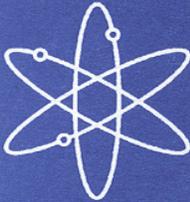




# **The Potential for Criticality Following Disposal of Uranium at Low-Level-Waste Facilities**



## **Containerized Disposal**



**Oak Ridge National Laboratory**



**U.S. Nuclear Regulatory Commission  
Office of Nuclear Regulatory Research  
Washington, DC 20555-0001**



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

## **Containerized Disposal**

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## 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 and test some reasonable 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 possible scenarios for uranium migration and concentration increase at LLW disposal facilities, (2) model groundwater transport and subsequent concentration increase via 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  $^{235}\text{U}$  in the present scope of work. The work documented in this report indicates that the potential for a criticality safety concern to arise in an LLW facility is extremely remote, but not impossible. Theoretically, conditions that lead to a potential criticality safety concern might arise. However, study of the hydrogeochemical mechanisms, the associated time frames, and the factors required for an actual criticality event indicate that proper emplacement of the SNM at the site can eliminate practical concerns relative to the occurrence and possible consequences of a criticality event.



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## EXECUTIVE SUMMARY

This work is Volume 2 of a two-volume study to address the potential for nuclear criticality resulting from increasing uranium concentrations in low-level-waste (LLW) disposal facilities. In contrast to Volume 1, which focused on  $^{235}\text{U}$  blended with soil, this report focuses on containerized waste with 10 weight percent (wt %)  $^{235}\text{U}$  in uranium, U(10). Hydrogeochemical modeling assumed precipitation of uranium in reducing zones instead of concentration on sorption sites as a means of forming a critical mass. As in the earlier study, several assumptions were made in developing the hydrogeochemical and criticality models. These are discussed in Section 5.

The criticality safety calculations showed that higher concentrations of  $^{235}\text{U}$  were needed for U(10) than U(100), as expected. Using the minimum concentration values necessary for a potential criticality, the mass of  $^{235}\text{U}$  as U(10) was 1.6 times greater than the  $^{235}\text{U}$  as U(100). Differences in  $^{235}\text{U}$  concentration have not been analyzed on a point-by-point basis and could be larger and smaller than the 1.6 value observed at the minimum concentration posing criticality concerns.

The mechanism of precipitation for increasing the concentration of uranium in assumed reducing zones under saturated conditions has been evaluated. These reducing zones formed very efficient barriers to uranium transport, precipitating nearly 100 wt % of the uranium in solution. The results of the geochemical modeling indicated that the reducing zone did not become oxidizing despite the influx of oxidized water. The source of reducing agents is postulated to be steel drums or wooden crates, thus serving as plausible locations for a buildup of uranium. Further study of the geometry of these reducing zones would be needed to evaluate the potential for concentrating relatively small critical masses (e.g., spherical masses). Other limitations may be identified through the evaluation of reducing zones.

Nonetheless, disposal practices at the Chem-Nuclear Systems, Inc., disposal facilities at Barnwell, S.C., restrict the possibility of criticality safety concerns in several ways. Very low-average  $^{235}\text{U}$  enrichments have been reported for most trenches, below the 1 wt % limit to produce a criticality concern under typical disposal conditions. For most trenches with higher  $^{235}\text{U}$  enrichments, the source term (e.g., mass) for uranium is too low to produce a slab of sufficient size with the required increase in concentration of  $^{235}\text{U}$  needed for criticality concern. One exception is Trench 23, which has a high enrichment (greater than 80 wt %) and a large source (175 kg) of  $^{235}\text{U}$ .

Even for the limited examples that potentially have sufficient  $^{235}\text{U}$ , very long times are needed to accumulate a critical mass. For the most conservative travel time, assuming one-dimensional (1-D) flow and no dispersion, tens to hundreds of thousands of years are needed. Three-dimensional (3-D) modeling indicates even longer times are needed when dispersion is incorporated. The flow paths would need to funnel the uranium from large trench volumes to relatively small reducing zones in order to increase the concentration of  $^{235}\text{U}$  to the level that would pose a criticality safety concern. If larger reducing zones form, either the  $^{235}\text{U}$  will be too diffuse to pose a criticality safety concern, or larger sources of  $^{235}\text{U}$  than are reported to be present in the trench are required. Uranium travel times are long enough to allow monitoring and possible mitigation of conditions that could pose criticality safety concerns.

This study results in the following recommendations for consideration of license reviews of LLW facilities:

1. Minimize those factors that enhance SNM accumulation.
  - Reduce groundwater infiltration
  - Reduce enrichment

## Executive Summary

- Minimize opportunities to create isolated zones of reducing conditions. Avoid organic matter in waste cells
  - Design trench to minimize focused flow
2. Limit the areal density of the fissile materials.
  3. Model trench performance using site-specific conditions on a scale that addresses the potential for criticality. Consequently, the observation that the average enrichment of a trench is less than 1 wt %  $^{235}\text{U}$  in the uranium does not necessarily eliminate a criticality concern for the trench. Burial reports may suggest that localized regions of a trench contain quantities of fissile material that greatly exceed the average enrichment.
  4. Continue to use sumps in disposal trenches to monitor for the presence of iron, organics, and uranium as indicators of mobility in the trenches. If uranium is observed in the sumps, determine its enrichment. Changes in redox conditions may be monitored by changes in different iron species. Even though it may take many years for sufficient buildup of uranium, early detection of mobile iron and uranium would indicate changes in the trench water chemistry.

Disposal trenches at the Barnwell, S.C., LLW facility have waste materials containing uranium with average  $^{235}\text{U}$  enrichments less than 1 wt %, insufficient masses of  $^{235}\text{U}$  at enrichments larger than 1 wt %, or distributions and mass proportions of  $^{235}\text{U}$  and  $^{238}\text{U}$  such that criticality safety concerns are not a realistic issue. For the single disposal trench, Trench 23, having a large mass of waste material containing highly enriched uranium, subcriticality is ensured by the physical distribution and commingling of the material with substantial quantities of "source material," which is typically normal or depleted uranium (e.g., 0.7 or 0.2 wt %  $^{235}\text{U}$  in uranium respectively). Uranium concentration factors (i.e., hydrogeochemical relocations and densification of uranium) larger than 10 are required to pose a potential criticality safety concern. As demonstrated by the evaluation of the reaction for hydrogeochemical cumulative uraninite precipitation for long time frames, it requires a minimum of 7,000 years to increase the  $^{235}\text{U}$  density from about 0.002 g/cm<sup>3</sup> to 0.02 g/cm<sup>3</sup>. It is judged that the same hydrogeochemical processes will redistribute and concentrate the commingled source material that is present, thereby further ensuring subcriticality through isotopic dilution of the SNM with normal or depleted uranium to  $^{235}\text{U}$  enrichments less than 1 wt %.

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# 1 PURPOSE

The purpose of this study was to evaluate the potential for hydrogeochemical processes to redistribute special nuclear material (SNM) in containerized low-level-waste (LLW) disposal facilities such that there is sufficient concentration increase and geometry reconfiguration to permit nuclear criticality. This particular evaluation is restricted to criticality safety concerns and geochemical processes associated with uranium. The approach was (1) to identify some reasonable scenarios for uranium migration and increase in concentration at LLW disposal facilities, (2) to model coupled groundwater transport and geochemical speciation of uranium, and (3) to evaluate the potential for nuclear criticality in terms of passive geometry configurations and increases in uranium concentration. A combination of hydrogeochemistry and criticality safety experts worked together to perform the evaluation.

This study extends the previous work reported in *The Potential for Criticality Following Disposal of Uranium at Low-Level Waste Facilities, Volume 1: Uranium Blended With Soil* (Toran et al., 1997). The present work emphasizes the disposal of containerized uranium instead of disposal of uranium blended in soil. Some different scenarios and concentrations are evaluated. In particular, the emphasis in this report is on the mobilization of uranium under oxidizing conditions, with immobilization under reducing conditions. In the previous report, uranium was desorbed from the soil and resorbed in a zone of high-sorption sites. This process did not increase the uranium concentration enough to cause nuclear criticality safety concerns, and the scenario will not be reexamined here. In addition, the previous criticality safety analysis assumed the uranium was enriched to 100 wt %  $^{235}\text{U}$  [referred to as U(100)]. In this report, additional calculations are presented for 10 wt %  $^{235}\text{U}$  in uranium [referred to as U(10)], and some comparisons are drawn between these U(10) calculations and the previous U(100) computational results reported in Vol. 1.

The Chem-Nuclear Systems, Inc. (CNSI), LLW disposal facility at Barnwell, S.C., was used as an example of site conditions for containerized disposal. Specific disposal practices at the site were evaluated as they related to the potential for the  $^{235}\text{U}$  concentration to increase.

However, the models were process-oriented rather than site-specific. That is, the models emphasized processes that could occur in disposal settings, rather than being a detailed construction of site conditions such as might be used in a performance assessment. Some details could not be addressed without a site-specific model that incorporates transient soil moisture conditions, or without additional data such as packing configurations and weathering rates. Assumptions were selected based on judgment regarding the potential conditions that would increase the possibility for criticality.

The questions addressed in this study are:

- Is there sufficient inventory for the available geometries requisite for criticality?
- How does the concentration of  $^{235}\text{U}$  needed for criticality compare with systems containing U(10) vs U(100)?
- What chemical conditions and physical aspects of trenches are conducive to increasing uranium concentration?
- Can reducing zones, which precipitate uranium, be sustained to enable critical masses to accumulate?
- How could disposal practices, in particular at Barnwell, S.C., enhance or mitigate the development of critical masses?

The questions addressed in this study reflect the important processes that could be evaluated with the available data, hydrogeochemical models and criticality safety analyses. The results provide bounds on conditions that could raise nuclear criticality safety concerns. Insights gained from the hydrogeochemical modeling and criticality safety analyses will be used as a basis for recommendations concerning future disposal practices for SNM.

## 2 PREVIOUS WORK

In the previous report, Vol.1 (Toran et al., 1997a), nuclear criticality evaluations and hydrogeochemical scenarios were based on licensed soil-contamination limits specified for Envirocare of Utah, Inc. The maximum average concentration of  $^{235}\text{U}$  permitted in disposed waste under the State of Utah license (UT 2300249) is 770 pCi  $^{235}\text{U}/\text{g}$  soil, which equates to about 0.0006 g of  $^{235}\text{U}$  per  $\text{cm}^3$  of soil given a soil bulk density of about 1.6 g soil/ $\text{cm}^3$ . If disposal occurred at this maximum concentration, there is the theoretical possibility of a nuclear criticality accident, given assumptions about hydrogeochemical influences on reconfiguration of the uranium. Only a narrow range of conditions resulted in sufficient increase in uranium concentration, and the length of time required to increase the concentration of uranium is expected to be many thousand of years. This slow approach to criticality will further mitigate consequences that occur with rapid approaches to critical conditions.

However, it is important to note that reviews of disposal records from Envirocare of Utah, Inc., indicate that concentrations of  $^{235}\text{U}$  in the waste material are more than a factor of 10 less than allowed by the license, and that the average site  $^{235}\text{U}$  enrichment is below the minimum 1 wt % (Pruvost and Paxton, 1996) required to achieve nuclear criticality. Thus the likelihood of a criticality accident is vanishingly small.

Because of the numerous combinations of parameters that could be considered in nuclear criticality evaluation and hydrogeochemical modeling, bounding and simplifying assumptions were used in the analysis. Nuclear criticality evaluations were performed for simple geometries using two generic soil types:  $\text{SiO}_2$  soil (the more conservative medium because pure  $\text{SiO}_2$  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.

Potential, direct radiation exposures were estimated for two postulated types of criticalities: one with a high concentration factor (large increase in  $^{235}\text{U}$  concentration) and one with a low concentration factor. The locations of the determined radiation exposures were for two positions 1 m above grade. One position was directly above the deposit, and the other position was 90 m away from the deposit. The assumed fission yields from both type systems were based upon the fission energy release necessary to remove the quantity of water that is necessary to moderate neutrons to sustain nuclear criticality throughout an over-moderated condition. These assumptions were predicated upon a geologically slow approach to a non-idealized critical geometry, thereby permitting localized steam generation and self regulation and shutdown of the fission-chain reaction. Alternative idealized assumptions have been postulated by others (Bowman and Venneri, 1994; Greenspan, Armel, Ahn and Vujic, 1997) that present more severe consequences.

The criticality evaluation showed that the  $\text{SiO}_2$ -soil results are similar to the nominal soil results. In terms of the hydrogeochemical processes that can increase uranium concentration, the critical slab configurations are more readily achieved than cylindrical or spherical configurations (i.e., lower concentration factors are required). The criticality evaluation also provided a minimum concentration needed to achieve criticality safety concerns, which was the target concentration for hydrogeochemical modeling.

Simplifying assumptions in the hydrogeochemical modeling included one-dimensional (1-D) transport and saturated conditions. The hydrogeochemical scenario was postulated based on the geometry of the Envirocare site with disposal in soil containing sorbed uranium, then mobilization to a zone of higher-sorption capacity below the disposal trench. This sorption zone could potentially contain uranium in a zone of higher concentration. A reducing zone to capture uranium was also hypothesized, but not explicitly modeled, in the previous study. A reducing zone was difficult to define given the limited supply of reducing agents in the trenches and the unsaturated conditions in soil, which would keep the system oxygenated. A sensitivity analysis

was performed to evaluate various factors, such as concentration of complexing agent, quantity of initial uranium source term, and groundwater velocity on the potential to increase uranium concentration.

The previous work noted that the concentration of complexing agent and the size of the source term were limiting factors in the reconfiguration of uranium. For most scenarios, once sufficient uranium was mobilized, the concentration of a complexing agent was important because it outcompeted sorption sites in the high-sorption zone and prevented increases in uranium concentration. The possibility of immobilizing uranium in reducing zones was presented as a more-likely scenario (to be evaluated in the present work). Furthermore, if the initial concentration of uranium could be limited during disposal (e.g., by limiting disposal thickness), it would not be possible to increase the uranium concentration sufficiently along a 1-D flow path to pose a criticality safety concern. Much uncertainty exists in the estimates of the time frame for the increase in uranium concentration, but analogs from soil-forming processes suggest that these processes can require thousands of years.

Volume 1 of this report provided the following recommendations for consideration during a license review of LLW facilities having uranium blended with soil:

1. Minimize the factors that enhance the increase in the concentration of uranium. For example, reduce water infiltration, dilute the  $^{235}\text{U}$  by reducing the enrichment, and minimize opportunities to create zones of reducing potential that precipitate uranium readily (e.g., by maintaining unsaturated conditions, and avoiding organic matter in waste cells to prevent methanogenesis).
2. Limit the areal density of uranium to a safe value by limiting the licensed depth of the disposal cell and the licensed disposal concentration. Results suggest that criticality safety concerns can be reduced or eliminated even under worst-case hydrogeochemical transport by reducing the disposal cell depth.

### 3 SITE DESCRIPTION

The parameters used in the models are based on site conditions at the CNSI LLW disposal facility in Barnwell, S.C., although not all physical and chemical conditions were explicitly modeled. A variety of geologic and hydrologic information on the site is available from previous studies (e.g., Weiss and Colombo, 1979; Cahill, 1982; Dennehy and McMahon, 1987; and data provided by CNSI).

The Barnwell facility was opened by CNSI in 1971. The facility receives approximately 8490 m<sup>3</sup> (300,000 ft<sup>3</sup>) to 11320 m<sup>3</sup> (400,000 ft<sup>3</sup>) of LLW per year. Barnwell receives Class A, B, and C waste. The majority of the SNM is contained in Class A waste. These waste classifications are defined in 10 CFR 61.55.

About two-thirds of the waste disposed at the Barnwell facility comes from nuclear power plants, with the other third coming from other industry and government sources (such as the U.S. Army). The SNM consists primarily of <sup>235</sup>U. The <sup>235</sup>U enrichment is typically less than 10 wt % for trenches with enrichment data available. Approximately 90 wt % of the SNM is dry active waste. Other waste includes resins, which have been separated from the dry waste since the mid-1980s. Scintillation vials and other organic liquids were banned in 1979.

Originally, waste was received and buried at the Barnwell facility in containers, such as cardboard boxes and drums. During the 1980s, shipments more commonly came in plywood boxes. Then disposal requirements became more stringent, and steel drums (then high-density polyethylene containers) became the standard in the mid to late 1980s. These containers are now overpacked in concrete vaults or cylinders. The containers are disposed in trenches that today are typically 30,480 cm (1000 ft) long, 6,096 cm (200 ft) wide, and 762 cm (25 ft) deep. Smaller trenches were used in the past and are used today for high-concentration radioactive waste (Class C).

The trenches are excavated through the surficial sand into clayey sand. Prior to waste placement, buffer sand is placed on the bottom of the trench. The bottom of the trench is sloped, and a French drain is used to move water to sumps, where it can be sampled for detection of contaminant movement (Fig. 3.1). In the past, the French drains were constructed on one side of the trenches and consisted of gravel. In the late 1980s to early 1990s, slotted plastic pipes within high-permeability material were used to collect water on both sides of the trench. When the trenches are full, they are backfilled with sand, then covered with earthen caps. New high-density polyethylene caps have been emplaced on some older trenches to further inhibit water from infiltrating the trenches.

From January 1970 to 1984, there were 12 amendments to the original SNM disposal specifications that influenced nuclear criticality safety. The original license included a 200-g possession limit for all SNM with no package limit. The license was amended to increase the possession limit and included package limits of 15 g to 50 g. Later amendments included mass and spacing limitations for accumulations of packages based on disposals from specific generators. In 1981, an areal density limit of 200 g/ft<sup>2</sup> was imposed. In addition, the waste form requirements and practices varied over time until 1984 when the 10 CFR Part 61 criteria were implemented.

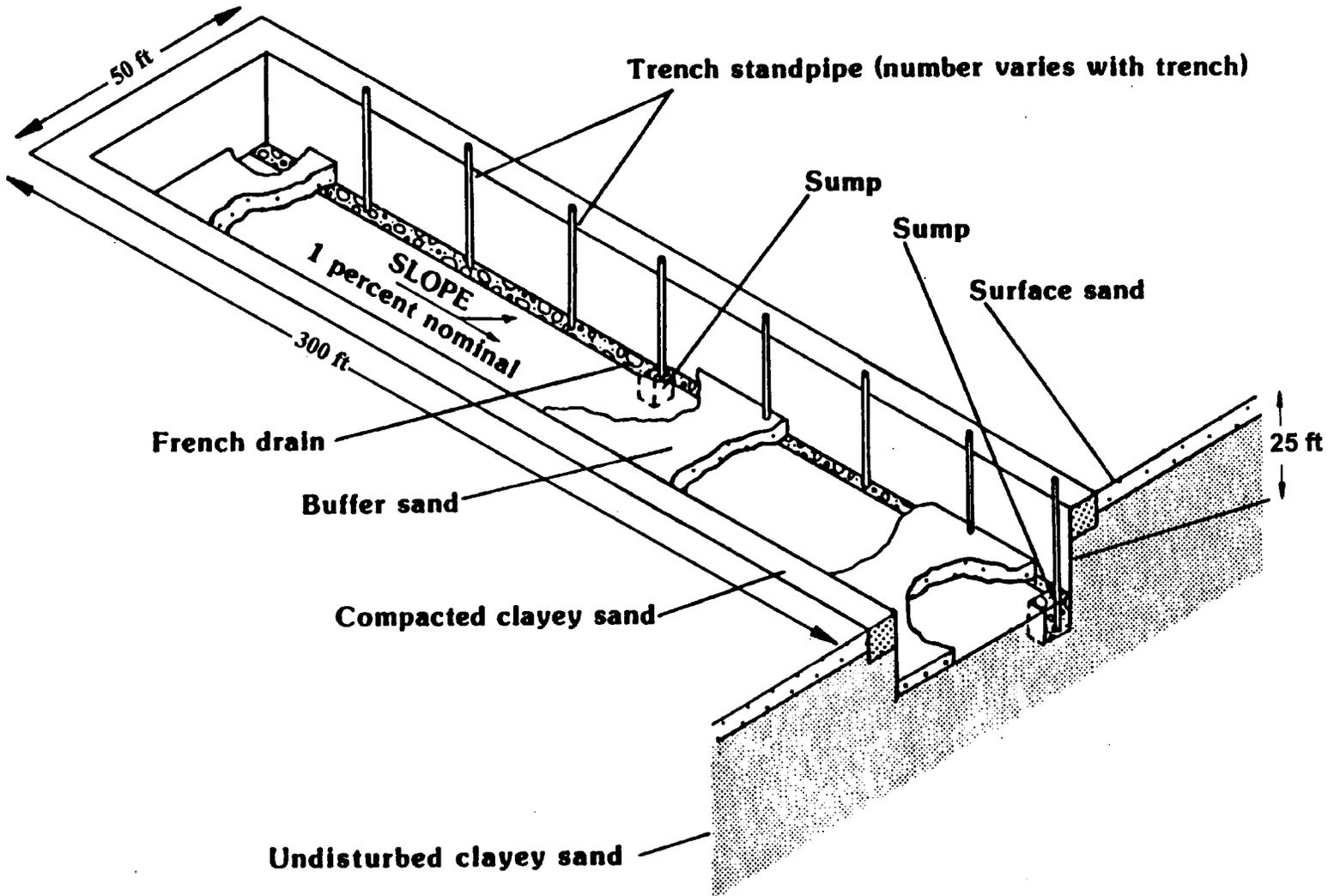


Figure 3.1 Typical construction of waste trenches. Graded bottom is lined with capture water for French drain. Location of drains varies. Some typical dimensions are shown. Taken from Dennehy and McMahon (1987)

CNSI's last Radioactive Material License (No. 12-13536-01) with the NRC allowed 350 g of  $^{235}\text{U}$  per package. The 350-g limit pertains to  $^{235}\text{U}$ , which represents the vast majority of the SNM waste. Other SNM, such as  $^{233}\text{U}$  and isotopes of plutonium, make up less than 1 wt % of the total grams of SNM. Shipments commonly contain less than 100 g per package. Any shipment with 1 g or more SNM is reportable. Shipments of SNM are presently placed only at the bottom of the trench if they contain at least 30 g of SNM. In the past, configuration of the SNM in trenches was determined by "operational randomness"; that is, packages of waste containing SNM were not placed on top of packages already emplaced with SNM. These blocks were typically 304.8 cm (10 ft)  $\times$  304.8 cm (10 ft)  $\times$  365.76 cm (12 ft) in size, then later 762 cm (25 ft)  $\times$  762 cm (25 ft)  $\times$  1524 (50 ft).

Disposal records provided for this project by CNSI vary in detail. Specifically, the records for Trenches 1–35 (herein referred to as the "old" trenches) provide only total SNM mass, whereas the records for "new" trenches (38–87), specific isotopes of uranium have been identified, so grams of  $^{235}\text{U}$  are available and average trench enrichment can be calculated. No data on disposal amounts were provided for Trenches 36 and 37. The disposal record data indicate that, in general, smaller quantities of SNM were disposed of in the "old" trenches. Most trenches have less than 40 kg, with masses ranging between 0.5 kg and 175 kg (Fig. 3.2). In the new trenches, disposed quantities of  $^{235}\text{U}$  range from just a few grams to 1600 kg, with most trenches containing less than 300 kg.

The Barnwell facility is located on the Atlantic Coastal Plain. The surficial deposit at the site is known as the Tobacco Road Formation (formerly the Hawthorn Formation). The deposit is approximately 1828.8 (60 ft) to 2438.4 cm (80 ft) thick, and contains dominantly a sandy clay [e.g., 85 wt % quartz (Pietrzak et al., 1982)] with coarse sand occasionally present near the base. The present-day water table is within this deposit, typically around 1066.8 cm (35 ft) below the land surface. Beneath the Tobacco Road Formation is the Dry Branch Formation (formerly the Barnwell Formation), which is a massive medium-grained sand. The permeability of the surficial deposit varies from  $3 \times 10^{-3}$  to  $2 \times 10^{-8}$  cm/s (Cahill, 1982, p. 38) based on laboratory tests on core collected in the region from a variety of depths. Dennehy and McMahon (1987, p. 28) analyzed shallow cores near experimental trenches and found a permeability range from  $7 \times 10^{-3}$  to  $7 \times 10^{-8}$  cm/s. Field measurements using slug tests tend to the upper end of the hydraulic conductivity (Cahill, 1982, p. 38). Porosity of the deposit is estimated to be around 40%. Effective porosity typically is somewhat lower, around 30%, but the porosity of the waste matrix could be higher, up to 40 to 50%, as reported at other waste sites (Spalding, 1987).

Soil moisture above the water table is typically high due to the humid, wet climate. Cahill (1982) reports soil moisture measurements made over 1.5 years, with values typically greater than 90%. The annual rainfall is about 114.3 cm (45 in.) per year, but only 35.56 cm (14 in.) to 43.18 cm (17 in.) per year is expected to infiltrate the regional flow system (Dennehy and McMahon, 1987). Estimates of infiltration in the disturbed area around the trenches with earthen caps have not been reported but may be higher due to runoff from the caps.

Groundwater velocity in the trenches has been estimated through tracer tests and groundwater modeling. Dennehy and McMahon (1987) constructed experimental trenches similar to waste disposal trenches and monitored water levels, soil moisture, and a salt tracer to estimate groundwater travel times from and within the trenches. Salt granules (NaCl) were placed at the bottom of the experimental trenches and at or near the land surface. Detection of the tracer in monitoring points was used to estimate the velocity of  $3 \times 10^{-6}$  cm/s in the cap and around  $6 \times 10^{-5}$  cm/s in the backfill material. Cahill (1982) estimated a similar lower vertical velocity of  $2.5 \times 10^{-6}$  cm/s in a regional groundwater flow model in the area. Both the upper and lower ranges of values were used in modeling here.

U-235 Inventories in the Barnwell LLW Facility

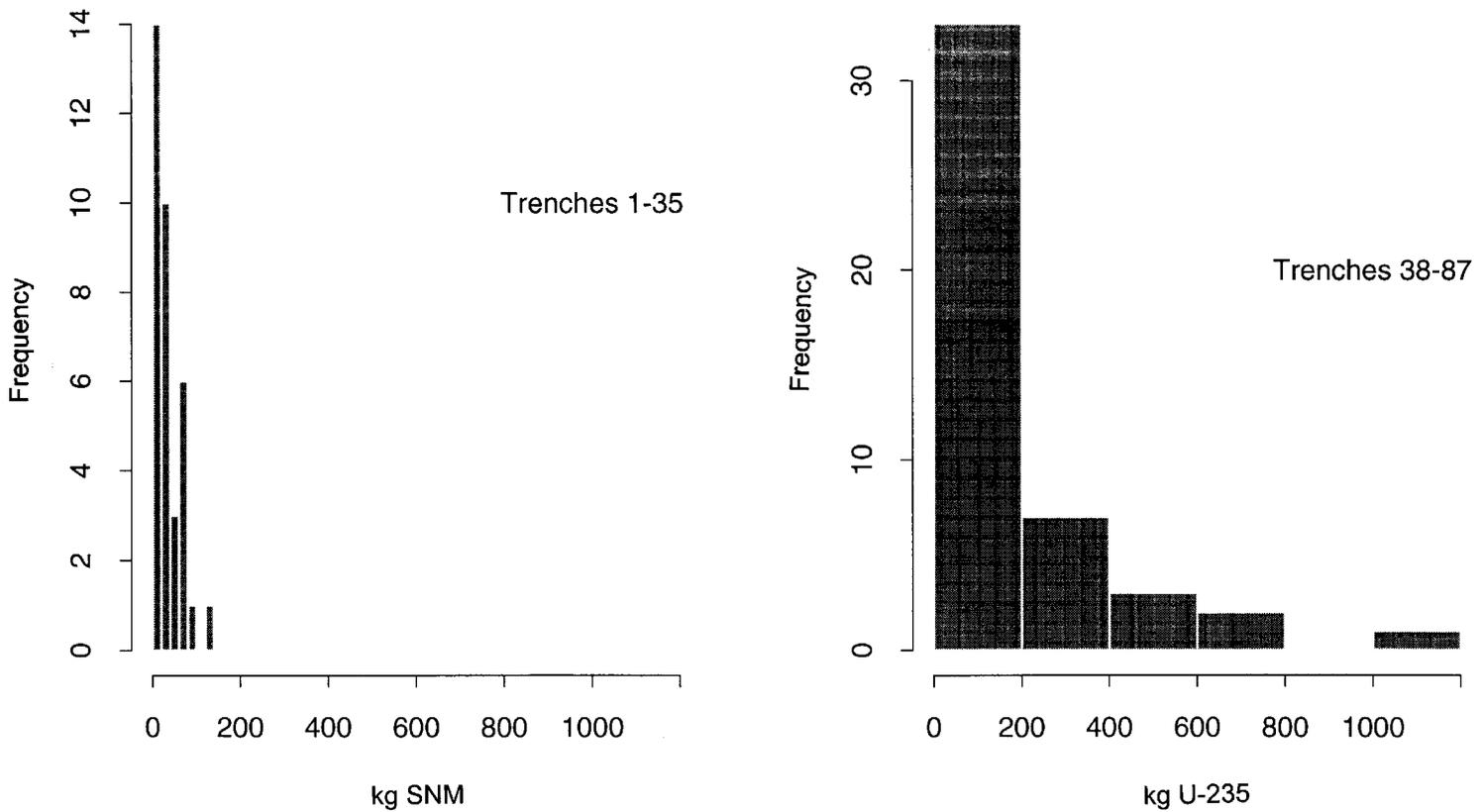


Figure 3.2 Histograms of source material in trenches. Grams of SNM or <sup>235</sup>U per trench are given for (a) pre-1981 trenches, SNM reported, and (b) post-1981 trenches, <sup>235</sup>U reported. Older trenches have lower source terms

Monitoring of the sumps and other wells surrounding the waste facilities has detected a tritium plume about 914.4 m (3000 ft) in length. Tritium is likely to travel at the velocity of groundwater, being an isotope of water, and provides an early warning system of leakage in the trenches. Tritium was first detected outside the trenches in 1978 and measured in an off-site well in 1990 along the base of the Tobacco Road Formation. Additional caps were installed over the oldest trench area first to help minimize future migration of tritium. Ichimura et al. (1994) report that the travel time of the tritium is similar to estimates of velocity in the fast horizontal flow zone of  $1 \times 10^{-4}$  cm/s beneath the trenches. Cobalt-60 and organics have also been detected below the site (Cahill, 1982).

Trench water chemistry reported by Weiss and Colombo (1979) is presented in Table 3.1. Although the chemistry is variable, it indicates moderately oxidizing conditions. However, this does not mean that reducing zones will not form. Zonation of redox species in landfills has been reported elsewhere, and analogues from other sites are useful to consider here.

Table 3.1 Concentration of dissolved nonmetals and metals in trench water samples taken at the LLW burial site near Barnwell, S.C. (Weiss and Colombo, 1979)

Dissolved component (mg/L)	Trenches				
	3	5	6	8	25/21 <sup>a</sup>
Total alkalinity (as CaCO <sub>3</sub> )	100	200	40	600	80
Inorganic carbon	24	-	11	130	38
Dissolved organic carbon (DOC)	7	-	2	170	12
Chloride	7	10	90	85	42
Nitrogen (N) (ammonia)	0.3	- <sup>b</sup>	1.4	59	25
Nitrogen (N) (NO <sub>2</sub> <sup>-</sup> + NO <sub>3</sub> <sup>-</sup> )	<0.04	<0.1	23	8.0	15
Silica	4.3	7.6	5.8	6.0	5.0
Sulfate	<5	7	18	34	56
Total anions (meq/L)	2.3	4.4	4	16	5.7
Calcium	4.0	3.2	16	34	21
Iron	0.15	1.5	0.4	1.2	0.2
Magnesium	2.5	3.3	1.0	18	3.3
Manganese	0.24	0.34	0.45	0.72	0.32
Potassium	1.0	4.6	1.4	12	3.5
Sodium	2.3	20	29	87	37
Total cations <sup>c</sup> (meq/L)	0.55	2.9	2.3	12	4.8

<sup>a</sup> Trenches 25 and 21 are reported together.

<sup>b</sup> Insufficient sample for analysis.

<sup>c</sup> Includes nitrogen as .NH<sub>4</sub><sup>+</sup>

Relatively few studies detail the redox conditions in landfills because of the problematic nature of obtaining reliable measurements. One of the main difficulties is that not all redox couples are in equilibrium, so a given measurement may not be relevant to all redox couples (Lindberg and Runnells, 1984). A series of redox reactions may occur in zones around landfill leachate (Baedecker and Back, 1979; Christensen et al., 1994). The zones can be identified by patterns in water chemistry, such as concentration of redox couples, evolving from the aerobic zone, to nitrate-reducing, iron-reducing, sulfate-reducing, then methanogenic (Fig. 3.3). The range of redox variation in landfills is quite large and controls the mobilization and concentration of many redox-sensitive species. The observed values range from -200 to +600 mV over distances of hundreds of meters. Similar redox variation has also been observed in natural systems (Champ et al., 1979) on the scale of hundreds of meters to kilometers.

Although the zones may be extensive horizontally, they are often quite thin vertically, limited by the lack of vertical mixing in the landfill plume. These zones develop over decades, but may initiate on the order of 5 years [e.g., the Bemidji spill (Baedecker et al., 1993)]. Some of the key factors in the development of zones are the mineralogy of the sediments [in particular, availability of iron minerals, according to Heron et al. (1994)], the organic carbon content, microbial degradation rates, and the moisture content. The size of the different zones also depends on the landfill size, vertical mixing, and rates of groundwater flow.

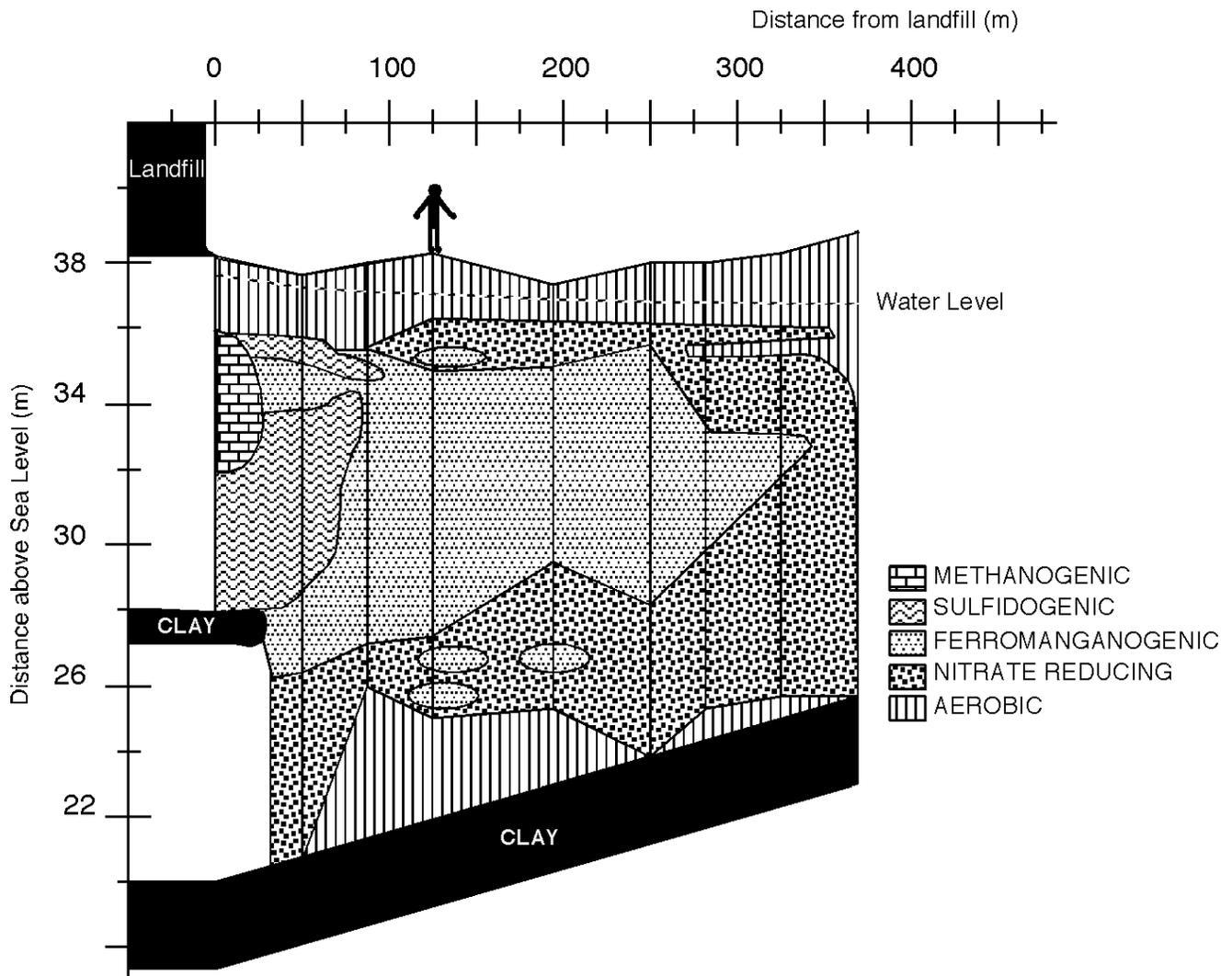


Figure 3.3 Example of redox zonation in a landfill cross section. Five zones from methanogenic to aerobic were mapped in multilevel piezometers, B1 through B9. Reducing zones extend over 350 m from landfill source. Adapted from Christensen et al., 1994



## 4 APPROACH

### 4.1 NUCLEAR CRITICALITY EVALUATION

#### 4.1.1 Code Description and Validation

The SCALE (1995) code system was used to calculate the  $k_{\text{eff}}$  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 is performed using the NITAWL and BONAMI codes. For this study, the XSDRNPM code was executed by the CSAS module to provide the  $k_{\text{eff}}$  values. XSDRNPM is a deterministic code that solves the Boltzmann equation for neutron transport in a 1-D mathematical 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 May 30, 1995. The Brookhaven Evaluated Nuclear Data File B Version V (ENDF/B-V) point cross-section library, which was collapsed to a 238-neutron-energy group library (Greene, 1994), named REF01.XN238, was created 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  $^{235}\text{U}$  systems before and after the evaluations performed for this study. The verification and validation benchmark calculations provided identical results for calculations performed before the study as those performed 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  $\sim 0.5\%$  of the experimental values; that is, the calculated  $k_{\text{eff}}$  values of the 14 critical experiment benchmarks were between 0.9954 and 1.0064. See Vol. 1 for more details.

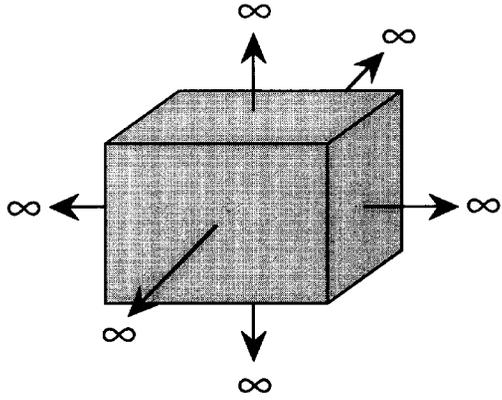
#### 4.1.2 Analytical Approach

The analytical approach taken for the nuclear critical evaluation was performed in two segments. The first segment was to evaluate the infinite-media multiplication constant,  $k_{\infty}$ , of a fixed-density  $\text{SiO}_2$  soil matrix having differing degrees of  $^{235}\text{U}$  and water contents or densities within the soil. These results provided indications of the combinations of  $^{235}\text{U}$ , soil, and water that could support self-sustaining nuclear fission chain reactions in an essentially infinite sea of material (i.e.,  $k_{\infty} \geq 0.95$ ). The second segment involved examining three geometries that have relevance to the evaluation: spheres, cylinders of infinite length, and slabs of infinite extent (Fig. 4.1). In Fig. 4.1, the dimension  $r + 4$  m refers to the determined critical radius plus 4 m of uncontaminated  $\text{SiO}_2$  and water, and the dimension  $h + 8$  m refers to the thickness of the determined critical slab plus 8 m of  $\text{SiO}_2$  and water. The evaluations of the infinite slabs approximate the effects of the  $^{235}\text{U}$ , contaminating the soil-like waste, settling vertically onto a waste-cell floor and are consistent with previous evaluations (Hopper et al., 1995) performed for reviewing LLW facilities.

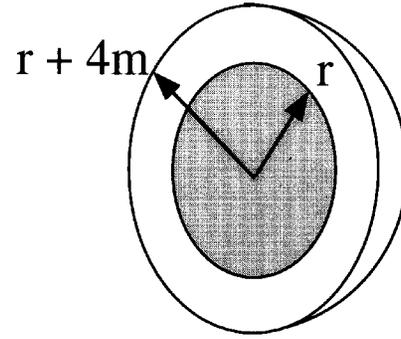
#### 4.1.3 Parameters

Consultations among Oak Ridge National Laboratory (ORNL) and U.S. Nuclear Regulatory Commission (NRC) staff, evaluating the CNSI disposal records, permitted the inference of a representative uranium enrichment for

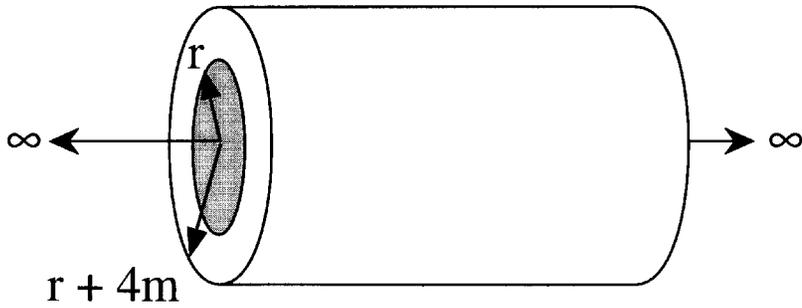
Infinite Media Model



Spherical Model



Infinite Cylinder Model



Infinite Slab Model

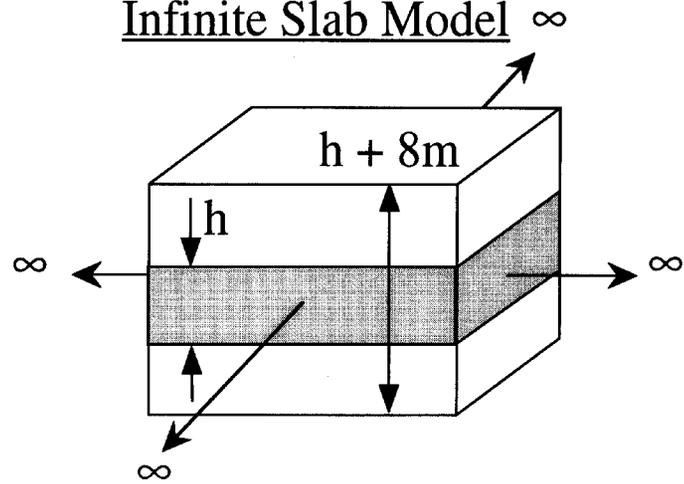


Figure 4.1 Conceptual configuration for nuclear criticality evaluations

the nuclear criticality computational evaluations. Though CNSI has been licensed to receive and dispose of any enrichment of uranium, the assumed arbitrary representative uranium enrichment for the computational evaluations was 10 wt %  $^{235}\text{U}$  in uranium.

The nuclear criticality computational studies that are reported in Vol. 1 were performed for 100 wt %  $^{235}\text{U}$  in uranium, U(100), and water in one of two hypothetical waste matrixes. Both waste matrixes without uranium or water had identical bulk densities (i.e.,  $1.6 \text{ g/cm}^3$ ) and equivalent 0.4 void fractions. The basic matrix called "Nominal Soil (N-S)" consisted of "average" weight fractions of earthen elements and the matrix called "SiO<sub>2</sub>-Soil, (S-S)" consisted of only SiO<sub>2</sub>. The nuclear criticality computational results in Vol. 1 demonstrated that the ratio of critical uranium areal densities for the S-S matrix, divided by the critical uranium areal densities for the N-S matrix, was generally on the order of 0.7. The S-S matrix was chosen for this study to provide conservative estimates for the lesser uranium enrichment of 10 wt %  $^{235}\text{U}$ . Furthermore, the sand at Barnwell may be more like S-S than N-S.

The reported results from this study are for various concentrations of 10 wt %  $^{235}\text{U}$  in uranium, U(10), and water in a  $1.6\text{-g SiO}_2/\text{cm}^3$  hypothetical waste matrix as it may relate to the LLW facility operated by CNSI near Barnwell, S.C. The results are presented in tabular and graphic form, followed by discussions about the relevance of the results to practical initial disposal conditions. Some comparisons are drawn between the U(10)-H<sub>2</sub>O-SiO<sub>2</sub> and previous U(100)-H<sub>2</sub>O-SiO<sub>2</sub> computational results that were reported in Vol. 1.

The lowest  $^{235}\text{U}$  concentration in the nuclear criticality evaluations is the concentration of  $^{235}\text{U}$  used in Vol. 1 of this study, which was the permissible State of Utah license limit for  $^{235}\text{U}$ . Although this concentration is not relevant to disposal as containerized waste, it is below the level of concern for nuclear criticality safety and can be used as a starting point. However, it is no longer a reference point for a concentration factor, as used in Vol. 1.

## 4.2 HYDROGEOCHEMICAL MODELING

### 4.2.1 Conceptual Model

As stated previously, a process-oriented model was developed to evaluate hydrogeochemical mechanisms that would increase the concentration of uranium in disposal settings. The model was not intended to provide site-specific predictions, but data (where available and applicable) from the Barnwell site were used in the model. The specific process modeled was the reduction and precipitation of oxidized uranium in reducing zones. The interplay between reducing zones and oxidizing water that infiltrates requires a coupled transport and geochemistry model because, instead of just a single component (uranium), the transport of multiple components (e.g., uranium, oxygen, complexing agents, competitive electron acceptors such as  $\text{Fe}^{3+}$ ) is involved.

This study focused on an increase in concentration of uranium at a hypothetical boundary between oxidized and reduced zones, rather than the development of zones. The reducing agent was assumed to be either elemental iron, which represented the 55-gal drums, or methane ( $\text{CH}_4$ ), which represents the organics contained in the waste and the cardboard, plastic, or wood containers. This study did not evaluate kinetic aspects or time variant infiltration.

One-dimensional (1-D) transport of uranium through the trench was assumed. Even though three-dimensional (3-D) transport is more realistic, dispersion will reduce the concentration of uranium transported from one point to another, as shown in selected runs. By using 1-D flow, the results of the modeling will be conservative: the

travel times will be shortest, and the concentrations will be maximized. Transport pathways that would mimic vertical flow through the trench and horizontal flow along the drainage systems can be modeled with two different 1-D legs.

In summary, conditions that tend to enhance the potential for increasing uranium concentration were modeled, but only if judged to be within reasonable bounds, or the limitations could be specified. A detailed description of model assumptions and further discussion are provided in Sect. 5.

#### 4.2.2 Models Used

Preliminary hydrogeochemical modeling was conducted using two codes: PHREEQC and ParSSim. PHREEQC (Parkhurst, 1995) is a chemical speciation code that models 1-D transport using mixing cells. It has a fairly complete geochemical database, but neglects transport effects such as dispersion, which can reduce concentrations. ParSSim (Wheeler et al., 1997) runs on a supercomputer and incorporates full transport and user-defined geochemical reactions in a multidimensional, multispecies transport code. This code permits more realistic simulations, but is also less stable numerically and more time-consuming to use (in terms of run time and output analysis). Not all cases that run for PHREEQC were run successfully with ParSSim. However, a representative 3-D problem has been run successfully with ParSSim, which provided some error bounds on the simplified PHREEQC modeling.

Both codes were tested by comparison with a field and modeling problem involving nitrate removal by oxidation of pyrite, which creates a sharp redox front (Engesgaard and Kipp, 1992). Numerical methods in the codes were selected to help code stability over the large concentration ranges resulting from redox problems (Toran et al., 1997b).

#### 4.2.3 Parameters and Model Grid

The model grid represented a 1-D flow field. A continuous input of oxidized water containing dissolved uranium was introduced into a reduced zone and allowed to flow through the reduced zone at velocities of  $1 \times 10^{-5}$  and  $1 \times 10^{-6}$  cm/s. The model grid was 5 m long and represented a reduced zone (Fig. 4.2). The 5-m length was selected arbitrarily to monitor movement of the reduction boundary during the injection. In most cases, the boundary stayed within the model grid. The grid began at the redox boundary because this is the location where most of the geochemical alterations are presumed to occur. This approach neglects some interactions within the oxidized zone in the natural flow field, but these interactions are likely to be less important than the reactions taking place at the redox boundary.

The oxidized water assumed in this modeling exercise was geochemically similar to trench water at the CNSI site (Trench 25/21, Table 3.1) with an assumed (rather than measured) concentration of uranium. The uranium was input as a dissolved species. The uranium concentration was varied in the different runs, with values ranging from 1 to 20 mg/L. The initial uranium concentration is limited by mineral solubility. For example, in this water, dissolution of schoepite results in an equilibrium concentration of about 20 mg/L (based on PHREEQC modeling). Furthermore, tens of mg/L of dissolved uranium has been observed in water running off of uranium mill tailing piles in oxidizing environments. A lower limit of 1 mg/L uranium was selected as the input concentration for most runs, a reference concentration used in previous studies (Sims et al., 1993). Infiltrate water in the model column is also represented by the geochemistry of Trench 25/21, with a low uranium concentration of 0.01 mg/L.

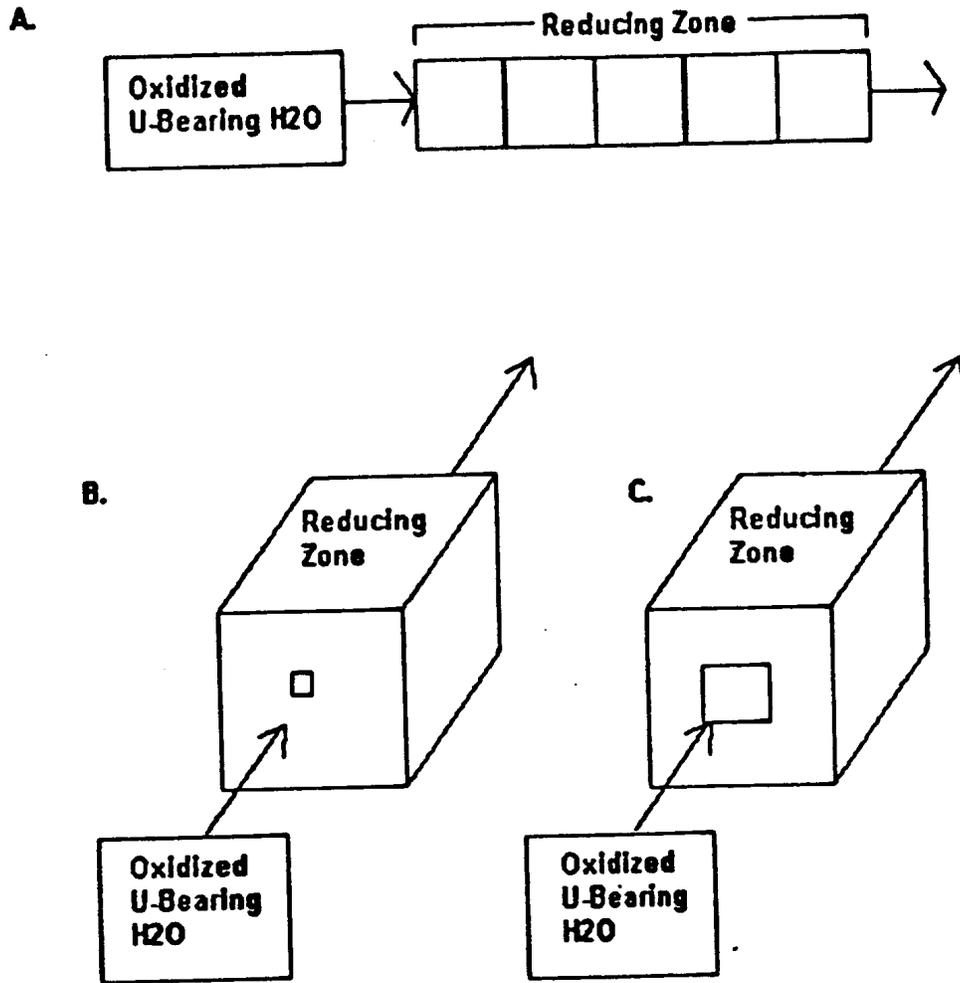


Figure 4.2 Schematic of model grids showing oxidized injection into reducing zone. A. One-dimensional model used in PHREEQC and 1-D ParSSim evaluation. B. Three-dimensional ParSSim model with small source term, allowing dispersion in three dimensions. C. Three-dimensional ParSSim model with larger source term, limiting 3-D dispersion along centerline of plume

The oxygen content of the trench water is not well known; Eh and dissolved oxygen were not reported in CNSI monitoring data or the hydrologic summary of Cahill (1982). A few (5) dissolved oxygen measurements were reported by Weiss and Colombo (1979), ranging from 0.1 to 1.5 mg/L. Calculations of the redox potential of the Trench 25/21 data also indicate an oxidizing Eh (on the order of 400 mV). The undersaturated conditions of the trench also suggest dissolved oxygen is present in the trench water. A base-case value of 2 mg/L dissolved oxygen was selected, and the value was varied in the sensitivity analysis (see below).

Ten components from this background water were selected to form the basic components for the geochemical modeling (Table 4.1). From these components, equilibrium species form and minerals can be selected for equilibration. For PHREEQC, the database contains an extensive list of complexed species. For ParSSim, the user identifies key complexes for equilibration, and a more limited number of complexes is favorable for faster convergence. The complexes selected for equilibration in ParSSim (Table 4.1) were based on PHREEQC modeling, which indicated those complexes formed in significant concentrations (at least one order of magnitude greater concentration than other complexes of the same component).

Table 4.1 Components and reactants (minerals and aqueous complexes) used in ParSSim<sup>a</sup>

Components	mg/L	Products	log(K <sub>eq</sub> )	Minerals/Phases	log(K <sub>sp</sub> )
H <sup>+</sup>	5.9 (pH)	OH <sup>-</sup>	-14.00	FEMETAL	-84.0
CO <sub>3</sub> <sup>-2</sup>	80	H <sub>2(aq)</sub>	-44.67	CH <sub>4</sub>	-130.9
Ca <sup>+2</sup>	21	HCO <sub>3</sub> <sup>-</sup>	10.35	UO <sub>2</sub>	-27.72
Na <sup>+</sup>	37	H <sub>2</sub> CO <sub>3</sub>	16.68	Fe <sub>2</sub> O <sub>3</sub>	- <sup>b</sup>
Mg <sup>+2</sup>	3.3	U(OH) <sub>4(aq)</sub>	-41.018	Fe(OH) <sub>3(a)</sub>	- <sup>b</sup>
Fe <sup>+3 c</sup>	0.2	UO <sub>2</sub> (CO <sub>3</sub> ) <sub>2</sub> <sup>-2</sup>	17.00	FeS <sub>2</sub>	- <sup>b</sup>
O <sub>2(aq)</sub>	2 (varied)	UO <sub>2</sub> (CO <sub>3</sub> ) <sub>(aq)</sub>	9.63		
SO <sub>4</sub> <sup>-2</sup>	9.0	Fe <sup>+2</sup>	-7.76		
UO <sub>2</sub> <sup>+2</sup>	1.0 (varied)	FeHCO <sub>3</sub> <sup>+</sup>	-5.76		
Cl <sup>-</sup>	42	Fe(OH) <sub>2</sub> <sup>+</sup>	-5.67		

<sup>a</sup> For PHREEQC components and complexes, see PHREEQC database.

<sup>b</sup> Used for PHREEQC only. K<sub>sp</sub> not calculated for ParSSim database.

<sup>c</sup> Iron input as Fe<sup>3+</sup> in ParSSim equilibrates to Fe<sup>2+</sup>.

In the models PHREEQC and ParSim, minerals will not dissolve or precipitate unless they are specifically selected for equilibration with the model solutions. Minerals selected for equilibration were uraninite (the reduced uranium mineral) for the case of a carbon-reducing agent (CH<sub>4</sub>), and uraninite, hematite, amorphous iron hydroxide, and pyrite for the case of an iron-reducing agent (FEMETAL). These iron minerals resulted in a

solution that was not supersaturated with respect to any remaining minerals; however, other combinations of iron-mineral equilibration could have been used.

The reducing agents, FEMETAL ( $\text{Fe}^0$ ) and  $\text{CH}_4$ , were selected to represent waste containers in the disposal site. FEMETAL represents 55-gal drums and other steel containers commonly used for waste, and  $\text{CH}_4$  is a surrogate for organic matter, such as wood crates used in disposal. The amounts of these reducing agents were varied in the model, but were typically less than the molar content of a single barrel or wooden crate.

The FEMETAL was introduced by setting an equilibration constant with respect to elemental iron (zero oxidation state). The simulations were conducted with FEMETAL undersaturated in the system. When saturation with respect to FEMETAL was assumed, the model predicted a large release of reduced iron in the system, along with very low  $p_e$ , and high pH ( $> 10$ ). More likely, a slower release of iron occurs, and this was represented in the model by setting the FEMETAL equilibrium to subsaturated conditions. A saturation index of  $-15$  produced a rate of release of reduced iron in solution that was reducing at near-neutral pH. Although the source concentration of FEMETAL in the model is well below the total iron content of containers in the trenches, it is not known if the iron in the trenches would become localized in a zone to form a redox boundary or what the rate of reduction would be.

At Barnwell, there are fewer sources of organic matter than at municipal or industrial landfills, but early disposal practices included wood and cardboard boxes for containers. In addition, small amounts of scintillation solutions were included in the waste prior to 1976. Thus there are potential sources of organic matter in the waste trenches that could serve as reducing agents or sorbing surfaces to increase the concentration of uranium. Organic matter is very important in the concentration and reduction of sedimentary uranium ores (Spirakis, 1996; Landais, 1996; Wood, 1996). In particular, sources (including tree trunks) are believed to be important (Wood, 1996). Kinetic inhibition of reduction may be overcome by heat or microbial activity. Organic matter is also common in landfills and is associated with methanogenic reducing zones.

Thermodynamic data are not available for wood or cardboard, so a surrogate species must be used. The use of  $\text{CH}_4$  as a reducing agent represents an end-member composition for these materials, although it is probably more reducing than wood or cardboard, and is readily dissolved in water. A partial pressure of  $\text{CH}_4$  gas was maintained such that there was 2 mg/L in solution.

Precipitation in the reducing zone was controlled by the mineral uraninite, a commonly observed uranium mineral in reducing zones of natural ore deposits. Model runs were typically 500 L of pore fluid, which represented 80 years at present-day infiltration rates. Runs representing times up to 1,000 and 10,000 years were also conducted. The longer times were run only with PHREEQC.

#### 4.2.4 Sensitivity Analysis

A series of runs were conducted to help provide bounds on conditions that could limit or enhance the potential for nuclear criticality safety concerns. The parameters selected for evaluation were the initial concentration of dissolved uranium, the concentration of oxygen, the amount and size of reducing zones, additional uranium complexes, precipitation of uranium silicate minerals, dispersion, and 3-D source terms. Most of the sensitivity analyses were conducted with PHREEQC. A more limited number of runs to evaluate dispersion, dimension, and size were done with ParSSim (Fig. 4.2). The range in parameters was selected to span likely field conditions rather than worst-case scenarios. Thus the uranium concentration was varied from 1 to 20 mg/L (an upper limit of observed values around waste sites); the oxygen concentration varied from 1 to 8 mg/L (the upper limit being

the solubility limit for oxygen dissolved in water); the leachate complexes were chosen based on observed values for the anions nitrate and fluoride at the Barnwell site.

## 5 ASSUMPTIONS

All models are simplifications of reality. Some of the processes not explicitly modeled in the simulations are evaluated here. The scenarios modeled typically represent bounding calculations.

- Saturated, steady-state flow

The models assumed saturated, steady-state flow to approximate conditions over the long time frame. These conditions represent worst-case scenarios because unsaturated, transient flow likely involves longer travel times.

- Geochemical complexes and solid phases

The geochemical complexes and solid phases that were selected for equilibration were based on a knowledge of important uranium species and what is known about site chemistry. The decision was made to focus on the reduced form of the uranium (i.e., uraninite). Reactions among species were assumed to be in equilibrium. Although additional complexes and phases could change the mobility of reactants, and kinetic considerations could change the rates, the calculations represent likely bounds and best available data.

- Simplistic deposit geometries

Simplistic deposit geometries, having no density gradients, were used in the criticality assessment. That is the  $^{235}\text{U}$  and source material were assumed to be uniformly distributed over the volume of the trenches. 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  $^{235}\text{U}$  as compared with a homogeneous core mass of 1162 g  $^{235}\text{U}$  in an equal volume.

- Enrichment

For the nuclear criticality calculations, a  $^{235}\text{U}$  enrichment of 10 wt % is assumed. Based on disposal records (see Section 8 and Table 8.1), this represents an arbitrary upward bound on likely enrichments in the trench. Because of incomplete records and a lack of regulatory limits on enrichment, a theoretical upper bound of 100 wt % enrichment is conceivable, but not likely. The case of 100 wt % enrichment was modeled previously in Vol. 1, and the calculations presented there are applicable to issues of this study. In evaluating individual trenches, the average enrichment was assumed to be appropriate.

Enrichment does not affect hydrogeochemical mobility of uranium. Relative enrichments of  $^{235}\text{U}$  are assumed to remain constant between the source and the precipitated secondary phases. The geochemical model accounts for total uranium, irrespective of the  $^{235}\text{U}$  isotopic enrichment. However, because the amount of dissolved uranium associated with the waste can span an order of magnitude from 1 to 10 mg/L, one can interpret the 1 mg/L as 100 wt % enrichment and the 10 mg/L as 10 wt % enrichment that have equivalent  $^{235}\text{U}$  densities (i.e., 1 mg  $^{235}\text{U}/\text{L}$ ). The geochemical models deal only with chemical species and not isotopes.

- Reducing zones

It was assumed in this analysis that reducing zones exist within the disposal environment. The development and longevity of reducing zones was not specifically modeled in this analysis because of a lack of site-specific information on weathering rates of containers, amount of organics in specific trenches, and 3-D data on chemical zonation within and around the trenches. Reducing zones also require a water-saturated environment, which is not currently present in the trenches. Thus the reducing zones represent a worst-case scenario of future conditions, which would enhance precipitation of uranium.

- Container degradation

For the reasons given above, the degradation of the waste containers was not modeled explicitly in this analysis; all of the source term was assumed to be exposed to migrating fluids. Realistically, degradation is fast relative to the time required for significant chemical migration. Steel can degrade in tens of years, concrete can crack, and high-density polyethylene (HDPE) degrades in hundreds of years.

- Horizontal and preferential flow paths

An important assumption regarding the mobilization of uranium in trenches is that multiple vertical flow paths can be funneled into a demobilization zone. This funneling occurs through a horizontal component of flow. Although the areal density disposal limits and the package limits mitigate development of a critical mass along a vertical path, horizontal flow along the base of the trench may result in the concentration of material within the drainage system or the sumps. In this exercise, two-dimensional (2-D) flow was modeled as a combination of 1-D segments.

## 6 CRITICALITY SAFETY EVALUATION RESULTS

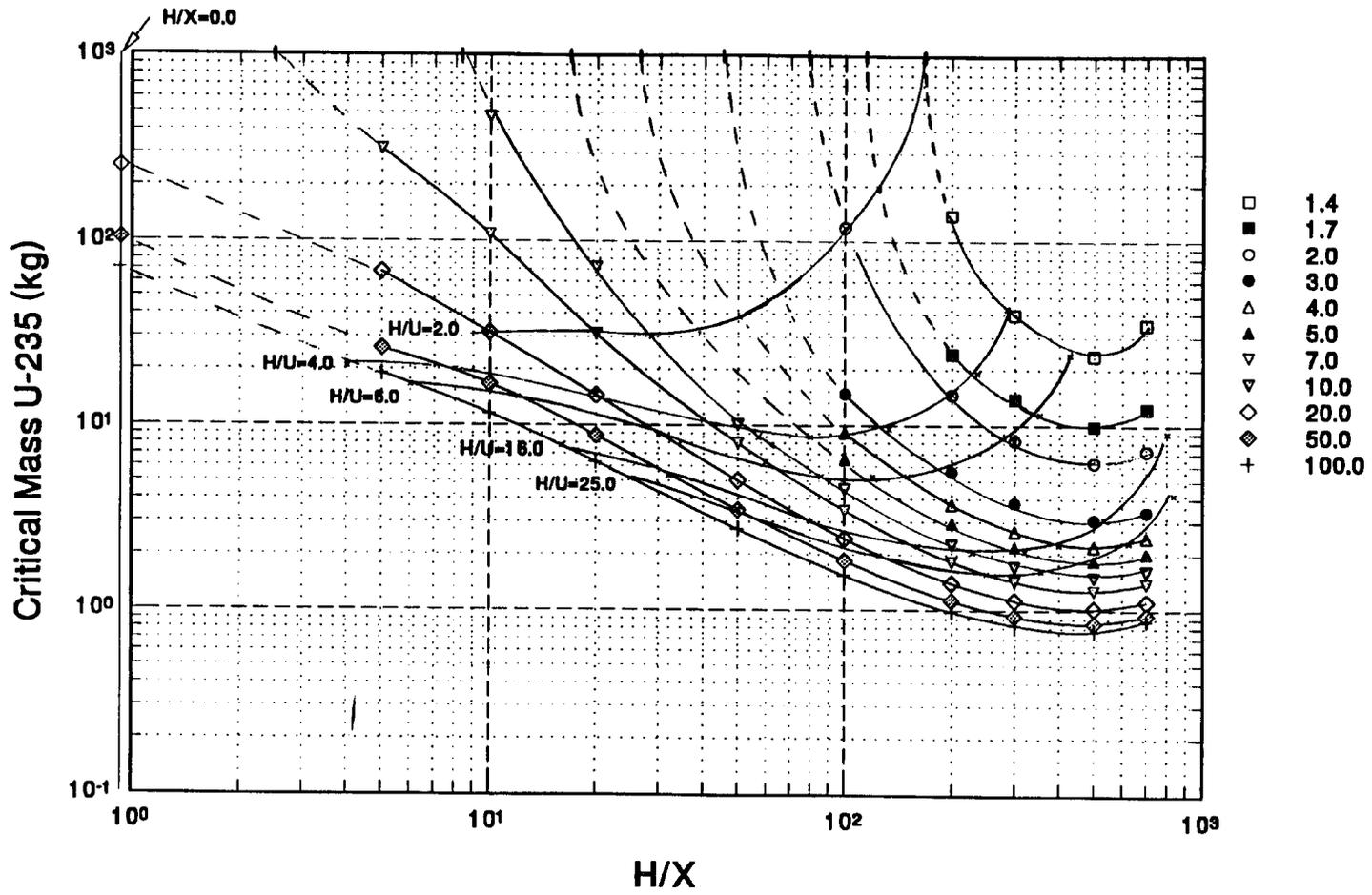
### 6.1 <sup>235</sup>U ENRICHMENT INFLUENCE ON CRITICAL MASS OF URANIUM

The critical mass of <sup>235</sup>U is inversely proportional to the <sup>235</sup>U enrichment of the uranium. That is, all else being equal (e.g., degree of neutron water moderation, volume, chemical composition, temperature, relative shape, etc.) as the weight percent <sup>235</sup>U enrichment in uranium decreases, the critical mass of <sup>235</sup>U increases. The following figures are taken from an illustrative study (Jordan and Turner, 1992) of this effect relative to the degree of neutron water moderation, expressed as the ratio of hydrogen atoms to <sup>235</sup>U atoms (H/X), for fully water-reflected spheres of homogeneous UO<sub>2</sub>F<sub>2</sub>-H<sub>2</sub>O solutions. The figures are provided to illustrate the difference between <sup>235</sup>U mass response to moderation and uranium mass response to moderation at various <sup>235</sup>U enrichments. Figure 6.1 demonstrates that for a given H/X of 500 the critical <sup>235</sup>U mass is estimated to be about 0.76 kg <sup>235</sup>U for U(100), whereas the critical <sup>235</sup>U mass is estimated to be about 1.2 kg <sup>235</sup>U for U(10). Figure 6.2 demonstrates that for a given H/X of 500 the critical uranium mass is also estimated to be about 0.76 kg uranium for U(100), whereas the critical uranium mass is estimated to be about 10.2 kg uranium for U(10). Thus for an H/X = 500, the ratio of the minimum critical masses of <sup>235</sup>U for U(100) and U(10) is about 0.63 (0.76/1.2). As can be observed in Figs. 6.1 and 6.2, for water moderation less than an H/X = 500, the optimum for minimum <sup>235</sup>U mass results in significantly smaller ratios of U(100):U(10) critical masses. For instance, at a poorly moderated H/X of 20 for U(100), the critical mass is estimated to be about 6.2 kg <sup>235</sup>U, whereas for U(10) the critical mass is estimated to be about 31.0 kg <sup>235</sup>U, a <sup>235</sup>U mass ratio of 0.2 or uranium mass ratio of 0.02. These effects are the direct result of the slowing-down power of water for high fission-energy neutrons escaping the neutron resonance capture characteristic of <sup>238</sup>U in the U(10). With less water present in the mixture, neutrons cannot be thermalized as rapidly as when present in optimum ratios of water to <sup>235</sup>U, about H/X = 500, and therefore the neutrons are captured by <sup>238</sup>U resonance capture.

### 6.2 TRENDS FOR U(10)-H<sub>2</sub>O-SiO<sub>2</sub> MIXTURES

Similar trends for increased uranium and <sup>235</sup>U mass are present for the mixtures of U(10)-H<sub>2</sub>O-SiO<sub>2</sub> presented in this report. The obvious differences between the above-referenced illustrative study and this study are the assumption of elemental uranium (i.e., no chemical compounds were considered) and the presence of silicon dioxide at a fixed density of 1.6 g SiO<sub>2</sub>/cm<sup>3</sup>. The presence of 90 wt % <sup>238</sup>U in the uranium in these calculations increased the importance of SiO<sub>2</sub> as a poor moderator of neutrons and a poor capturer of neutrons. For example, in the studies with U(10) having no water, the resonance capture of neutrons by the <sup>238</sup>U was important and suppressed the infinite-media neutron multiplication factor,  $k_{\infty}$ , from 0.955 for U(100) to 0.713 for U(10) at a <sup>235</sup>U concentration of 0.000886 g <sup>235</sup>U/g SiO<sub>2</sub>. A slight introduction of water to the matrix (i.e., 0.01813 g H<sub>2</sub>O/g SiO<sub>2</sub>) for both systems decreased the  $k_{\infty}$  for the U(100) to 0.867, whereas the  $k_{\infty}$  was increased for the U(10) to 0.779. This response is the direct result of the SiO<sub>2</sub> affording excess neutron slowing down for the U(100) matrix, thereby allowing hydrogen neutron capture to suppress  $k_{\infty}$ . In the case of the U(10), the SiO<sub>2</sub> provides excess neutron slowing down for the U(10) matrix, but these neutrons are forced to slow in energy into the <sup>238</sup>U resonance capture region and be captured. The slight addition of water to the U(10) matrix can slow neutrons by large amounts and thereby permit neutrons to be moderated or slowed to energies that are less than the <sup>238</sup>U resonance capture energies, thereby offsetting some of the negative effects of the SiO<sub>2</sub> over moderation.

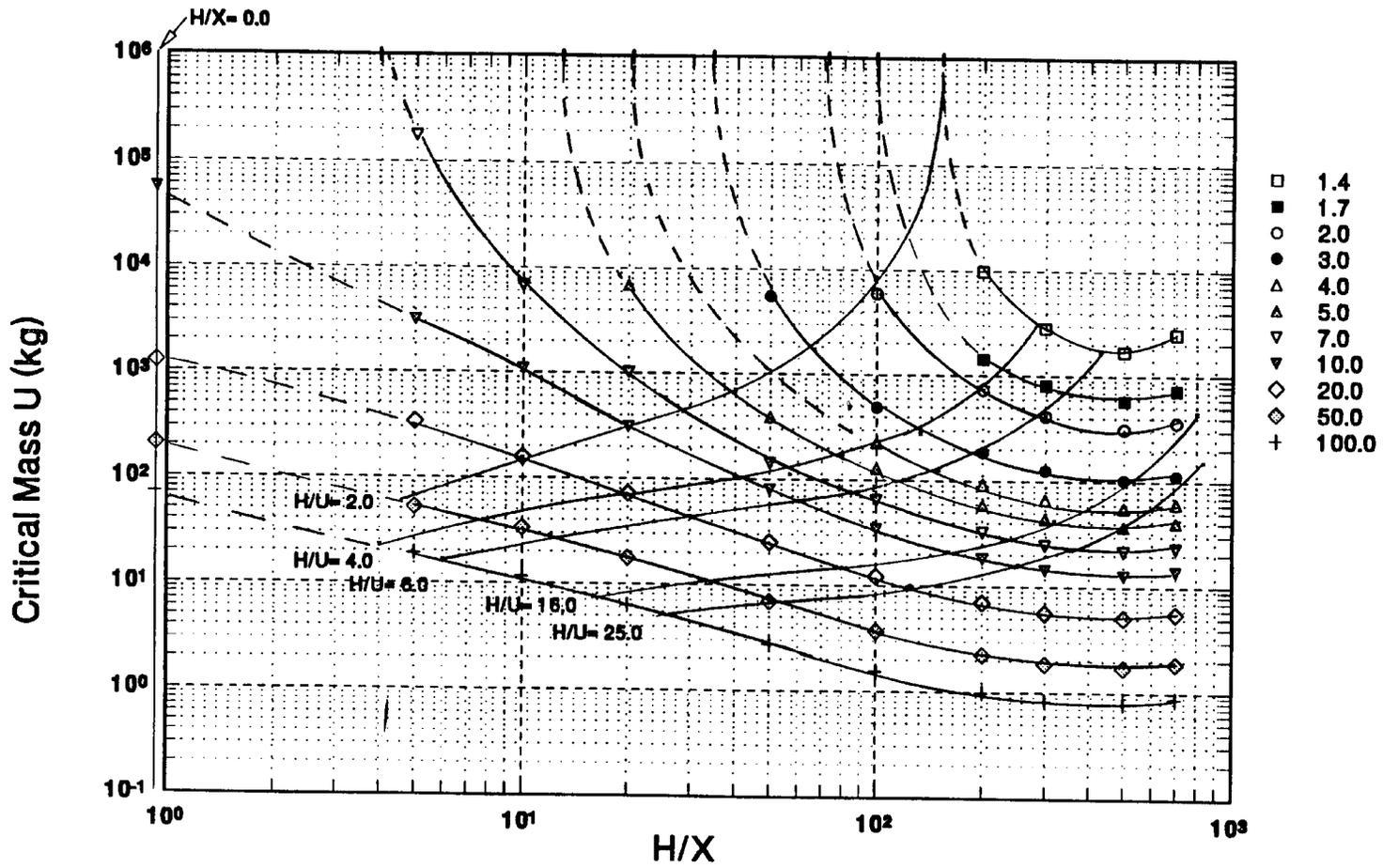
Table 6.1 provides extracted results of the computations to highlight the extremes of this study. The complete listing of the results of this study are provided in Table A.1 of the Appendix to this report. The results are provided in the same format that was used in Vol. 1, Appendix C. Figure 6.3 provides an interpolated and smoothed surface plot for the critical infinite slab areal density vs H<sub>2</sub>O and <sup>235</sup>U concentrations relative to SiO<sub>2</sub>.



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Figure 6.1 Critical mass  $^{235}\text{U}$  vs H/X for  $\text{UO}_2\text{F}_2\text{-H}_2\text{O}$  in spherical  $\text{H}_2\text{O}$ -reflected systems



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Figure 6.2 Critical mass U vs H/X for  $UO_2F_2-H_2O$  in spherical  $H_2O$ -reflected systems.

Table 6.1 U(10) plus H<sub>2</sub>O plus SiO<sub>2</sub>-soil (S-S) results

Line entry	<sup>235</sup> U content		Water content		k <sub>∞</sub> or k-inf	Critical infinite slab		Critical infinite cylinder		Critical sphere	
	g <sup>235</sup> U/cm <sup>3</sup>	g <sup>235</sup> U/gS-S	g H <sub>2</sub> O/cm <sup>3</sup>	g H <sub>2</sub> O/gS-S		Thickness (cm)	Areal density (kg <sup>235</sup> U/m <sup>2</sup> )	Diameter (cm)	Linear density (kg <sup>235</sup> U/m)	Diameter (cm)	Mass (kg <sup>235</sup> U)
31	0.00163	0.0010188	0.324	0.324	0.445						
69	0.0024761	0.0015476	0.029	0.01813	0.982	464.22	11.4946	758.34	111.84	1020.78	1378.9967
110	0.0032722	0.0020451	0.0885	0.05531	1.031	174.48	5.7093	328.62	27.7534	439.7	145.64911
156	0.0057142	0.0035714	0.324	0.2025	0.965	234.28	13.3872	401.80	72.4544	484	339.22677
162	0.0063	0.0039375	0.119	0.07438	1.222	66.02	4.1593	123.50	7.54681	174.32	17.473536
208	0.2817	0.1760625	0.029	0.01813	1.110	13.83	38.9591	90.94	182.9674	143.62	436.9495
212	0.2817	0.1760625	0.251	0.15688	1.334	11.24	31.6631	33.84	25.3352	53.06	22.0337
213	0.2817	0.1760625	0.324	0.2025	1.379	9.82	27.6629	28.80	18.3510	44.96	13.4049

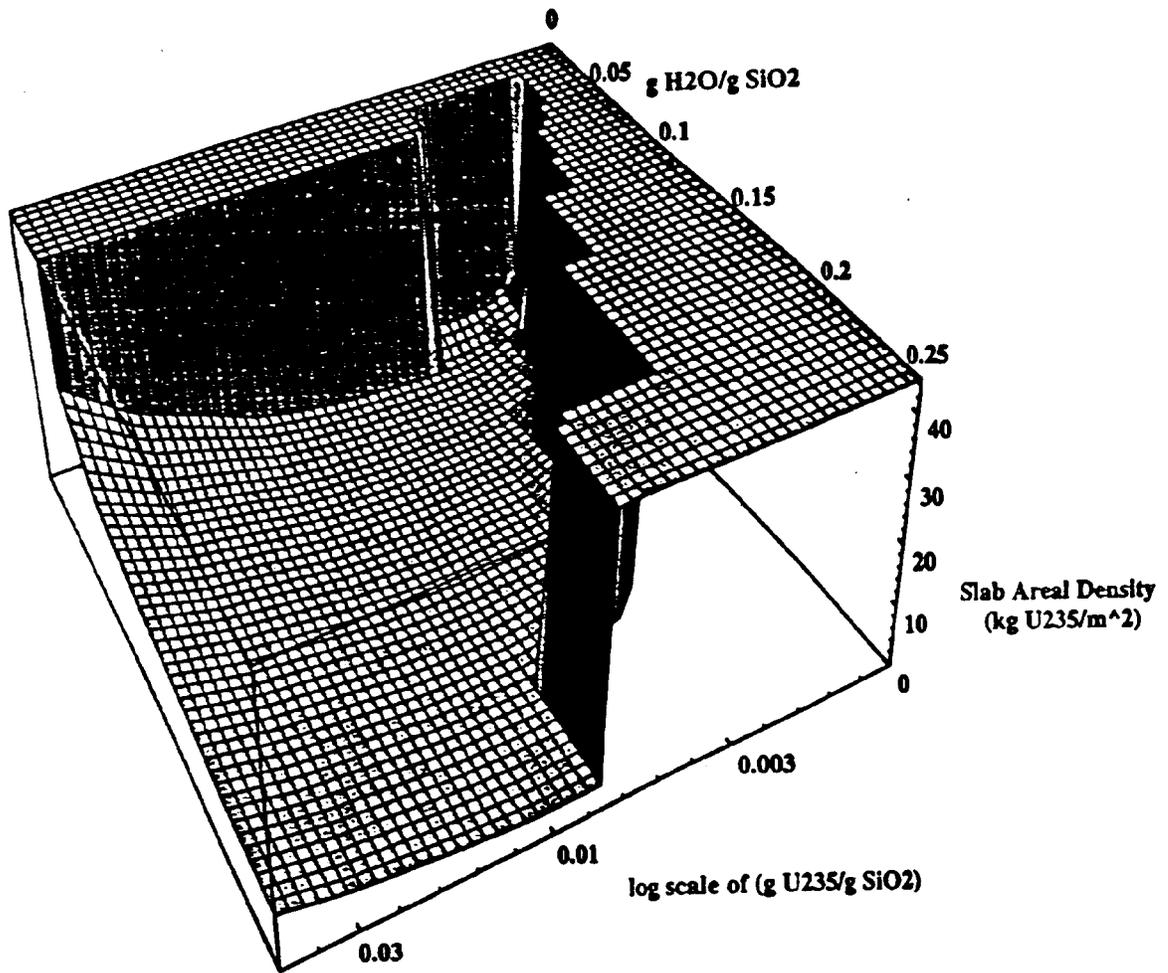


Figure 6.3 Infinite slab areal density (kg <sup>235</sup>U/m<sup>2</sup>) vs g H<sub>2</sub>O/g SiO<sub>2</sub> and log scale of g <sup>235</sup>U/g SiO<sub>2</sub>

Figures A.1 through A.11 provide interpolated and smoothed surface plots of the data provided in Table A.1. These plots are provided in the same format that was used in Vol. 1, Appendix D. In future comparisons, care should be exercised to recognize that even though there are similarities between the general appearances of the surface plots of the U(100) and the U(10), the significantly larger  $^{235}\text{U}$  values are for uranium enriched to only 10 wt %  $^{235}\text{U}$  in the uranium.

### **6.3 COUPLING OF NUCLEAR CRITICALITY AND HYDROGEOCHEMICAL MODELING**

The nuclear criticality safety calculations were used to establish the configurations and associated uranium concentration increases that are required to reach a level of concern. Then, using the hydrogeochemical modeling approach and assumptions discussed in Sections 4 and 5, the time required to increase the uranium concentration from the reported disposal values to one of the minimum concentration levels of concern was determined. Several minimum values of the critical mass were selected to provide a conservative scenario as benchmarks for evaluating the potential of developing a critical mass of  $^{235}\text{U}$  under the conditions of  $\text{SiO}_2$  matrix, water, and 10 wt % enrichment  $^{235}\text{U}$ . The lowest uranium concentration that could support a nuclear reaction was in a nearly dry system:  $0.0024761 \text{ g } ^{235}\text{U}/\text{cm}^3$  and  $0.029 \text{ g H}_2\text{O}/\text{cm}^3$  (Table A.1, line entry 69). This concentration requires a slab 464.2 cm thick, or 1379 kg, if in a spherical configuration. By contrast, concentrations of  $0.0224 \text{ g } ^{235}\text{U}/\text{cm}^3$  and  $0.2817 \text{ g } ^{235}\text{U}/\text{cm}^3$  under almost fully saturated conditions would require slabs of 19.7 cm and 9 cm thick, respectively. The hydrogeochemical model assumes a water-filled porosity of 40%. If uranium precipitates under wet conditions, then the system would need to dry out in order to achieve the minimal configuration necessary to support criticality. In order to develop the higher uranium concentrations, funneling of source material by horizontal transport is necessary.

## 7 HYDROGEOCHEMICAL MODELING RESULTS

Hydrogeochemical modeling results are presented as a comparison of simulations with different parameters. The evolution of uranium precipitation over time is used as the basis for comparison (Table 7.1). PHREEQC modeling is conducted in terms of pore volumes, which can be translated into years by assuming a groundwater velocity. A pore volume in the model was assumed to be 1 L of water. Using an assumed groundwater velocity of  $10^{-5}$  cm/s (1 m/year), most simulations were conducted for 80 years (500 pore volumes) with selected simulations up to thousands of years. Relevance of the modeling to site conditions at Barnwell is discussed in Section 8.

### 7.1 REDUCING ZONES

The reducing zones maintained a low redox state over the duration of modeled influx of oxidized water (up to 30,000 L for the longest simulation; typically 500 L). The  $\text{CH}_4$  created a reducing zone that had a pe of  $-2.7$  with a pH of 6, whereas FEMETAL ( $\text{Fe}^0$ ) affected a reducing zone with a pe of  $-1.9$  and a pH of 6. The rate of release of the reducing agents was on the order of 25 mg/L per year for methane (Fig. 7.1) and 114 mg/L per year for FEMETAL (a small fraction of a barrel). The values are derived from the modeled conditions and are not meant to represent the actual release rates in a disposal setting. These modeled release rates are not likely to consume the supply of wood from disposal crates or metal from weathering drums. However, the kinetics of weathering of these disposal containers is not well known. Both types of reactions could be mediated by bacteria, providing accelerated weathering rates and formation of reducing zones.

The reducing zones as modeled capture very close to 100% of the uranium input (e.g., 99.998% for Run 0), producing a linear increase in the amount of uranium precipitated over time (Fig. 7.1). One reason the reducing zones are efficient at capturing uranium is that reduced uranium minerals, such as uraninite, have low solubility. Another important factor is that there were modest amounts of complexing agents present in the observed trench water.

### 7.2 SENSITIVITY ANALYSIS

For the conditions simulated, the most significant parameter in predicting the amount of uranium precipitation was the initial source of uranium or the dissolved concentration associated with wastewater filtering through dry waste.

The uraninite precipitation was directly related to the initial concentration of uranium, and complete precipitation occurred for initial concentrations up to 20 mg/L (maximum modeled). The uraninite precipitation was not sensitive to the oxygen concentration in the infiltrating water, which has the potential to compete with uranium for electron acceptor sites. The concentration of oxygen dissolved in water can vary only over a small range; so it is not a sensitive parameter. The model predicted essentially the same amount of precipitation for cases of half the concentration of the reducing agent. Small, but realistic, amounts of complexing agents were added with no significant effect. In particular, increasing the bicarbonate concentration from 80 to 400 mg/L did not inhibit uraninite precipitation. The uranyl-carbonate complex is not dominant under reducing conditions. Sensitivity to the precipitation of silicate minerals and hydrodynamic dispersion is discussed in the following sections.

Table 7.1 Parameter variation and results of hydrogeochemical modeling

Run No.	Amendment	Value	U PPT CH4	U PPT FE
0	Base case (one set of runs for each of two reducing agents: FeMetal and CH <sub>4</sub> ) <sup>a</sup>	See below	$2 \times 10^{-4}$	$2 \times 10^{-4}$
1	Vary uranium in leachate	2 mg/L	$4 \times 10^{-4}$	$4 \times 10^{-4}$
2		10 mg/L	$2 \times 10^{-3}$	$2 \times 10^{-3}$
3		20 mg/L	$4 \times 10^{-3}$	$4 \times 10^{-3}$
4	Vary oxygen in leachate	2 mg/L	$2 \times 10^{-4}$	$2 \times 10^{-4}$
5		8 mg/L	$2 \times 10^{-4}$	$2 \times 10^{-4}$
6	Vary reducing zone	1 mg/L CH <sub>4</sub> or 1 M FeMetal	$1.4 \times 10^{-4}$ ( $1.4 \times 10^{-4}$ )	$2 \times 10^{-4}$
7		1 cell reducing	$2 \times 10^{-4}$	$2 \times 10^{-4}$
8	Vary carbonate concentration	240 mg/L HCO <sub>3</sub> <sup>-</sup> 400 mg/L HCO <sub>3</sub> <sup>-</sup>	$2 \times 10^{-4}$ $2 \times 10^{-4}$	$2 \times 10^{-4}$ $2 \times 10^{-4}$
9a	Vary leachate complexes	Add 2 mg/L F	$2 \times 10^{-4}$	$2 \times 10^{-4}$
9b		Add 20 mg/L NO <sub>3</sub>	$2 \times 10^{-4}$	$2 \times 10^{-4}$
10	Vary precipitate (ppt in zone of higher Si, equilibrated with quartz)	ppt. coffinite	$2 \times 10^{-4}$	$2 \times 10^{-4}$
11		ppt. soddyite	$1.0 \times 10^{-4}$	
12a	Incorporate transport (dispersion)	1-D transport in ParSSim	$1.8 \times 10^{-4}$	
12b		1-D, slower velocity	$3.8 \times 10^{-5}$	
12c		1-D reducing zone halfway down grid	$1 \times 10^{-4}$	
13		3-D transport with small source area	$2.6 \times 10^{-5}$	
14		3-D transport with large source area	$1.3 \times 10^{-5}$	
15		Larger dispersivity	$2.9 \times 10^{-6}$ ( $1.4 \times 10^{-5}$ )	

<sup>a</sup>Base Case: 1 mg/L U in leachate

Reducing Agent 1: 10 M FeMetal (equilibrated, but undersaturated to create a slow release)

Reducing Agent 2: 2 mg/L CH<sub>4</sub>, constant source

2 mg/L O<sub>2</sub>

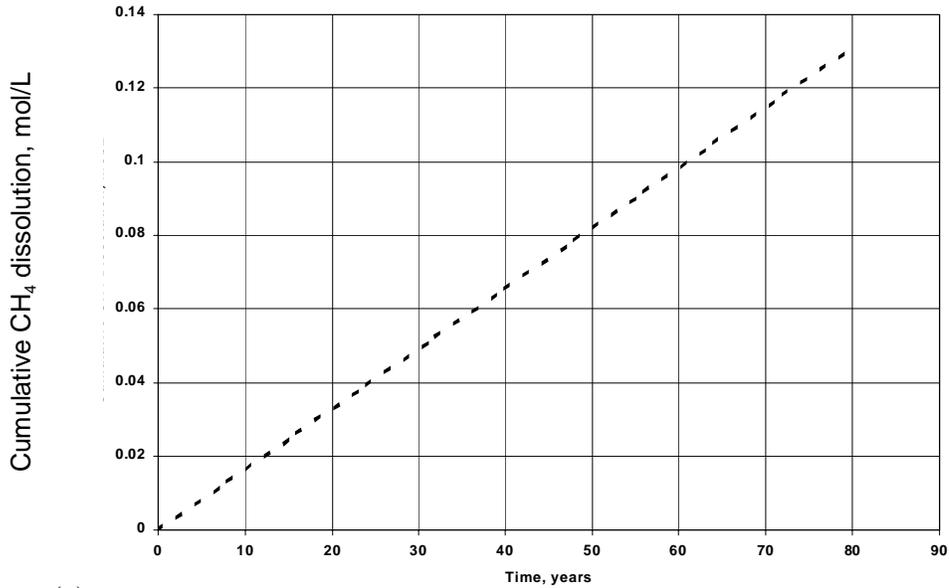
80 mg/L HCO<sub>3</sub><sup>-</sup>

5 cells reducing

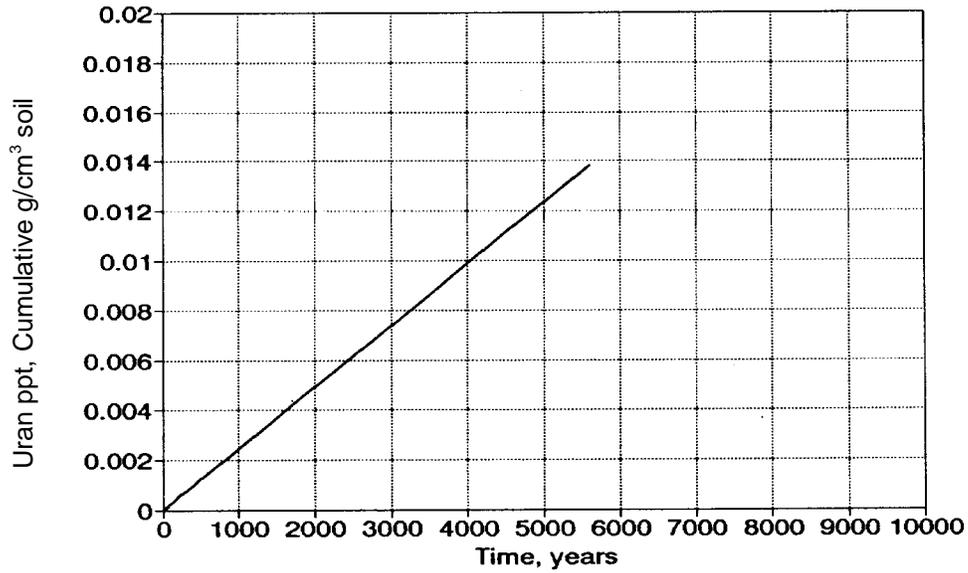
Equilibration minerals: uraninite, also for FeMetal: pyrite, hematite, Fe(OH)<sub>3</sub>

U PPT = g U-mineral precipitated per cm<sup>3</sup> soil using CH<sub>4</sub> or Fe<sup>0</sup> as reducing agents.

For runs with number shown in parentheses, the run did not go to completion. Value should be compared with value shown in parentheses for equivalent timestep.



(a)



(b)

Figure 7.1 Reaction progress for hydrogeochemical model: (a) Cumulative CH<sub>4</sub> release vs time for short time frame; (b) Cumulative uraninite precipitation for long time frame. Times shown are based on a velocity of  $3 \times 10^{-6}$  cm/s and a porosity of 0.4

### 7.3 PRECIPITATION OF SILICATE MINERALS

Some alternative uranium mineral precipitates were considered by including silica in the model. The silica in the infiltrating water is expected to be relatively low, given that the source area contains waste rather than soil. However, when the leachate reaches the drains at the base of the trench they will encounter a zone of backfill containing sand. This is likely to result in water in equilibrium with an amorphous silicon dioxide such as chalcedony. The scenario modeled was that of water infiltrating into a zone in which chalcedony dissolution occurs, increasing the silica concentration up to about 6 mg/L. Precipitation of uranyl silicates was modeled with and without reducing agents present. When the uranium infiltrates this zone, uranium silicates precipitate. The minerals considered in this analysis were soddyite, an oxidized uranium silicate, and coffinite, a partially reduced uranium silicate. The soddyite does not completely precipitate all of the available uranium because it has a higher solubility than uraninite. The coffinite captured the uranium as efficiently as the uraninite.

### 7.4 3-D HYDROGEOCHEMICAL MODEL

The main error induced by using a mixing cell model, such as PHREEQC, instead of a coupled geochemistry and transport code, such as ParSSim, is that dispersion is neglected in the mixing cell model. Dispersion is a function of the dispersivity and the velocity. It can occur in three dimensions, although the two transverse directions dispersivity tends to be smaller in the transverse directions rather than the longitudinal direction (in the direction of flow). The effects of dispersion on transport and reconcentration were evaluated and compared in a variety of ways: (1) 1-D vs 3-D dispersion, representing an infinite source and a point source; (2) dispersion along the flow path before the reducing zone is encountered; (3) longitudinal and transverse dispersion (ratio 3:1) after release from a 1-m by 1-m source area; (4) longitudinal and transverse dispersion after release from a larger source, 3 m by 1 m; and (5) fast vs slow velocities. These runs were conducted with ParSSim, using the same background solutions and the same equilibration minerals as in the CH<sub>4</sub> reducing zone case.

The results (Table 7.1) indicated that 1-D dispersion reduces the amount of uraninite precipitated, but that the amounts are reduced less than an order of magnitude, assuming typical scaled values for dispersivity (0.2 m). The errors due to neglecting dispersion in one dimension are relatively small compared with other uncertainties, such as the kinetics of weathering, mineral dissolution, and precipitation, and the size and distribution of source term. Three-dimensional dispersion results in a decrease in the amount of uranium precipitation of approximately an order of magnitude. This decline in concentration of precipitated uranium indicates that isolated sources of uranium that can disperse in three dimensions (Fig. 4.2) will require (1) much longer times to reach levels of concern for nuclear criticality safety, (2) larger source terms, or (3) may never develop concentrations sufficient to pose a criticality safety concern. Table 7.1 indicates that the model results are not highly sensitive to certain parameters [e.g., velocity (Run Nos. 12a and 12b), source term (Run Nos. 13 and 14) and flow path length (Run Nos. 12a and 12c)].

## 8 DISCUSSION

Several conditions restrict the potential for a criticality safety concern in specific disposal trenches at the Barnwell LLW disposal facility. Among these are the enrichment, the source term, the chemistry of water within the trenches, and the potential flow paths. Because of uncertainties associated with the disposal conditions, the uncertainties associated with the results of this analysis need to be underscored. The basic geochemical process, precipitation in reducing zones, does not appear to be a limiting factor based on the current model assumptions. However, long times may be needed. Uncertainties and other mitigating factors are also discussed.

### 8.1 ENRICHMENT

The average enrichment of homogenized material within each trench is available only for “new trenches,” Nos. 38 through 87. For the trenches with enrichment data (Fig. 8.1), the average  $^{235}\text{U}$  enrichment is 0.5 wt %, and only six trenches have more than the minimum 1 wt %  $^{235}\text{U}$  enrichment needed to reach a critical configuration. Trench 66 has a reported  $^{235}\text{U}$  enrichment of 100 wt %. It is judged from historic and current disposal practices and data (see Section 3 and Appendix B) that SNM and source material disposals are comingled in the trenches. Because of this comingling and dispersion of SNM, very few of these trenches need to be considered as criticality safety concerns. Again, this is based upon the reasonable assumption that  $^{235}\text{U}$  and nonfissile  $^{238}\text{U}$  behave in a similar geochemical fashion.

For the older trenches (trench numbers less than 38), only grams of SNM (Table 8.1) were reported. Since uranium is the most common SNM, to simplify the calculations, it was assumed that all of the SNM is  $^{235}\text{U}$  even though minor amounts of plutonium may have been disposed of in the trenches. No data were available on amounts of nonfissile uranium, so again the conservative assumption is that the uranium is all  $^{235}\text{U}$  and the enrichment is 100 wt % (Fig. 8.1). Two of these trenches have no reported SNM, which leaves 35 trenches with uncertain enrichment. Most of these trenches are unlikely to have high enrichments, although Trench 23 is an exception. The response to a specific NRC staff inquiry resulted in the identification of relative quantities and proportions of SNM, source material (SM) and byproduct material activity (BPM) that were placed in Trench 23. The reference to and summary of that response are provided in Appendix B, Subcriticality Evaluation for Chem-Nuclear Systems, Inc., Trench 23.

### 8.2 SOURCE TERM

The minimum mass of  $^{235}\text{U}$  to achieve criticality was calculated and compared with the total mass disposed in each trench. The minimum mass for a sphere provides a lower bound on source calculations, and these are shown in Table A.1. For slabs, the minimum mass for the source can be calculated assuming the length of each side of an “infinite” slab is ten times the calculated critical thickness. These finite volumes are multiplied by the critical uranium concentration to give total mass required for a potential criticality concern. These uranium mass values were used to determine the time frames needed to achieve three selected uranium densities: the minimum concentration to achieve criticality ( $0.0024 \text{ g/cm}^3$ ), approximately 10 times that density ( $0.022 \text{ g/cm}^3$ ), and approximately 100 times that density (the maximum calculated in Table A.1,  $0.28 \text{ g/cm}^3$ ).

For spheres, the critical masses range from 3 kg to 2000 kg over these density ranges (Fig. 8.2). For slabs, the critical masses vary from 20 kg to 6000 kg (Fig. 8.2). With the exception of Trench 66, the “new” trenches that contain SNM with  $^{235}\text{U}$  enrichments greater than 1 wt % have less than 30 g of  $^{235}\text{U}$  (Table 8.1), which is well below the minimum critical masses calculated. Trench 66 has a reported source term of 2.3 kg of 100 wt %

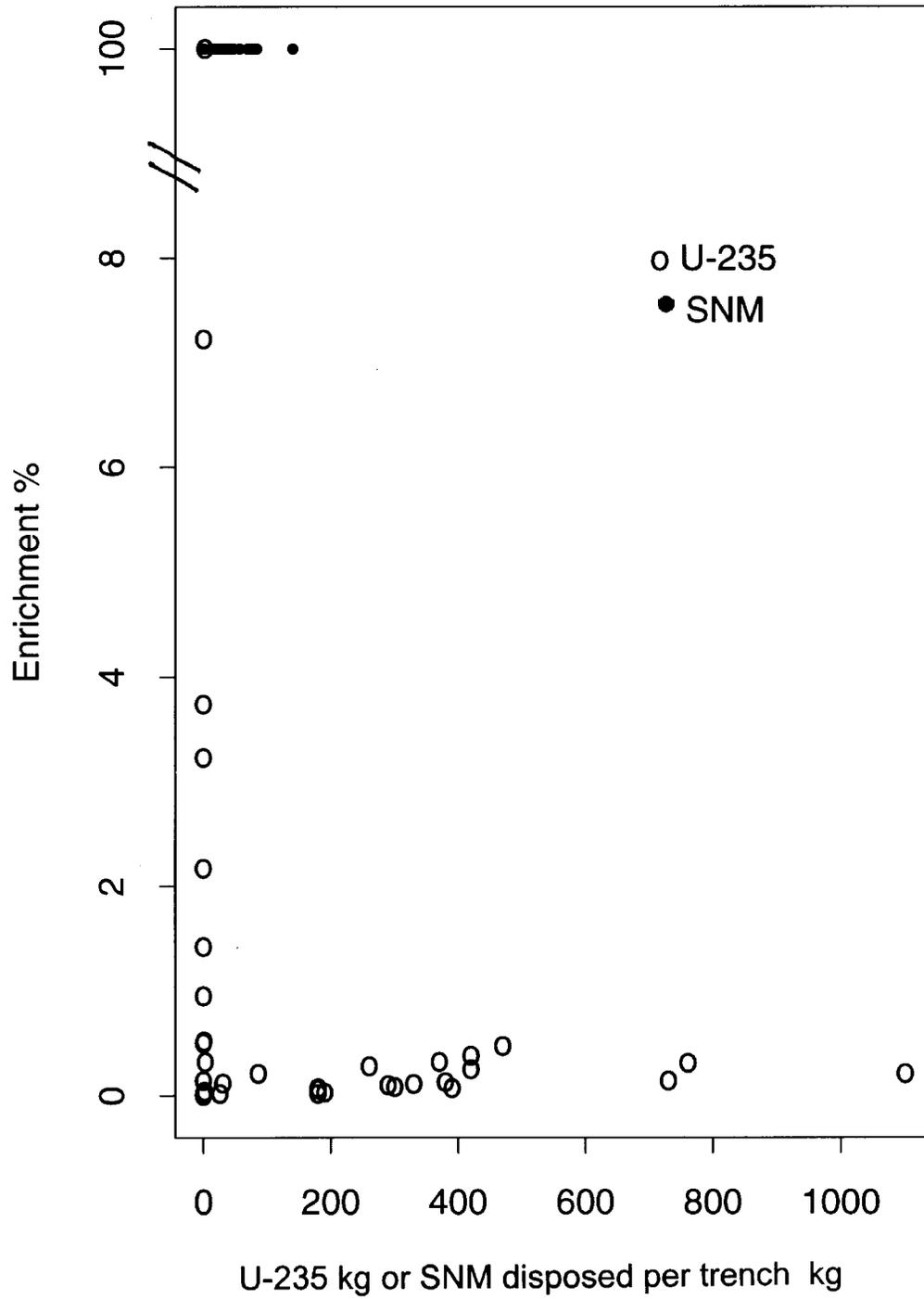


Figure 8.1 Relationship between enrichment and source term

Table 8.1 Disposal records from Barnwell, S.C., and calculated enrichments and density<sup>a</sup>

No.	SNM mass (g)	<sup>235</sup> U (g)	<sup>238</sup> U (g)	<sup>235</sup> U Enrichment (wt %)	Disposal volume (ft <sup>3</sup> ) <sup>b</sup>	Density <sup>c</sup> g/cm <sup>3</sup>
1	1.32e+04 <sup>d</sup>				4.2e+04	1.11e-05
2	3.73e+04				1.1e+05	1.20e-05
3	1.01e+04				2.3e+04	1.52e-05
4	4.66e+04				1.3e+05	1.26e-05
5	1.71e+04				1.8e+05	3.36e-06
6	2.53e+04				2.4e+05	3.66e-06
7	3.35e+04				2.4e+05	5.00e-06
8	2.45e+04				2.0e+05	4.29e-06
9	4.10e+04				1.9e+05	7.47e-06
10	2.91e+04				2.0e+05	5.18e-06
11	2.91e+04				1.5e+05	6.81e-06
12	1.73e+04				2.0e+05	3.09e-06
13	1.49e+04				2.1e+05	2.50e-06
14	2.46e+04				2.2e+05	3.95e-06
15	3.43e+04				2.8e+05	4.32e-06
16	1.62e+04				2.6e+05	2.20e-06
17	1.30e+04				1.8e+05	2.57e-06
18	1.78e+04				1.7e+05	3.63e-06
19	2.51e+04				2.4e+05	3.69e-06
20	1.07e+04				2.4e+05	1.61e-06
21	1.42e+04					
22	8.04e+04				9.2e+05	3.10e-06
23	1.75e+05				9.8e+05	4.91e-06
24	5.01e+02				2.6e+05	6.73e-08
25	1.42e+04				2.7e+05	1.83e-06
26	7.31e+04				8.9e+05	2.90e-06
27	1.44e+04				2.1e+05	2.39e-06
28	8.30e+04				8.9e+05	3.29e-06
29	7.31e+04				1.2e+06	2.24e-06
30	6.86e+04				4.8e+05	5.07e-06
31	1.39e+04				2.5e+05	1.93e-06
32	5.56e+04				7.1e+05	2.76e-06
33	7.60e+04				6.6e+05	4.07e-06
34	3.78e+04				5.4e+05	2.47e-06
35	7.04e+04				7.2e+05	3.47e-06
36						

Table 8.1 (continued)

No.	SNM mass (g)	<sup>235</sup> U (g)	<sup>238</sup> U (g)	<sup>235</sup> U Enrichment (wt %)	Disposal volume (ft <sup>3</sup> ) <sup>b</sup>	Density <sup>c</sup> g/cm <sup>3</sup>
37						
38	9.18e+07	2.59e+05	9.16e+07	0.28	5.1e+05	1.79e-05
39	1.10e+08	4.16e+05	1.09e+08	0.38	5.5e+05	2.66e-05
40	9.16e+04		9.16e+04		1.8e+04	
41	1.68e+08	4.17e+05	1.68e+08	0.25	5.8e+05	2.55e-05
42	2.93e+08	2.87e+05	2.92e+08	0.10	6.1e+05	1.67e-05
43	3.88e+08	3.02e+05	3.88e+08	0.08	7.8e+05	1.37e-05
44	2.81e+08	3.79e+05	2.81e+08	0.13	7.2e+05	1.85e-05
45	3.42e+03	4.63e+00	3.42e+03	0.14	1.0e+05	1.61e-09
46	1.14e+08	3.66e+05	1.14e+08	0.32	5.7e+05	2.28e-05
47	5.65e+02		5.65e+02		4.1e+04	
48	1.96e+03	2.78e+01	1.93e+03	1.42	6.4e+04	1.54e-08
49					6.7e+04	
50					9.4e+04	
51	9.93e+07	4.70e+05	9.88e+07	0.47	6.0e+05	2.74e-05
52	7.95e+06		7.95e+06		8.2e+04	
53	2.61e+08	1.78e+05	2.61e+08	0.07	5.3e+05	1.19e-05
54	6.02e+08	1.90e+05	6.02e+08	0.03	6.4e+05	1.05e-05
55	2.13e+02	4.63e+00	2.08e+02	2.17	8.6e+04	1.91e-09
56	3.87e+02		3.87e+02		9.9e+04	
57	6.30e+03		6.30e+03		8.3e+04	
58	5.68e+08	3.94e+05	5.68e+08	0.07	7.3e+05	1.90e-05
59						
60					1.1e+03	
61	9.90e+08	1.76e+05	9.90e+08	0.02	7.7e+05	8.05e-06
62	5.14e+08	7.27e+05	5.13e+08	0.14	7.5e+05	3.40e-05
63	5.11e+08	1.07e+06	5.10e+08	0.21	7.7e+05	4.90e-05
64	2.42e+08	7.57e+05	2.41e+08	0.31	5.1e+05	5.22e-05
65					2.2e+04	
66	2.32e+03	2.32e+03	0.00e+00	100.0	2.0e+04	4.12e-06
67	1.24e+02	4.63e+00	1.19e+02	3.74	1.4e+04	1.15e-08
68	4.30e+02	1.39e+01	4.16e+02	3.23	1.9e+04	2.61e-08
69					5.7e+03	
70	1.22e+08	2.61e+04	1.22e+08	0.02	2.2e+05	4.23e-06
71	3.00e+08	3.30e+05	3.00e+08	0.11	6.1e+05	1.90e-05
72	3.05e+08	1.83e+05	3.05e+08	0.06	1.0e+06	6.30e-06

Table 8.1 (continued)

No.	SNM mass (g)	<sup>235</sup> U (g)	<sup>238</sup> U (g)	<sup>235</sup> U Enrichment (wt %)	Disposal volume (ft <sup>3</sup> ) <sup>b</sup>	Density <sup>c</sup> g/cm <sup>3</sup>
73	2.51e+07	3.08e+04	2.51e+07	0.12	3.7e+05	2.91e-06
74	4.00e+07	8.59e+04	4.00e+07	0.21	3.8e+05	8.06e-06
75					5.4e+03	
76					1.3e+04	
77	6.41e+01	4.63e+00	5.95e+01	7.22	1.2e+04	1.36e-08
78					6.0e+03	
79	1.95e+03	1.85e+01	1.93e+03	0.95	3.3e+04	2.00e-08
80						
81	2.13e+04	1.06e+02	2.12e+04	0.50	3.1e+04	1.20e-07
82	3.68e+05	6.01e+01	3.68e+05	0.02	3.0e+04	7.16e-08
83	2.21e+05	1.14e+03	2.20e+05	0.52	2.6e+04	1.54e-06
84						
85	4.52e+06	2.01e+03	4.52e+06	0.04	2.1e+05	3.46e-07
86	9.88e+05	3.16e+03	9.84e+05	0.32	5.7e+04	1.96e-06
87	1.95e+04		1.95e+04		2.5e+04	

<sup>a</sup> SNM masses for Trench 38 and greater are determined by summing uranium isotope masses. Trench numbers less than 38 are reported SNM. Blank indicates data not reported or could not be calculated (enrichment or density) from available data.

<sup>b</sup> Values reported in units of cubic feet. Cubic meters can be determined by multiplying cubic feet by 0.0283.

<sup>c</sup> Density for Trench 38 and greater is the density of <sup>235</sup>U in g/cm<sup>3</sup>. For trench numbers less than 38, the density is the g/cm<sup>3</sup> of SNM. Blank values for the density indicate the volume or required mass was not reported.

<sup>d</sup> Read as  $1.32 \times 10^4$ .

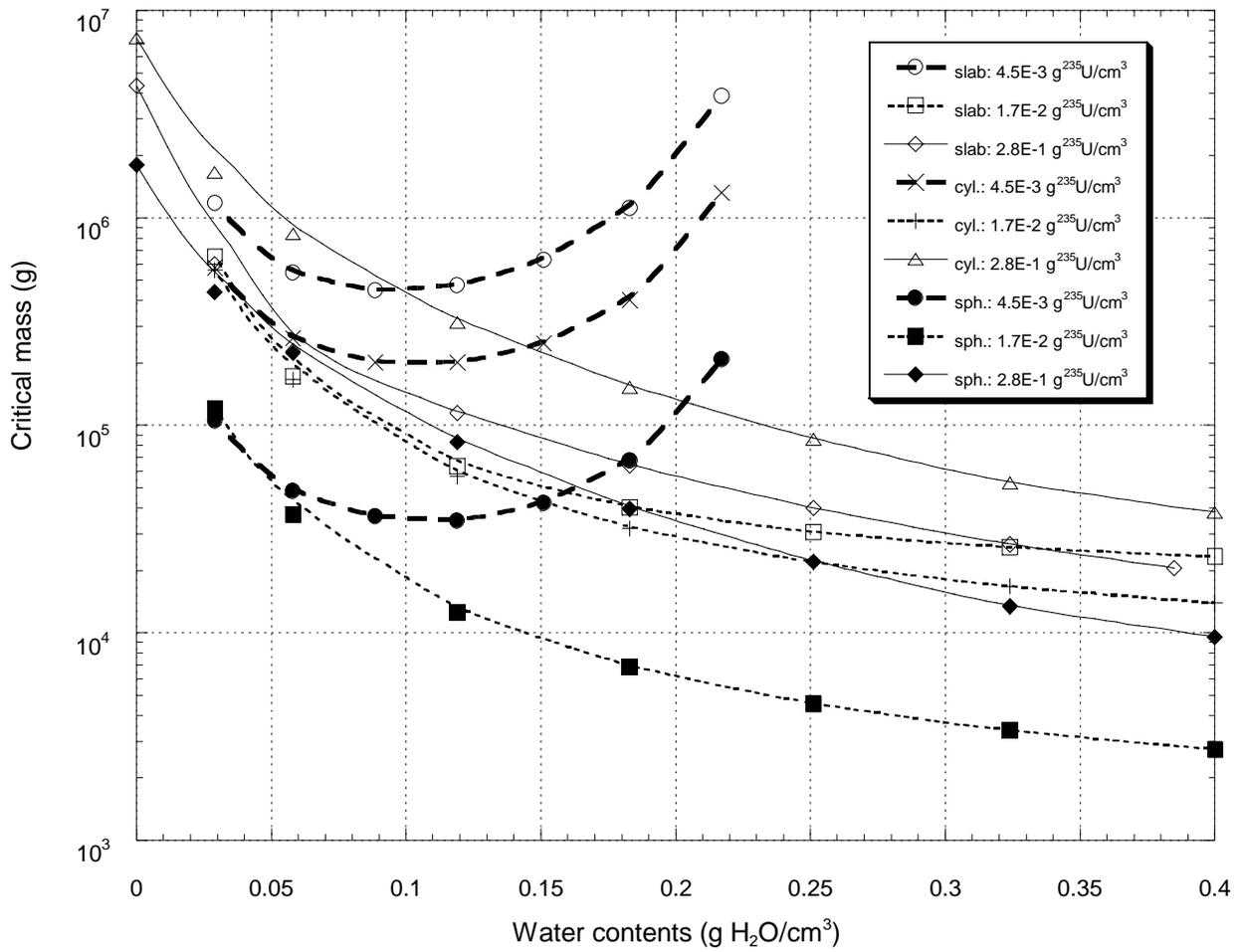


Figure 8.2 Critical ( $k_{\text{eff}, \infty} \geq 0.95$ ) masses of spheres and “pseudo-infinite” slabs and cylinders as a function of water content for different densities of  $^{235}\text{U}$  in waste matrix

enriched  $^{235}\text{U}$ , within the range for critical spheres. Because it is difficult to form a sphere under the transport conditions existing in a disposal trench, this small mass does not pose a criticality safety concern. The “old” trenches with unreported enrichments contained 10 to 175 kg of SNM (Trench 23), with a mean value of 30 kg. These masses are within the range of critical masses for spheres and for some slab configurations, assuming  $^{235}\text{U}$  enrichments of 10 wt %. Note that larger masses will be needed for enrichments between 1 and 10 wt %.

Distribution of the source term within a given trench is an important component in the potential to develop a critical mass. License conditions in effect between 1970 and 1981 limited the mass of  $^{235}\text{U}$  in a package, as well as gave some constraints on disposal. Prior to 1977, package limits varied between 15 and 50 g. The areal density of the material in the “old” trenches is less than  $0.05\text{ kg/m}^2$ , less than one-third of the minimum “new” trench areal density of  $0.16\text{ kg/m}^2$  (see Fig. 8.3). Although the enrichment of the early disposed material is not known, the smaller masses of SNM, the lower areal densities, and lower package limits indicate that the SNM is more dispersed in the “old” trenches (1–37) than in the “new” trenches (38–87). Therefore, while the older trenches have an inventory of SNM that *theoretically* would be sufficient to form a critical mass at 100 wt % enrichment, it is likely that the SNM is dispersed enough that significant funneling would be required to reconcentrate the  $^{235}\text{U}$  into a critical mass.

Trench 23 is an exception. Trench 23 contains 175 kg of highly enriched material and sufficient quantity to form either a sphere or slab with a critical mass. However, a subsequent evaluation of Trench 23 was performed (see Appendix B, “Subcriticality Evaluation for Chem-Nuclear Systems, Inc. Trench 23”) that demonstrates that criticality would not be achieved because of:

- (1) the limited thickness of the disposal burials (i.e., approximately 4 to 11 ft),
- (2) the conservatively evaluated low  $^{235}\text{U}$  densities (e.g., between a maximum of about  $0.00168\text{ g }^{235}\text{U/cm}^3$  for 350 g SNM in a single 55-gal container to a generally expected global density of less than about  $2.5 \times 10^{-5}\text{ g }^{235}\text{U/cm}^3$  within the trench), and
- (3) the significant quantity of source material (i.e., 210 metric tons of normal or depleted uranium or thorium), that is, codisposed with the 175 kg SNM that results in an homogenized trench averaged  $^{235}\text{U}$  enrichment of about 0.08 wt %.

### 8.3 GEOCHEMICAL PROCESSES

To increase the concentration of uranium, a geochemical process is needed that will capture uranium in a solid phase. Evidence from ore bodies indicates that uranium concentrations can increase through sorption onto a substrate or by precipitation as reduced uranium minerals. Because sorption was evaluated under a variety of conditions in Volume 1 of this study, this analysis assumed precipitation of uranium minerals.

One important result of the geochemical modeling is that given reasonable amounts of reducing agents, reducing zones are stable in a water-saturated environment (with limited oxygen). A more realistic approach to modeling the redox conditions in the trench would be to model transient wet/dry cycles, and weathering of drums and wood crates. It is known that such containers weather fast enough to be breached in recent disposal history. However, the weathered containers are still present, not completely weathered away. The remnants of such containers will serve as surfaces for sorption or redox-driven precipitation of transported uranium.

Concentrations of U-235 in Waste/ Barnwell LLW Facility

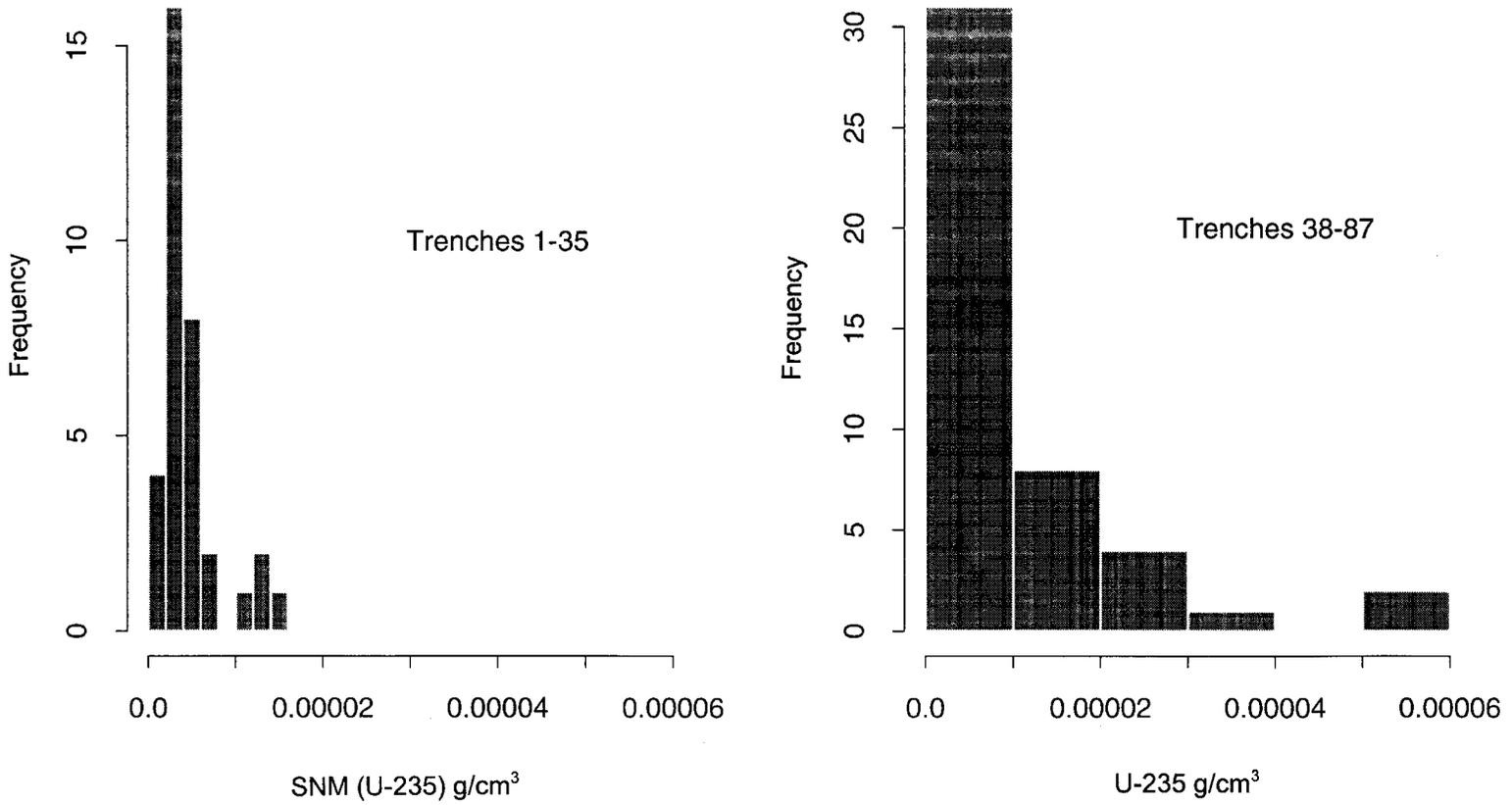


Figure 8.3 Histograms of calculated densities of <sup>235</sup>U in old and new disposal trenches using reported SNM or <sup>235</sup>U mass and reported disposal volumes (not total trench volume)

The longevity of the transport processes needed to increase the concentration of uranium can be estimated by the amount of water transporting dissolved uranium. Because of solubility limitations, the concentration of uranium dissolved in pore water is significantly less than the initial concentration on the waste. Thus the amount of fluid required to flush through the waste matrix in order to dissolve sufficient uranium for developing a critical mass is quite large.

To mobilize the 4.4-kg requisite to form a slab with a critical density of  $0.0224 \text{ g/cm}^3$  (line 190), with a solubility of  $10 \text{ mg U/L}$  (or  $1 \text{ mg } ^{235}\text{U/L}$ , assuming 10 wt % enrichment),  $3.5 \times 10^6 \text{ L}$  is needed. For a flow rate of  $1 \text{ m/year}$ , the volume of water to pass through  $1 \text{ m}^3$  ( $1\text{-m}^2$  footprint) of waste with 40% porosity is  $400 \text{ L}$  per year. The critical slab has a footprint of approximately  $4 \text{ m}^2$  and a thickness of approximately  $20 \text{ cm}$ . Assuming a flow rate of  $1 \text{ m/year}$ , almost 11,000 years are needed to mobilize the 4.4 kg. Unsaturated flow, or intermittent flow through the trench will also lengthen this estimated mobilization time.

The amount of reducing agent available was not a limitation in this analysis. Only a few grams are consumed in uranium reduction per pore volume, much less than the mass of iron- or organic-rich containers present in the trenches. Because the reductant was not a limiting reactant, the modeled geochemical reactions were not able to provide a constraint on the conditions for mobilizing and immobilizing  $^{235}\text{U}$ . However, other changes in hydrogeochemical conditions may occur over the time period of disposal.

## 8.4 HORIZONTAL-VS-VERTICAL FLOWPATHS

The initial areal densities within the waste trenches were estimated from the disposal records. The areal densities were calculated by assuming that the waste concentration is projected on a  $1\text{-m}^2$  footprint (Fig. 8.4). The average areal densities for the Barnwell facility trenches are significantly less than the minimal areal density required for criticality in a slab configuration (Table A.1, Appendix A), indicating that simple 1-D vertical flow paths will be insufficient to produce a critical areal density. Therefore, funneling of vertical flow paths or channeling of flow paths into horizontal flow (drainage system) must be considered.

The potential for horizontal flow in unsaturated soils has been evaluated by Kung (1990a,b). Kung (1990a,b) delineates three types of preferential flow in the unsaturated zone: short circuiting, fingering, and funneling. Short circuiting is the concentration of flow in macropores or fractures; fingering is the splitting of flow paths due to instabilities; and funneling is the horizontal movement and combining of flow paths caused by heterogeneities. Kung gives field evidence for funneling using dye tracers in sandy soils. Typically, a coarse unit will form a barrier to movement because of the large suction required to penetrate the larger pores; the penetration of this barrier is related to the hydraulic conductivity, the slope of the unit, and the height of the available water column. If funneling occurs, there is no need for saturated flow to develop. If flow occurs in 50% of the matrix, this amount is equivalent to increasing the capture zone for potential source material by a factor of 2, and if flow occurs in 1% of the matrix, the capture zone could be nearly a factor of 100 larger than assumed for strictly 1-D flow.

In general, the flow through the trenches at Barnwell is vertical. However, due to local heterogeneity and the transient nature of vertical groundwater flux, some horizontal flow may occur. Horizontal flow could occur within the waste zone or at the base of the trenches (Fig. 8.5). Horizontal flow is anticipated in the drainage systems along the base of the trench.

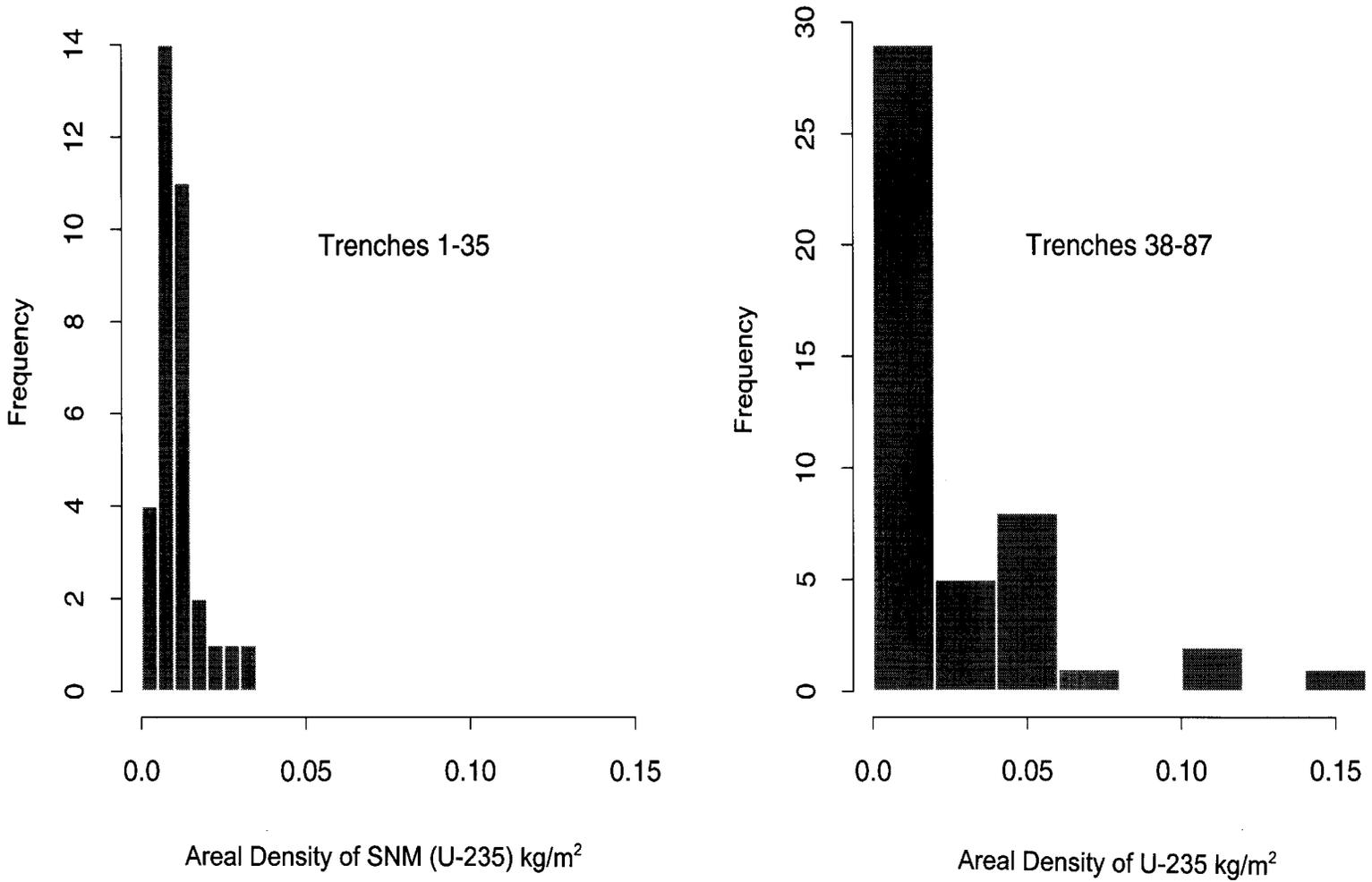


Figure 8.4 Histograms of calculated areal densities of <sup>235</sup>U in old and new disposal trenches using reported SNM or <sup>235</sup>U mass and reported disposal volumes

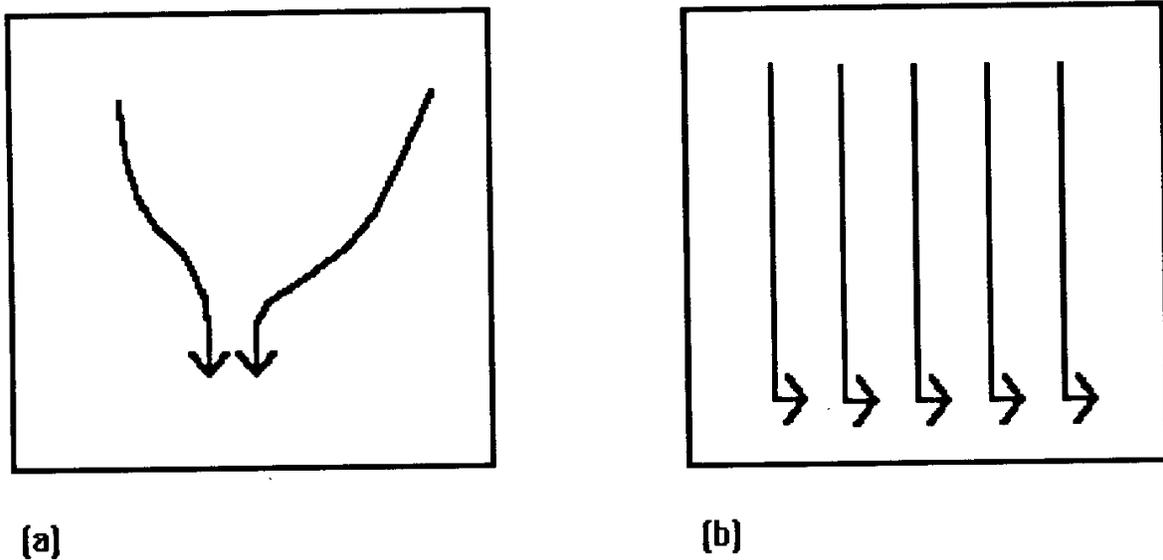


Figure 8.5 Cross sections showing horizontal flow (a) within the waste zone due to preferential flow paths and (b) at the base of the trench due to less-permeable sediments at the bottom boundary and trench drainage

Direct evidence indicates that horizontal flow occurs in the Barnwell trenches. Tracer tests were conducted by the U.S. Geological Survey in experimental trenches (constructed like the actual trenches) at Barnwell. Dennehy and McMahon (1987) report a bromide tracer experiment in which “the data from the probes ... indicated the response ... was due to ponded water in the trench moving laterally into the undisturbed sediments.” They attribute lateral flow to the impermeable nature of the “clayey” sand barrier, and also report evidence for temporary perched water, which has potential for saturated lateral flow.

A second piece of evidence for lateral flow is the appearance of tritium-contaminated water in the sumps that monitor trench discharge (Ichimura et al., 1994; see their Fig. 5). These sumps collect water at the base of the trenches from lateral movement along a sand buffer, which is part of the trench design (Fig. 3.1). Quantitative estimates of flow in the sumps were not available for this analysis. Much of the sump data are suspect because (1) video logs indicate that sump monitoring wells have filled in, and (2) discontinuities in water levels within the same trench indicate that monitoring wells act as conduits for surface water infiltration.<sup>1</sup> In either case, once the sump monitoring well is damaged, neither wet nor dry monitoring data can be used to understand water movement at the location. Thus, accumulation of wastewater in the sumps cannot be ruled out.

An estimate of the potential amount of horizontal flow at the base of the trenches was conducted by Mark Thaggard<sup>2</sup> of the NRC using the HELP code, a 1-D water-routing code developed to analyze percolation into waste facilities (Schroeder et al., 1994). Thaggard used three layers to represent a waste trench and input regional precipitation data from Cahill (1982). The three layers represented waste backfill, a drainage zone at the base of the trench (used to collect water at sumps) and undisturbed soils beneath the trench. The hydraulic conductivity of the basal unit was an important variable in determining the amount of horizontal flow that would occur. The hydraulic conductivities measured in four samples of undisturbed clayey sand vary from  $6.7 \times 10^{-8}$  to  $4.8 \times 10^{-7}$  (Dennehy and McMahon, 1987). Using hydraulic conductivity of  $1 \times 10^{-7}$  cm/s, Thaggard estimated 90% of recharge moves horizontally at the base of the trench and using  $10^{-5}$  cm/s, 0.03% of the flow is horizontal. These values might be considered bounding estimates, although Thaggard points out that the higher conductivity value affects horizontal flow, which corresponds to recharge estimates that better match regional recharge. It is not known whether recharge beneath the trenches should match regional recharge, however. The calculation leaves a large range in uncertainty for quantifying horizontal flow. However, the sensitivity of the calculation to hydraulic conductivity suggests that more *in situ* characterization of hydraulic conductivity in the clayey sand would help to better define the nature of flow in the bottom of the Barnwell facility trenches.

The extent of lateral flow needed to increase the areal density of the current configuration of  $^{235}\text{U}$  in the Barnwell trenches is unknown and is difficult to estimate given the uncertainties concerning distribution of the source term in the trench, weathering of containers and vault materials, and flow through the vaults. In the new trenches (38–87) if one vault contains SNM, then the adjacent vaults do not contain SNM. The areal densities of  $^{235}\text{U}$  in the trenches are significantly less than the areal densities necessary to sustain a nuclear reaction. In order to achieve a minimum areal density of  $4 \text{ kg/m}^2$  (Appendix A; Table A.1), significant lateral flow and/or funneling must occur. For new trenches, the lateral flow would have to be directed along the bottom of the trench in the drainage system, directly below the SNM. It is not known to what extent this occurs. The areal densities for the older trenches suggest that the distribution of SNM in the trenches is probably more diffuse than in the newer

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<sup>1</sup>Vernon Ichimura, Chem-Nuclear Systems, Inc., personal communication to Laura Toran, September 10, 1996.

<sup>2</sup>Mark Thaggard, NRC, personal communication to Laura Toran, October 25, 1996.

trenches (Figure 8.4). However, disposal practices during operation of the older trenches were less restrictive, suggesting that SNM in the older trenches may not be homogeneously distributed. The primary collector for water in the older trenches is the French drain system, which runs along the side of the trench. In order for a significant increase in concentration of  $^{235}\text{U}$  to occur, a combination of lateral and funneled flow will be necessary.

In summary, it will be important to continue to monitor the reliable sumps draining the waste trenches for evidence of waste mobilization. Although the sumps are meant to provide a collection point for wastewater and may enhance horizontal flow, they also can serve as a means of detecting uranium transport. If possible it would be useful to evaluate potential formation of reducing zones and the size of such zones. If the drainage system to the sumps becomes water-saturated, there is the potential for reducing conditions to occur. Since reducing zones would not form instantaneously, monitoring of the trenches would indicate changes in water chemistry over time. By keeping the trenches unsaturated and oxidized, it is possible to prevent buildup of uranium precipitation to thicknesses that would be of concern for nuclear criticality safety.

## 8.5 OTHER MITIGATING FACTORS

Other factors in the disposal setting may mitigate the reconfiguration of fissile uranium into critical masses. Because these factors cannot be quantified with the present information and scope of work, the extent to which they limit criticality is not presently known. Nonetheless, these considerations can be discussed in general terms and used to identify any concerns that need to be addressed.

In all trenches at the Barnwell facility, there are multiple barriers specifically included to physically separate the SNM. These barriers, which include disposal containers, vaults, and fill material, will inhibit reconfiguration of uranium.

The reducing zones, if they form, will require a water-saturated environment without dissolved oxygen. Presently, the trenches are not saturated with respect to water and the water chemistry appears to be slightly oxidizing. The effects of changing rainfall rates and trench capping could be modeled to evaluate the potential for saturating all or part of the trenches. Several trenches have high-density polyethylene caps, and future capping is planned. Another effect of assuming saturated conditions is faster estimates of travel time.

The potential distribution (amount and location) of reducing zones in this setting is a large source of uncertainty in this analysis. As stated previously, if the reducing zones are extensive, the precipitated uranium will be disperse and the concentration will be lower than needed to provide a criticality safety concern. If reducing zones do not form, the mechanism for increasing the concentration of uranium needs to change. Sorption on corroded waste packages was not modeled in this analysis. Sorption on mineral surfaces was considered in Volume 1 of this study, and did not result in sufficient increase in uranium concentration.

Small masses of  $^{235}\text{U}$  may be considered a criticality safety concern (Figure 8.2); however, they must form a sphere, which is difficult to achieve given the generally dispersive nature of flow. Nonspherical geometries, with the same uranium density, require considerably greater masses of  $^{235}\text{U}$ . Even though it may be easier to form a slab or cylinder by flow paths in a trench, the masses required to form such geometries are, in many cases, greater than the inventory of an entire trench.



## 9 CONCLUSIONS

This study presented calculations and assumptions to evaluate the potential for a nuclear criticality accident at an LLW site where containerized waste is buried. Many operational conditions were considered. Data needs for further evaluation are suggested. Even though conditions specific to Barnwell were used in the calculations, the work was also intended to provide an example for types of analyses for determining the potential for criticality at other disposal sites. The answers to questions posed in this study are as follows:

- Is there sufficient inventory for a criticality safety concern?

The inventory of  $^{235}\text{U}$  in the trenches is sufficient to form a critical geometry. However, the  $^{235}\text{U}$  inventory,  $^{235}\text{U}$  enrichment and the spatial distribution of the uranium must be considered in combination to judge whether there is a criticality safety concern in a LLW trench. The critical mass of  $^{235}\text{U}$  for pseudo-infinite slabs (length and width at least 10 times the thickness) comprised of silica soil ( $1.6 \text{ g SiO}_2/\text{cm}^3$  with variable densities of enriched uranium and water) ranged from a minimum of about 11.3 kg to well over 2000 kg of  $^{235}\text{U}$ , depending upon the concentration of water and uranium contamination in the available spaces of the soil porosity. This broad range of critical masses coincides with a broad range of uranium density and system volumes or sizes. The smaller critical masses require a greater concentration (i.e., densification) of enriched uranium than the larger critical masses. For example, a  $60 \times 60 \times 6 \text{ m}$  pseudo-infinite slab of silica soil contaminated with  $0.00155 \text{ g }^{235}\text{U}/\text{g}$  of silica soil requires about 53,000 kg  $^{235}\text{U}$  as 10 wt % enriched uranium to be critical, whereas a  $1.31 \times 1.31 \times 0.131 \text{ m}$  pseudo-infinite slab of silica soil contaminated with  $0.032 \text{ g }^{235}\text{U}/\text{g}$  of silica soil requires about 11.3 kg  $^{235}\text{U}$  as 10 wt % enriched uranium to be critical. This differential is a nearly 21-fold increase in uranium density and a nearly 5,000-fold decrease in uranium mass.

The available mass of  $^{235}\text{U}$  in the Barnwell trenches ranges from less than 1 kg to about 1070 kg with assumed homogenized enrichments of 100 and 0.21 wt %. Taken together with the available information on the spatial distribution of the uranium in the trenches (including average  $^{235}\text{U}$  density), these combinations of mass and enrichment were not judged to present a criticality safety concern for the Barnwell LLW site. However, for a single Barnwell disposal trench, Trench 23, the 175 kg of highly enriched uranium contained in the trench caused sufficient concern that a trench-specific investigation was performed (see Appendix B). The resulting investigation indicated subcriticality was ensured by the physical distribution of the fissile material and the commingling of the material with substantial quantities of SM. Without site-specific information regarding the spatial distribution and mass of disposals, such as for the Barnwell Trench 23 disposal, there could be concern for disposal sites having sufficient inventory to pose a potential criticality safety concern.

- How does U(10) compare to U(100)? How much more concentration of uranium is needed for U(10) than for U(100)?

The criticality safety calculations showed that higher concentrations of  $^{235}\text{U}$  were needed for U(10) than U(100), as expected. At the minimum concentration that achieved safety concern, the concentration ratio of  $^{235}\text{U}$  for U(10) to that for U(100) was 1.6. Variances in this ratio have not been analyzed on a point-by-point basis and could be larger and smaller than the value at the minimum concentration. Furthermore, the differences in the minimum potential critical mass of  $^{235}\text{U}$  for U(10) and U(100) are small. Only when enrichments drop below 10 wt % do minimum masses increase significantly.

- What chemical conditions and physical aspects of trenches are conducive to increasing uranium concentration?

Some trenches have sufficient disposal masses to pose a criticality safety concern, and the conditions are potentially available to mobilize and precipitate uranium. At the Barnwell LLW disposal facility, the water is slightly oxidizing, which enhances the potential for the transport of uranium. The presence of wood and iron containers within the trenches increases the potential for reducing zones to form. Transport could occur as water flushes through the waste and forms uranyl carbonate complexes. Degradation of waste containers can produce a reducing zone in the waste trench, if a water-saturated environment is present to limit oxygen concentrations. These hypothetical reducing zones can precipitate uranium, as discussed below. The zones are hypothetical because they have not actually been observed at the site.

The hypothetical nature of the reducing zones is an important limitation to the current work because the mass required to cause a criticality safety concern is dependent upon the geometry of the fissile material, which in turn is dependent on specific geometries of the reducing zones. Multiple flow paths would need to funnel the uranium from large trench volumes (*if* the SNM is distributed across the trench volume) to very small reducing zones in order to increase the concentration of  $^{235}\text{U}$  to a criticality safety concern. If larger reducing zones form, either the  $^{235}\text{U}$  will be too diffuse, or larger sources of  $^{235}\text{U}$  than are reported to be present in the trench are required.

It is difficult to calculate travel time for the accumulation of uranium. Since kilogram quantities of  $^{235}\text{U}$  are needed, and transporting waters have only mg/L of  $^{235}\text{U}$ , millions of liters of water need to flush through a reducing zone. Speculation about flow rates and conditions is needed to make estimates, but travel times are long. Tens of thousands of years are needed, assuming a 1-m/year velocity, a 40-vol % porosity, and no dispersion.

- Can reducing zones, which precipitate uranium, be sustained to enable critical masses to accumulate?

Yes. In the analysis, reducing zones formed very efficient barriers to uranium transport, precipitating nearly 100% of the uranium in solution. The reducing zones did not become oxidizing despite the influx of oxidized water over the range of pore volumes modeled: 500 to 30,000 L. The source of the reducing agent was postulated to be steel drums or wooden crates.

- How could disposal practices, in particular at the Barnwell LLW disposal facility, enhance or mitigate the development of critical masses of SNM?

Some disposal practices and trench designs can increase the potential for criticality and others can reduce it. Trenches dug in impermeable material tend to retain the fissile material within their confines, increasing the potential for criticality. Trenches that allow for contaminant release reduce the potential for criticality by increasing the volume of the geologic medium into which the uranium can spread. At Barnwell, tritium release has occurred, but uranium might or might not be released from these trenches, depending on the geochemical conditions. Trenches whose floors are sloped can enhance focused flow, increasing the potential for accumulation of uranium. Consequently, French drains and sumps can be sites for SNM accumulation, indicating the desirability to restrict (when possible) their geometries and size to avoid potential critical geometries. Maintaining relatively impermeable caps on the trenches should tend to reduce the uranium migration by limiting the quantity of water available and the potential for locally reducing conditions where  $^{235}\text{U}$  could accumulate by promoting aeration of the trench due to its unsaturated conditions. Commingling of SNM and SM

may reduce the enrichment of the mobilized uranium if the two are leached similarly and well blended. To assume similar leaching and thorough blending, it would be necessary to consider the types of packages containing SNM and SM and their locations in the trench to estimate the time when packages start leaking and the rate of leaching. The Trench 23 records that were examined in detail indicate that homogenization of SNM and SM would likely occur because of the relatively uniform intermingling of SNM and SM emplacements. The presence of materials that can act as effective neutron absorbers can reduce the potential for criticality. Materials that sorb uranium can slow the transfer process and disperse the SNM if the sorbing material is dispersed. On the other hand, materials such as bulk carbon and beryllium can act as low-neutron-absorbing moderators, thereby reducing the needed density, but increasing the needed mass, of uranium for criticality.

Although conditions that permit criticality safety concerns are not impossible, disposal practices have limited the potential concern. This study results in the following recommendations for consideration of license reviews of LLW facilities:

1. Minimize those factors that enhance SNM accumulation.
  - Reduce groundwater infiltration
  - Reduce enrichment
  - Minimize opportunities to create isolated zones of reducing conditions. Avoid organic matter in waste cells
  - Design trench to minimize focused flow
2. Limit the areal density of the fissile materials.
3. Model trench performance using site-specific conditions on a scale that addresses the potential for criticality. Consequently, the observation that the average enrichment of a trench is less than 1 wt %  $^{235}\text{U}$  in the uranium does not necessarily eliminate a criticality concern for the trench. Burial reports may suggest that localized regions of a trench contain quantities of fissile material that greatly exceed the average enrichment.
4. Continue to use sumps in disposal trenches to monitor for the presence of iron, organics, and uranium as indicators of mobility in the trenches. If uranium is observed in the sumps, determine its enrichment. Changes in redox conditions may be monitored by changes in different iron species. Even though it may take many years for sufficient buildup of uranium, early detection of mobile iron and uranium would indicate changes in the trench water chemistry.

In summary, the concentration of dissolved uranium in reducing zones is a possible mechanism for reconcentrating SNM within LLW disposal facilities. Further study of the geometry of these reducing zones is needed to evaluate the potential for concentrating relatively small critical masses (e.g., spherical deposits) and the associated predicted consequences. Further evaluation of flow through the trenches would better define the effect of the source term and its distribution within the trench on the development of a critical mass. Evaluation of mitigating factors, such as multiple-barrier disposal in the new trenches and the effect of sorption by organics and iron oxyhydroxides to inhibit transport out of the waste containers, will also better define the analyses presented here.



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**APPENDIX A**

**Criticality Study Results**



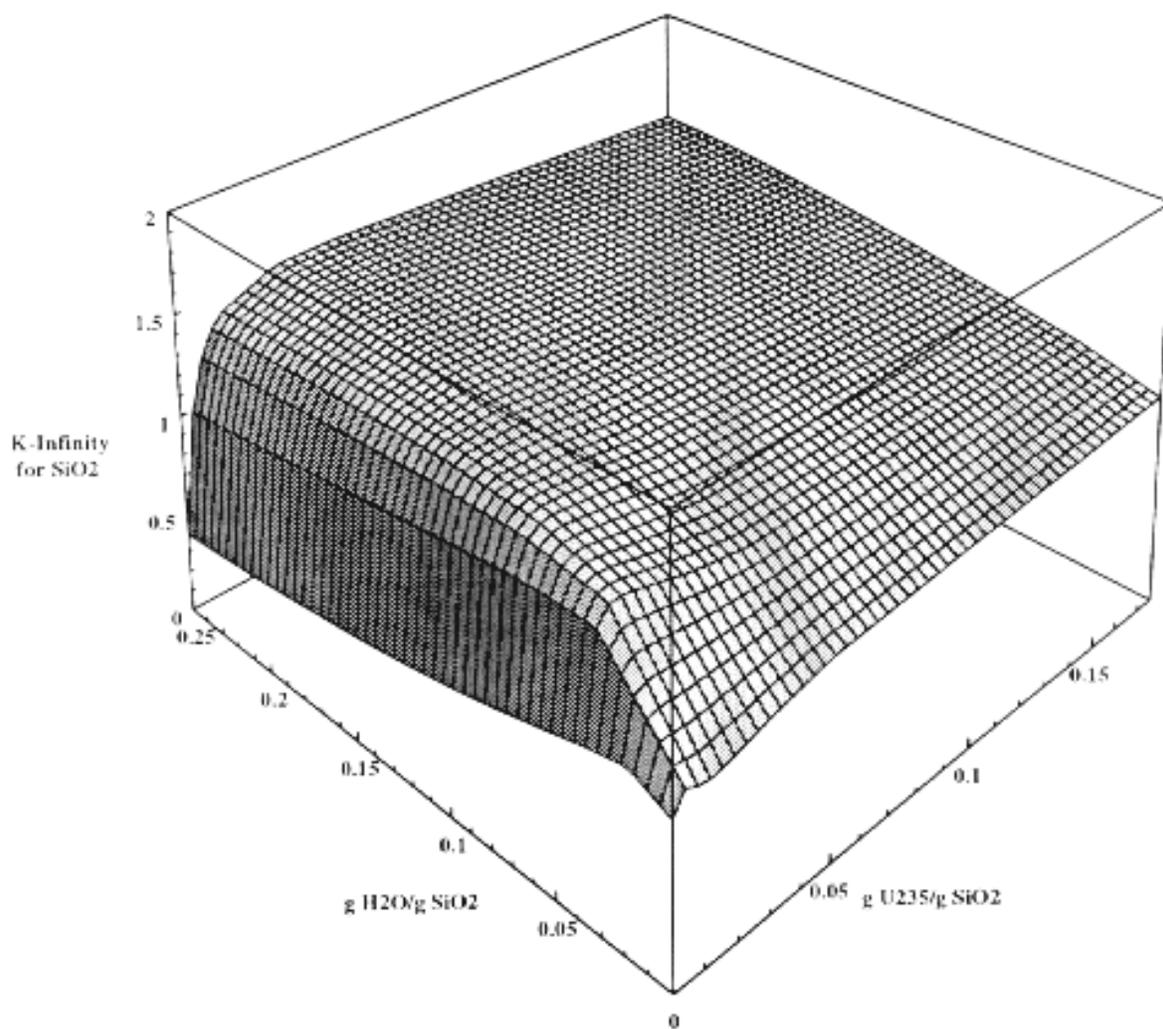


Figure A.1 Infinite media neutron multiplication factor ( $k_{\infty}$ ) vs g  $\text{H}_2\text{O}$ /g  $\text{SiO}_2$  and g  $^{235}\text{U}$ /g  $\text{SiO}_2$

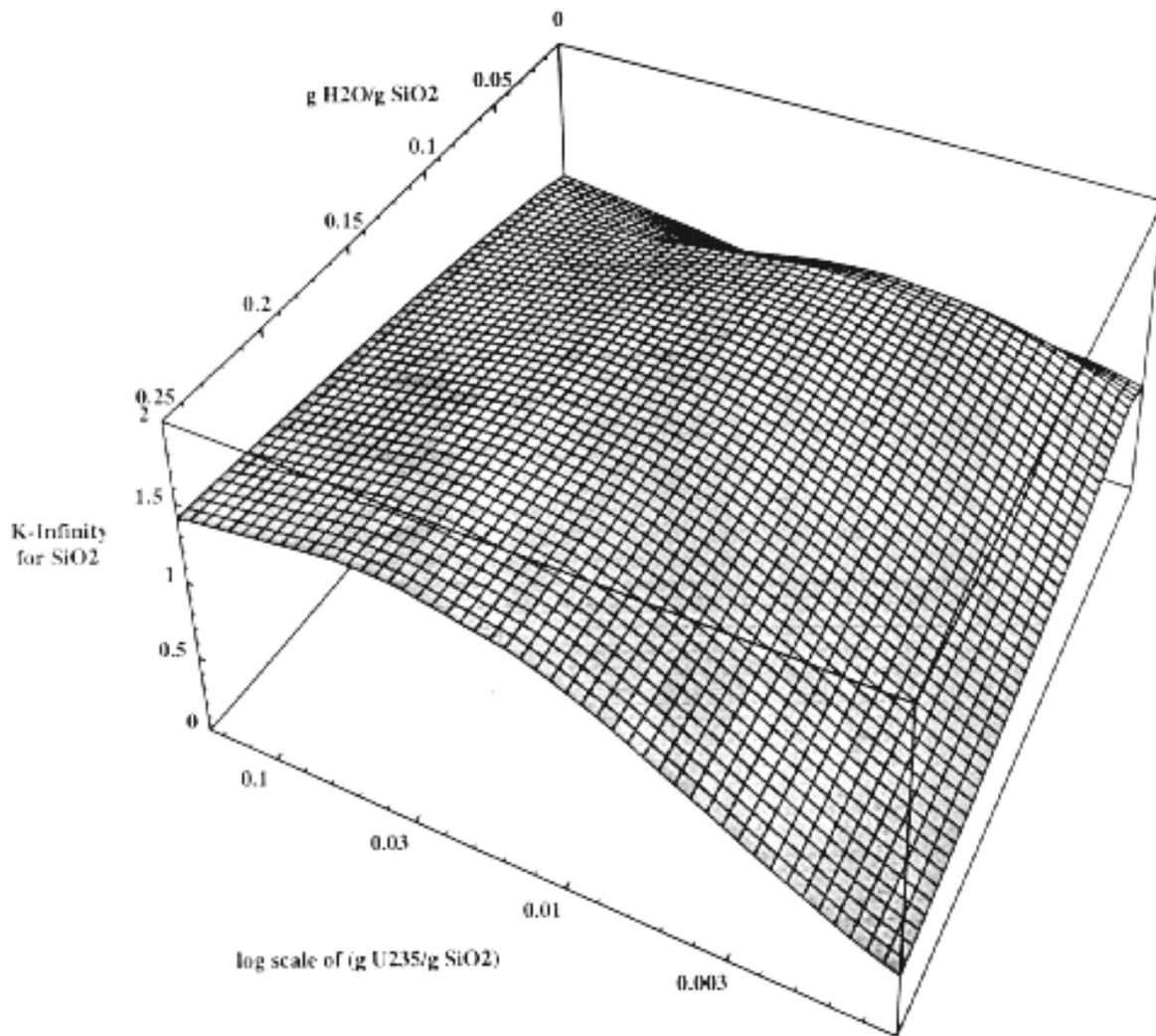


Figure A.2 Infinite media neutron multiplication factor ( $k_{\infty}$ ) vs  $\text{g H}_2\text{O/g SiO}_2$  and log scale of  $\text{g } ^{235}\text{U/g SiO}_2$

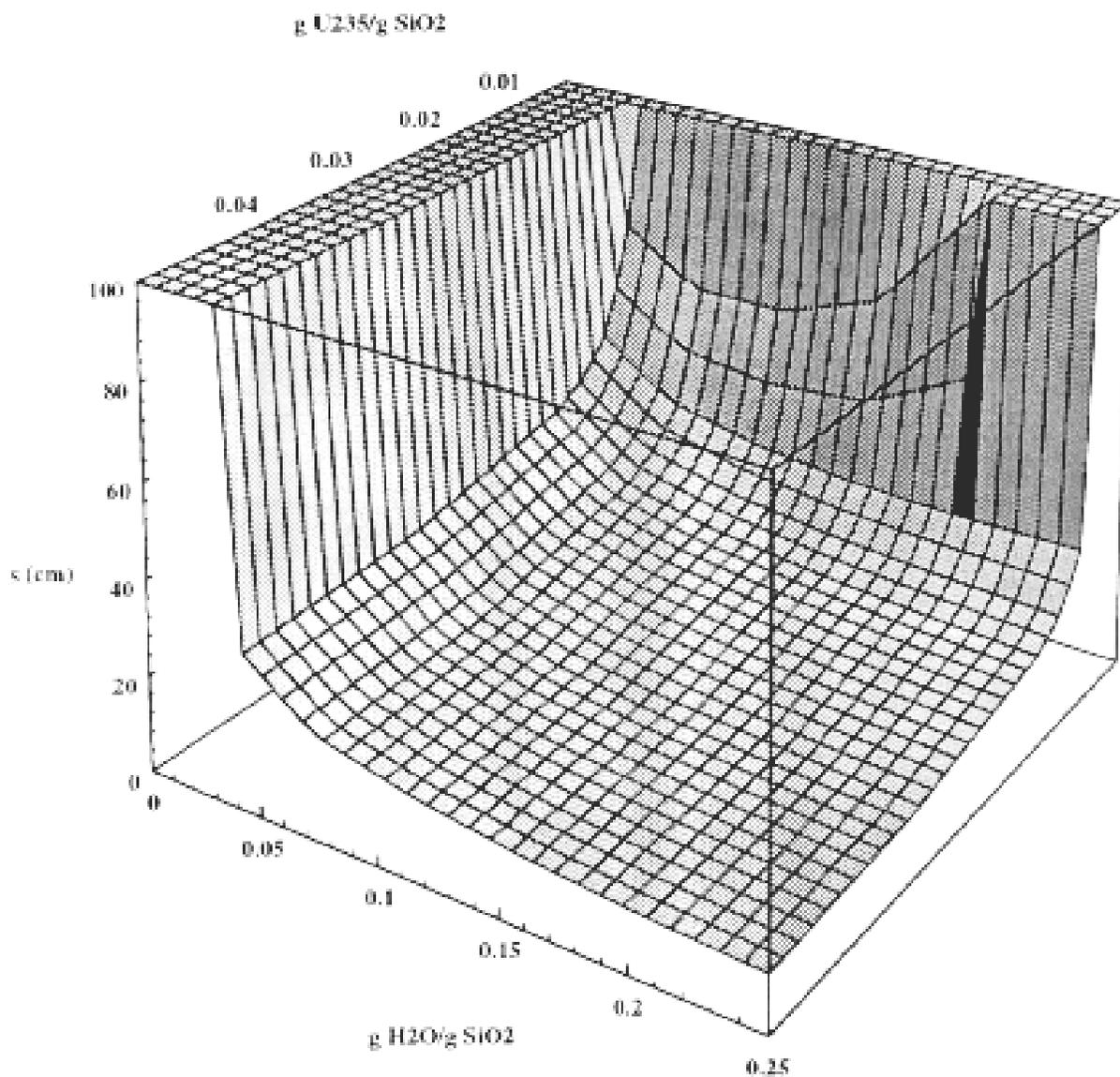


Figure A.3 Infinite slab thickness (cm) vs g H<sub>2</sub>O/g SiO<sub>2</sub> and g <sup>235</sup>U/g SiO<sub>2</sub>

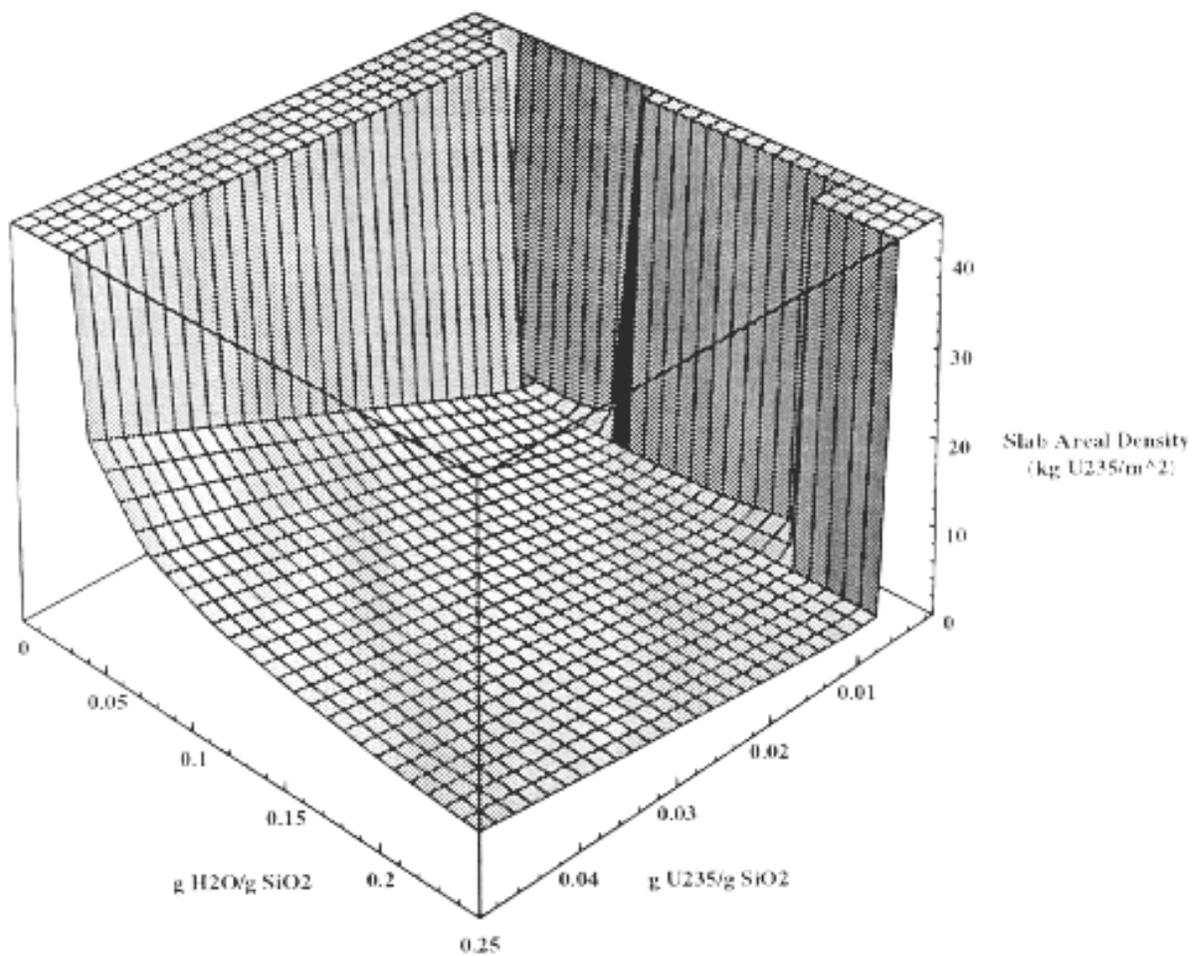


Figure A.4 Infinite slab areal density ( $\text{kg } ^{235}\text{U}/\text{m}^2$ ) vs  $\text{g H}_2\text{O}/\text{g SiO}_2$  and  $\text{g } ^{235}\text{U}/\text{g SiO}_2$

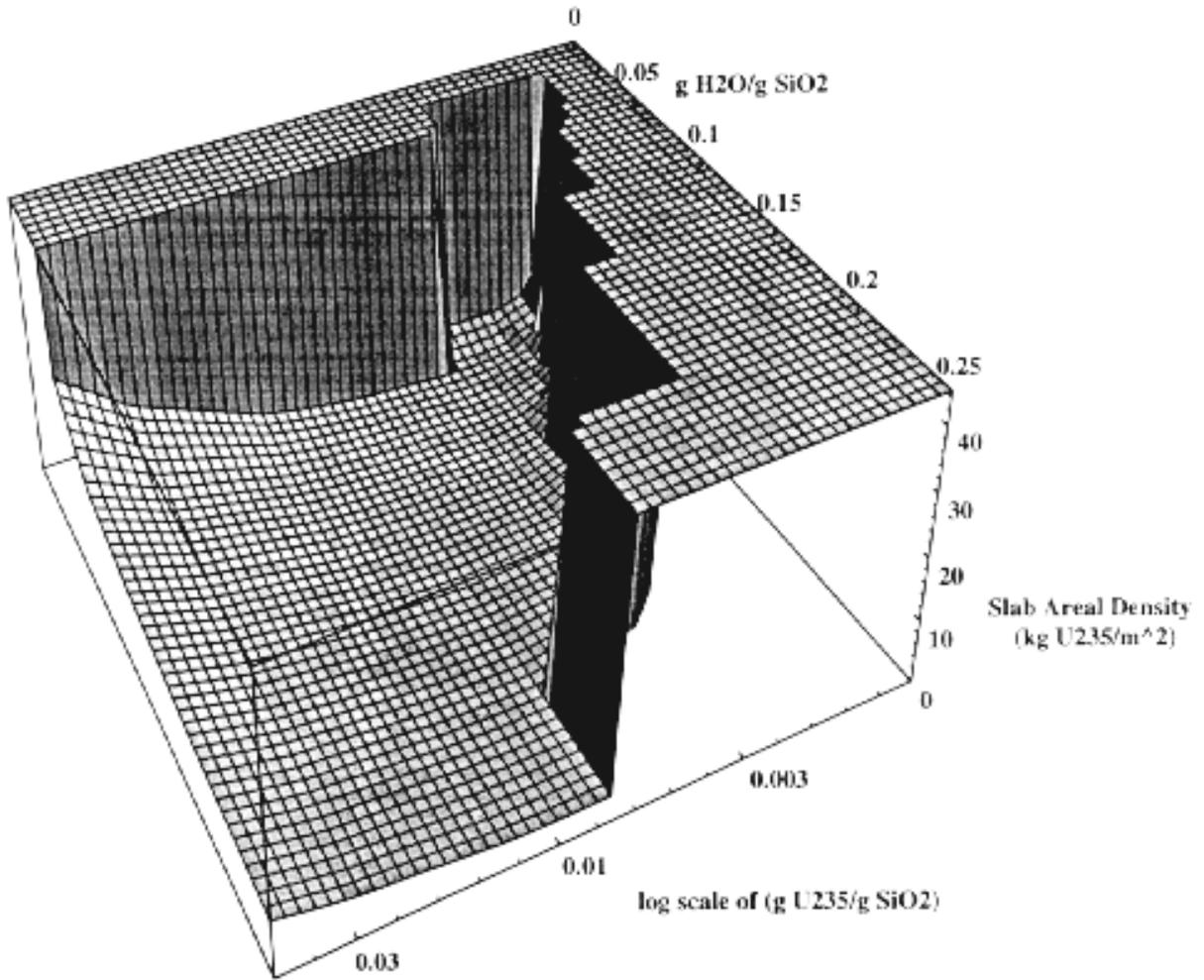


Figure A.5 Infinite slab areal density (kg <sup>235</sup>U/m<sup>2</sup>) vs g H<sub>2</sub>O/g SiO<sub>2</sub> and log scale of g <sup>235</sup>U/g SiO<sub>2</sub>

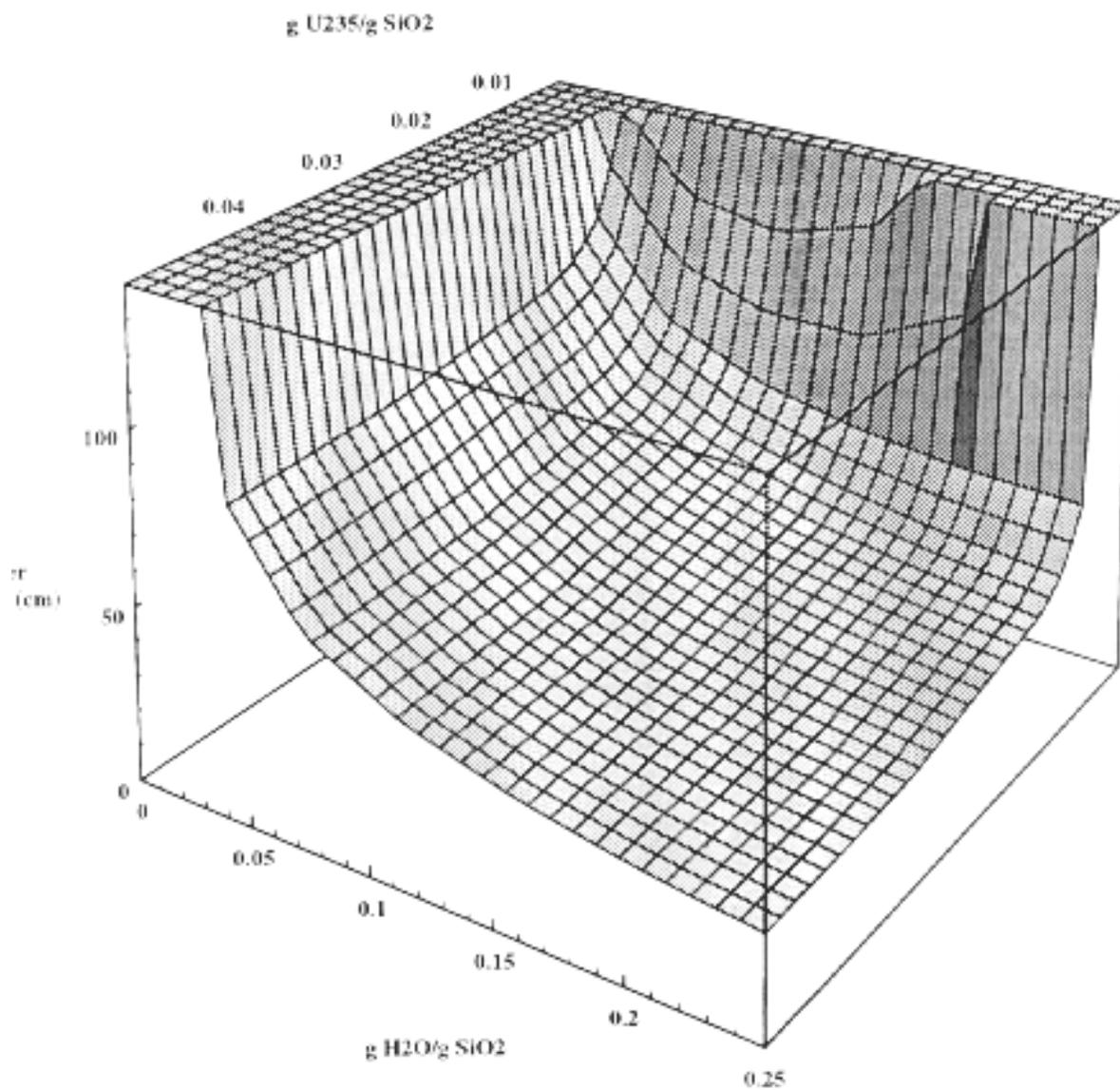


Figure A.6 Infinite cylinder diameter (cm) vs g H<sub>2</sub>O/g SiO<sub>2</sub> and g <sup>235</sup>U/g SiO<sub>2</sub>

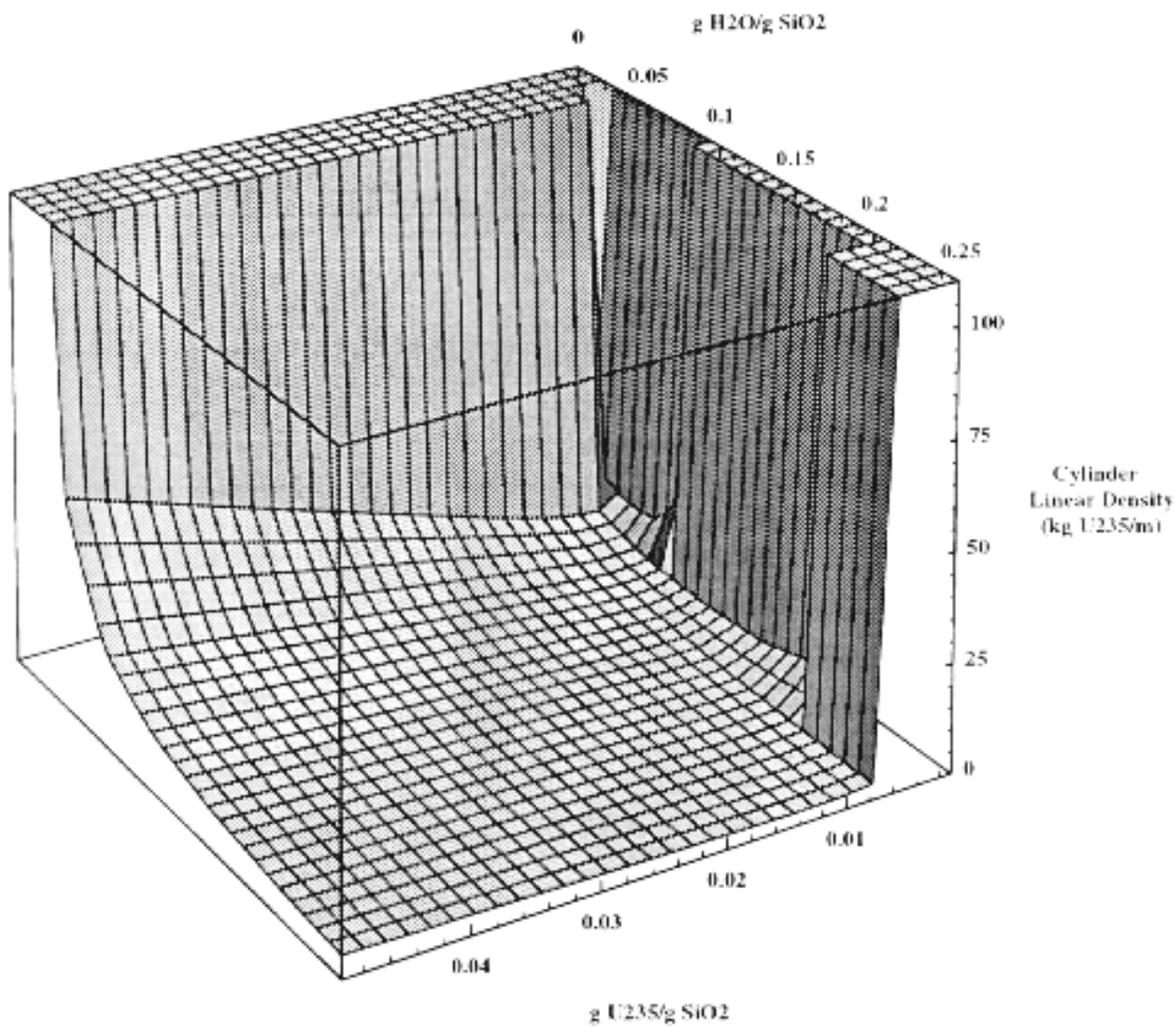


Figure A.7 Infinite cylinder linear density (kg <sup>235</sup>U/m) vs g H<sub>2</sub>O/g SiO<sub>2</sub> and g <sup>235</sup>U/g SiO<sub>2</sub>

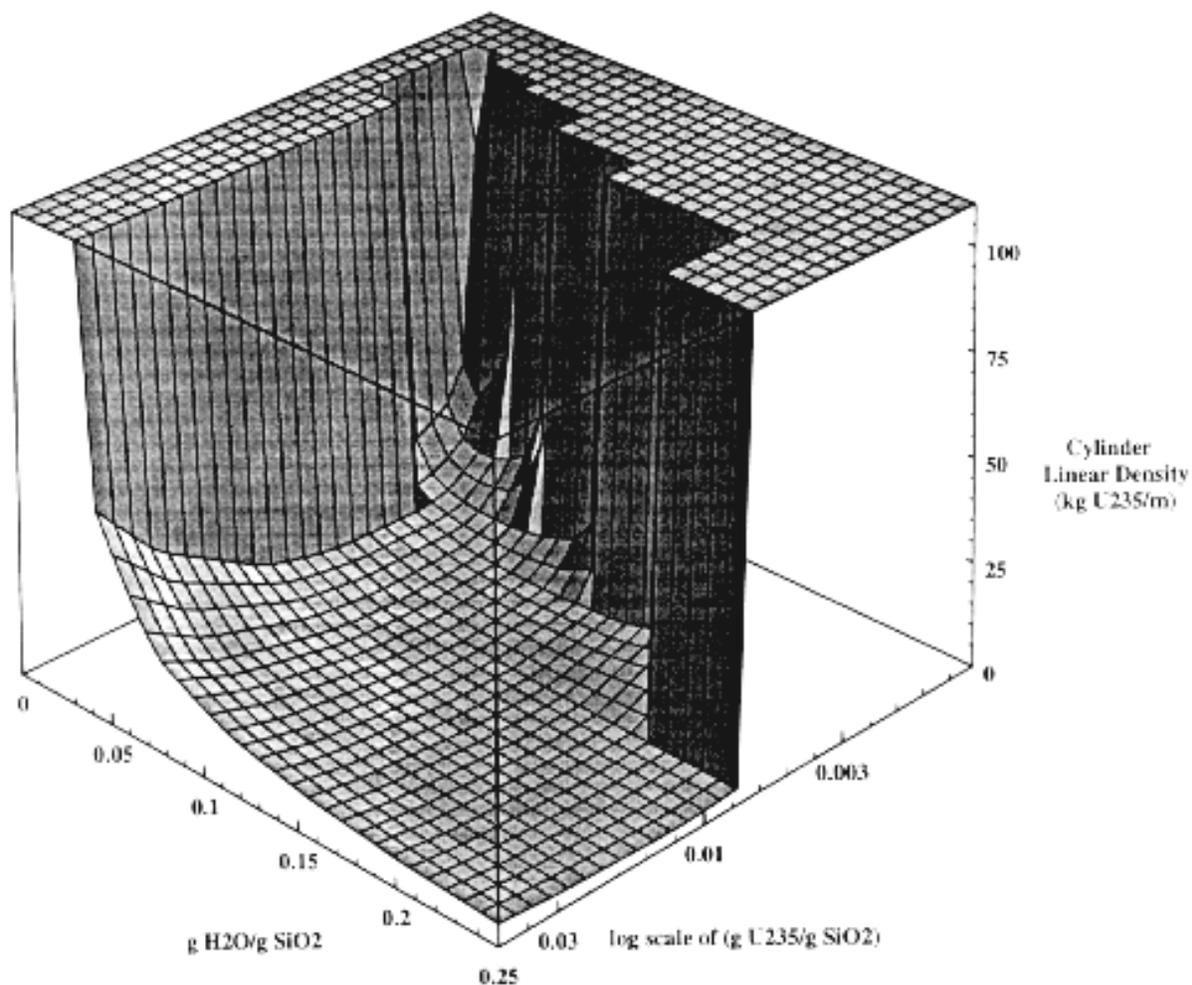


Figure A.8 Infinite cylinder linear density (kg <sup>235</sup>U/m) vs g H<sub>2</sub>O/g SiO<sub>2</sub> and log scale of g <sup>235</sup>U/g SiO<sub>2</sub>

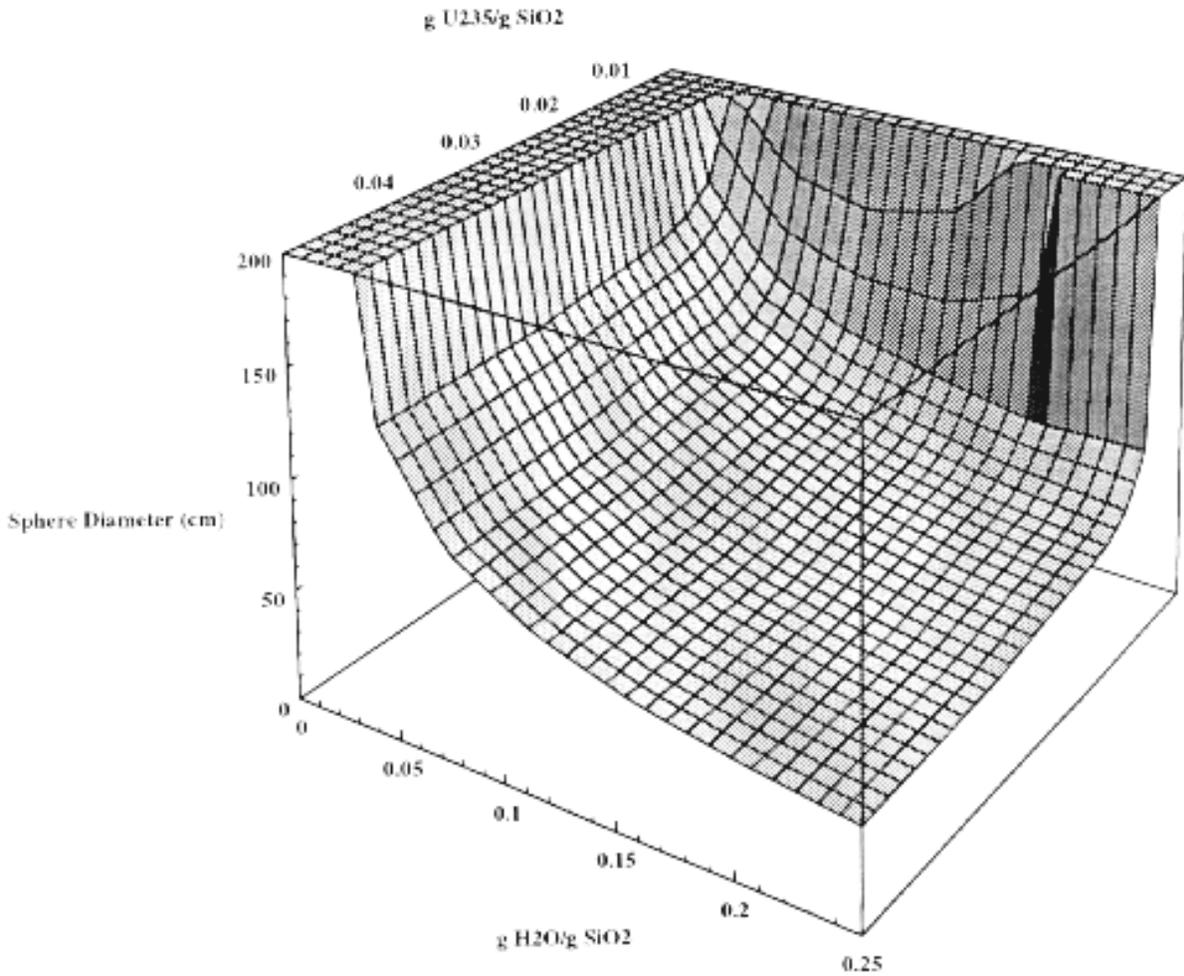


Figure A.9 Sphere diameter (cm) vs g H<sub>2</sub>O/g SiO<sub>2</sub> and g <sup>235</sup>U/g SiO<sub>2</sub>

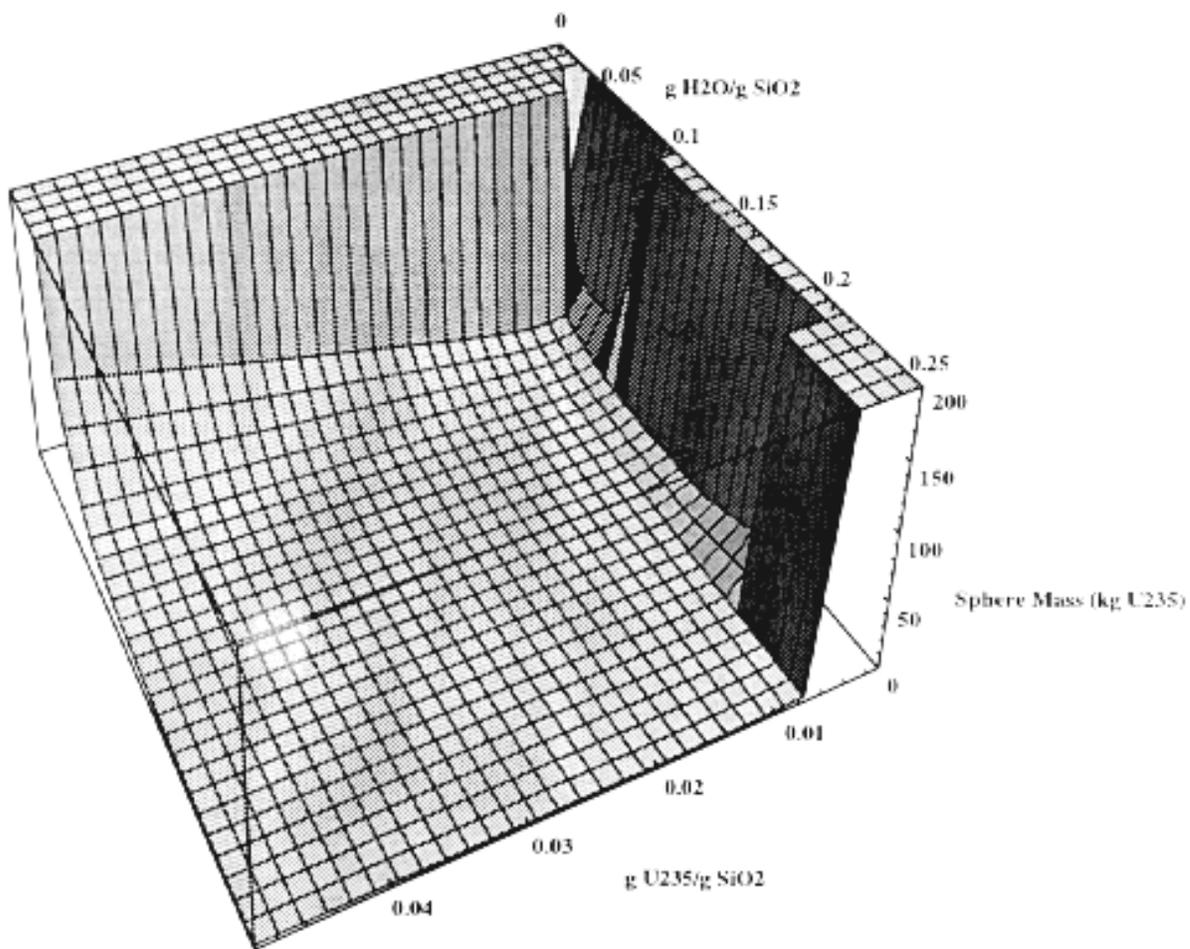


Figure A.10 Sphere mass (kg <sup>235</sup>U) vs g H<sub>2</sub>O/g SiO<sub>2</sub> and g <sup>235</sup>U/g SiO<sub>2</sub>

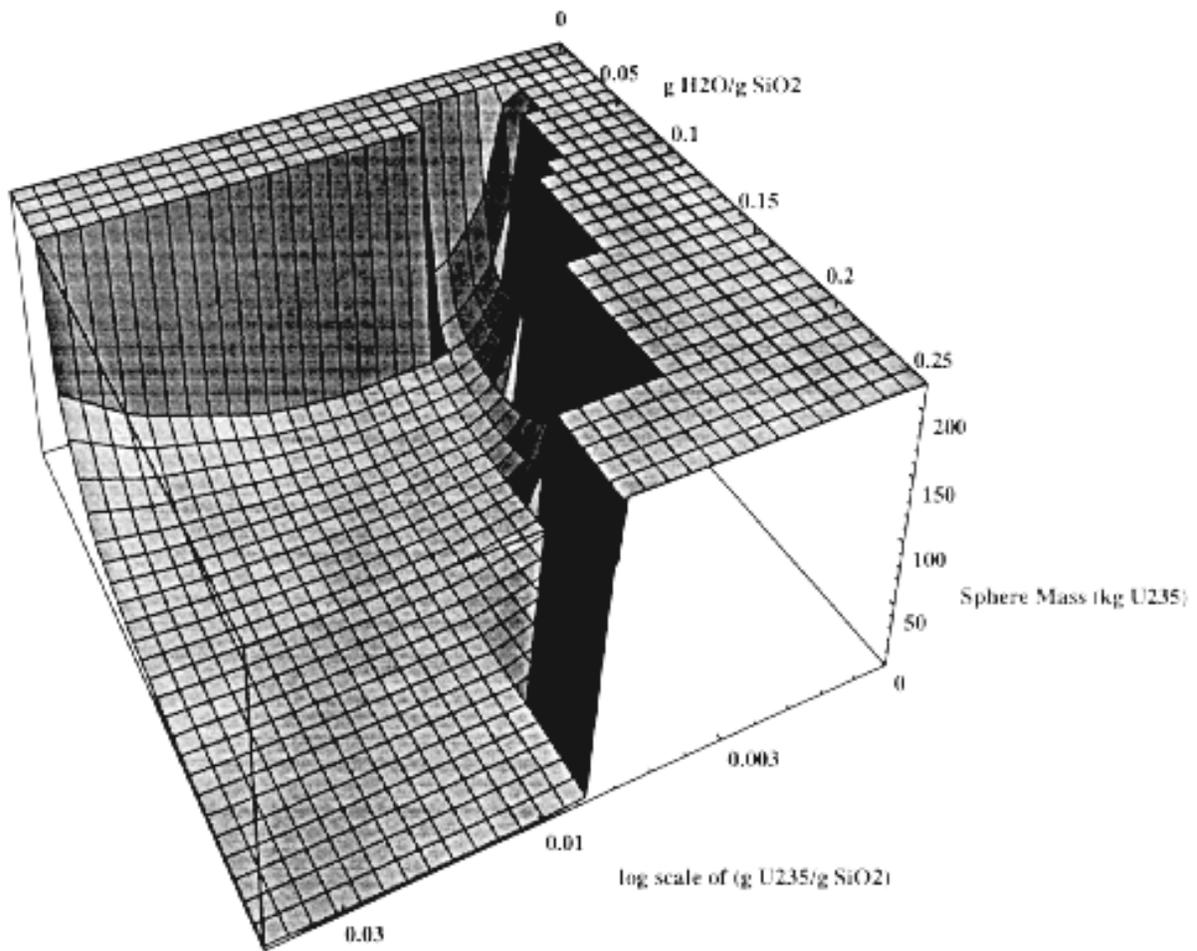


Figure A.11 Sphere mass (kg <sup>235</sup>U) vs g H<sub>2</sub>O/g SiO<sub>2</sub> and log scale of g <sup>235</sup>U/g SiO<sub>2</sub>

Table A.1 U(10) plus H<sub>2</sub>O plus SiO<sub>2</sub>-soil (S-S) results

Line entry	<sup>235</sup> U content		Water content		k <sub>∞</sub> or k-inf	Critical infinite slab		Critical infinite cylinder		Critical sphere	
	g <sup>235</sup> U/cm <sup>3</sup>	g <sup>235</sup> U/gS-S	g H <sub>2</sub> O/cm <sup>3</sup>	g H <sub>2</sub> O/gS-S		Thickness (cm)	Areal density (kg <sup>235</sup> U/m <sup>2</sup> )	Diameter (cm)	Linear density (kg <sup>235</sup> U/m)	Diameter (cm)	Mass (kg <sup>235</sup> U)
1	0.0005	0.0003125	0	0.	0.430						
2	0.0005	0.000313	0.058	0.03625	0.356						
3	0.0005	0.000313	0.119	0.07438	0.282						
4	0.0005	0.000313	0.183	0.11438	0.230						
5	0.0005	0.000313	0.251	0.15688	0.193						
6	0.0005	0.000313	0.324	0.2025	0.164						
7	0.0005	0.000313	0.4	0.25	0.142						
8	0.0014179	0.000886	0	0	0.713						
9	0.0014179	0.000886	0.029	0.01813	0.779						
10	0.0014179	0.000886	0.058	0.03625	0.73						
11	0.0014179	0.000886	0.0885	0.05531	0.672						
12	0.0014179	0.000886	0.119	0.07438	0.62						
13	0.0014179	0.000886	0.151	0.09438	0.572						
14	0.0014179	0.000886	0.183	0.11438	0.53						
15	0.0014179	0.000886	0.217	0.13563	0.492						
16	0.0014179	0.000886	0.251	0.15688	0.458						
17	0.0014179	0.000886	0.2875	0.17969	0.427						
18	0.0014179	0.000886	0.324	0.2025	0.4						
19	0.0014179	0.000886	0.362	0.22625	0.375						
20	0.0014179	0.000886	0.4	0.25	0.353						
21	0.00163	0.0010188	0.	0.	0.744						
22	0.00163	0.0010188	0.029	0.029	0.832						
23	0.00163	0.0010188	0.058	0.058	0.787						

Table A.1 (continued)

Line entry	<sup>235</sup> U content		Water content		k <sub>∞</sub> or k-inf	Critical infinite slab		Critical infinite cylinder		Critical sphere	
	g <sup>235</sup> U/cm <sup>3</sup>	g <sup>235</sup> U/gS-S	g H <sub>2</sub> O/cm <sup>3</sup>	g H <sub>2</sub> O/gS-S		Thickness (cm)	Areal density (kg <sup>235</sup> U/m <sup>2</sup> )	Diameter (cm)	Linear density (kg <sup>235</sup> U/m)	Diameter (cm)	Mass (kg <sup>235</sup> U)
24	0.00163	0.0010188	0.0885	0.0885	0.73						
25	0.00163	0.0010188	0.119	0.119	0.677						
26	0.00163	0.0010188	0.151	0.151	0.627						
27	0.00163	0.0010188	0.183	0.183	0.583						
28	0.00163	0.0010188	0.217	0.217	0.543						
29	0.00163	0.0010188	0.251	0.251	0.507						
30	0.00163	0.0010188	0.2875	0.2875	0.474						
31	0.00163	0.0010188	0.324	0.324	0.445						
32	0.00163	0.0010188	0.362	0.362	0.418						
33	0.00163	0.0010188	0.4	0.4	0.394						
34	0.0018	0.001125	0.	0.	0.763						
35	0.0018	0.001125	0.029	0.029	0.870						
36	0.0018	0.001125	0.058	0.058	0.828						
37	0.0018	0.001125	0.119	0.119	0.719						
38	0.0018	0.001125	0.183	0.183	0.623						
39	0.0018	0.001125	0.251	0.251	0.544						
40	0.0018	0.001125	0.324	0.324	0.479						
41	0.0018	0.001125	0.4	0.4	0.426						
42	0.0018738	0.0011711	0.	0.	0.771						
43	0.0018738	0.0011711	0.029	0.01813	0.884						
44	0.0018738	0.0011711	0.058	0.03625	0.845						
45	0.0018738	0.0011711	0.0885	0.05531	0.789						
46	0.0018738	0.0011711	0.119	0.07438	0.736						
47	0.0018738	0.0011711	0.151	0.09438	0.684						

Table A.1 (continued)

Line entry	<sup>235</sup> U content		Water content		k <sub>∞</sub> or k-inf	Critical infinite slab		Critical infinite cylinder		Critical sphere	
	g <sup>235</sup> U/cm <sup>3</sup>	g <sup>235</sup> U/gS-S	g H <sub>2</sub> O/cm <sup>3</sup>	g H <sub>2</sub> O/gS-S		Thickness (cm)	Areal density (kg <sup>235</sup> U/m <sup>2</sup> )	Diameter (cm)	Linear density (kg <sup>235</sup> U/m)	Diameter (cm)	Mass (kg <sup>235</sup> U)
48	0.0018738	0.0011711	0.183	0.11438	0.639						
49	0.0018738	0.0011711	0.217	0.13563	0.597						
50	0.0018738	0.0011711	0.251	0.15688	0.560						
51	0.0018738	0.0011711	0.2875	0.17969	0.525						
52	0.0018738	0.0011711	0.324	0.2025	0.494						
53	0.0018738	0.0011711	0.362	0.22625	0.465						
54	0.0018738	0.0011711	0.4	0.25	0.439						
55	0.002154	0.0013463	0.	0.	0.794						
56	0.002154	0.0013463	0.029	0.01813	0.935						
57	0.002154	0.0013463	0.058	0.03625	0.902						
58	0.002154	0.0013463	0.0885	0.05531	0.848						
59	0.002154	0.0013463	0.119	0.07438	0.795						
60	0.002154	0.0013463	0.151	0.09438	0.744						
61	0.002154	0.0013463	0.183	0.11438	0.697						
62	0.002154	0.0013463	0.217	0.13563	0.654						
63	0.002154	0.0013463	0.251	0.15688	0.615						
64	0.002154	0.0013463	0.2875	0.17969	0.578						
65	0.002154	0.0013463	0.324	0.2025	0.546						
66	0.002154	0.0013463	0.362	0.22625	0.515						
67	0.002154	0.0013463	0.4	0.25	0.487						
68	0.0024761	0.0015476	0.	0.	0.811						
69	0.0024761	0.0015476	0.029	0.01813	0.982	464.22	11.4946	758.34	111.84	1020.78	1378.9967
70	0.0024761	0.0015476	0.058	0.03625	0.957	966.8	23.9389	1684.72	551.97	2867.82	30578.9410
71	0.0024761	0.0015476	0.0885	0.05531	0.907						

Table A.1 (continued)

Line entry	<sup>235</sup> U content		Water content		k <sub>∞</sub> or k-inf	Critical infinite slab		Critical infinite cylinder		Critical sphere	
	g <sup>235</sup> U/cm <sup>3</sup>	g <sup>235</sup> U/gS-S	g H <sub>2</sub> O/cm <sup>3</sup>	g H <sub>2</sub> O/gS-S		Thickness (cm)	Areal density (kg <sup>235</sup> U/m <sup>2</sup> )	Diameter (cm)	Linear density (kg <sup>235</sup> U/m)	Diameter (cm)	Mass (kg <sup>235</sup> U)
72	0.0024761	0.0015476	0.119	0.07438	0.856						
73	0.0024761	0.0015476	0.151	0.09438	0.804						
74	0.0024761	0.0015476	0.183	0.11438	0.757						
75	0.0024761	0.0015476	0.217	0.13563	0.713						
76	0.0024761	0.0015476	0.251	0.15688	0.673						
77	0.0024761	0.0015476	0.2875	0.17969	0.635						
78	0.0024761	0.0015476	0.324	0.2025	0.600						
79	0.0024761	0.0015476	0.362	0.22625	0.568						
80	0.0024761	0.0015476	0.4	0.25	0.539						
81	0.0028465	0.0017791	0.	0.	0.827						
82	0.0028465	0.0017791	0.029	0.01813	1.027	275.92	7.8541	461.7	47.6563	629.16	371.18729
83	0.0028465	0.0017791	0.058	0.03625	1.011	258.9	7.3696	429.26	41.1947	576.8	286.01284
84	0.0028465	0.0017791	0.0885	0.05531	0.965	486.26	13.8414	829.7	153.901	1188.78	2503.8843
85	0.0028465	0.0017791	0.119	0.07438	0.916						
86	0.0028465	0.0017791	0.151	0.09438	0.865						
87	0.0028465	0.0017791	0.183	0.11438	0.818						
88	0.0028465	0.0017791	0.217	0.13563	0.774						
89	0.0028465	0.0017791	0.251	0.15688	0.732						
90	0.0028465	0.0017791	0.2875	0.17969	0.693						
91	0.0028465	0.0017791	0.324	0.2025	0.658						
92	0.0028465	0.0017791	0.362	0.22625	0.624						
93	0.0028465	0.0017791	0.4	0.25	0.593						
94	0.0028639	0.0017899	0	0.	0.828						
95	0.0028639	0.0017899	0.029	0.01813	1.029	270.62	7.7503	456.82	46.9394	621.22	359.49490

Table A.1 (continued)

Line entry	<sup>235</sup> U content		Water content		k <sub>∞</sub> or k-inf	Critical infinite slab		Critical infinite cylinder		Critical sphere	
	g <sup>235</sup> U/cm <sup>3</sup>	g <sup>235</sup> U/gS-S	g H <sub>2</sub> O/cm <sup>3</sup>	g H <sub>2</sub> O/gS-S		Thickness (cm)	Areal density (kg <sup>235</sup> U/m <sup>2</sup> )	Diameter (cm)	Linear density (kg <sup>235</sup> U/m)	Diameter (cm)	Mass (kg <sup>235</sup> U)
96	0.0028639	0.0017899	0.058	0.03625	1.013	253.3	7.2543	420.06	39.6889	488.7	175.01830
97	0.0028639	0.0017899	0.0885	0.05531	0.967	452.94	12.9717	741.02	123.511	1045.96	1715.9383
98	0.0028639	0.0017899	0.119	0.07438	0.918						
99	0.0028639	0.0017899	0.151	0.09438	0.868						
100	0.0028639	0.0017899	0.183	0.11438	0.821						
101	0.0028639	0.0017899	0.217	0.13563	0.776						
102	0.0028639	0.0017899	0.251	0.15688	0.735						
103	0.0028639	0.0017899	0.2875	0.17969	0.696						
104	0.0028639	0.0017899	0.324	0.2025	0.66						
105	0.0028639	0.0017899	0.362	0.22625	0.626						
106	0.0028639	0.0017899	0.4	0.25	0.596						
107	0.0032722	0.0020451	0	0.	0.839						
108	0.0032722	0.0020451	0.029	0.01813	1.076	206.12	6.7447	356.8	32.7174	488.7	199.97028
109	0.0032722	0.0020451	0.058	0.03625	1.071	205.9	6.7375	299.36	23.0312	410.46	118.48161
110	0.0032722	0.0020451	0.0885	0.05531	1.031	174.48	5.7093	328.62	27.7534	439.7	145.64911
111	0.0032722	0.0020451	0.119	0.07438	0.975	314.6	10.2943	513.12	67.6654	696.78	579.59612
112	0.0032722	0.0020451	0.151	0.09438	0.926						
113	0.0032722	0.0020451	0.183	0.11438	0.880						
114	0.0032722	0.0020451	0.217	0.13563	0.835						
115	0.0032722	0.0020451	0.251	0.15688	0.793						
116	0.0032722	0.0020451	0.2875	0.17969	0.754						
117	0.0032722	0.0020451	0.324	0.2025	0.717						
118	0.0032722	0.0020451	0.362	0.22625	0.682						
119	0.0032722	0.0020451	0.4	0.25	0.651						

Table A.1 (continued)

Line entry	<sup>235</sup> U content		Water content		k <sub>∞</sub> or k-inf	Critical infinite slab		Critical infinite cylinder		Critical sphere	
	g <sup>235</sup> U/cm <sup>3</sup>	g <sup>235</sup> U/gS-S	g H <sub>2</sub> O/cm <sup>3</sup>	g H <sub>2</sub> O/gS-S		Thickness (cm)	Areal density (kg <sup>235</sup> U/m <sup>2</sup> )	Diameter (cm)	Linear density (kg <sup>235</sup> U/m)	Diameter (cm)	Mass (kg <sup>235</sup> U)
120	0.0036054	0.0022534	0	0.	0.844						
121	0.0036054	0.0022534	0.029	0.01813	1.094	178.4	6.432	314.58	28.0223	435.52	155.94676
122	0.0036054	0.0022534	0.058	0.03625	1.095	144.80	5.2206	254.54	18.3465	351.28	81.829957
123	0.0036054	0.0022534	0.0885	0.05531	1.059	159.32	5.7441	255.18	18.4389	348.92	80.191741
124	0.0036054	0.0022534	0.119	0.07438	1.015	179.48	6.471	300.34	25.5428	405.2	125.59153
125	0.0036054	0.0022534	0.151	0.09438	0.968	338.54	12.2057	559.48	88.6363	770	861.83520
126	0.0036054	0.0022534	0.183	0.11438	0.923						
127	0.0036054	0.0022534	0.217	0.13563	0.878						
128	0.0036054	0.0022534	0.251	0.15688	0.836						
129	0.0036054	0.0022534	0.2875	0.17969	0.796						
130	0.0036054	0.0022534	0.324	0.2025	0.760						
131	0.0036054	0.0022534	0.362	0.22626	0.724						
132	0.0036054	0.0022534	0.4	0.25	0.692						
133	0.004539	0.0028369	0.	0.	0.848						
134	0.004539	0.0028369	0.029	0.01813	1.147	137.54	6.2429	252.60	22.7465	354.52	105.89635
135	0.004539	0.0028369	0.058	0.03625	1.168	106.26	4.8231	194.66	13.5083	272.8	48.249396
136	0.004539	0.0028369	0.0885	0.05531	1.143	99.46	4.5145	178.30	11.3331	248.16	36.320692
137	0.004539	0.0028369	0.119	0.07438	1.107	101.48	4.6062	178.04	11.3001	244.74	34.839639
138	0.004539	0.0028369	0.151	0.09438	1.064	111.42	5.0574	191.08	13.0160	261.02	42.264928
139	0.004539	0.0028369	0.183	0.11438	1.022	134.88	6.1222	224.38	17.9480	304.96	67.404271
140	0.004539	0.0028369	0.217	0.13563	0.98	204.24	9.2705	333.30	39.6022	444.22	208.33062
141	0.004539	0.0028369	0.251	0.15688	0.939						
142	0.004539	0.0028369	0.2875	0.17969	0.899						
143	0.004539	0.0028369	0.324	0.2025	0.862						

Table A.1 (continued)

Line entry	<sup>235</sup> U content		Water content		k <sub>∞</sub> or k-inf	Critical infinite slab		Critical infinite cylinder		Critical sphere	
	g <sup>235</sup> U/cm <sup>3</sup>	g <sup>235</sup> U/gS-S	g H <sub>2</sub> O/cm <sup>3</sup>	g H <sub>2</sub> O/gS-S		Thickness (cm)	Areal density (kg <sup>235</sup> U/m <sup>2</sup> )	Diameter (cm)	Linear density (kg <sup>235</sup> U/m)	Diameter (cm)	Mass (kg <sup>235</sup> U)
144	0.004539	0.0028369	0.362	0.22625	0.826						
145	0.004539	0.0028369	0.4	0.25	0.793						
146	0.0057142	0.0035714	0.	0	0.845						
147	0.0057142	0.0035714	0.029	0.01813	1.187	113.84	6.505	217.88	21.3049	309.16	88.410517
148	0.0057142	0.0035714	0.058	0.03625	1.229	85.06	4.8605	162.50	11.8509	230.72	36.745974
149	0.0057142	0.0035714	0.0885	0.05531	1.218	76.08	4.3474	142.68	9.13631	201.26	24.390822
150	0.0057142	0.0035714	0.119	0.07438	1.190	73.42	4.1954	134.86	8.16227	189.08	20.225106
151	0.0057142	0.0035714	0.151	0.09438	1.154	74	4.2285	133.02	7.94106	184.84	18.894783
152	0.0057142	0.0035714	0.183	0.11438	1.117	77.36	4.4205	135.94	8.29353	187.6	19.753885
153	0.0057142	0.0035714	0.217	0.13563	1.078	84.16	4.8091	145.14	9.45407	198.28	23.323341
154	0.0057142	0.0035714	0.251	0.15688	1.039	97.18	5.5531	163.42	11.99	222.14	32.797016
155	0.0057142	0.0035714	0.2875	0.17969	1.001	125.58	7.1759	206.36	19.11	277.76	64.115516
156	0.0057142	0.0035714	0.324	0.2025	0.965	234.28	13.3872	401.80	72.4544	484	339.22677
157	0.0057142	0.0035714	0.362	0.22625	0.929						
158	0.0057142	0.0035714	0.4	0.25	0.896						
159	0.0063	0.0039375	0.	0	0.841						
160	0.0063	0.0039375	0.029	0.01813	1.200	106.76	6.7259	207.36	21.2749	296.06	85.6011
161	0.0063	0.0039375	0.058	0.03625	1.251	78.62	4.9531	152.84	11.5585	218.36	34.344595
162	0.0063	0.0039375	0.119	0.07438	1.222	66.02	4.1593	123.50	7.54681	174.32	17.473536
163	0.0063	0.0039375	0.183	0.11438	1.154	66.58	4.1945	119.22	7.03280	165.88	15.056403
164	0.0063	0.0039375	0.251	0.15688	1.081	75.82	4.7767	130.56	8.43432	178.8	18.855658
165	0.0063	0.0039375	0.324	0.2025	1.008	108.14	6.8128	177.52	15.5928	240.68	45.989549
166	0.0063	0.0039375	0.4	0.25	0.940						
167	0.01167	0.0072938	0.	0.	0.809						

Table A.1 (continued)

Line entry	<sup>235</sup> U content		Water content		k <sub>∞</sub> or k-inf	Critical infinite slab		Critical infinite cylinder		Critical sphere	
	g <sup>235</sup> U/cm <sup>3</sup>	g <sup>235</sup> U/gS-S	g H <sub>2</sub> O/cm <sup>3</sup>	g H <sub>2</sub> O/gS-S		Thickness (cm)	Areal density (kg <sup>235</sup> U/m <sup>2</sup> )	Diameter (cm)	Linear density (kg <sup>235</sup> U/m)	Diameter (cm)	Mass (kg <sup>235</sup> U)
168	0.01167	0.0072938	0.029	0.01813	1.232	80.08	9.3694	170.30	26.6497	249.76	95.4452
169	0.01167	0.0072938	0.058	0.03625	1.336	54.32	6.3391	117.66	12.6887	173.48	31.901984
170	0.01167	0.0072938	0.119	0.07438	1.376	40.88	4.7707	85.8	6.74737	125.52	12.083931
171	0.01167	0.0072938	0.183	0.11438	1.351	36.38	4.2455	73.4	4.93801	106.14	7.3064548
172	0.01167	0.0072938	0.251	0.15688	1.307	34.48	4.0238	67.14	4.13164	96.	5.4060885
173	0.01167	0.0072938	0.324	0.2025	1.255	34.02	3.9701	64.08	3.76361	86.6	3.9684702
174	0.01167	0.0072938	0.4	0.25	1.201	34.68	4.0472	63.54	3.70045	88.82	4.2815573
175	0.01704	0.0106500	0.	0	0.798						
176	0.01704	0.01065	0.029	0.01813	1.216	72.62	12.3454	161.24	34.7114	238.58	120.8789
177	0.01704	0.01065	0.058	0.03625	1.346	46.56	7.9338	107.44	15.4486	160.9	37.165187
178	0.01704	0.01065	0.119	0.07438	1.425	33.44	5.6982	75.14	7.55616	111.94	12.514805
179	0.01704	0.01065	0.183	0.11438	1.428	28.69	4.8888	62.06	5.15445	91.56	6.8483436
180	0.01704	0.01065	0.251	0.15688	1.406	26.2	4.4645	54.68	4.00143	79.92	4.5544361
181	0.01704	0.01065	0.324	0.2025	1.372	24.76	4.2191	50.02	3.34847	72.4	3.3859761
182	0.01704	0.01065	0.4	0.25	1.332	23.94	4.0794	47.06	2.96389	67.5	2.7439709
183	0.0224	0.014	0.	0.	0.801						
184	0.0224	0.014	0.029	0.01813	1.192	69.72	15.6173	158.62	44.2630	235.40	152.9914
185	0.0224	0.014	0.058	0.03625	1.335	42.74	9.5738	102.72	18.5630	154.88	43.574522
186	0.0224	0.014	0.119	0.07438	1.44	29.72	6.6573	70.08	8.64024	105.68	13.842834
187	0.0224	0.014	0.183	0.11438	1.463	25.02	5.6045	56.78	5.15445	84.86	7.1673019
188	0.0224	0.014	0.251	0.15688	1.457	22.44	5.0266	49.10	4.00144	72.76	4.5177759
189	0.0224	0.014	0.324	0.2025	1.436	20.78	4.6547	44.08	3.34847	64.74	3.1824728
190	0.0224	0.014	0.399	0.24938	1.407	19.66	4.4038	40.58	2.9639	59.16	2.4284608
191	0.0509	0.0318125	0	0.	0.873						

Table A.1 (continued)

Line entry	<sup>235</sup> U content		Water content		k <sub>∞</sub> or k-inf	Critical infinite slab		Critical infinite cylinder		Critical sphere	
	g <sup>235</sup> U/cm <sup>3</sup>	g <sup>235</sup> U/gS-S	g H <sub>2</sub> O/cm <sup>3</sup>	g H <sub>2</sub> O/gS-S		Thickness (cm)	Areal density (kg <sup>235</sup> U/m <sup>2</sup> )	Diameter (cm)	Linear density (kg <sup>235</sup> U/m)	Diameter (cm)	Mass (kg <sup>235</sup> U)
192	0.0509	0.0318125	0.029	0.01813	1.107	68.04	34.6324	160.24	102.6478	241.14	373.7009
193	0.0509	0.0318125	0.058	0.03625	1.253	36.34	18.4971	95.92	36.7912	147.06	84.761779
194	0.0509	0.0318125	0.119	0.07438	1.404	23.1	11.7579	61.74	15.2384	94.96	22.821201
195	0.0509	0.0318125	0.183	0.11438	1.474	18.56	9.447	48.16	9.27214	73.86	10.738516
196	0.0509	0.0318125	0.251	0.15688	1.51	16.00	8.144	40.22	6.46683	61.46	6.1871985
197	0.0509	0.0318125	0.324	0.2025	1.525	14.28	7.2685	34.86	4.85805	53.02	3.9722406
198	0.0509	0.0318125	0.397	0.24813	1.53	13.06	6.6475	30.98	3.83681	46.82	2.7353355
199	0.0794	0.049625	0.	0	0.929						
200	0.0794	0.049625	0.029	0.01813	1.078	65.12	51.7053	157.32	154.3354	237.98	560.3270
201	0.0794	0.049625	0.058	0.03625	1.2	34.34	27.266	94.38	55.5482	145.82	128.90518
202	0.0794	0.049625	0.119	0.07438	1.353	21.14	16.7852	59.50	22.0772	92.28	32.669452
203	0.0794	0.049625	0.183	0.11438	1.437	16.66	13.22	45.76	13.0582	70.74	14.716829
204	0.0794	0.049625	0.251	0.15688	1.487	14.18	11.2589	37.8	8.91033	58.24	8.2126578
205	0.0794	0.049625	0.324	0.2025	1.518	12.48	9.9091	32.44	6.56254	49.8	5.1346060
206	0.0794	0.049625	0.396	0.2475	1.536	11.3	8.9722	28.68	5.129	43.88	3.5125212
207	0.2817	0.1760625	0	0	1.044	53.61	151.019	149.16	492.2307	230.28	1801.1676
208	0.2817	0.1760625	0.029	0.01813	1.110	27.66	77.918	90.94	182.9674	143.62	436.9495
209	0.2817	0.1760625	0.058	0.03625	1.146	20.06	56.509	72.38	115.9047	114.88	223.6242
210	0.2817	0.1760625	0.119	0.07438	1.217	16	45.072	52.16	60.1920	82.54	82.9428
211	0.2817	0.1760625	0.183	0.11438	1.281	13.13	36.9872	41.00	37.1904	64.58	39.7264
212	0.2817	0.1760625	0.251	0.15688	1.334	11.24	31.6631	33.84	25.3352	53.06	22.0337
213	0.2817	0.1760625	0.324	0.2025	1.379	9.82	27.6629	28.80	18.3510	44.96	13.4049
214	0.2817	0.1760625	0.385	0.24063	1.41	9	25.353	25.82	14.7499	40.20	9.5821
215	1.	0.625	0.	0	1.151						

Table A.1 (continued)

Line entry	<sup>235</sup> U content		Water content		k <sub>∞</sub> or k-inf	Critical infinite slab		Critical infinite cylinder		Critical sphere	
	g <sup>235</sup> U/cm <sup>3</sup>	g <sup>235</sup> U/gS-S	g H <sub>2</sub> O/cm <sup>3</sup>	g H <sub>2</sub> O/gS-S		Thickness (cm)	Areal density (kg <sup>235</sup> U/m <sup>2</sup> )	Diameter (cm)	Linear density (kg <sup>235</sup> U/m)	Diameter (cm)	Mass (kg <sup>235</sup> U)
216	1.	0.625	0.058	0.03625	1.197						
217	1.	0.625	0.119	0.07438	1.216						
218	1.	0.625	0.183	0.11438	1.23						
219	1.	0.625	0.251	0.15688	1.245						
220	1.	0.625	0.324	0.2025	1.261						
221	1.	0.625	0.346	0.21625	1.267						



## **APPENDIX B**

### **Subcriticality Evaluation for Chem-Nuclear Systems, Inc., Trench 23**



## APPENDIX B

### Subcriticality Evaluation for Chem-Nuclear Systems, Inc., Trench 23

#### Introduction

A criticality safety review of the Barnwell, S.C., Chem-Nuclear Systems, Inc. (CNSI) Trench 23 disposal information (Autry, 1998) was performed to evaluate the subcriticality of the burial trench as compared with recent nuclear criticality safety studies (Toran et al., 1997). The information summarized burial information for Trench 23 (100 feet wide, by 20 feet deep, by approximately 992 feet long) for the months of September 1977 through April 1978. The monthly "Burial Activity Report(s)" included:

<u>Information</u>	<u>Abbreviation used in this evaluation</u>
Byproduct Material, millicuries <sup>3,4</sup>	BPM
Source Material, pounds <sup>5</sup>	SM(lbs.)
Special Nuclear Material, grams	SNM(g)
Total Volume Buried, cubic feet	Tot Vol.(ft <sup>3</sup> )
# SNM packages	#SNM Pkgs.
Location of materials in trench by	
I SNM Shipments (i.e., no. shipment groupings)	#SNM Grps
II Source Material Shipments (i.e., no. shipment groupings)	#SM Grps
Burial position (ft) of Shipments along the length of Trench 23 from	
beginning of month to	Start ft
end of month	End ft

No information was provided regarding specific individual burial volumes of SNM, Source Material, or Byproduct Material. Additionally, no information was provided regarding the mass or placement of Byproduct and matrix Material within the trench. It was necessary to apply some assumptions to evaluate the subcriticality of Trench 23. Those evaluation assumptions and their effects on the evaluation are provided below.

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<sup>3</sup>Title 10 of the U.S. Code of Federal Regulations Part 30, §30.4 Definitions, "Byproduct Material means any radioactive material (except special nuclear material) yielded in or made radioactive by exposure to the radiation incident to the process of producing or utilizing special nuclear material," December 31, 1997.

<sup>4</sup>Title 10 of the U.S. Code of Federal Regulations Part 40, §40.4 Definitions, "Byproduct Material means the tailings or wastes produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content, including discrete surface wastes resulting from uranium solution extraction processes," December 31, 1997.

<sup>5</sup>Title 10 of the U.S. Code of Federal Regulations Part 70, §70.4 Definitions, "Source Material means: (1) Uranium or thorium, or any combination thereof, in any physical or chemical form, or (2) ores which contain by weight one-twentieth of one percent (0.05%) or more or: (1) Uranium, (ii) thorium, or (iii) any combination thereof," December 31, 1997.

## Assumptions

In order to perform the comparative evaluation of Trench 23, relative to information provided in NUREG-6505, Vol. 1 or the Nuclear Criticality Safety Guide (Pruvost and Paxton, 1996), it was necessary to assume that the containerized waste matrix, contaminated with SNM, was either a hydrogenous material like plastic, water, wood, and paper or was a relatively ineffective neutron-absorbing material like SiO<sub>2</sub>. Other assumptions, and their effects on the subcriticality evaluations, are provided in Table B.1 below.

## Burial Information

The Trench 23 information used for the comparative evaluation was derived from Autry, 1998, and is summarized in Table B.2. Footnotes to the table provide explanations as to how the reference data were used to derive the values used in the comparative evaluation. The derived values were then compared with information published in Toran et al., 1997 (Vol. 1) and Hopper et al., 1995 to demonstrate subcriticality of Trench 23. The primary values of interest for the comparative evaluation were g <sup>235</sup>U/cm<sup>3</sup> (or g SNM/cm<sup>3</sup>) and kg <sup>235</sup>U/m<sup>2</sup> (or kg SNM/m<sup>2</sup>).

## Results of Comparative Evaluation

Information from Table C-2 SiO<sub>2</sub>-soil (S-S) results provided in Toran et al., 1997 (Vol. 1); and guidance from Hopper et al., 1995, was compared with information extracted from Table B.2 above (highlighted cells) to determine the magnitude of SNM density increase (i.e., concentration factor) to approach a criticality concern. The following information was extracted from Table B.2 for the comparative evaluation:

- maximum density is  $2.6 \times 10^{-5}$  g SNM/cm<sup>3</sup>,
- maximum “infinite” slab areal density is  $5.0 \times 10^{-2}$  kg SNM/m<sup>2</sup>,
- maximum single package SNM mass is less than 350 g <sup>235</sup>U (by license) but calculated to be less than 45 g SNM/Pkg,
- minimum package volume calculated is 60.6 ft<sup>3</sup>,
- maximum single “SNM Shipments” burial is 52369.5 SNM(g), and
- total SNM mass in Trench 23 is 174.93 kg SNM.

The information presented in Table B.3 below provides comparisons between the maximum extracted values from Table B.2 above and information from Toran et al., 1997 (Table C-2) and Hopper et al., 1995 (Table 1). The concentration factor necessary to alter the Trench 23 values to the reference values is also provided.

As determined from information in Table B.2, the ratio of #SNM Grps to #SM Grps is about 1.8 to 1. However, the Eff wt % of <sup>235</sup>U is about 0.08%. Given the reported maximum calculated value of 45 g <sup>235</sup>U per package (typically larger than 4 ft × 4 ft × 4 ft), significant concentration of <sup>235</sup>U (via hydrogeochemical processes after breach of package walls) will necessarily involve the very large masses of SM (nearly 1200 g SM per g SNM) in the trench. The concentration and migration of SNM and SM will be effected through repeated dissolution and reduction of the uranium, thereby significantly reducing the effective enrichment of the uranium.

Table B.1 Subcriticality evaluation assumptions and ramifications

Subcriticality evaluation assumption	Ramification
(1) The specified mass of SNM is assumed to be grams of $^{235}\text{U}$ without $^{238}\text{U}$ .	Likely an excessively conservative assumption since most NRC fuel cycle facility waste uranium enrichments are on the order of less than 5 wt % $^{235}\text{U}$ ; however, many burials in Trench 23 are reported to be in excess of 90 wt % $^{235}\text{U}$ . (conservative – actual enrichment is typically less than 90 wt % $^{235}\text{U}$ )
(2) The volume of byproduct material was ignored as being inconsequential.	Based upon information for Trenches 38 and later (after October 1981) (House, 1996) the “Total Fuel Cycle Volume Percent” was commonly 78 – 98% of the total burial volume with the balance burial volume being “Non Fuel Cycle” (Byproduct Material). (competing conservative/ nonconservative – ignoring volume reduces density of $^{235}\text{U}$ , thereby reducing reactivity of $^{235}\text{U}$ but including the volume increases density of $^{235}\text{U}$ but provides neutron absorption)
(3) The number of SNM packages per “SNM Shipments” groups (Grps) placement on the trench plat map is equivalent to the number of SM packages per “Source Material Shipments” Grps placements on the trench plat map. This permits the determination of SM packages and trench volume occupied by the SM packages.	Because of the US NRC license limitation of 350 g $^{235}\text{U}$ per package during this 1977 – 1978 period and because of the essentially unrestricted mass of $^{238}\text{U}$ or thorium as Source Material (SM) per package, it is likely that the actual volume per gram of SM would be less than the actual volume per gram of SNM thereby providing greater volumes for the SNM, thereby reducing the determined SNM densities in the assumptions. (conservative in that the volume per gram of SM is likely smaller per “Source Material Shipments”)
(4) One evaluation assumed that all SNM Grps were placed together at the determined average depth of waste for the trench.	The CNSI burial records indicate that the SNM Shipments Grps were generally intermingled with Source Material Shipments Grps, thereby reducing the density of $^{235}\text{U}$ and providing neutron absorption with $^{238}\text{U}$ . (conservative in that the SNM was not dispersed with SM)
(5) One evaluation assumed that all SNM Grps and all SM Grps were intermingled at the determined average depth of waste for the trench.	The CNSI burial records indicate that the SNM Shipments Grps were generally intermingled with Source Material Shipments Grps, thereby reducing the density of $^{235}\text{U}$ and providing neutron absorption with $^{238}\text{U}$ . (competing conservative/ nonconservative – providing the diluting volume of SM Grps is nonconservative; however, the inclusion of the SM does provide unaccounted-for neutron absorption)
(6) The average burial thickness of about 10.2 ft was assumed for the determination of kg $^{235}\text{U}/\text{m}^2$ .	The CNSI burial records only show volume of burial and approximate length of trench used for a specific burial month. (competing conservative/ nonconservative in that some burials will be more deep or more shallow)

Table B.2 Raw and transformed data from Autry, 1998.

Month	Sep-77	Oct-77	Nov-77	Dec-77	Jan-78	Feb-78	Mar-78	Apr-78	Sums/Averages
SM(lb)	14.0	79807.2	9746.5	18716.6	56251.4	196346.9	34121.3	67625.1	462628.9
SNM(g)	1102.0	19646.7	29131.4	28986.4	27127.8	<b>52369.5</b>	7228.7	9337.7	<b>174930.2</b>
Eff wt % (1)	14.779%	0.054%	0.653%	0.340%	0.106%	0.059%	0.047%	0.030%	0.083%
Tot Vol. (ft <sup>3</sup> )	17101.5	125224.8	180667.7	159599.2	110198.8	129574.8	104820.7	158087.7	985275.1
#SNM Pkgs.	64	1273	1026	1339	1153	1176	366	886	7283
g SNM/Pkg.	17.2	15.4	28.4	21.6	23.5	<b>44.5</b>	19.8	10.5	24.0
#SNM Grps	3	29	49	53	36	33	20	23	246
#SM Grps	3	16	17	19	20	27	11	18	131
#SM Pkgs. (2)	64	702	356	480	641	962	201	693	4100
#Pkgs. (3)	128	1975	1382	1819	1794	2138	567	1579	11383
ft <sup>3</sup> /Pkg. (4)	133.6	63.4	130.7	87.7	61.4	<b>60.6</b>	184.8	100.1	86.6
g SNM/cm <sup>3</sup> (5)	4.6E-06	8.6E-06	7.7E-06	8.7E-06	1.4E-05	<b>2.6E-05</b>	3.8E-06	3.7E-06	9.8E-06
g SM/cm <sup>3</sup> (6)	2.6E-05	2.9E-02	3.4E-03	7.1E-03	2.3E-02	5.4E-02	1.5E-02	1.6E-02	2.1E-02
g SNM/cm <sup>3</sup> -T (7)	2.3E-06	5.5E-06	5.7E-06	6.4E-06	8.7E-06	1.4E-05	2.4E-06	2.1E-06	6.3E-06
g SM/cm <sup>3</sup> -T (8)	1.3E-05	1.0E-02	8.7E-04	1.9E-03	8.2E-03	2.4E-02	5.2E-03	6.9E-03	7.5E-03
Start ft (9)	0	0	105	280	300	550	756	750	0.0
End ft (10)	30	105	280	456	525	754	850	964	964
Depth ft (11)	5.7	11.9	10.3	9.1	4.9	6.4	11.2	7.4	10.2
kg SNM/m <sup>2</sup> (12)	7.9E-03	3.1E-02	2.4E-02	2.4E-02	2.0E-02	<b>5.0E-02</b>	1.3E-02	8.4E-03	3.1E-02
kg SNM/m <sup>2</sup> (13)	3.7E-04	1.9E-03	1.7E-03	1.6E-03	1.2E-03	2.6E-03	7.7E-04	4.4E-04	1.8E-03

## Notes:

- (1) One hundred times the mass of SNM divided by the sum of the SNM plus SM masses in grams.
- (2) The #SM Grps times the #SNM Pkgs. divided by the #SNM Grps assuming equivalent number of packages per group irrespective of SNM or SM.
- (3) The sum of #SNM Pkgs. plus #SM Pkgs.
- (4) The Total Volume Buried, cubic feet, divided by the #Pkgs.
- (5) The mass of SNM divided by the product of #SNM Pkgs. times ft<sup>3</sup>/Pkg.
- (6) The mass of SM divided by the product of #SM Pkgs. times ft<sup>3</sup>/Pkg.
- (7) The mass of SNM divided by the trench volume (i.e., the trench width times the burial depth times the difference of the End minus the Start), expressed in g SNM/cm<sup>3</sup>.
- (8) The mass of SM divided by the trench volume (i.e., the trench width times the burial depth times the difference of the End minus the Start), expressed in g SM/cm<sup>3</sup>.
- (9) Starting position within the trench for a burial, in feet.
- (10) Ending position within the trench for a burial, in feet.
- (11) Effective depth of a burial determined from Total Volume Buried divided by the product of the trench width (100 ft) times the difference of the Start minus the End of the burial, expressed in feet of depth.
- (12) The mass of SNM divided by the volume of SNM Pkgs. (i.e., #SNM Pkgs. times ft<sup>3</sup>/Pkg) times the burial Depth, expressed in kg <sup>235</sup>U/m<sup>2</sup>.
- (13) The mass of SNM divided by the Total Volume Buried, expressed in kg <sup>235</sup>U/m<sup>2</sup>.

Table B.3 Concentration factors for criticality concern

	Subcritical reference values and concentration factors for Trench 23			
Information extracted from Table B.2 above (Trench 23 study)	Table C-2 (Toran et al, 1997)	Concentration factor	Table B.1 (Hopper et al., 1995)	Concentration factor
$2.6 \times 10^{-5}$ g SNM/cm <sup>3</sup>	$\sim 1.4 \times 10^{-3}$ g <sup>235</sup> U/cm <sup>3</sup>	~54	$1.16 \times 10^{-2}$ g <sup>235</sup> U/cm <sup>3</sup>	446
$5.0 \times 10^{-2}$ kg SNM/m <sup>2</sup>	$3.1 \times 10^0$ kg <sup>235</sup> U/m <sup>2</sup>	62	$4.0 \times 10^0$ kg <sup>235</sup> U/m <sup>2</sup>	80
45* g SNM/Pkg (unit)	$2.02 \times 10^3$ g <sup>235</sup> U	44	$7.6 \times 10^2$ g <sup>235</sup> U	17.3
* 350 g SNM/Pkg (unit) by License	$2.02 \times 10^3$ g <sup>235</sup> U	5.7	$7.6 \times 10^2$ g <sup>235</sup> U	2.2

## Hydrogeochemical Potential for Uranium Concentration

The results of this study for determining the potential for criticality following the disposal of uranium at low-level-waste facilities as containerized waste provide information regarding the cumulative uraninite precipitation for long time frames in an environment consistent with the CNSI Barnwell, S.C. site (see Figure 7.1). The results, as shown in Figure 7.1, show that increasing cumulative uraninite precipitation from about 0.002 g/cm<sup>3</sup> to about 0.02 g/cm<sup>3</sup> (a tenfold increase) would require about 7000 years. It is judged that, though somewhat preferential, the concentration of the SNM within Trench 23 will become blended with the SM by the same mechanisms and in the same uranium proportions placed in the trench.

## Conclusions

We conclude that the areal density of the buried SNM (disregarding the commingling of SM) in Trench 23 is sufficiently small (i.e., one-eightieth or one-sixty-second of the areal density of concern for criticality) that criticality cannot be achieved as placed in the trench. Even though SNM concentration factors of potential concern could develop over approximately 7,000 years, the same hydrogeochemical mechanisms that could cause vertical and horizontal migration of SNM will also migrate SM, thereby further reducing the potential for criticality by reducing the effective <sup>235</sup>U enrichment in the blended materials to well below 1 wt % <sup>235</sup>U (~0.08 wt % <sup>235</sup>U).



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<b>11. ABSTRACT</b> <i>(200 words or less)</i>  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 and test some reasonable 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 possible scenarios for uranium migration and concentration increase at LLW disposal facilities, (2) model groundwater transport and subsequent concentration increase via 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 <sup>235</sup> U in the present scope of work. The work documented in this report indicates that the potential for a criticality safety concern to arise in an LLW facility is extremely remote, but not impossible. Theoretically, conditions that lead to a potential criticality safety concern might arise. However, study of the hydrogeochemical mechanisms, the associated time frames, and the factors required for an actual criticality event indicate that proper emplacement of the SNM at the site can eliminate practical concerns relative to the occurrence and possible consequences of a criticality event.						
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