criticality safety evaluations. Parameter estimates were based on assumptions about site geology and analogs from soil-forming processes and uranium ore deposition.



NOT TO SCALE

Figure 1.5-2 Schematic illustration of waste soil column evaluated by hydrogeochemical reactive transport modeling

Introduction

These calculations provided some preliminary estimates of the infinite-media neutron multiplication constant (k_∞) by using realistic geologic parameters and "nominal soil" but not accounting for the likelihood that increases in concentration of uranium could occur. The second stage incorporated the limitations suggested by the hydrogeochemical modeling of scenarios to increase the concentration, which supported alternative simple geometries, and expanded to include an SiO_2 waste matrix. This afforded additional realism and conservatism in the assessment of the criticality risk.

There are numerous combinations of variables that may lead to or support nuclear criticality in a waste matrix, herein referred to as soil. They include

- the composition of the soil (e.g., SiO_2, concrete debris, contaminated combustibles, and iron scrap),
- the enrichment of ^{235}U mass relative to the total uranium mass (e.g., less than 5 wt % ^{235}U from commercial power reactor fuel fabrication processes, ~93 wt % ^{235}U from research reactor fuel fabrication processes, 0.7 wt % from natural uranium, and less than about 0.2 wt % from enrichment process tails),
- the density of the soil (e.g., grams of compacted debris per cubic centimeter or tons of compacted debris per cubic yard),
- the density of the ^{235}U within the soil (e.g., grams of ^{235}U per cubic centimeter),
- the degree of neutron moderation in the soil (e.g., typically grams of H_2O per cubic centimeter, grams of H_2O per gram of soil, and hydrogen-to-^{235}U atom ratio),
- the degree of neutron reflection (e.g., thickness and density of reflecting materials such as concrete, soil, and moist or saturated soil),
- the geometry or distribution of ^{235}U in the soil (e.g., neutronically infinite deposits, finite geometries typically characterized as spheres, infinite-length cylinders, and finite thickness slabs of infinite length and width), and
- critical configurations were assumed to be at trench half-depth positions as opposed to near trench bottom positions, thereby reducing the thickness of overburden that could provide additional radiation shielding.

The nuclear criticality evaluations began with the permissible state of Utah license condition limit for ^{235}U contamination in waste (i.e., 770 pCi of ^{235}U per gram of soillike waste) at the Envirocare facility in Clive, Utah. Because of the permissive license condition, the nuclear criticality evaluations considered 100 wt % enriched uranium (i.e., pure ^{235}U), thereby safely bounding lesser uranium enrichments.

Because of the numerous combinations of parameters that could be considered in the nuclear criticality evaluations, bounding and simplifying assumptions were used. Nuclear criticality evaluations were performed for simple finite-media geometries and infinite media assuming various densities of the uranium ^{235}U isotope and water for two generic soil media (both having a dry and uncontaminated density of 1.6 g of soil media per cubic centimeter): "SiO_2 soil" (the most conservative media because SiO_2 is the least likely soil composition to absorb neutrons, thereby enhancing the potential for criticality) and a nominal soil composed of minerals and secondary phases representative of a world-average soil composition. The infinite-media neutron-multiplication constants, k_∞, were determined for each mixture of nominal soil/^{235}U/water and SiO_2 soil/^{235}U/water. Additionally, sphere diameters, infinitely long cylinder diameters, and thicknesses of slabs (infinite in two dimensions) were determined for finite-media geometry neutron-multiplication constants, k_{eff}, equal to a fiducial "critical" value of 0.95.

(Here, and throughout this report, 0.95 is used as the upper limit for neutron-multiplication factors because it provides a 5% margin of subcriticality that accounts for bias and uncertainties in the calculational methods and data.) The finite-media geometry calculations included a 2-m-thick "neutron reflector" on the surfaces of each type geometry. The neutron reflector consisted of uncontaminated soil having identical water content as the contaminated soil. The calculations were performed with a 1-D discrete-ordinates neutron transport theory code in SCALE (1995) using two sets of realistic geologic parameters for various densities of ^{235}U. Calculated critical densities of ^{235}U for various finite-media geometries were translated into CFs (i.e., the ratio of the required ^{235}U density to sustain criticality divided by the maximum authorized ^{235}U density allowed for burial at Envirocare).

1.5.3 Direct Dose Radiation Transport Analysis for Criticality Consequences

Criticality is not likely to occur at the Envirocare LLW facility because of the small likelihood for sufficient hydrogeochemical concentration of uranium and because of the low enrichment of uranium indicated by historic records. However, because there are no license operating constraints on the enrichment of the uranium disposed, there is a theoretical possibility for criticality. Therefore, direct-dose radiation transport calculations were performed to estimate the neutron and gamma radiation doses near the soil surface as a consequence of two postulated critical configurations at the Envirocare facility—one at near minimum ^{235}U critical density and the other at the near maximum ^{235}U density. Both were assumed to be large-diameter (i.e., 18 m), disklike cylindrical deposits of overly water-moderated uranium in SiO_2. Both configurations were vertically centered in a trench. Both configurations were constrained by the maximum allowable 0.0006 g $^{235}U/cm^3$ disposal value (based upon a soil density of about 1.6 g soil/cm^3) that corresponds to an areal density of about 5.7 kg $^{235}U/m^2$ as projected to the bottom of a trench. The first postulated critical configuration was an 18-m-diam disk that was approximately 2.5 m in thickness, which corresponds to a CF of about 3.6. The second postulated critical configuration was an 18-m-diam disk that was approximately 0.9 m in thickness, which corresponds to a CF of about 10.5.

The direct-dose evaluations were performed for two different locations, both at 1 m above the ground surface, for both critical configurations. The first location was directly above the center of the cylindrical deposit, and the second location was 90 m from the first.

The maximum direct total dose was estimated to be about 160 rem at 1 m above ground level directly above the center of the low-density, 2.5-m-thick cylindrical deposit. The minimum direct total dose was estimated to be about 0.06 rem at 1 m above ground level, ~90 m from the center of the 0.9-m-thick cylindrical deposit. Though the estimated fission yield (4.1×10^{21} fissions) for the 0.9-m-thick deposit was nearly twice the estimated fission yield (2.8×10^{21} fissions) for the 2.5-m-thick deposit, the maximum direct doses were observed from the lower ^{235}U density 2.5-m-thick deposit because of the smaller thickness of SiO_2 overburden that provided less shielding for the fission radiation.

1.5.4 Basis for Data Interpretation

Both the nuclear criticality safety analysis and the hydrogeochemical modeling assume that the maximum permissible limits for ^{235}U are disposed of (0.0006 g/cm^3). For the hydrogeochemical modeling, ^{235}U enrichment is not a factor in transport behavior, but total uranium concentration could influence uranium mobilization. Therefore, the total uranium concentration was increased above 0.0006 g/cm^3 to an upper limit of 0.06 g/cm^3. Thus, assuming a fixed ^{235}U concentration of 0.0006 g/cm^3, the range of uranium concentrations used in the hydrogeochemical modeling can be interpreted to span the ^{235}U enrichment range of 1 to 100 wt %. However, because LLW regulations do not limit enrichment, a worst-case scenario of 100% enrichment is assumed in the nuclear criticality safety analysis.

The criticality safety analysis provided information on the concentrations of uranium (grams of uranium per grams of soil) in various shapes and sizes that would be required to achieve $k_{eff} = 0.95$ in nominal and SiO_2 soil. The hydrogeochemical modeling provided information on the extent to which uranium would be mobilized and concentrated under varying hydrologic and geochemical conditions. The analogs (ore deposits, soil genesis) provided additional information on geologically reasonable sizes and shapes of concentrated uranium. By comparing these data, the potential for reasonable hydrogeochemical mechanisms to configure uranium in a concentration and geometry that pose a criticality safety concern was evaluated.

Radiation exposures were calculated for postulated criticalities that were based upon geometric models that could conservatively envelop uranium concentrations in disposals that were considered in the hydrogeochemical scenarios.

1.6 ORGANIZATION OF REPORT

This report is organized into 12 sections. The introduction describes the purpose, rationale, and objectives of the study and outlines the general approach to integrating the results of the hydrogeochemical process modeling, criticality safety analyses, and postulated criticality radiation transport consequence evaluation. The background of the problem is outlined in more detail in Sect. 2. The background section emphasizes the concern that regulatory limits may not adequately address the potential effect of hydrogeochemical processes on mobilization and the increase in concentration of SNM in a disposal cell. The NRC, in commissioning this study, expressed special interest in the LLW disposal facility at Clive, Utah, operated by Envirocare of Utah, Inc. This study addressed some site-specific conditions at that facility, which are described in Sect. 3.

The spectrum of possible geochemical and hydrologic processes that could be relevant to an increase in uranium concentration is broad, and it is not feasible to examine all these potential scenarios. Therefore, to limit the range of geologically reasonable scenarios, uranium hydrogeochemical processes were reviewed (Sect. 4) to provide a realistic frame of reference for key processes, geometry, and time frames that might be expected in LLW disposal cells. Following consideration of natural analogs, scenarios for the mobilization and concentration increase of SNM were developed by ORNL staff and approved by the NRC. The approved scenarios were tested by hydrogeochemical and criticality safety modeling over a range of conditions to evaluate the likelihood of an increase in uranium concentration to densities that pose a criticality safety issue. These scenarios are described in Sect. 5.

The processes of uranium mobilization and concentration increase considered in the modeled scenarios are described in Sect. 6. The details of hydrogeochemical process modeling and nuclear criticality evaluation are described in Sect. 7, and the assumptions underlying the modeling and the parameters used in the models are addressed in Sects. 8 and 9, respectively. The results of the model simulations are presented in Sect. 10 and are discussed within the context of site-specific conditions at the Envirocare site and as they pertain to the more general issue of developing regulatory guidelines to minimize criticality safety risks.

The geological realism and complexity of the modeled geochemical transport processes incorporated into this study exceed that of previous analyses of the potential for concentrated SNM to reach nuclear criticality. However, simplifications still exist that create uncertainties in estimating risks. Limitations of the models and the consequences of uncertainties associated with interpretation of the results are discussed in Sect. 11, and the data required to reduce the uncertainties are identified. Finally, Sect. 12 provides a summary and statement of the conclusions of the study.

2 DESCRIPTION OF PROBLEM

The amount of SNM that can be received, possessed, or transferred by a licensee is restricted by 10 CFR P. 150 to quantities not sufficient to form a critical mass. Part 150.11, Critical Mass, specifically defines limits as follows:

> uranium enriched in the isotope ^{235}U in quantities not exceeding 350 grams of contained ^{235}U; ^{233}U in quantities not exceeding 200 grams; plutonium in quantities not exceeding 200 grams; or any combination of them in accordance with the following formula: For each kind of special nuclear material, determine the ratio between the quantity of that special nuclear material and the quantity specified above for the same kind of special nuclear material. The sum of such ratios for all kinds of special nuclear materials in combination shall not exceed unity.

These restrictions are applicable to waste materials in individual waste packages (e.g., 55-gal drums and boxes) or soil prior to burial. However, the total inventory of SNM contained in all buried waste packages or contaminated soils at a disposal facility can greatly exceed the critical mass limits stated previously.

If disposal packages remain sealed or the disposal facility remains dry, no potential exists for significant movement of SNM. Low probability, potential exceptions include site disturbance or modification caused by tornado, earthquake accompanied by liquefaction of soil, and deliberate or accidental excavation. The potential for movement of SNM increases when the disposal facility is wet. Under wet conditions, uranium can potentially be transported with the advecting fluid as dissolved uranium in a variety of oxidation states: as cationic or anionic complexes with inorganic or organic ligands; as colloidal particles formed by homogeneous precipitation of hydrolyzed uranium or plutonium; or by adsorption or precipitation of the SNM on natural colloids such as layer silicates, mineral oxides, or natural organic material. Physical and chemical characteristics at a waste site may vary both spatially and temporally, effecting changes that block physical migration of solutions or promote chemical exchange, sorption, or precipitation reactions. Both physical barriers and chemical reactions may have the potential to concentrate SNM sufficiently to pose a criticality safety concern. Waste form degradation accompanied by the transport and increase in concentration of SNM form the basis of postulated scenarios that have raised concerns regarding criticality safety at high-level waste facilities (Bowman and Venneri 1994). However, previous analyses have not attempted to account for the hydrogeochemical feasibility of increases in SNM concentration.

3 SITE CHARACTERISTICS

The site characteristics described here are limited to those of the Envirocare site near Clive, Utah. General descriptive remarks were excerpted from the NRC Final Safety Evaluation Report (1994). Additional site information used in this evaluation (including waste cell water leachate chemistry, native soil mineralogy and chemistry, Utah Radioactive Material License to Envirocare of Utah, and SNM inventory) was obtained from the state of Utah, Department of Environmental Quality, Division of Radiation Control, or from Envirocare of Utah, Inc., at the request of the Division of Radiation Control. NRC and ORNL staff conducted a site visit on August 8, 1995, to obtain additional information on site characteristics and LLW disposal operations.

The Envirocare site near Clive, Utah, is located approximately 120 km west of Salt Lake City, Utah, at the eastern margin of the Great Salt Lake Desert in the Basin and Range province of North America. Topography in the Basin and Range is controlled by block-faulted, north-south trending mountain ranges separated by alluvial-filled basins. The lithology of the mountains includes limestone, dolomite, shale, quartzite, and sandstone. Basin sediments consist of Quaternary lacustrine Lake Bonneville deposits and Tertiary colluvial and alluvial materials eroded from adjacent mountains. Unconsolidated to semiconsolidated valley fill sediments are 240 to 300 m thick in the central portions of valleys within the Great Salt Lake Desert. The materials consist of intercalated colluvium, alluvium, lacustrine, and fluvial deposits with some aeolian material.

Precipitation records from Wendover, Utah, provide an estimate of 13 cm/year average annual total precipitation at the Clive site, indicative of a dry climate.

Topographic relief at the South Clive site is ~3 m over the 250-ha tract, sloping from northeast to southwest. The Envirocare facility is underlain by Quaternary lake bed deposits of Lake Bonneville. On-site logs of the subsurface indicate that these lake deposits extend to depths of at least 75 m. Soils at the facility have been mapped as Iosepa silt loam by the U.S. Soil Conservation Service. Translocated clay and sodium minerals are concentrated 10 to 38 cm below the surface, resulting in a relatively impervious soil.

The design of waste cells is modeled after that used by the U.S. Department of Energy (1984) for disposal of uranium mill tailings in the adjacent “Vitro Cell” at the South Clive, Utah, site. The waste cell is constructed in a continuous cut and fill operation such that excavation, fill, and capping take place in a limited area of the planned waste cell. Design features of the waste disposal cells relevant to the evaluation of SNM mobilization and concentration increase (Fig. 1.5-2 schematically illustrates this design) are summarized as follows:

(1) The existing low-relief terrain is excavated to a depth of 2.4 m, and the native soil materials removed are stockpiled for use in incremental capping of the filled waste embankment.

(2) A 0.6-m-thick liner is constructed. The liner consists of scarified and recompacted native soil. The liner is designed to be more permeable than the soil cap to prevent retention of infiltrated water.

(3) Waste material is placed on the liner in 0.3-m (1-ft) lifts and compacted in place to a maximum fill height of 11 m above the original ground elevation. (Note: The vertical dimensions of the Envirocare waste cells presented in various schematic design illustrations obtained from the Utah Division of Radiation Control are not consistent. For modeling purposes, an 8-m source thickness plus a 2-m-thick sorption zone was selected in consultation with NRC staff).

(4) A 2-m-thick layer of native soil treated with sodium tripolyphosphate and compacted in 0.3-m lifts is placed on top of the compacted waste to form a radon and water infiltration barrier. This cap is constructed incrementally as portions of the waste cell are filled to design capacity.

(5) An erosion protection barrier is placed over the radon barrier. The erosion barrier consists of a 0.15-m-thick bedding layer of fine gravel that acts as a filter zone. This is overlain by a 0.46-m (1.5-ft)-thick layer of riprap having a minimum medium-sized rock size (D_{50}) of 2.5 cm on the top slope and 8.75 cm on the side slopes to complete erosion protection. (D_{50} = 2.5 cm and D_{50} = 8.25 cm designate effective rock size for an aggregate having a distribution of sizes with 50% of the "particles" less than 2.5 or 8.25 cm in diameter, respectively.)

Because wastes disposed of at the site may originate anywhere in the United States, mean world soil compositions were used for modeling (Table 3.1-1). The composition of water percolating through the waste was modeled as rainwater equilibrated with the chemically treated and compacted native soil cap. These water compositions compared well with the chemistry of "leachate" runoff waters collected at the margins of active, uncovered portions of the disposal cell (based on data obtained from L. Morton, State of Utah, Department of Environmental Quality). The model water compositions were used in place of the state of Utah site water chemistry because the site water analyses were not charge balanced. Waste disposed of is limited to less than 10% by volume of debris distributed uniformly in a 0.3-m lift (Utah Department of Environmental Quality, 1995, p. 14). Noncompactible debris (i.e., concrete, metal, and stone) distributed uniformly to minimize voids may compose up to 25% by volume of a 0.3-m lift (Utah Department of Environmental Quality, 1995, p. 14). In practice, waste exceeding these criteria are mixed with native soil to reduce the debris content of a lift.

The mineralogy and chemistry of the native soil used to construct the liner and radon barrier cap is dominated by carbonates (4% dolomite, 8% calcite, and 53% aragonite). Clays are secondary (2% kaolinite, 1% illite, 15% smectite), and feldspars and quartz are relatively minor constituents (2% plagioclase, 3% K-feldspar, and 12% quartz). The analysis was provided to Envirocare by Applied Geotechnical Engineering Consultants, Inc., as part of an evaluation of the long-term permeability of the waste cell liner. The high proportion of carbonates contained in the native soil supported the selection of CO_3^{2-} as the most significant complexing ion for geochemical modeling. The addition of sodium tripolyphosphate as a flocculating agent indicated the need to include PO_4^{2-} in geochemical modeling.

Table 3.1-1 Elemental composition for nominal soil

Element	Weight percent of dry bulk density (1.6 g/cm^3)
Carbon	4.290
Oxygen	49.000
Sodium	0.680
Magnesium	0.600
Aluminum	7.100
Silicon	33.000
Potassium	1.360
Calcium	1.370
Iron	2.600

4 RELEVANT ANALOGS

Naturally occurring processes have mobilized (dissolution and hydrogeochemical transport) and concentrated (sorption and precipitation) uranium and other elements to form ore deposits or other mineral segregations (concretions, clay-enriched soils, caliche layers, and fracture fillings). Some of these processes can serve as analogs for migration and concentration of uranium in an LLW facility. The size and shape of these naturally occurring deposits, as well as the time scale required to increase the concentration, place broad bounding limits on the geometry and time periods expected for concentration of uranium in an LLW facility. Literature describing uranium ore and soil-forming processes were reviewed to identify the processes and mechanisms responsible for producing natural mineral concentrations.

After completing the first-stage criticality safety analyses with realistic soil constituents, the sizes and shapes of the predicted critical masses were compared with the geometry of naturally occurring mineral deposits. This comparison was done to evaluate the potential for obtaining the calculated shape and mass of uranium in a waste cell. Planar shapes can be compared to development of enriched clay layers and cement fillings in sedimentary rocks, or salt beds of evaporative deposits, which are natural manifestations of mechanisms to increase concentrations. Planar shapes may be produced by vertical transport and subsequent immobilization of uranium in planar horizons created by variations in chemistry of the thin layers (lifts) making up the waste cell. Transport accompanying flow of surface water through a breech in the radon barrier cap can be anticipated to be dominantly vertical. Spherical shapes may be compared to concretionary nodules that have developed in subsoil horizons. Cylindrical shapes can develop as a result of mineral replacement of organic material accompanying petrification of trees. Spherical and cylindrical concentrations of uranium of sufficient size and density needed to attain nuclear criticality are considered to have a lower probability of formation in LLW facilities; both vertical and lateral transport of uranium would be required. Furthermore, the size of spherical concretions that are observed to develop in soil horizons are not large enough to include a critical mass of uranium, and the limitations on the size of debris included in the waste precludes formation of a cylindrical critical mass by replacement reactions. Spheres and cylinders are not modeled using the 1-D hydrogeochemical codes but were evaluated using nuclear criticality safety codes that treat infinite slabs, infinite cylinders, and spheres.

The following sections discuss uranium ore formation processes and aspects of the processes relevant to increasing uranium concentration in an LLW facility (4.1) and soil genesis processes (4.2) that concentrate minerals, their time scales of operation (4.2.1), and the implications of soil genesis for disposal of SNM in LLW facilities (4.2.2).

4.1 ORE DEPOSITION

A review by Kimberley (1978; Table 4.1-1) summarizes uranium deposit size and geometry. Planar ore deposits range from a few centimeters to several meters in thickness. The horizontal extent is typically kilometers. According to Nash, Granger, and Adams (1981), sandstone is the most common host rock for economic deposits of uranium in the United States (95% of U.S. resources) and worldwide (41% of world's resources). These are generally deposits formed as a result of precipitation of uranium under reducing conditions, although examples of concentrations of oxidized uranium in sorption zones have also been found. Methane and sulfur are believed to be the most common reducing agents involved in uranium precipitation (see also Raffensberger and Garvin 1995). Methanogenic reduction was treated in hydrogeochemical modeling of precipitation under chemically reducing conditions (see Sect. 10.2.1).

Table 4.1-1 Summary of ore body sizes[a]

Thickness	Extent	Name
36 cm	About 35 km	Basal Welkom, South Africa
3 cm	N/A	Dominion, South Africa
1–3 m	13 km	Elliot Lake, Ontario
>50 cm	80 km	Serra De Jacobin, Brazil
1.5 cm	Not continuous	Moeda, Brazil
Several meters	N/A	Mt. Eclipse, Australia
Several meters	11 km	Fieberbrunn, Austria
8 m	6 km	Yellirrie, Australia
5–10 m	1 km^2	Sabatini, Italy
< 15 m	N/A	Pene Blanca, Mexico

[a] Source: Kimberley, 1978.

Nash, Granger, and Adams (1981) state that since oxygenation of the earth's atmosphere 2.2 billion years ago, the genesis of uranium ore deposits has been dominated by three geochemical processes: (1) oxidation of uranium to soluble U(VI) species that are transported in aqueous solutions, commonly as uranyl-carbonate complexes; (2) reduction of U(VI) by C, S^{2-}, or Fe^{2+} species to U(IV) accompanied by precipitation of uraninite (UO_2) and coffinite ($USiO^4$); and (3) igneous and metamorphic differentiation resulting from the exclusion of uranium from the crystal structure of most rock-forming minerals. The first two processes are relevant to potential mobilization and increase in concentration of uranium in LLW disposal facilities such as the Envirocare facility. Uranyl-carbonate complexes are emphasized in this report because of their importance in ore-forming processes and as a component of "leachate" waters at the Envirocare facility.

Langmuir (1978) found that results of equilibrium calculations for uranium species in groundwater solutions typical of those in the Eocene Wind River Formation of Wyoming are dominated by the $UO_2(HPO_4)_2^{2-}$ complex in the pH range of 4 to 7.5 when even small amounts (0.1 ppm) of PO_4^{2-} are present. The same groundwater compositions are in general dominated by uranyl-carbonate complexes for pHs higher than 7.5. In the presence of cations, such as Na^+, K^+, Ca^{2+}, Mg^{2+}, Cu^{2+}, and Fe^{2+}, the stability of uranyl complexes decreases and uranyl minerals precipitate. These uranium minerals form a series of decreasing solubility: carbonates > sulfates > phosphates and arsenates > silicates > vanadates. Uranyl ions are also removed from solution by sorption or coprecipitation from dilute solutions by silicate gels, clays, zeolites, iron hydroxides, and oxyhydroxides.

Nash, Granger, and Adams (1981), in summarizing the interaction of uranium with organic materials, state that although low concentrations of uranium (<100 ppb) can be dissolved in the presence of humic and fulvic acids, higher uranium concentrations (1000 ppm) promote precipitation of uranyl humates and fulvates from neutral or weak acid solutions. Andreyev and Chumachenko (1964) note that in environments containing abundant humic material and H_2S, the organic matter will initially concentrate uranium by adsorption, and, subsequently, the uranium is reduced by the combined effects of H_2S and organic matter.

Nash, Granger, and Adams (1981) describe the genesis of sandstone-type uranium deposits that may serve as one analogue for the increase in concentration of uranium at LLW facilities. Sandstone-type deposits are hosted by sandstones created by the deposition of fine-to-medium grain sands in rivers and lakes adjacent to eroding mountains. The resulting sedimentary layers are porous and permeable. These sandstones typically contain organic matter and fossilized wood that reflect rapid burial and preservation under anaerobic conditions below the groundwater table. Important physical and chemical factors controlling the deposition of uranium from groundwater solutions in these sedimentary units are (1) permeability of the sediments, (2) the presence of adsorptive constituents such as coalified wood, humic material, and titanium oxides in the sediments, and (3) reducing agents such as carbonaceous matter and sulfur species produced by biogenic sulfate reduction or partial oxidative destruction of sulfide minerals (pyrite-marcasite). The most important ore minerals are uraninite (UO_2), coffinite ($USiO_4$), and uraniferous organic matter composed of reduced uranium, U(IV). Ore bodies are typically tabular, sheetlike, or tubular and are bounded by the original sedimentary layers of the host sandstone. In one minor variety of sandstone-type uranium ore deposits, uranium is apparently sorbed as U(VI) by iron hydroxides, zeolites, or clays.

The physical and chemical factors described previously exist in part at the Envirocare site. The waste materials in currently active cells contain dark soils (suggesting the presence of organic matter) and can contain a variety of debris (including wood). Anaerobic conditions may be imposed when the waste cell is capped by the compacted soil radon barrier and subsequent bacterial activity produces CH_4. The permeability of the waste materials is significantly less than for a porous sand because of the average grain size of waste soils and compaction that accompanies waste disposal. The existence of naturally occurring uranium ore deposits formed as a result of U(VI) sorption is an important natural analog that is evaluated with respect to the concentration of uranium at the Envirocare facility.

Another type of uranium ore deposit described by Nash, Granger, and Adams (1981) is associated with calcrete (caliche). Calcrete is gravel, sand, or desert debris cemented by porous calcium carbonate. Calcrete develops below the ground surface in response to solution transport of calcium carbonate during alternating wet and dry cycles. When fully developed, a calcrete layer may be more than 15 cm thick and is impervious to further groundwater flow. Host rocks for the calcrete uranium deposits are lenticular bodies of alluvium, soil, or detritus cemented by carbonate and other minerals. Uranium, usually as carnotite [$K_2(UO_2)_2V_2O_8 \bullet 3H_2O$], is deposited in voids and fractures in calcrete and is disseminated in underlying clay-quartz-bearing rocks. Similar uranium deposits occur in salt lakes.

In Australia, economic calcrete uranium deposits occur in Quaternary or modern playa lakes or river trunk valleys (Mann and Deutscher 1978) in areas of internal drainage where evaporation exceeds rainfall. In these environments, carnotite precipitates where groundwater flow is restricted by barriers and is caused to move upward, becoming oxidized or mixing with deeper groundwater that contains reducing agents [i.e., vanadium (IV)]. Carnotite precipitates when vanadium is oxidized to V(V) and/or mixed with a solution containing U(VI). An alternate mechanism for carnotite precipitation in calcrete is advocated by Hambleton-Jones and Toens (1978), who believe that carnotite precipitates above the water table in response to upward diffusion caused by evaporative soil suction accompanied by dissociation of uranyl complexes. The groundwater solutions are undersaturated with respect to carnotite, but metastable nucleation on montmorillonite is postulated. The discussion of carnotite precipitation is included here as an example of uranium mineralization associated with calcrete layers in natural systems. Carnotite precipitation was not modeled because vanadium is not reported as a constituent of leachate water at the Envirocare facility. However, the disposal of uranium mill tailings (or similar waste, e.g., from Formerly Utilized Sites Remedial Action Program sites) and fissile uranium materials in the same disposal cell could provide the chemical constituents required for carnotite precipitation.

The development of calcrete layers is discussed in Sect. 4.2, which describes soil genesis processes (movement of soluble components in soils, such as calcium carbonate and calcic horizons) and provides insight from studies of natural soils concerning the physical conditions and time required for generation of calcium carbonate concentrations in soils.

The Oklo uranium deposits in the Gabon Republic were the site of a natural fission reactor 2 billion years ago. Cowan (1976) summarizes the isotopic and geochemical investigations that led to the discovery of this natural fission reactor. Nash, Granger, and Adams (1981) categorize the uranium deposits at Oklo as related to unconformity-type deposits. This type of ore probably formed in many stages, spanning more than a billion years, and its genesis is complex. The long history of ore body formation and the hydrothermal processes involved preclude the Oklo deposits from consideration as a natural analog for the mobilization and increase in concentration of uranium at an LLW disposal facility. However, characteristics of the Oklo deposits and the interpreted operational history of natural fission reactors do provide important implications for nuclear criticality in an LLW facility. A descriptive scenario of a critical event(s) and the magnitude of the event at a radioactive waste facility that has been compared to nuclear criticality at Oklo is provided in Apps et al. (1983).

> Magnitude of a critical event. The attainment of a critical configuration may be expected to be a slow process resulting from a gradual accumulation of fissile material. To achieve criticality, a flow of water must be introduced into the repository. This water is necessary to move the fissile nuclides and to moderate and reflect neutrons. A system approaching criticality will produce energy at an increasing rate, and this will cause the temperature and pressure in the critical region to increase. When the pressure exceeds the hydrostatic gradient of the local formation, the influx of water will cease and the reactor will go subcritical. The maximum temperature that can be reached is the saturation temperature at the hydrostatic pressure in the repository. A temperature in excess of this would evaporate the water from the critical region.
>
> The power level limit is that which maintains these limiting temperature and pressure conditions. The critical system is thus maintained in a quasi-steady-state condition. Consequently, an explosive reaction, like that associated with the rapid accumulation of fissile material and moderator, is not possible. The event would be more like that which occurred at Oklo.

For shallow burial, such as that provided by the Envirocare waste cells, the limiting temperature and pressure conditions are those that produce saturated steam from modified rainwater entering the waste. These conditions are essentially P = 100 kPa (1 atmosphere) and T = 100°C. The Oklo reactor may have operated at P = 8000 kPa (80 atmospheres) to 100 MPa (1000 atmospheres) and T = 400 to 700°C in a high-grade ore zone estimated to contain greater than 800 m^3 of 70% uranium (Brookins, 1979).

Another factor limiting applicability of the Oklo natural reactor to disposal sites is that the estimated ^{235}U concentration is many times higher than disposal limits or practices. For example, 1.9 million g ^{235}U at Oklo is estimated to be present in 800 m^3 (or 2400 g/m^3). At the Envirocare facility, the disposal limit is 6 g/m^3 (or 0.0006 g/cm^3) and disposal practices are estimated to be on the order of 0.03 g/m^3, both well below Oklo concentrations.

Alteration of uraninite (UO_2) at the Nopal I uranium deposit in the Pena Blanca District of Mexico has been compared by Pearcy et al. (1994) to degradation of spent fuel in the proposed Yucca Mountain, Nevada, high-level nuclear waste repository. Two features of secondary mineralization at Pena Blanca, Mexico, suggested that uranium silicates should be included in the hydrogeochemical modeling conducted for this evaluation: (1) the alteration of uraninite (UO_2) having a low trace element content in an oxidizing environment above the water

table and (2) the presence of uranium silicate phases as secondary minerals. Soddyite [$(UO_2)_2SiO_4 2H_2O$] was used as a model for uranium silicates.

4.2 SOIL ALTERATION

The following discussion of soil-forming processes is provided to introduce the reader to relevant chemical and physical processes and the probable periods of time required to modify the chemistry and physical configuration of engineered soil caps and underlying waste at the Envirocare facility. The soil-forming or modifying processes that can be anticipated to take place in a waste cell include those resulting from biocolonization, physical changes that control the influx of water, and chemical changes that affect the transport of uranium.

Soil-forming processes operate at highly variable rates that are largely dependent on interactions of soil parent material with climate. The soil profile is the product of a net integration of various processes during hundreds to many thousands of years. Five basic factors control development of a soil and the associated chemical, physical, and mineralogical properties: parent material, relief, climate, biology, and time (Wilding, Smeck, and Hall, 1983a and 1983b).

4.2.1 Development of Chemical Horizons

The development of chemical horizons in subsoils may be analogous to processes of uranium mobilization and immobilization in waste cells.

A way to visualize soil genesis is to consider soil, at a given location in the landscape, as a chromatographic column with rainfall as the fluid medium. At time = 0, the column is filled with parent material, which may be quite uniform in composition or very heterogeneous or even stratified. In a humid environment where water is available for leaching, visible changes occur that can be observed within less than 50 years. One of the earliest changes is the addition of organic matter to the soil surface and to the upper soil via root mass. This produces the genetic “A” horizon, which is defined as the surficial soil that has been darkened by organic matter. As time proceeds in a leaching system, soluble components or very fine clay particles are translocated or deleted from the soil column. In the humid parts of the United States, which have net leaching, a clay-enriched subsoil “B” horizon forms within 1800 to 2000 years. As time continues, and without climate changes that alter the basic soil-forming process, soil horizons become more visually evident and thicker. Subsoil horizons move downward into the column as time increases, as long as surface geomorphic stability exists or until a steady state is reached. At depth, saturated water flow becomes channelized along preferred pathways. This movement of water causes increased chemical weathering and the subsequent translocation of soluble components deep into the soil; while in those areas without water movement or with unsaturated water flow, little transport occurs (Wilson et al., 1991; Jardine, Jacobs, and Wilson, 1988 and 1990; Luxmoore et al., 1990).

Gile, Peterson, and Grossman (1966) started an intensive study of how carbonate moves in soils and how long it takes for carbonate to be either removed or translocated to certain depths to form carbonate-enhanced genetic soil horizons. These authors identified several stages that describe the movement and subsoil accumulation of calcium carbonate:

Stage 1: Scattered grain coatings in gravely soils and filaments in fine-grained soils

Stage 2: Carbonate nodules separated by lower carbonate content soil

Stage 3: Subsoil horizon of accumulation plugged throughout

Stage 4: An indurated laminar horizon formed above the plugged horizon

While working on the Desert Project located near Las Cruces, New Mexico, Gile, Hawley, and Grossman (1970) were able to correlate the stages of subsoil carbonate accumulations with dateable geomorphic surfaces. The following general time frame has been established on the basis of the Desert Project:

Stage 1: <1000 to about 5000 years

Stage 2: 5000 to 15,000 years

Stage 3: >15,000 years

Stage 4: >100,000 years

Gile (1995) estimated that the average rate of carbonate accumulation was about 5.1 kg/m^2 per 1000 years.

The formation of clay-enriched subsoil horizons has been studied for many years. While studying soils in Pennsylvania, Bilzi and Ciolkosz (1977) found that between 2000 and 3000 years were required on a stable geomorphic surface and in noncalcareous parent materials for processes of soil genesis to translocate enough clay particles from surface soil horizons to form a detectable subsoil clay-enriched horizon. Cremeens (1995) reported the results of a study of soil genesis on a 2100-year-old Native American mound in southwest West Virginia. On the geomorphically stable mound crest, soil genesis had produced horizons to a depth of 0.75 to 1.0 m. He found that very little clay movement took place during 2100 years. Cremeens also documented the presence of redoximorphic features in the mound, including both areas depleted in iron-manganese and areas of concentration increase. He also found iron-manganese nodules, but these may have been inherited from the older alluvium used to construct the mound.

4.2.2 Implications of Soil Alteration in LLW Disposal Facilities

Factors that may accelerate soil genesis processes include the following.

1. Rainfall will eventually infiltrate into a compacted surface cap, resulting in subsurface transport and initiating soil genesis/modification processes. LLW mound sideslopes are subject to accelerated rates of erosion. Impervious caps on LLW mounds eventually crack or otherwise become breached. Impervious caps greatly increase the rate of erosion, especially on lower sideslopes, because all rainfall must flow off.

2. LLW must be compacted to the same extent as the final impervious cap. If not, differential settlement of the waste will occur as well as eventual settlement of the cap, resulting in cracks forming or in the ponding of water, reducing the time before water penetrates.

3. Rates of infiltration and alteration may be slow, on the order of thousands of years.

4.3 SUMMARY

The evaluation of natural mineral concentration processes (ore deposit formation and soil genesis) places constraints on the shape and size of zones of increased uranium concentration in LLW facilities and on the time scale of the chemical and physical concentrating processes. Processes that form ore-deposit grade concentrations of uranium are dominated by precipitation reactions, but chemical sorption is also a plausible mechanism for increasing concentrations.

Under present climatic conditions, the high evaporation-transportation rates relative to rainfall accumulation at the Utah site will limit the depth of carbonate accumulation to less than 1 m below the top of the waste cell (Jenny and Leonard, 1934). Therefore, in the event that uranium and calcium carbonate accumulate in sufficient quantity to form a subsoil horizon greater than 15 cm thick (stage 3 subsoil carbonate accumulation, >15000 years), the top 1 m of waste soil fill will have an insufficient uranium inventory to produce a critical mass of ^{235}U. In this same environment, spherical nodules greater than a few centimeters in diameter are unlikely to form during accumulation of carbonates in the top 1 m of the waste cell. Subsequent analysis will show that this dimension is too small for spherical shapes to form a criticality safety concern. The infinite cylinder might be postulated to form by chemical reduction and replacement of organic material such as wood. The state of Utah license for Envirocare limits the size of debris to less than 25 cm in at least one dimension and no longer than 2.4 m in any dimension. This limit will minimize the potential for uranium accumulation by chemical replacement of carbonaceous material because subsequent analysis will show at least 20.8 cm diam or greater are needed for criticality safety concern.

The evaluation of ore deposits and mineral-concentrating processes operating during soil genesis provides insight concerning the general processes and mechanisms that are operating to produce mineral concentrations. However, each ore deposit or mineral concentration has site-specific characteristics that require a generic set of analytical tools to be applied to understand the details of its genesis. In the same manner, a set of generic tools must be applied to evaluate the migration and increase in concentration of SNM in a waste disposal cell.

5 SCENARIO DEVELOPMENT

SNM in contaminated soils that are buried and not contained or protected from meteorological, chemical, and geological influences could migrate and subsequently concentrate at sites of natural opportunity (adsorption zones, reducing zones, or zones of low permeability that result in a concentration of perched water) within the buried media. Though such migration and increase in concentration can occur laterally, the gravity-driven flow of moisture and chemicals through the buried media is expected to be vertically downward under conditions of transient saturation. For some circumstances where disposal of permitted concentrations of SNM occurs by burial, potentially critical areal densities of SNM may be predicted if the mass of SNM in the vertical profile of the contaminated soil migrates to the floor of waste cells or other natural heterogeneous features. The redistributed SNM could concentrate into a spherelike region because of some chemically extractant material or a void region permitting the influx of solutions containing SNM. Another assumption could be that the location of concentrated material along the intersection of a burial-cell wall with the burial-cell floor forms a cylinderlike region. The last assumed simple geometry results from the downward migration and increase in concentration of SNM into a generally horizontal slab. The horizontal slab geometry was produced by 1-D hydrogeochemical modeling of vertical flow in a column. Spherical and cylindrical geometry cannot be formed using a 1-D hydrogeochemical model.

Material can be concentrated into a semirandom geometry that is dependent upon the waste matrix geometry. However, information concerning the details of geochemistry and geometry of the waste matrix typically will not be available. Therefore, reasonable parameters must be determined that may lead to increases in concentration, given existing license conditions, and bounding models must be approximated for performing nuclear criticality calculations.

Hydrogeochemical modeling and criticality safety assessment modeling were conducted over a range of parameters to evaluate the likelihood of each of the outcomes given in Sect. 1.5. The parameters considered in these analyses reflect the range of possible parameter values for hydrologic and geochemical conditions at the LLW disposal sites where soils are nonnative and for which little geochemical or hydrologic data are available.

Hydrogeochemical modeling considered two events. First, the uranium present in the soil must be mobilized. Second, the uranium moves to a new location where it is immobilized and concentrated. These processes were modeled in a 10-m-long simulated column (Fig. 1.5-2). Column length was determined from probable waste cell construction.

- The study assumed vertical infiltration of water through a 10-m-deep cell of uranium-contaminated soil.

- The uranium was considered to be initially distributed uniformly throughout the soil profile; both adsorption and precipitation were considered to be the mechanisms for the initial association of uranium with the soil matrix.

- Mobilization of sorbed uranium was modeled as a (slug) volume of water (rain events) entering through the cell cap. The range of aqueous chemistry (pH, ionic strength, and composition) of the water entering the top of the simulated column was selected on the basis of the assumption that rainwater infiltrating through the 2-m-thick cap over the cells reached chemical equilibrium with the minerals in the cap before entering the soil column. The validity of that assumption was tested by comparing calculated equilibrium concentrations to an analysis of the chemistry of leachate at the Envirocare site.

- The simulated column contained one or several zones of altered chemistry that could serve as a zone for an increase in uranium concentration, including

  - a single zone at the bottom of the column with a greater abundance of adsorption sites than the rest of the soil column;
  - multiple zones of higher sorption distributed at different levels within the column; or
  - a reducing zone at the bottom of the column.

The zonation modeled is not based on observed site conditions but is considered an evaluation of configurations of maximum uranium concentration. A zone of higher sorption at the base of a cell could be the result of material used as a soil. Thickness and sorption capacity of liners can be determined for specific sites. Multiple zones of sorption could be the result of variable chemical and physical properties of waste disposed of in different lifts (layers) within the waste cell. Formation of redox zones is known to occur in aquifers, but there have been few studies of landfill zonation. Reducing zones are typically associated with degradation of organic waste (Christensen et al., 1994; Baedecker, Cozzarelli, and Eganhouse, 1993; Lyngkilde and Christensen, 1992; and Baccini, 1989), which is unlikely to be a significant factor in LLW facilities. These zones can be on the order of meters thick. Redox gradients of 40 mV/m have been observed in groundwater (Barcelona et al., 1989).

Chemical changes and parameter values considered in these simulations are discussed in more detail in subsequent sections.

6 MOBILIZATION AND IMMOBILIZATION MECHANISMS

6.1 MOBILIZATION MECHANISMS

Mobilization of uranium can occur as a result of ligand complexation, and this can be enhanced under conditions of subneutral pH and high Eh. Complexation by CO_3^{2-} will be considered in detail and can be considered as a surrogate for other complexing agents such as organic ligands, although different concentrations of organic ligands may be required to bring about a similar mobilizing effect. Like carbonate, organic ligands are postulated to increase the solubility of uranium minerals and enhance the transport of uranium (because of mobile complexed uranium cations). For example, natural organic matter (NOM) has been shown to complex actinides and promote their transport in groundwater downgradient of transuranic disposal cells at ORNL in Oak Ridge, Tennessee (McCarthy, Marsh, and Tipping, 1995). At the same site, lanthanides—which have a chemistry similar to many actinides— injected in a groundwater tracer experiment were complexed with NOM and moved rapidly downgradient at rates similar to those of nonreactive tracers (Knowles et al., 1995). Anthropogenic organic complexants disposed in shallow soil trenches at ORNL also mobilized cobalt and uranium, and the uranium-EDTA complex was recovered in monitoring wells outside the disposal cell (Means, Crerar, and Duguid, 1978). The assumption that organically complexed uranium will be transported in a manner similar to carbonate complexes is a worst-case scenario. The NOM-uranium complex can possibly be retarded as a result of adsorption of NOM on the soil particles, thus reducing the extent of uranium migration (McCarthy, Marsh, and Tipping, 1995).

The eluant in the model column was based on leachate chemistry at disposal sites and at other landfills where more detailed waste chemistry is available. The site data were obtained from reports on groundwater sampling or from equilibration of water with the matrix. The production of CO_2, caused by microbial activity would be important in potential uranium complexation and was estimated from other sites. A range of concentrations was tested because of uncertainty in parameter estimates and model sensitivity. For example, knowing if a particular range of pH and ionic strengths were uniquely effective in uranium mobilization would be useful.

Colloids have also been considered, at least conceptually, as a potential transport mechanism. However, a preliminary analysis indicated that colloid transport would not be a significant mobilization mechanism, especially under oxidizing conditions. Colloid transport is important only for highly insoluble or strongly adsorbing species whose transport as a dissolved species is limited. Because of the high solubility of the carbonate complex of uranium, colloids would not contribute significantly to transport under oxidizing conditions. Colloids could, however, contribute to some vertical spreading of uranium under reducing conditions. Furthermore, colloids may play a much more significant role for other SNM such as plutonium, whose aqueous solubility may limit transport of the dissolved species.

6.2 IMMOBILIZATION MECHANISMS

Immobilization of uranium in zones of altered chemistry or porosity can occur through a variety of mechanisms, including adsorption, precipitation, filtration, and evaporation, and it can occur with and without changes in aqueous chemistry of the advecting fluid (pH, Eh). Although fundamental understanding of transport processes suggests that these are reasonable mechanisms, the quantitative effect of these processes on the increase in concentration of uranium cannot be predicted a priori. The zones of altered chemistry were modeled as zones within the column that have a different sorption capacity or as reducing zones. The modeling was designed with adsorption as the primary potential mechanism for increasing the concentration under oxidizing conditions

(although the significance of precipitation under oxidizing conditions was considered). Precipitation was emphasized as the potential mechanism for increasing concentration under reducing conditions.

Filtration and evaporation mechanisms were not formally modeled. The potential importance of these processes is addressed in the discussion of natural analogs of increasing uranium concentration (Sect. 4).

7 ANALYTICAL APPROACHES

Because nuclear criticality can exist over a broad range of ^{235}U-contaminated soil parameters and because a broad range of hydrogeochemical processes can affect the concentration of ^{235}U, both analytical approaches required the development of parametric surface responses to examine intersections of these surfaces that could mutually define parameter space that can potentially lead to nuclear criticality. In addition, other ^{235}U concentrations were evaluated to span less likely, yet credible, concentrations independent of the hydrogeochemical values.

7.1 HYDROGEOCHEMICAL MODELING

7.1.1 Reactive Transport Modeling

Uranium geochemistry is complex and requires a sophisticated modeling approach that considers simultaneous and interacting processes of complexation, sorption, and precipitation occurring under conditions of pH, Eh, ionic strength, and composition that vary over time and space (Langmuir, 1978; Toran, 1994). An increase in uranium concentration is postulated to result from sequential processes of mobilization of uranium by formation of soluble complexes, followed by immobilization of the soluble species by processes of adsorption and precipitation. Geochemical modeling can calculate the distribution of soluble, adsorbed, and precipitated uranium species under a given aqueous chemistry and mineral composition, but this type of modeling will not account for how the uranium moves and interacts with chemicals along a flow path; transport modeling is required to account for those processes. However, most transport codes do not account for detailed geochemical reactions but simply lump multiple geochemical processes into a term called the retardation factor. Because the retardation factor only slows transport and does not consider mobilization mechanisms or variable chemistry along a flow path, concentrations greater than initial conditions in solution cannot be obtained by using such a simplified approach.

Hence, a multispecies transport code was used to examine the speciation and transport behavior of uranium. A multispecies transport code models groundwater flow, chemical dispersion, and a suite of selected geochemical reactions. Reactive transport modeling has not yet reached the application stage in groundwater modeling. Most of the available codes are considered research codes (van der Heijde and Elnawaway, 1993; National Research Council, 1992; Mangold and Tsang, 1991), which are primarily used by the developers (or someone working closely with developers). Only a handful of examples exist of the use of these codes on problems based on real sites. Furthermore, reactive transport modeling is computationally intensive (Yeh and Tripathi, 1991a) because a full suite of geochemical calculations must be conducted at each node in the problem domain.

A parallelized version of a reactive transport code was used with the assistance of the code developers in setting up problems. PARSim (Arbogast, Dawson, and Wheeler, 1994) was developed by researchers from the Computational Mathematics Department at Rice University (the group has recently moved to the University of Texas at Austin). PARSim was developed as an efficient flow and transport model designed to run on a supercomputer configured with a parallel processor architecture to speed simulations having repetitive calculations. A chemical reaction code, KEMOD, (Yeh et al., 1995) based on MINTEQ was coupled to PARSim to create a multispecies reactive transport code. The user defines which chemicals, complexes, and solids are to be considered by the model. For the flow and transport code, the domain is divided into a finite element mesh.

At every node in the mesh, the full set of selected geochemical reactions are modeled. All parallel runs were conducted on a 66-processor Intel Paragon supercomputer at ORNL. Run times were typically 1 to 2 hours.

Results from PARSim have been compared to results from other multispecies transport codes (HYDROGEOCHEM, Yeh and Tripathi, 1991b; DYNAMIX, Liu and Narasimhan, 1989) to test its reliability. Analytical solutions do not exist for this type of problem; therefore, the only way to test a code is to compare its results with those produced by other codes. PARSim matched trends well and matched absolute concentrations in most cases. Some discrepancies occurred for processes that were not used in this modeling.

In addition to PARSim, geochemical modeling without transport was performed using the U.S. Geological Survey code PHREEQE (Parkhurst, Thorstenson, and Plummer, 1982), which is a well-established code for modeling chemical speciation and reactions. This modeling was used to establish initial conditions and identify important complexes and appropriate phases for the precipitation. Stability diagrams (as discussed in Sect. 7.1.2) were also used to select geochemical phases to model. PHREEQE was especially important for consideration of redox reactions, where it was used as the primary tool for chemical reactions. Redox reactions occur over such a wide range of concentrations that the reactive flow and transport models frequently become unstable. PHREEQE could thus be used more efficiently than a full multispecies transport code. Although this model does not include transport or mixing caused by hydrodynamic dispersion, the key features of the scenario are adequately captured by this approach, and any error introduced by this simplification is small relative to other model uncertainties.

In summary, PARSim was used to model mobilization and transport of uranium as well as immobilization under oxidized conditions. PARSim was again used for mobilization studies under initially oxidized conditions, but PHREEQE was used to determine reactions in the reduced zones.

7.1.2 Solubility/Phase Definition

The study used phase diagrams and geochemical modeling with PHREEQE to select key components and minerals in the model (Table 7.1-1). Results of this modeling and other modeling indicates only certain species dominate. A variety of possible species was included to cover changing geochemical conditions, but some limitations had to be imposed on the number of species to prevent the model convergence time from becoming prohibitive.

Dominant uranium complexes of hydroxyl and carbonate have been identified by Langmuir (1978) and Tripathi (1983). The dominant uranyl-phosphate complex at circum-neutral pH has been identified by Tripathi (1983), Nash, Granger, and Adams, 1981, and Lee, Elless, and Hoffman, 1993, as $UO_2(HPO_4)_2^{2-}$. Several solid phases were considered in modeling, as suggested by stability diagrams. The oxidized uranium minerals modeled included rutherfordine (UO_2CO_3), and for selected runs, soddyite [$(UO_2)_2SiO_42H_2O$] and U-hydroxide [$UO_2(OH)_2$-beta]. Reliable thermodynamic data could not be obtained for some uranium minerals {e.g., uranophane [$Ca(H_2O)_2(UO_2)_2\ (SiO_2)_2(OH)_6$]}. For the reduced mineral phase, uraninite (UO_2) was the selected phase (Fig. 7.1-1), although others were considered in preliminary calculations. Uraninite is a frequently cited mineral in ore zones and precipitates readily under modeled conditions.

For other complexes, standard carbonate and hydroxyl species were considered. Calcite ($CaCO_3$) and hydroxyapatite [$Ca_5OH(PO_4)_3$] were selected as solubility controls for CO_3^{2-} and PO_4^{2-}, respectively.

Table 7. 1-1 Chemical species considered in sensitivity analysis[a]

Species	log K	Stoichiometry					
		Component	Value	Component	Value	Component	Value
H^+	0	H					
CO_3^{-2}	0	CO_3					
Ca^{+2}	0	Ca					
UO_2^{+2}	0	UO_2					
$Si(OH)_4$	0	$Si(OH)_4$					
HPO_4^{-2}	0	HPO_4					
SOH	0	SO-H					
OH^-	−14	H					
$CaCO_3(aq)$	3.22	Ca	1	CO_3	1		
$CaHCO_3^+$	11.43	Ca	1	CO_3	1	H	1
$CaHPO_4(aq)$	2.74	Ca	1	P	1		
$CaOH^+$	−12.85	Ca	1	H	−1		
UO_2OH^+	−5.3	UO_2	1	H	−1		
$(UO_2)_2(OH)_2^{+2}$	−5.68	UO_2	2	H	−2		
$(UO_2)_3(OH)_4^{+2}$	−11.88	UO_2	3	H	−4		
$(UO_2)_3(OH)_5^+$	−15.82	UO_2	3	H	−5		
$(UO_2)_4(OH)_7^+$	−21.9	UO_2	4	H	−7		
$(UO_2)_3(OH)_7^-$	−28.34	UO_2	3	H	−7		
$UO_2CO_3(aq)$	9.65	UO_2	1	CO_3	1		
$UO_2(CO_3)_2^{-2}$	17.08	UO_2	1	CO_3	2		
$UO_2(CO_3)_3^{-4}$	21.7	UO_2	1	CO_3	3		
$(UO_2)_2CO_3(OH)_3^-$	−1.18	UO_2	2	CO_3	1	H	−3
$UO_2(HPO_4)_2^{-2}$	18.56	UO_2	1	HPO_4	2		
HCO_3^-	10.32	H	1	CO_3	1		
$H_2CO_3(aq)$	16.67	H	2	CO_3	1		
H_3PO_4	9.35	H	2	HPO_4	1		
SO^-	−10.3	SO-H	1	H	−1		
SOH_2^+	5.4	SO-H	1	H	1		
$SO\text{-}UO_2OH^+$	−7.1	SO-H	1	UO_2	1	H	−2
$SOH_2^+(UO_2)_3(OH)_7^-$	−31	SO-H	1	UO_2	3	H	−8
Rutherfordine (UO_2CO_3)	14.4	CO_3	1	UO_2	1		
Calcite ($CaCO_3$)	8.48	Ca	1	CO_3	1		

[a] Source: Yeh and Tripathi, 1991b.

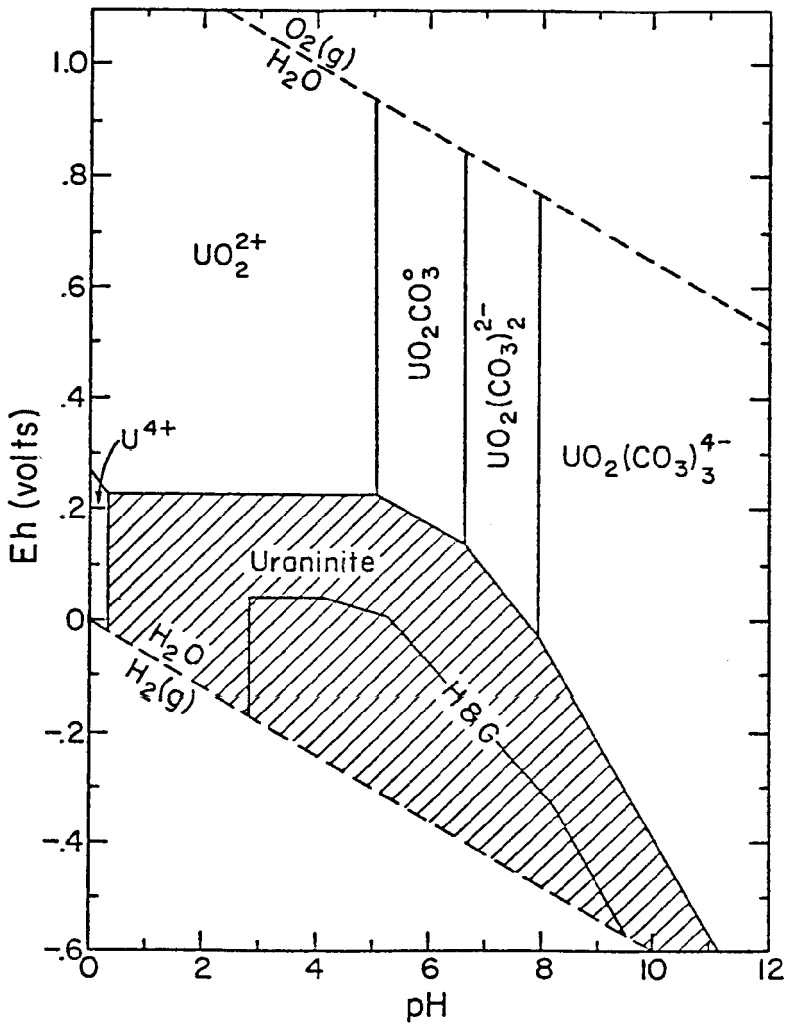


Figure 7.1-1 Eh-pH diagram for the U-O_2-CO_2-H_2O system at 25°C for $P_{CO2} = 10^{-2}$ atm. Uraninite, UO_2(c), solution boundaries are drawn at 10^{-6} *M* (0.24 ppm) dissolved uranium species. (After: Langmuir, 1978)

7.1.3 Sensitivity Analysis

A sensitivity analysis was needed for this model because a large number of the parameters (e.g., concentrations of carbonate and uranium and abundance of sorption sites) could vary over a range of values. This uncertainty created a large number of conditions that needed to be considered to adequately evaluate the potential for increasing uranium concentration. The sensitivity analysis addresses the question of which parameters create the most variation in model output. A common method of conducting a sensitivity analysis is to change one parameter at a time while holding other parameters constant. This method may neglect parameter sensitivity if the constant parameters fix the system in a particular range.

To avoid creating gaps in the sensitivity analysis, all uncertain parameters were varied systematically to better analyze sensitivity to parameter ranges. A Latin Hypercube sampling selected parameter values for six uncertain parameters (see Sect. 9.1 on parameters for more details). The Latin Hypercube sampling maximizes the relationship between parameters because no value is selected more than once, but it also minimizes the total number of runs required (McKay, Beckman, and Conover, 1979). The number of runs should be at least five times the number of parameters tested. The Monte-Carlo-like set of runs that were created contained 40 simulations for 6 parameters. The results are analyzed by calculating the multivariate regression between parameters and a selected output variable (such as uranium concentration). A rank regression, which is more robust than a linear regression (Iman and Conover, 1979), was used.

7.2 NUCLEAR CRITICALITY EVALUATION

7.2.1 Code Description and Validation

The SCALE (1995) code system was used to calculate the k_{eff} of the designated systems. SCALE is a modular system of codes that provides criticality safety analysis sequences (CSAS) to calculate the neutron-multiplication factor of a system. Problem-dependent processing of the cross sections to account for temperature effects and resonance self-shielding are performed using the NITAWL and BONAMI codes. For this study, the XSDRNPM code was executed by the CSAS module to provide the k_{eff} values used in this study. XSDRNPM is a deterministic code that solves the Boltzmann equation for neutron transport in a 1-D system using a discrete-ordinates approach. SCALE was used because of its historic and recognized success in the performance of benchmark and applications analyses for licensing activities.

The stationary system of the SCALE codes used for this study and validation, CSAS, BONAMI, NITAWL, XSDRNPM, and KENO V.a, were created on May 30, 1995. The Brookhaven Evaluated Nuclear Data File B Version V point cross-section library, which was collapsed to a 238 neutron-energy group library (Greene et al., 1994), named REF01.XN238, was created on May 26, 1995, and resided on the same hardware platform as the SCALE suite of codes during the period of this study. The 238-energy group library was used because of its currency of evaluation, testing, and benchmarking. The hardware platform, the SCALE computational codes, and the 238-energy group library used were validated through the computation of verification and validation benchmarks involving ^{235}U systems before and after the evaluations performed for this study. The verification and validation benchmark calculations provided identical results for calculations performed both before and after the study, thereby demonstrating the stability of the software and data throughout the study. The bias and uncertainties of the benchmark calculations were within ~0.5% of the experimental values; that is, the calculated

k_{eff} of the 14 critical experiment benchmarks were between 0.9954 and 1.0064. Results of the calculations are provided in Table 7.2-1. Note that the k_{eff} for some of the cases in Table 7.2-1 were obtained using the KENO V.a code, a code that uses the Monte Carlo approach to solve the Boltzmann transport equation for multidimensional systems.

Table 7.2-1 Computational benchmark results

XSDRNPM result filename	**k_{eff}**
17CSB.OUTPUT [a]	1.00350
18CSB.OUTPUT [a]	1.00639
19CSB.OUTPUT [a]	1.00316
110CSB.OUTPUT [a]	1.00453
111CSB.OUTPUT [a]	1.00129
ORNL1.OUTPUT [a]	0.998680
ORNL2.OUTPUT [a]	0.998468
ORNL3.OUTPUT [a]	0.995463
ORNL4.OUTPUT [a]	0.996905
ORNL10.OUTPUT [a]	0.997975
KENO V.a result filename	**k_{eff} ± sigma**
OR260901.OUTPUT [b]	1.0059 ± 0.0014
OR260906.OUTPUT [b]	1.0041 ± 0.0012
ORFP2710R.OUTPUT [c]	1.0025 ± 0.0021
RFP2710U.OUTPUT [c]	1.0047 ± 0.0023

[a] See Brookhaven National Laboratory, 1974.
[b] See J. K. Fox, 1958.
[c] See R. E. Rothe, 1978.

7.2.2 Analytical Approach

The analytical approach used for the nuclear criticality evaluation was performed in two segments. The first segment was to evaluate the infinite-media multiplication constant, k_∞, of two fixed-density soil matrices having differing degrees of ^{235}U and water contents or densities within the soils. These results provided indications of the combinations of ^{235}U, soil, and water that could support self-sustaining nuclear fission chain reactions in an

essentially infinite sea of material (i.e., $k_\infty \geq 1.0$). The second segment involved examining three geometries that have relevance to the evaluation: spheres, cylinders of infinite length, and slabs of infinite extent. Conceptual configurations of the geometries are provided in Fig. A.1-1 of Appendix A. The evaluations of the infinite slabs approximate the effects of the ^{235}U, contaminating the soillike waste and settling vertically onto a waste-cell floor, and are consistent with previous evaluations (Hopper et al., 1995) performed for reviewing LLW facilities.

The two waste matrices assumed for the evaluations were nominal soil and SiO_2. The nominal soil was an approximation of soils reported by Sposito (1989) and Shacklette and Boerngen (1984). Before contamination with water and ^{235}U, both waste matrices were assumed to have fixed densities of 1.6 g/cm^3, thereby leaving void space for variable densities of ^{235}U contaminant and water. For the nominal soil (N-S) cases, seven ^{235}U densities (i.e., grams of ^{235}U/cm^3 of soil) or concentrations (i.e., grams of ^{235}U/g soil) were evaluated. Six different densities (i.e., g H_2O/cm^3 of soil) or concentrations (i.e., g H_2O/g soil) of water were assumed for each of the seven ^{235}U concentrations. For the SiO_2 soil (S-S) cases, 18 ^{235}U densities or concentrations were evaluated. Seven different water densities or concentrations were assumed for each of 7 ^{235}U densities or concentrations, and 13 different water densities or concentrations were assumed for each of 11 ^{235}U densities or concentrations.

7.3 CRITICALITY CONSEQUENCES: DIRECT DOSE FROM RADIATION TRANSPORT

7.3.1 Calculational Models

Direct-dose radiation transport calculations were performed to estimate the neutron and gamma radiation doses near the soil surface as a consequence of two postulated critical configurations at the Envirocare facility—one at near-minimum ^{235}U critical density and the other at the near-maximum ^{235}U density. Both were assumed to be large-diameter (i.e., 18 m), disklike cylindrical deposits of overly water-moderated uranium in SiO_2. Both configurations were selected so that they would be subcritical when void of the water moderation. Both configurations were vertically centered in a trench. Both configurations were constrained by the maximum allowable 5.7 kg of ^{235}U per m^2 areal density in a disposal trench. The first postulated critical configuration was an 18-m-diam disk that was 2.5 m in thickness, which corresponds to a CF of about 3.6. The second postulated critical configuration was an 18-m-diam disk that was ~0.9 m in thickness, which corresponds to a CF of about 10.5. The calculations were performed using the DORT code (Rhoades and Childs, 1988), which solves the 2-D discrete-ordinates radiation transport equations and the SCALE 27-neutron/18-gamma group cross-section library. The code was utilized in a two-step procedure for computational efficiency. The first step solved an effectively 1-D criticality problem to obtain the magnitude and energy distribution of the neutrons and gamma rays leaking through the ground above the deposit. The second step solved the 2-D air-over-ground problem with the leakage source (a tabulation of the neutrons/gamma rays leaking from the ground) from the first step. This second step determines the population of neutrons/gamma rays both in the ground and at all modeled locations above ground. The specific doses were then read from the code output at locations corresponding to 1 m above the ground surface, directly over the center of the assumed deposit and 90 m away from the vertical centerline of the assumed deposit.

7.3.2 Estimate of Potential Fission Yields

The actual number of fissions that occur (the so-called fission yield) is a difficult quantity to determine. The approach taken in this study was to assume that the concentration of fissile material occurs under wet conditions, followed by a dryout period in which the critical conditions are met, with event termination due to the evaporation of the full inventory of water. Using a widely accepted rule of thumb that 10^{17} fissions are required to remove 1 L water (G. Tuck, 1974), the amounts of water present under the various cases were used to estimate postulated fission yields. The resulting fission yield estimates were 4.1×10^{21} and 2.8×10^{21} fissions for the high and low concentrations, respectively.

8 ASSUMPTIONS

The assumptions implicit in the approach to the problem are specified as follows. All models are simplifications of reality, and the processes not explicitly modeled in the simulations must be evaluated. Beneath each assumption the significant implications of the assumptions to the realism and general applicability of the results are identified in italics.

The flow-field was 1-D (simple in-one-end and out-the-other hydraulics).

One-dimensional flow neglects tortuous paths and reduces travel times. Concentration of uranium into a slab was the only configuration tested with the flow-field. One-dimensional flow requires vertical recharge, which may be limited in arid environments. Furthermore, uranium can be flushed out of the bottom of the column, reducing the mass available for concentration within the column. A corollary assumption is that any uranium leaving the engineered barriers of the disposal cell will disperse or be concentrated by mechanisms similar to those within the disposal cell. This assumption may require further examination, and the modeling here did not consider transport outside the disposal cell.

For the sensitivity analysis, uranium was disposed of at the maximum allowable concentration of ^{235}U.

Lower disposal concentrations (such as those reported at the Utah site) require greater CFs to reach levels of concern. This relationship is essentially linear. The worst-case scenario was modeled with some additional runs at values reported from the Utah site.

The hydraulic conductivity was uniform (which resulted in assumed values for velocity, under saturated and unsaturated conditions).

Velocities had little effect on peak concentrations and were not the most important control; the possible range in travel times for variations in saturation are so extreme as to make heterogeneity an insignificant factor. That is, uncertainty in saturation conditions is the most important unknown variable in estimating travel times.

Unsaturated travel times can be estimated from consideration of saturated travel times.

This assumption creates one of the largest sources of uncertainty. Travel times increase greatly in unsaturated conditions, but how much to increase them is unknown because of uncertainty in unsaturated conditions.

Unsaturated concentrations can be approximated by a saturated model.

Some errors in the prediction of uranium concentration will occur because of different hydrodynamic dispersion and transient effects, but these are expected to be small compared to errors resulting from the uncertainty in saturation conditions.

Steady-state conditions were attained.

By limiting the model to a specified number of time steps, an early or late peak of uranium may have been missed. After evaluating this factor, only limited cases were found that require shorter or longer run times.

A fixed, stable reducing zone is assumed.

In reality, redox zones can change as a result of infiltrating water. Although sustained reducing conditions can occur, this was not evaluated, and thus the worst-case scenario was modeled.

Assumptions

Selection of the solid phases (e.g., minerals and sorption surfaces) introduces uncertainty.

Other mobilization and demobilization agents should be modeled to evaluate the importance of alternate scenarios. The modeling here provides a general approach.

A limited number of complexes were considered.

The most influential complexes were considered, and significant error is not anticipated. Mobility would be greater than modeled as a result of neglecting important complexes and competition for sorption sites. Mobility would be overpredicted if precipitation or sorption were neglected.

Chemical equilibrium conditions were attained during transport.

Consideration of kinetic factors could change estimates of travel time. However, the time frame modeled is fairly long, so equilibrium is likely to be approached. Little or no kinetic data are available to evaluate this factor.

The EQ3/EQ6 and PHREEQE databases were used for most equilibrium constants and provided self-consistent, appropriate datasets.

Well-known databases were used when possible, and no major errors are expected to occur. Again, increases or decreases in mobility could occur depending on the magnitude and direction of the errors in the databases. Uranophane $[Ca(H_2O)_2(UO_2)_2\ (SiO_2)_2(OH)_6]$ *was not used in calculations because of a reported error in its equilibrium constant.*

The soil matrix (at a bulk density of 1.6 g/cm^3) with varying degrees of ^{235}U contamination and varying degrees of water content was chosen as a realistic bounding composition for the purpose of this scoping study. Discrete values were selected for evaluation.

The primary influence affecting the nuclear criticality evaluation is the density of the ^{235}U contaminant in the soil. There are two secondary influences affecting the nuclear criticality evaluation; the water content within the soil and the composition of the soil. The water content and ^{235}U were chosen arbitrarily to span the parameter ranges of interest to this scoping study. A third secondary influence affecting the nuclear criticality evaluation is the enrichment of the ^{235}U. There are, however, no licensed constraints on uranium enrichment at the Envirocare facility. Therefore, 100 wt % ^{235}U was assumed for these evaluations. Lesser enrichments require greater concentrations of uranium (to increase ^{235}U concentrations to compensate for ^{238}U neutron absorption) and more restrictive water concentrations to permit nuclear criticality. The discrete values chosen do not provide all the necessary intermediate parametric values to evaluate nuclear criticality. That is, the transition from subcritical infinite soil matrices to realistically dimensioned disposal/burial sites having burial depths greater than about 4 m, and ^{235}U CFs less than 3 have not been characterized in detail. Additionally, soil and waste matrix conditions can be substantially different, chemically and neutronically; therefore, this study has limited or no applicability to other waste matrices involving bulk quantities of polyethylene, carbon/graphite, beryllium, or heavy hydrogen compounds.

Simplistic deposit geometries, having no density gradients, were used in the criticality assessment.

Smaller quantities of fissile material in equivalent volumes may be required to reach criticality for certain density gradients. An extreme, but actual, critical experiment performed by Morfitt (1953), was the assembly of five concentric cylindrical uranyl fluoride solution regions having variable densities of 93 wt % enriched uranium. Solution uranium densities were selected to produce a nearly uniform thermal neutron core flux. Doing so produced a critical system with 1061 g ^{235}U as compared to a homogeneous core mass of 1162 g ^{235}U in an equal volume.

The radiation transport models that were used for the criticality consequence evaluation assumed that the concentrated uranium deposits were vertically centered at the midplane of the disposal trenchs and were shaped like cylindrical disks having vertical axes.

More conservative models (deposit at surface of disposal trench) or less conservative (deposit at floor of disposal trench) could be assumed regarding radiation dose determination at the surface of the disposal trench. Justifying such models would require extended periods of study. The midplane location was selected as a compromise to issues regarding uranium density gradients and unlikely alternative uranium transport/concentration mechanisms.

The two uranium deposit concentrations (minimum and maximum) selected for the criticality consequence evaluation were those that were determined to be practical by the hydrogeochemical scenarios and that would also permit criticality.

More precise limiting deposit concentrations could be determined and selected for the evaluation, but the probability of such precise conditions to support criticality were not judged worthy of further criticality safety analyses.

9 PARAMETERS USED AS INPUT SOURCES

The range of input parameters of potentially mutual concern to the hydrogeochemical simulations and nuclear criticality evaluations was established for this task. A nominal soil composition was used for both analytical studies. Chemistries of water contained in soil considered within the hydrogeochemical simulations were not considered significant to the neutronic calculations because the dry nominal soil composition contains the overwhelming fractions of elemental constituents for damp or water-saturated soils. However, water content (i.e., grams of H_2O/grams of soil concentration or water density, grams of H_2O/cm^3 of bulk soil density) was considered significant and was included in the neutronic calculations.

9.1 PARAMETERS USED IN HYDROGEOCHEMICAL SIMULATIONS

The only SNM included in this study was ^{235}U, which is the most abundant fissile isotope at the disposal site being considered. Furthermore, because the thermodynamic databases are far less reliable for plutonium than for uranium, the results of model simulation with plutonium would be subject to far greater uncertainties than with uranium.

Flow of water and other chemicals occurred into the top and out of the bottom of the 10-m-long column. A 1-m grid spacing simulated transport, with a dispersion coefficient of 0.1, which is typical for this scale (Gelhar, Welty, and Rehfeldt, 1992). The velocity and hydraulic conductivity were uniform within the column, but a range of parameter values were examined. All model runs were conducted in the saturated mode. The travel times and pore volumes for different simulations are reported. Travel times for unsaturated conditions would be longer and were calculated as a simple linear reduction factor (as discussed subsequently). Output was obtained for up to 14,600 time steps for all runs. Selected runs were modeled for 73,000 time steps to determine when steady-state conditions had been achieved; many of the runs reached steady state by 14,600 time steps. Because of the small time step (0.5 days) needed for reactive transport, longer run times were not part of the standard output. Again, this time step does NOT represent travel times for unsaturated conditions.

Six aqueous components (H^+,CO_3^{2-},Ca^{2+}, UO_2^{2+}, Na^+, and HPO_4^-), 1 surface component, 18 aqueous species, 4 surface species, and 2 minerals were considered in most of the simulations. The equilibrium constants and stoichiometric relationship of the species and minerals were input to the model (Table 7.1-1). The primary mobilizing agent was CO_3^{2-}. The pH is important in determining the extent of uranium complexation by CO_3^{2-}, and Ca^{2+} and Na^+ were included as cations to balance the CO_3^{2-}. An additional complexing agent included was PO_4^{2-} because tripolyphosphate was used to stabilize caps at the Envirocare site. The initial conditions were equilibrated with PHREEQE under oxidizing conditions, and the phosphate concentration was limited by hydroxyapatite solubility; the CO_3^{2-} was limited by calcite solubility.

The parameters selected for sensitivity analysis were uranium concentration on initial sorption sites, CO_3^{2-} concentration of the influent water, pH of the influent water, velocity, concentration of high adsorption sites, and the pattern of the adsorption sites (one or multiple sorption zones). In general, the ranges of parameters were chosen on the basis of typical soil and waste site conditions, not any particular site data. Except for the sorption pattern, the parameters were sampled uniformly from their logarithmic values to cover the full range of expected values.

The total uranium was equal to or greater than the maximum allowable ^{235}U for disposal. Because the geochemical transport behavior of uranium is independent of the isotope, enrichment is not an influence on

hydrogeochemical processes. Increasing the total uranium concentration, while assuming a fixed ^{235}U, means the total uranium used in the hydrogeochemical transport models can be interpreted to span a range of ^{235}U enrichments.

The model input was relative to pore water (even for sorbed species), so uranium concentrations are converted to molar as follows. The state of Utah regulatory limit for ^{235}U is 770 pCi/g soil. Assuming a soil density of 1.6 g/cm^3 and using the specific activity for ^{235}U to be 2.16×10^6 pCi/g ^{235}U, then the allowable concentration of ^{235}U in the soil is 0.0006 g/cm^3. If the saturated porosity of the soil is 33%, the concentration is 1.2 g ^{235}U/L of pore water or 5×10^{-3} *M*. Uranium concentrations representative of ^{235}U enrichments of 100 to 1% were modeled. Thus the full range of uranium is 5×10^{-3} *M* for 100% ^{235}U to 0.5 *M* for 1% ^{235}U (0.0006 to 0.06 g/cm^3). Initially, uranium was sorbed to the soil with a low sorption capacity (see the following). Some runs were conducted using a uranium mineral as the initial form of uranium in the soil.

In addition, a series of runs were conducted with the uranium concentration reported at the Envirocare site. The total uranium concentration (with the same soil density assumptions) is 4.02×10^{-3} *M* with an estimated ^{235}U enrichment of only 0.42%. These concentrations are below the range used in the sensitivity analysis, so the sensitivity analysis concentration presented a test of regulatory concerns, not site conditions. A summary of Envirocare disposal records on an annual basis is provided in Appendix B.

The expected range of total inorganic carbon in landfill leachate is from 1×10^{-4} to 1×10^{-2} *M* (the range of compositions reported by Baedecker and Back, 1979; Staubitz et al., 1989; and data from L. Morton, State of Utah, Department of Environmental Quality). Although high CO_3^{2-} levels increase mobility of uranium, they also can maintain the uranium as a soluble complex under otherwise immobilizing conditions. Thus it was not clear a priori what the effects of high vs low CO_3^{2-} would be. The concentration of available CO_3^{2-} is also controlled by pH, so the pH was varied from 6.5 (HCO_3^- dominant) to 9.5 (CO_3^{2-} dominant). This was considered a reasonable range for CO_3^{2-}-buffered systems, although slightly wider ranges could be considered.

The flow velocity was varied over two orders of magnitude for the sensitivity analysis, although natural soils can vary over an even wider range. However, this range provided information on model sensitivity, and extremely low velocities take a long time to run. Thus a fast upper value was selected to speed run times, 0.5×10^{-3} to 0.5 m/d. The approach used to estimate transport times under unsaturated conditions was based on simulations under saturating conditions that necessarily involve simplifications and relatively large uncertainties. To calculate unsaturated conditions, a decrease in velocity of 2 to 3 orders of magnitude was assumed (Baver, Gardner, and Gardner, 1971; Hillel, 1991) because of lower hydraulic conductivities (but higher gradients, which limit the reduction in velocities). This estimate was crude, but the uncertainty in velocities points to the need to better understand hydraulic conditions at specific sites. Although this approach neglects the effects of dispersion under unsaturated conditions (which could increase concentrations somewhat; Jardine, Jacobs, and Wilson, 1993) and does not account for conditions of transient saturation, the calculation does provide a rough estimate of travel times. To conduct transient, saturated/unsaturated modeling would (1) require significantly more model input (for soil conditions), (2) increase model uncertainty (because the soil data are not available), and (3) be more computationally intensive. Furthermore, no test problems are available for saturated/unsaturated multispecies transport to provide confidence in model calculations.

Two scenarios for immobilization and increase in concentration of uranium were considered to encompass both oxidizing and reducing conditions. The first scenario is sorption of uranium within a zone having a higher adsorption capacity than the bulk soil. The sorption site equilibria were obtained from Yeh and Tripathi (1991b). Concentrations of sorption sites were compared to literature values for uranium sorption on iron hydroxides.

Reported values range from 0 to 2.4 mg uranium/g soil (Hsi and Langmuir, 1985), which is equivalent (see Table 9.1-1) to a maximum concentration of binding sites of 3.2×10^{-2} mol/L (units relative to pore water were required by the model). A slightly higher upper range was considered, and sensitivity was examined over 2 orders of magnitude by using a range of sorption site density from 1.0×10^{-3} to 1.0×10^{-1} *M*.

Table 9.1-1 Assumptions and calculations for conversion from milligrams of uranium per gram of soil to moles of uranium per liter of water

Concentration of uranium	Comment
2.4 mg U/g soil	Maximum reported value
1×10^{-4} mmol U/g soil	235 g/mol U
1×10^{-4} mmol U/0.625 cm^3 soil	Assuming soil density of 1.6 g/cm^3
1×10^{-4} mmol U/0.937 cm^3 porous media	Assuming soil volume of 67% gives the calculated total volume
1×10^{-4} mmol U/0.312 cm^3 water	Assuming porosity of 33%, water saturated
3.2×10^{-2} mol/L	cm^3 = ml, 1000 ml = L

Some runs under oxidizing conditions were conducted with mineral precipitation as the immobilizing mechanism. Rutherfordine (UO_2CO_3) was used in all runs in conjunction with high sorption zones because of the importance of CO_3^{2-} species. Soddyite [$(UO_2)_2SiO_42H_2O$] and $UO_2(OH)_2$-beta were used in conjunction with sorption for some runs, but these minerals did not concentrate uranium because sorption dominated; these were not considered further. Precipitation of $UO_2(OH)_2$-beta without sorption was also modeled in one run.

For the second immobilization scenario, reduction and precipitation were considered. Presumably, essentially all the uranium precipitated when it reached the reducing zone at the end of the column. This assumption was based on stability diagrams (e.g., Langmuir, 1978) as well as modeling using PHREEQE, which indicated that less than 1×10^{-9} *M* uranium was in equilibrium with reduced uraninite (UO_2). The mobilization portion of this scenario made use of the existing runs with the parameters described previously.

9.2 PARAMETER RANGES OF SOIL COMPOSITION AND POROSITY FOR CRITICALITY ASSESSMENT

The first step in the criticality safety assessment involved selection of a composition of a nominal geologically realistic soil for the initial neutronics calculations. The elemental composition of soils can vary greatly depending on individual soils. The composition (mean and range) of major elements in soil are listed in Table 9.2-1 and served as a basis for selection of the composition of the nominal soil. For neutronic calculations, the weight percents of individual elements were selected to minimize neutron capture and maximize neutron moderators to create a soil that is highly reactive from a criticality perspective.

Table 9.2-1 Mean and range of elemental compositions in soil

Element	Range of reported "mean" soil composition (wt %)[a]	Range of soil composition (wt %)[b]
O	49	–
Si	31 – 33	1.6 – 45
Al	7.1 – 7.2	0.07 – >10
Fe	2.6 – 3.8	0.01 – >10
Ca	1.4 – 2.4	0.01 – 32
Na	0.68 – 1.2	<0.05 – 10
K	1.36 – 1.5	0.005 – 6.3
Mg	0.6 – 0.9	0.005 – >10
B	0.001 – 0.003	0.002 – 0.03
C	1.0 – 2.5	0.06 – 37

[a]Ermolenko (1972); Bohn, McNeal, and O'Connor (1985); Sposito (1989); Shacklette and Boerngen (1984).
[b]Shacklette and Boerngen (1984).

The water content of the soils is limited by the porosity of the matrix containing the waste. The lower limit on porosity could be zero, or potentially could be the percent water contained by the clay mineral structure (5 to 20%). A typical mean porosity of 33% was assumed. Thus each bulk "soil" composition could accommodate approximately 0.33 g of H_2O/cm^3 or ~0.21 g of H_2O/g soil. Because of the high theoretical density of ^{235}U and $^{235}UO_2$ (~18 g and ~9 g of U/cm^3, respectively), the void fraction within the soil could easily accommodate upwards to 0.5 g of $^{235}U/cm^3$ with little impact on available void fraction for water.

The nuclear criticality calculations were performed by merely inserting fiducial values of water and ^{235}U densities into the input of a calculation and determining the k_∞ of the mixture. These fiducial values were selected to span the considered range of SiO_2 water and uranium densities (i.e., $0 \leq$ g of $H_2O/cm^3 \leq 0.4$, and about $0.0005 \leq$ g of $^{235}U/cm^3 \leq 1.0$). The same mixtures were then used to determine the dimensions of "critical" ($k_{eff} = 0.95$) single spheres, single infinitely long cylinders, and single infinitely distributed planes surrounded with 2 m of the same soil mixture without the ^{235}U contaminant (see Appendix A for schematics of computational models). The 0.95 value was assumed for the critical condition to account for uncertainties in the modeling and evaluation.

The two soil compositions used for this study were the "nominal" soil (derived from data in Table 9.2-1) and an "SiO_2" soil, both of which were taken to have a dry bulk density of 1.6 g/cm^3. The elemental composition of these soils is provided in Tables 3-1 and 9.2-2.

Table 9.2-2 Elemental composition for SiO_2 soil

Element	Weight percent of dry bulk density (1.6 g/cm^3)
Oxygen	53.257
Silicon	46.743

9.3 PARAMETRIC INPUT FOR THE DIRECT-DOSE CRITICALITY CONSEQUENCE EVALUATION

Assumed Deposit Conditions

Two postulated criticalities were defined that corresponded to high and low concentration factors of 10.5 and 3.6 relative to the regulatory limit of 0.0006 g of ^{235}U per cm^3. The high concentration factor (HCF) corresponds to the upper limit indicated as possible based on hydrogeochemical scenarios. The low concentration factor (LCF) roughly corresponds to the minimal concentration conditions under which a criticality can occur. The criticality events were assumed to occupy a localized region with a thickness equal to the slab geometry thickness defined as "critical" (actually k = 0.95) for the purposes of this study. While the cases studied for criticality purposes allowed for both dry and wet systems, the radiation transport studies to determine criticality consequences only analyzed dry systems. The dry systems are assumed to be limiting since water is a very good shield for the large number of neutrons released from a criticality event.

10 RESULTS

Observations of the consolidated results and intersections of parameter surface responses from the hydrogeochemical scenarios and nuclear criticality evaluations yielded a small number of simulated geological circumstances that could potentially support nuclear criticality. That is, few sets of hydrogeological conditions yielded ^{235}U CFs greater than 3, and CFs greater than 10 are not judged credible based upon the stated hydrogeochemical simulation assumptions. Criticality cannot be achieved with less than a CF of about 2 for the assumed SiO_2 soil or less than a CF of about 6 for the assumed nominal soil. Results of the two criticality consequence evaluations provide ranges of radiation dose estimates at the ground surface centered above the disklike concentrated uranium deposits (56 to 157 rem) and at the ground surface but translated 90 m from the vertical axes of the disklike concentrated uranium deposits (57 to 165 mrem).

10.1 NUCLEAR CRITICALITY EVALUATION

Specific values from the nuclear criticality evaluations are presented in tabular format in Appendix C (Table C-1 for the nominal soil and Table C-2 for the SiO_2 soil). Corresponding 3-D surface plots (Appendix D) provide a broader view of the relationship and responses of the systems to the water and ^{235}U content in the soils; the figures in Appendix D are cited in the column headings of the tables in Appendix C.

10.1.1 Interpretation of Results

The state of Utah provides a license disposal limit of 770 pCi of ^{235}U/g of soil-like waste for Envirocare. Assuming a soil-like waste density of 1.6 g/cm^3 of soil, this translates to 0.0006 g of ^{235}U/cm^3 of soil, subsequently referred to as the state of Utah limit. Although increases in concentrations of ^{235}U densities are limited by hydrogeochemical conditions, CFs ten times the authorized initial uranium elemental density could result in critical systems having densities of about 0.006 g of ^{235}U/cm^3 of soil (see line entries 13 through 17 in Table C-1 and line entries 34 and 158 through 164 in Table C-2, which are extracted in the following).

Additional critical densities and infinite-media multiplication constants are provided in Tables C-1 and C-2. As acknowledged by the state of Utah (1994), the licensee may construct disposal cells to accommodate 10-m depths of waste materials. Under the assumption that the soillike materials have a density of about 1.6 g/cm^3, the permitted parameters for the burial cell can result in an areal density (i.e., the mass of ^{235}U projected downward to any 1-m^2 area on the floor of the cell) of about 5.2 kg/m^2. Given hydrogeochemical CFs of 3 or 10 for allowed ^{235}U disposal densities, critical ($k_{eff} \geq 0.95$ in this study) infinite-slab systems of "SiO_2" or "Nominal Soil," respectively, can be created (see Tables C-1 and C-2).

In nominal soil, the vertical migration of ^{235}U into infinite planar configurations can produce critical concentrations, as shown in Table 10.1-1, line entries 16 and 17. Although spherical or cylindrical geometries can also occur in the waste matrix, they require greater concentrations of ^{235}U to become critical because of the geometric effect of increased neutron leakage from cylinders and spheres. For example, the areal density of an infinite slab of nominal soil at 0.006 g of ^{235}U/cm^3 (line entry 15) is critical at 5.286 kg of ^{235}U/m^2 (very nearly the assumed critical value of 5.2 kg of ^{235}U/m^2) in a planar configuration. However, the ^{235}U linear density of the infinite cylinder in line entry 15 requires a projected areal density of about 7.8 kg of ^{235}U/m^2 (i.e., 12.9 kg of ^{235}U/1.65-m cylinder diameter × 1-m cylinder length = 12.9/1.65 kg of ^{235}U/m^2, or 7.8 kg of ^{235}U/m^2). Thus achieving criti-cality in a cylindrical geometry requires substantial lateral migration as well as vertical migration of the ^{235}U.

Table 10.1-1 Extracted nominal-soil (N-S) results[a]

		^{235}U content		Water content			Critical infinite slab[b]		Critical infinite cylinder[b]		Critical sphere[b]	
Line entry	Conc. factor	g ^{235}U per cm^3	g ^{235}U per g N-S	g H_2O per cm^3	g H_2O per g N-S	k_∞ or k-infinity	Thickness (cm)	^{235}U areal density (kg/m^2)	Diameter (cm)	^{235}U linear density (kg/m)	Diameter (cm)	^{235}U mass (kg)
						Fig. D-1	Fig. D-2	Fig. D-3	Fig. D-4	Fig. D-5	Fig. D-6	Fig. D-7
7	3.3	0.0020	0.001250	0.000	0.000000	0.825						
8	3.3	0.0020	0.001250	0.020	0.012500	0.803						
9	3.3	0.0020	0.001250	0.040	0.025000	0.770						
10	3.3	0.0020	0.001250	0.080	0.050000	0.706						
11	3.3	0.0020	0.001250	0.160	0.100000	0.602						
12	3.3	0.0020	0.001250	0.330	0.206250	0.457						
13	10.0	0.0060	0.003750	0.000	0.000000	1.292	142.950	8.577	270.140	34.389	382.220	175.424
14	10.0	0.0060	0.003750	0.020	0.012500	1.310	101.720	6.103	192.520	17.466	272.700	63.710
15	10.0	0.0060	0.003750	0.040	0.025000	1.292	88.100	5.286	165.280	12.873	233.480	39.985
16	10.0	0.0060	0.003750	0.080	0.050000	1.239	78.040	4.682	143.000	9.636	200.600	25.360
17	10.0	0.0060	0.003750	0.160	0.100000	1.131	78.860	4.732	137.920	8.964	190.040	21.562
18	10.0	0.0060	0.003750	0.330	0.206250	0.946						

[a]Figures presented in Appendix D.

[b]Dimensions based on system k_{eff} = 0.95 rather than k_{eff} = 1.0 to conservatively account for methods and data uncertainty.

Achieving criticality in a spherical geometry requires a projected areal density of about 9.34 kg of $^{235}U/m^2$ (i.e., 39.985 kg of $^{235}U/4.28\ m^2$ of projected area of the sphere = 39.985/4.28 kg of $^{235}U/m^2$, or 9.34 kg of $^{235}U/m^2$), a density that requires significant lateral migration of ^{235}U. In summary, a unidirectional migration of SNM into a slablike configuration requires less concentration of ^{235}U to pose nuclear criticality safety concerns than do cylinders and spheres.

The SiO_2-soil results are similar to the nominal soil results. For line entry 160 in Table 10.1-2, the critical areal density for the infinite slab is 3.156 kg/m^2. The approximate projected areal density of the infinite cylinder is 4.85 kg of $^{235}U/m^2$ (i.e., 4.748 kg of $^{235}U/0.9796\ m^2$ of projected area). The approximate projected areal density of the sphere is 5.87 kg of $^{235}U/m^2$ (i.e., 9.106 kg $^{235}U/1.55\ m^2$ of projected area).

For cases in which localized voids (e.g., such as under slabs of broken concrete that cannot be compacted) might provide regions for ^{235}U-bearing solutions (e.g., aqueous or organic based) to collect without soil dilution, another reference exists (Paxton and Pruvost, 1987) for predicting critical masses and volumes with concentrations greater than 0.013 g of $^{235}U/cm^3$. The concentration yielding the smallest critical mass is approximately 0.05 g of $^{235}U/cm^3$ for a critical mass of ~0.83 kg of ^{235}U in an ~17-L volume; such a mass yields a projected areal density of about 10.38 kg of $^{235}U/m^2$. The larger critical masses observed for the soils is due to the dilution of fissile material increased neutron leakage and some neutron capture.

The seemingly anomalous circumstances where nominal soil or SiO_2 soil can produce more sensitive results (i.e., lesser critical concentrations) than pure aqueous solutions of ^{235}U is founded in the low neutron-capture values for the assumed soils in comparison with the values for aqueous solutions. In dilute systems, the neutron capture in hydrogen can offset the hydrogen influence in slowing down and thermalizing neutrons for more effective thermal fission. Table 10.1-2 shows that for relatively dilute systems, the addition of water to the ^{235}U and SiO_2 systems reduces the infinite-media multiplication constant, k_∞.

10.1.2 Interpretation of Results in Relation to Envirocare of Utah, Inc.

The uranium concentration in soil was estimated from reports provided by D. L. Finefrock of the State of Utah, Department of Environmental Quality. The inventory reports provide estimates of picocuries of uranium and volume of soil for the years 1988–95 (Appendix B). These data have been converted into the overall average concentration of $9.5 \times 10^{-4}\ g/cm^3$ total U. The calculated average enrichment from the inventory is estimated at only 0.42%. This average enrichment is below the minimum 1% enrichment required to achieve nuclear criticality in a homogeneous water-moderated system. The inventory of the waste pit would need to be examined to further evaluate nuclear criticality safety. However, this concentration ($9.5 \times 10^{-4}\ g/cm^3$) was used to model site conditions and to provide an evaluation for slightly larger enrichments that could exist in disposal cells.

This study did not evaluate the validity of the “unity rule” prescribed with either the state of Utah license condition 16.A for combinations of fissile nuclides (i.e., ^{235}U, ^{239}Pu, and ^{241}Pu) or license condition 16.B for quantities of other sole fissile nuclides that contaminate soils (i.e., ^{239}Pu and ^{241}Pu). However, the authorized concentrations of ^{239}Pu and ^{241}Pu are nearly seven orders of magnitude smaller than the permissible concentration of ^{235}U. Because the critical masses of ^{239}Pu and ^{241}Pu are not much less than one quarter that of ^{235}U, it is highly improbable that ^{239}Pu or ^{241}Pu will pose a nuclear criticality hazard at the Envirocare site at the specified disposal limits.

Table 10.1-2 Extracted SiO_2-soil (S-S) results[a]

		^{235}U content		Water content			Critical infinite slab[b]		Critical infinite cylinder[b]		Critical sphere[b]	
Line entry	Conc. factor	g ^{235}U per cm^3	g ^{235}U per g S-S	g H_2O per cm^3	g H_2O per g S-S	k_∞ or k-infinity	Thick-ness (cm)	^{235}U areal density (kg/m^2)	Diameter (cm)	^{235}U linear density (kg/m)	Diameter (cm)	^{235}U mass (kg)
						Fig. D-8	Fig. D-9	Fig. D-10	Fig. D-11	Fig. D-12	Fig. D-13	Fig. D-14
34	3.00	0.0011250	0.0011250	0.00000	0.00000	1.060	367.1300	6.6083	621.5	54.6065	844.74	355.0752
35	3.00	0.0018000	0.0011250	0.05800	0.03625	0.894						
36	3.00	0.0018000	0.0011250	0.11900	0.07438	0.751						
37	3.00	0.0018000	0.0011250	0.18300	0.11438	0.642						
38	3.00	0.0018000	0.0011250	0.25100	0.15688	0.556						
39	3.00	0.0018000	0.0011250	0.32400	0.20250	0.487						
40	3.00	0.0018000	0.0011250	0.40000	0.25000	0.431						
158	10.50	0.0063000	0.0039375	0.00000	0.00000	1.512	94.2100	5.9352	205.28	20.8508	303.38	92.1086
159	10.50	0.0063000	0.0039375	0.05800	0.03625	1.478	54.5300	3.4354	113.3	6.3517	165.3	14.8990
160	10.50	0.0063000	0.0039375	0.11900	0.07438	1.367	50.1000	3.1563	97.96	4.7482	140.28	9.1060
161	10.50	0.0063000	0.0039375	0.18300	0.11438	1.261	50.9400	3.2092	94.72	4.4393	133.44	7.8379
162	10.50	0.0063000	0.0039375	0.25100	0.15688	1.163	55.9600	3.5255	99.6	4.9085	138.1	8.6880
163	10.50	0.0063000	0.0039375	0.32400	0.20250	1.075	68.4900	4.3149	117.08	6.7826	159.62	13.4153
164	10.50	0.0063000	0.0039375	0.40000	0.25000	0.995	111.8700	7.0478	174.66	15.0945	246.4	49.3471

[a]Figures presented in Appendix D.

[b]Dimensions based on system k_{eff} = 0.95 rather than k_{eff} = 1.0 to conservatively account for methods and data uncertainty.

10.1.3 Discussion of Results in Relation to the NRC Division of Fuel Cycle Safety and Safeguards Proposed Limits

Results of this nuclear criticality evaluation demonstrate that the ^{235}U limit proposed in the rule change petition (Federal Register, 1993) will yield subcritical conditions based on three specified assumptions:

1. the SNM is uniformly distributed throughout the soil,

2. the soil matrix is SiO_2, and

3. the SNM-contaminated soil matrix has a spherical geometry and an optimal water content for nuclear criticality.

As noted in Sect. 1.2, the basic assumption for uniform distribution at the proposed SNM limit of 5000 g of soil/ 1 g of ^{235}U (100% enriched), or 0.0002 g of ^{235}U/g SiO_2, cannot be assured through hydrogeochemical influences, but the SNM limit does provide a factor of 3 reduction from the Utah license limit. However, the probability of transporting the ^{235}U and concentrating it into a suitable geometry and density to achieve criticality is very low. The results do confirm that SiO_2 is a conservative soil matrix for nuclear criticality evaluations.

The fact that a spherical geometry requires multidirectional, convergent movement and greater concentrations than a slab geometry of ^{235}U indicates that a sphere may not be representative of realistic migrations of SNM within a disposal cell. In fact, since total mass is not restricted at an LLW site, a slab configuration seems the most likely to yield a potential for criticality (see Sect. 10.2, Geochemical Transport, and Sect. 4, Relevant Analogs).

10.2 GEOCHEMICAL TRANSPORT

The nuclear criticality safety analysis above makes it clear that a key uncertainty in evaluating safety is the potential for redistributing SNM to zones of higher concentration by hydrogeochemical processes. The likelihood that uranium would concentrate to levels of concern for criticality safety analysis must be evaluated to determine whether specific configurations of uranium can occur. Through hydrogeochemical modeling, project participants examined uranium's potential to mobilize and concentrate as a slab. Results are presented in terms of the extent to which uranium is concentrated by immobilization processes. Results are also described in terms of the spatial distribution of mobile uranium (dissolved species) or immobilized uranium (adsorbed species) in the simulated columns as a function of the number of pore volumes of eluant passing through the soil profile; the number of pore volumes is related to time and flow velocity. Data from 40 simulations were examined as part of the formal sensitivity analysis (Table 10.2-1). Note that each run had unique input variables of pH, etc. Output is summarized as CFs resulting from sorption and from flushing. The definitions of these CFs are given in the table. The results of the 40 simulations are discussed in Sects. 10.2.1 and 10.2.3 and are discussed within the context of the sensitivity analysis in 10.2.4. In addition, the effects of some alternate scenarios were examined, such as precipitation of oxidized minerals and uranium disposal at levels below the regulatory limit (i.e., from disposal records at the Envirocare site).

Table 10.2-1 Summary of sensitivity analysis runs

Input data								Output	
Run No.	pH	log CO_3^{-2} *M*	U conc, *M*	Enrich %	log sorp conc, *M*	Sorp patt	Vel m/day	Flush CF	Sorp CF
1	7.27	-3.23	0.0539	4.64	-2.18	2	0.0090	1.72	0.09
2	9.50	-3.18	0.1385	1.80	-2.33	1	0.2462	1.86	0.78
3	7.81	-2.00	0.0092	27.28	-1.31	2	0.0080	7.84	1.38
4	8.81	-2.87	0.1754	1.43	-1.51	1	0.3509	2.73	0.79
5	6.81	-2.51	0.0209	11.94	-3.00	2	0.1535	8.00	0.81
6	8.88	-3.74	0.0025	100.00	-1.56	2	0.0419	3.48	1.04
7	7.73	-3.33	0.0057	43.75	-2.38	2	0.0372	3.01	0.80
8	7.96	-2.26	0.0036	70.18	-2.79	2	0.2188	8.00	0.00
9	9.19	-3.90	0.0103	24.24	-1.10	1	0.1364	2.04	0.99
10	8.50	-2.41	0.0972	2.57	-1.00	1	0.1212	5.43	0.87
11	9.27	-2.92	0.2500	1.00	-1.41	2	0.0261	1.66	0.90
12	8.12	-3.69	0.0081	30.70	-1.36	1	0.0294	7.91	0.02
13	7.88	-3.64	0.0298	8.38	-2.54	2	0.0183	0.34	0.92
14	7.04	-4.00	0.1974	1.27	-2.49	1	0.0471	1.70	0.87
15	8.19	-3.85	0.0682	3.67	-1.77	1	0.0063	1.31	0.90
16	6.58	-3.28	0.0032	78.96	-2.08	1	0.0114	4.05	0.88
17	7.65	-2.36	0.2221	1.13	-1.97	2	0.0206	2.07	0.90
18	9.04	-2.05	0.0236	10.61	-1.67	2	0.0129	6.30	1.11
19	7.19	-2.77	0.1559	1.60	-1.21	2	0.0756	2.45	1.06
20	7.35	-3.03	0.0864	2.89	-1.62	2	0.0102	1.47	0.97
21	7.12	-2.62	0.0147	17.01	-1.72	2	0.1944	8.00	1.09
22	8.42	-2.15	0.0265	9.43	-2.59	1	0.0050	3.06	0.86
23	6.73	-2.72	0.0040	62.36	-2.23	2	0.3948	8.00	0.00
24	6.50	-2.97	0.0336	7.44	-1.46	2	0.3118	8.00	0.39
25	8.58	-3.95	0.0479	5.22	-2.74	1	0.0597	1.94	0.90
26	9.12	-2.67	0.1231	2.03	-2.69	1	0.0232	1.94	0.89
27	8.04	-2.56	0.0768	3.26	-1.15	1	0.1728	3.77	0.68
28	7.42	-2.31	0.0606	4.12	-2.85	1	0.5000	8.00	0.78
29	8.27	-3.38	0.0378	6.61	-1.05	2	0.0530	1.85	1.14
30	7.50	-3.59	0.0072	34.55	-1.87	1	0.0957	2.66	0.73
31	8.73	-2.46	0.0186	13.43	-2.13	1	0.0851	8.00	0.78
32	7.58	-3.44	0.1094	2.29	-2.44	2	0.0071	1.28	0.90
33	6.96	-3.08	0.0045	55.41	-2.28	1	0.0163	4.79	0.83
34	9.35	-2.21	0.0064	38.88	-1.92	2	0.0331	8.00	1.02
35	8.65	-2.82	0.0131	19.15	-2.03	2	0.0672	4.52	0.98
36	9.42	-3.79	0.0165	15.12	-2.90	1	0.1077	1.74	0.81
37	6.65	-2.10	0.0116	21.54	-1.82	1	0.0056	5.22	0.84
38	8.35	-3.54	0.0028	88.86	-2.64	1	0.0145	4.52	0.57
39	6.88	-3.13	0.0051	49.24	-1.26	1	0.2771	8.00	0.81
40	8.96	-3.49	0.0425	5.88	-2.95	2	0.4443	2.00	0.00

KEY: Enrich % = effective enrichment = 100 × (regulatory limit)/(total U input) where the regulatory limit is 0.005 *M*.
U conc * 2 = total concentration on sorption sites.
Sorp patt = the pattern for sorption sites.
1 is high sorption zone at the end of the column.
2 is four high sorption zones interspersed with low sorption zones.
Vel = velocity.
Sorp CF = conc factor on sorption sites
= (peak concentration)/(input U concentration).
Flush CF = conc factor calculated after flushing.

10.2.1 Sorption

The effect of sorption on uranium mobilization and immobilization was examined in 40 simulations, summarized in Table 10.2-1. Sorption of uranium did not produce a sufficient increase in concentration to be of concern for nuclear criticality in these simulations. The CF of concern for criticality safety was between 3 and 10 (see Table C-1). Interpolation within the tables revealed that an approximately sixfold concentration is the lowest CF to raise a criticality safety concern. None of the simulations had CFs above 6 for the time period modeled (Table 10.2-1). The CFs were less than 2 for all cases modeled, and most were less than 1, indicating that uranium has flushed past the sorption zone. The mobilization of uranium was dominant over sorption of uranium.

In the presence of high concentrations of CO_3^{2-}, most of the uranium is quickly flushed out the column. Formation of highly mobile uranium-carbonate complexes is favored, even in the zone with the higher concentrations of sorption sites; under these conditions, CO_3^{2-} complexation outcompetes the sorption sites. This result can be seen in plots of concentration vs distance in the column for various times (breakthrough curves). Time was expressed as a relative unit termed as pore volumes or the number of times the column is flushed with water. The breakthrough curves of dissolved and sorbed uranium move down the model column with time. After only 24 pore volumes, the break-through curves are becoming flat and the uranium is nearly flushed from the column (Fig. 10.2-1). Modest CO_3^{2-} concentrations produce favorable conditions for concentration because uranium can be mobilized, but CO_3^{2-} does not outcompete sorption sites. With these lower CO_3^{2-} concentrations, uranium becomes sorbed in the 8- to 10-m range of the column by the high concentration of sorption sites (Fig. 10.2-2). The concentration of dissolved uranium remains fairly constant through time (pore volumes) when CO_3^{2-} concentrations are lower. At the first time step shown, the dissolved uranium is higher at the bottom of the column because it is still flushing out. The sorbed uranium has not increased over the initial concentration of 0.02 g U total/cm^3, but the concentration is higher at the bottom of the column than at the top.

Multiple zones of sorption in the column were modeled to see the effect of intermediate accumulation of uranium. The peak concentration at each zone of high sorption (at 3, 5, 7, and 9 m) in the column reached nearly the same level; as a result, the changes between the values in the high- and low-sorption concentration zones creates a sawtooth pattern (Fig. 10.2-3). The peak concentration was lower than the run with only one sorption zone. During flushing, the concentration in the third zone increased slightly for some simulations.

Another factor influencing the CF in the sorption zone is the size of the fissile uranium source term. The role of the cell depth (column length) was examined in more detail in additional simulations. There is an 8-m column with an even distribution of initial ^{235}U for the sensitivity analysis. For a simulation based on run 9 except with 0.004 *M* total U (a lower concentration than in Table 10.2-1), this column length produced a CF of 3.5. When the source column increased to 14 m in length, the CF increased to 4.8; for a 19-m column the CF is 5.8. Although the increase in CF does not cause a 1:1 linear increase (Fig. 10.2-4), the column length is a significant influence.

10.2.2 Influence of Oxidized Minerals

When mineral precipitation was included in the model, little or no change occurred in the uranium concentrations on the sorption sites. For the minerals examined {rutherfordine (UO_2CO_3), soddyite [$(UO_2)_2SiO_4 2H_2O$], and $UO_2(OH)_2$-beta}, adsorption dominated precipitation as an immobilizing mechanism in oxidizing conditions.

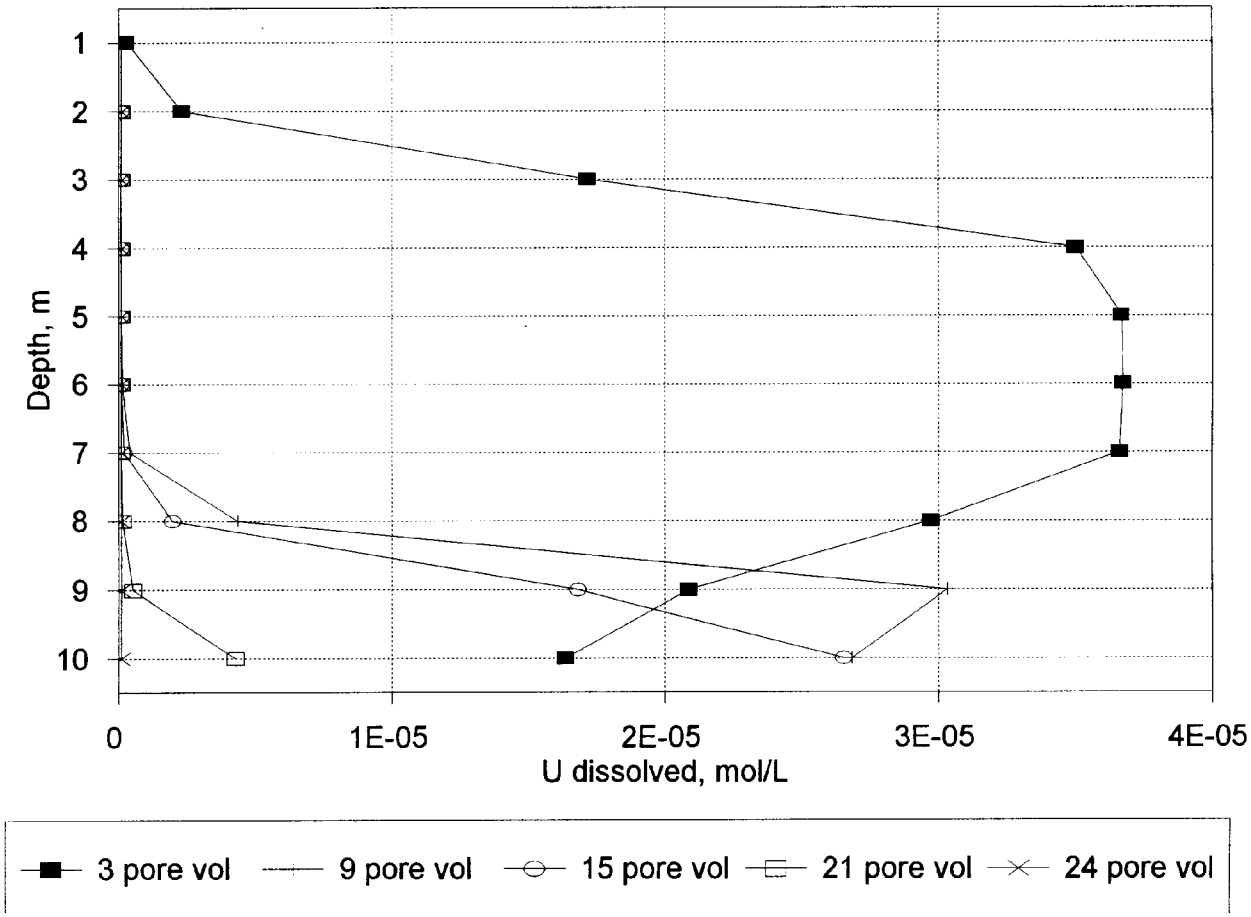


Figure 10.2-1 Example of flushing of uranium in solution and on sorption sites. Breakthrough curves are shown for several different time steps, expressed as the number of pore volumes that have passed through the column. The last timestep (8) is essentially flat. Run conditions are those for run number 39 which has an initial U_{TOT} = 0.01 *M*.

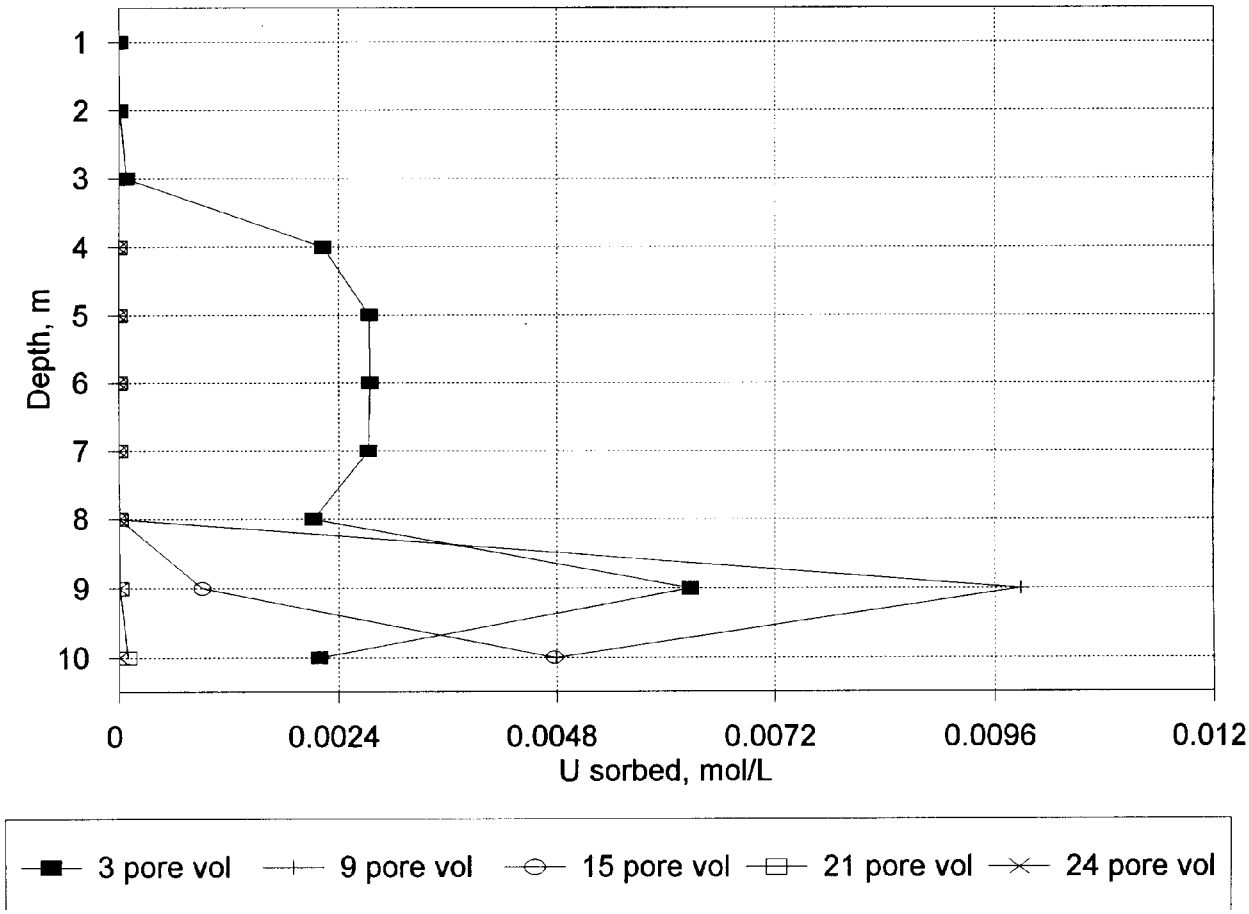


Figure 10.2-1 (continued)

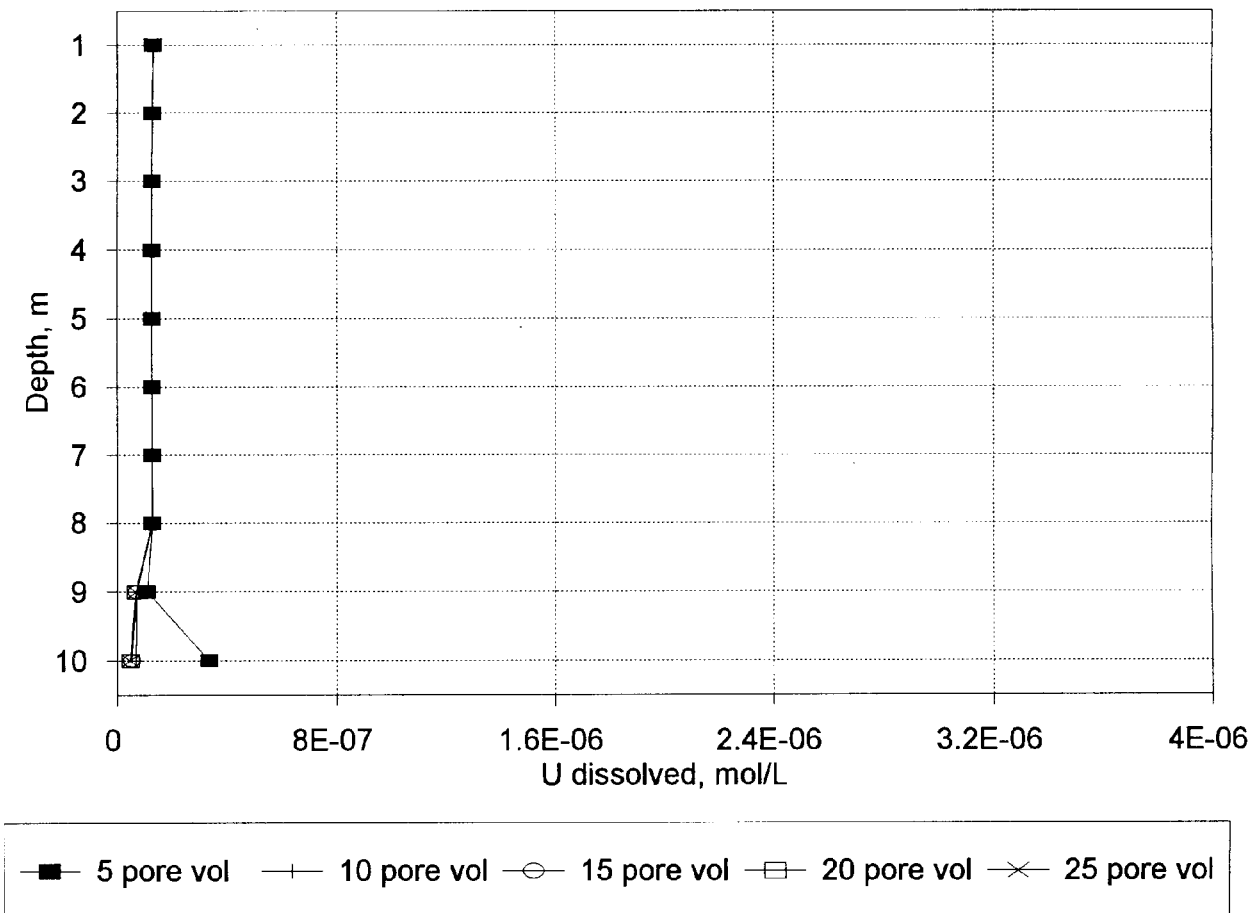


Figure 10.2-2 Example of sorption on zone at bottom of column with lower CO_3^{-2} concentration than shown in Fig. 10.2-1. Breakthrough curves are shown for several different time steps, expressed as the number of pore volumes that have passed through the column. The sorbed uranium concentrates in a localized zone shown by the peak, but the dissolved uranium has a fairly level concentration. Run conditions are for run number 9 which has an initial U_{TOT} = 0.02 *M*. Note the change in scale for dissolved uranium relative to Fig. 10.2-1.

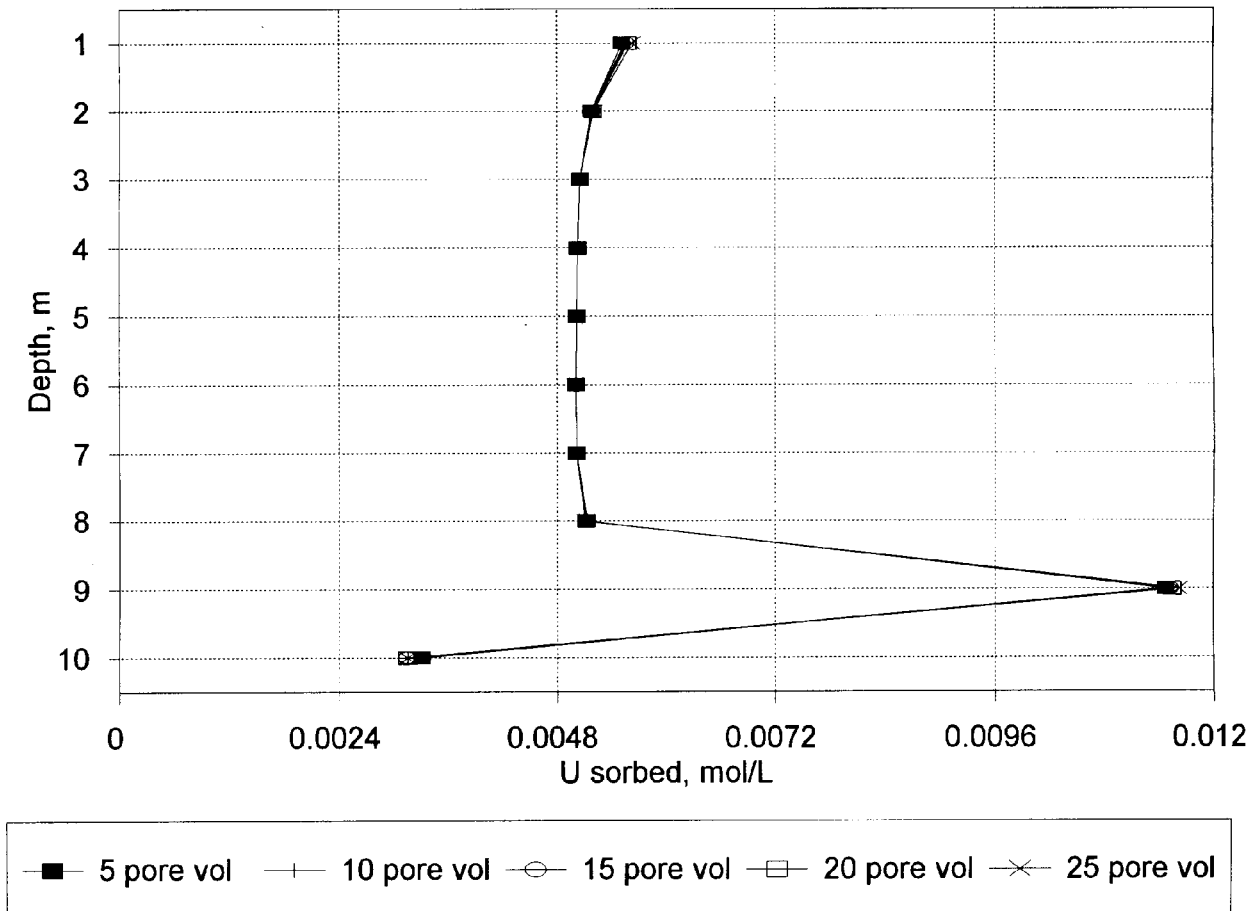


Figure 10.2-2 (continued)

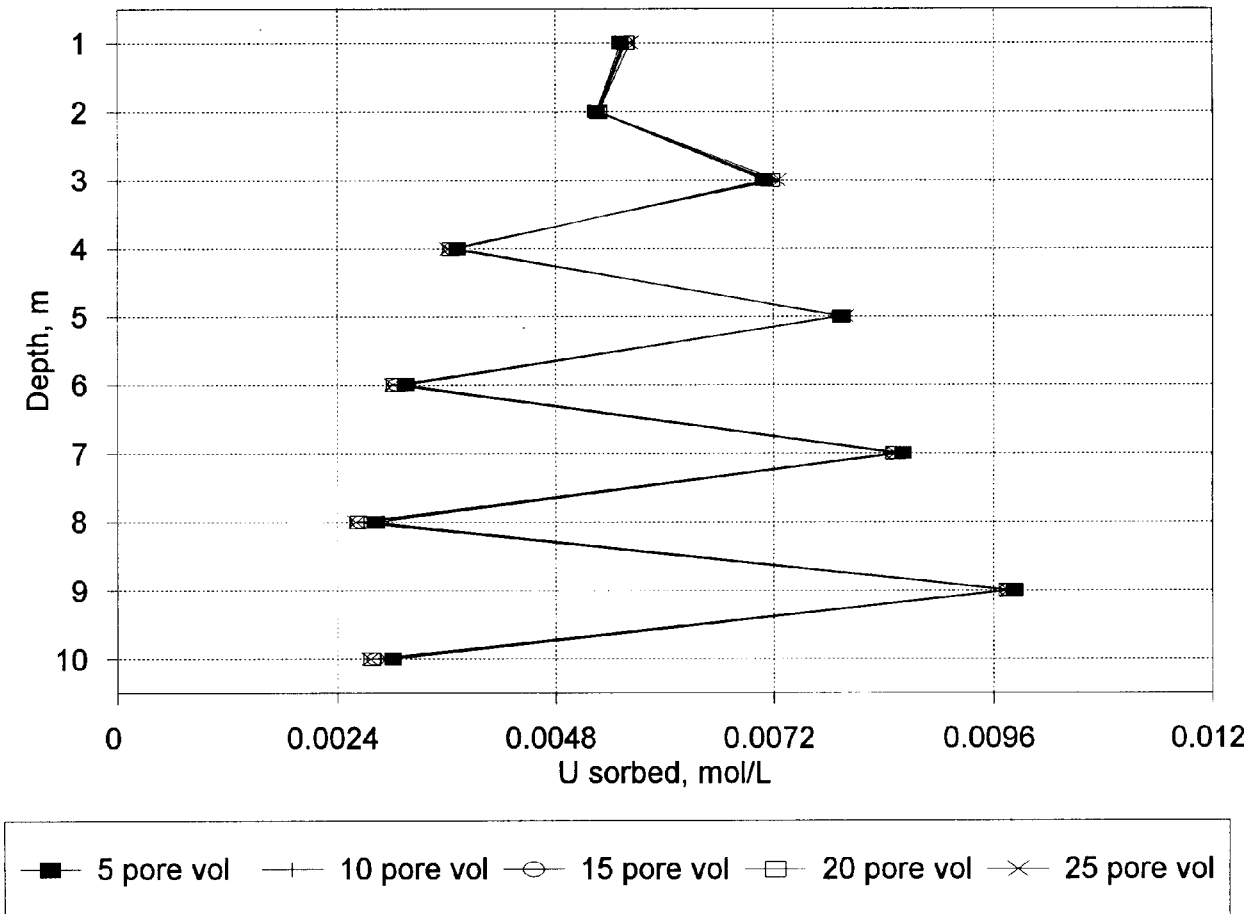


Figure 10.2-3 Example of sorption on multiple zones of high sorption within the column. The sawtooth pattern is created by alternation of high- and low-sorption site concentrations. The run is based on run 9 with an initial U_{TOT} = 0.02 *M*. The peak concentration was similar in the different zones and lower than the run with only one sorption zone (Fig. 10.3-2)

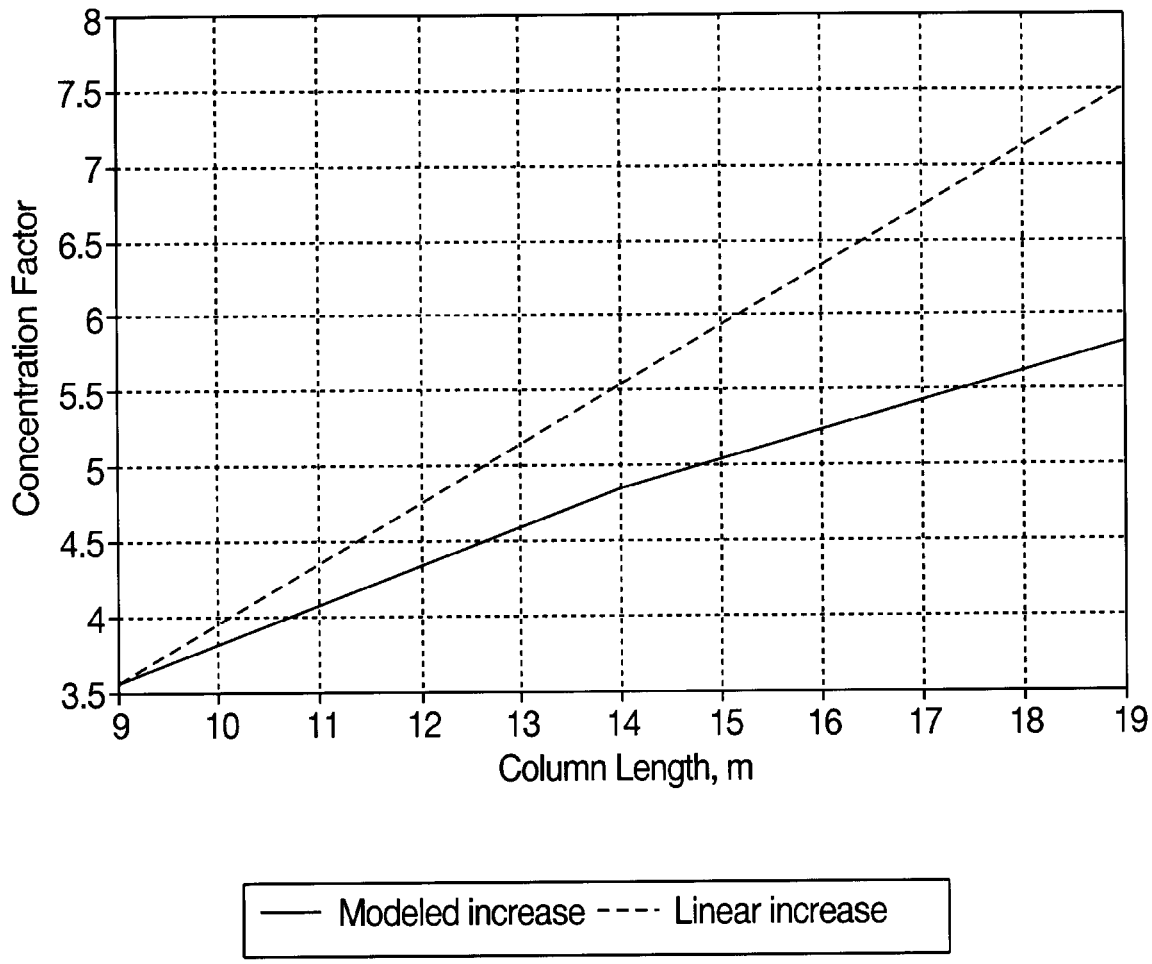


Figure 10.2-4 The effect of column length (or source thickness) on increase in uranium concentration. The modeled increase has a lower slope than a linear 1:1 increase but nonetheless has a significant slope

Results

The minerals did not precipitate because the equilibration of uranium with sorption sites left little uranium in solution. When the sorption zone was at the end of the column, the mobile uranium concentrated into a smaller zone than the initial configuration throughout the column, and the concentration reached 0.004 g of total U/cm^3 for the example shown (Fig. 10.2-5). However, when the solution was equilibrated with $UO_2(OH)_2$-beta in the absence of sorption sites, precipitation occurred throughout the column. The concentration reached only 0.00032 g of total U/cm^3 for this same example because the precipitate was spread over the top 8 m of the model columns.

10.2.3 Reducing Conditions

The presence of zones with reducing conditions constitutes an important immobilization scenario because the geochemical CF is not limited by the availability of sites as in the sorption scenario. Furthermore, complexation with the mobilizing agent (CO_3^{2-}) is less important given the low solubility of reduced uranium minerals. Of the 40 runs shown in Table 10.2-1, 12 runs had potential for increasing the concentration of uranium.

Transport was not modeled explicitly in the reducing scenario because of stability problems in redox zones for the first version of the code. Instead, the increase in uranium concentration in a reducing zone was estimated by evaluating the factors that would enhance the mobilization of uranium so that uranium would flush to the bottom of the column and concentrate in the reducing zone. This strategy was based on two factors: (1) the nearly complete demobilization of uranium by precipitation of reduced phase minerals and (2) the need to mobilize a large portion of the uranium initially spread out in the model column. Geochemical modeling of the extent of precipitation of uranium under reducing conditions (pe of –4 or less) clearly demonstrated that most uranium entering a reduced zone would be immobilized (e.g., Fig. 7.1-3). Reducing zones precipitate uranium readily. However, a strongly reducing (e.g., methanogenic) environment is preferable for precipitating reduced uranium (Raffensberger and Garvin, 1995). Because of the low concentrations of uranium ($<1 \times 10^{-9}$ *M*) in solution when equilibrated with reduced minerals, it was assumed that all of the uranium in a reducing zone was precipitated. The second hypothesis, that nearly all of the uranium in the disposal cell needed to be mobilized, is based on the geometry of the zones. For an 8-m thickness of initially uniformly distributed uranium to concentrate in a 1-m-thick reducing zone, the maximum CF is 8 (if all of the uranium is mobilized). This geometric CF has the potential to produce nuclear criticality under some conditions (Tables 10.1-1 and 10.1-2). With these assumptions, the problem was reduced to mobilizing the uranium and flushing nearly all of it to a hypothetical reducing zone.

The amount of uranium mobilized was estimated by doing a mass balance on uranium in the model. The modeled uranium concentration was summed over each of the model cells. The total initial uranium minus the final uranium gave the amount mobilized. The concentration of mobilized uranium is multiplied by the geometric factor of 8 to give the concentration in a hypothetical reducing zone. Then it is divided by the initial concentration to obtain the total CF (Table 10.2-1). For 12 of the 40 runs, this CF is greater than 6, and most of these are close to the maximum of 8 allowed by the geometry. Additional flushing could occur at longer times, but this sampling is fairly representative. Thus mobilization to a reducing zone seems to be a possible scenario for increasing uranium concentration.

10.2.4 Sensitivity Analysis

A sensitivity analysis was conducted to evaluate uranium mobility over a range of parameter values and to try to quantify the influence of different parameters. The effects of six parameters were evaluated for sorption of uranium and four parameters (excluding the two parameters related to sorption) for flushing of uranium.

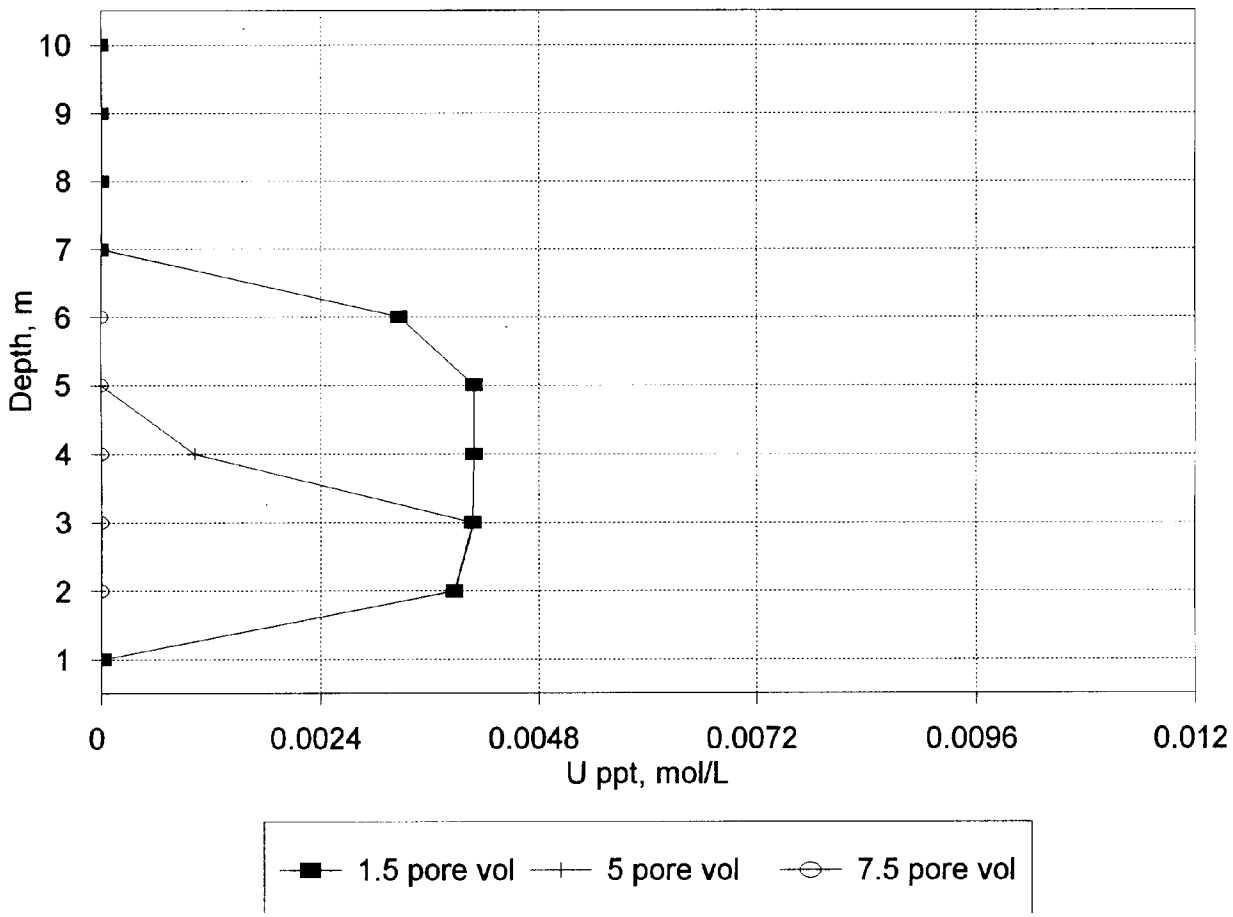


Figure 10.2-5 Example of uranium demobilization on sorption sites vs a mineral precipitate [$UO_2(OH)_2$-beta]. Sorption sites at the end of the model column concentrate uranium. Precipitation occurs throughout the column and results in a lower concentration of uranium. High sorption sites are omitted from the run with mineral equilibration only. Run conditions are for run 9 with an initial U_{TOT} = 0.02 *M*.

Results

Multivariate analysis was used to calculate regression coefficients for each variable. The higher the coefficient, the greater the influence of the variable. Rank regression was used.

The multivariate regression was not used to evaluate the importance of parameters for sorption. Although coefficients were calculated for the CFs from sorption, the correlation coefficient is low (0.65), indicating a poor fit to data. The CFs are low, and the data are clustered. See, for example, the relationship between initial uranium (input as uranium molarity) and the CF resulting from uranium absorption (Fig. 10.2-6).

The multivariate regression for CFs from mobilization to a reducing zone had a correlation coefficient of 0.9. The most important factor in moving the uranium to the reducing zone is the CO_3^{2-} concentration, which is the primary mobilizing complex. The CO_3^{2-} was directly correlated with a coefficient of 0.59 (Fig. 10.2-7). Uranium concentration was also significant but had an inverse correlation coefficient of -0.52. This inverse relationship suggests that high concentrations of uranium can limit the CF because more uranium must be mobilized. However, it is important to note that the relationship between initial uranium and flushing is influenced by other factors as well and cannot provide the only limit on resulting CFs, as evidenced by some scatter in the plot of Fig. 10.2-8. Velocity is also a significant variable, and slower velocities mobilized less uranium. It is possible that longer times would eventually lead to additional mobilization to the reducing zone in these cases. The pH was not as important, apparently because the total CO_3^{2-} concentration was sufficient to create free anions for uranium complexation.

In summary, the sensitivity analysis was useful in examining the interplay between various parameters. The behavior over the range of parameters modeled indicated that a narrow combination of factors led to conditions that could be of concern for criticality for the case of flushing uranium to a reducing zone. For the case of sorption, no runs resulted in CFs of concern, but longer disposal columns (i.e., thicker waste disposal cells) could cause larger increases in uranium concentration. No single variable dominated the behavior of uranium, so it is important to evaluate multiple factors.

10.2.5 Relevance to the Envirocare of Utah, Inc., Site

The Envirocare site was not explicitly modeled; however, the sensitivity analysis and one additional set of simulations provided useful information for evaluating the site and suggested that the concentration of ^{235}U at the site is unlikely to be a nuclear safety concern. A site-specific model of the Envirocare site would require more detailed information on hydraulics (e.g., can recharge occur and lead to significant downward migration of solutions?) and geochemistry (e.g., what are the available mobilizing and binding agents? what are the competing complexes?). The present analysis assumes vertical flow through a 10-m column and sorption at the bottom of the column.

For the simulations here, the sorption site concentration was set at a high value to model a worst-case scenario. The CO_3^{2-} concentration was varied. Other selected parameter values presented worst-case scenarios: (1) high pH to dissociate CO_3^{2-} complexation from HCO_3^- and (2) high velocity. However, the total concentration of uranium was modeled with a low value that approximated site-specific conditions. The determined uranium concentration estimated from disposal records tabulated in Appendix B is 9.5×10^{-4} g/cm^3 (8×10^{-3} *M* total U) and the estimated average enrichment is 0.42% or only 4×10^{-6} g/cm^3 (3.3×10^{-5} *M*) ^{235}U.

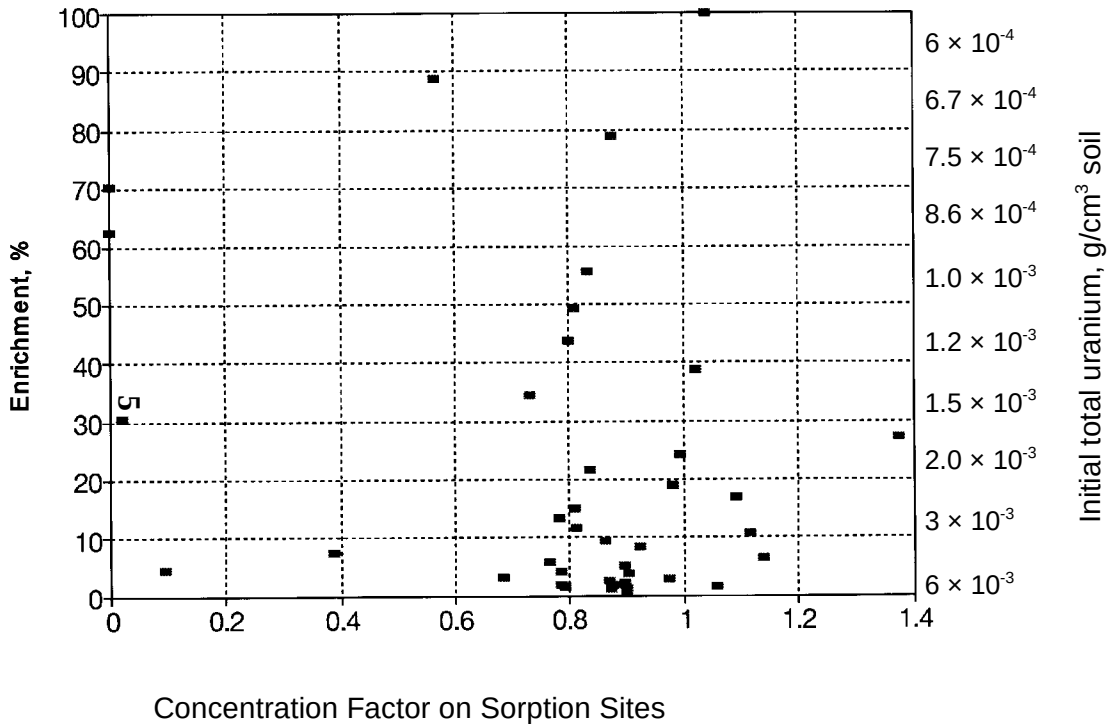


Figure 10.2-6 Initial uranium (in terms of ^{235}U enrichment relative to 6×10^{-4} g/cm^3 limit on ^{235}U) vs CF for uranium on sorption sites. There is no distinct trend because most of the runs showed uranium flushing past the sorption sites

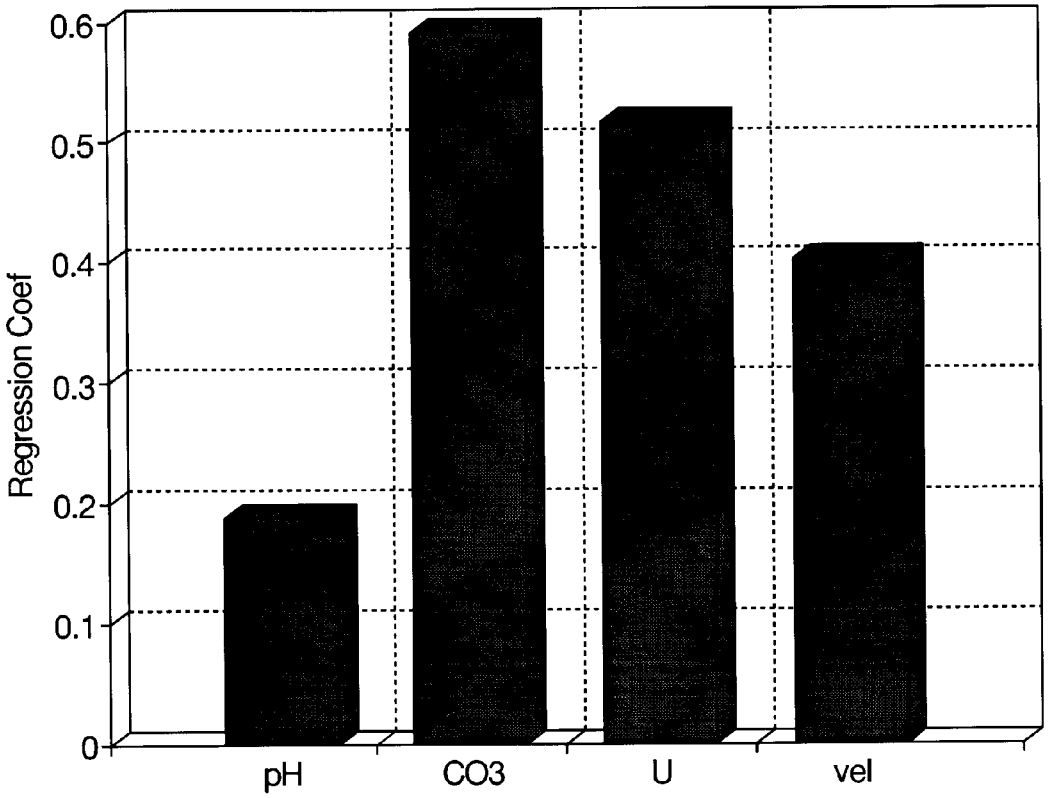


Figure 10.2-7 The absolute values of the regression coefficients for multivariate regression on the CF for mobilization of uranium to a reducing zone shows that several parameters were important in predicting the CF. The height of the bar shows the relative importance of the parameters

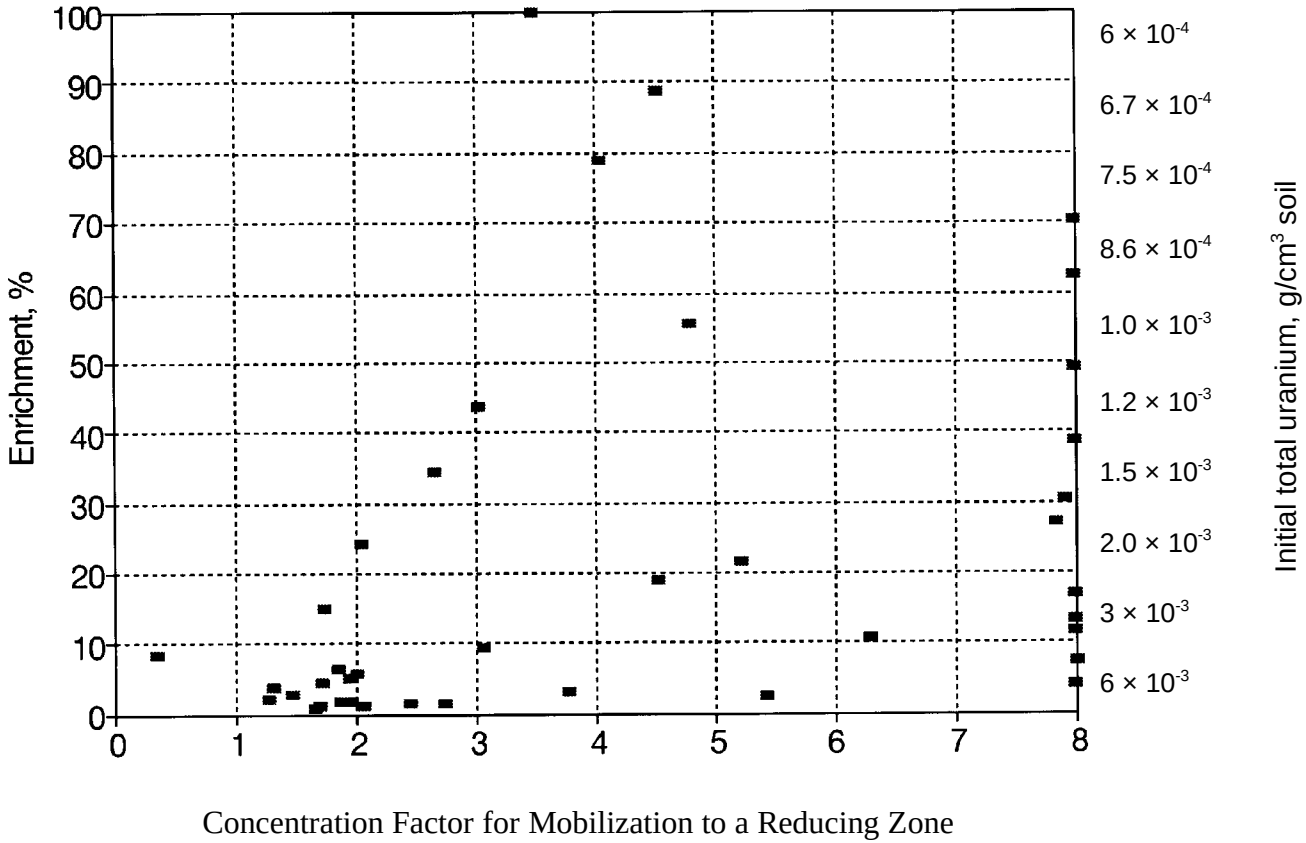


Figure 10.2-8 Initial uranium (in terms of ^{235}U enrichment relative to 6×10^{-4} g/cm^3 limit on ^{235}U) vs CF for mobilization to a reducing zone shows that the CF tends to increase with reduced initial uranium loading (higher enrichment), but there is considerable scatter in the data. This scatter indicates the importance of other parameters as well in predicting the CF for mobilization to a reducing zone. A single parameter did not dominate the trends

Results

The breakthrough curves of uranium concentration in the modeled column (Fig. 10.2-9) showed different peaks for the various concentrations of CO_3^{2-} (the mobilizing agent). The largest peak observed with the reported disposal concentrations was 0.085 *M*. Although this is an increase of a factor of about 10 using the low 0.42% enrichment, the ^{235}U concentration is only 3.6×10^{-4} *M* (or 0.4×10^{-5} g/cm^3). This concentration is about two orders of magnitude below any concentration of concern for nuclear criticality.

For ^{235}U at this initial concentration (4×10^{-6} g/cm^3) to be of concern in criticality safety analysis, the concentration would have to increase to a value between 0.0036 and 0.006 g/cm^3 or a CF (CF) from 900 to 1500. An increase in uranium concentration of this magnitude is not possible. First, it would exceed the sorption capacity of the soil. Secondly, even assuming immobilization by precipitation in a reducing zone, there simply is not a sufficient mass of uranium in a vertical column of the waste cell. For example, line 42 (CF = 833.3) and line 49 (CF = 1666.7) in Table C-1 require areal densities of 28.8 and 38.7 kg/m^2 to reach nuclear criticality.

These areal densities exceed the available source of uranium. Other scenarios can and should be considered, but any analysis will have to overcome the very low initial concentrations of ^{235}U. Scenarios might increase the CF by including precipitation in a reducing zone; however, the site is unsaturated and likely has oxidizing conditions through most of the soil. Scenarios that might also increase the CF include the migration of uranium from multiple directions (e.g., creation of a spherical body) to increase the source mass. In any case, the unsaturated conditions would greatly increase travel time at the site, although behavior during storm events should be studied. As seen from the example given here, the low initial mass is a significant inhibition to the increase in concentration of ^{235}U at this site for any scenario.

10.2.6 Timing

In these simulations, the number of pore volumes required to flush uranium from the oxidized zone varied from 2 to 50. Corresponding travel times are from 1 to 10 years for saturated conditions. For a setting that is typically unsaturated, these fast travel times represent unrealistic, worst-case scenarios. Travel times in the unsaturated zone are assumed to be two to three orders of magnitude slower than for saturated conditions. This assumption results in a range of unsaturated travel times from about 100 to 10,000 years. These estimates contain considerable uncertainty but are comparable to the long times observed for formation of concentration zones in soil (Sect. 4). Velocities drop sharply as soil moisture content drops, but the exact relationship between soil moisture content and velocity is nonlinear and dependent on the particular soil. Furthermore, unsaturated conditions are inherently transient; thus, velocity will vary over time. If a few periods of saturation occur, solutes can move quickly and then remain stable for a period of time. It has been estimated at ORNL that up to 90% of contaminant flushing occurs during brief periods of saturation. Although modeled sorption of uranium did not produce increases in uranium sufficient for criticality, peak uranium concentration was reached at times similar to flushing times.

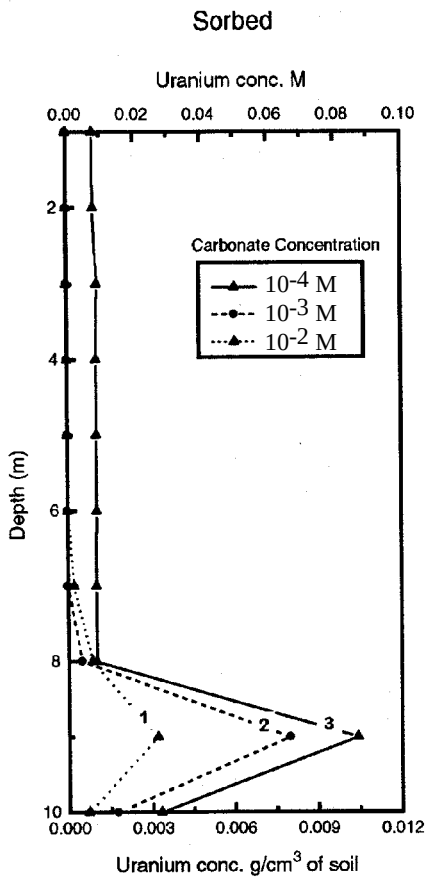


Figure 10.2-9 Results of a scenario with worst-case conditions for sorption (high concentration), but low initial concentration of uranium (based on disposal conditions reported at Envirocare, Utah); breakthrough curve of uranium concentration on sorption sites vs distance for three different carbonate concentrations. The low carbonate concentration of carbonate produces the highest peak. The inflow to the model column is at the top, and outflow is at the bottom

10.3 INTEGRATION OF GEOCHEMICAL MODELING AND CRITICALITY EVALUATION

There are four questions to address in integrating the geochemical modeling and the criticality evaluation:

1. Is the initial mass of uranium sufficient to provide the mass calculated for critical configurations?

2. Are the thicknesses calculated for critical configurations reasonable in comparison to analogs seen in nature?

3. Are the CFs calculated for critical configurations comparable to hydrogeochemical CFs? Do the observed CFs for geochemical processes produce criticality safety concerns?

4. How do the transport rates modeled affect approach to criticality? How does the hypothesized approach to criticality affect the criticality event?

These questions are discussed in the following sections, along with a summary of the implications of the presented work.

10.3.1 Initial Mass of Uranium

The criticality calculations for slab geometry provide a thickness and also a density of uranium needed to reach nuclear criticality. The density of uranium in a critical thickness can be evaluated to consider whether disposal levels provide a sufficient source term. The areal density can be converted to a disposal thickness, assuming a soil concentration of 0.0006 g/cm^3 (Fig. 10.3-2).

The areal density for the nominal soil varied between 4.7 and 8.6 kg/m^2 to attain critical conditions for a CF of 10 (Table C-1). If the dry soil areal density is spread out through a volume of soil to attain a volume density of 0.0006 g/cm^3, then the thickness needs to be greater than 15 m. This is somewhat greater than the average thickness of an Envirocare disposal cell. The average thickness of a disposal cell is approximately 10 m. The cell thickness required as a source for the minimum areal density under wet conditions for CFs of 10 and 33 (Tables C-1 and C-2) is somewhat less than 10 (Fig. 10.4-2). All of the other CFs would require greater thicknesses to provide sufficient uranium for a critical thickness (Fig. 10.3-3). The areal densities for SiO_2 soil tend to be lower; thus they don't require as large a thickness for the initial deposit of uranium. In summary, the disposal thickness limits the range of configurations that can reach a critical concentration, but there is sufficient uranium mass for criticality safety concern.

10.3.2 Calculated Thicknesses

Evaluation of reasonable thicknesses can be addressed through consideration of analogue studies of ore deposits and landfills. For the slab shapes, the calculated thicknesses are less than 2 m (Fig. 10.3-1). Layers of mineral concentration exceeding this thickness are observed in soils and mineral deposits. In landfill construction, liners

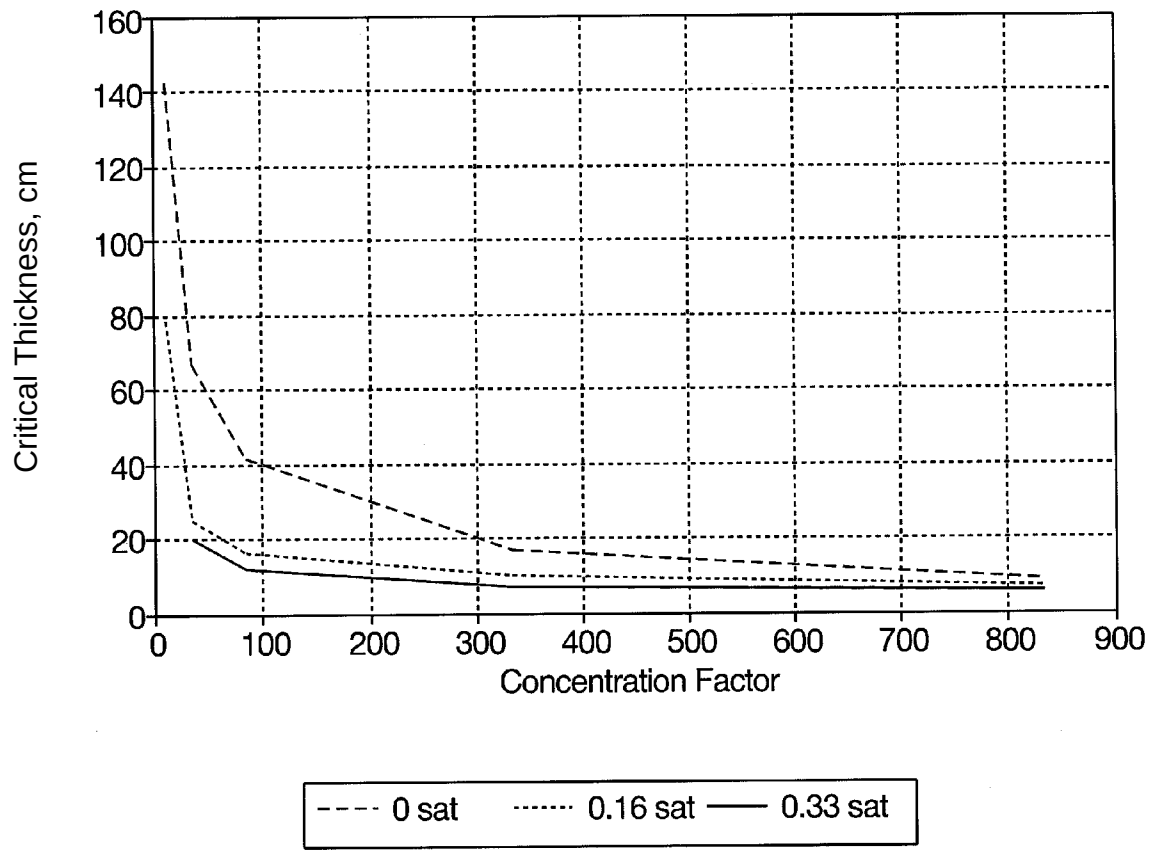


Figure 10.3-1 Calculated thicknesses for slabs with a critical configuration. The figure shows that slabs of less than 2 m (200-cm) in thickness are needed, even for low CFs. Three different water saturation contents are shown; dry configurations require somewhat thicker slabs

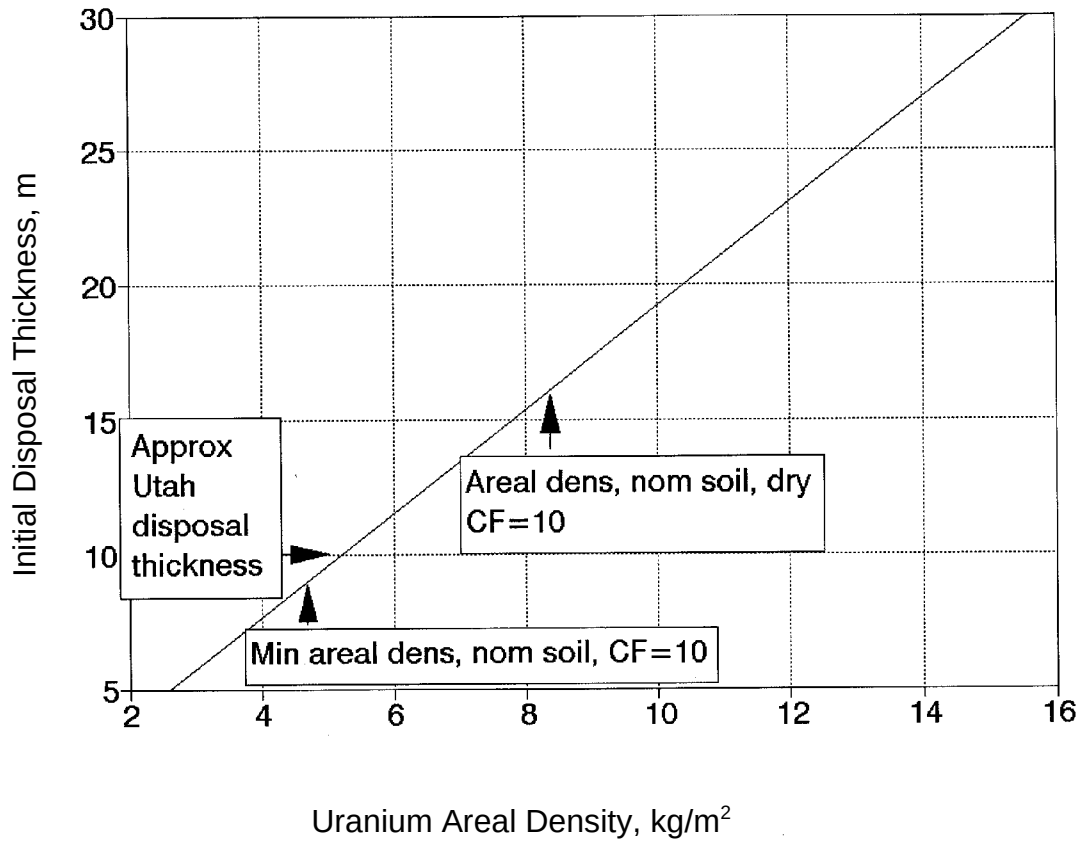


Figure 10.3-2 Initial disposal thickness needed for concentration to a critical slab as a function of uranium areal density assuming 0.0006 g U/cm^3 soil. The figure shows that fairly large disposal-cell thicknesses would be required for many of the critical configurations summarized in Table C-1. This information could be used to place limits on disposal-cell thicknesses

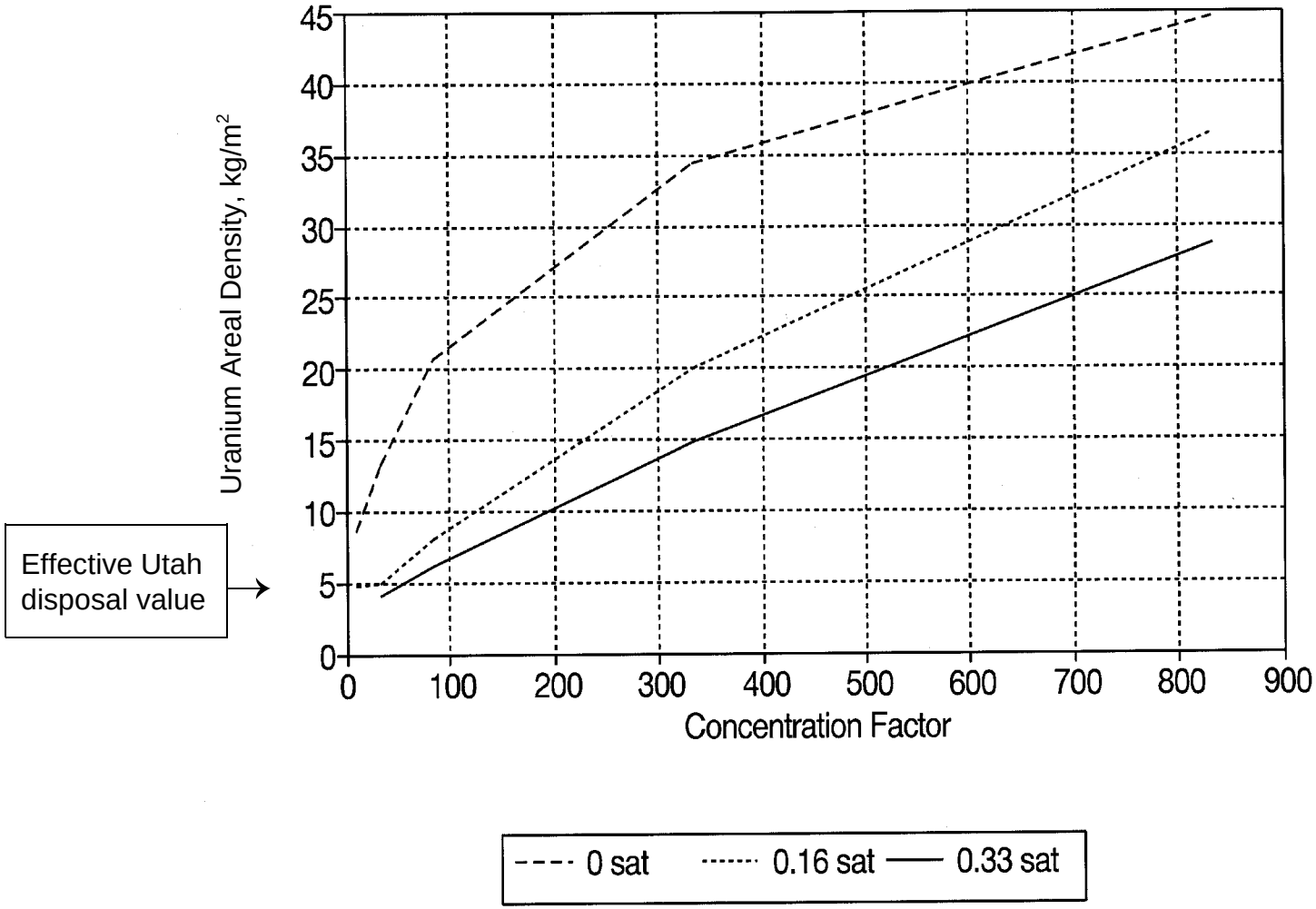


Figure 10.3-3 Uranium areal density for a critical slab (see Table C-1) as a function of CF at various water saturation values. The figure shows that areal density (and hence disposal thickness) increases as the water saturation content decreases, and that it usually increases with the CF. The regulatory disposal limit for Utah is shown to indicate there are some areal densities at or below the limit with potential for criticality safety concern.

of this thickness that create geochemical differentiation could also be found. For cylindrical and spherical shapes, the sizes are somewhat larger than observed bodies in soil-forming processes.

10.3.3 Comparison of Concentration Factors

Two geochemical processes were considered: concentration in a zone of higher sorption and concentration in a redox zone. The modeled processes were sorption and flushing of the uranium.

For the case of sorption, the modeled CFs did not produce critical configurations. The CFs were less than 2 for all cases modeled. The mobilization of the uranium overwhelmed the sorption process.

However, mobilization of uranium can lead to precipitation in reducing zones, if they exist. The extent of mobilization calculated by the models provides a limit to the concentration in the reducing zone. Mobilization and redeposition of uranium uniformly distributed in an 8-m thickness of contaminated soil into a 1-m-thick reducing zone results in a geometric maximum CF of 8. If uranium is deposited instead over a 2-m-thick zone, then a maximum CF is 4. Other waste thicknesses can also be considered. The case modeled here is an 8-m-thick section of waste and a hypothetical 1-m-thick reducing zone. For 12 of the 40 runs evaluated, sufficient uranium is flushed to reach CFs between 6 and the geometric maximum of 8. The other runs did not flush sufficient uranium to reach CFs that could attain criticality within the model time.

10.3.4 Evaluation of Results

One additional question that needs to be addressed is what is the probability that a criticality event would occur? This question cannot be addressed quantitatively with the present data. However, some general sense about confidence can be gained by evaluating the different states of modeling and events. The first stage to consider is the model processes that lead to an increase in uranium concentration. The present modeling indicates that a narrow set of conditions would lead to significant concentration increases. This narrow set of conditions may imply a low probability for occurrence, but site-specific data would be the key to evaluating probability. The probability could be zero at some sites. Hence, there is a high degree of uncertainty in this modeling because of gaps in data and possible large variations between specific sites. For the criticality calculation, there is also a narrow set of conditions that lead to a criticality event when geologically reasonable configurations are considered. The calculations have a high degree of certainty when evaluated from a mathematical viewpoint, but there is again uncertainty in whether the full range of natural conditions has been considered. For instance, concentration regions have been assumed to have sharp boundaries, and moisture content has been assumed to be uniform. Natural variations are likely to occur, which have not yet been evaluated. A third stage of releasing fissile uranium that presents a dose health threat could occur before or after a criticality event.

10.3.5 Summary of Criticality Potential

Although only a small number of cases reached CFs of concern, these cases modeled realistic ranges of geochemical conditions. Further analysis should examine the realism of the hydraulic conditions and any geochemical factors that have been neglected that could inhibit increases in concentration (e.g., competing complexes that prevent mobilization or inhibit precipitation). At Envirocare, the reported low initial mass of ^{235}U and the low enrichment were important considerations for ensuring subcriticality.

Because many hydrogeochemical simulations do not present CFs of concern, these examples also suggest measures that can be taken to inhibit increases in concentration of uranium. Limiting water content can prevent fast flow and help maintain oxidizing conditions (avoid local reducing zones). Limiting the enrichment increases the total amount of uranium that must be mobilized, and in some cases nonfissile uranium limits the CFs attained. Limitations on the source available for mobilization through the disposal-cell dimensions (related to the density of uranium in critical configurations) can also be calculated.

10.4 CRITICALITY CONSEQUENCES: DIRECT DOSE FROM RADIATION TRANSPORT

The purpose of this study was to evaluate whether or not fissile uranium concentration in LLW facilities can be increased by hydrogeochemical processes to permit nuclear criticality. Criticality is a precise condition wherein the rate of neutron production, excluding neutron sources whose strengths are not a function of fission rate, is equal to the rate of neutron loss. Because criticality is affected by delayed neutrons (neutrons that are produced at delayed times following the fission process), it is possible to change from subcritical to supercritical through the delayed neutron contribution. However, a very slow approach to criticality can produce substantial, nearly infinite, neutron multiplication as revealed in the subcritical relationship for neutron source multiplication, $M_n \approx [1/(1 - k_{\infty, eff})]$. This is to say that whatever inherent neutron source is present (e.g., spontaneous fission from ^{235}U – 6.6×10^{-1} neutrons/kg-min, ^{238}U – 8.1×10^{2} neutrons/kg-min, or other nuclides present) would be multiplied by M_n. Depending on the initial neutron flux density within the system at the time of an increase in system reactivity from an action such as the slumping of a deposit, the criticality “switch” could be “snapped on” resulting in rapid fission heating and feedbacks that tend to expand and “turn off” the criticality.

An element in the depletion of the critical condition is whether or not water moderation is required for criticality. If so, approximately 10^{16} fissions are required to raise 1 L of water from room to boiling temperature, and a total of about 10^{17} fissions are required to vaporize 1 L of water. If criticality were approached from an overly water-moderated condition, it is conceivable that a fairly rapid fission process would occur until sufficient water is removed, via evaporative steam or physical expulsion, to render the system subcritical. Such energy releases could mimic geyser behavior cycling over longer time periods or a single steam explosion within a few tenths of a second. For the purposes of the direct-dose radiation transport evaluations, the “first-pulse” fission yields correspond to the thermal fission energy required to remove that quantity of water/moisture providing the over-moderated condition. Therefore, further migration of water back into such a deposit could result in further fissions for extended periods—well beyond the original estimated fission yields. The radioactive-decay half-life of ^{235}U is 7×10^{8} years. Depending upon the beginning enrichment and the excess mass of ^{235}U available to continue to contribute to the fission process, radioactive decay in geological time may or may not be a contributor to the termination of criticality. A large number of scenarios preventing or supporting criticality may be postulated.

Postulating that a criticality event could occur at an LLW site, the immediate consequences of such an event to the public were considered as part of this study. Using the models, methods, and sources described in Sect. 7.3, the dose results under the assumed fission yields for the high concentration factor (HCF) case (i..e, 4.1×10^{21}

fissions) and the low concentration factor (LCF) case (i.e., 2.8×10^{21} fissions) were determined. The results are shown in Table 10.4-1 for all conditions analyzed.

Results

Table 10.4-1 Dose results for assumed high and low CFs at locations near and far from potential critical deposits

High or low concentration	Distance (m) from deposit center[a]	Neutron dose (rem)[b]	Gamma-ray dose (rem)	Total dose (rem)
High	0	34.4	21.7	56.1
Low	0	98.6	58.8	157.4
High	90	0.045	0.012	0.057
Low	90	0.131	0.034	0.165

[a]All detector locations assumed to be located 1 m above ground.
[b]ANSI/ANS-6.1.1-1977, 1977.

These results show that total doses are higher for the LCF case than for the HCF case by about a factor of 3. The total dose-per-fission values (not shown) for the LCF are about a factor of 4 higher than for the HCF. This higher dose is because the LCF deposit is thicker and hence closer to the ground surface. This higher LCF dose per fission is somewhat offset by the LCF fission yield, which is about 30% smaller than the HCF fission yield.

Also shown in Table 10.4-1 are the neutron and gamma-ray dose components. These values sum to the total dose and are given primarily for completeness. Note that the neutron doses near the deposit account for about 60% of the total dose for both HCF and LCF cases, while the neutrons comprise about 80% of the total dose at the 90-m location. Thus it appears that the neutron dose portion of the total dose increases with distance away from the deposit, although the total dose decreases at a rapid rate with increasing distance from the axis of the critical deposit.

11 DISCUSSION OF LIMITATIONS ASSOCIATED WITH THE ANALYSIS

Because of the broad parametric phase space of the study (i.e., soil types, soil densities, range of ^{235}U contamination density in soil, degree of water moderation, and geometric configurations) and various combinations of the parameters, the study was confined to the grid of parameters used in the nuclear criticality evaluation (Tables 10.1-1 and 10.1-2). The parameter grid was selected to span the parametric ranges of concern but was, by necessity, limited in the number of cases evaluated. The results of this scoping study provide general bounds for parametric combinations of criticality safety concern. For the simple case of vertical migration, a prior study (Hopper et al., 1995) of SNM burial limits should be considered for addressing licensing issues.

The overall limitations of the geochemical transport modeling were the necessary simplifications to develop a model and the many assumptions and estimates that had to be made for unknown parameters. A large number of unknowns existed because of the complexity of the problem. These uncertainties ranged from the hydrologic conditions (moisture content, velocities, and inlets and outlets of the system) to geochemical conditions. Many complexes and minerals can be considered for this problem. However, the thermodynamic database quality is sometimes questionable, which can limit consideration of some complexes and some potentially important scenarios (e.g., involving organic complexation). A further problem is that coupled models are time consuming to debug and run. In particular, simulations evaluating variable redox conditions present calculation convergence problems because of the extreme concentration ranges involved. The 1-D flow-field modeled here limits analysis to formation of slabs in a single disposal cell.

Despite these limitations, this work is an important first step toward gaining an understanding of the principles of study. Starting with simple cases was important to determine constraining factors. The range in possible conditions of concern can be narrowed by further study of the key factors identified here. Some additional tasks for future work include the following:

- conduct criticality calculations for other soil matrices suggested at specific sites;
- conduct criticality calculations for plutonium;
- search for databases to evaluate the potential to increase plutonium concentration;
- search for databases to evaluate organic complexation/colloids;
- consider other mobilizing agents, such as colloids;
- further consider redox scenarios to examine the stability of redox fields;
- consider lateral flow to transport uranium from several disposal cells to a zone of increased concentration beneath the cells;
- consider precipitation under oxidizing conditions by evaluating geochemical factors that could change to create precipitating and nonprecipitating zones;
- model saturated and unsaturated conditions; and

- conduct laboratory or field work to evaluate site conditions: study thermodynamics, zonation, and saturation conditions.

12 SUMMARY AND RECOMMENDATIONS

Nuclear criticality evaluations based upon licensed soil-contamination limits specified for Envirocare reveal a theoretical possibility of a nuclear criticality accident, given reasonable soil assumptions and hydrogeochemical influences on the concentration of SNM, specifically ^{235}U. The length of time required to concentrate uranium is expected to be long (e.g., many years). This slow approach to criticality will further mitigate rapid approaches to critical or supercritical conditions.

However, reviews of disposal/burial records from Envirocare reveal that concentrations of ^{235}U in the waste material are more than a factor of 10 less than licensed concentrations of ^{235}U and that the average enrichment is below the minimum 1% required to achieve nuclear criticality. Thus the likelihood of a criticality accident is vanishingly small.

The results presented here identify important factors in uranium concentration within a disposal cell, for example, the concentration of the mobilizing agent (CO_3^{2-} in our simulations) and the size (thickness) of the source term. Although these processes were modeled under saturated conditions, the study estimates that relatively long time frames (perhaps thousands of years) would be needed to concentrate uranium under unsaturated conditions. Much uncertainty exists in these temporal estimates because soil conditions have not been explicitly modeled and can vary both spatially and temporally. Analogs provided by studies of soil-forming processes suggest that increasing the concentration is a long-term process (e.g., requiring thousands of years).

This study results in the following recommendations for consideration of license review of LLW facilities.

1. Minimize those factors that enhance the concentration of uranium.

    - Reduce water infiltration. Water increases mobilization and transport rates; unsaturated conditions greatly reduce opportunities for concentration.

    - Where possible, reduce enrichment of ^{235}U. The presence of nonfissile uranium decreases the extent of ^{235}U mobilization. The regulations do not specify any enrichment factors, so 100% enrichment is permissible.

    - Minimize opportunities to create isolated zones of extreme reducing potential if possible because uranium precipitates readily under reducing conditions. Unsaturated conditions would tend to create more oxidizing environments, but further study is needed to determine the importance of microenvironments within disposal cells. Also, avoid organic matter in waste cells to prevent methanogenesis.

2. Limit the areal density of uranium by limiting the depth of the disposal cell. Results demonstrate that criticality concerns can be minimized even if worst-case hydrogeochemical transport and concentration mechanisms are assumed. If the depth of the disposal cell is limited so that there is not a sufficient mass of ^{235}U within a cross section of a vertical flow pathway, criticality cannot occur.

The simplified models used in this study have brought researchers one step closer to understanding the potential for criticality at an LLW facility using conservative but realistic conditions. The approach followed here, integrating hydrogeochemical modeling and nuclear criticality safety expertise, provides a rationale and basis for formulating rule changes to minimize criticality safety concerns.

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GLOSSARY

Term	Discussion
Areal density	Mass per unit area used to characterize the ^{235}U contaminant of an infinite slab of soil.
Concretions	Nodular or irregular mineral concentration in sedimentary rocks, developed by the localized deposition of material from solution.
Effective neutron multiplication factor, k_{eff}	Ratio of the total number of neutrons produced during a time interval to the total number of neutrons lost by absorption and leakage during the same interval.
Infinite cylinder	Conceptually, a cylinder of finite diameter but of infinite length.
Infinite media	Conceptually, material that fills an infinite volume.
Infinite slab	Conceptually, a slab of finite thickness but of infinite length and width.
Infinite neutron multiplication factor, k_∞	Ratio of the total number of neutrons produced during a time interval to the total number of neutrons lost by absorption during the same interval.
Lift	Layer of loose, uncompacted waste not exceeding a thickness of 0.3 m that is mechanically manipulated to ensure uniform density. Thinner lifts are required if necessary to meet compaction requirements. The minimum surface area for a lift is 929 m^2 (10,000 ft^2).
Linear density	Mass per unit length used to characterize the ^{235}U contaminant of an infinitely long cylinder of soil.
M_n	Neutron source multiplication factor for subcritical fissile material systems. $M_n \approx [1/(1 - k_{\infty, eff})]$.
Moderation	Reduction of neutron energy by scattering neutrons without appreciable neutron capture by neutron scattering material.
Neutron multiplication factor, k	Ratio of the total number of neutrons produced during a time interval to the total number of neutrons lost during same interval.
Nuclear criticality	Condition of being critical. For special nuclear material systems, the condition when the rate of neutron production, excluding neutron sources whose strengths are not a function of fission rate, is equal to the rate of neutron loss; k_{eff} or $k_{eff} = 1.0$. For studies in this report, 0.95, rather than 1.0, was selected as a critical value to conservatively account for materials and data uncertainty.
Reflection	Scattering of neutrons back to a system that would otherwise escape from the system.
Subcritical (subcriticality)	For special nuclear material systems, the condition when the rate of neutron production, excluding neutron sources whose strengths are not a function of fission rate, is less than the rate of neutron loss; k_{eff} or $k_{eff} < 1.0$. For studies in this report, values less than 0.95, rather than less than 1.0, are assumed to conservatively account for materials and data uncertainty.
Uranium concentration	Ratio of uranium mass to soil or water mass.

Term	Discussion
Uranium concentration factor (CF)	Ratio of the uranium concentration (resulting from a uranium densification or dilution process) divided by the original, authorized/licensed uranium concentration within the soil or waste matrix. In this report, 0.0006 g/cm^3 is used as the original ^{235}U concentration.
Uranium enrichment	Mass ratio of the ^{235}U isotope of uranium to total mass of uranium typically expressed in weight percent ^{235}U.

APPENDIX A

Conceptual Configurations for Nuclear Criticality Evaluations

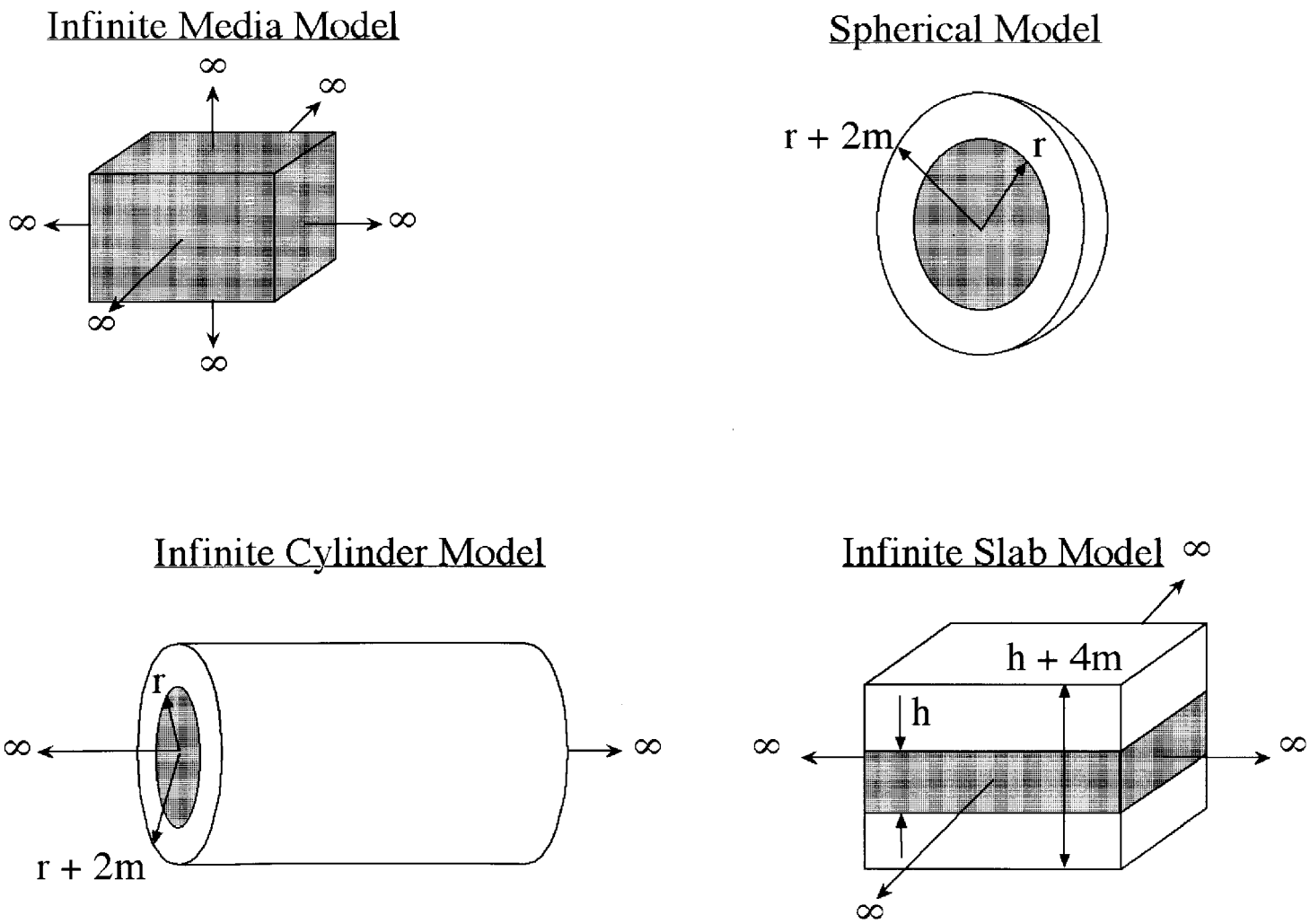

Figure A.1-1 Conceptual configurations for nuclear criticality evaluations

APPENDIX B

Envirocare of Utah Disposal Records

Table B-1 Envirocare of Utah disposal records, mCi

Year	DU[a]	NU[b]	^{234}U	^{235}U	^{238}U	^{236}U
1988		622				
1989		22.8				
1990		7.94				
1991	7450	10				
1992	4170	17900	1490	82	643	
1993	16300	1620	1200	55.7	324	1.24
TOT	27920	20182.7	2690	137.7	967	1.24

[a]Depleted uranium (< 0.7 wt % ^{235}U in total uranium).
[b]Normal uranium (0.7 wt % ^{235}U in total uranium).

Table B-2 Envirocare of Utah disposal records, grams,and volumetric

Year	DU[a]	NU[b]	^{234}U	^{235}U	^{238}U	^{236}U	Total U	Total ^{235}U[c]	Enrichment, %	Volume yards^3	g $^{235}U/cm^3$	Total g U/cm^3
1988		883240					883240	6359.328	0.72	5610	1.48E-06	0.000206
1989		32376					32376	233.1072	0.72	26200	1.16E-08	1.62E-06
1990		1127438					11274.8	81.17856	0.72	46900	2.26E-09	3.14E-07
1991	24808500	14200					24822700	67085.19	0.270257	68000	1.29E-06	0.000477
1992	13886100	25418000	239.89	37966	1916140		41258446	258468.1	0.626461	197000	1.72E-06	0.000274
1993	54279000	2300400	193.2	25789.1	965520	19.22	57570922	188905.3	0.328126	74100	3.33E-06	0.001016
Total							1.25E+08	521132.2	0.418315	417810		
g/mCi[d]	3330	1420	0.161	463	2980	15.5						

[a]Depleted uranium (< 0.7 wt % ^{235}U in total uranium, 0.27 wt % ^{235}U in total uranium assumed).
[b]Normal uranium (0.7 wt % ^{235}U in total uranium).
[c]Total ^{235}U is the sum of 0.0027(DU) + 0.007(NU) + ^{235}U.
[d]Factors used for converting reported mCi in Table B-1 to g.

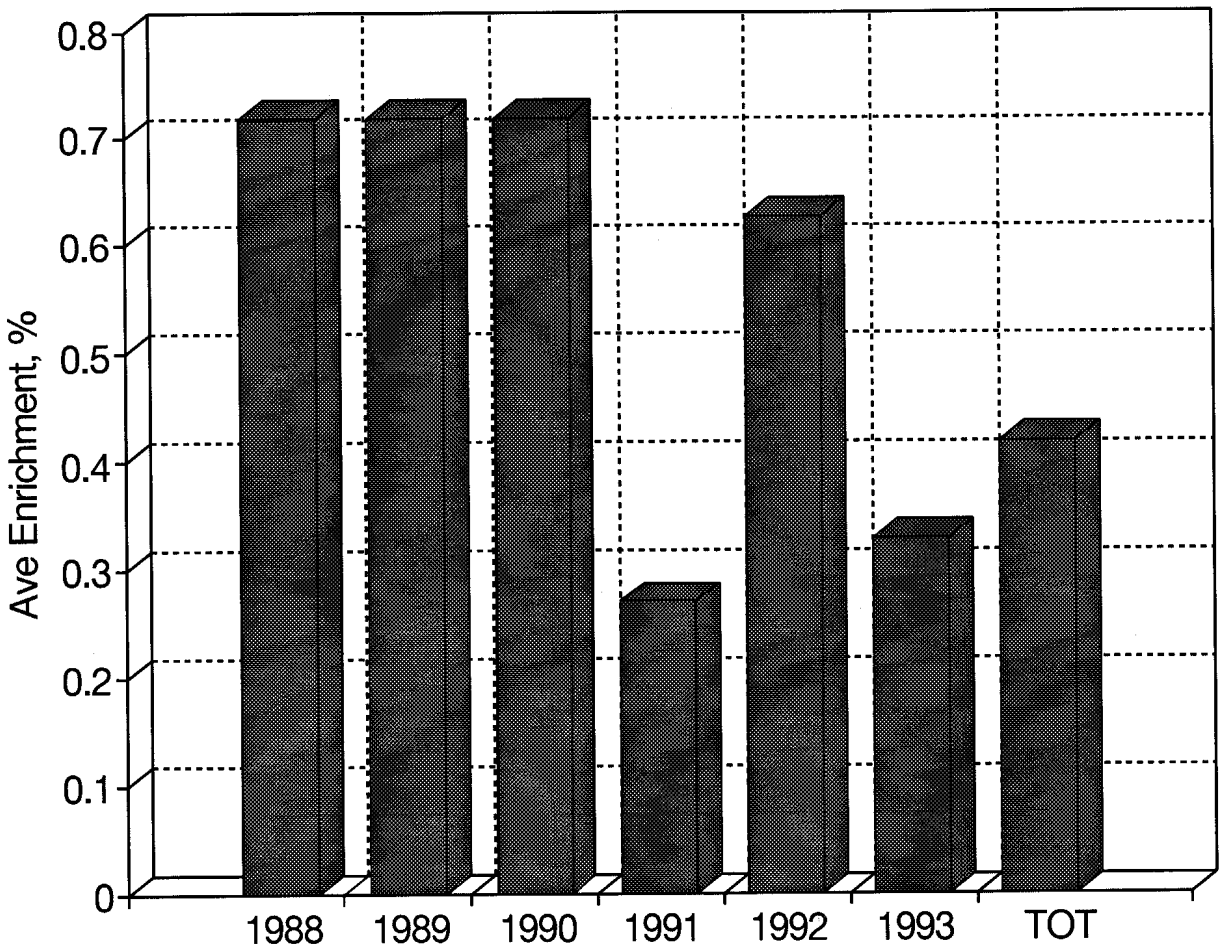

Figure B.1-1 Yearly and six-year average (TOT) ^{235}U enrichment of uranium disposed of at Envirocare of Utah

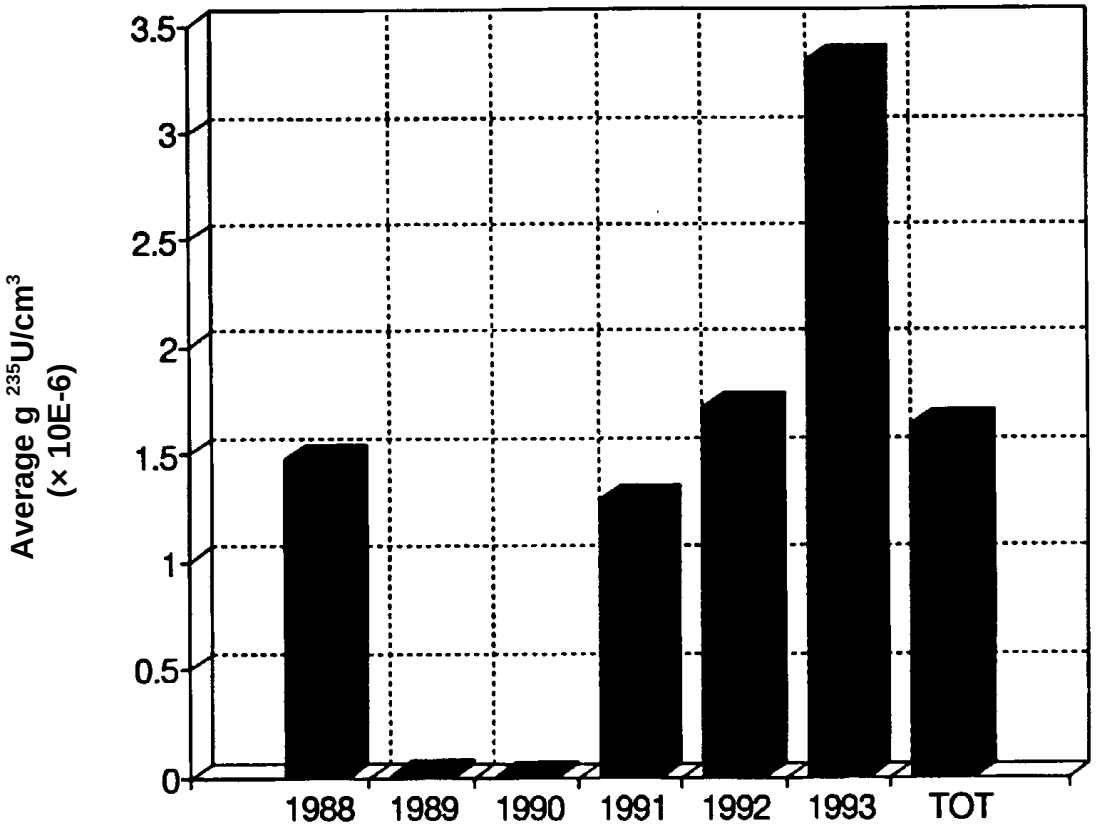

Figure B.1-2 Yearly and six-year average (TOT) of the concentration of ^{235}U disposed of at Envirocare of Utah

APPENDIX C

Expanded Tables 10.1-1 and 10.1-2

Table C-1 Nominal-soil (N-S) results[a]

Line entry	Conc. factor	^{235}U content		Water content		k_∞ or k-infinity	Critical infinite slab[b]		Critical infinite cylinder[b]		Critical sphere[b]	
		$^{235}U/cm^3$ g	^{235}U/g N-S g	H_2O/cm^3 g	H_2O/g N-S g		Thickness (cm)	^{235}U areal density (kg/m^2)	Diameter (cm)	^{235}U linear density (kg/m)	Diameter (cm)	^{235}U mass (kg)
						Fig. D-1	Fig. D-2	Fig. D-3	Fig. D-4	Fig. D-5	Fig. D-6	Fig. D-7
1	1.0	0.0006	0.000375	0.000	0.000000	0.357						
2	1.0	0.0006	0.000375	0.020	0.012500	0.338						
3	1.0	0.0006	0.000375	0.040	0.025000	0.317						
4	1.0	0.0006	0.000375	0.080	0.050000	0.281						
5	1.0	0.0006	0.000375	0.160	0.100000	0.228						
6	1.0	0.0006	0.000375	0.330	0.206250	0.163						
7	3.3	0.0020	0.001250	0.000	0.000000	0.825						
8	3.3	0.0020	0.001250	0.020	0.012500	0.803						
9	3.3	0.0020	0.001250	0.040	0.025000	0.770						
10	3.3	0.0020	0.001250	0.080	0.050000	0.706						
11	3.3	0.0020	0.001250	0.160	0.100000	0.602						
12	3.3	0.0020	0.001250	0.330	0.206250	0.457						
13	10.0	0.0060	0.003750	0.000	0.000000	1.292	142.950	8.577	270.140	34.389	382.220	175.424
14	10.0	0.0060	0.003750	0.020	0.012500	1.310	101.720	6.103	192.520	17.466	272.700	63.710
15	10.0	0.0060	0.003750	0.040	0.025000	1.292	88.100	5.286	165.280	12.873	233.480	39.985
16	10.0	0.0060	0.003750	0.080	0.050000	1.239	78.040	4.682	143.000	9.636	200.600	25.360
17	10.0	0.0060	0.003750	0.160	0.100000	1.131	78.860	4.732	137.920	8.964	190.040	21.562
18	10.0	0.0060	0.003750	0.330	0.206250	0.946						
19	33.3	0.0200	0.012500	0.000	0.000000	1.529	66.680	13.336	157.080	38.758	236.220	138.032
20	33.3	0.0200	0.012500	0.020	0.012500	1.625	46.670	9.334	110.800	19.284	167.120	48.878
21	33.3	0.0200	0.012500	0.040	0.025000	1.652	38.710	7.742	91.540	13.163	138.160	27.617
22	33.3	0.0200	0.012500	0.080	0.050000	1.657	31.180	6.236	72.360	8.225	108.820	13.494
23	33.3	0.0200	0.012500	0.160	0.100000	1.619	24.970	4.994	55.540	4.845	82.640	5.910
24	33.3	0.0200	0.012500	0.330	0.206250	1.506	20.360	4.072	42.160	2.792	61.520	2.438
25	83.3	0.0500	0.031250	0.000	0.000000	1.567	41.420	20.710	119.380	55.966	187.440	172.407
26	83.3	0.0500	0.031250	0.020	0.012500	1.663	31.160	15.580	88.620	30.841	138.620	69.734
27	83.3	0.0500	0.031250	0.040	0.025000	1.717	26.010	13.005	73.500	21.215	114.820	39.630
28	83.3	0.0500	0.031250	0.080	0.050000	1.766	20.740	10.370	57.500	12.984	89.600	18.832
29	83.3	0.0500	0.031250	0.160	0.100000	1.788	16.050	8.025	42.660	7.147	66.120	7.568
30	83.3	0.0500	0.031250	0.330	0.206250	1.759	12.100	6.050	29.860	3.501	45.680	2.495
31	333.3	0.2000	0.125000	0.000	0.000000	1.693	17.210	34.420	74.980	88.310	126.300	210.979
32	333.3	0.2000	0.125000	0.020	0.012500	1.672	16.120	32.240	64.100	64.541	106.120	125.147
33	333.3	0.2000	0.125000	0.040	0.025000	1.687	14.670	29.340	56.180	49.577	92.340	82.451

Table C-1 (continued)

Line entry	Conc. factor	^{235}U content		Water content		k_∞ or k-infinity	Critical infinite slab[b]		Critical infinite cylinder[b]		Critical sphere[b]	
		$^{235}U/cm^3$ g	$^{235}U/g$ N-S g	H_2O/cm^3 g	H_2O/g N-S g		Thickness (cm)	^{235}U areal density (kg/m^2)	Diameter (cm)	^{235}U linear density (kg/m)	Diameter (cm)	^{235}U mass (kg)
						Fig. D-1	Fig. D-2	Fig. D-3	Fig. D-4	Fig. D-5	Fig. D-6	Fig. D-7
34	333.3	0.2000	0.125000	0.080	0.050000	1.730	12.520	25.040	45.660	32.749	74.440	43.196
35	333.3	0.2000	0.125000	0.160	0.100000	1.790	10.010	20.020	34.140	18.308	55.100	17.518
36	333.3	0.2000	0.125000	0.330	0.206250	1.847	7.410	14.820	23.240	8.484	37.020	5.313
37	833.3	0.5000	0.312500	0.000	0.000000	1.808	8.310	41.550	51.560	104.396	91.640	201.476
38	833.3	0.5000	0.312500	0.020	0.012500	1.763	8.910	44.550	47.560	88.827	82.060	144.665
39	833.3	0.5000	0.312500	0.040	0.025000	1.741	8.820	44.100	43.800	75.337	74.640	108.864
40	833.3	0.5000	0.312500	0.080	0.050000	1.731	8.330	41.650	37.800	56.110	63.560	67.223
41	833.3	0.5000	0.312500	0.160	0.100000	1.750	7.310	36.550	29.820	34.920	49.380	31.523
42	833.3	0.5000	0.312500	0.330	0.206250	1.800	5.760	28.800	20.860	17.088	33.980	10.272

[a] Figures are presented in Appendix D.
[b] Dimensions based on system $k_{eff} = 0.95$ rather than $k_{eff} = 1.0$ to conservatively account for methods and data uncertainty.

Table C-2 SiO_2-soil (S-S) results[a]

Line entry	Conc. factor	^{235}U content $^{235}U/cm^3$ g	^{235}U content $^{235}U/gS\text{-}S$ g	Water content H_2O/cm^3 g	Water content $H_2O/gS\text{-}S$ g	k_∞ or k-inf	Critical infinite slab[b] Thick-ness (cm)	Critical infinite slab[b] ^{235}U areal density (kg/m^2)	Critical infinite cylinder[b] Diameter (cm)	Critical infinite cylinder[b] ^{235}U linear density (kg/m)	Critical sphere Diameter (cm)	Critical sphere ^{235}U mass (kg)
						Fig. D-8	Fig. D-9	Fig. D-10	Fig. D-11	Fig. D-12	Fig. D-13	Fig. D-14
1	0.83	0.0005000	0.0003125	0.00000	0.00000	0.497						
2	0.83	0.0005000	0.0003125	0.05800	0.03625	0.370						
3	0.83	0.0005000	0.0003125	0.11900	0.07438	0.288						
4	0.83	0.0005000	0.0003125	0.18300	0.11438	0.234						
5	0.83	0.0005000	0.0003125	0.25100	0.15688	0.195						
6	0.83	0.0005000	0.0003125	0.32400	0.20250	0.166						
7	0.83	0.0005000	0.0003125	0.40000	0.25000	0.143						
8	2.36	0.0014179	0.0008862	0.00000	0.00000	0.955	2131.7200	30.2257	3351.58	1250.9348	4540.5	69495.2729
9	2.36	0.0014179	0.0008862	0.02900	0.01813	0.867						
10	2.36	0.0014179	0.0008862	0.05800	0.03625	0.783						
11	2.36	0.0014179	0.0008862	0.08850	0.05531	0.710						
12	2.36	0.0014179	0.0008862	0.11900	0.07438	0.648						
13	2.36	0.0014179	0.0008862	0.15100	0.09438	0.594						
14	2.36	0.0014179	0.0008862	0.18300	0.11438	0.548						
15	2.36	0.0014179	0.0008862	0.21700	0.13563	0.506						
16	2.36	0.0014179	0.0008862	0.25100	0.15688	0.471						
17	2.36	0.0014179	0.0008862	0.28750	0.17969	0.437						
18	2.36	0.0014179	0.0008862	0.32400	0.20250	0.409						
19	2.36	0.0014179	0.0008862	0.36200	0.22625	0.382						
20	2.36	0.0014179	0.0008862	0.40000	0.25000	0.359						
21	2.72	0.0016300	0.0010188	0.00000	0.00000	1.021	481.5000	7.8485	791.82	80.2658	1067.33	1037.7252
22	2.72	0.0016300	0.0010188	0.02900	0.01813	0.936						
23	2.72	0.0016300	0.0010188	0.05800	0.03625	0.851						
24	2.72	0.0016300	0.0010188	0.08850	0.05531	0.775						
25	2.72	0.0016300	0.0010188	0.11900	0.07438	0.711						
26	2.72	0.0016300	0.0010188	0.15100	0.09438	0.654						
27	2.72	0.0016300	0.0010188	0.18300	0.11438	0.606						
28	2.72	0.0016300	0.0010188	0.21700	0.13563	0.561						
29	2.72	0.0016300	0.0010188	0.25100	0.15688	0.523						
30	2.72	0.0016300	0.0010188	0.28750	0.17969	0.487						
31	2.72	0.0016300	0.0010188	0.32400	0.20250	0.456						
32	2.72	0.0016300	0.0010188	0.36200	0.22625	0.427						
33	2.72	0.0016300	0.0010188	0.40000	0.25000	0.402						
34	3.00	0.0011250	0.0011250	0.00000	0.00000	1.060	367.1300	6.6083	621.5	54.6065	844.74	355.0752
35	3.00	0.0018000	0.0011250	0.05800	0.03625	0.894						
36	3.00	0.0018000	0.0011250	0.11900	0.07438	0.751						
37	3.00	0.0018000	0.0011250	0.18300	0.11438	0.642						
38	3.00	0.0018000	0.0011250	0.25100	0.15688	0.556						
39	3.00	0.0018000	0.0011250	0.32400	0.20250	0.487						
40	3.00	0.0018000	0.0011250	0.40000	0.25000	0.431						
41	3.12	0.0018738	0.0011711	0.00000	0.00000	1.086	321.3800	6.0220	547.78	44.1596	748.13	410.8214
42	3.12	0.0018738	0.0011711	0.02900	0.01813	1.005	358.7500	6.7223	585.18	50.3955	785.23	475.0207
43	3.12	0.0018738	0.0011711	0.05800	0.03625	0.920						

Table C-2 (continued)

Line entry	Conc. factor	^{235}U content $^{235}U/cm^3$ g	^{235}U content $^{235}U/gS\text{-}S$ g	Water content H_2O/cm^3 g	Water content $H_2O/gS\text{-}S$ g	k_∞ or k-inf	Critical infinite slab[b] Thick-ness (cm)	Critical infinite slab[b] ^{235}U areal density (kg/m^2)	Critical infinite cylinder[b] Diameter (cm)	Critical infinite cylinder[b] ^{235}U linear density (kg/m)	Critical sphere[b] Diameter (cm)	Critical sphere[b] ^{235}U mass (kg)
						Fig. D-8	Fig. D-9	Fig. D-10	Fig. D-11	Fig. D-12	Fig. D-13	Fig. D-14
44	3.12	0.0018738	0.0011711	0.08850	0.05531	0.843						
45	3.12	0.0018738	0.0011711	0.11900	0.07438	0.777						
46	3.12	0.0018738	0.0011711	0.15100	0.09438	0.717						
47	3.12	0.0018738	0.0011711	0.18300	0.11438	0.666						
48	3.12	0.0018738	0.0011711	0.21700	0.13563	0.620						
49	3.12	0.0018738	0.0011711	0.25100	0.15688	0.579						
50	3.12	0.0018738	0.0011711	0.28750	0.17969	0.541						
51	3.12	0.0018738	0.0011711	0.32400	0.20250	0.507						
52	3.12	0.0018738	0.0011711	0.36200	0.22625	0.477						
53	3.12	0.0018738	0.0011711	0.40000	0.25000	0.449						
54	3.59	0.0021540	0.0013463	0.00000	0.00000	1.149	248.1100	5.3443	436	32.1594	602.2	246.3012
55	3.59	0.0021540	0.0013463	0.02900	0.01813	1.074	218.2200	4.7005	370.78	23.2578	504.94	145.1990
56	3.59	0.0021540	0.0013463	0.05800	0.03625	0.990	344.4100	7.4186	556.18	52.3319	741.84	460.4421
57	3.59	0.0021540	0.0013463	0.08850	0.05531	0.912						
58	3.59	0.0021540	0.0013463	0.11900	0.07438	0.844						
59	3.59	0.0021540	0.0013463	0.15100	0.09438	0.783						
60	3.59	0.0021540	0.0013463	0.18300	0.11438	0.730						
61	3.59	0.0021540	0.0013463	0.21700	0.13563	0.681						
62	3.59	0.0021540	0.0013463	0.25100	0.15688	0.638						
63	3.59	0.0021540	0.0013463	0.28750	0.17969	0.598						
64	3.59	0.0021540	0.0013463	0.32400	0.20250	0.562						
65	3.59	0.0021540	0.0013463	0.36200	0.22625	0.529						
66	3.59	0.0021540	0.0013463	0.40000	0.25000	0.500						
67	4.13	0.0024761	0.0015476	0.00000	0.00000	1.210	204.0800	5.0532	369.2	26.5083	515.19	177.2838
68	4.13	0.0024761	0.0015476	0.02900	0.01813	1.142	162.6200	4.0266	285.8	15.8848	393.88	79.2243
69	4.13	0.0024761	0.0015476	0.05800	0.03625	1.059	186.5000	4.6179	314.22	19.2011	426.66	100.6961
70	4.13	0.0024761	0.0015476	0.08850	0.05531	0.982	326.3400	8.0805	527.06	54.0229	706.88	457.9350
71	4.13	0.0024761	0.0015476	0.11900	0.07438	0.914						
72	4.13	0.0024761	0.0015476	0.15100	0.09438	0.851						
73	4.13	0.0024761	0.0015476	0.18300	0.11438	0.797						
74	4.13	0.0024761	0.0015476	0.21700	0.13563	0.746						
75	4.13	0.0024761	0.0015476	0.25100	0.15688	0.701						
76	4.13	0.0024761	0.0015476	0.28750	0.17969	0.659						
77	4.13	0.0024761	0.0015476	0.32400	0.20250	0.621						
78	4.13	0.0024761	0.0015476	0.36200	0.22625	0.586						
79	4.13	0.0024761	0.0015476	0.40000	0.25000	0.555						
80	4.74	0.0028465	0.0017791	0.00000	0.00000	1.267	174.0600	4.9546	323.74	23.4312	456.09	141.4039
81	4.74	0.0028465	0.0017791	0.02900	0.01813	1.208	130.8700	3.7252	237.26	12.5849	331.06	54.0792
82	4.74	0.0028465	0.0017791	0.05800	0.03625	1.128	134.4300	3.8265	235.14	12.3610	323.6	50.5051
83	4.74	0.0028465	0.0017791	0.08850	0.05531	1.052	163.0300	4.6406	273.7	16.7475	371.38	76.3423
84	4.74	0.0028465	0.0017791	0.11900	0.07438	0.984	271.8900	7.7393	436.33	42.5629	583.66	296.3395
85	4.74	0.0028465	0.0017791	0.15100	0.09438	0.921						
86	4.74	0.0028465	0.0017791	0.18300	0.11438	0.865						

Table C-2 (continued)

Line entry	Conc. factor	^{235}U content $^{235}U/cm^3$ g	^{235}U content $^{235}U/gS\text{-}S$ g	Water content H_2O/cm^3 g	Water content $H_2O/gS\text{-}S$ g	k_∞ or k-inf	Critical infinite slab[b] Thick-ness (cm)	Critical infinite slab[b] ^{235}U areal density (kg/m^2)	Critical infinite cylinder[b] Diameter (cm)	Critical infinite cylinder[b] ^{235}U linear density (kg/m)	Critical sphere[b] Diameter (cm)	Critical sphere[b] ^{235}U mass (kg)
						Fig. D-8	Fig. D-9	Fig. D-10	Fig. D-11	Fig. D-12	Fig. D-13	Fig. D-14
87	4.74	0.0028465	0.0017791	0.21700	0.13563	0.813						
88	4.74	0.0028465	0.0017791	0.25100	0.15688	0.766						
89	4.74	0.0028465	0.0017791	0.28750	0.17969	0.722						
90	4.74	0.0028465	0.0017791	0.32400	0.20250	0.683						
91	4.74	0.0028465	0.0017791	0.36200	0.22625	0.646						
92	4.74	0.0028465	0.0017791	0.40000	0.25000	0.613						
93	5.45	0.0032722	0.0020451	0.00000	0.00000	1.321	151.9000	4.9705	290.4	21.6732	412.87	120.5809
94	5.45	0.0032722	0.0020451	0.02900	0.01813	1.271	109.9000	3.5961	205.21	10.8225	289.54	41.5876
95	5.45	0.0032722	0.0020451	0.05800	0.03625	1.196	106.8200	3.4954	192.49	9.5224	268.17	33.0422
96	5.45	0.0032722	0.0020451	0.08850	0.05531	1.121	115.6700	3.7850	201.17	10.4005	276.79	36.3320
97	5.45	0.0032722	0.0020451	0.11900	0.07438	1.054	139.2500	4.5565	233.95	14.0662	317.8	54.9921
98	5.45	0.0032722	0.0020451	0.15100	0.09438	0.991	214.9100	7.0323	346.32	30.8237	463.16	170.2282
99	5.45	0.0032722	0.0020451	0.18300	0.11438	0.935						
100	5.45	0.0032722	0.0020451	0.21700	0.13563	0.882						
101	5.45	0.0032722	0.0020451	0.25100	0.15688	0.834						
102	5.45	0.0032722	0.0020451	0.28750	0.17969	0.788						
103	5.45	0.0032722	0.0020451	0.32400	0.20250	0.747						
104	5.45	0.0032722	0.0020451	0.36200	0.22625	0.709						
105	5.45	0.0032722	0.0020451	0.40000	0.25000	0.674						
106	4.77	0.0028639	0.0017899	0.00000	0.00000	1.269	172.9200	4.9523	322.11	23.3376	453.93	140.2565
107	4.77	0.0028639	0.0017899	0.02900	0.01813	1.210	129.8400	3.7185	235.59	12.4842	328.89	53.3468
108	4.77	0.0028639	0.0017899	0.05800	0.03625	1.131	132.9900	3.8087	232.43	12.1516	320.5	49.3674
109	4.77	0.0028639	0.0017899	0.08850	0.05531	1.055	159.9900	4.5820	268.98	16.2738	365.3	73.0980
110	4.77	0.0028639	0.0017899	0.11900	0.07438	0.987	258.7600	7.4106	415.9	38.9068	556.21	258.0316
111	4.77	0.0028639	0.0017899	0.15100	0.09438	0.924						
112	4.77	0.0028639	0.0017899	0.18300	0.11438	0.868						
113	4.77	0.0028639	0.0017899	0.21700	0.13563	0.816						
114	4.77	0.0028639	0.0017899	0.25100	0.15688	0.769						
115	4.77	0.0028639	0.0017899	0.28750	0.17969	0.725						
116	4.77	0.0028639	0.0017899	0.32400	0.20250	0.685						
117	4.77	0.0028639	0.0017899	0.36200	0.22625	0.649						
118	4.77	0.0028639	0.0017899	0.40000	0.25000	0.615						
119	6.01	0.0036054	0.0022534	0.00000	0.00000	1.356	139.5500	5.0313	271.92	20.9375	388.95	111.0794
120	6.01	0.0036054	0.0022534	0.02900	0.01813	1.314	98.9200	3.5665	188.45	10.0562	267.73	36.2279
121	6.01	0.0036054	0.0022534	0.05800	0.03625	1.242	93.6800	3.3775	172.29	8.4055	241.79	26.6850
122	6.01	0.0036054	0.0022534	0.08850	0.05531	1.169	97.2000	3.5044	172.93	8.4681	239.82	26.0380
123	6.01	0.0036054	0.0022534	0.11900	0.07438	1.103	108.3500	3.9065	186.69	9.8693	256.04	31.6866
124	6.01	0.0036054	0.0022534	0.15100	0.09438	1.040	134.1900	4.8381	223.61	14.1588	302.57	52.2913
125	6.01	0.0036054	0.0022534	0.18300	0.11438	0.984	215.0600	7.7538	345.17	33.7372	462.48	186.7372
126	6.01	0.0036054	0.0022534	0.21700	0.13563	0.930						
127	6.01	0.0036054	0.0022534	0.25100	0.15688	0.882						
128	6.01	0.0036054	0.0022534	0.28750	0.17969	0.836						
129	6.01	0.0036054	0.0022534	0.32400	0.20250	0.794						

Table C-2 (continued)

Line entry	Conc. factor	^{235}U content $^{235}U/cm^3$ g	^{235}U content $^{235}U/gS\text{-}S$ g	Water content H_2O/cm^3 g	Water content $H_2O/gS\text{-}S$ g	k_∞ or k-inf	Critical infinite slab[b] Thick-ness (cm)	Critical infinite slab[b] ^{235}U areal density (kg/m^2)	Critical infinite cylinder[b] Diameter (cm)	Critical infinite cylinder[b] ^{235}U linear density (kg/m)	Critical sphere[b] Diameter (cm)	Critical sphere[b] ^{235}U mass (kg)
						Fig. D-8	Fig. D-9	Fig. D-10	Fig. D-11	Fig. D-12	Fig. D-13	Fig. D-14
130	6.01	0.0036054	0.0022534	0.36200	0.22625	0.754						
131	6.01	0.0036054	0.0022534	0.40000	0.25000	0.719						
132	7.56	0.0045390	0.0028369	0.00000	0.00000	1.430	116.7900	5.3011	238.07	20.2050	345.33	97.8727
133	7.56	0.0045390	0.0028369	0.02900	0.01813	1.409	79.7700	3.6208	159.45	9.0636	230.09	28.9502
134	7.56	0.0045390	0.0028369	0.05800	0.03625	1.346	72.4900	3.2903	140.1	6.9972	200.07	19.0329
135	7.56	0.0045390	0.0028369	0.08850	0.05531	1.279	71.0700	3.2259	133.04	6.3098	187.88	15.7616
136	7.56	0.0045390	0.0028369	0.11900	0.07438	1.216	72.8700	3.3076	132.46	6.2549	185.18	15.0918
137	7.56	0.0045390	0.0028369	0.15100	0.09438	1.156	77.7400	3.5286	137.37	6.7272	190.12	16.3321
138	7.56	0.0045390	0.0028369	0.18300	0.11438	1.101	86.5400	3.9281	149.1	7.9251	204.16	20.2242
139	7.56	0.0045390	0.0028369	0.21700	0.13563	1.047	103.8400	4.7133	173.8	10.7684	235.54	31.0565
140	7.56	0.0045390	0.0028369	0.25100	0.15688	0.999	144.4800	6.5579	234.41	19.5886	314.43	73.8807
141	7.56	0.0045390	0.0028369	0.28750	0.17969	0.951	1234.4000	56.0294	2057.26	1508.7887	2840.9	54491.2343
142	7.56	0.0045390	0.0028369	0.32400	0.20250	0.908						
143	7.56	0.0045390	0.0028369	0.36200	0.22625	0.867						
144	7.56	0.0045390	0.0028369	0.40000	0.25000	0.829						
145	9.52	0.0057142	0.0035714	0.00000	0.00000	1.490	100.0400	5.7165	213.5	20.4570	313.84	92.4866
146	9.52	0.0057142	0.0035714	0.02900	0.01813	1.492	66.5200	3.8011	139.6	8.7461	204.36	25.5353
147	9.52	0.0057142	0.0035714	0.05800	0.03625	1.440	58.9300	3.3674	119.65	6.4250	173.46	15.6154
148	9.52	0.0057142	0.0035714	0.08850	0.05531	1.381	56.0100	3.2005	110.13	5.4432	158.08	11.8191
149	9.52	0.0057142	0.0035714	0.11900	0.07438	1.323	55.2800	3.1588	105.61	5.0056	150.2	10.1383
150	9.52	0.0057142	0.0035714	0.15100	0.09438	1.267	55.9700	3.1982	104.12	4.8653	146.73	9.4517
151	9.52	0.0057142	0.0035714	0.18300	0.11438	1.214	57.9200	3.3097	105.16	4.9630	146.99	9.5021
152	9.52	0.0057142	0.0035714	0.21700	0.13563	1.163	61.4900	3.5137	109.02	5.3341	151.09	10.3196
153	9.52	0.0057142	0.0035714	0.25100	0.15688	1.115	67.0600	3.8319	116.23	6.0629	159.94	12.2412
154	9.52	0.0057142	0.0035714	0.28750	0.17969	1.068	76.8100	4.3891	129.96	7.5799	176.99	16.5882
155	9.52	0.0057142	0.0035714	0.32400	0.20250	1.025	94.5700	5.4039	156.4	10.9779	210.46	27.8909
156	9.52	0.0057142	0.0035714	0.36200	0.22625	0.983	142.4500	8.1399	227.79	23.2871	305.69	85.4669
157	9.52	0.0057142	0.0035714	0.40000	0.25000	0.945						
158	10.50	0.0063000	0.0039375	0.00000	0.00000	1.512	94.2100	5.9352	205.28	20.8508	303.38	92.1086
159	10.50	0.0063000	0.0039375	0.05800	0.03625	1.478	54.5300	3.4354	113.3	6.3517	165.3	14.8990
160	10.50	0.0063000	0.0039375	0.11900	0.07438	1.367	50.1000	3.1563	97.96	4.7482	140.28	9.1060
161	10.50	0.0063000	0.0039375	0.18300	0.11438	1.261	50.9400	3.2092	94.72	4.4393	133.44	7.8379
162	10.50	0.0063000	0.0039375	0.25100	0.15688	1.163	55.9600	3.5255	99.6	4.9085	138.1	8.6880
163	10.50	0.0063000	0.0039375	0.32400	0.20250	1.075	68.4900	4.3149	117.08	6.7826	159.62	13.4153
164	10.50	0.0063000	0.0039375	0.40000	0.25000	0.995	111.8700	7.0478	174.66	15.0945	246.4	49.3471
165	37.33	0.0224000	0.0140000	0.00000	0.00000	1.604	48.4500	10.8528	139.54	34.2559	218.62	122.5508
166	37.33	0.0224000	0.0140000	0.05800	0.03625	1.765	25.6900	5.7546	71.18	8.9136	111.22	16.1360
167	37.33	0.0224000	0.0140000	0.11900	0.07438	1.751	21.3300	4.7779	54.94	5.3103	84.76	7.1420
168	37.33	0.0224000	0.0140000	0.18300	0.11438	1.713	19.1400	4.2874	46.48	3.8008	70.78	4.1589
169	37.33	0.0224000	0.0140000	0.25100	0.15688	1.668	17.7300	3.9715	41.1	2.9718	61.9	2.7818
170	37.33	0.0224000	0.0140000	0.32400	0.20250	1.619	16.7700	3.7565	37.38	2.4582	55.64	2.0203
171	37.33	0.0224000	0.0140000	0.39900	0.24938	1.569	16.0900	3.6042	34.64	2.1110	51.14	1.5687
172	132.33	0.0794000	0.0496250	0.00000	0.00000	1.623	24.0000	19.0560	98.82	60.8976	164.52	185.1293

Table C-2 (continued)

Line entry	Conc. factor	^{235}U content: g ^{235}U per cm^3	^{235}U content: g^{235}U per gS-S	Water content: gH_2O per cm^3	Water content: gH2O per gS-S	k_∞ or k-inf	Critical infinite slab[b]: Thick-ness (cm)	Critical infinite slab[b]: ^{235}U areal density (kg/m^2)	Critical infinite cylinder[b]: Diameter (cm)	Critical infinite cylinder[b]: ^{235}U linear density (kg/m)	Critical sphere[b]: Diameter (cm)	Critical sphere[b]: ^{235}U mass (kg)
						Fig. D-8	Fig. D-9	Fig. D-10	Fig. D-11	Fig. D-12	Fig. D-13	Fig. D-14
173	132.33	0.0794000	0.0496250	0.05800	0.03625	1.767	14.8200	11.7671	56.02	19.5703	91.62	31.9735
174	132.33	0.0794000	0.0496250	0.11900	0.07438	1.826	12.3700	9.8218	42.44	11.2321	68.52	13.3743
175	132.33	0.0794000	0.0496250	0.18300	0.11438	1.847	10.9200	8.6705	34.92	7.6043	55.8	7.2231
176	132.33	0.0794000	0.0496250	0.25100	0.15688	1.853	9.8900	7.8527	29.88	5.5676	47.42	4.4331
177	132.33	0.0794000	0.0496250	0.32400	0.20250	1.849	9.0800	7.2095	26.2	4.2807	41.32	2.9329
178	132.33	0.0794000	0.0496250	0.39600	0.24750	1.841	8.4600	6.7172	23.52	3.4497	36.88	2.0854
179	469.50	0.2817000	0.1760625	0.00000	0.00000	1.754	9.6200	27.0995	63.82	90.1136	113.54	215.8898
180	469.50	0.2817000	0.1760625	0.05800	0.03625	1.716	8.3400	23.4938	45	44.8024	76.96	67.2327
181	469.50	0.2817000	0.1760625	0.11900	0.07438	1.757	7.6600	21.5782	35.52	27.9140	59.66	31.3210
182	469.50	0.2817000	0.1760625	0.18300	0.11438	1.793	7.0800	19.9444	29.48	19.2279	48.88	17.2258
183	469.50	0.2817000	0.1760625	0.25100	0.15688	1.820	6.5400	18.4232	25.18	14.0278	41.36	10.4358
184	469.50	0.2817000	0.1760625	0.32400	0.20250	1.841	6.0400	17.0147	21.96	10.6694	35.76	6.7449
185	469.50	0.2817000	0.1760625	0.38500	0.24063	1.854	5.6800	16.0006	19.88	8.7440	32.18	4.9152
186	1666.67	1.0000000	0.6250000	0.00000	0.00000	1.923	3.1600	31.6000	36.14	102.5808	69.12	172.9059
187	1666.67	1.0000000	0.6250000	0.05800	0.03625	1.818	3.7000	37.0000	31.14	76.1600	55.86	91.2644
188	1666.67	1.0000000	0.6250000	0.11900	0.07438	1.780	4.0400	40.4000	26.98	57.1707	47.14	54.8488
189	1666.67	1.0000000	0.6250000	0.18300	0.11438	1.768	4.1800	41.8000	23.64	43.8919	40.62	35.0928
190	1666.67	1.0000000	0.6250000	0.25100	0.15688	1.770	4.1600	41.6000	20.54	33.1353	34.88	22.2192
191	1666.67	1.0000000	0.6250000	0.32400	0.20250	1.776	4.0400	40.4000	18.66	27.3472	31.32	16.0866
192	1666.67	1.0000000	0.6250000	0.34600	0.21625	1.779	3.8700	38.7000	18.04	25.5601	30.2	14.4218

[a] The figures are presented in Appendix D.

[b] Dimensions based on system k_{eff} = 0.95 rather than k_{eff} = 1.0 to conservatively account for methods and data uncertainty.

APPENDIX D

Surface Response Figures for Tables A-1 and A-2 Data

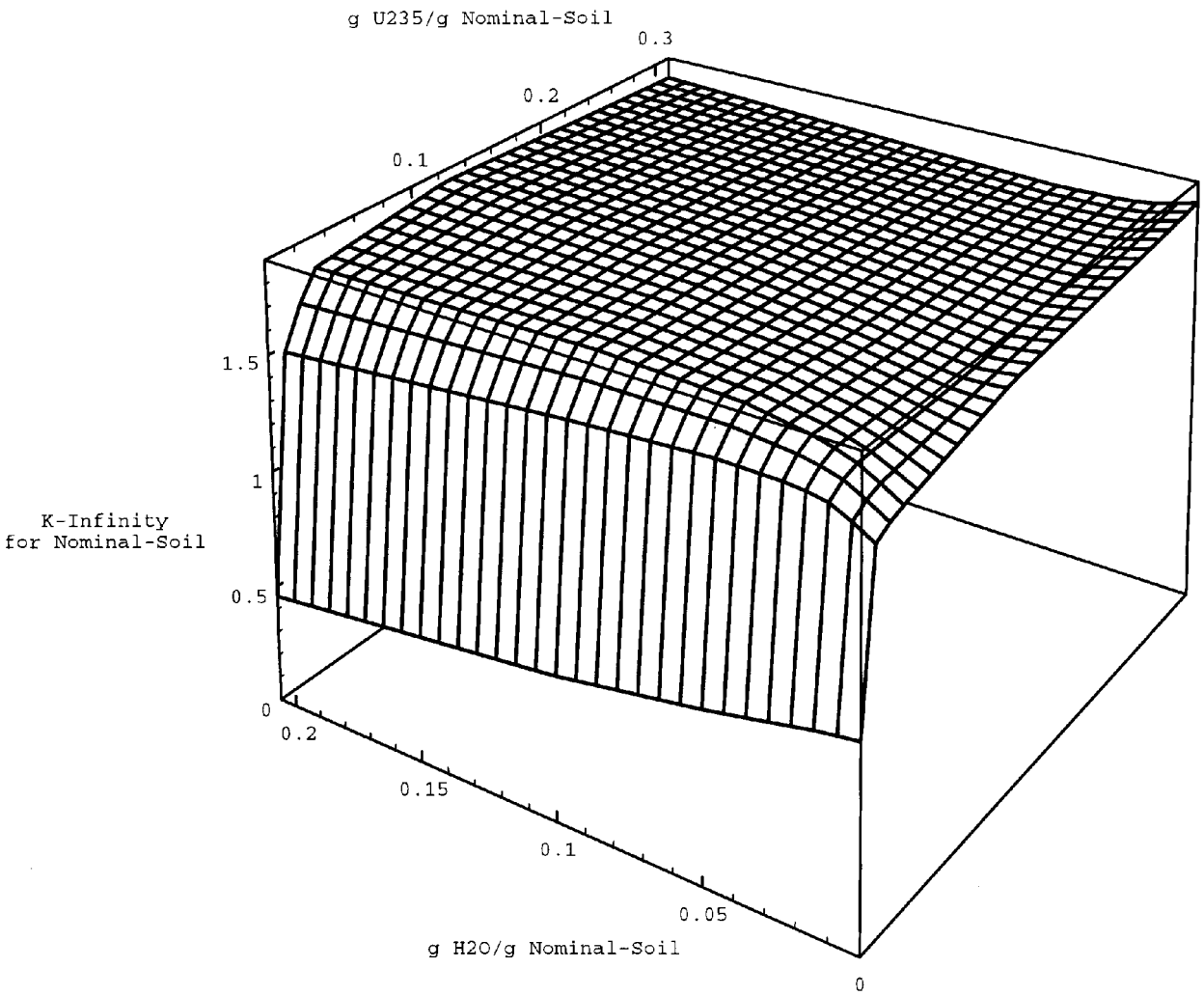

Figure D.1-1 Nominal soil k-infinity vs ^{235}U and H_2O

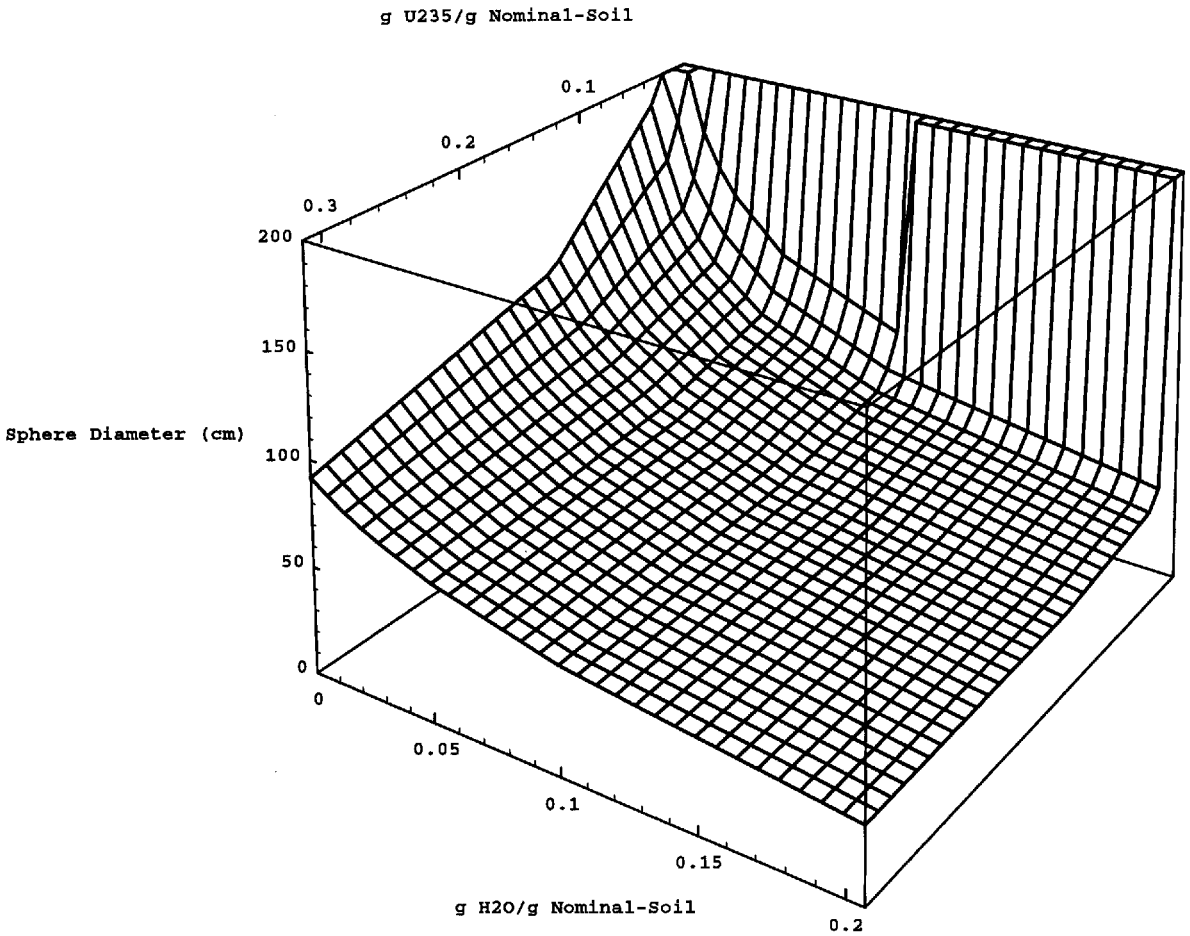

Figure D.1-2 Nominal-soil critical sphere diameter vs ^{235}U and H_2O concentration

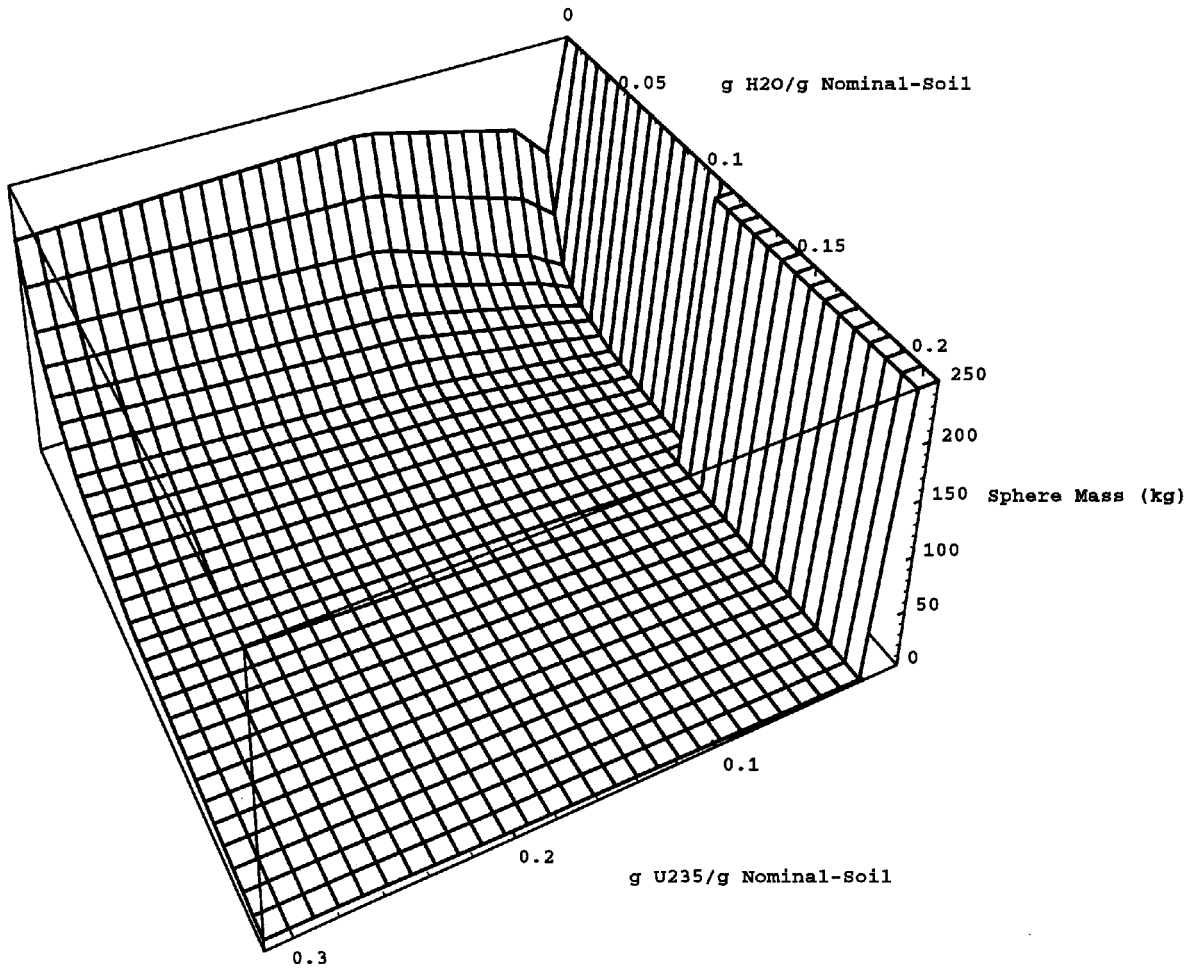

Figure D.1-3 Nominal-soil critical sphere mass vs ^{235}U and H_2O concentration

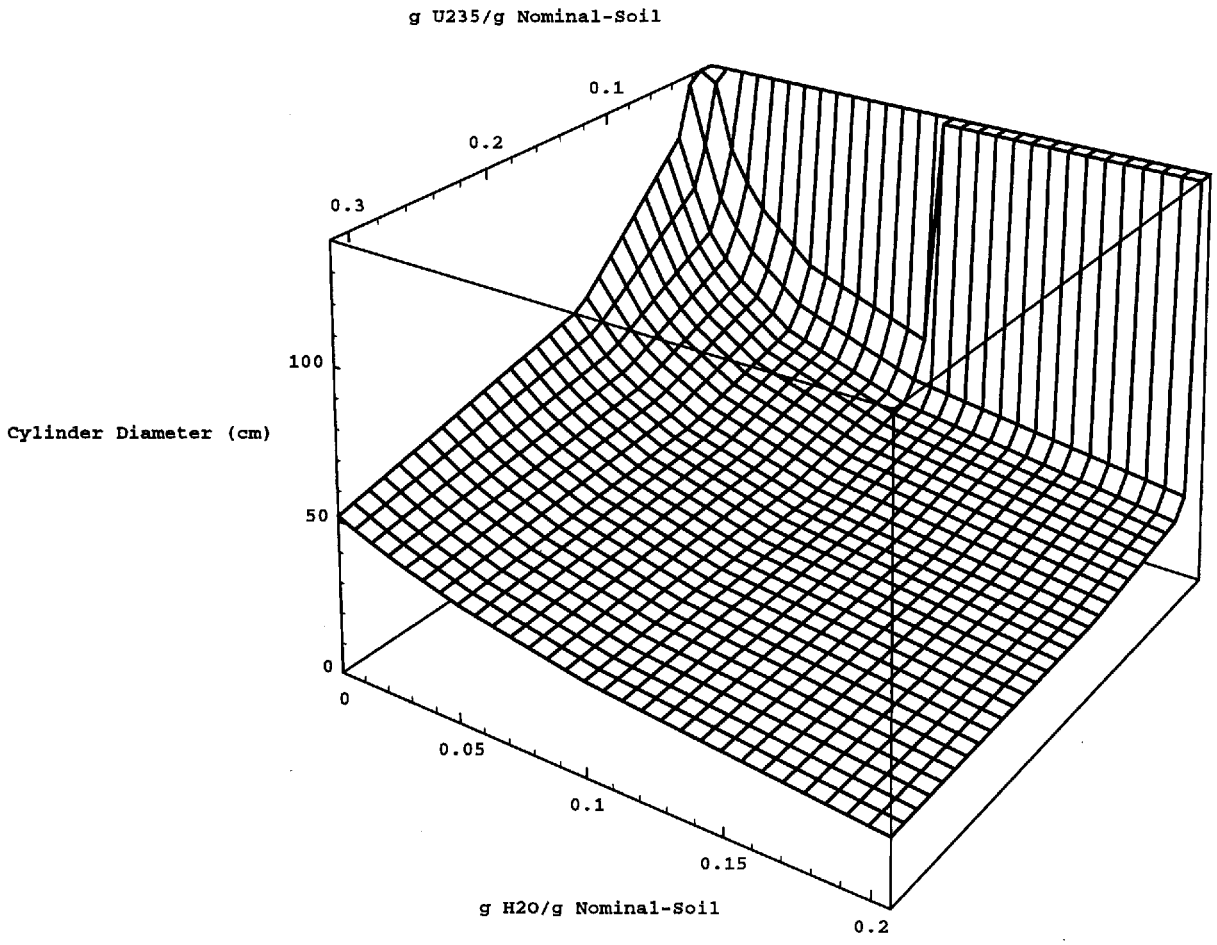

Figure D.1-4 Nominal-soil critical infinite cylinder diameter vs ^{235}U and H_2O concentration

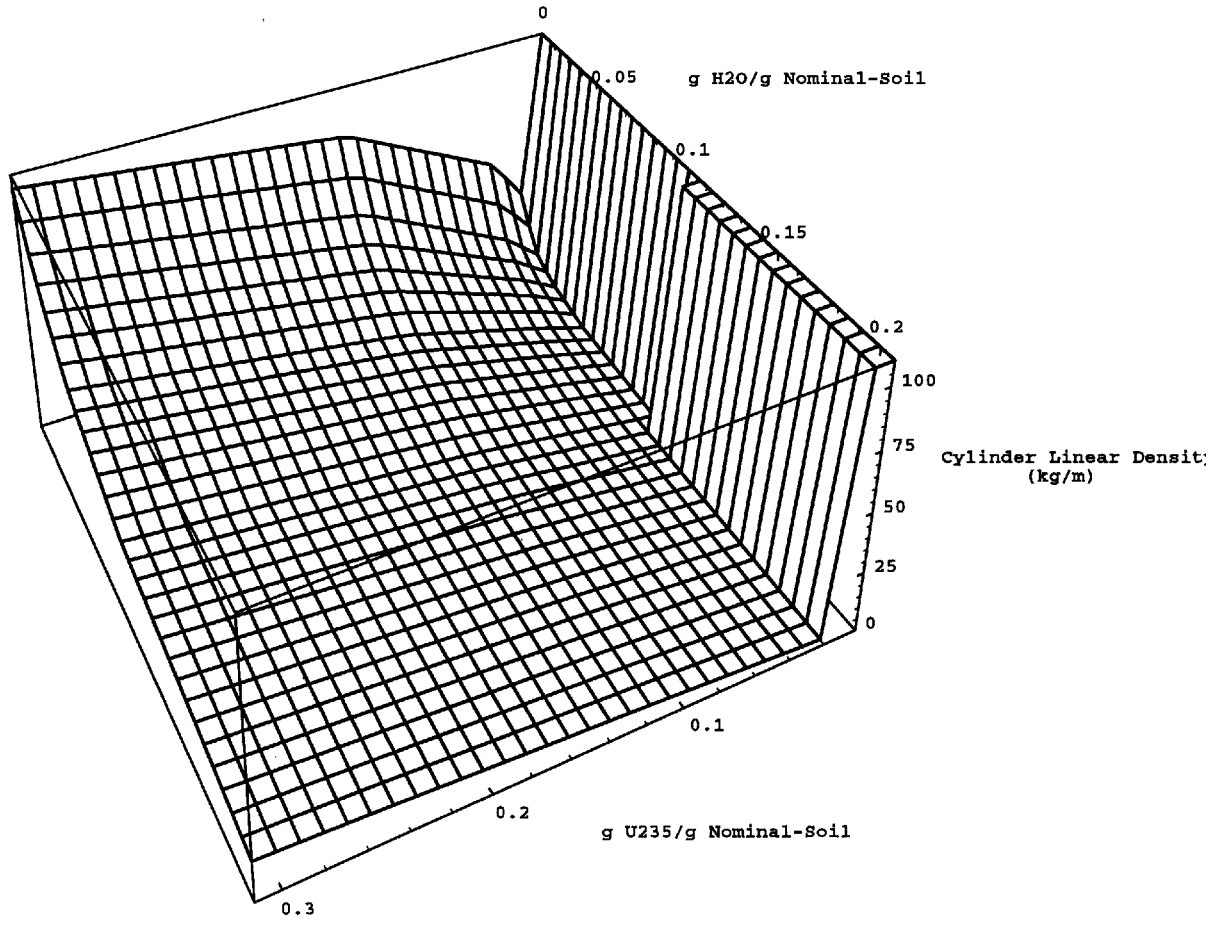

Figure D.1-5 Nominal-soil critical infinite cylinder linear density vs ^{235}U and H_2O concentration

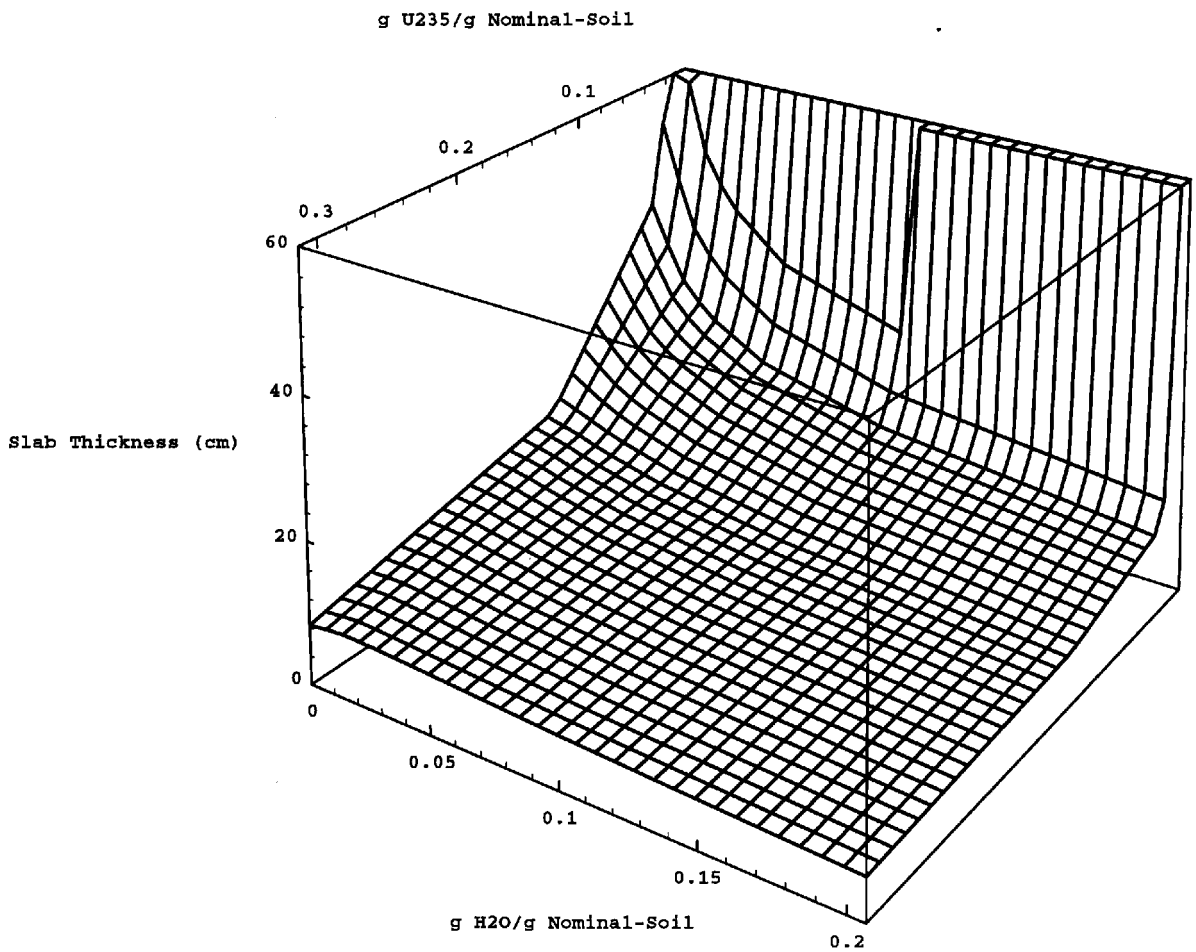

Figure D.1-6 Nominal-soil critical infinite slab thickness vs ^{235}U and H_2O concentration

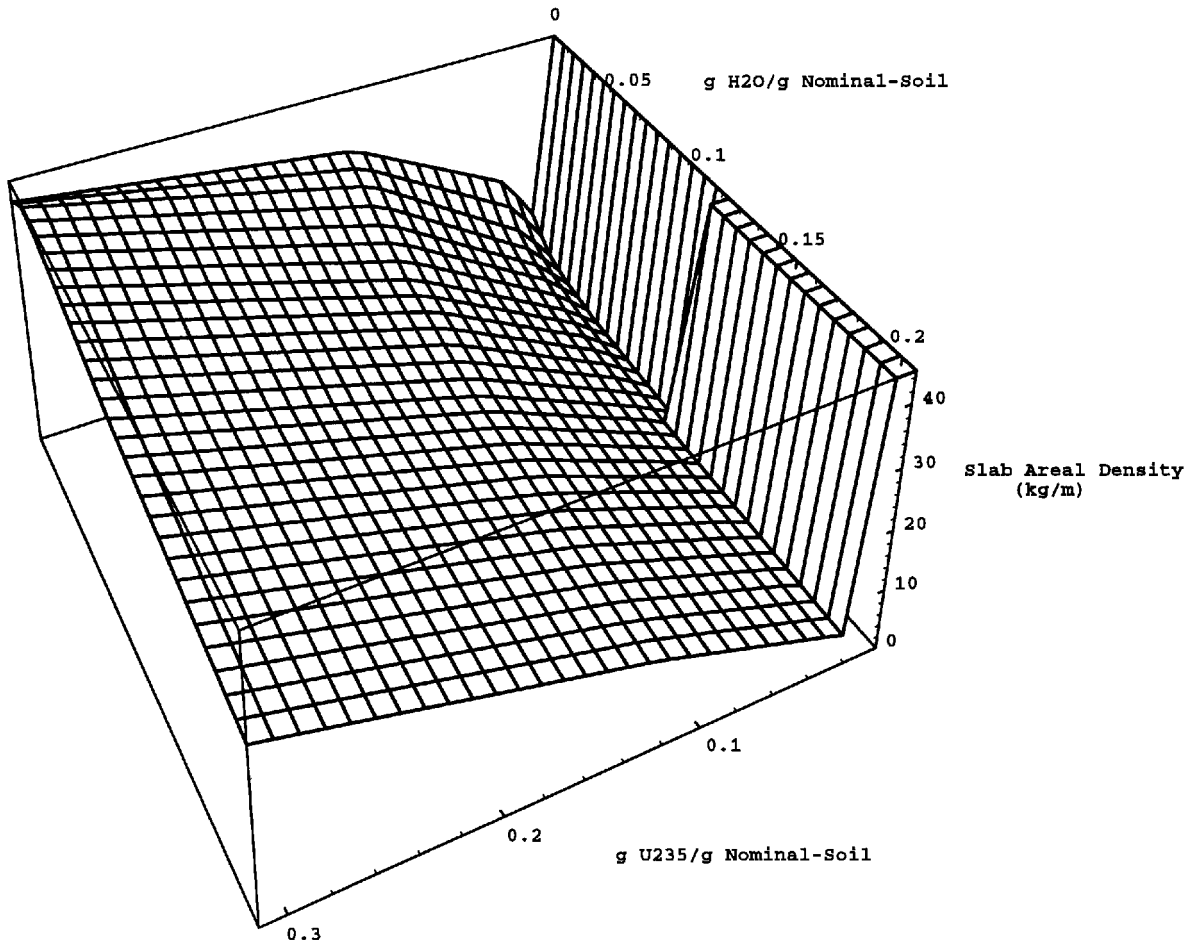

Figure D.1-7 Nominal-soil critical infinite slab areal density vs ^{235}U and H_2O concentration

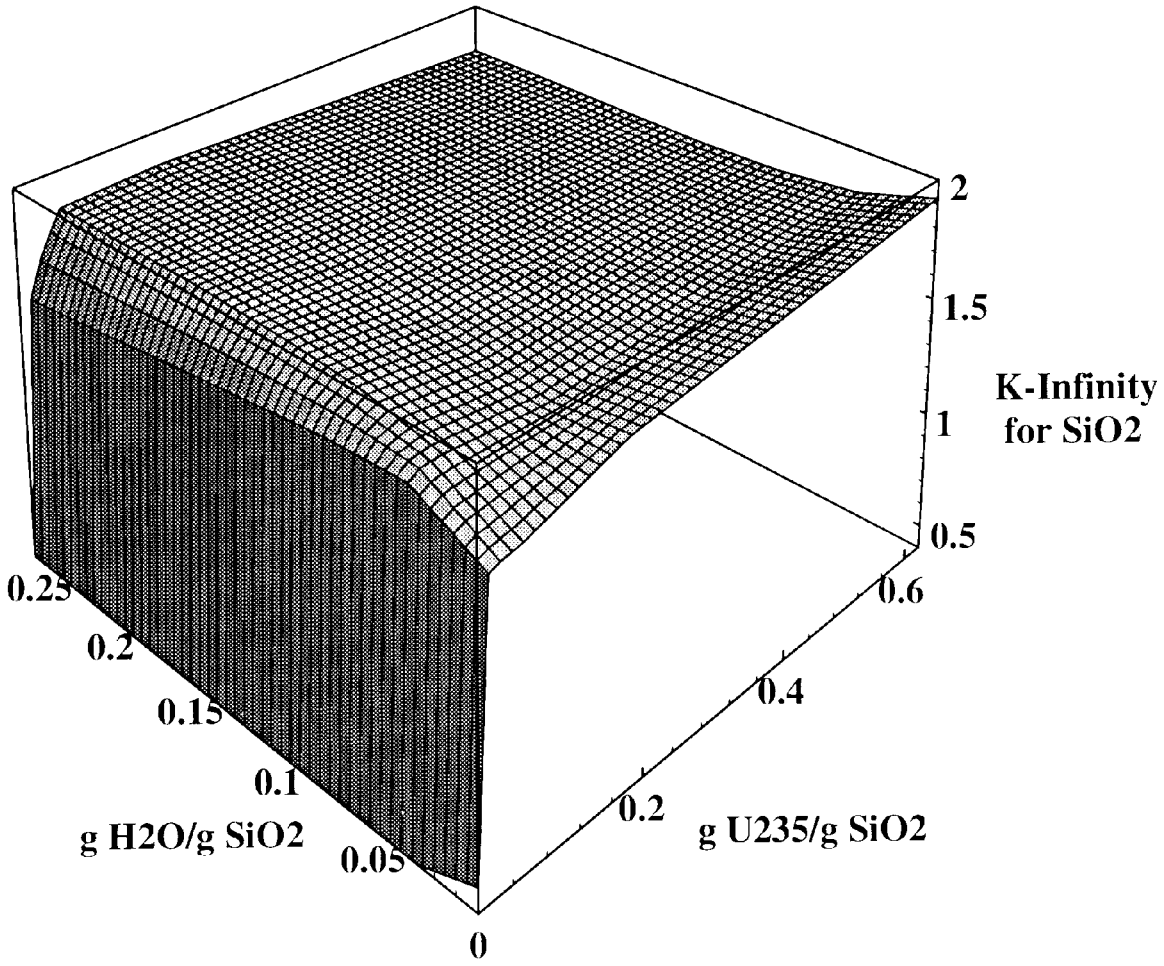

Figure D.1-8 SiO_2-soil k-infinity vs H_2O and ^{235}U concentration linear plot

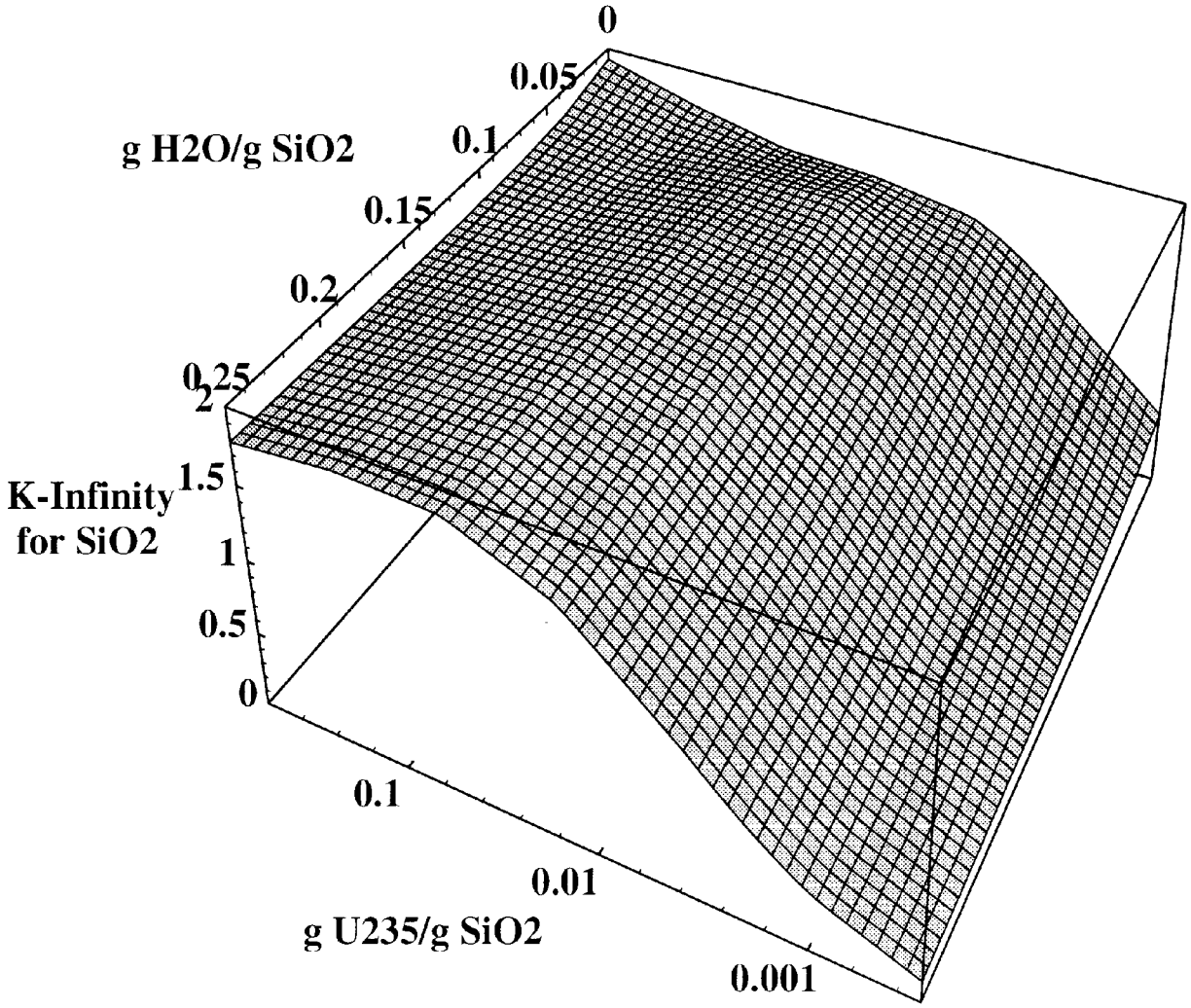

Figure D.1-9 SiO_2-soil k-infinity vs H_2O and ^{235}U concentration log plot

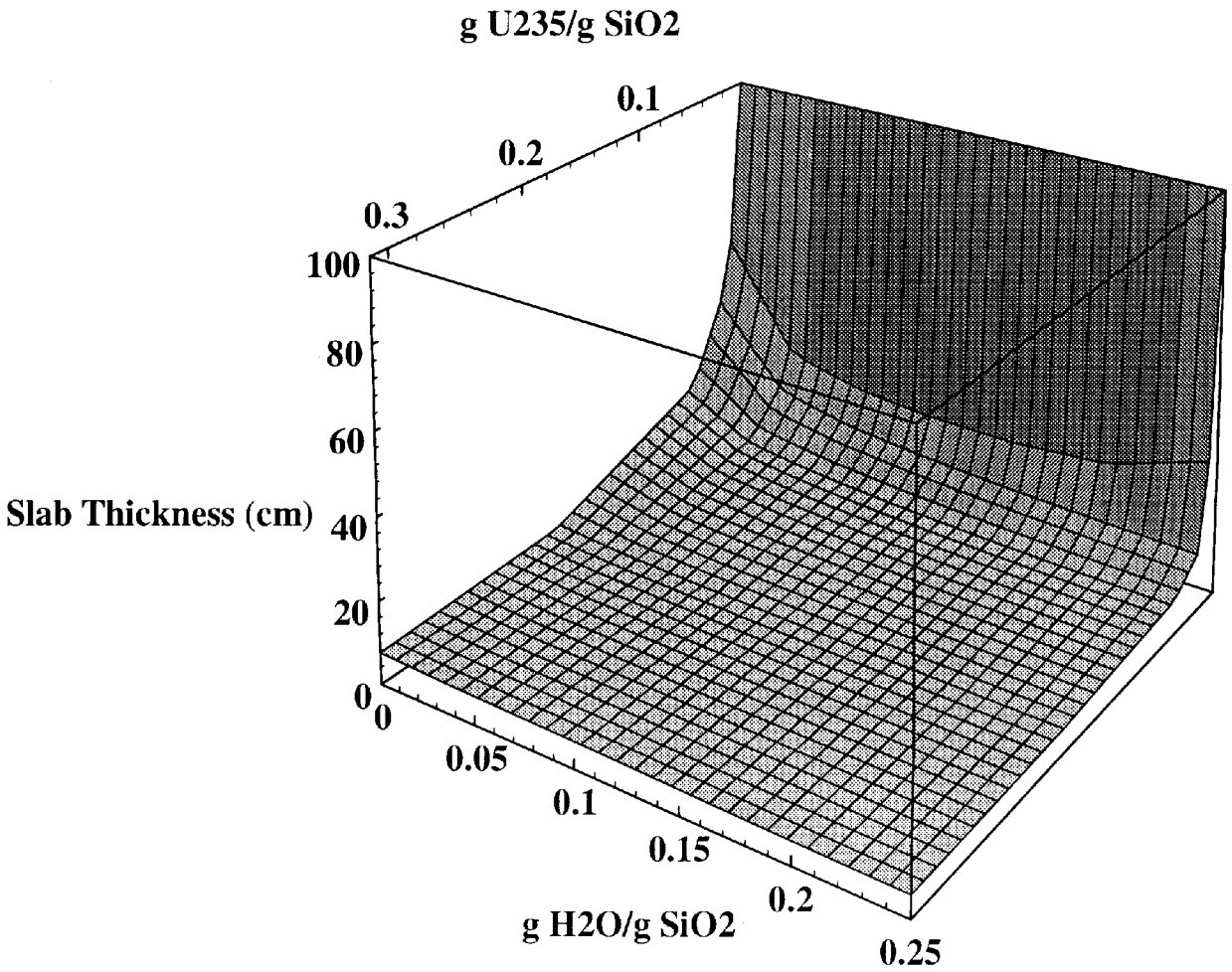

Figure D.1-10 SiO_2-soil critical infinite slab thickness vs H_2O and ^{235}U concentration linear plot

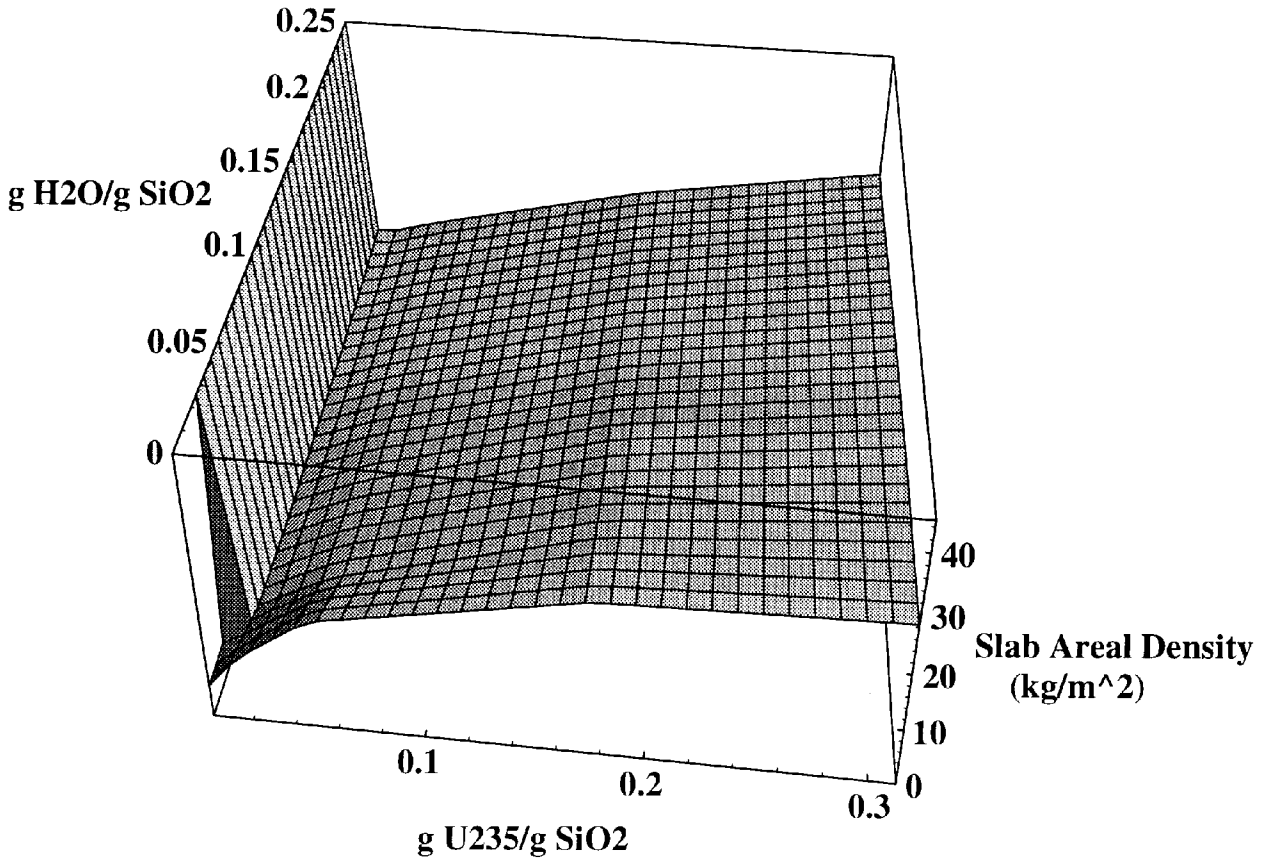

Figure D.1-11 SiO_2-soil critical infinite slab areal density vs H_2O and ^{235}U concentration linear plot

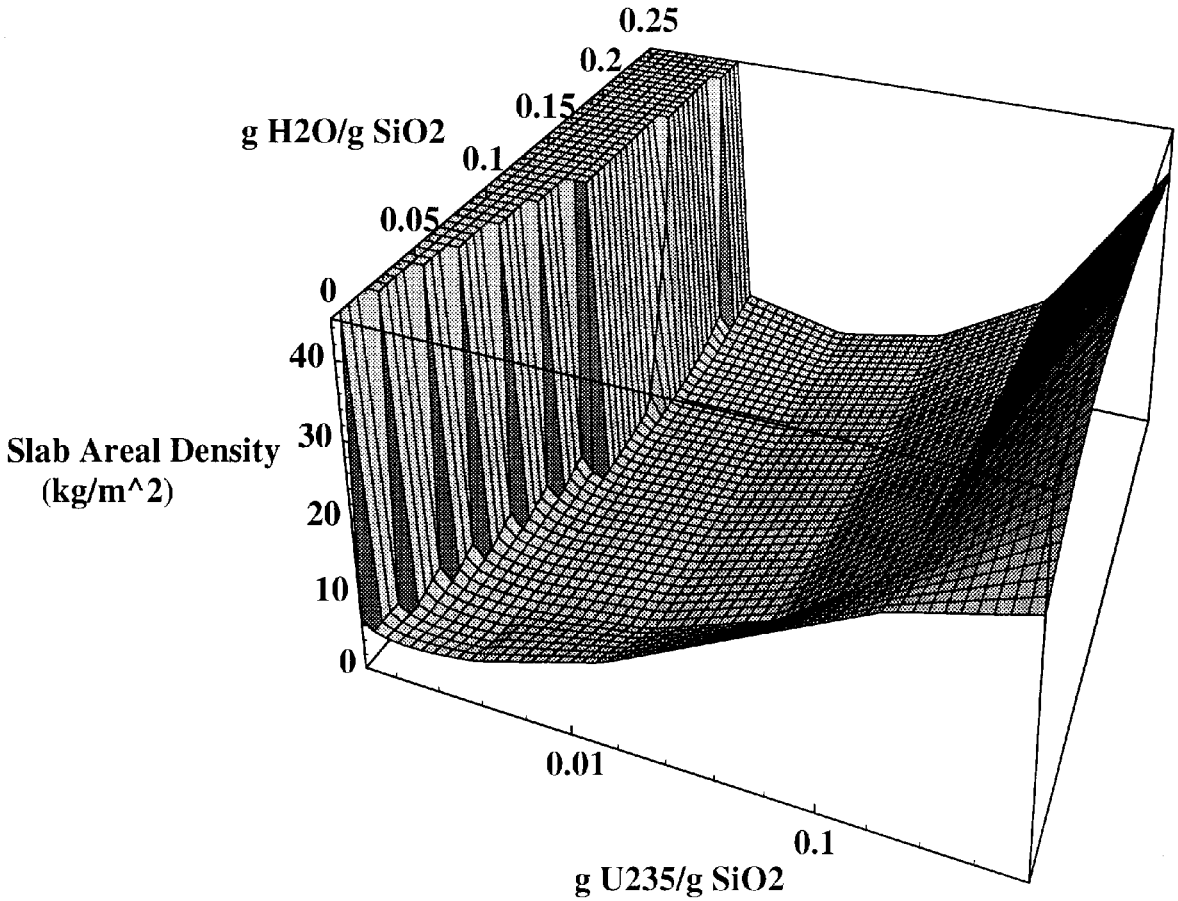

Figure D.1-12 SiO_2-soil critical infinite slab areal density vs H_2O and ^{235}U concentration log plot

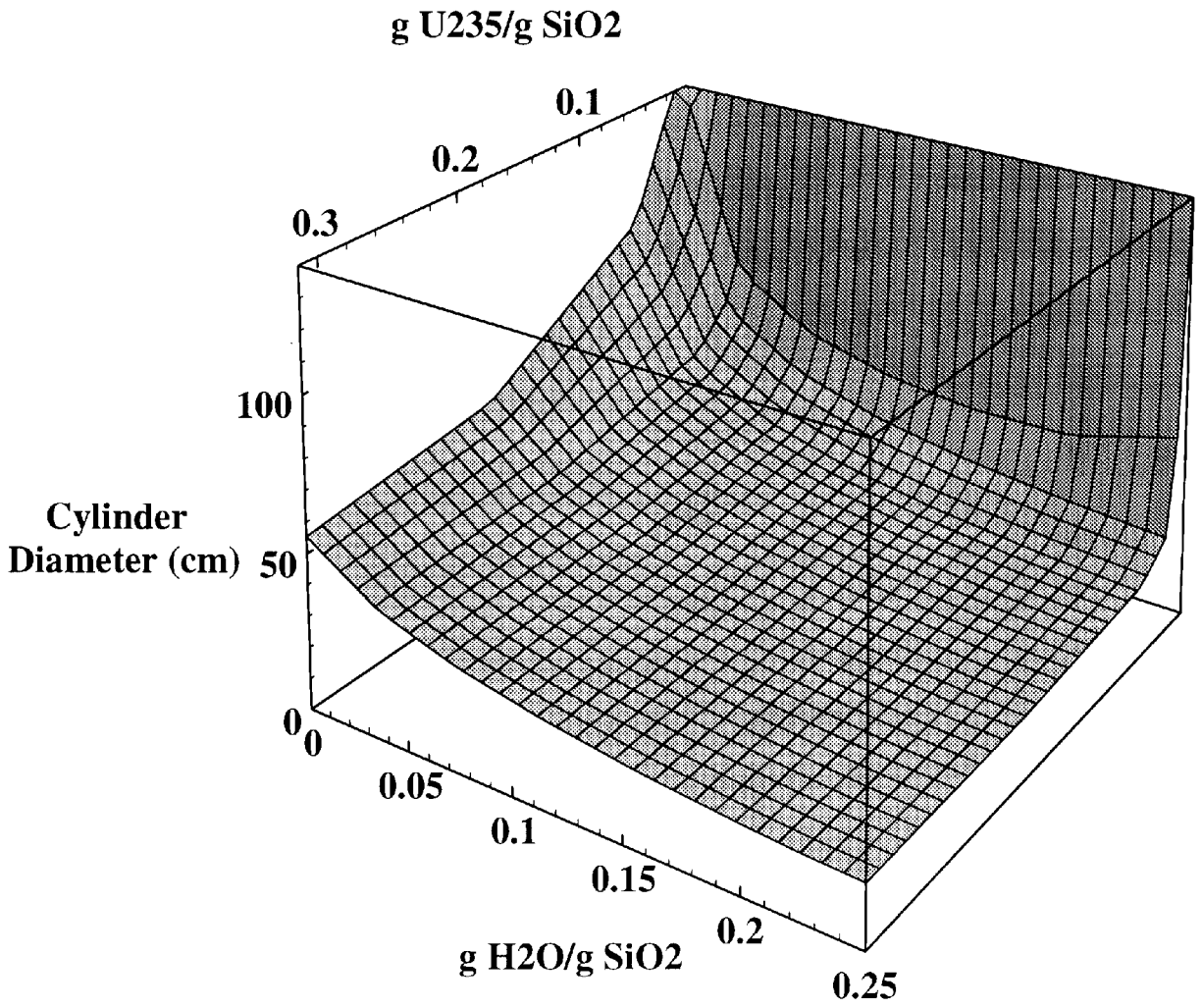

Figure D.1-13 SiO_2-soil critical infinite cylinder diameter vs H_2O and ^{235}U concentration linear plot

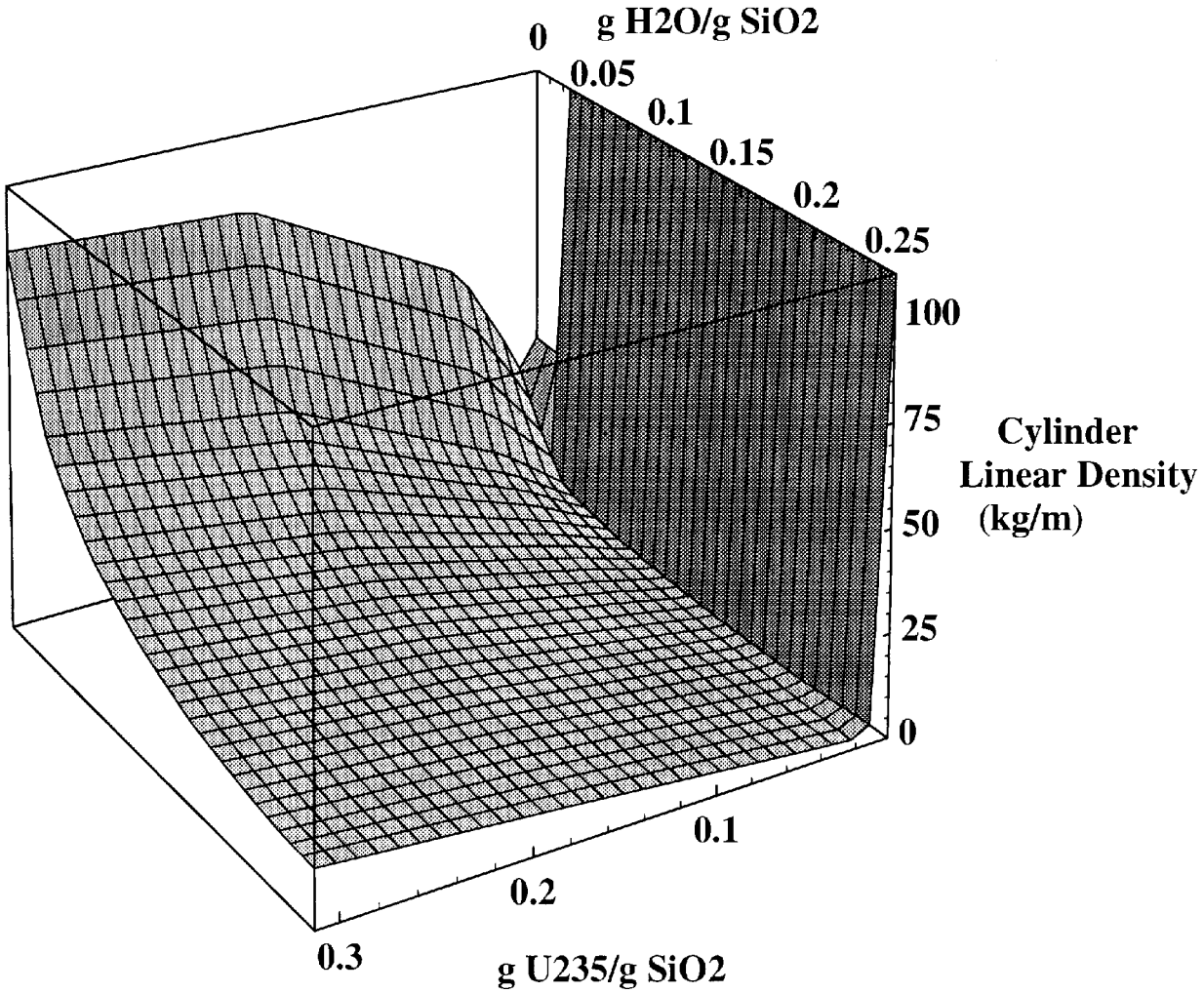

Figure D.1-14 SiO_2-soil critical infinite cylinder linear density vs H_2O and ^{235}U concentration linear plot

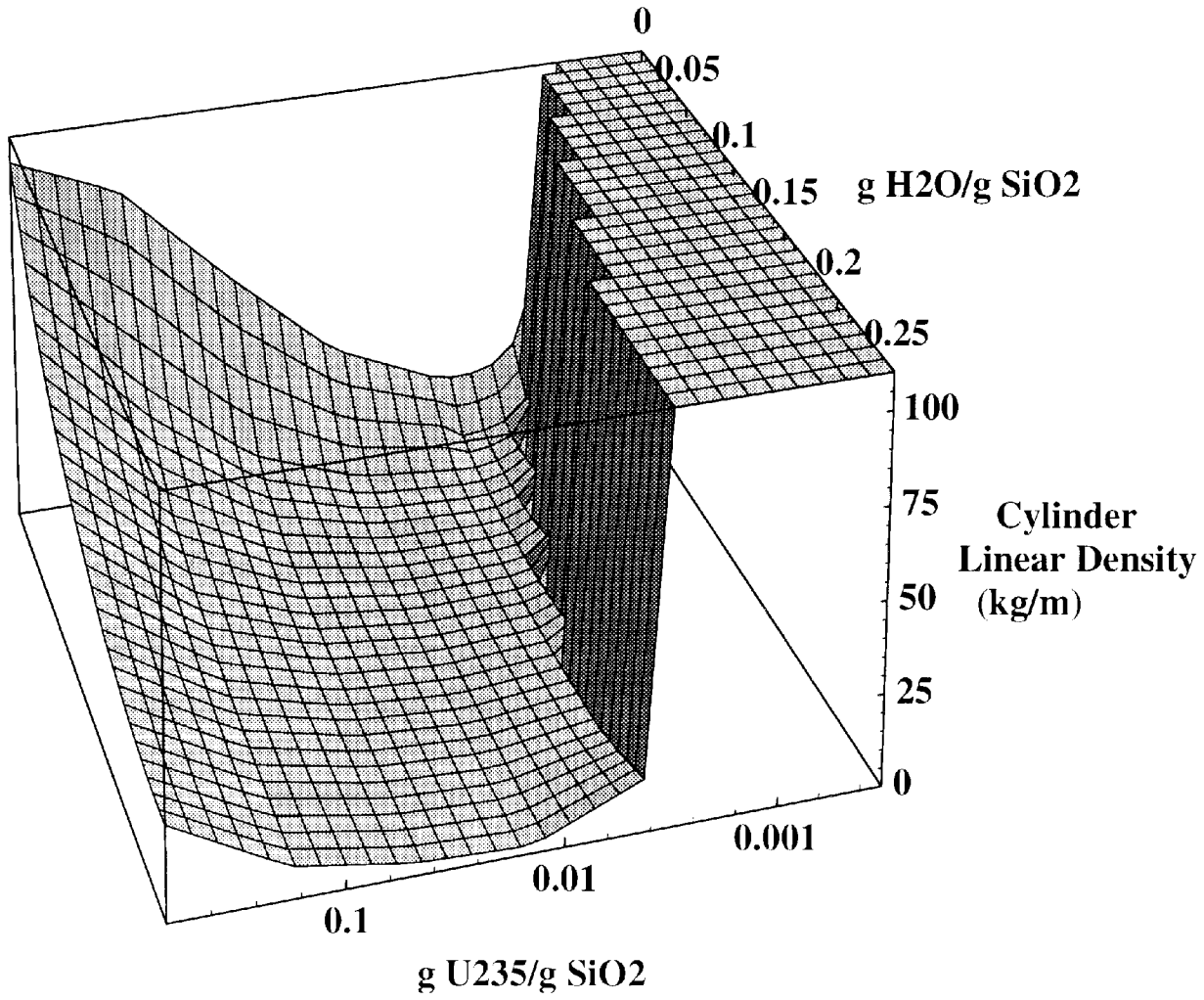

Figure D.1-15 SiO_2-soil critical infinite cylinder linear density vs H_2O and ^{235}U concentration log plot

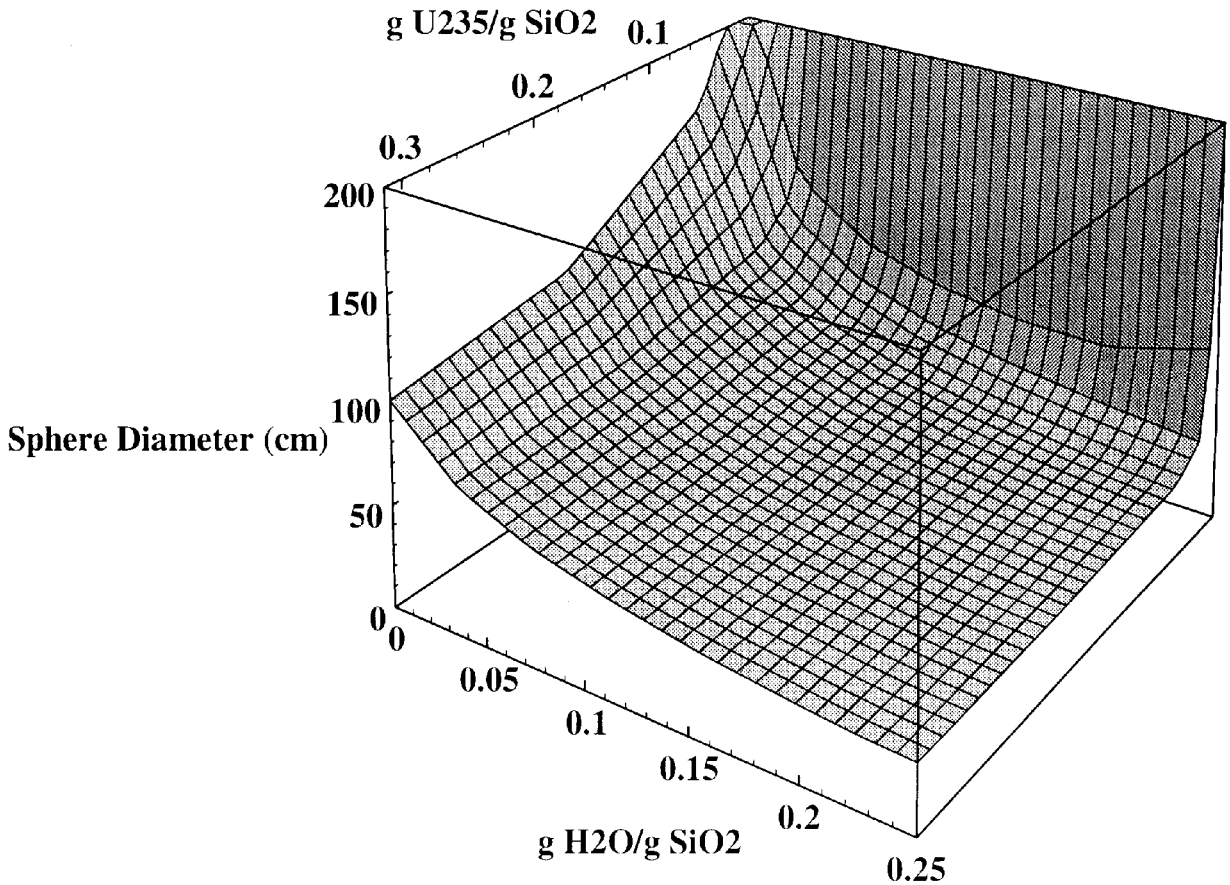

Figure D.1-16 SiO_2-soil critical sphere diameter vs H_2O and ^{235}U concentration linear plot

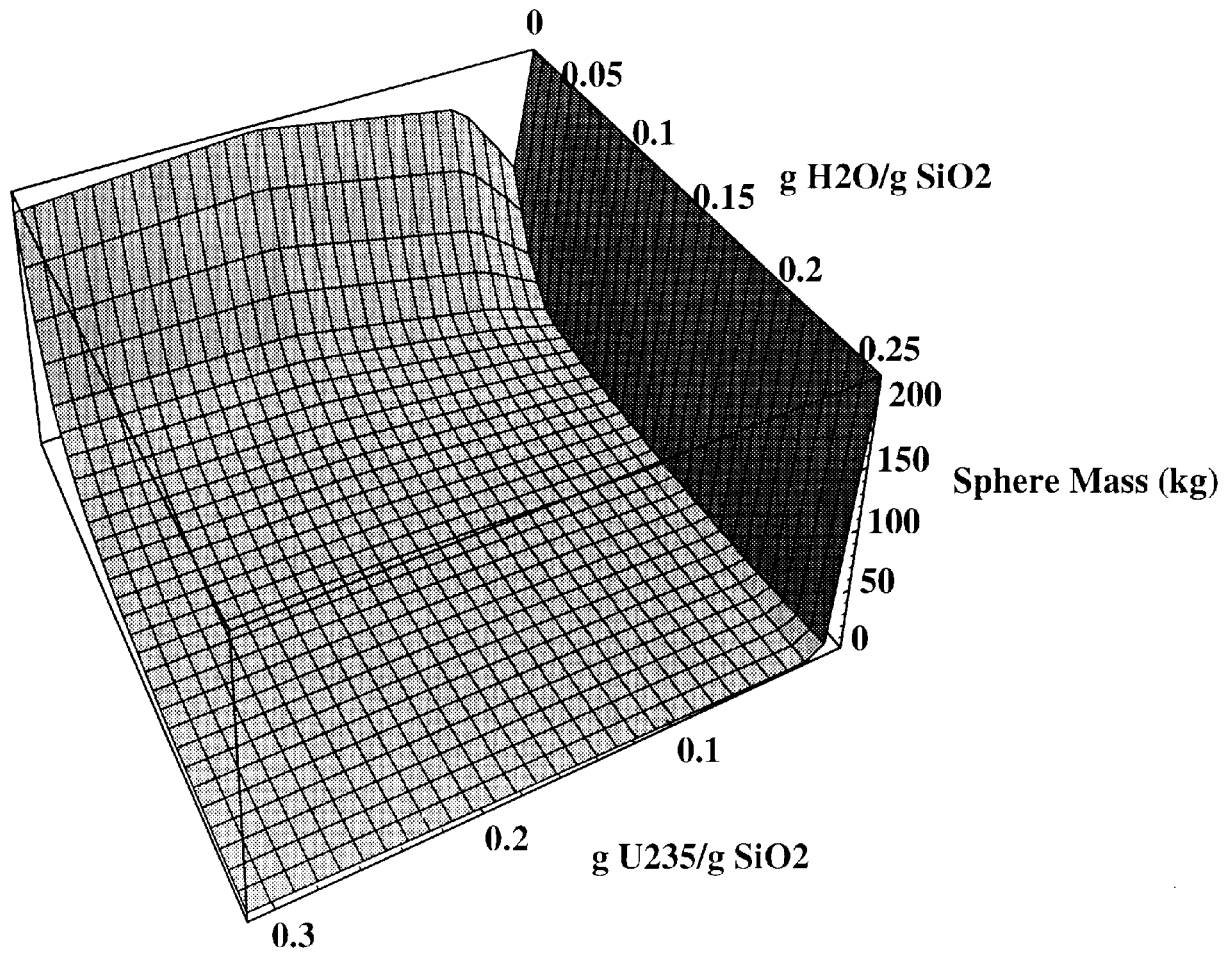

Figure D.1-17 SiO_2-soil critical sphere mass vs H_2O and ^{235}U concentration linear plot

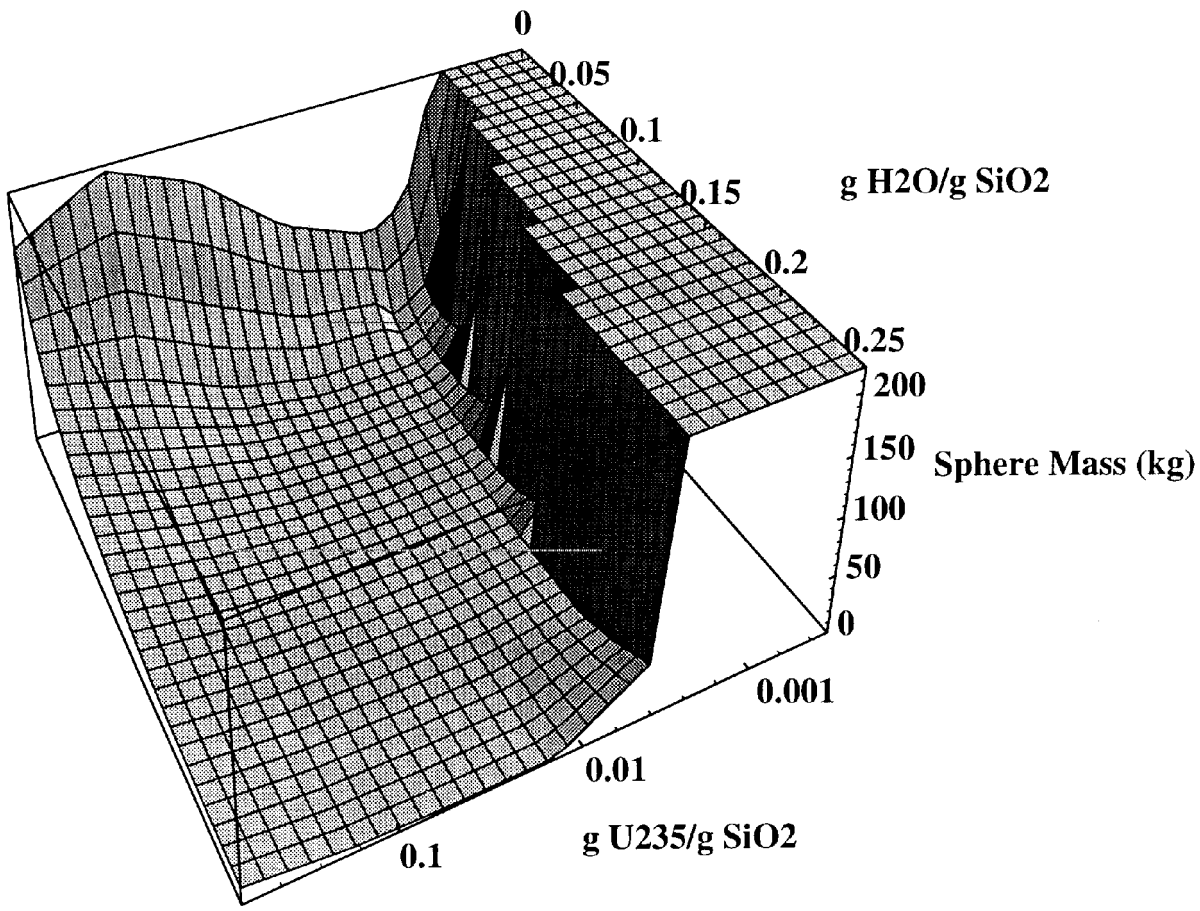

Figure D.1-18 SiO_2-soil critical sphere mass vs H_2O and ^{235}U concentration log plot

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(2-89)
NRCM 1102
3201, 3202

U.S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

(See instructions on the reverse)

1. REPORT NUMBER (Assigned by NRC, Add Vol., Supp., Rev., and Addendum Numbers, if any.)

NUREG/CR-6505, Vol. 1
ORNL/TM-13323/V1

2. TITLE AND SUBTITLE

The Potential for Criticality Following Disposal of Uranium at Low-Level-Waste Facilities

Uranium Blended with Soil

3. DATE REPORT PUBLISHED

MONTH	YEAR
June	1997

4. FIN OR GRANT NUMBER

L1376

5. AUTHOR(S)

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6. TYPE OF REPORT

Technical

7. PERIOD COVERED ***(Inclusive Dates)***

8. PERFORMING ORGANIZATION — NAME AND ADDRESS ***(If NRC, provide Division, Office or Region, U.S. Nuclear Regulatory Commission, and mailing address; if contractor, provide name and mailing address.)***

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P.O. Box 2008
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9. SPONSORING ORGANIZATION — NAME AND ADDRESS ***(If NRC, type "Same as above"; if contractor, provide NRC Division, Office or Region, U.S. Regulatory Commission, and mailing address.)***

Division of Waste Management
Office of Nuclear Material Safety and Safeguards
U.S. Nuclear Regulatory Commission
Washington, DC 20555-0001

10. SUPPLEMENTARY NOTES

11. ABSTRACT ***(200 words or less)***

The purpose of this study was to evaluate whether or not fissile uranium in low-level-waste (LLW) facilities can be concentrated by hydrogeochemical processes to permit nuclear criticality. A team of experts in hydrology, geology, geochemistry, soil chemistry, and criticality safety was formed to develop achievable scenarios for hydrogeochemical increases in concentration of special nuclear material (SNM), and to use these scenarios to aid in evaluating the potential for nuclear criticality. The team's approach was to perform simultaneous hydrogeo-chemical and nuclear criticality studies to (1) identify some achievable scenarios for uranium migration and concentration increase at LLW disposal facilities, (2) model groundwater transport and subsequent concentration increase via sorption or precipitation of uranium, and (3) evaluate the potential for nuclear criticality resulting from potential increases in uranium concentration over disposal limits. The analysis of SNM was restricted to ^{235}U in the present scope of work. The outcome of the work indicates that criticality is possible given established regulatory limits on SNM disposal. However, a review based on actual disposal records of an existing site operation indicates that the potential for criticality is not a concern under current burial practices.

12. KEY WORDS/DESCRIPTORS ***(List words or phrases that will assist researchers in locating the report.)***

uranium, low-level waste (LLW), special nuclear material (SNM), nuclear criticality, uranium migration, hydrogeochemical modeling

13. AVAILABILITY STATEMENT

unlimited

14. SECURITY CLASSIFICATION

(This Page) unclassified

(This Report) unclassified

15. NUMBER OF PAGES

16. PRICE