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Consideration of Criticality when Directly Disposing Highly Enriched Spent Nuclear Fuel in Unsaturated Tuff: Bounding Estimates

Rob P. Rechard, Martin S. Tierney, Larry C. Sanchez, Mary-Alena Martell

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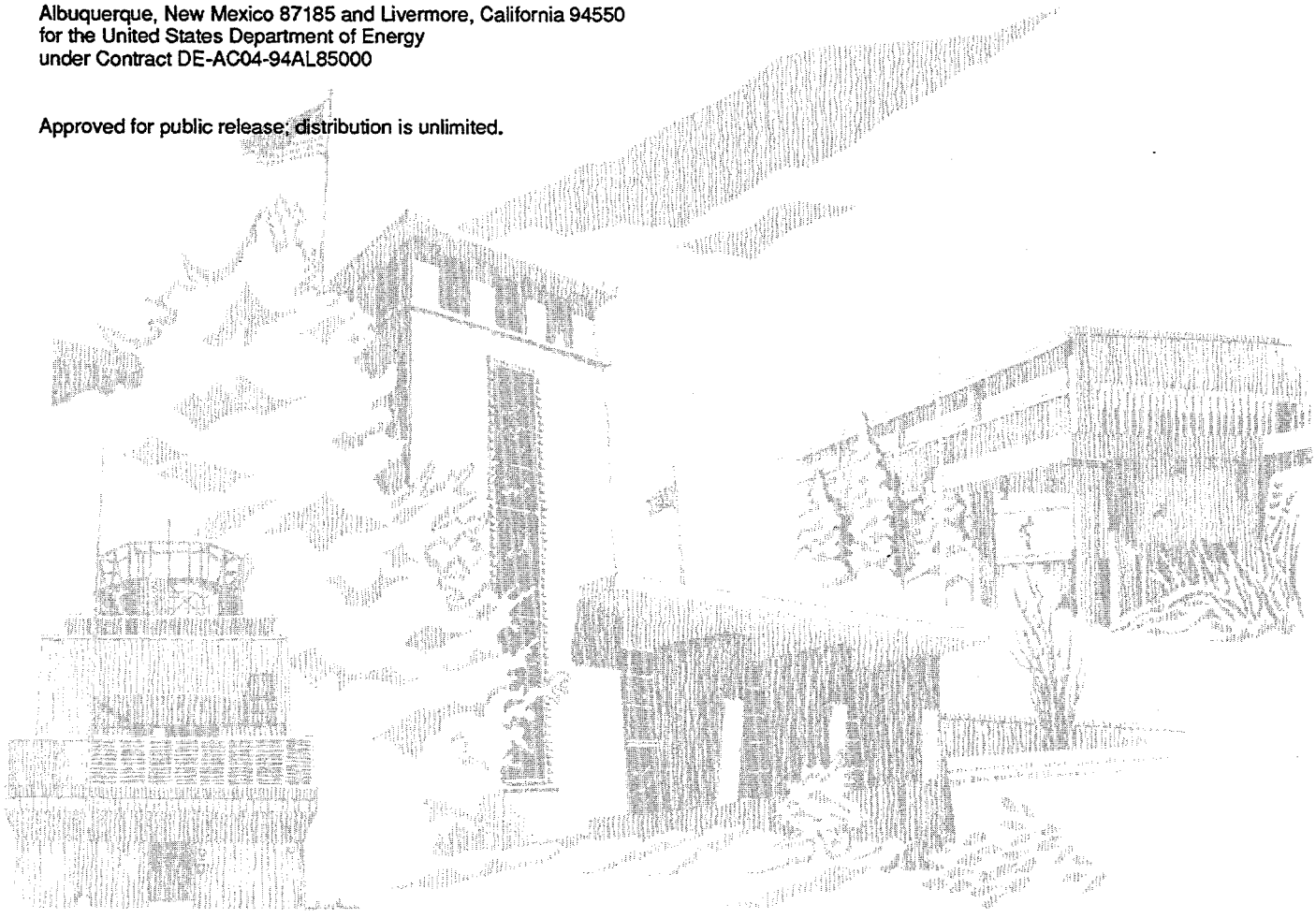

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Abstract

Although idealized calculations of the potential for an atomic explosion within a repository can make headlines, a more technically useful assessment is a systematic, multidisciplinary, integrated analysis that uses a set of consistent assumptions of disposal system performance. The analysis described here, called a performance assessment, employs the same general approach to study the potential of a critical mass assembly as has been used to examine other potentially disruptive scenarios in a nuclear waste disposal system. This report presents one of two approaches—bounding calculations—which were used in a major study in 1994 to examine the possibility of a criticality in a repository. The bounding probabilities in this study are rough and do not entirely dismiss the possibility of a critical condition; however, they do point to the difficulty of creating conditions under which a critical mass could be assembled (i.e., corrosion of containers, separation of neutron absorbers from the fissile material, and collapse or precipitation of the fissile material) and, more important, how significant the geochemical and hydrologic phenomena are in examining this criticality issue. Furthermore, the study could not conceive of a mechanism that was consistent with conditions under which an atomic explosion could occur, i.e., first, the manner in which fissile material could be collected and, then, how it would be assembled (or diffused outward) within microseconds. In addition, should a criticality occur in or near a container in the future, the bounding consequence calculations in this study showed that fissions from one critical event ($<\sim 10^{20}$ fissions, if similar to aqueous and metal accidents and experiments) are quite small compared to the amount of fissions represented by the spent nuclear fuel itself. Also, if it is assumed that the containers necessary to hold the highly enriched spent nuclear fuel in this study went critical once per day for 1 million years, creating an energy release of about 10^{20} fissions, the number of fissions equals about 10^{28}, which corresponds to only 1% of the fission inventory in a repository containing 70,000 metric tons of heavy metal (MTHM) (the expected size for the proposed repository at Yucca Mountain, Nevada).

Acknowledgments

This report is based in part on a study, commissioned by the Office of Environmental Management of the U.S. Department of Energy (DOE), to guide the spent fuel technology development program at the Idaho National Engineering Laboratory (INEL) and elsewhere that was begun a year before the controversy concerning the Bowman-Venneri calculations. Because the original report is about 1400 pages, this report was written to make the criticality aspects of the study (and the bounding estimates in particular) more accessible. In addition, a short summary of the criticality aspects of this study was presented at the International Conference on Criticality Safety, Albuquerque, NM, September 17-21, 1995.

The authors wish to acknowledge the input of J.L. Powell of Sandia on fission product yields, the input of J.S. Rath and D. Morrison, of New Mexico Engineering Research Institute, Albuquerque, on scaling underground nuclear explosions, and the editorial support of J. Chapman, of Tech Reps, Inc., Albuquerque. The authors also wish to thank the Sandia reviewers, R.W. Bild and G.E. Barr, for their helpful suggestions to the text.

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1.0 INTRODUCTION

The primary purpose of this report is to present bounding calculations as one approach taken to date to explore the potential of a critical configuration developing in or near a container of highly enriched uranium spent nuclear fuel disposed without treatment directly into an unsaturated volcanic tuff repository. The highly enriched uranium fuel comes from special reactors owned by the U.S. government. Although the initial configuration of the spent fuel will change over time, whether a critical configuration can or will develop is uncertain. Since the publication of articles in *The New York Times* about Bowman and Venneri's belief that a potential exists for an atomic explosion in a nuclear waste repository (Bowman and Venneri, 1996; Broad, 1995; Taubes, 1995), several commentators have criticized the Bowman and Venneri calculations and called for a "... probabilistic risk assessment of the hypothesis" (Geppert, 1995), or stated that a "risk-based analysis of possible scenarios and neutron transport ... is needed" (Van Konynenburg, 1996) or that a "... systematic and multi-disciplinary approach is recommended ... based on risk and decision theories" (Paperiello, 1995). A risk analysis has as its hallmark a balanced, systematic, multidisciplinary, integrated analysis of a technological system to evaluate the potential and consequences of unwanted outcomes. This type of analysis takes into account actual features, events, and processes of the system rather than relying on simplistic calculations of conjectured initial conditions. The remainder of the introduction discusses the risk analysis methodology, previous criticality work, and disposal options for the highly enriched fuel considered herein.

1.1 Performance Assessment

The overall risk analysis process for a nuclear waste disposal system is usually called a "performance assessment" (PA). In the United States, a performance assessment is intimately tied to the general scientific process of stochastic modeling. The analysis differs in practical details, but its theory is identical to the probabilistic risk assessment (PRA) performed for a nuclear reactor, an analysis technique that was developed concurrently with the performance assessment (Rechard, 1995a). The use of stochastic modeling to incorporate uncertainty in outcomes has, until about 1990, set PAs and PRAs apart from the risk assessment traditionally performed to evaluate chemical toxicity and carcinogenicity. Furthermore, PAs and PRAs have also used more formal methods to construct scenarios—an example of which is shown in this report.

Quantifiable measures from a performance assessment (like a PRA) are derived from answers to three questions: What unwanted conditions may occur? What is the probability of each occurrence? What are the consequences of each occurrence (Kaplan and Garrick, 1981; Helton et al., 1993)? In a performance assessment, the answer to the first question is generated by characterizing the system and then identifying various scenarios. The scenario discussed in this report is the assembly of a mass of fissile material in or near a waste container after disposal in a geologic repository such that a critical condition occurs.[1] The second question is answered by estimating probabilities based on empirical data or modeling the physical processes necessary to assemble a critical mass in various configurations. An important aspect of this step is to establish probable initial conditions. For the criticality issue, the answer to the third question is provided from estimates of consequences based on empirical data.

1.2 Previous Work

Because of the general importance of considering criticality scenarios in a risk-based analysis of any nuclear facility, consideration of criticality scenarios at a nuclear disposal site is not new. For example, the Waste Isolation Pilot Plant (WIPP) Project, which is studying the effects of disposal of waste contaminated with transuranic radionuclides, has included criticality in its scenarios since 1979 (Bingham and Barr, 1980; 1979), and the international community has included it since 1981 in its generic list of events to consider (IAEA, 1981). Carter (1973) evaluated the potential for criticality as part of a safety analysis of nuclear waste disposal in trenches at the Hanford site. In 1979, Clayton (1979) suggested a positive feedback mechanism for a critical

1 In nuclear engineering, a critical condition or event (commonly called "a criticality") denotes the assembly of sufficient fissile mass such that a threshold is reached whereby, over the long term, sufficient neutrons are generated from fissions to create a self-sustaining nuclear chain reaction and readily generate heat energy. However, the meaning of the word "critical" to people outside the nuclear community is most often associated with the way it is used in the medical field, that is, to denote a crisis (actually the threshold of a crisis). As described further in Section 5.1.6, for a nuclear criticality to become a "crisis" (i.e., create an atomic explosion) the system must pass through the critical state extremely fast and reach a prompt supercritical state whereby the number of prompt neutrons (not including delayed neutrons) increases faster than the fissile material can expand from the energy release, thereby allowing massive numbers of chain carrying neutrons to be produced. Because the intended audience of this report is outside the nuclear community, the phrase "sustained nuclear chain reaction" is occasionally used to counteract the understandable but mistaken tendency to associate a nuclear criticality with a grave crisis.

event in the trenches at the Hanford site based on the calculations by Carter (1973). In 1983, Stratton (1983) dismissed the allegations that a nuclear explosion had occurred at a waste disposal site in the Ural mountains in Russia. Finally, recent comments on the Bowman calculations have provided qualitative arguments about the difficulty of achieving a critical condition in a repository and the apparent impossibility of an atomic explosion (Van Konynenburg, 1996; Canavan et al., 1995). The study described in this report differs from these others by using the risk analysis framework to provide a quantitative evaluation of the potential for a criticality scenario including, especially, the chemical and physical phenomena necessary to alter the waste container and the fuel. Although many of the qualitative arguments presented by van Konynenburg (1996) and Canavan et al. (1995) could be placed within this risk framework, this has not been done here.

1.3 Disposition Options for Waste Containing Fissile Material (Highly Enriched Spent Nuclear Fuel)

The amount of fissile material (i.e., primarily ^{233}U, ^{235}U, and ^{239}Pu, which are fissioned by slow [thermal] neutrons) in nuclear waste varies from practically none for reprocessed fuel to ~2% for commercial spent nuclear fuel to ~90% for weapons-grade plutonium and some highly enriched uranium spent nuclear fuel before irradiation. Therefore, the disposition options and implications of developing a criticality vary considerably. This report pertains to the highly enriched uranium spent nuclear fuel category; however, some of the results qualitatively apply to other waste categories.

Several disposition options exist for the highly enriched spent nuclear fuel, each with a different potential for developing a criticality condition. This report examines direct disposal without treatment. However, prudent design measures are included to preclude a criticality immediately after disposal, such as including boron in stainless steel to absorb neutrons that otherwise could contribute to a nuclear chain reaction and mixing of containers such that containers with highly enriched uranium spent fuel are not placed next to each other. (Previous calculations [Rechard, 1995b] also showed that the mass of the containers was such that if two containers of highly enriched uranium were placed end-to-end, the neutron population in one container would not significantly contribute to the neutron population normally present in the other container.) In addition, several design features are included that moderately reduce the probability of a critical condition over the long term: (a) containers with an average long design life are used, (b) containers are spaced close enough so that the heat generated by the decay of radioisotopes heats the rock such that sufficient vaporization occurs in order to keep the containers dry for several hundred years, and (c) the repository is located in an unsaturated zone of volcanic tuff similar to that proposed for commercial, low enriched spent nuclear fuel at Yucca Mountain, Nevada.

Because decisions on construction and packaging of the containers for highly enriched fuel are not final, many options for preventing a criticality with moderate or high assurance are available for consideration. Other moderate assurance measures that have been considered, but are not discussed further here, include (1) using neutron poisons with similar geochemical behavior to that of the geologic setting of the repository, (2) limiting fissile mass in each individual container, (3) using a salt repository, and (4) use of naturally occurring phosphate minerals with adsorbed rare earth metals. Rechard (1993) reports on the impact to the disposal system of the moderate assurance measure of limiting fissile mass in each container and the use of a salt repository. Within the salt, the likelihood of a criticality is practically zero; hence, a criticality can occur only outside the salt mass which, as shown in Rechard (1993) and elsewhere (Helton et al., 1993; WIPP PA Department, 1992), requires a human intrusion. The fourth option, using phosphates mixed with rare earth metals (i.e., naturally occurring apatite) outside the container to greatly reduce the mobility of actinides and, simultaneously, to prevent a criticality, has been only proposed.

High assurance measures for preventing a critical condition that have been or will be considered, but are not discussed further here, include (1) surface storage of the spent fuel assemblies; (2) reprocessing and reuse of the fissile material in a nuclear reactor, or transmutation in a neutron flux generated by either a nuclear reactor or particle accelerator; and (3) mixing of the spent fuel with depleted uranium. While the first option certainly precludes a criticality in the geologic repository, it requires active institutional control on the surface to prevent accidental and terrorist-caused criticalities and dispersal of the fissile material. Rechard (1993) reports on the uses of the high assurance measures of removing all fissile material from the spent fuel and then using the fissile material elsewhere, or mixing the fuel with depleted or natural uranium. As discussed by Rechard (1993), a negative impact of the latter option is the increased number of containers required, but a positive benefit (besides providing a greater assurance of preventing a criticality) is the disposal of stockpiles of depleted uranium.

In summary, several options exist for providing moderate to high assurance of preventing a criticality during disposal of large masses of fissile material. In this report, one technique, borated stainless steel, for preventing a criticality in the short term (i.e., storage, transportation, and operational phase of the repository) is considered. If other engineered features are eventually adopted to prevent a criticality over the long term, the arguments presented here will continue to be useful for conveying the level of assurance that the disposal system can provide.

1.4 Analysis Overview

In the following description of the analysis, possible regulatory criteria regarding a criticality are first mentioned briefly, and then the discussion is organized around answers to the three questions of a performance assessment: (1) system characterization and scenarios, (2) probability and process of assembling the fissile material into a configuration that promotes a nuclear chain reaction, and (3) consequences of the continued or cyclic operation of a nuclear chain reaction within the fissile mass assembly. Although neither the assembly process nor its consequences could be explored completely within the time available for the study on which this report is based (Rechard, 1995b), both aspects were investigated because of their interrelationship. A useful analysis requires that consistency be maintained between these two aspects (i.e., the processes that assemble the fissile mass establish the initial conditions under which a nuclear reaction would operate). Furthermore, because current regulations lack specific guidance on criticality issues over the long term, the assembly and operation of the fissile mass (items 2 and 3 above) were studied here through bounding calculations. The bounding calculations on the probability and consequences of a critical condition in a geologic repository are presented in this report in order to provide alternatives for (1) rational policy decisions concerning criticality and (2) procedures for examining the possibility of a criticality in a repository.

2.0 REGULATORY SETTING

Although a risk-based analysis is not limited to developing metrics for comparison with regulatory criteria, the regulations do establish important criteria and an accepted modeling style. Several laws and regulations affect the disposal of radioactive waste in geologic repositories.

2.1 Policy Laws

Three laws of particular importance with regard to defining policy are the *Nuclear Waste Policy Act of 1982* (NWPA, 1983), subsequent amendments to this act in 1987 (NWPAA, 1987), and the *Energy Policy Act of 1992* (Energy Policy Act, 1992). Although the NWPA of 1982 and the NWPAA of 1987 do not establish performance criteria for a repository, they do establish the policy that the current generation is obliged to bear the political and financial costs of developing a permanent disposal option. A specific requirement of this policy in NWPA of 1982 is that U.S. Department of Energy (DOE) defense and experimental waste, with the exception of transuranic defense waste, should be disposed of with commercial nuclear waste and thus must comply with all requirements of the Nuclear Regulatory Commission (NRC).

In 1985, in accordance with suggestions in the *Nuclear Waste Policy Act of 1982* and after study of the issue, President Reagan concurred that the DOE high-level waste (waste from reprocessing spent fuel) be placed in the same repository as commercial spent nuclear fuel. Disposal options for the direct disposal of DOE spent nuclear fuel were not explicitly studied for some time because it was implicitly assumed that the fuel would be reprocessed to extract the highly enriched uranium (^{235}U) or fissile plutonium (^{239}Pu). However, in 1992, the U.S. government established a policy to end all reprocessing of DOE-owned spent nuclear fuel except in special circumstances. Hence, disposal of the spent nuclear fuel without reprocessing might be necessary, and therefore a need existed to study the impact on the repository of its disposal. One such impact was the criticality issue, which is reported here.

The *Energy Policy Act of 1992* set a new policy that is expected to generate substantial changes in the regulatory setting. Generally, the act requires that the Environmental Protection Agency (EPA) seek advice from the National Academy of Sciences (NAS) to promulgate a site-specific standard for the proposed nuclear waste repository at Yucca Mountain, Nevada, and a revision of the NRC regulation to agree with the new EPA standard. The NAS issued its recommendations (NAS, 1995) after this study was completed; three suggestions that will affect future studies are that (1) the maximum annual effective dose equivalents be used as the criteria for protecting public health, (2) the maximum annual effective dose equivalent be determined over a million year period, and (3) the probability of an inadvertent human intrusion event *not* be formally estimated, instead only potential consequences of a few selected situations

would be evaluated. The influence of these recommendations on the present study are qualitatively noted.

2.2 Regulations

As noted above, the regulations for geologic disposal are in flux. However, current regulations are discussed here briefly because they represent the only guidance at present.

2.2.1 40 CFR 191

In 1985, in response to NWPA of 1982, the EPA promulgated the standard, *40 CFR Part 191—Environmental Standards for the Management and Disposal of Spent Nuclear Fuel, High-Level and Transuranic Radioactive Wastes; Final Rule* (EPA, 1985), which had been under study since 1977. The regulation was remanded shortly thereafter but repromulgated with only minor changes in 1993 (EPA, 1993). The EPA standard, 40 CFR 191, establishes criteria for the disposal system as a whole. One criterion in 40 CFR 191, the Containment Requirements, requires an analysis to evaluate probabilities of integrated release at the disposal system boundary for 10,000 yr and compare results against the probabilistic criterion in these requirements.

2.2.2 10 CFR 60

The NRC is responsible for ensuring that a disposal system for commercially generated spent fuel meets the requirements of EPA's 40 CFR 191. Prior to promulgation of 40 CFR 191, but cognizant of its likely contents, the NRC promulgated in 1983 the technical criteria to be used in evaluating a geologic repository and thus implementing 40 CFR 191 as "Disposal of High-Level Radioactive Wastes in Geologic Repositories," *Code of Federal Regulations 10, Part 60.* The technical criteria established minimum requirements for subsystems of the disposal system (thereby treating the disposal system like a nuclear reactor). The requirements are stringent: a 300- to 1000-yr period for substantially complete containment of radionuclides by the waste container; and a maximum engineered barrier fractional release rate of 10^{-5}/yr of the 1000-yr inventory for any radionuclide released at a rate greater than 0.1% of the calculated total release rate of all radionuclides. In addition, 10 CFR 60 flatly prohibits explosive, pyrophoric, and chemically reactive material and free liquids and also requires solidification of all radioactive waste, consolidation of particulates, and reduction of combustible radioactive waste forms to noncombustible forms. Provided these criteria on subsystems remain in the revised 10 CFR 60, they might have been used to form the basis for setting initial conditions for a criticality analysis. Because of anticipated changes to 10 CFR 60, however, herein they are used only as a means of comparing calculated performance in the original study (Rechard, 1995b).

2.2.3 Criticality

40 CFR 191 lacks specific guidance regarding the occurrence of a criticality after closure of the repository. The only mention of a criticality is in 10 CFR Part 60, Section 60.131, "General design criteria for the geologic repository *operations area*" (emphasis added), where geologic repository operations area means "a high-level radioactive waste facility that is part of a geologic repository, including both surface and subsurface areas, where waste *handling* activities are conducted" (emphasis added). For the operational phase of the repository, paragraph (b)(7) of this section adopts standard criticality control similar to nuclear power plants, stating[2] "all systems for processing, transporting, handling, storage, retrieval, emplacement, and isolation of radioactive waste shall be designed to ensure that a nuclear criticality accident is not possible unless at least two unlikely, independent, and concurrent or sequential changes have occurred in the conditions essential to nuclear criticality safety ... ."

Although the current regulations lack either specific guidance or provide guidance that would be difficult to apply to the post-closure phase regarding the acceptability of a self-sustaining nuclear chain reaction, the risks associated with a critical condition still must be evaluated under the general provisions of the regulations.

3.0 TYPE OF NUCLEAR WASTE AND DISPOSAL SYSTEM STUDIED

It is beyond the scope of this report to provide a complete description of the system. Only a few parameters are mentioned in this report to provide the reader a measure of understanding of some facets of the disposal system and the bounding arguments presented.

2 Although this section of 10 CFR 60 is describing the operational phase of the repository, the word "isolation" is used. As defined by 10 CFR 60, "isolation" means "inhibiting the transport of radioactive material so that amounts and concentrations of this material entering the accessible environment will be kept within prescribed limits." Because of this ambiguity and the potential difficulty of applying standard criticality control to the post-closure phase of an underground repository, the authors anticipate that when 10 CFR 60 is modified in the future the NRC will clarify either the type of criticality control or the criticality calculations necessary for the post-closure evaluation.

The nuclear waste considered in this study is spent nuclear fuel and high-level waste owned by the DOE. Of primary interest in this study is the potential for the highly enriched spent nuclear fuel to assemble into a critical mass. Although various disposal options are possible, the option studied here is direct disposal in an unsaturated volcanic tuff repository similar to the proposed repository at Yucca Mountain, Nevada, as mentioned earlier.

3.1 Highly Enriched Uranium Spent Nuclear Fuel

Although the majority of spent fuel by volume and mass is produced commercially by nuclear power plants, the DOE has about 1400 m^3 or 2700 metric tonnes of uranium (heavy metal) (MTHM) of spent fuel for disposal (DOE, 1994). Of this spent fuel, about 210 MTHM (8%) was originally highly enriched (i.e., the mass of fissile uranium isotope ^{235}U is >20%). The many types of highly enriched fuel were placed in three categories for this study (Table I): (1) graphite spent fuel (28 MTHM in 83 containers), (2) a chemically reactive highly enriched spent fuel (72 MTHM in 33 containers), and (3) a robust highly enriched spent fuel (110 MTHM in 256 containers).

The graphite fuel was represented by the Fort St. Vrain (Colorado) reactor fuel, consisting of uranium and thorium carbide fuel particles coated with silicon carbide in a graphite matrix. The inventory includes radionuclides from the Peach Bottom 1 (Pennsylvania) reactor fuel. Uranium-aluminum metal alloy advanced test reactor (ATR) fuel with aluminum cladding represented the chemically reactive spent fuel category, because ATR is a plurality among this type of fuel and also is one of the most chemically reactive highly enriched spent fuels. Some of the ATR fuel has been damaged from corrosion during storage in the water basin and thus may be treated before disposal by placement in stainless steel capsules to guard against creating conditions conducive to a rapid chemical oxidation of the uranium metal or a criticality during storage and transportation. The zircaloy-clad, uranium-dioxide Shippingport fuel represented the robust highly enriched fuel category. The inventory of the fuel from the Navy propulsion reactors is included in this category.

3.2 Other Nuclear Waste Included in Study

In addition to the 210 MTHM of highly enriched uranium spent fuel in 372 containers, the hypothetical repository under study included (1) 2262 MTHM of low-enriched pressurized water reactor (PWR) fuel and weapons-production fuel in 95 containers; (2) 320 MTHM (equivalents) of high-level calcine waste from reprocessing of spent nuclear fuel at Idaho National Engineering Laboratory (INEL) in 296 containers (original study used 2037 containers); and (3) 9268 MTHM (equivalents) of high-level sludge waste from reprocessing spent nuclear fuel within the DOE complex (Table I) in 1880 containers. Although this waste is important with regard to other aspects of the original study, for the purposes of this report, the addition of this waste can be viewed primarily as permitting an examination of the influence of highly enriched uranium fuel in a moderately sized repository.

3.3 Nuclear Waste Not Included in Study

Five categories of spent nuclear fuel were not included in this study. First, although up to 70,000 MTHM of commercial spent nuclear fuel would possibly be present in the repository (NWPA, 1983), it was not included in the original study because, if it were, the DOE highly enriched spent nuclear fuel would represent only about 0.3% of the mass and it would be difficult to assess its influence on repository performance in an initial study. Future studies can remove this type of conservatism if warranted. Second, sufficient information about foreign spent fuel was not available in time to include in this study, which was performed in fiscal year 1994. This DOE-owned spent fuel is similar to the ATR fuel but was used in foreign reactors and was shipped (prior to 1986) to Savannah River Plant, South Carolina, for storage (and, beginning this year, will again be shipped there). Third, high-level waste from the single-shelled tanks at the Hanford was not included, because the inventory was not available. Fourth, potential high-level nuclear waste from decontamination and destruction of DOE facilities was not included. Finally, the more than 50 MTHM of plutonium and hundreds of tons of highly enriched uranium that will become surplus as U.S. nuclear weapons are dismantled as the result of the Strategic Arms Reduction Treaties (START I and II) and unilateral pledges (NAS, 1994, p. 1) was not included. The weapons uranium is generally not contaminated with fission products (unlike the highly enriched uranium discussed in this report), and so it can be blended with depleted uranium to produce standard low-enriched fuel for commercial reactors. The more important question is the disposition of the excess ^{239}Pu. The NAS has recommended exploring three options for its disposal; all three ultimately involve geologic disposal, but two options involve direct disposal either by vitrification in borosilicate glass and then disposal in a commercial

Table I. Waste Types Included in 12,060-MTHM Hypothetical Repository in Unsaturated Volcanic Tuff (Rechard, 1995b, Table ES-2).

Waste Type	Specific Fuel Used to Represent Waste Type	Fuel Construction	MTHM*
Spent Nuclear Fuel			
1. Graphite	Fort St. Vrain, reactor in Colorado	Small particles of uranium and thorium carbide coated with silicon carbide in graphite matrix	28
2. Highly enriched uranium (HEU), i.e., fuel originally with over 20% by mass fissile ^{235}U isotope	Advanced Test Reactor (ATR)	Uranium-aluminum alloy fuel clad by aluminum**	72
3. Highly enriched uranium in robust assemblies	Shippingport fuel from Light Water Breeder Reactor research program, Shippingport, Pennsylvania; inventory from Naval propulsion reactors included	Uranium dioxide fuel wafers and clad by zircaloy	110
4. Low enriched uranium (LEU) spent fuel, i.e., fuel originally with less than 20% by mass fissile ^{235}U isotope	Commercial pressurized water reactor (PWR) fuel	Uranium-dioxide fuel pellets shaped into rods and clad by zircaloy	162
5. Weapons material fuel	N-Reactor fuel from Hanford, Washington; spent fuel from Savannah River not included	Uranium-metal fuel shaped into two concentric circles each clad with zircaloy	2100
High-Level Waste			
6. Calcine	Idaho Chemical Processing Plant at INEL	Calcination in a fluidized bed of liquid waste from reprocessing spent fuel to recover uranium; 3-18% powder, the remainder, granular	320***
7. Sludge Wastes	– Savannah River (Defense Waste Processing Facility at Savannah River Plant in South Carolina)	Sludge liquids containing radionuclides from reprocessing spent fuel to recover uranium and plutonium	8036***
	– Hanford (Hanford Waste Vitrification Plant in Washington; double-shelled tanks only)		1160***
	– West Valley (West Valley Demonstration Project in New York)		72***

* Another initialism, MTIHM (metric tons of initial heavy metal), is used by some authors who wish to emphasize that the measurement is the initial mass of heavy metal rather than the current mass of heavy metal. In this report, we use the designation, MTHM, to mean initial mass, not only because it is found more frequently in the literature, but also because 40 CFR 191 defines "heavy metal" as "... all uranium, plutonium, or thorium placed *into* a reactor..." (40 CFR 191.12; emphasis added). Thus, the use of MTIHM, while useful in calling attention to this fact, is not necessary.

** Much of the foreign research reactor spent fuel being returned to the U.S. for disposal is aluminum clad.

***The equivalent mass of uranium, plutonium, and thorium represented by the high-level waste was calculated for all high-level waste as a group, using the most conservative procedure described in 40 CFR 191.

repository or emplacement in very deep boreholes in granite without first consuming ^{239}Pu in a commercial reactor. Evaluating the criticality concerns of these options was beyond the scope of this report but could involve arguments similar to those reported here except that plutonium will be substituted for uranium. (The discussion presented here would apply directly if the containers, waste matrix, and lower solubility of ^{239}Pu delay release assembly until it decays to ^{235}U, as is considered likely at Yucca Mountain.)

3.4 Containers

For the arguments presented, the container for the highly enriched fuel was a multi-purpose canister of 304L stainless steel, 25.4 mm thick, 5.14 m long, and 1.77 m in diameter. This handling canister was then assumed to be overpacked with a 20-mm-thick corrosion-resistant material (Incoloy 825) and a 100-mm-thick corrosion-allowance material (i.e., ASTM Type A-516, Grade 60 carbon steel) (Fig. 1). To preclude criticalities even when voids in an intact container are fully filled with water, the container design was assumed to use borated stainless steel neutron absorber as the support structure for the spent nuclear fuel.

The corrosion rate of these protective materials in the container is a strong function of the concentration of oxygen, water saturation, and temperature. The simulation in the original study (Rechard, 1995b), accounts for this functional dependence; here, however, the distribution of rates at oxic conditions, 26°C (ambient) and 100°C, at 100% saturation, are presented for use in the bounding calculations (Fig. 2).

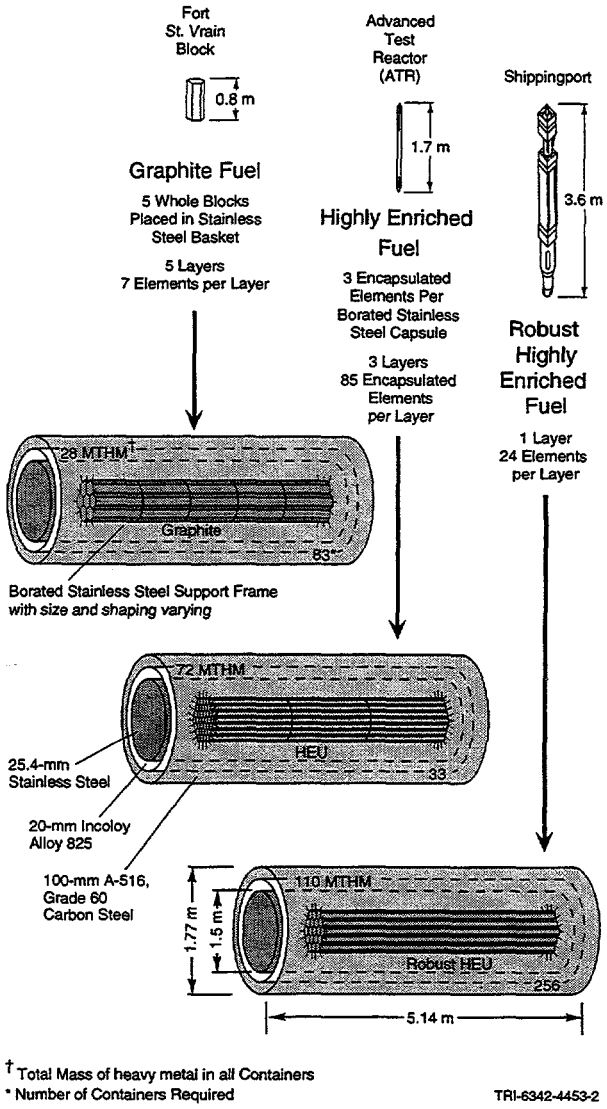

Fig. 1. Proposed packaging of the three categories of highly enriched uranium spent nuclear fuel in 25.4-mm-thick stainless steel multi-purpose canisters that are then overpacked with 20-mm-thick Incoloy 825 and 100-mm-thick carbon steel containers (after Rechard, 1995b, Figure ES-4).

3.5 Repository Layout and Waste Emplacement

The engineered barriers consist of the human-design elements that are intended to isolate the waste from the accessible environment. Two design components are the waste form itself and the containers (discussed above). Components of the engineered barriers that are specific to the repository include (1) the subsurface facility layout, (2) the design for waste emplacement, and (3) any specially prepared backfill (a subset of which is often called a "seal") in the drifts and ramps. Many precise details of the repository—such as the need for ramps or ventilation, and the size of the operational area—depend more on the operational phase of the repository than behavior after closure. Thus, this discussion highlights only the dimensions considered pertinent to post-closure behavior, such as the minimum spacing of containers and rooms.

The disposal region of the repository was sized to accommodate 12,060 MTHM of spent fuel or equivalent high-level waste. The mine design consists of long rooms ("tunnels"), 7.65 m in diameter, that are surrounded by and connected to access drifts, which are in turn connected to ramps that lead to the surface (Fig. 3).

The large waste containers are assumed to be transported to and within the repository by rail and then

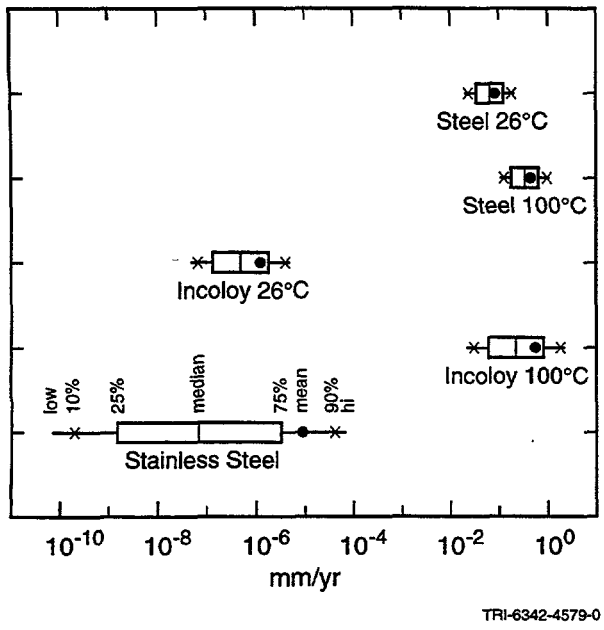

Fig. 2. Corrosion penetration rates of carbon steel, Incoloy 825, and stainless steel at 100% water saturation for oxic conditions (water in equilibrium with air with 21% oxygen) and a temperature of 26°C and 100°C. (Generalized data for carbon steel from Kirby, 1979, and Soo, 1984; localized corrosion data for Incoloy from Wilson et al., 1994, Table 13-5; generalized data for stainless steel from McCright et al., 1987.)

parked horizontally, end to end with no gap, in the tunnels for disposal. The tunnels were spaced 4.3 m apart (8.6 m between tunnel centers). This spacing resulted in an areal power density of 23.5 W/m^2 for the nuclear waste if it is placed in the year 2030. The containers are closely packed ("hot" repository concept) to ensure the absence of water in the majority of the repository for a number of years. Backfill was placed only in the drifts and ramps; no backfill was placed around the containers in the rooms. (For modeling [Rechard, 1995b], this void was added to the porosity of a numerical mesh element containing the waste parcel.) At emplacement, neutron interaction between containers does not occur because sufficient material exists at the ends of the container. Furthermore, in simulations (Rechard, 1995b), the highly enriched spent nuclear fuel containers were assumed to be uniformly mixed with the other nuclear waste included in the study and placed throughout the entire repository.

3.6 Unsaturated, Volcanic Tuff Geologic Barrier

The geologic barrier isolates the repository from the accessible environment. The general technical criterion for a geologic barrier is an ability to physically isolate radionuclides from the biosphere or, for regulatory purposes, the accessible environment; 40 CFR 191 defines the accessible environment as any location (including the surface and subsurface) that is 5 km from the repository. The geologic barrier that is discussed comprises a sequence of tuff (formed from welding of hot volcanic ash as it is deposited from a volcanic eruption) at Yucca Mountain, Nevada, located in a basin and range topographical province. General advantages of tuff as a geologic barrier are that it readily adsorbs many radionuclides and that groundwater becomes saturated with silica, which reduces the solubility of radionuclides. Advantages of this particular tuff are that it is located relatively near the surface in areas remote from human populations. Also, because the tuff deposit is located in a semi-arid climate, the repository can reside in the large unsaturated zone, which limits the amount of water that can degrade containers and transport radionuclides to the accessible environment. The advantage of limited water in the unsaturated zone may be offset somewhat by the potential of being an oxic environment, which can promote higher corrosion rates and radionuclide solubilities; however, as seen in preliminary simulations (to be discussed in a future report), the large amounts of iron in the container considered herein greatly reduce the amount of oxygen available for tens of thousands of years.

Frequently in simulations (e.g., Rechard, 1995b), the stratigraphy of the tuff disposal system is idealized as a series of constant thickness hydrologic modeling units ("pancakes") with a dip of 4.6°. The modeling units consist of consecutive layers of tuff with degrees of porosity that are similar, based on three wells near Yucca Mountain, USW GU-3/G-3, USW G-4, and UZ-16. Although the modeling units roughly correspond to the formal geologic stratigraphy, some units contain only a portion of a formation member or overlay several formation members. Herein, the repository is assumed to be placed 286 m below the surface and 333 m above an aquifer. The current saturation of the volcanic tuff is about 65% (Wilson et al., 1994) (Fig. 4). The average intact matrix porosity of the 333-m-thick tuff surrounding the repository (TS modeling units) is ~0.14 (Wilson et al., 1994, Table 7-3). The average matrix porosity of the entire column of tuff used in the study is ~0.19 (Fig. 4) (Rautman, 1995). Although not apparent from the coarse porosity data shown in Fig. 4, the average and

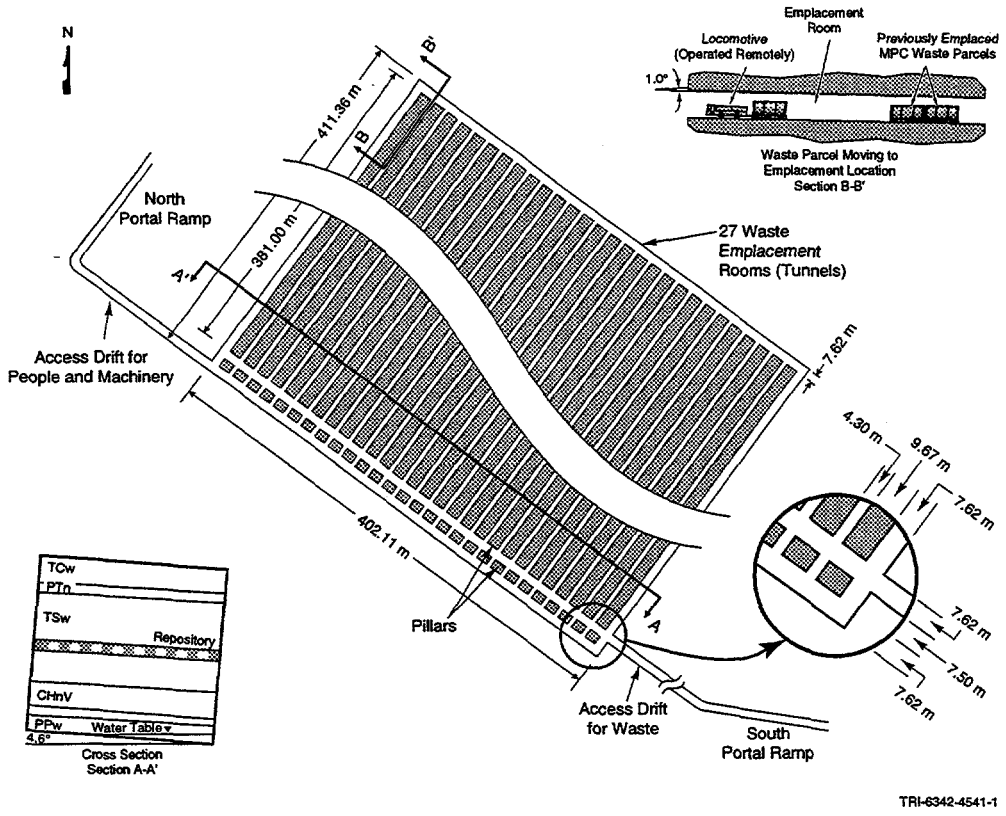

Fig. 3. Layout of waste disposal rooms and access drifts in volcanic tuff for hypothetical repository containing DOE spent fuel and high-level waste (after Rechard, 1995b, Figure ES-8). (Original study had disposal rooms rotated 90° from those used in this report, eliminating the need to tilt the stratigraphy in the two-dimensional simulation.)

maximum intact matrix porosity of the nonlithophysal tuff (TSMn) at a proposed repository horizon are 0.085 and 0.12, respectively, based on porosity data from UZ-16. The average and maximum intact matrix porosity in a lithophysal tuff zone (TSL1) between 20 and 130 m below the repository are 0.12 and 0.26, respectively. Only in the zeolitic tuff (CHnz), near the water table, does the intact porosity reach an average and maximum of 0.35 and 0.50, respectively.

4.0 ANALYSIS APPROACH AND SCENARIO DEVELOPMENT

4.1 Analysis Approach

In analyzing the risk of a critical condition developing after emplacement, two approaches were used. The first approach assumed that a critical condition would develop and investigated the consequences. The critical condition was treated as an event, and scenarios were developed that contained this event. However this approach, which was pursued in response to interest in the consequences of such an event, required that a probability be assigned to the critical event. The probabilities and consequences of scenarios (features, events, and processes of likely interest) are evaluated to determine whether the scenario should be considered. A small probability or consequence can be used as the basis for removing the scenario from consideration. For systems that are still being characterized and thus are not well understood, however, such an evaluation may give overly pessimistic results which, while useful for relative comparisons of similar systems, are suspect for evaluating absolute probabilities. In evaluating the probabilities and consequences, a purposely imposed constraint was to avoid the use of detailed simulations and, instead, to use general scientific knowledge and natural analogues as information sources.

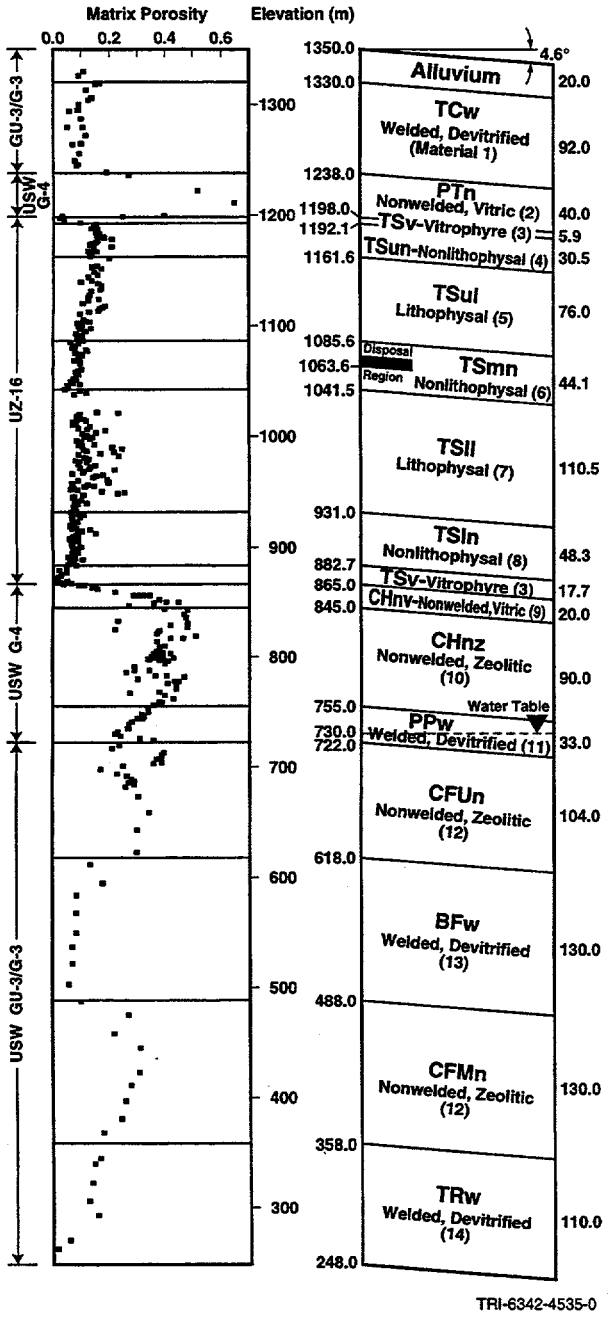

Fig. 4. Correspondence of stratigraphic column of Yucca Mountain tuffs and 12 hydrologic modeling units with similar porosity and/or welding that were used in the simulations (after Rautman, 1995, Figure 8, and Rechard, 1995b, Figure ES-10). Original study (Rechard, 1995b) used 11 modeling units; here the modeling unit containing the repository has been subdivided.

The second approach was to fully simulate the evolution of the disposal system as the container degrades to monitor whether conditions exist under which a criticality could occur. The second approach evaluates a critical condition by examining the natural phenomena that may promote it. This approach permits a better but not complete understanding of the phenomena necessary to promote a nuclear chain reaction in a repository and the likely initial and boundary conditions for a critical event. Although fascinating, a drawback is that the modeling (i.e., Rechard, 1995b) is complex and thus difficult to convey in a report such as this, which is primarily devoted to emphasizing the analysis approach taken. In the following sections, this report describes both the difficulties encountered in evaluating a probability of a criticality event for the first approach, and the success of bounding the consequences (Fig. 5).

4.2 Scenario Development with Criticality as an Event

Scenario development is the second phase of model development (the first, disposal system characterization, has already been discussed) and is the general process of deciding what may happen to the disposal system in the future and how to model it. In conjunction with system characterization, the scenario development process establishes how the real world will be represented by conceptual model(s). Although not often a separate task in the analysis of small systems, in the analysis of a large, complex disposal system it is important to treat this part of model development (even though it is mostly a heuristic process) as a distinct analysis task consisting of the following general steps (Rechard, 1995a): (1) Identifying and listing the universe of features, events, and processes of the disposal system, where events are short-term phenomena and processes are long-term phenomena; (2) selecting those features, events, and processes to model including those elements that have a high probability of occurrence and are likely to contribute to unwanted consequences, while omitting those elements that (a) have exceedingly low probabilities of occurrence, (b) lead to exceedingly low consequences, or (c) have no role in calculations based on regulatory guidance; (3) grouping those features, events, and processes into summary scenarios; (4) creating conceptual models of the disposal system based on system characterization and incorporating the retained elements; and (5) designing performance assessment calculations based on the conceptual models.

In the present study, three events were judged to be important in summary scenarios for the first 10,000 yr: basaltic volcanism (V); human intrusion by exploratory

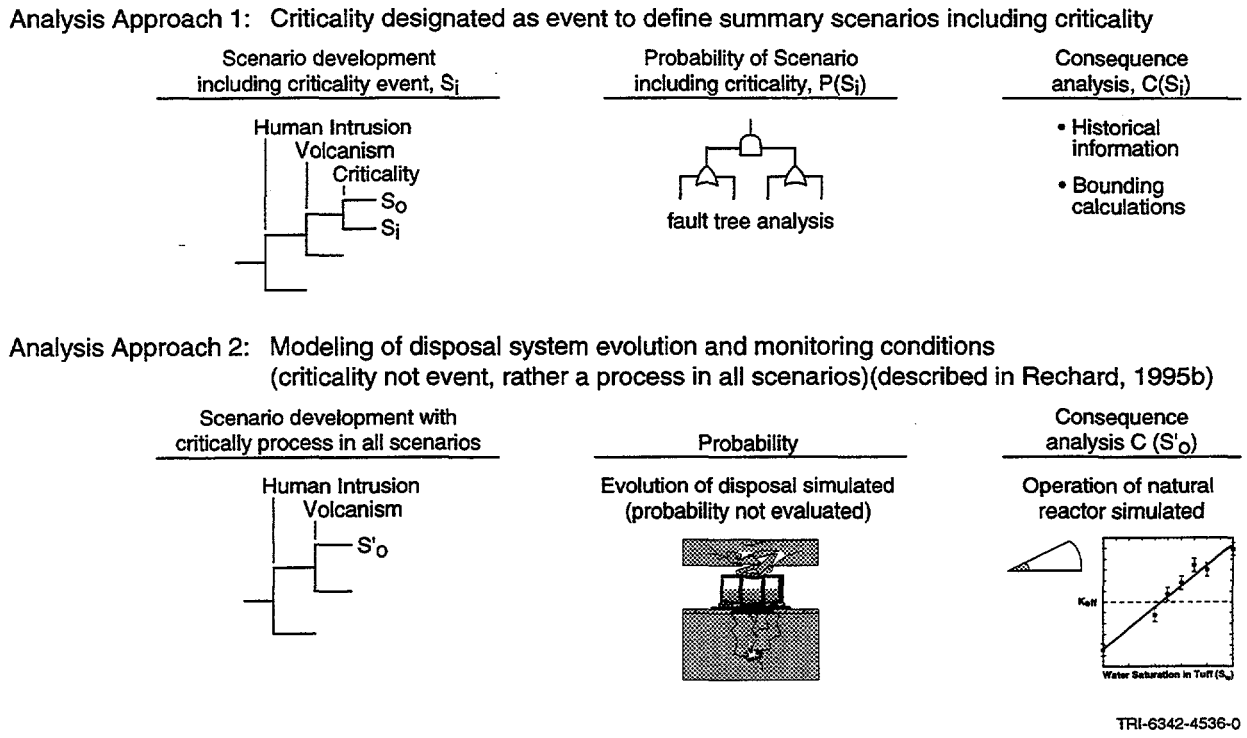

Fig. 5. Analysis approach for evaluating probability and consequences of a criticality in or near a large, horizontally emplaced, multi-purpose canister in a tuff disposal system.

drilling (HI); and criticality (C) (Fig. 6). All other events, processes, and features were treated as either certainties or impossibilities. (A description of the features and processes included in the analysis is presented in Rechard, 1995b.) The elementary probability of basaltic volcanism near Yucca Mountain, Nevada, in the first 10,000 yr was conservatively taken as $\sim 2.4 \times 10^{-4}$ (Barnard et al., 1992, pp. 7-5 to 7-7). The elementary probability of at least one inadvertent human intrusion in the first 10,000 yr was taken as $\sim 5.6 \times 10^{-2}$. (The original study, Rechard, 1995b, used 7.9×10^{-2} because the repository area was larger [2.6×10^4 m^2 versus 1.9×10^4 m^2] due to the greater number of containers needed for high-level waste stored as calcine; see Section 3.2.) The probability of at least one inadvertent human intrusion, $P\{N > 0\}$, uses a Poisson discrete analytic distribution function for exploratory drilling with a drilling rate (λ) of 3 boreholes/area of waste (km^2)/10,000 yr as recommended in Appendix C of 40 CFR 191 (EPA, 1993), i.e., $P\{N > 0\} = 1 - e^{-\lambda \Delta t}$, where Δt = 10,000 yr. The calculation of the 10,000-yr probability of a criticality is calculated in Section 5.3 and is $\sim 2 \times 10^{-3}$. The resulting probabilities of the summary scenarios (Fig. 6) show that scenarios S_3, S_6, and S_7 can be neglected by virtue of EPA guidance in 40 CFR 191 that allows omission of categories of features, events, and processes that have probabilities of occurrence of less than 10^{-4} in 10,000 yr. For example, a criticality with volcanism (S_3) has a very low probability of occurrence, $\sim 10^{-6}$ in the first 10,000 yr, assuming the events are independent.

Eliminating these scenarios leaves the base case scenario (S_0), a scenario involving only repository criticality (S_1), a scenario involving only volcanism (S_2), a scenario involving only human intrusion (S_4), and a scenario involving repository criticality *and* human intrusion (S_5). Only the scenarios involving criticality are of interest here, and only the scenario with just the criticality event (S_1) is discussed further. A critical event on the surface, caused by drilling into a waste container, was not considered in this initial study because the deposition of the fissile material on the surface already represented a release, i.e., a criticality in a mud pit might be no more hazardous to drilling operators than that caused by just bringing radionuclides to the surface because the water would certainly shield the operators to an extent. A definitive analysis of the consequences would require information on the time of intrusion, detailed knowledge of the setup for the future drilling event, and models of

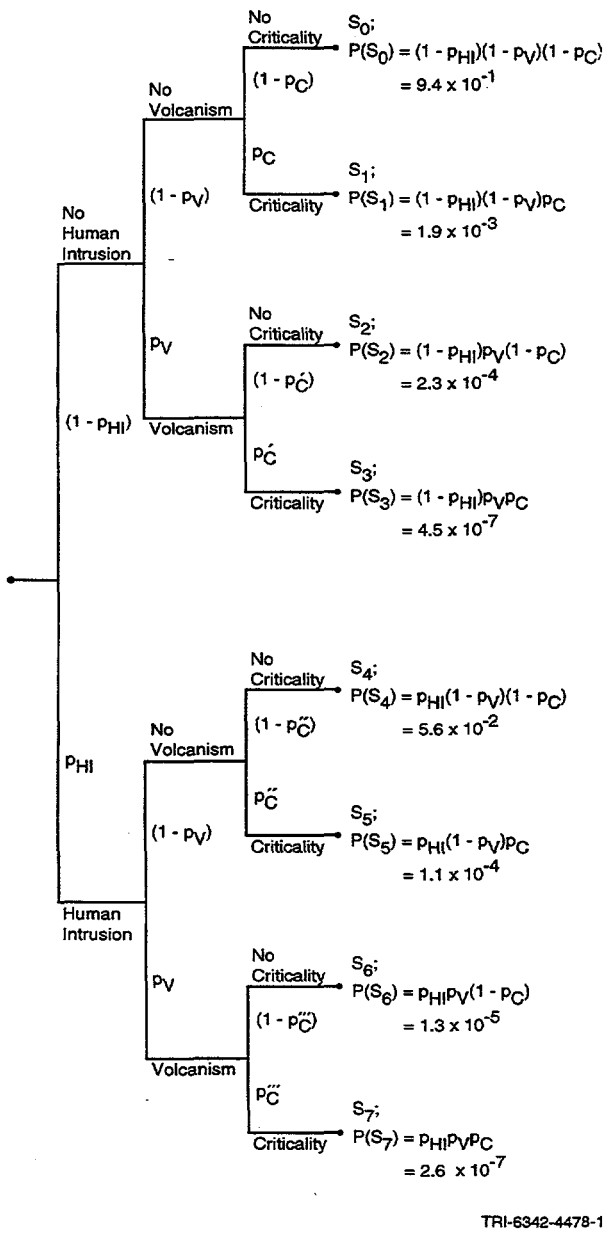

Fig. 6. Logic diagram to enumerate summary scenarios, S_j, for first 10,000 yr of repository performance (after Rechard, 1995b, Figure 7-1). The probabilities of the summary scenarios, $P(S_j)$, are the product of the event probabilities, that is, the probability (or complement of the probability) of at least one occurrence of human intrusion by exploratory drilling, p_{HI}, basaltic volcanism, p_V and any criticality in or near the repository, p_C. See text for numerical values of event probabilities, p_{HI}, p_V and p_C.

the circulating fluid to examine the ability of the fluid to carry significant quantities of the very dense uranium fuel pellets, and was beyond the scope of this study. More important, the human intrusion event is a release mechanism for which the NAS no longer suggests that a probability be assigned (NAS, 1995) (see Section 2.1), and the scenario probability was near the threshold ($1.1 \times 10^{-4} \cong 10^{-4}$). Also, the probability of an intrusion into the necessary number of containers with highly enriched uranium for a criticality at the surface is much lower than general intrusion into any container (i.e., $p_c'' \neq p_c$ because only 14% of the containers contain highly enriched uranium).

4.3 Categories of the Criticality Event

For bounding probabilities and consequences, the criticality event was divided into two basic conditions leading up to a self-sustaining nuclear chain reaction in or near a container. The two basic conditions are (1) moderation (e.g., sufficient or insufficient water to efficiently moderate a nuclear chain reaction) and (2) assembly (i.e., slow, fast, and explosive assembly of the mass of fissile material, where slow refers to processes occurring over geologic time such as chemical precipitation, fast refers to processes on a short time scale such as gravity-driven collapse of spent fuel in a container, and explosive refers to processes occurring in microseconds)[3] (Fig. 7). The first condition categorizes the most important feature regulating the operation of an assembly in the natural environment (e.g., water moderator, see Section 5.1.1). The second condition categorizes the time required (and thus power [energy/time] expanded) to assemble the critical mass. A similar categorization of criticality conditions is reported by Evans et al. (1994) and Rechard (1995b) but omits the explosive assembly.

Examples of the situations follow, together with a review of standard criticality calculations, natural analogues, and criticality experiments and accidents that are relevant to a criticality in a repository and can be used to satisfactorily bound the consequences of a critical condition. As discussed below, the consequences of all, except possibly explosive assembly, have similar consequences even though the probability that any of these situations could occur within a repository varies substantially.

3 The "explosion" descriptor also relates to the basic concept that a small energy release is often necessary to trigger a large energy release. For example, small chemical or electrical blasting caps are used to initiate large chemical explosives; chemical explosives are used to initiate fission explosions; and a fission explosion is used to initiate a fusion explosion.

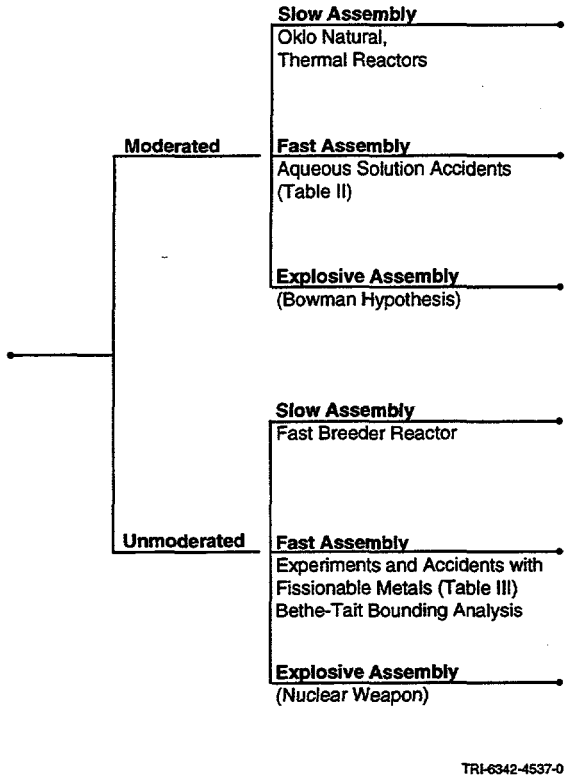

Fig. 7. Logic diagram to further refine those scenarios with a criticality event. Similarities with natural or engineered reactor systems and possible analysis techniques are noted (after Rechard, 1995b, Fig. 10-3).

5.0 BOUNDING PROBABILITY OF CRITICAL EVENT

The assembly process (which includes the process of collecting fissile material in one general location and the final process of bringing together the fissile material into a critical mass) determines the probability. Although the details for this assembly process as modeled are discussed elsewhere (Rechard, 1995b), in this report we present information on analogous events and then general heuristic arguments regarding the probability of subevents (i.e., general categories of processes for which we have removed the dependence on time) of the criticality event to estimate a bounding probability.

5.1 Probabilities of Criticality Conditions

5.1.1 High Moderation, Slow Rate of Assembly (Oklo Natural Reactors)

The moderated, slow rate of assembly type of criticality is represented by the natural reactors in uranium ore deposits in Gabon, Africa. The deposits are located within the Francevillian basin, located along Africa's west coast, which is divided into two regions (Gauthier-Lafaye and Weber, 1989): the northwest edge and central part. Eighty percent of Gabon's known uranium is found in the northwest edge, which holds three main ore deposits. The most economically important is the Oklo ore deposit, which contains 17,300 Mg or metric tonnes (19,070 short tons). Most of the ore is low grade, 0.2% to 1% uranium, with an average of ~0.3%; a high grade ore of 20% to 60% is located at tectonic structures.

The ore deposits occur within an unmetamorphosed sedimentary layer, the oldest high-grade sedimentary deposit known. The uranium-rich layer lies between a sandstone and conglomerate formation (designated FA) and a thick black shale and sandstone formation (designated FB). As proposed by Gauthier-Lafaye and Weber (1989), the FA formation was formed as a deltic deposit about 2.1×10^9 years ago (2.1 Ga), during erosion of the Chaillu massif (Archean-age basement rock, 2.7 Ga); the FA formation is the likely source of the uranium. The FB formation was formed during settling of the basin, possibly 2.06 Ga, allowing seas to transgress across the basin and deposit 3000- to 5000-m-thick marine sediments, rich in organic material, thus burying the FA formation. Subsequent tectonic activity faulted and uplifted the basin. A second porosity was created when highly pressurized fluids hydrofractured the sandstone, allowing hydrocarbons and water to migrate; the hydrocarbons moving through the FB formation were trapped by the faults. Massive amounts of oxygen-rich waters in the wet equatorial climate circulating through the conglomerates in the FA formation dissolved and transported uranium. High concentrations of uraninite (UO_2) (20% to 60%) were precipitated as this uranium-rich water came in contact with the hydrocarbon fault traps because of the reducing environment from the organics, between the time that the FB formation was formed and the startup of the natural reactors.

About 16 lenticular regions have been identified within the Oklo ore body that operated as natural reactors about 1.97 Ga, when natural uranium had a ^{235}U content of ~3.68% (Fig. 8) (Nagy, 1993; Smellie, 1995). The first six natural reactor regions are typically 10 to 20 m in length and width, and less than 1 m thick.

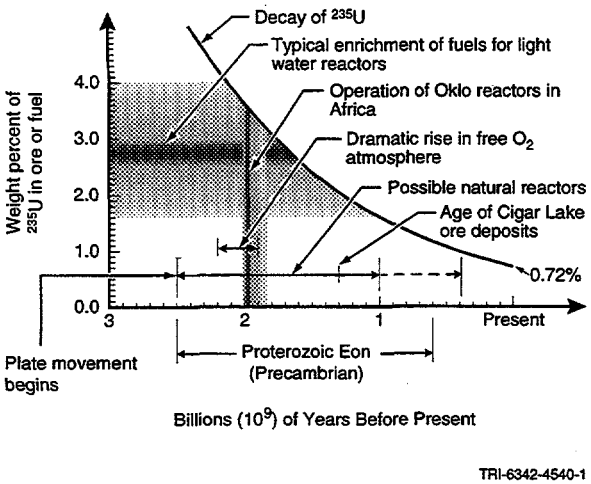

Fig. 8. Operation of natural, pressurized reactor in Oklo uranium deposit in Gabon, Africa, occurred about 2 billion years ago when weight percent of ^{235}U was above three. Illustration indicates typical enrichment of fuels for light waster reactors (Lamarsh, 1984), operation of Oklo reactors (Gauthier-Lafaye et al., 1989), rise in free O_2 atmosphere (Holland, 1994), plate movement (Locardi, 1988), and age of Cigar Lake ore deposits (Cramer and Smellie, 1995).

Pressurization of the water (30 to 40 MPa) from geologic conditions (Blanc, 1995) was such that the temperature ranged between 440 and 640 K (Gauthier-Lafaye et al., 1989). The content of ^{235}U (uranium enrichment) and temperature are similar to a modern, engineered, pressurized water reactor (Fig. 8). (It should be noted that pressurization of water is not possible in the unsaturated zone at Yucca Mountain; hence, the power output would be significantly less than a pressurized system such as Oklo.)

Because the optimum water content of the reactor was quite low (12% to 15% by volume, 6% by weight) (Brookins, 1984; Cowan, 1976), the reactors may have formed in an overmoderated condition, with sporadic drying as the cause of the criticality. Thereafter, each assembly may have operated in a cyclic manner: first, shutting down, since the k_{eff} of the assembly would decrease below one as the water (which served as a moderator for the assembly) expanded near its critical point (Cowan and Norris, 1978) or turned to steam, and then sustaining a nuclear chain reaction when the water cooled or sufficient liquid water percolated down through fractures in the rock. The convective circulation of water heated by the natural reactor continued to concentrate the ore to between 20% and 60% uranium around the reactor (Nagy et al., 1991, reports on fission products precipitated with uranium), altered the hydrocarbons to form bitumen that enclosed some natural reactors (Nagy et al., 1993), and dissolved quartz in the sandstones. However, reactor analysis indicates that the continued enrichment of the uranium ore was not as important to sustained reactor operation as the ability to increase the amount of moderating water and transmute any neutron poisons ("burnable poisons") to compensate for creation of fission products that absorbed neutrons (Reuss, 1975; Naudet, 1975; Maeck et al., 1975; Cowan, 1976). The reactor behavior continued for about 2×10^5 to 8×10^5 yr (Cowan et al., 1975; Gauthier-Lafaye et al., 1989) until conditions changed significantly, e.g., when the reactor cavity collapsed because of hydrothermal dissolution of silica in the sandstone and conglomerate, with a subsequent lack of an optimum configuration in the collapsed cavity (Smellie, 1995). For the first six reactors, researchers have estimated that 6 Mg (6.6 short tons) of ^{235}U was consumed out of ~800 Mg (880 short tons) of high grade uranium (Gauthier-Lafaye et al., 1989; Cowan and Norris, 1978). Consumption of 6 Mg corresponds to $\sim 10^{28}$ fissions or ~15,000 MW-yr (see App. B; Walker et al., 1989). Although up to 3 Mg (3.3 short tons) of ^{239}Pu was also produced in the natural reactor (Cowan and Norris, 1978) from neutron absorption by ^{238}U, very little fissioned (2% to 6%) (Maeck et al., 1975). Most decayed to ^{235}U; a large portion of this ^{235}U inventory was then subsequently fissioned.

If a criticality in the highly enriched fuel can occur in or near a repository, we believe the Oklo type of criticality behavior is the most likely, provided enough water becomes available to change conditions within the repository and to slowly dissolve and remove either the boron or the uranium and moderate the reaction. Consequently, the probability of this type of criticality was used for this report (Section 5.2). Although thought to be a possibly common occurrence soon after its discovery, the Oklo phenomenon remains unusual because other uranium deposit sites of the Proterozoic System of the Precambrian Era show no evidence of forming natural reactors. Any other ore deposits that may have formed natural reactors during this period must have undergone erosion and redistribution (Cowan, 1976) or been mined without detection.

To elaborate, the richest uranium ore deposit in the world at Cigar Lake in Canada has been present for ~1.3 Ga (Cramer and Smellie, 1994; Cramer and Sargent, 1994), but shows no sign of a criticality. This deposit, with an enrichment of ~2% when formed, has an enrichment somewhat greater than the plausible enrichment boundary of 1.6% proposed by Naudet (1977), which corresponds to the lowest enrichment used in portions of commercial reactors (Lamarsh,

1983). The minimum possible boundary for natural reactors is 1% enrichment (400 Ma) (Naudet, 1978). Consequently, the Nopal I ore deposit in northern Mexico (Pearcy et al., 1995), which formed 7.9 Ma as the result of hydrothermal solutions precipitating uraninite in highly fractured regions below the water table of a ~43.8 Ma volcanic tuff formation, is too young to have had natural reactors.

5.1.2 High Moderation, Fast Rate of Assembly (Critical Events in Aqueous Solutions)

A moderated, fast rate of assembly corresponds to the critical aqueous-type accidents in nuclear fuel processing plants (i.e., ^{235}U and ^{239}Pu aqueous solutions) (Table II) or moderated metal and metal-oxide system accidents and experiments (Table III). The reported events generated $\leq 10^{20}$ fissions (~350 kW-hr, ~1250 MJ, or an equivalent consumption of ~16 mg of ^{235}U), with an average of about 10^{18} fissions (~5 kW-hr, ~19 MJ, or burnup of ~0.23 mg ^{235}U). Also note that the maximum number of immediate fissions is also only about 10^{18} or a maximum quick release of energy of 18.7 MJ. This type of criticality excursion could be the result of a human intrusion situation that generates a slurry consisting of fissile material and water (moderator)—a scenario not evaluated because the intrusion already represented a release, as noted earlier, and precise details of the drilling operation were needed to evaluate consequences. (A situation in which water dripped onto a mass of fissile material [previously assembled through, for example, collapse of a partially corroded container] that was not critical until the addition of water would resemble an Oklo situation, discussed above.) No characteristic of these situations makes it more probable than a high moderated, slow rate of assembly (discussed above) or an unmoderated, fast rate of assembly (discussed below) because an additional condition, such as an earthquake or human intrusion, is required.

5.1.3 High Moderation, Explosive Assembly

A highly moderated, explosive assembly can be represented by one of the events that was recently hypothesized by Bowman and Venneri (1996).[4] Bowman and Venneri (1996) describe the effects of six situations, which occur as the result of an arbitrary assembly of some fissile mass of ^{239}Pu, water content, and pure silica content (SiO_2) (instead of tuff). The six situations differ primarily in whether the assumed collected mass would experience negative or positive feedback during a critical event. Bowman and Venneri further suppose that all situations with positive feedback could result in nuclear explosions.

Briefly, the situations can be summarized as follows. Situation 1 involves movement of water into an already deposited sphere of fissile material, which then goes critical with negative feedback. A modification to Situation 1 was also included, in which the sphere of fissile material contains more mass than is required to go critical. After being deposited into a small region, the mass goes critical and then fissile material is dispersed out into a *dry* matrix through "water-steam explosions," creating a new configuration that can experience positive feedback. Situation 2 involves the transport of 10 kg of fissile material from the container and uniform distribution of the material into a 0.5-m-radius region of silica with a 0.1 water mole fraction (an uncommon method of describing water saturation) that goes critical with negative feedback. In Situation 3, a 70-kg mass of fissile material is transported and deposited in a 2-m-radius region with 0.15 mole fraction of water, resulting in a configuration that can experience positive feedback. Situation 4 is similar to Situation 3 except that 15 kg of fissile material is deposited in a 1-m-radius region with 0.27 mole fraction of water. Situation 5 involves collecting 70 kg of fissile material in a 2-m radius; the fissile material is assumed to remain noncritical because of too much moderator (i.e., overmoderated with water). When water is removed through drying, the fissile mass goes critical and experiences positive feedback. Finally, Situation 6 involves a 2-kg mass of fissile material deposited into a 0.3-m-radius region with a water mole fraction of 0.60, which experiences positive feedback. Note that a water mole fraction of 0.60 is beyond the 0.16 to 0.26 water mole fractions that are considered physically possible in the lithophysal and nonlithophysal tuff near the repository horizon.[5]

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4 In order to obtain the nuclear explosion proposed by Bowman and Venneri (1996), the assembly must occur in microseconds; thus the assembly is termed "explosive." The need for such rapid assembly is explained in Section 5.1.6, Low Moderation, Explosive Assembly. Also, note that in this report, assembly means assembling into a configuration that is supercritical. This assembly can occur either from fissile material "coming together" or from material diffusing uniformly "out" into the moderator.

5 As noted in Section 3.6, the maximum intact matrix porosity of the nonlithophysal tuff at a proposed repository horizon is 0.12 (0.16 water mole fraction), and the maximum intact matrix porosity in a lithophysal tuff zone between 20 and 130 m below the repository is 0.26 (0.33 water mole fraction). These conversions assume that the pores are fully filled with water and that the pure silica matrix is cristobolite. The intact porosity reaches a maximum of 0.50 (0.58 water mole fraction) only near the water table; however, the travel time of the moderately adsorbed uranium can average 50,000 years. (Also, plutonium is well adsorbed and takes much longer.) Furthermore, extensive mixing with the low enriched spent fuel and high-level waste will likely have occurred at this distance and time.

Table II. Criticality Accidents in Processing Plants

Date	Plant	Total Fissions	Prompt Fissions	Doses (Rads)	Cause
6/16/58	Y-12	1.3×10^{18}	7×10^{16}	365, 339, 327, 270, 236, 69, 69, and 23	^{235}U solution washed into drum
12/30/58	LASL	1.5×10^{17}	1.5×10^{17}	4400 (fatal), 135, and 3	Plutonium concentrated in solvent layer
10/16/59	ICPP	4×10^{19}	10^{17}	50 and 32 (primarily beta)	^{235}U solution siphoned into tank
1/25/61	ICPP	6×10^{17}	6×10^{17}	None	^{235}U solution forced into cylinder by air
4/7/62	Hanford Recuplex	8.2×10^{17}	10^{16}	87, 33, and 16	Plutonium solution in sump sucked into tank
7/24/64	Wood River Junction	1.3×10^{17}	10^{17}	10,000 (fatal), Two 60 to 100	^{235}U solution poured into tank
8/24/70	Windscale	10^{15}	10^{15}	Negligible	Plutonium concentrated in trapped solution
10/17/78	ICPP	3×10^{18}	Unknown	None	^{235}U buildup due to diluted scrub solution

Taken from Knief (1985), original reference, Paxton (1983). (Data values shown here are in good agreement with those in Stratton and Smith, 1989).

Table III. Criticality Accidents Involving Moderated Metal and Oxide Systems (after Stratton and Smith, 1989)

Date	Plant	Total Fissions	Prompt Fissions	Doses (Rads)	Cause
6/06/45	Los Alamos	4×10^{16}	3×10^{15}	66, 66, and 7.4	Water leaked into assembly
1950	Chalk River	Unknown	NA	NA	Excess moderator added
6/02/52	Argonne National Lab	1.22×10^{17}	NA	136, 127, 60, and 9	Control removed, water not removed
12/12/52	Chalk River	1.2×10^{20}	NA	low	Positive void coefficient
7/22/54	Idaho National Lab	4.68×10^{18}	NA	NA	Planned transient extended
1015/58	Vinca, Yugoslavia	2.6×10^{18}	NA	205, 320, 410, 415, 422, and 433	Faulty power monitoring
3/15/60	Saclay, France	3×10^{18}	NA	NA	Removal of absorber rod
1/03/61	Idaho National Lab	4.4×10^{18}	NA	3 fatalities	Removal of control rod
11/05/62	Idaho National Lab	1×10^{18}	NA	NA	Planned transient exceeded
12/30/65	Mol, Belgium	4.3×10^{17}	NA	NA	Misoperation plus not draining tank
9/23/83	Buenos Aires, Argentina	4×10^{17}	NA	low	Failure to drain tank

Critics of the analyses put forward by Bowman and Venneri focus their attention either on the consequences (usually critics with weapons experience) or the assembly process (usually those with geologic background). With regard to consequences, critics wonder (1) whether regions of positive feedback actually exist in a volcanic tuff system versus a pure silica system, (2) whether a positive feedback mechanism that exists briefly in a fissile mass implies a nuclear explosion, and (3) how much energy is potentially released from a positive feedback mechanism. In answering the first concern, volcanic tuff is far from being pure silica, containing several elements that can significantly absorb neutrons. Also, the assembly is not a homogeneous mixture, but is instead heterogeneous so that material can actually shield the fissile mass from neutrons ("self-shielding"). Given this information, there are few situations where positive feedback could occur; also much more fissile mass than that purported by Bowman and Venneri (1996) is required to go critical (Parsons et al., 1995).

In replying to the second concern, credible means for changing the system naturally—e.g., removing the moderator from an overmoderated system by evaporation or water vaporization—are slow in relation to the microseconds required for nuclear explosions (see Section 5.1.6; Stratton, 1983; Sanchez et al., 1995). For example, even the maximum transfer rates achievable through pumping in nuclear fabrication facilities for PuO_2 and PuO_2—UO_2 are incapable of causing and sustaining a nuclear explosion (Hansen et al., 1976).

With regard to the third concern, the energy release calculations are suspect because the initial conditions for the calculations are neither justified nor are they the same as the initial conditions used to describe the situations (i.e., a $k_{eff} = 1.1$ is used in the energy release calculations, but no explanation is given of how a natural system moves from just critical at $k_{eff} = 1$ to $k_{eff} = 1.1$, an excess reactivity of ~14). For example, the porosity of the tuff matrix is too great to confine the critical assembly, thus virtually all of the energy generated would be heat rather than kinetic energy (Kimpland, 1996; Parsons et al., 1995). Finally, there is no natural analogue, accident, or experiment that can be cited as an example of a system, which starts at near critical ($k_{eff} \cong 1$), and then has a nuclear explosion initiated solely as the result of positive feedback.

Critics of the assembly process (Canavan et al., 1995; Van Konynenburg, 1996) are concerned with its improbability because of geologic concerns. Because the six situations are only generally described (e.g., terms like "water-steam explosion" and "driven by fission heat through rock" are used), they must be translated into known geologic phenomena and a complete conceptual model developed so that the corresponding hypothesis can be tested for reasonableness. For example, we note that in order to collect such a large mass of fissile material, all six situations require groundwater to promote corrosion, dissolution, transport, and deposition, although these conditions are not specifically stated.

Furthermore, we note that as the hypothesized fissile material is collected (probably through precipitation, though Bowman and Venneri use the phrase "water carrying plutonium oxide particles and depositing them somewhere else," possibly implying colloid transport), the groundwater would be present to act as a moderator, resulting in either a high moderation, slow rate of assembly (e.g., Oklo natural reactors) or high moderation, high rate of assembly (aqueous accidents). To illustrate, Bowman and Venneri's Situation 3 assumes 70 kg of ^{239}Pu, 2-m radius, 0.15 water mole fraction, and positive feedback; however, earlier, Bowman and Venneri showed that only negative feedback occurs with about 2 kg of ^{239}Pu in a 0.3-m-radius region and a water mole fraction of 0.15. Thus, to obtain initial conditions with positive feedback, the Bowman-Venneri explosion hypothesis (which has not been observed) requires the system to pass through a possible Oklo-like situation (which has been observed) requiring less fissile mass.

A similar problem, in which the system must pass from an Oklo-like situation to another situation, is more explicit in the variation of Situation 1 hypothesized by Bowman, in which the system is silica-moderated rather than water-moderated. They hypothesize the system could experience positive feedback either as the silica expanded because of its positive coefficient of thermal expansion or as the fissile mass expanded in diameter (Taubes, 1995). (The increase in multiplication factor [k_{eff}] as the fissile mass expands was first studied by Carter [1973].) Although SiO_2 has the classical characteristics of a moderator, it is very inefficient. Assembly of fissile mass with silica as the moderator for the chain reaction would require, first, water to create the optimal mixture of the fissile material with the silica (simple diffusion of the fissile material into the tuff would not occur within a reasonable time scale). Consequently, prior to removing the water, the conditions would favor startup of the nuclear chain reaction with water as the moderator instead. Thus a new mechanism must then be invoked ("water-steam explosions"), but it is not a credible mechanism to reach the hypothesized *dry* conditions (Van Konynenburg, 1996). In essence, the conditions necessary to produce the explosion hypothesis are inconsistent with the conditions necessary to produce the initial conditions for the assembly. This inconsistency in assumptions is a very serious error in

developing a scientifically acceptable conceptual model of a phenomenon (Rechard, 1995a).

Based on these criticisms, we argue that the Bowman and Venneri situations resemble highly moderated, slow or fast rates of assembly discussed in the previous two sections, even though in rare instances the system could experience positive feedback for a time; we assigned a probability of zero to the highly moderated, explosive assembly category. Although not used in the above arguments, the mechanisms also may be improbable. General geochemical processes for separating the fissile mass from poisons and reconcentrating it are difficult to achieve. Examination of these geochemical processes has been presented when the dissolution of the spent nuclear fuel was simulated along with its consequences (Rechard, 1995b).

5.1.4 Low Moderation, Slow Rate of Assembly

A low or unmoderated, slow rate of assembly would roughly correspond to conditions in a breeder reactor. In a natural setting, a credible method for *slowly* assembling the fissile material without water or human intervention is unknown and probably thermodynamically impossible because it would require unreasonably large decreases in local entropy. Consequently, the probability was set to zero. However, a fast rate of assembly from a sudden collapse of the fuel after sufficient corrosion of the support matrix is considered in the next section.

5.1.5 Low Moderation, Fast Rate of Assembly (Experiments with Fissionable Materials)

Low moderation corresponds to assemblies that contain little or no water or any other moderating materials, i.e., elements such as hydrogen, carbon, and beryllium, which have a high probability of scattering neutrons while simultaneously not absorbing many neutrons. This situation would create a "hard" critical nuclear assembly; the "hard" descriptor means that the neutrons in this nuclear assembly have a "hard" (high energy) spectrum. As mentioned earlier, this situation might develop from partial corrosion of the support structure followed by a sudden collapse, e.g., an earthquake ($r_E \cong 10^{-3}$ yr^{-1}). An unmoderated, fast rate of assembly would likely correspond to accidents and experiments carried out at Los Alamos National Laboratory (e.g., Godiva Reactor [Paxton, 1983]). Some of the accidents yielded between 10^{16} and 10^{17} fissions. The number of fissions produced by prompt neutrons was calculated to be 6×10^{17}, which is comparable to the number of prompt fissions estimated to have been produced in aqueous accidents (Table II). "Bethe-Tait" bounding type calculations done for evaluating the safety of two fast reactors yield upper bounds of 3×10^{19} fissions for assemblies with a large mass of fissionable material (Meyer et al., 1967; Bethe and Tait, 1956). A hypothetical criticality accident in a waste supercompactor also represents an unmoderated, fast rate of assembly. Calculations showed hypothetical total yields between 10^{18} and 10^{19} fissions and bulk temperatures between 412 and 912 K (Plaster et al., 1995).

5.1.6 Low Moderation, Explosive Assembly

Low moderation, with explosive assembly, is similar to a nuclear weapon but requires special design features—a situation that cannot occur by accident; it is mentioned here for completeness. To elaborate, although a $k_{eff} > 1$ is a necessary condition for an atomic explosion, it alone is not sufficient (see Appendix A). Special conditions must exist such that the rate of increase in k_{eff} is very rapid. To obtain a rapid increase in k_{eff} requires, in turn, that sufficient energy be added to the system such that the fissile mass be assembled very rapidly to allow for many generations of neutrons (each uranium fission and thus each generation requiring about 10 nanoseconds) to produce massive numbers of fissions and thereby sufficient heat and kinetic energy before negative feedback changes the configuration and limits the number of neutron generations. For ^{239}Pu and ^{235}U, an atomic explosion is typically produced by imploding a sphere of fissile mass together through detonating surrounding explosives at precisely the same time and injecting a large number of neutrons (not just stray neutrons) at precisely the correct moment (i.e., basic concept of Fat Man). For just ^{235}U, a somewhat simpler design (i.e., basic concept of Little Boy) is possible in which a large number of neutrons is injected precisely at the same time that two subcritical amounts of ^{235}U are explosively brought together to form a supercritical mass (an explosive assembly with a velocity between 300 and 1000 m/s [1000 and 3000 ft/s].[6] This velocity is ~300 to 1000 times faster than the "fast" assembly caused by the acceleration of gravity over ~1 m. More details on the design of nuclear weapons can be found elsewhere (Serber, 1992; Hansen, 1988; Bickel, 1979).

6 This design is not possible with ^{239}Pu because stray neutrons from ^{240}Pu prevent it.

5.2 Fault Tree for High Moderation, Slow Rate of Assembly

Since their development by Bell Labs and subsequent use by Boeing on the Minuteman Missile in 1963 (Haasl, 1965; Hixenbaugh, 1968), fault trees have been used to evaluate the probability of failures within complex human-engineered systems. Consequently, a method for developing an understanding about the likelihood of the assembly of fissile material is to develop a fault tree in which several distinct events leading up to a critical condition are postulated, and the corresponding probabilities of occurrence for each event are then combined to arrive at overall probabilities. Although it is possible to use fault trees to evaluate changes in states of components within a geologic disposal system, their use requires care. An advantage of fault trees in this type of analysis is that their structured approach can quickly identify probabilities of a criticality in a repository, and it was this advantage that prompted their use for developing hypotheses of behavior in this study (Fig. 9). Because of the proposed design for the multi-purpose canister assumed in this study, a crucial step to developing a critical condition is corrosion of the container and subsequent separation of the boron (in the borated stainless steel support structure) and the highly enriched uranium in the spent fuel. Once the separation occurs, various types of criticalities, as previously described, may be feasible. Consequently, the important group of events in the fault tree involves the separation of boron from the highly enriched uranium and is described below. One method for evaluating this probability, which is used here, is to determine the rate of formation of conditions conducive to a critical event. The conditions for container failure, removal of boron and general assembly are described below for the most probable branch (Fig. 9).

5.2.1 Container Failure

The only credible manner of destroying the container and removing the boron from around the spent fuel is through corrosion and transport by water. The importance of water is similar to that for general release from the disposal of radionuclides in the repository without a critical event. The following discussion assumes that the failure mode of concern is from corrosion as the result of fractures that drip water. Certainly failure from corrosion due to humid air, or corrosion from the partially saturated tuff matrix contacting the container, can occur. However, these two modes of container corrosion would not likely provide enough water

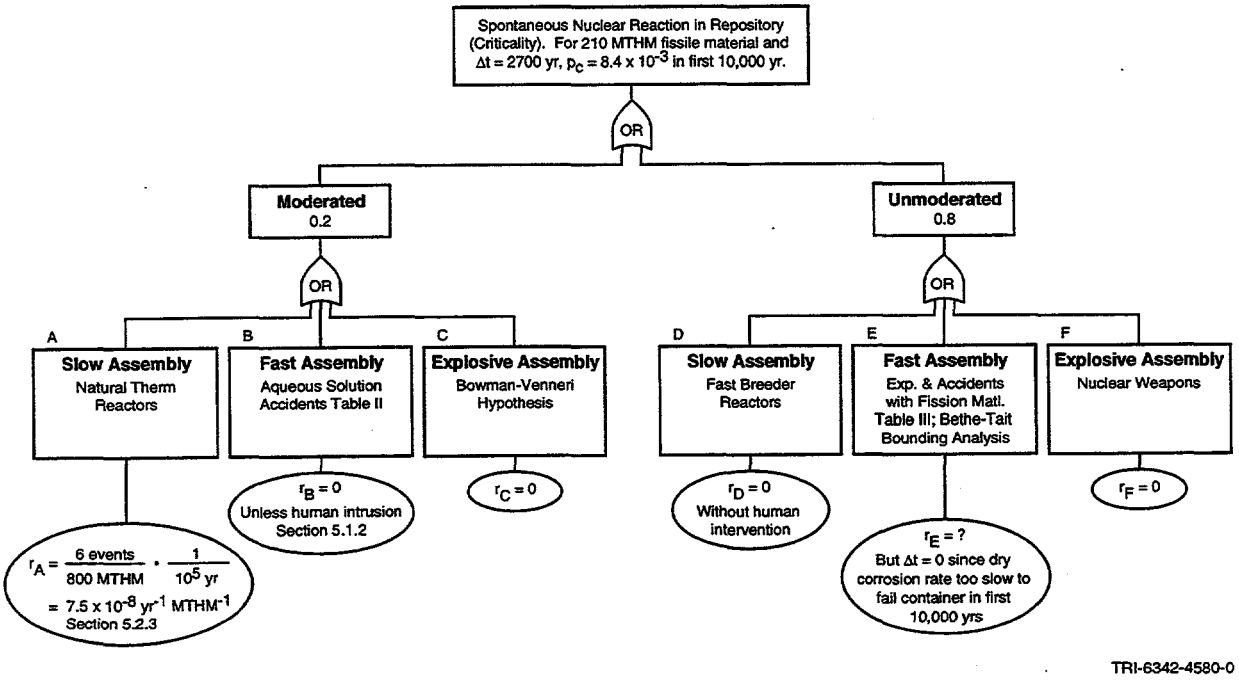

Fig. 9. Fault tree to evaluate probability of a criticality event, showing that the branch with high moderation and slow rate of assembly is most probable. As evident from the text, the great difficulty is assigning probabilities to the initiating conditions.

to transport and separate the boron and uranium, and thus allow a critical event; hence, they are not discussed further.[7]

Analysts currently assume that the repository at Yucca Mountain would be dry initially or, if not completely dry, that containers would not be placed near any seepage found. Hence, for corrosion to occur from fractures that drip water, new seepage areas would have to appear. Possible sources for this water include increased precipitation at the site, as the result of changes in climate; the water could then percolate down and intersect the repository. Although the probability of this change in initiating events is unknown, here we assume substantial change cannot occur within 10,000 yr (consistent with EPA's intent in limiting analysis to this time period [40 CFR 191]). However, a very similar effect can occur with the hot repository concept modeled here (see Section 3.5), in that vaporization and subsequent condensation can cause redistribution of the water in the matrix pores (~65% saturated) such that parts of the repository have dripping fractures. Simulations can model this effect (Rechard, 1995b), but in this report we use climatic change as an analogue for this effect and, in turn, use the information from Rainier Mesa, a nearby region of the Nevada Test Site (NTS) with precipitation twice as great as at Yucca Mountain (Wang et al., 1993; Russell et al., 1978). Although the spacing of the potentially wet fractures is not known, here we used ~25 m, which is a frequent spacing of wet fractures in the E and O tunnels, which are located under Rainier Mesa (Carnahan et al., 1991), even though the number of connected fractures has been increased by nuclear testing (Clebsch, 1960; Russell et al., 1978). In these tunnels, the spacing of moist fractures can extend up to 300 m; in G tunnel at the NTS, only 1 or 2 wet fractures exist. Consequently, a 25-m spacing represents a reasonably tight spacing for the volcanic tuff at Yucca Mountain in a wetter climate. With a fracture spacing of 25 m and a container length of 5 m, approximately 20% of the containers would eventually fail from wet fractures.

The time to container breach is about 300 yr, based on a fully immersed generalized corrosion rate of 0.4 mm/yr at 100°C for carbon steel (Kirby, 1979; Soo, 1984) and a fully immersed local penetration rate of 2 mm/yr at 100°C for Incoloy 825 (Fig. 2) (Wilson et al., 1994, Table 13-5). (The maximum rate for Incoloy 825 is high, because above 80°C Incoloy 825 is sensitive to localized corrosion from pitting.) Faulty containers could shorten this life (Rechard, 1995b), but for bounding calculations the container life is already very short and cannot significantly influence the probability of a critical event.

5.2.2 Physical Removal of Boron

Once the container has been penetrated, the uranium and/or boron can be transported. Boron in molten borated stainless steel is not very soluble and forms precipitate borides, frequently chromium boride, within the stainless steel. In general, metal borides are very hard, refractory, and chemically inert because of the strong covalent metal-boron bond (Cotton and Wilkinson, 1972). Because of this bond, the bulk stainless steel will likely corrode faster than the borides, which will remain as small unreacted particles within the corrosion products. Consequently, whether the boron, which is highly soluble once released from the chromium boride particles, is released faster from the boride particles than the low soluble uranium depends on the amounts of water and oxygen present; these questions were examined in a simulation (Rechard, 1995b). In the heuristic arguments presented in this section, however, the quantities of water and oxygen are not evaluated and so either boron or uranium transport has to be assumed.

Because transport of uranium requires a reconcentration of the uranium (because the solution is not critical), which is inherently less probable (ore bodies are not ubiquitous in the earth's crust), boron removal is assumed in this section. In the container design considered here, the amount of boron that must be removed before a critical event could occur is quite large. In the designs ~0.02% boron in the stainless steel of the support structure was sufficient to prevent sustained nuclear chain reactions; yet, commercially available borated stainless steel is 2% boron—100 times that required.[8] Here, the boron release is assumed to be congruent with the generalized corrosion of the stainless steel, which is highly conservative because the generalized corrosion of stainless steel would still leave the chromium boride particles, from which the boron would still have to escape as noted above. For stainless steel, McCright et al. (1987) report a fully immersed, generalized corrosion rate at 100°C of about 0.1 microns/yr; this is likely near the high end of possible corrosion rates (Fig. 2).

7 For the simulation (Rechard, 1995b), this inability to transport was not assumed *a priori* because any type of corrosion prior to having enough water to fill fractures could reduce the time to failure; other conservative conditions in the analysis, such as fully wet corrosion rates, are assumed to compensate for this simplifying but non-conservative assumption.

8 "Burn up of boron" (boron depletion by capturing a neutron and then conversion to lithium) is not an issue because the container is not in an intense neutron flux as occurs in a reactor. Rather, the boron is present to prevent any stray neutrons from starting a chain reaction.

About 7000 yr would be required to corrode all of the stainless steel at this maximum rate. (At the mean of the distribution used in this study, it would take 10^5 yr, and at the median of the distribution, it would take 7×10^6 yr.)

5.2.3 Probability of Conditions Conducive to a Critical Event

The only known basis for determining a rate of formation of critical events that accounts for phenomena not specifically mentioned above (i.e., fracture spacing and climatic change) is the number of uranium sites in the world that have gone critical. Only about 16 reactor zones in the Oklo ore deposit in the Francevillian basin are known to have gone critical. At the Oklo, the rate of formation of the reactors is unknown, but the upper bound is the time required to form the uranium-rich layer, about 10^8 years (2.06 Ga - 1.97 Ga). The lower bound is possibly the minimum operating life of the reactors, about 2×10^5 years (see Section 5.1.1). Furthermore, six of the reactor zones involved about 800 MTHM. Thus, the maximum formation rate is $\sim 3.75 \times 10^{-8}$ events/yr/MTHM (6 events/[800 MTHM • 2×10^5 yr]).

Clearly, given the heuristic arguments described above that do not depend upon simulation of the container, and given enough time, conditions conducive to a sustained nuclear chain reaction could occur. This probability, however, is conditional on the container being under a dripping fracture and more infiltration occurring through the mountain than is thought now to occur. As noted above, the probability of greater infiltration from fluid redistribution was conservatively set to one, and the probability of being under a dripping fracture was set to 0.2. Finally, the amount of fissile material that can go critical is only highly enriched uranium (210 MTHM). The remainder of the DOE waste is either high-level waste that does not contain fissile material or is similar to commercial spent nuclear fuel whose enrichment is below 2%. Fig. 8 graphically shows that for about 2×10^9 years, no known uranium ore deposits with enrichments less than 2% have gone critical in a natural setting. Commercial spent nuclear fuel will also have enrichments less than 2% and, furthermore, contain numerous fission fragments that readily absorb chain-carrying neutrons. Therefore, the rate of formation, $r(t)$, is 1.6×10^{-6} yr^{-1} (0.2 • 210 MTHM • 3.75×10^{-8} events/yr/MTHM).

In the above argument, the rate of 1.6×10^{-6} events/yr is sensitive to the assumed time required to form a natural reactor. (Recall that 2×10^5 yr was used, based on the time of operation of an Oklo reactor.) Alternately, one can use the uranium solubility in tuff and possible flow rates of water through the repository to determine a rate. However, evaluating a flow rate in a fracture system is difficult because both the infiltration and the surface area contributing to the fracture flow are highly uncertain. Hence, the following text solves for the flow rate (q) based on the 1.6×10^{-6} events/yr to check for reasonableness. The solubility of uranium (S) in an oxic environment is typically measured at 10^{-5} mM (Cramer and Smellie, 1994), but analysis of the volcanic tuff repository frequently uses theoretically calculated values for an oxic environment with a mean value of $10^{-1.5}$ mM (Wilson et al., 1994). Based on this latter solubility, the flow rate (q) is 29 m^3/yr or 2.9×10^5 m^3 over 10,000 yr [1.6×10^{-6} events/yr = (6 events/800 Mg)(10^{-3} mM)(q)]. Using an infiltration rate of 24 mm/yr that occurs at Rainier Mesa (Wang et al., 1993) (equivalent to complete desaturation ~87 m above the hypothetical repository) yields a contributing area of 1200 m^2. This contributing area contradicts the 25-m spacing of wet fractures assumed initially and so, to be consistent, the area must be reduced by a factor of two to 625 m^2. Hence, the time to form a natural reactor in a Yucca Mountain-like environment is underestimated by at least two times and probably by two or more orders of magnitude (the latter because the theoretically calculated solubility of uranium is likely high [Cramer and Smellie, 1994]).

Next, we determine the probability in the first 10,000 yr to be consistent with the probabilities cited for the volcanism and human intrusion in the logic tree for developing scenarios described in Section 4.2. The probability model is based on the failure-rate function (Elandt-Johnson and Johnson, 1980, p. 51) defined by $r(t) = -d/dt\ ln[1-F(t)]$, where t is time elapsed since the disposal system was closed and $F(t)$ denotes the cumulative distribution function (CDF) for the first time, T, when failure occurs (i.e., $F(t) = P\{T \leq t\}$). This expression can be integrated to give

$$F(t) = 1 - exp\left[-\int r(\tau) d\tau\right] \qquad [1]$$

Here $r(t)$ is a constant and equal to 8×10^{-7} (1/2 of 1.6×10^{-6}) over a period of 10,000 yr. In the first 10,000 yr, however, the containers must first fail and the boron be separated from the uranium, which requires at least 7300 yr to occur, as described above (300 yr for container breach and 7000 yr for boron removal). Integrating equation 1 from 7300 to 10,000 yields a probability of 2×10^{-3}, which was used in evaluating probabilities of scenarios in the above section (Section 4.0 and Fig. 6) (10^{-2} was used in the original study). The probability evaluated by this fault tree applies primarily to the "no intrusion, no volcanism" branch in the event

tree (Fig. 6). A fault tree, applicable to each major condition of the event tree in Fig. 6 is needed (e.g., human intrusion, but no volcanism); however, each fault tree provides only an order of magnitude number that does not change substantially and so they will not be shown here. The above arguments noted the possibility of at least a two order of magnitude drop in the rate, which would reduce the probability to less than 10^{-4} over 10,000 yr. Such a low value eliminates the critical event from further consideration, according to the guidance of 40 CFR 191. However, 40 CFR 191 no longer applies, and the time frame of interest will likely increase to 10^6 yr (see Section 2.1). Although the authors believe that the probability is likely low even over 10^6 yr, based on experience in analyzing geologic disposal systems, very low values over 10^6 yr cannot be easily demonstrated for scenarios containing a critical event, without a more fundamental simulation of the phenomena potentially creating a critical condition. For example, Rechard (1995b) shows the extreme difficulty in separating fissile material, with natural phenomena, from any neutron absorbers (e.g., fission fragments and/or boron) and to collect fissile material to initiate any type of critical condition.

6.0 BOUNDING CONSEQUENCES OF CRITICAL EVENT FOR SLOW AND FAST RATES OF ASSEMBLY

In the above presentation, explosive assembly was argued to have a probability of zero. However, slow and fast rates of assembly had probabilities greater than 10^{-4} over 10,000 yr. Consequently, bounding calculations of the consequences of a critical condition were performed (assuming the boron and uranium had separated) so that, if the consequences were negligible, a basis would be established for neglecting the scenario. (Note that when this approach is used, it is important to hypothesize reasonable initial conditions.) Consequences of a critical event can include (1) creating a greater radiological hazard by producing more radioisotopes from the fission process, including more fission gases, and (2) damaging the tuff repository by compromising the isolating and stabilizing functions of the geologic medium by generating kinetic or additional heat energy. These consequences are discussed in the following four sections.

6.1 Bounds on Total Energy (Fissions)

Both moderated and unmoderated fast rates of assembly release similar amounts of energy (as represented by fissions) of between 10^{15} and 10^{20} fissions. These rates provide an empirical bound on the energy release from a critical event because in an unsaturated repository, the criticality is assumed to occur at near atmospheric pressure with a breached container. For comparison, a 70,000-MTHM Yucca Mountain-type repository of spent fuel with burn-up of 40,000 MWd/MTHM would represent on the order of 10^{31} fissions, and the 12,060-MTHM repository used in this study would represent on the order of 10^{30} fissions. (Fuel in commercial nuclear reactors typically has a burnup range of 25,000 to 40,000 MWd/MTHM, with an average of 30,000 MWd/MTHM [Foster and Wright, 1977; Appendix A, 40 CFR 191].) Clearly, an increase in fissions from one critical event of 10^{20} would be negligible; only radioisotopes with very short half-lives would show any appreciable increase in comparison to the original inventory (see Section 6.2).

Because the fast rate of assembly, such as a gravity-driven collapse of spent fuel in a container, would occur sporadically, if at all, the slow rates of assembly such as those found in the Oklo phenomena, which can be considered continuous, could produce more total fissions. Both the magnitude and the rate of the Oklo phenomena are unknown. For this study, we assume a maximum number of fissions per event equal to the unmoderated, fast assembly (10^{20}) and a rate of one event per day (this rate is defended in Section 6.3). One critical event occurring every day for 10,000 yr would amount to ~10^{26} fissions, and one critical event occurring every day for one million years would amount to ~10^{28} fissions, which is only 1% and 0.1% of the fission inventory represented by the 12,060- and 70,000-MTHM repositories, respectively.

Another bounding approach that yields similar amounts of energy is to assume that the 210-Mg fissile contents of all 372 containers of highly enriched fuel undergo a burnup of 40,000 MWd/MTHM (which corresponds to a fission of ~4.3% of the ^{235}U atoms in commercial PWR fuel); this situation would result in ~10^{28} fissions. Alternately, using the fact that six events at Oklo occurred in 800 MTMH, consuming 6 MTHM (i.e., 1 MTHM consumed/event), then for 210 MTHM of highly enriched fuel, 10^{28} fissions occur. The 10^{28} fissions are identical to the total power estimated for the Oklo reactors (Section 5.1.1).

6.2 Increased Radioisotope Inventory

The radioisotope inventory is an important component when evaluating the performance of a geologic disposal system. A sustained nuclear chain reaction would increase the radioisotope inventory, but only by a very small amount, as follows. While the initial evaluation (see Evans et al., 1994) was made using

ORIGEN2.1 (Croff, 1983; ORNL, 1995), a relatively simple expression is presented here from which to calculate increases, assuming constant power generation from criticality.

The additional inventory caused by a constant fission rate and accounting for depletion from radioactive decay is the solution of the equation, $dN/dt = C_f - \lambda N$, where N is the number of atoms of one product from a fission, λ is the decay constant and equal to $\ln 2/t_{1/2}$, where $t_{1/2}$ is the half-life of radioisotope product, and C_f is the growth rate (Ci/s) and equal to the fission rate (r_f) (expressed as Ci/s) times the standard fission product yield (f_y). Solving this equation and adding in the initial inventory of the fission product yields

$$N_t = N_o e^{\lambda t} + (r_f f_y/\lambda)(1 - e^{-\lambda t}). \quad [2]$$

A possible gaseous radioisotope of concern is ^{85}Kr, but as is easily calculated based on a $t_{1/2}$ of 10.73 yr, a standard fission product yield (f_y) from ^{235}U of 0.01317 (Walker et al., 1989), a fission rate r of 10^{20} fissions/day or ~10^{15} fissions/s, which converts to 5.5×10^{-6} Ci of ^{85}Kr/s, and an initial inventory (N_o) of 5.379×10^2 Ci, the inventory is largest when initially placed in the repository (Evans et al., 1994; Rechard, 1995b).

Neptunium-237 is a long-lived isotope ($t_{1/2}$ = 2.14×10^6 yr) that is of potential concern in a tuff repository (Rechard, 1995b), because it is mobile in very oxic solutions. However ^{237}Np is created by neutron absorption rather than as a fission product (i.e., created primarily by ^{235}U absorbing neutrons that first form ^{236}U and then ^{237}U, followed by beta decay to ^{237}Np). Assuming neutron flux conditions in the natural reactor are bound by the ATR spent nuclear fuel (which represents the highly enriched spent fuel category), then the increase in ^{237}Np can be bounded. For the ATR fuel of 72 MTHM, the ^{235}U was reduced from 93% to 82% (~7.9 Mg of ^{235}U was fissioned) and produced 17 Ci ^{237}Np out of a total ^{237}Np activity of 1264 Ci (Rechard, 1995b). The 10^{28} fissions assumed for a critical event in a repository represents ~3.9 Mg ^{235}U. Hence, taking the ratio, the ^{237}Np increase would be ~8 Ci or only 0.6%.

6.3 Increased Heat

When modeling the unmoderated, fast assembly conditions, the theoretical local temperature from a burst of thermal energy rose to between 400 and 900 K (Plaster et al., 1995). However, the thermal energy released from a moderated, slow assembly situation in unsaturated tuff would be much less because the fissile material would be at atmospheric pressure. Thus the maximum temperature before the criticality event stopped would be below 373 K in order to maintain the presence of the water moderator. This maximum temperature helps determine the rate (power) of approximately one criticality event per day as follows.

As mentioned previously, the criticality event may occur either at the container after removal of the boron, or outside the container in the volcanic tuff after transport and precipitation of the uranium. For the criticality event at the container, we assume the input must equal the radiative energy transfer from the surface of the cylindrical container through the air gap to the surface of the tunnel, which yields a steady-state power, q, of (Incropera and DeWitt, 1981, p. 655)

$$q = 2\pi r l\, \sigma\left(T_w^4 - T_t^4\right)\Big/\left[1/\varepsilon_w + (r/R)\left(1/\varepsilon_t - 1\right)\right] = 13\ kW \quad [3]$$

where

- r = radius of container (0.885 m)
- R = radius of tunnel (2.15 m)
- T_w = absolute temperature of waste container surface (373 K)
- T_t = absolute temperature of tuff (far-field temperature is 303 K)
- l = container length (5.1 m)
- σ = Stefan-Boltzmann constant (5.67×10^{-8} W/[m^2 K^4])
- ε_t = emissivity of oxidized carbon steel container at 100°C (0.8)
- ε_w = emissivity of welded tuff (0.8).

In reality, the container would be corroded and the fissile mass would be on the tunnel floor, but the approximation is consistent with the rough estimations. The thermal energy of 13 kW is ~10^{20} fissions/day (see App. B) and was used in Section 6.1 for the moderated, slow assembly when evaluating total fissions possible over 10^6 yr. This value of 10^{20} fissions is also consistent with the rate assumed for the Oklo natural reactors for evaluating the probability (i.e., 6 Mg/2×10^5 yr = 2×10^{20} fissions/day). Assuming 1.6 critical events ([6 events/ 800 MTHM] • 210 MTHM), to be consistent with Section 5.2.3, then the power from criticality in the repository is ~21 kW. For the hypothetical 12,060-MTHM repository, the thermal energy in the year 2030 would be 4000 kW. Hence, the 13-kW to 21-kW thermal power produced is at most 0.5% of the thermal power normally produced from radioactive decay 30 yr from the time at

which the waste was placed in the geologic repository and not sufficient, by itself, to significantly alter the unsaturated fluid flow near the repository. Therefore, the heat energy produced is well within that anticipated in the repository. Furthermore, the heat energy is not sufficient to create exotic conditions such as vaporizing fissile material to reach the initial condition claimed by Bowman and Venneri (1996).

The indefinite geometry for the situation of criticality in the volcanic tuff after transport and precipitation of the uranium makes the calculation of a steady-state power more difficult. Here we assume a point power source in water-saturated tuff. For steady-state, the power is determined by the radial distance from the source, where ambient temperatures are assumed. If a 40-m radius is assumed (100 times the radius of a 0.4-m "point source" of an approximately 6-kg uranium isotope mixture from the ATR fuel—see Fig. 1), then the power over 10^6 yr is ~20 kW, similar to that used above for a criticality in the repository tunnel (Carslaw and Jaeger, 1959; Rechard, 1995b, p. 13-1):[9]

$$Q = 4\pi r\, \Delta T \,/\, \text{erfc}\,[r/(4\alpha t)] \cong 20 \text{ kW} \qquad [4]$$

where

- r = radial distance to point of ambient temperature (40 m)
- ΔT = change in temperature from point source to ambient temperature (373 K - 303 K)
- α = diffusivity = $K/\rho C_p$
- K = thermal conductivity of tuff/water mixture (1.6 W/(m K) = $\phi K_w + (1 - \phi)\, K_t$ = 0.139 (0.68) + 0.861(1.7756))
- ρ = density of tuff/water mixture (2273 kg/m^3 = 0.139 (958.4) + 0.861(2485))
- C_p = specific heat capacitance (1325 J/(kg K) = 0.139(4217) + 0.861(858))
- t = time (10^6 yr).

This estimate of 20 kW for a 0.4-m-radius sphere is 1000 times greater than the estimated power density of 0.1 kW/m^3 at Oklo (Cowan and Norris, 1978) and thus likely very conservative.

6.4 Potential Physical Damage to Tuff

Noticeable physical damage to the tuff would require a rapid energy release per unit time (i.e., high power). The moderated, slow assembly is inherently limited in power in an unsaturated environment at atmospheric pressure because lack of water stops the nuclear reaction. Indeed, it was this inherent power limit caused by boiling water that was used to establish a maximum power of about 10^{20} fissions/day. A criticality from a sudden, gravity-assisted collapse of a partially corroded container could possibly release energy over a short time period similar to the aqueous and unmoderated, fissile metal accidents. The maximum energy release from prompt neutrons for the aqueous accident and calculated by Bethe-Tait analysis was 6×10^{17} fissions or 18.7 MJ (Section 5.1.5). This small amount of rapidly produced energy release per event (~4.5 kg TNT [~10 lbs] [Walker et al., 1989]) would be unlikely to cause any significant damage to the immediate rock 300 m (~1000 ft) under the surface.

This result can be shown by using an empirical equation developed for designing buildings that may experience explosive mishaps (Baker et al., 1980). The true radius of a potential spherical void (or camouflet) formed beneath the surface (the void minus the debris) from this explosion is

$$R_T = 1.053\, (W)^{0.252}\, d^{0.135}, \qquad [5]$$

where R_T is true radius (ft), d is depth below surface (ft), and W is weight of explosive (lbs). The weight, rather than the energy from the explosive, is used in the empirical equation because the exponent is so small (~1/4); the type of rock is not used for rough estimates because it has a secondary effect. The potential void radius is 1.4 m (4.8 ft)—more than the 0.9-m radius of the uncollapsed container but less than the 2.15-m radius of the original tunnel.

This estimated radius is larger than the radius obtained by scaling down from underground nuclear tests.[10] Specifically, assuming that damage occurs below a specific failure stress (Table IV) and then using this failure stress (σ), a material density (ρ), and a conservative estimate of the peak wave speeds (w_s) (~1/2 seismic velocity), the peak particle velocity (V_p) can be

9 If a 300-m radius is assumed, which is the maximum radius for a self-sustaining reaction near the container, and a repository depth of ~300 m, then the power is ~150 kW.

10 In using this scaling method, we do not mean to imply that we are now endorsing the nuclear explosion hypothesis (i.e., from a massive number of nuclear chain reactions in microseconds), which was discounted in Section 5.1.3. Rather, the scaling method allows us to use empirical data on explosive damage in tuff and demonstrates, regardless of probability, the low consequences of a small nuclear chain reaction ("critical excursion") whose energy output, inherently limited by the natural environment, is "miraculously" converted into kinetic energy rather than heat energy.

Table IV. Data and Results from Scaling Underground Nuclear Tests to 18.7 MJ of Energy

	Parameters			Results	
Material	Failure stress (σ_f) (MPa)	Density (ρ) (kg/m^3)	Peak wave speed (w_s) (m/s)	Peak particle velocity (V_p) (m/s)	Failure radius (r_f) (m)
Intact tuff	155	2219	1065	66	<0.5
Fractured tuff	24	1800 (80%)	750	16	0.8
Salt	70	2000	2050	17	1.3

calculated ($V_p = \sigma/[\rho \cdot w_s]$). Using this peak particle velocity, V_p, and a yield of 18.7 MJ, empirically obtained scaling of underground nuclear tests in salt and tuff (Crawford et al., 1974) yields an estimate of the radius of the failed material. For intact salt, it is ~1.3 m, as found using equation 5. For intact tuff, the radius is less than 0.5 m; for already disturbed tuff, it is 0.8 m–both less than the 0.9 radius of the container.

For salt

$$\log (R_s/E^{1/3}) = -0.4831 \log V_p - 1.705.$$

For tuff

$$\log (R_T/E^{1/3}) = -0.5034 \log V_p - 1.899$$

where

R_s or R_T = radius (m) for salt or tuff, respectively, E = yield (J), and V_p = peak particle velocity (m/s).

7.0 DISCUSSION

Because the bounding probability of a critical event for all but the explosive assembly was greater than 10^{-4}, we can say only that a criticality event is not easily dismissed through simple logical arguments. This situation occurs because the sequence of events (primary components of a fault or event tree) for a geologic disposal system is usually composed of complex natural processes. Evaluating the probabilistic effect of a natural process is difficult because, while the probability of a natural process occurring may be high (e.g., corrosion), whether it will induce adverse conditions that promote a critical condition is not easy to discern. Furthermore, the probability of natural processes occurring will change with time (e.g., climate change). Finally, the probability of a criticality changes spatially. Hence, the probabilities from the fault tree are both time and spatially dependent. These difficulties with the fault tree methodology were also noted for other aspects of probabilistic simulations of a repository in the late 1970s (Campbell et al., 1978) and explain why complex event and fault trees have not traditionally been used for performance assessments (Rechard, 1995a) and, instead, are used primarily for organizing thoughts.

Consequently, in Rechard (1995b), an effort was made to explore the underlying processes leading up to conditions that may be conducive to a self-sustaining nuclear reaction. In this manner, we could obtain a sense of the probability of a critical mass assembly indirectly by modeling phenomena relevant to the processes of assembly and then observing whether conditions conducive to a self-sustained nuclear chain reaction occurred. Given the design of the container, a critical condition can occur after disposal only if, first, the container (and any other protective layer such as cladding) corrodes and, then, either the boron is leached from the container, or the fissile material goes into solution and is transported from the vicinity of the boron. In the latter case, the fissile material must then be reconcentrated into a shape and mass to promote a sustained nuclear chain reaction. A mechanism to reconcentrate a purified fissile material is particularly difficult to envision, but only detailed modeling can provide an answer.

A possible drawback, however, of a detailed analysis of the alteration process of the container and fuel, and the amount of fissile material necessary to be assembled together, is that arguments are not succinct—fully comprehending such a detailed analysis requires a full understanding of the complex models used. In this regard, bounding calculations on the probability and consequences of a criticality as presented herein are useful because the arguments can be comprehended by a wider audience, although the argument is, perhaps, less definitive. In this study, the calculated bounding consequences are particularly useful in pointing out the small amount of fissions generated from critical events over a million years (~10^{28} fissions) in relation to the amount of fissions already represented by the spent nuclear fuel

itself (between 10^{30} and 10^{31} fissions). In short, concerns about a criticality on the surface where humans can be exposed either directly from the event or indirectly from cleaning up contaminated material do not apply to conditions in a deep, closed, geologic repository.

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APPENDIX A: CONCEPTS OF CRITICALITY AND RELATED TERMINOLOGY

To properly understand the results of these boundary calculations, which is that the consequence of a criticality occurring in an underground repository of radioactive waste is low, it is important to understand the concept of a critical condition. The following is a discussion of basic principles of a nuclear chain reaction based upon the discussion in Harper (1961, p. 5-9).

A.1 Nuclear Chain Reaction

Fission (splitting of atoms) of the nucleus of fissionable material (e.g., ^{235}U) may occur when the fissionable material absorbs a neutron. For example, after absorbing a neutron, ^{235}U will usually (~5 out of 6 times) split into other elements of unequal mass. In a fission reaction, more neutrons are emitted than are required to start it (e.g., fission of ^{235}U releases between 2 and 3 neutrons). If loss of neutrons is not too great, a chain reaction can build up. Loss of neutrons takes two forms: (1) loss of neutrons from the system and (2) sorption of neutrons by fissile material without the production of fission and sorption of neutrons by other material (Fig. A-1). In a small piece of fissionable material, one stray neutron will not start a nuclear chain reaction, even if a nucleus is fissioned, because the neutrons emitted by this initial fission will likely escape before starting another fission. In a larger piece of fissionable material, neutrons would have less chance of escape, and in an infinite block they could never escape (they could be lost only to sorption by elements that do not fission). With no loss of neutrons at all, and more than one neutron liberated per fission, the rate of fission increases until a change in physical conditions imposes a limit.

A.2 Multiplication Factor

As discussed above, in a large block of fissile material (such as ^{235}U), a nuclear chain reaction can occur whereby some of the original neutrons will produce second-generation neutrons, some third, a few fourth, and so on. Ultimately, allowing for all the generations, there may be many more fissions produced than neutrons initially present. To be precise, if n "stray" neutrons are introduced into the fissile material initially, and there are kn in the second generation, there will be k^2n in the third, k^3n in the fourth, and so on. In all, the total number of neutrons that would appear due to n initial neutrons is given by

$$\textit{total number of neutrons that would appear} = n \sum_{r=0}^{r=\infty} k^r . \quad \text{(A-1)}$$

If we can measure the neutron population in one neutron generation and again in the next generation, then the ratio of these numbers would be the multiplication factor, k. (In real systems, it is the effective multiplication factor, most commonly denoted as k_{eff}):

$$\textit{Multiplication factor}\left(k_{eff}\right) = \frac{\textit{Number of neutrons in one generation}}{\textit{Number of neutrons in preceding generation}} \quad \text{(A-2)}$$

Because the number of fission neutrons in any generation is proportional to the number of fissions during each generation, then k_{eff} corresponds to the ratio of fissions from generation to generation (and hence ratio of power between generations). Three possible situations exist for k_{eff}:

$k_{eff} < 1$ subcritical (zero or decreasing fission rate [power])

$k_{eff} = 1$ critical (constant, but could be at any fission rate [power])

$k_{eff} > 1$ supercritical (increasing fission rate [power])

If the fissile mass is just that size for which $k_{eff} = 1$, any neutron present in one generation will, on average, leave exactly one survivor in the next generation, and so on through the generations (assuming no additional neutron source). Thus any population of neutrons that happens to be present survives indefinitely, the number of neutrons remains constant, and the rate of fission continues unchanged. The assembly (configuration of fissile material) is said to be critical, and in this condition, remains in a steady state irrespective of the size of the initial population of neutrons (n). If fissile material is removed from the block, or k_{eff} becomes less than unity for any other reason (see Section A.4), each generation contains fewer neutrons than the preceding one, thus there is a limit to the total number of fissions and the reaction stops (i.e., if $k < 1$, the sum converges [Equation A-1] and is equal to n/[1 - k]).

Nuclear fission occurs very quickly (~10 nanoseconds for ^{235}U); thus the generations succeed each other swiftly. Unless k_{eff} is nearly equal to unity, the first few terms in the series contribute most of the sum (see Equation A-1), and the corresponding generations of neutrons are born and die in a short interval of time. If the multiplication factor k_{eff} is less than but not too close to 1.0,

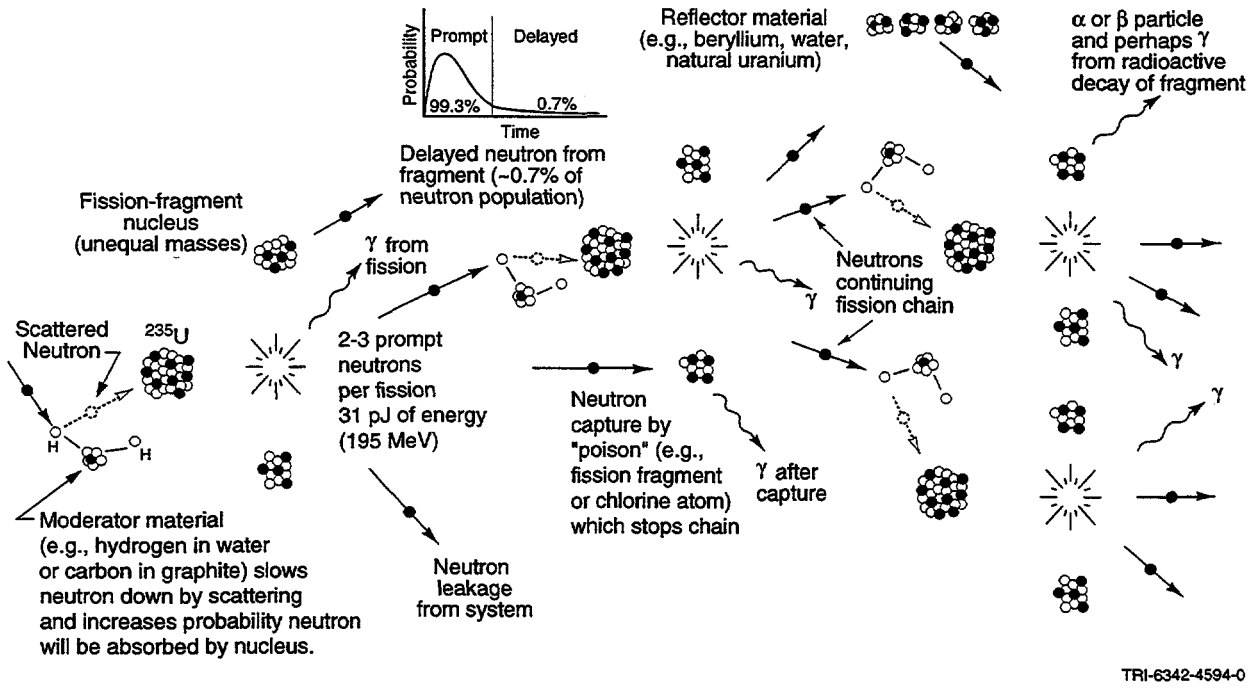

Fig. A-1. Schematic of ^{235}U fission chain reaction showing three generation of neutrons. The diagram assumes that each fission forms two fission fragments and two to three neutrons. The neutrons released as the nuclear fissions are "prompt" neutrons; the neutrons released from a few of the fission fragments are "delayed" neutrons. Although the delayed neutrons represent ~0.7% of the neutron population, they are very important to dynamic reactor analysis.

spontaneous fissions in the block will give rise to more fissions, but the total number will remain fairly small. Each spontaneous fission, and each arrival of a stray (e.g., cosmic ray) neutron, will provoke activity.

If more fissile material is added so that $k_{eff} > 1$, any activity already present, however small, can multiply indefinitely. As the interval between generations is small, the multiplication is rapid and the activity can rise rapidly. In such circumstances, however, the resultant liberation of heat quickly expands or breaks apart the supercritical mass (negative feedback), reducing k_{eff} below unity, and stopping the reaction before all the fissile material can be used up (see Section A.4). As described in Section 5.1.6.1, although $k_{eff} > 1$ is necessary for an atomic explosion, it alone is not sufficient. The rate of assembly must be fast enough to counteract negative feedback for a sufficient amount of time.

A.3 Reflection and Moderation to Promote Critical Conditions

In reactor design, materials are frequently placed around a nuclear reactor core containing the fissile materials to reflect back into the core many of the neutrons that would otherwise escape. Common reflector materials, which may also be moderators, in a nuclear reactor are graphite, beryllium, water, and natural or depleted uranium (Fig. A-1). Materials (neutron moderators) are also used directly in the reactor core to promote critical conditions by slowing down the high-velocity neutrons from the fission process because in fissile material (a subset of fissionable material, consisting primarily of ^{235}U and ^{235}Pu) slow (thermal) neutrons have a much greater probability of being absorbed by a nucleus. Common neutron moderators in a nuclear reactor are natural ("light") water, "heavy" water (water with significant quantities of the deuterium isotope), and graphite, because the hydrogen and carbon atoms of these materials slow down neutrons effectively through scattering while absorbing few of them (Fig. A-1).

A.4 System Feedback that Tends to Arrest Critical Conditions

Usually when $k_{eff} > 1$, the power and temperature (since most of the energy is heat) will rise until negative feedback becomes large enough to reduce the multipli-

cation effects to a subcritical condition. The most important negative feedback from temperature increase are (1) density decrease of fissile material from thermal expansion (which is important for metal systems, in which the neutron leakage from the assembly is increased), (2) thermal expansion of the moderator (which is a major feedback mechanism for dissolved systems in which fewer neutrons are slowed down), and (3) Doppler effects (since broadening of "resonance points" increases the probability that neutrons are absorbed by nucleus without fission). Included in the latter two mechanisms is desaturation of groundwater. If supercriticality ($k_{eff} > 1$) cannot be counteracted by these negative feedback effects, enough heat energy may be produced to melt the fissile material metal (assemblies) or boil the moderator (solutions), or enough heat energy may be converted to kinetic energy to break the assembly apart. As is argued in this report, the most likely criticality event is one that corresponds to a negative feedback mechanism in which the water moderator is removed, thus desaturating the porous media from within the remains of the waste container or a nearby fissile material precipitation zone. This is a very effective negative feedback mechanism, because a natural reactor within tuff would likely be at atmospheric pressure and not pressurized.

A.5 System Feedback that Promotes Critical Conditions

It is possible that the components of the system, such as the neutron moderator, can actually cause k_{eff} to increase momentarily as the power and temperature of the system rise. For example, positive feedback can occur when too much neutron moderator is present in the system that is already at $k_{eff} = 1$ such that removal of any of the moderator (e.g., boiling) increases k_{eff} to above 1. This instability continues until the system configuration changes. As noted in Section 5.1.1 of this report, Oklo natural reactors may have started from an overmoderated condition.

Those constructing the first atomic bombs flirted with the idea of using positive feedback (autocatalytic "method") to increase k_{eff} from ~1 to that needed for an explosion, but never pursued it because the scheme was found to require at least five times more material (and considerably more to achieve efficiency) than that used in other methods. More important, the fissile material could easily go critical prematurely, aborting a nuclear explosion (Serber, 1992). As noted in Section 5.1.3 of this report, earth scientists strongly criticize the theory that a natural system is able to avoid "premature" assembly to achieve critical conditions and yet attain conditions in which an atomic explosion may occur.

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APPENDIX B: ENERGY EQUIVALENTS

	^{235}U Fissions[a]	MJ	kW-hr	MWd	Btu	^{235}U Burn-up (kg)
Fissions	1	3.124×10^{-17}	8.679×10^{-18}	3.616×10^{-22}	2.961×10^{-14}	3.902×10^{-25}
MJ	3.201×10^{16}	1	0.2778	1.157×10^{-5}	9.478×10^{2}	1.249×10^{-8}
kW-hr	1.152×10^{17}	3.600	1	4.166×10^{-5}	3.412×10^{3}	4.496×10^{-8}
MWd	2.765×10^{21}	8.640×10^{4}	2.400×10^{4}	1	8.189×10^{7}	1.079×10^{-3}
Btu (IST)[b]	3.377×10^{13}	1.055×10^{-3}	2.931×10^{-4}	1.221×10^{-8}	1	1.318×10^{-11}
U^{235} Burn-up (kg)	2.563×10^{24}	8.006×10^{7}	2.224×10^{7}	9.266×10^{2}	7.859×10^{10}	1

(a) Calculated using the useful energy release value of 195 MeV/fission.
(b) International steam table

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