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Bounding Estimates for Critical Events When Directly Disposing Highly Enriched Spent Nuclear Fuel in Unsaturated Tuff

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This paper examines the possibility of criticality in a nuclear waste repository. The estimated probabilities are rough bounds 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). In addition, should a criticality occur in or near a container, the bounding consequence calculations showed that fissions from one critical event are quite small ($<\sim 10^{20}$ fissions, if similar to aqueous and metal accidents and experiments). Furthermore, a reasonable upper bound of total critical events of 10^{28} fissions corresponds to only 0.1% of the number of fissions represented by the spent nuclear fuel inventory in a repository containing 70,000 metric tons of heavy metal (MTHM) (the expected size for the proposed repository at Yucca Mountain, Nevada).

KEY WORDS: Criticality; nuclear waste repository; highly enriched spent nuclear fuel; risk assessment; performance assessment.

1. INTRODUCTION

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[1-3] several commentators have criticized the Bowman and Venneri calculations and called for a "... probabilistic risk assessment of the hypothesis,"[4] or stated that a "risk-based analysis of possible scenarios and neutron transport ... is needed."[5]

The overall risk-based analysis process for a nuclear waste disposal system is usually called a "performance assessment" (PA). Quantifiable measures from a performance assessment (like a probabilistic risk assessment or 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?[6,7] In a performance assessment, the answer to the first question is generated by characterizing the system and then identifying various scenarios. The scenario discussed is the assembly of a mass of fissile material in or near a waste container after disposal of highly enriched uranium fuel without treatment in an unsaturated volcanic tuff repository such that a critical condition occurs.[2] The second question is

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[2] A critical condition or event (commonly called "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 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 4.1.6, for a nuclear criticality to become a "crisis" (i.e., create an atomic explosion) the system must pass through the critical state extremely fast whereby the number of neutrons increases faster than the fissile material can expand due to the heat energy released and thereby allows massive numbers of chain carrying neutrons to be produced.

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answered by estimating probabilities, based on empirical data, for assembling a critical mass in various configurations. Similarly, the answer to the third question is provided from estimates of consequences based on empirical data.

1.1. Previous Work

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,[8] and the international community has included it since 1981 in its generic list of events to consider.[9] In 1979, Clayton[10] speculated on a positive feedback mechanism for a critical event in trenches at the Hanford site based on criticality calculations. In 1983, Stratton[11] dismissed allegations that a nuclear explosion had occurred at a waste disposal site in the Ural mountains in Russia. More recently, commentors 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.[5,12]

1.2. Disposition Options for 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 paper pertains to direct disposal without treatment of the highly enriched uranium spent nuclear fuel category; however, some of the results qualitatively apply to other waste categories. Prudent design features are included to preclude a criticality during storage and transportation, and immediately after disposal, such as including borated stainless steel in the container and mixing of containers such that containers with highly enriched uranium spent fuel are not placed next to each other. Other design features include (a) containers with long average design life, (b) containers spaced close enough so that heat generated by the decay of radioisotopes heated the rock such that sufficient vaporization occurred to keep the containers dry for several hundred years, and (c) locating the disposal region for the U.S. Department of Energy (DOE) waste in an unsaturated zone of volcanic tuff similar to that proposed for commercial, low enriched spent nuclear fuel at Yucca Mountain, Nevada.

Other assurance measures that have been or will be 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 (e.g., naturally occurring phosphate minerals with adsorbed rare earth metals), (2) limiting fissile mass in each individual container,[13] (3) using a salt repository, (4) surface storage of the spent fuel assemblies, (5) 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 (6) mixing of the spent fuel with depleted uranium.[14] If these 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 a level of assurance for the disposal system itself.

1.3. Analysis Overview

In the following description of the analysis, the discussion is organized around answers to the three questions of a performance assessment: (1) system characterization and scenarios (i.e., collections of features, events, and processes of interest), (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. The assembly process and its consequences were investigated because of their interrelationship. 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. (The use of detailed calculations to fully simulate the evolution of the disposal system as the container degrades, which is another approach to determine whether conditions exist under which a criticality could occur, is described elsewhere).[15,16]

2. TYPE OF NUCLEAR WASTE AND DISPOSAL SYSTEM STUDIED

2.1. Highly Enriched Uranium Spent Nuclear Fuel

The DOE has about 1400 m^3 or 2700 metric tonnes heavy metal (MTHM) of spent fuel for disposal.[14,15] Of

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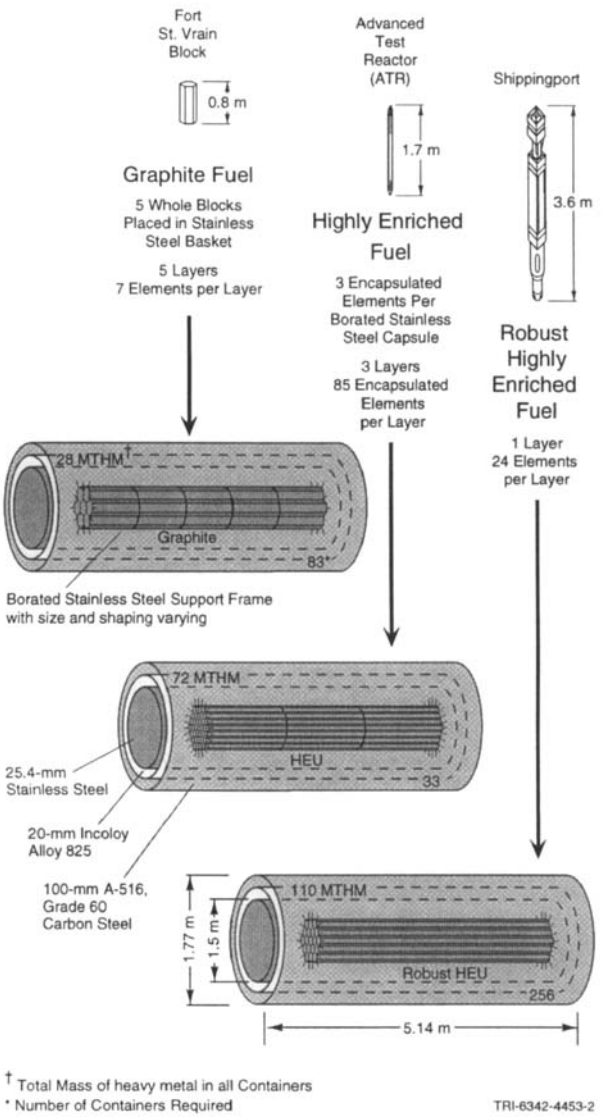


Fig. 1. Proposed packaging of the three categories of highly enriched uranium spent nuclear fuel in 25.4-mm-thick stainless steel multi-purpose canisters that are then overpacked with 20-mm-thick Incoloy 825 and 100-mm-thick carbon steel containers.[14]

this spent fuel, about 210 MTHM (8%) was originally highly enriched (i.e., of the 210 MTHM mass, more than 20% is fissile uranium isotope ^{235}U). The many types of highly enriched fuel were placed in three categories for this study: (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. Uranium–aluminum metal alloy advanced test reactor (ATR) fuel with aluminum cladding represented the chemically reactive spent fuel category. The zircaloy-clad, uranium-dioxide Shipping-port fuel represented the robust highly enriched fuel category. The inventory of fuel from Navy propulsion reactors is included in this category.

2.2. Other DOE 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 in 296 containers; and (3) 9268 MTHM (equivalents) of high-level vitrified sludge waste from fuel reprocessing within the DOE complex in 1880 containers.

2.3. Containers

The container for the highly enriched fuel was a multipurpose canister of 304L stainless steel, 25.4 mm thick, 5.14 m long, and 1.77 m in diameter. This handling canister was 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 filled with water, the container design used borated stainless steel as the support structure for the spent nuclear fuel to absorb neutrons that otherwise could contribute to a nuclear chain reaction.

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 accounts for this functional dependence;[15] here, however, the distribution of rates in the presence of free oxygen (oxic conditions), 26°C (ambient) and 100°C, at 100% saturation, is used (Fig. 2).

2.4. Repository Layout and Waste Emplacement

The disposal region of the repository for DOE waste was sized to accommodate 12,060 MTHM of spent fuel or equivalent high-level waste. The repository design consists of long rooms (''tunnels''), 7.65 m in

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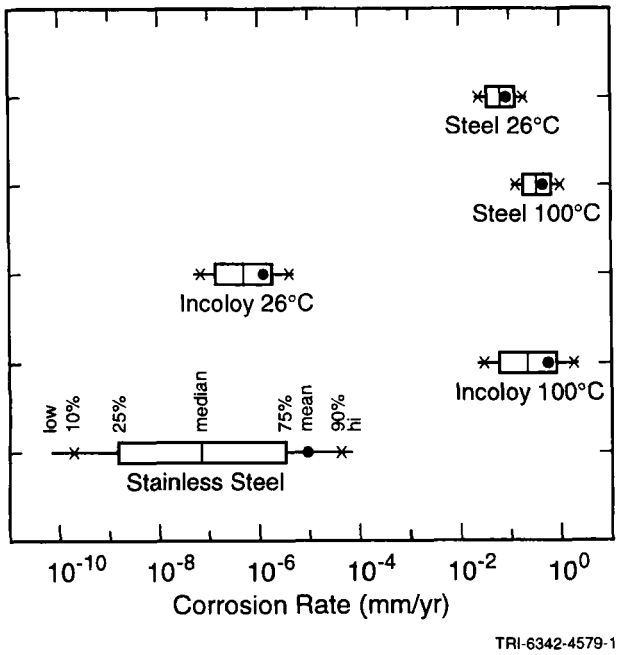


Fig. 2. Corrosion penetration rates of carbon steel, Incoloy 825, and stainless steel at 100% water saturation for oxic conditions (water in equilibrium with air with 21% oxygen) and a temperature of 26°C and 100°C (Fig. 2 in Ref. 14).

diameter, that are surrounded by and connected to access drifts, which are in turn connected to ramps that lead to the surface. The large waste containers are assumed to be parked horizontally, end to end with no gap, in the tunnels for disposal. At emplacement, neutron interaction between containers does not occur because sufficient material exists at the ends of the container.[15] The tunnels were spaced 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.

2.5. Unsaturated, Volcanic Tuff Geologic Barrier

Based on previous modeling experience,[15,17] 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 (Fig. 3). The repository is assumed to be placed 286 m below the surface and 333 m above an aquifer. The average intact matrix porosity of the 333-m-thick tuff surrounding the repository (TS modeling units) is ~0.14.[17] The average matrix porosity of the entire column of tuff used in the study is ~0.19 (Fig. 4), but the average and maximum intact matrix porosities of various layers deviate from this average (Table I).

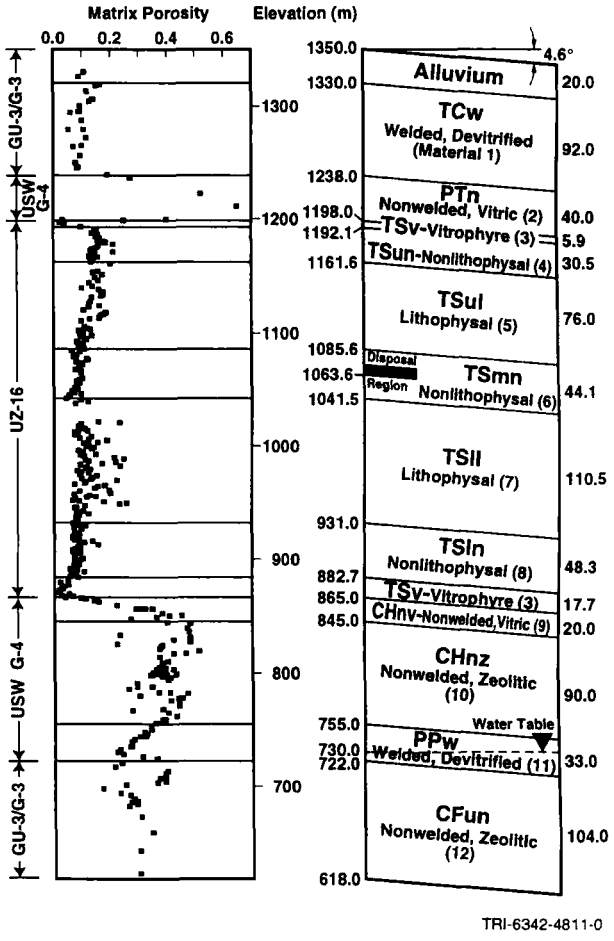


Fig. 3. Idealized stratigraphic column of Yucca Mountain tuffs defined by similar porosity (Fig. 4 in Ref. 14).

3. SCENARIO DEVELOPMENT

3.1. Scenario Development with Criticality as an Event

Three events were judged to be important in summary scenarios for the first 10,000 yr: basaltic volcanism

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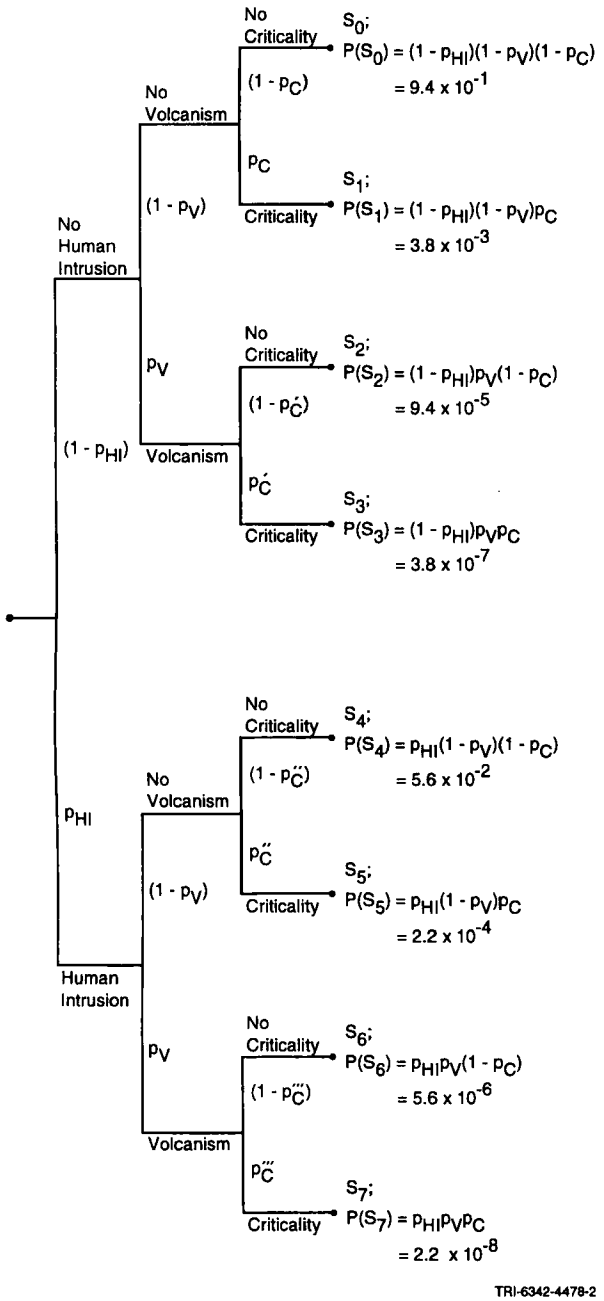


Fig. 4. Logic diagram to enumerate summary scenarios, S_j, for first 10,000 yr of repository performance (Fig. 6 in Ref. 14). See text for numerical values of event probabilities, p_{HI}, p_V and p_C.

(V); human intrusion by exploratory drilling (HI); and criticality (C) (Fig. 4). All other events, processes, and features were treated as either certainties or impossibilities. The elementary probability of basaltic volcanism near Yucca Mountain, Nevada, in the first 10,000 yr was

Table I. Matrix Porosity for TSmn, TSll, and CHnz

Modeling layer	Matrix porosity		Water mole fraction[a]
	Average	Maximum	
TSmn	0.085	0.12	0.16
TSll	0.12	0.26	0.33
CHnz	0.35	0.50	0.58

[a] The conversions to water mole fractions assume that the pores are filled with water and that the matrix is *pure cristobalite* (a form of silica).

conservatively taken as $\sim 1.0 \times 10^{-4}$ for the "hot repository."[17] The elementary probability of at least one inadvertent human intrusion in the first 10,000 yr was taken as $\sim 5.6 \times 10^{-2}$.[14] The probability of at least one inadvertent human intrusion, $P\{N > 0\}$, and one volcanic event is an exponential distribution function. As an example for exploratory drilling, a drilling rate (λ) of three bore holes/area of waste (km^2)/10,000 yr as recommended in Appendix C of 40 CFR 191, i.e., $P\{N > 0\} = 1 - e^{-\lambda\Delta t}$, was used in the Poisson distribution. The 10,000-yr probability of a criticality is calculated in Section 4.3 and is $\sim 4 \times 10^{-3}$. The resulting probabilities of the summary scenarios (Fig. 4) show that scenarios S_3, S_6, and S_7 can be neglected by virtue of Environmental Protection Agency (EPA) guidance in Appendix C of 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. [Also 10,000 yr is the period of interest according to the EPA's current regulation, 40 CFR 191, but is likely to increase to one million years (1 Ma)].[18]

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 critical 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. More important, the National Academy of Sciences (NAS) suggests that probabilities not be assigned to the human intrusion release mechanism.[18]

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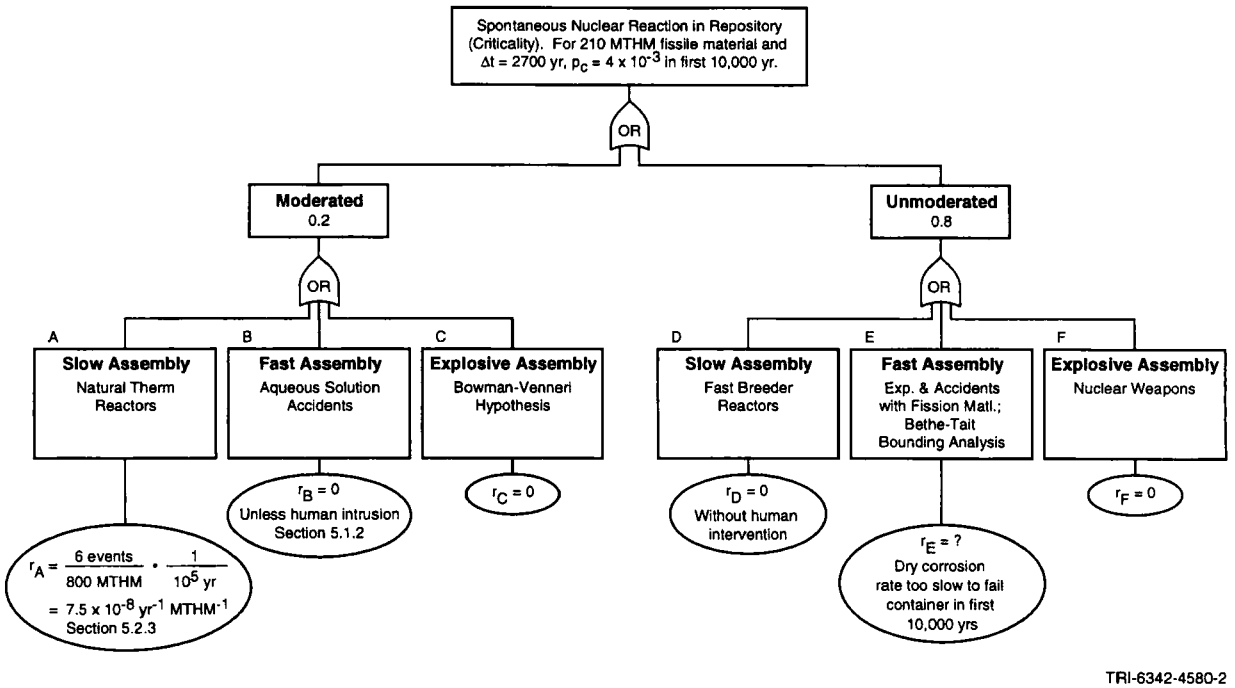


Fig. 5. Fault tree to evaluate probability of a criticality event, showing that the branch with high moderation and slow rate of assembly is most probable. Similarities with natural or engineered system and possible analysis techniques are also noted (Fig. 9 in Ref. 14).

3.2. Categories of Criticality Event

For bounding probabilities and consequences, the criticality event[3] was divided into two basic conditions: (1) moderation (e.g., sufficient or insufficient neutron moderating material[4] 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) (Fig. 5). The first condition categorizes the most important feature regulating the operation of an assembly in the natural environment (e.g., water moderator, see Section 4.1.1). The second condition categorizes the time required [and thus power (energy/time) expanded] to assemble the critical mass. Examples of the situations follow that are relevant to a criticality in a repository and can be used to bound the consequences of a critical condition. All criticality event categories, except possibly explosive assembly, have similar consequences.

[3] To more precisely describe the critical event, a multiplication factor is mathematically defined as follows: If n "stray" neutrons are introduced into the fissile mass initially, and there are kn in the second generation, there will be k^2n in the third, and so on. In all, the total number of neutrons that would appear due to n initial neutrons is given by $n \sum_{r=0}^{r=\infty} k^r$. If we can measure the neutron population in one neutron generation and again in the next generation, then the ratio of these numbers is the effective multiplication factor, k_{eff}. Three possible situations exist for k_{eff}: $k_{eff} < 1$, subcritical (zero or decreasing fission rate [decreasing power]); $k_{eff} = 1$, critical (constant fission rate, but could be at any fission rate [constant power]); and $k_{eff} > 1$, supercritical [increasing fission rate (increasing power)].

[4] Neutron moderators slow down the high-velocity neutrons from the fission process through scattering while absorbing few of them and thereby promote critical conditions [slow (thermal) neutrons have a much greater probability of being absorbed by a nucleus of fissile material]. Common neutron moderators are natural ("light") water, "heavy" water (water with significant quantities of the deuterium isotope), and graphite.

4. BOUNDING PROBABILITY OF CRITICAL EVENT

In this paper, we estimate a bounding probability from information on analogous events and heuristic arguments regarding the probability of subevents of the critical event.

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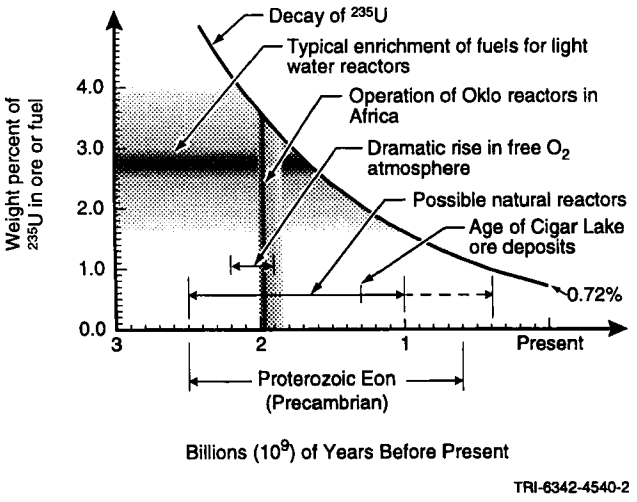


Fig. 6. Operation of natural, pressurized reactor in Oklo uranium deposit in Gabon, Africa, occurred about 2 billion years ago when weight percent of ^{235}U was above three (Fig. 8 in Ref. 14).

4.1. Probabilities of Criticality Conditions

4.1.1. Moderated, 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 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,[19] the FA formation was formed as a deltaic deposit about 2.1×10^9 years ago (2.1 Ga), during erosion of the Archean-age basement rock (2.7 Ga); the FA formation is the likely source of the uranium. The FB formation was formed during subsidence 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 included faulting and uplift of the basin. Tectonic faulting and hydrofracturing of the sandstone by highly geopressurized fluids enhanced the primary porosity and permeability of the rock allowing hydrocarbons and water to migrate through the FB formation to fault traps. Massive amounts of meteoric (oxygen-rich) waters in the wet climate zone circulating through the conglomerates in the FA formation dissolved and transported uranium. High concentrations of uraninite (UO_2) (>20%) were precipitated as this uranium-rich water came in contact with the fault-trapped hydrocarbons 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 17,300 Mg Oklo ore deposit that operated as natural reactors about 1.97 Ga, when natural uranium had a ^{235}U content of ~3.68% (640 Mg of ^{235}U) (Fig. 6). The first six natural reactor regions are typically 10–20 m in length and width, and less than 1 m thick. Pressurization of the water (30–40 MPa) from geologic conditions was such that the temperature ranged between 440–640 K in most of the reactors (some may have operated close to the surface and near atmospheric pressure).[20] The content of ^{235}U (uranium enrichment) and temperature are similar to a modern, engineered, pressurized water reactor (Fig. 6). (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 the pressurized reactors at Oklo).

Because the optimum water content of the reactor was low (12–15% by volume, 6% by weight),[21] several of 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 from negative feedback such as when the water (which served as a moderator for the assembly) expanded near its critical point[22] 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.[5] The convective circulation of water heated by the natural reactor continued to concentrate the ore around the reactor (up to 60%), altered the hydrocarbons to form bitumen that enclosed some natural reactors, and dissolved quartz in the sandstones, causing compaction in the reactor zone.[23] Reactor analysis indicates that sustained reactor operation depended on the ability to increase the

[5] Usually when $k_{eff} > 1$, the power and temperature (since most of the energy is heat) will rise until negative feedback becomes large enough such that $k_{eff} < 1$. Negative feedback effects from temperature increase include (1) density decrease of fissile material from thermal expansion (important for metal systems, because the neutron leakage from the assembly is increased), (2) thermal expansion of the moderator (a major feedback mechanism for dissolved systems in which fewer neutrons are slowed down), and (3) Doppler effects (i.e., broadening of "resonance points" that increase the probability that neutrons are absorbed by a fissile nucleus without fission). Desaturation is included in the latter two mechanisms and would be very effective because a natural reactor within tuff would likely be at atmospheric pressure and not pressurized.

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amount of moderating water and transmute any neutron absorbers (''burnable poisons'') to compensate for creation of fission products that absorbed neutrons.[21,24] The reactor behavior continued for about 2×10^5 to 8×10^5 yr,[25,20] 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. For the first six reactors, researchers have estimated that 6 MTHM (6.6 short tons) of ^{235}U was consumed out of ~800 MTHM (880 short tons) of high grade uranium.[20,22] Consumption of 6 MTHM corresponds to $\sim 10^{28}$ fissions or ~15,000 MW-yr. Although up to 3 MTHM (3.3 short tons) of ^{239}Pu was also produced in the natural reactor[22] from neutron absorption by ^{238}U, very little fissioned (2–6%).[24] Most decayed to ^{235}U; a large portion of this ^{235}U inventory was then subsequently fissioned.

If 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 since the media is similar (i.e., 84% versus 74% SiO_2, 10% versus 8.5% porosity, and media is fractured), 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 paper (Section 4.2).

4.1.2. Moderated, 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)[14,26] or moderated metal and metal-oxide system accidents and experiments.[27] 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, 18.7 MJ, or burnup of ~0.23 mg ^{235}U). Furthermore, the maximum number of immediate fissions is also only about 10^{18} (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.

4.1.3. Moderated, Explosive Assembly

A moderated, explosive assembly can be represented by one of the events that was recently hypothesized by Bowman and Venneri.[1] Bowman and Venneri 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. Herein, assembly of a critical configuration can occur either from fissile material ''coming together'' or from material diffusing ''out'' into the moderator.

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 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; in Situation 4, 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 it is 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 0.16–0.26 water mole fractions that are considered physically possible in the lithophysal and nonlithophysal tuff near the repository horizon.

Critics of the Bowman and Venneri analyses focus 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 vs. a pure silica system, (2) whether a positive feedback mechanism that exists briefly in a fissile mass implies a

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nuclear explosion, and (3) how much energy is potentially released from a positive feedback mechanism. In answering the first concern, volcanic tuff, far from being pure silica, contains several elements that can significantly absorb neutrons. Also, the assembly is a heterogeneous mixture that 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[1] is required to go critical.[28] 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 4.1.6).[11] 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.[29] 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 credible explanation is given for how a natural system moves from just critical at k_{eff} = 1 to k_{eff} = 1.1, an excess reactivity of ~$14 in a ^{235}U system). For example, the porosity of the tuff matrix is too great to confine the critical assembly, thus virtually all of the energy generated would be heat rather than kinetic energy.[28,30] Finally, there is no natural analogue, accident, or experiment that can be cited as an example of a system, which starts at near critical ($k_{eff} \cong 1$), and then has a nuclear explosion initiated solely as the result of positive feedback.

Critics of the assembly process are concerned with its improbability because of geologic concerns. 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 Bowman and Venneri also show 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 requires the system to pass through a possible Oklo-like situation requiring less fissile mass.[6]

A similar problem 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.[3] 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. Thus a new mechanism must then be invoked (''water-steam explosions''), but it is not a credible mechanism to reach the hypothesized *dry* conditions.[5] In essence, the conditions necessary to produce the explosion hypothesis are inconsistent with the conditions necessary to produce the initial conditions for the assembly.[31]

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.

4.1.4. Unmoderated, Slow Rate of Assembly

Unmoderated corresponds to assemblies that contain little or no water or any other moderating materials, i.e., elements such as hydrogen, carbon, and beryllium,

[6] 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 4.1.1, some 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.[34] As noted here, earth scientists strongly criticize the theory that a natural system is able to avoid criticality ''prematurely''.

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which have a high probability of scattering neutrons while simultaneously not absorbing many neutrons. This situation would create a "hard" critical nuclear assembly, i.e., neutrons with a high energy spectrum. 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.

4.1.5. Unmoderated, Fast Rate of Assembly (Experiments with Fissionable Materials)

An unmoderated, fast rate of assembly 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}), and would likely correspond to accidents and experiments carried out at Los Alamos National Laboratory (e.g., Godiva Reactor).[32] Some of the accidents yielded between 10^{16} and 10^{17} fissions. "Bethe-Tait" bounding calculations for evaluating the safety of two fast reactors yield upper bounds of 10^{19} fissions for assemblies with a large mass of fissionable material. 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. 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.[33]

4.1.6. Unmoderated, Explosive Assembly

Unmoderation, 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. Although a $k_{\text{eff}} > 1$ is a necessary condition for an atomic explosion, it alone is not sufficient. 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 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 are injected precisely at the same time that two subcritical amounts of ^{235}U are explosively brought together to form a supercritical mass.[34] (This design is not possible with ^{239}Pu because stray neutrons from ^{240}Pu prevent it.)

4.2. Fault Tree for Moderated, Slow Rate of Assembly

Fault trees have been used since 1963 to evaluate the probability of failures within complex human-engineered systems.[31] The likelihood of the assembly of fissile material is estimated here by 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. 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 (Fig. 5). Because of the proposed design for the multipurpose 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. 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.

4.2.1. 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 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

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provide enough water to transport and separate the boron and uranium, and thus allow a critical event; hence, they are not discussed further.

Analysts currently assume that the repository at Yucca Mountain would be dry initially or that containers would not be placed near existing seeps. Hence, for corrosion to occur from fractures that drip water, new seeps would have to appear. Possible sources for seeps include increased precipitation at the site, as the result of changes in climate; the water would infiltrate fractures and intersect the repository. Although the probability of this increased precipitation is unknown, we here 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 2.5), 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. In this paper, 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.[35] Although the spacing of the potentially wet fractures is not known, we here used ~25 m, which is a typical spacing of wet fractures in the E and O tunnels, located under Rainier Mesa.[36] 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 and a fully immersed local penetration rate of 2 mm/yr at 100°C for Incoloy 825 (Fig. 2). (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,[15] but in bounding calculations the container life is already very short and cannot significantly influence the probability of a critical event.

4.2.2. Physical Removal of Boron

Once the container has been penetrated, the uranium and/or boron can be transported into the host rock. However the rates of removal for uranium and boron may differ. During production of borated stainless steel, the molten boron is not very soluble and forms precipitate borides, frequently chromium boride, within the stainless steel. In general, these metal borides are very hard, refractory, and chemically inert because of the strong covalent metal–boron bond. Because of this bond, the bulk stainless steel may corrode faster than the borides, which may 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 than the low soluble uranium depends on the amounts of water and oxygen present. In this section, the quantities of water and oxygen are not evaluated and so either boron or uranium transport has to be assumed.

Time for boron removal is estimated 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.[7] Here, the boron release is assumed to be congruent with the generalized corrosion of the stainless steel, which is 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, the reported fully immersed, generalized corrosion rate at 100°C is about 0.1 microns/yr; this is likely near the high end of possible corrosion rates (Fig. 2). About 7000 yr would be required to corrode all of the stainless steel at this maximum rate.

4.2.3. 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 the reactor zones in the Oklo ore deposit 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. The lower bound is possibly the minimum operating life of the reactors, about 2×10^5 years (see Section 4.1.1). 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 [i.e., 6 events/(800 MTHM $\cdot$ 2×10^5 yr)] which gives an indication of the efficiency in

[7] "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 stray neutrons from starting a chain reaction.

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nature of forming a critical mass in an area ideal for forming an ore deposit.

Given the heuristic arguments described above, conditions conducive to a sustained nuclear chain reaction could occur in time. The probability of occurrence is conditional on the container being under a dripping fracture and more infiltration occurring through Yucca 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%. Figure 6 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} (i.e., $0.2 \cdot 210$ MTHM $\cdot 3.75 \times 10^{-8}$ events/yr/MTHM).

Next, we determine the probability of criticality in the first 10,000 yr for consistency with the probabilities cited for the volcanism and human intrusion. The probability model is based on a failure-rate function[38] 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

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

Here $r(t)$ is a constant and equal to 1.6×10^{-6} per year. 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 4×10^{-3}, which was used in evaluating probabilities of scenarios (Section 3, Fig. 4). The probability evaluated by this fault tree applies primarily to the "no intrusion, no volcanism" branch in the event tree (Fig. 4). A fault tree, applicable to each major condition of the event tree (in Fig. 4) is needed; however, each fault tree provides only an order of magnitude number that does not change substantially and so they will not be shown here.

To evaluate the probability of the alternative situation—uranium migrating away from the container and into the tuff—the criticality incidence is estimated from Oklo and uranium solubility and possible flow rates of water through the repository are used to determine a hazard rate ($r(t)$). The solubility of uranium (S_U) in tuff has been measured at 10^{-2} mM.[37] A flow rate of 15 m^3/yr can be estimated from a 25-m spacing of wet fractures and an infiltration rate of 24 mm/yr, which occurs at Rainier Mesa[35] (equivalent to complete desaturation ~87 m above the Yucca Mountain repository). The corresponding hazard rate ($r(t)$) is 2.6×10^{-7} yr^{-1}. The probability in the first 10,0000 yr from Eq. (1) is 2.5×10^{-3} when integrating from 300 to 10,000 yr with assumed instantaneous release of uranium after corrosion of the container. This probability is less than that estimated for a criticality at the container and so was not used in the event tree. Also, the probability bound is likely high because criticality away from the container requires a reconcentration of uranium (because the solution is not critical), which is inherently less probable (ore bodies are not ubiquitous in the earth's crust). The above arguments can be extended to plutonium with a solubility of $\sim 10^{-5}$ mM.[17] This calculation results in a three order of magnitude drop in the rate, which, in turn, reduces the probability to less than 10^{-4} over 10,000 yr. Such a low value removes the criticality event with migration of plutonium away from the container from further consideration, according to guidance of 40 CFR 91. However, 40 CFR 191 no longer applies to the Yucca Mountain repository and the period of interest will likely increase to 10^6 yr.[18]

5. BOUNDING CONSEQUENCES FOR SLOW AND FAST RATES OF ASSEMBLY

In the above presentation, explosive assembly was argued to have probability zero. However, slow and fast rates of assembly of uranium 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. 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. A surrogate for these two consequence categories, bounds on total fissions, is discussed first, followed by discussion of direct consequences.

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5.1. Bound on Total Fissions

Both moderated and unmoderated fast rates of assembly release similar amounts of energy, 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 burnup of 40 GWd/MTHM would represent on the order of 10^{31} fissions, and the 12,060-MTHM DOE portion of the repository would represent on the order of 10^{30} fissions. Clearly, an increase in fissions from one critical event of 10^{20} would be negligible without any appreciable increase in radioisotopes in comparison to the original inventory (see Section 5.2).

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 5.3). One critical event occurring every day for 10,000 yr would amount to $\sim 10^{26}$ fissions, and one critical event occurring every day for one million years would amount to $\sim 10^{28}$ 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-MTHM fissile contents of all 372 containers of highly enriched fuel undergo a burnup of 40 GWd/MTHM (which corresponds to a fission of ~4.3% of the ^{235}U atoms in commercial PWR fuel); this situation would result in $\sim 10^{28}$ fissions. Alternately, using the fact that six events at Oklo occurred in 800 MTHM, consuming 6 MTHM, 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 first six Oklo reactors (Section 4.1.1).

5.2. Increased Radioisotope Inventory

A sustained nuclear chain reaction from uranium would increase the radioisotope inventory, but only by a very small amount, as follows. Neptunium-237 is a long-lived isotope ($t_{1/2} = 2.14 \times 10^6$ yr) that is of potential concern in a tuff repository,[15] because it is mobile in oxic solutions. Neptunium-237 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 (Fig. 1), 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.[15] The 10^{28} fissions assumed for a critical event in a repository represents ~3.9 MTHM ^{235}U. Hence, taking the ratio, the ^{237}Np increase would be ~8 Ci or only 0.6%.

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

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

A possible gaseous radioisotope of concern is ^{85}Kr, but as is easily calculated based on a $t_{1/2}$ of 10.73 a standard fission product yield (f_y) from ^{235}U of 0.01317,[39] a fission rate r of 10^{20} fissions/day or $\sim 10^{15}$ 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.[15]

Consequently, the influence of criticality from ^{235}U on the inventory of fission products is minor when measured directly, as in this section, or indirectly as total fissions, as in the previous section. The influence of criticality from ^{239}Pu on the inventory is similar. In addition, because the fission products have fewer health effects 100 years following criticality than the products present prior to criticality (Table II), criticality is actually beneficial as measured by the EPA standard. Provided the relative hazards of plutonium versus the fission products are correct, a health benefit occurs when transmuting plutonium in a particle accelerator, when burning plutonium in a nuclear reactor, or when plutonium criticality occurs underground in a repository. The primary difference is the efficiency of the transmutation, which is likely far less efficient in the repository although the possibility of direct human exposure is practically zero.

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Table II. Summed EPA Units vs. Time Based on Release Limits (Surrogate of Health Risk) in 40 CFR 191 for Fission Products of Initially One EPA Unit of ^{239}Pu

Time (yr)	Summed EPA units of ^{239}Pu or fission products[a]
0	1.0000
0.0003	11.1790
10	8.8538
100	1.1047
110	0.8794
1000	0.0004
10000	0.0003

[a] Release limit (Ci) = 100, halflife (s) = 7.594×10^{11}, mass (kg) = 1.60091.

5.3. 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.[33] 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[40]

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

where r is radius of container (0.885 m); R is radius of tunnel (2.15 m); T_w is absolute temperature of waste container surface (373 K); T_t is absolute temperature of tuff (far-field temperature is 303 K); l is container length (5.1 m); σ is Stefan–Boltzmann constant (5.67×10^{-8} W/(m^2 K^4)); ε_t is emissivity of oxidized carbon steel container at 100°C (0.8); and ε_w is 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 $\sim 10^{20}$ fissions/day and was used in Section 5.1 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 $\times$ 10^5 yr = 2 $\times$ 10^{20} fissions/day). Assuming 1.6 critical events [(i.e., 6 events/800 MTHM) · 210 MTHM], to be consistent with Section 4.2.3, 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 at 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.[1]

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 10-m radius is assumed (10 times the typical thickness of the Oklo lenticular reactors), then the power over 10^6 yr is ~7 kW similar to that used above for criticality in the repository:[41]

$$Q = 4\pi K r\, \Delta T/erfc[r/(4\alpha t)^{1/2}] \simeq 4\pi K r \Delta T = 7\ kW \quad (4)$$

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

This estimate of 7 kW for a 0.5-m-radius sphere is 100 times greater than the estimated power density of 0.1 kW/m^3 at Oklo[22] and thus likely very conservative.

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5.4. 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 was 6×10^{17} fissions (18.7 MJ) (Section 4.1.5). This small amount of rapidly produced energy release per event (~4.5 kg TNT [~10 lb]) would be unlikely to cause any significant damage to the immediate rock 300 m (~1000 ft) under the surface. Confidence in this result can be gained by using an empirical equation developed for designing buildings that may experience explosive mishaps.[42] 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 (lb). 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.[14] 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.

6. DISCUSSION

Because of their ability to be comprehended by a wider audience, bounding calculations on probability and consequence are used to explore the potential of a critical configuration developing in or near a container of highly enriched uranium spent nuclear fuel disposed without treatment in an unsaturated volcanic tuff repository. These bounds also apply to plutonium disposal in some cases. Except for explosive assembly,the possibility of criticality in the first 10^4 yr cannot be easily dismissed (i.e., $<10^{-8}$ yr^{-1}) through simple logical arguments using general information. (However, plutonium criticality away from the repository can be dismissed because of low solubility.) The same difficulty occurs for the first 10^6 yr—the likely period of interest for the new regulations that will apply to Yucca Mountain.[18] The 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 probabilities of a natural process is notoriously 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) and spatially. Therefore, detailed, site-specific calculations are needed to explore the underlying processes leading up to conditions that may be conducive to a self-sustaining nuclear reaction.

However, the 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 (10^{31} fissions for a 70,000-MTHM repository). These fissions are an indirect metric of such phenomena affecting human health risk such as the increase in radionuclide inventory, heat generation, and potential physical damage to the host rock. Estimates of direct metrics confirm this finding: the radionuclide inventory does not greatly increase (e.g., <1% for ^{237}Np from fissioning of ^{235}U) and the health hazards decrease (e.g., summed EPA units decreased from original inventory after 110 yr from fissioning of ^{239}Pu); the heat produced from criticality of ~13 kW is at most 0.5% of the thermal power normally produced from radioactive decay; and the zone of potential damage is less than that caused by the placement of 2-m tunnel. In short, general concerns about nuclear criticality on the surface where human exposure can occur either directly from the event or indirectly from the cleanup do not apply to conditions in a deep, closed, geologic repository.

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REFERENCES

1. C. D. Bowman and F. Venneri, "Underground Supercriticality from Plutonium and Other Fissile Material," *Sci. Global Security* **5**, 279–302 (1996).
2. W. J. Broad, "Scientists Fear Atomic Explosion of Buried Waste," *New York Times*, March 5, 1995, p. 1, Column 1 (1995).
3. G. Taubes, "Blowup at Yucca Mountain," *Science* **268** (5219), 1826–1839 (1995).
4. L. Geppert, "Showdown at Yucca Mountain," *IEEE Spectrum* **32** (5), 19 (1995).
5. R. A. Van Konynenburg, "Comments on the Draft Paper 'Underground Supercriticality from Plutonium and Other Fissile Material,' written by C. D. Bowman and F. Venneri (LANL)," *Sci. Global Security*, **5**, 303–322 (1996).
6. S. Kaplan and B. J. Garrick, "On the Quantitative Definition of Risk," *Risk Anal.* **1** (1), 11–27 (1981).
7. J. C. Helton, M. G. Marietta, and R. P. Rechard, "Conceptual Structure of Performance Assessments Conducted for the Waste Isolation Pilot Plant," in *Scientific Basis for Nuclear Waste Management XVI, Materials Research Society Symposium Proceedings, Boston, MA, November 30–December 4, 1992*, Vol. 294 (Materials Research Society, Pittsburgh, PA, 1993), pp. 885–898.
8. F. W. Bingham and G. E. Barr, "Development of Scenarios for the Long-Term Release of Radionuclides from the Proposed Waste Isolation Pilot Plant in Southeastern New Mexico," in C. J. M. Northrup, Jr. (ed.), *Scientific Basis for Nuclear Waste Management, Proceedings of the International Symposium, Boston, MA, November 27–30, 1979*, Vol. 2, SAND79-0955C (Plenum Press, New York, 1980), pp. 771–778.
9. IAEA (International Atomic Energy Agency), *Safety Assessment for the Underground Disposal of Radioactive Wastes*, Safety Series No. 56 (International Atomic Energy Agency, Vienna, 1981).
10. E. D. Clayton, *Anomalies of Nuclear Criticality*, PNL-SA-4868, Rev. 5 (Pacific North-west Laboratory, Richland, WA, 1979).
11. W. R. Stratton, *The Myth of Nuclear Explosions at Waste Disposal Sites*, LA-9360 (Los Alamos National Laboratory, Los Alamos, NM, 1983).
12. G. H. Canavan, S. A. Colgate, O. P. Judd, G. H. McCall, A. G. Petschek, J. C. Solem, T. F. Stratton, W. R. Stratton, and P. P. Whalen, *Comments on "Nuclear Excursions" and "Criticality Issues,"* LA-UR-95-0851 (Los Alamos National Laboratory, Los Alamos, NM, 1995).
13. R. P. Rechard (ed.), *Initial Performance Assessment of the Disposal of Spent Nuclear Fuel and High-Level Waste Stored at Idaho National Engineering Laboratory*, Vols. 1–2, SAND93-2330/1/2 (Sandia National Laboratories, Albuquerque, NM, 1993).
14. R. P. Rechard, M. S. Tierney, L. C. Sanchez, and M-A. Martell, *Consideration of Criticality When Directly Disposing Highly Enriched Spent Nuclear Fuel in Unsaturated Tuff: Bounding Estimates*, SAND96-0866 (Sandia National Laboratories, Albuquerque, NM, 1996).
15. R. P. Rechard (ed.), *Performance Assessment of the Direct Disposal in Unsaturated Tuff of Spent Nuclear Fuel and High-Level Waste Owned by U.S. Department of Energy*, Vols. 1–3, SAND95-2563/1/2/3 (Sandia National Laboratories, Albuquerque, NM, 1995).
16. R. P. Rechard, L. C. Sanchez, C. T. Stockman, J. L. Ramsey, Jr., and M. Martell, "Consideration of Criticality in a Nuclear Waste Repository." *Proceedings of Fifth International Conference on Nuclear Criticality Safety, Albuquerque, New Mexico, September 17–21, 1995* (Sandia National Laboratories, University of New Mexico, Los Alamos National Laboratories, Albuquerque, NM, 1995).
17. M. L. Wilson, J. H. Gauthier, R. W. Barnard, G. E. Barr, H. A. Dockery, E. Dunn, R. R. Eaton, D. C. Guerin, N. Lu, M. J. Martinez, R. Nilson, C. A. Rautman, T. H. Robey, B. Ross, E. E. Ryder, A. R. Schenker, S. A. Shannon, L. H. Skinner, W. G. Halsey, J. D. Gansemer, L. C. Lewis, A. D. Lamont, I. R. Triay, A. Meijer, and D. E. Morris, *Total-System Performance Assessment for Yucca Mountain—SNL Second Iteration (TSPA-1993)*, Vols. 1–3, SAND93-2675 (Sandia National Laboratories, Albuquerque, NM, 1994).
18. NAS (National Academy of Sciences), *Technical Bases for Yucca Mountain Standards* (National Academy Press, Washington, D.C., 1995).
19. F. Gauthier-Lafaye and F. Weber, "The Francevillian (Lower Proterozoic) Uranium Ore Deposits of Gabon," *Econ. Geol.* **84** (8), 2267–2285 (1989).
20. F. Gauthier-Lafaye, F. Weber, and H. Ohmoto, "Natural Fission Reactors of Oklo," *Econ. Geol.* **84** (8), 2286–2295 (1989).
21. G. A. Cowan, "A Natural Fission Reactor," *Scientific American* **235** (1), 36–47 (1976).
22. G. A. Cowan and A. E. Norris (eds.), *Investigations of the Natural Fission Reactor Program; October, 1977–September, 1978*, Report No. 7530 (Los Alamos Scientific Laboratory, Los Alamos, NM, 1978).
23. B. Nagy, F. Gauthier-Lafaye, P. Holliger, D. W. Davis, D. J. Mossman, J. S. Leventhal, M. J. Rigali, and J. Parnell, "Organic Matter and Containment of Uranium and Fissiogenic Isotopes at the Oklo Natural Reactors," *Nature* **354** (6353), 472–475 (1991).
24. W. J. Maeck, F. W. Spraktes, R. L. Tromp, and J. H. Keller, "Analytical Results, Recommended Nuclear Constants and Suggested Correlations for the Evaluation of Oklo Fission-Product Data," in *Le Phenomene d'Oklo/The Oklo Phenomenon, Proceedings of a Symposium, Libreville, Gabon, June 23–27, 1975* (International Atomic Energy Agency, Vienna, 1975), pp. 319–339.
25. G. A. Cowan, E. A. Bryant, and W. R. Daniels, "Some United States Studies of the Oklo Phenomenon," in *Le Phenomene d'Oklo/The Oklo Phenomenon, Proceedings of a Symposium, Libreville, Gabon, June 23–27, 1975* (International Atomic Energy Agency, Vienna, 1975), pp. 341–356.
26. R. A. Knief, *Nuclear Criticality Safety: Theory and Practice* (American Nuclear Society, La Grange Park, IL, 1985).
27. W. R. Stratton and D. R. Smith, *A Review of Criticality Accidents*, DOE/NCT-04 (U.S. Department of Energy, Assistant Secretary of Environment, Safety and Health, Washington, D.C., 1989).
28. D. K. Parsons, K. Despain, T. N. Dey, and J. L. Kammerdiener, "Los Alamos Review of the Bowman-Venneri Paper Regarding Possible Underground Criticality," in *Oral Presentation to the "Special ANS Panel: Criticality Stability of Fissile Material Underground," American Nuclear Society Annual Meeting, Philadelphia, PA, June 24–29, 1995*, LA-UR-95-2152 (Los Alamos National Laboratory, Los Alamos, NM, 1995).
29. G. E. Hansen, C. C. Byers, and J. J. Koelling, "Fission and Explosive Energy Releases of PuO_2 and PuO_2—UO_2 Assemblies," *Trans. Am. Nucl. Soc.* **24**, 208–209 (1976).
30. R. H. Kimpland, "Dynamic Analysis of Nuclear Excursions in Underground Repositories Containing Plutonium," *Sci. Global Security* **5**, 323–332 (1996).
31. R. P. Rechard, *An Introduction to the Mechanics of Performance Assessment Using Examples of Calculations Done for the Waste*

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Isolation Pilot Plant Between 1990 and 1992, SAND93-1378 (Sandia National Laboratories, Albuquerque, NM, 1995).
32. H. C. Paxton, *A History of Critical Experiments at Pajarito Site*, LA-9685-H (Los Alamos National Laboratory, Los Alamos, NM, 1983).
33. M. J. Plaster, B. Basoglu, C. L. Bentley, M. E. Dunn, A. E. Ruggles, A. D. Wilkinson, T. Yamamoto, and H. L. Dodds, "Analysis of a Hypothetical Criticality Accident in a Waste Supercompactor," *Nucl. Technol.* **111** (2), 219–226 (1995).
34. R. Serber, *The Los Alamos Primer: The First Lectures on How to Build an Atomic Bomb* (University of California Press, Berkeley, CA, 1992).
35. J. S. Y. Wang, N. G. W. Cook, H. A. Wollenberg, C. L. Carnahan, I. Javandel, and C. F. Tsang, "Geohydrologic Data and Models of Rainier Mesa and Their Implications to Yucca Mountain," in *High Level Radioactive Waste Management, Proceedings of the Fourth Annual International Conference, Las Vegas, NV, April 26–30, 1993*, Vol. 1 (American Nuclear Society, La Grange Park, IL; American Society of Civil Engineers, New York, NY, 1993), pp. 675–681.
36. C. L. Carnahan, E. L. Majer, N. G. W. Cook, T. V. McEvilly, S. Flexser, L. R. Myer, I. Javandel, C. F. Tsang, K. H. Lee, J. S. Y. Wang, M. J. Lippmann, and H. A. Wollenberg, *A Review of Rainier Mesa Tunnel and Borehole Data and Their Possible Implications to Yucca Mountain Site Study Plans*, LBL-32068 (Lawrence Berkeley Laboratory, Earth Sciences Division, Geologic Repository Project, Berkeley, CA, 1991).
37. B. W. Leslie, E. C. Pearcy, and J. D. Prikryl, "Oxidative Alteration of Uraninite at the Nopal I Deposit, Mexico: Possible Contaminant Transport and Source Term Constraints for the Proposed Repository at Yucca Mountain," *Mater. Res. Soc. Symp. Proc.* **294**, 505–512 (1993).
38. R. C. Elandt-Johnson and N. L. Johnson, *Survival Models and Data Analysis* (John Wiley & Sons, New York, NY, 1980).
39. F. W. Walker, J. R. Parrington, and F. Feiner, *Nuclides and Isotopes*, 14th Ed. (General Electric Company, San Jose, CA, 1989).
40. F. P. Incropera and D. P. DeWitt, *Fundamental of Heat Transfer* (John Wiley & Sons, New York, NY, 1981).
41. H. S. Carslaw and J. C. Jaeger, *Conduction of Heat in Solids*, 2nd Ed. (Clarendon Press, Oxford, 1959).
42. W. E. Baker, J. J. Kulesz, P. S. Westine, P. A. Cox, and J. S. Wilbeck, "*A Manual for the Prediction of Blast and Fragment Loading on Structures*," DOE/TIC-11268, Prepared by Southwest Research Institute, San Antonio, TX, Albuquerque Operations Office, U.S. Department of Energy, Albuquerque, NM (1980).

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