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Long-Term Criticality Control in Radioactive Waste Disposal Facilities Using Depleted Uranium

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LONG-TERM CRITICALITY CONTROL IN RADIOACTIVE WASTE DISPOSAL FACILITIES USING DEPLETED URANIUM

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ABSTRACT

Plant photosynthesis has created a unique planetary-wide geochemistry—an oxidizing atmosphere with oxidizing surface waters on a planetary body with chemically reducing conditions near or at some distance below the surface. Uranium is four orders of magnitude more soluble under chemically oxidizing conditions than it is under chemically reducing conditions. Thus, uranium tends to leach from surface rock and disposal sites, move with groundwater, and concentrate where chemically reducing conditions appear. Earth's geochemistry concentrates uranium and can separate uranium from all other elements except oxygen, hydrogen (in water), and silicon (silicates, etc).

Fissile isotopes include ^{235}U, ^{233}U, and many higher actinides that eventually decay to one of these two uranium isotopes. The potential for nuclear criticality exists if the precipitated uranium from disposal sites has a significant fissile enrichment, mass, and volume. The earth's geochemistry suggests that isotopic dilution of fissile materials in waste with ^{238}U is a preferred strategy to prevent long-term nuclear criticality in and beyond the boundaries of waste disposal facilities because the ^{238}U does not separate from the fissile uranium isotopes. Geological, laboratory, and theoretical data indicate that the potential for nuclear criticality can be minimized by diluting fissile materials with ^{238}U to 1 wt % ^{235}U equivalent.

I. INTRODUCTION

Nuclear criticality is an important issue associated with the disposal of wastes containing fissile materials. These wastes include spent nuclear fuel (SNF), transuranic wastes, and some types of low-level waste (LLW). Waste disposal facilities that may contain fissile materials include both shallow-land disposal facilities and geological repositories. Nuclear criticality can be initially prevented in a disposal facility by using engineering techniques such as geometry control and neutron absorbers. However, ultimately waste packages (WPs) will fail, the geometry of the WP and disposal facility will change, and selective materials will leach from the WP. Long-term criticality control refers to the control of nuclear criticality after WP failure. Classical engineering approaches to criticality control may not be effective under these changing conditions.

II. WASTE DISPOSAL FACILITY CRITICALITY ISSUES

Long-term nuclear criticality is a concern because of environmental, legal, and philosophical considerations. Current law[1] requires avoidance of nuclear criticality in a disposal facility. Furthermore, the National Academy of Sciences,[2] in its recommendations on performance standards for the proposed Yucca Mountain repository in the United States, recommends that disposal site performance be evaluated out beyond the time of maximum public exposure—this time is after WP failure. Such performance can be influenced by the potential for nuclear criticality events.

Nuclear criticality should be avoided in a waste disposal facility to minimize any potential for release of radionuclides to the biosphere. Naturally occurring nuclear reactors in the geological past[3–6] indicate that such events generate added radioactivity and heat over hundreds of thousands of years. The heat generated causes higher temperatures that, in turn, (1) accelerate chemical reactions, which then accelerate the degradation of WPs; (2) cause water movement that may transport radioactivity to the open environment;[7] and (3) create uncertainties in disposal facility

performance.[8] Heat can accelerate water movement in both unsaturated[9] and saturated geological environments. Because heat is a driver for groundwater movement, it is also a driver for radionuclide transport.

Nuclear criticality produces fission products and actinides that must be isolated from the environment. Criticality can increase the long-term hazards of the waste. Disposal sites are designed to contain radionuclides; however, long-term criticality, if it occurred, would occur after WP failure and degradation of the site capabilities for radionuclide containment. If the fissile materials are transported beyond the facility boundaries, nuclear criticality may occur beyond the specific geology chosen to contain the radionuclides.

III. CONDITIONS REQUIRED FOR NUCLEAR CRITICALITY

Nuclear criticality will occur if sufficient amounts and concentrations of fissile materials exist with appropriate geometry. The required concentrations depend upon the types of fissile materials, availability of moderators, and concentrations of other neutron absorbers. The primary fissile isotope of concern is ^{235}U, which is the dominant fissile isotope in many waste streams and is the decay product of ^{239}Pu—the other major fissile isotope in waste streams.

IV. GEOCHEMICAL BEHAVIOR OF URANIUM AND OTHER FISSILE MATERIALS

Uranium in any disposal site will ultimately dissolve into the groundwater, be transported, and reprecipitate. The specifics depend upon the local geochemistry and its evolution over time. The same processes that have created uranium ore bodies over the last several billion years of the evolution of the earth still operate on fissile materials within and beyond a disposal site. Nuclear criticality may occur if these geochemical processes concentrate the uranium and the other fissile materials. To understand the potential for long-term nuclear criticality events, we must first understand the geochemistry of uranium.

The general geochemistry of earth is well known. The planet as a whole exists under chemically reducing conditions and initially had a chemically reducing atmosphere.[10] With the evolution of life and photosynthesis, the atmosphere was converted from a reducing atmosphere to an oxidizing atmosphere about two billion years ago.[11] On land, the oxidizing conditions extend from the atmosphere to below the earth's surface for distances measured from <1 cm (coal deposits, swamps, etc.) to >1000 m. The ocean is oxidizing, but sludges on the bottom are for the most part under chemically reducing conditions because of the decomposition of organic material.

The solubility of uranium compounds is strongly dependent upon the oxidation conditions of the environment. Under geochemical reducing conditions, uranium is in the +4 valence state, is highly insoluble (<1 ppb), and is often found as uranium dioxide (UO_2). Under chemically oxidizing conditions, uranium is in the +6 valence state, is 2 to 4 orders of magnitude more soluble than under reducing conditions, and is often found as a uranyl (UO_2^{+2}) ion. Because of these chemical characteristics[3,12–16], many uranium ore deposits are formed by (1) oxidation of uranium from the +4 to the +6 valence state by oxidizing groundwater (from rain), (2) subsequent dissolution in groundwater, (3) transport in groundwater, and (4) reprecipitation when the groundwater flows through a chemically reducing environment which reduces uranium from the +6 to the +4 valence state (Fig 1). Chemically reducing environments[12,15–17] are primarily, but not exclusively, created by organic materials and iron in the +2 and metal state.

This relatively unusual redox chemistry implies that, unlike most other types of ore deposits, many uranium ore deposits migrate over time.[12] Flowing oxidizing groundwater will also oxidize other reducing agents within the rock. Uranium precipitated by chemical reduction will then be reoxidized, dissolved, transported, and reprecipitated. The uranium remains at the boundary between chemically oxidizing and reducing rock conditions and moves as oxidizing groundwater alters the location of this chemical interface. Such uranium ore deposits are known as "roll-front" deposits and some of these deposits have moved many kilometers over long time periods.

These mechanisms have extracted uranium from rock at concentrations of a few parts per million and created ore bodies with uranium concentrations of several tens of percent by weight. The concentration process separates uranium from most other elements including boron, cadmium, and rare earths. The only elements consistently found with natural uranium ore bodies are silicon, oxygen, and hydrogen in the chemical forms of silica, silicates, and water. Several other mechanisms also concentrate uranium over long time periods.

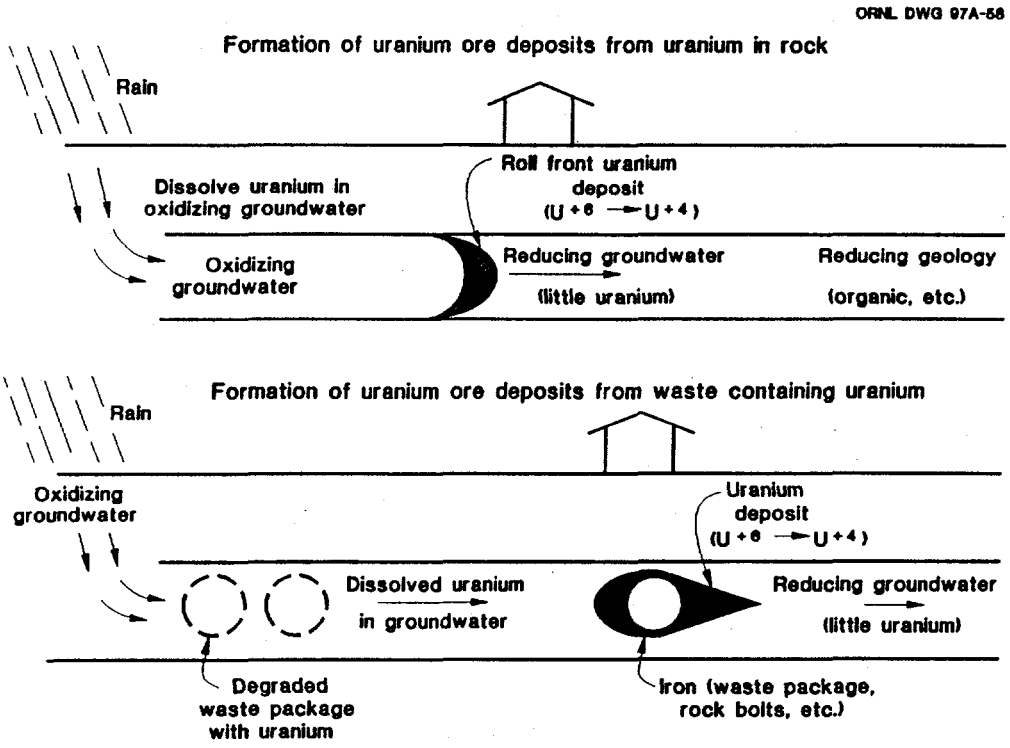

Fig. 1. Natural and man-made formation of uranium ore deposits.

V. IMPLICATIONS FOR LONG-TERM CRITICALITY CONTROL

The planet has an oxidizing atmosphere and at its depth a chemically reducing geology. The earth's geochemistry creates conditions for a dissolution of uranium in oxidizing groundwater at disposal sites, transport of the uranium in groundwater, and precipitation (concentration) of that uranium when chemically reducing conditions are encountered. Most shallow-land disposal facilities and some proposed repositories (e.g., Yucca Mountain in the United States) have chemically oxidizing conditions. Many other sites will ultimately have oxidizing conditions. It is noted that many WP systems contain iron-based alloys or carbon-containing compounds that provide man-made reducing agents within the disposal facility. The same geological processes will occur within the WP and the facility with cycles of uranium oxidation, dissolution, transport, and precipitation. After WP failure, the geometry and concentration of the uranium within the WP, within the disposal facility, and beyond the boundary of the facility will continuously change over time. Whether or when the uranium precipitation will result in sufficient concentrations of uranium such as to cause nuclear criticality is dependent upon the long-term geochemical evolution of the specific site, region, the quantity and shape of the uranium, and the enrichment of the uranium.

In time, dissolved uranium in groundwater with fissile concentrations different from those of natural uranium will isotopically exchange with natural uranium in the rock and be isotopically diluted to ~0.7 wt % ^{235}U. This phenomenon is, however, a slow process and presents the likelihood that the uranium will be concentrated by precipitation processes multiple times before full natural isotopic dilution occurs.

Determination of the probability of nuclear criticality for a specific disposal site depends upon (1) the total fissile inventory at the disposal site, (2) the fissile concentration of the uranium, (3) the climate of the disposal site and its variation with time, (4) the design of the waste facility, (5) the local geology, and (6) the geochemical evolution of the disposal facility and local geology (uplift, erosion, etc.) over time. Man does not currently have the capability to predict climate over long geological time frames and has only limited ability to predict the evolution of disposal sites and geology over time. These uncertainties make it difficult to rely on conventional engineered systems for long-term criticality control.

Lowering the isotopic fissile concentration of uranium in the disposal site to low levels greatly reduces long-term probabilities of nuclear criticality. That is, we need to add sufficient depleted uranium

(DU), uranium high in ^{238}U and low in fissile isotopes, such that the uranium enrichment levels are below those that can cause nuclear criticality. Having the same chemistry as fissile uranium, DU is the only neutron absorber that cannot separate from fissile uranium isotopes via operating geochemical processes over geological time.

The earth's geochemistry concentrates uranium and has resulted in naturally occurring nuclear reactors in the distant past. However, natural occurring reactors have not occurred in recent geological times because the fissile enrichment of uranium has been lowered to ~0.7 wt % ^{235}U by radioactive decay. Isotopic dilution with ^{238}U is the only demonstrated method for long-term control of nuclear criticality in the geological environment.

VI. ALLOWABLE FISSILE CONCENTRATIONS

If isotopic dilution (addition of ^{238}U) is to be used to minimize the potential for nuclear criticality, the technical question is: How much DU is required?

A. Uranium-235

The combinations of uranium enrichment, uranium mass, geometry, and neutron moderation that can result in criticality are known for ^{235}U. The minimum enrichment required to support criticality in a geological environment can be deduced from the following:

1. History. The historical geological record[3,6,16,18,19] shows that nuclear criticality has occurred in natural uranium ore bodies. For example, 16 natural nuclear reactors have been identified at Oklo, Gabon, Africa. The nuclear chain reactions began when the ^{235}U enrichment of natural uranium was about 3.6 wt %. After operation and the ensuing generation of heat and fission products, ^{235}U enrichments of the uranium were as low as 1.3 wt %—equivalent to the fissile enrichment of full-burnup light-water reactor (LWR) SNF. Today, natural uranium deposits have a ^{235}U enrichment level of 0.71 wt % because of the long-term decay of ^{235}U. Nuclear criticality can no longer occur in natural uranium ore bodies because of these low enrichment levels (Fig. 2).

2. Geochemical modeling of uranium ore deposits. The Commissariat a L'Energie Atomique (French Atomic Energy Commission) has studied the conditions during which natural nuclear reactors (ore deposits) form during the concentrating of uranium from rock.[20] The analysis indicates that nuclear criticality may occur at enrichments as low as 1.28 wt % with criticality becoming reasonably probable in some geological environments as enrichments approach 1.64 wt %. Others have estimated that criticality may occur in natural environments with enrichments approaching only 1 wt %.[3]

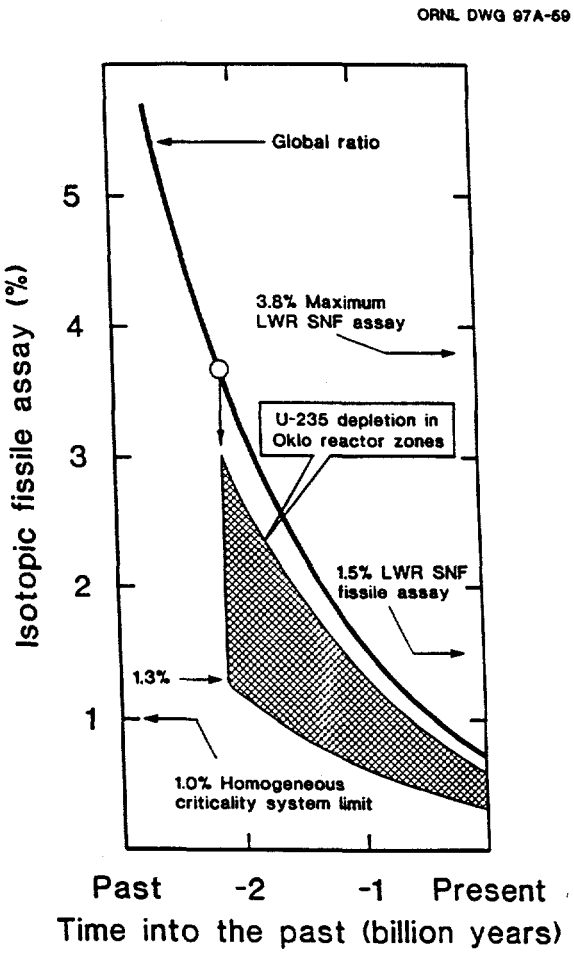

Fig. 2. Natural uranium enrichment levels over geological time.

3. Engineering experiments and analysis. Criticality calculations[21] and laboratory experiments[22] with the types of materials found in the natural environment indicate that nuclear criticality could, in theory, occur with fissile enrichment concentrations as low as ~1 wt % ^{235}U, but there is no experimental evidence that this has occurred in nature.

4. Environmental analysis. An analysis[23] for disposition of surplus high-enriched uranium (that is declared as waste) concluded that the uranium enrichment may be reduced to as low as 0.9 wt % ^{235}U for long-term criticality control. This is a conservative number chosen to bound environmental impacts of process operations to dilute enriched uranium with DU.

5. Waste facility safety assessments. Waste process equipment, waste tanks, LLW disposal sites, and other waste management facilities contain wastes with small quantities of enriched uranium. In many facilities, it is not practicable to control either the system geometry or the chemistry (neutron absorbers). Selective precipitation of uranium into significant deposits is possible. Based on analysis and experiments, many sites require isotopic dilution of ^{235}U with ^{238}U to ensure that nuclear criticality does not occur. For example, Oak Ridge National Laboratory (ORNL)[24] requires that any uranium sent to the liquid process waste system contain at least 100 parts ^{238}U per part ^{235}U.

The previous data suggest that a reasonable strategy to prevent nuclear criticality in disposal facilities is to require sufficient DU fill in each waste package such that the ratio of fissile materials to ^{238}U is <1 wt % ^{235}U equivalent. This assumes homogeneous mixing of fissile materials (see following). The 1 wt % is also the homogeneous criticality limit for ^{235}U in an optimized system containing water and silicon oxide. Such a limit can be extended to any other fissile material by determining the corresponding homogeneous criticality limit for that specific isotope in a system containing the isotope, water, and silicon oxide.

B. Plutonium-239

The disposition of wastes containing ^{239}Pu presents two types of long-term criticality issues. The initial concern is nuclear criticality caused by the ^{239}Pu itself. The longer term concern is that ^{239}Pu decays to ^{235}U. The primary plutonium isotope, ^{239}Pu, has a half-life of 24,000 years (i.e., the decay rate is 3×10^{-5}/year). Other plutonium isotopes have shorter half-lives.

If the ^{239}Pu does not move until it has decayed to ^{235}U, the long-term criticality issue is then with ^{235}U. Long-term criticality control can be addressed by requiring that ^{239}Pu be mixed with ^{238}U to minimize the potential for long-term ^{235}U criticality concerns. In many cases of interest, evidence suggests that ^{239}Pu will decay to ^{235}U before significant transport (i.e., the rate of plutonium decay to uranium is faster than are geochemical processes that separate uranium from plutonium).

The regulatory requirement in the United States[1] for WP system performance is that the maximum allowable radionuclide release rate from the engineered barrier system shall not exceed 10^{-5}/year (1 part in 100,000/year) of the inventory of that radionuclide calculated to be present at 1000 years following permanent repository closure. In a WP system that meets these regulatory requirements and has been filled with DU, most of the plutonium will have decayed to ^{235}U and thus allow isotopic dilution by the DU before significant quantities of uranium or plutonium leave the WP.

Theoretical, laboratory, and field data from naturally occurring reactors and other sources[4,5,14,16,18,19,25] indicate that in proposed facilities for plutonium disposal, the plutonium will decay to uranium before large quantities of uranium or plutonium can be transported from the waste form. It is widely accepted that plutonium will be disposed of in deep geological repositories because it is a long-term hazard. In these geological environments, geochemical changes occur very slowly. However, this assumption of slow change would not necessarily be valid in a near-surface disposal facility.

The inverse premise—that plutonium will not decay to uranium before uranium is transported to and within the local geological environment—must also be considered. Plutonium has very low solubilities in groundwater and thus is expected to be less mobile than uranium. In a geological repository containing significant quantities of DU, the local geological environment will become saturated with the DU. This saturation has longer-term implications. If it is assumed that, in a particular WP, because of some special local conditions, the uranium partly separates from the plutonium before complete plutonium decay to uranium occurs, then residual plutonium will decay to uranium (primarily ^{235}U) and isotopically mix with any remaining uranium in the environment near the WP. As this uranium is transported within the geological environment, it will most likely follow the flow path of the uranium that was earlier leached from the WP if conditions have not changed substantially. This movement will bring it into contact with the uranium that was transported earlier. This fissile uranium will then be isotopically diluted by the DU in the local geological environment below enrichment levels at which nuclear criticality can occur. In such cases,

additional DU can be placed in the WP to compensate for DU loses before the ^{239}Pu decays to ^{235}U.

C. Uranium-233

A recent study[26] evaluated the level of isotopic dilution required for ^{233}U that is equivalent to 1 wt % ^{235}U. The analysis indicates that the corresponding value for ^{233}U is 0.67 wt %.

D. Minor Isotopes

An analysis of isotopic dilution requirements for minor isotopes for criticality control has not been done. Most of these isotopes ultimately decay to ^{233}U or ^{235}U; thus, DU is an appropriate agent to prevent long-term nuclear criticality in a repository by isotopic dilution of the uranium fissile decay products from these actinides. Curium-245, ^{241}Am, and ^{237}Np ultimately decay to ^{233}U. Curium-243, ^{243}Am, and ^{239}Pu ultimately decay to ^{235}U.

E. Limits

To minimize the probability of long-term nuclear criticality in disposal sites as a result of geochemical phenomena, the ^{235}U and ^{239}Pu enrichment levels should be below ~1 wt % . The ^{233}U enrichment levels should be below ~0.67 wt %. In each case, the diluent is ^{238}U. If the material contains a mixture of fissile isotopes, appropriate mixing rules must be used.

The quantity of DU required is higher than that indicated by these fissile concentrations because some of the ^{238}U in the DU must be used for isotopic dilution of the residual ^{235}U in the DU. For example, if 1000 kg of 0.2 wt % DU is used, it contains 2 kg of ^{235}U. This ^{235}U will require 198 kg of ^{238}U for criticality control by isotopic dilution. This leaves 800 kg of ^{238}U available to isotopically dilute the fissile material being treated. Thus, there is an incentive to use DU with the lowest ^{235}U content.

VII. ENGINEERING IMPLICATIONS

The preferred treatment option when using DU for long-term criticality control is to mix (on an atomic scale) the DU with the waste form when it is processed. This option is applicable for wastes that are treated by vitrification, cementation, and plasma processing.

Some wastes, such as SNF will not be processed further and will exceed the 1 wt % ^{235}U equivalent value. For example, the average fissile concentration of LWR SNF is 1.47 wt % and increasing.[27] This SNF contains fissile ^{235}U, ^{239}Pu, ^{241}Pu, and other fissile isotopes. For LWR SNF, options exist to address long-term criticality concerns by using DU (a) to fill the void space in a WP containing SNF[28,29], (b) as a shield material component of the WP, and (c) as a fill around the waste package. In most such applications, the preferred chemical form of the DU will be UO_2. Filling the SNF coolant channels with DU is preferred in terms of criticality control because the DU and fissile materials are mixed on the scale of a centimeter. This minimizes concerns about the dissolution and transport of the DU before the dissolution and transport of the ^{235}U from the SNF occurs. When the DU and SNF uranium are in the same chemical form, uncertainties are minimized because both are expected to evolve together over time.

For other waste forms containing fissile materials, additional DU may be required for greater confidence that the DU will mix in time with the WP fissile materials. For example, ^{239}Pu decays to ^{235}U; however, sufficient ^{238}U must be in the waste with the ^{239}Pu to ensure isotopic dilution over the time it takes for the ^{239}Pu to decay to ^{235}U. If the DU leaches significantly faster than the ^{239}Pu, more DU should be used in the WP to cover expected loses, or the chemistry of the system can be altered to ensure that the DU remains with the other fissile, non-uranium materials.

VIII. AVAILABILITY OF DU

A large excess of DU exists worldwide that far exceeds the potential needs for criticality control. In LWR fuel production, natural uranium with a ^{235}U content of 0.71 wt % is separated into a DU fraction and an enriched uranium fraction. The enriched uranium (3–4% ^{235}U) is fabricated into fuel. Typically, 4 to 6 t of DU with a fissile content of 0.2–0.35 wt % ^{235}U are produced per ton of enriched uranium nuclear fuel. Currently, no substantial use exists for this material. In the United States, about 400,000 t of DU is in storage.[30] The total world inventory exceeds 1×10^6 t. Additional quantities are available from other enrichment facilities in Russia, France, Great Britain, Netherlands, Germany, and China.

IX. OTHER CONSIDERATIONS

Other approaches are available to address nuclear criticality concerns in a waste disposal facility.

Examples include geometry control and conventional neutron absorbers. However, these other options will be difficult to use because of the difficulty of predicting the chemical and geometric conditions in a disposal site over long time periods. In this context, the Nuclear Waste Technical Review Board,[8] mandated by U.S. law to review the technical aspects of U.S. repository design, recently analyzed long-term repository criticality issues associated with the proposed Yucca Mountain repository and stated: "Although external [to the WP] criticality may be unlikely, it can not be dismissed without thorough analysis. The Board understands that the U.S. Department of Energy intends to use probablistic risk analysis methodology to address external criticality. While such an approach is appealing, it may turn out to be costly and time-consuming to the point of impracticality in a repository context because of the very large number of events and geometric configurations possible in a repository. . . ." The board recommended the consideration of the use of DU for control of nuclear criticality in such facilities.

In recent years, speculation has arisen that autocatalytic, high-energy criticality events might occur in disposal facilities[31] in addition to the type that occurred at Oklo. The use of DU eliminates any criticality concerns that might be associated with these theoretical events.[32]

X. CONCLUSIONS

The geochemistry of earth makes it difficult to assure with high confidence that nuclear criticality will not occur in a waste disposal facility if the nuclear criticality control strategy is based on geometric control or use of standard neutron absorbers. However, isotopic dilution of fissile materials with ^{238}U can provide reasonable assurance that nuclear criticality will not occur in a disposal facility.

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