

**Criticality in a high level waste repository****A review of some important factors and an assessment of the lessons that can be learned from the Oklo reactors**Virginia M Oversby  
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June 1996

# **CRITICALITY IN A HIGH LEVEL WASTE REPOSITORY**

## **A REVIEW OF SOME IMPORTANT FACTORS AND AN ASSESSMENT OF THE LESSONS THAT CAN BE LEARNED FROM THE OKLO REACTORS**

*Virginia M. Oversby*

**VMO Konsult**

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This report concerns a study which was conducted for SKB. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

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## ABSTRACT

The conditions and scenarios that might allow sufficient  $^{239}\text{Pu}$  and/or  $^{235}\text{U}$  to accumulate together with enough water to allow for moderation of neutron energies and thereby achieving a state where neutron-induced fission reactions could be sustained at a rate significantly above the natural rate of spontaneous fission is discussed. The uranium deposit in Oklo, Gabon, which was the site of naturally-occurring neutron-induced fission reactions approximately 2000 My ago is described. The chemistry, mineralogy, and conditions of the nuclear reactor operations are reviewed. Results of modelling the conditions for criticality at Oklo are used to estimate the amounts of spent fuel uranium that must be assembled in a favorable geometry in order to produce a similar reactive situation in a geologic repository. The amounts of uranium that must be transported and redeposited to reach a critical configuration are extremely large in relation to those that could be transported under any reasonably achievable conditions. In addition, transport and redeposition scenarios often require opposite chemical characteristics. It is concluded that the likelihood of achieving a critical condition due to accumulation of a critical mass of uranium outside the canisters after disposal is negligible. Criticality inside the canister is rendered impossible by the use of low-solubility materials inside the canisters that fill space and prevent the entry of enough water to allow moderation of neutron energies. Criticality due to plutonium outside the canister can be ruled out because it requires a series of processes, each of which has a vanishingly small probability.

## SAMMANFATTNING

De förhållanden och scenarier som skulle kunna tillåta tillräckliga mängder  $^{239}\text{Pu}$  och/eller  $^{235}\text{U}$  att ansamlas tillsammans med tillräckligt mycket vatten för att möjliggöra moderation av neutronenergierna och därigenom få ett tillstånd där neutroninducerade fissionsreaktioner kan upprätthållas på en nivå som är påtagligt högre än den för spontana fissioner diskuteras. Uranförekomsten i Oklo, Gabon, som var platsen för naturligt förekommande neutroninducerade fissioner för ungefär 2000 millioner år sedan beskrivs. En översikt ges över kemin, mineralogin och förhållandena kring kärnreaktorernas verksamhet. Resultaten från modellering av förhållandena för kriticitet vid Oklo används för att uppskatta mängderna uran från använt bränsle som måste ansamlas i gynnsam geometri för att åstadkomma en liknande reaktiv situation i ett geologiskt förvar. Mängden uran som måste transporteras och återdeponeras för att nå en kritisk konfiguration är utomordentligt stor i förhållande till vad som kan transporteras under några förhållanden, som rimligen kan uppnås. Dessutom kräver transport och återdeponering ofta motsatta kemiska karakteristika. Slutsatsen är att sannolikheten för att nå kriticitet på grund av en kritisk uranmängd utanför kapseln efter deponering är försumbar. Kriticitet inuti kapseln omöjliggörs genom att använda material med låg löslighet som fyller utrymmet inne i kapseln och förhindrar att tillräckligt mycket vatten för att moderera neutronenergierna kan tränga in. Kriticitet utanför kapseln på grund av plutonium kan uteslutas därför att det kräver en serie av processer där var och en har försvinnande liten sannolikhet.

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## SUMMARY AND CONCLUSIONS

The likelihood that a critical mass of either uranium or plutonium might be accumulated at some time after disposal of spent nuclear fuel in a geologic repository is one of the safety analysis cases that must be done to assess the long term performance of the repository. This report reviews previous analyses of the possibility for and consequences of reaching critical configurations in an older Swedish repository concept that used a canister design that differed from the present design concepts. In addition, the conditions of formation and operation of the natural ore deposit fission reactor zones at Oklo, Gabon, are reviewed.

The conclusions reached are that:

Criticality due to plutonium inside the canister is not possible due to lack of sufficient plutonium in a single canister and due to lack of space in which to accumulate water to moderate neutron energies.

Criticality due to plutonium outside the canisters is shown to be impossible because of the lack of a mechanism to dissolve and transport the plutonium out of the canisters and then accumulate it in the needed geometry under any reasonable repository disposal conditions.

Criticality due to uranium inside the canister is rendered impossible by the use of low solubility materials to fill the space inside the canister and make it impossible to have enough water inside the canister to moderate neutron energies.

Criticality due to uranium outside the canister would require dissolution and transport of uranium under oxidizing conditions and deposition of uranium under reducing conditions. There is no credible mechanism to achieve both oxidizing and reducing conditions in the near-field repository host rock in the long term, after decay of the majority of alpha active isotopes. Thus, criticality due to uranium in the long term has a vanishingly small probability, requiring the application of two or more highly improbable events.

# 1 INTRODUCTION

Disposal of high level radioactive waste that contains substantial amounts of actinide elements carries with it the need to assess the potential for criticality to occur after disposal. This paper will address the issue of criticality for the case of disposal using the combined copper-steel canister described in the SKB 1993 Annual Report (SKB, 1994) and the KBS-3 disposal concept. Emphasis will be placed on evaluations using the results of studies of the natural uranium deposits in Gabon at Oklo, which sustained nuclear chain reactions approximately 2000 million years ago.

## 1.1 SCENARIOS UNDER WHICH CRITICALITY MIGHT BE REACHED

A range of scenarios might potentially lead to criticality, including entry of water into a failed waste canister and achieving a chain reaction inside the canister. For the case where the canister is largely intact and the water enters through a relatively small opening, the control and/or prevention of criticality can be achieved by inclusion of suitable materials inside the canister.

Another possibility for criticality to occur could present itself after the canister has failed to an extent that the spent fuel can be directly attacked by aqueous corrosion. One pathway for fuel corrosion would be the destruction of Zircaloy cladding by corrosion in high chloride waters, followed by dissolution of the fuel, assisted by radiolytic reaction products of the water and dissolved salts. In this case, the spent fuel and residual cladding material would form a somewhat heterogeneous mixture with any other materials that were in the canister, including corrosion products of the inner steel canister wall. If the canister were filled with glass beads prior to sealing to limit the potential for criticality, the final mixture would consist of silica, iron oxides, zirconium oxides and hydroxides, and uranium dioxide with approximately three percent fission products and one percent plutonium.

The outer copper canister will respond to pressure from the surroundings by plastic flow once the inner steel component has corroded to the extent that it can no longer provide structural strength. The pressure will be transmitted to the failed canister by the bentonite buffer material surrounding the canister. While a complete safety assessment would probably need a more detailed analysis of the

pressure field in the disposal setting, this is not possible until a disposal site is selected and detailed rock mechanical properties of the specific site are measured. In the absence of such information, we will assume that the pressure load will be hydrostatic and that the canister will be compressed onto the inner materials. The major variable in this case would be the residual porosity and, thus, the volume available for water inside the canister.

The second way in which criticality might be achieved after canister failure is through the dissolution and redeposition of fissile materials outside the canister. Both the geometry of redeposition and the chemistry in terms of uranium and plutonium concentrations would need to be evaluated as well as the chemistry of the medium in which deposition occurred.

## **1.2 PREVIOUS ASSESSMENTS OF CRITICALITY IN A SPENT FUEL REPOSITORY IN WET CRYSTALLINE ROCK**

Behrenz and Hannerz (1978) reviewed the physical and chemical conditions that might possibly produce criticality for the KBS disposal concept, which had a 200 mm thick copper canister filled with 498 BWR spent fuel rods with all free space between the rods filled with lead cast into place. They concluded the "the scenarios leading to criticality incidents in the repository are based on so unlikely assumptions that they border on physical impossibility and would hardly merit consideration in an ordinary safety analysis. The sole reason for discussing them at length in this report is the fact that statements are frequently appearing in the literature and public debate claiming that criticality represents a major hazard in geologic disposal of spent fuel." The scenarios considered by Behrenz and Hannerz are reviewed in Appendix 1. Differences between the canister design in 1978 and in 1994 are discussed there to show the implications for criticality scenarios.

## **1.3 RECENT DISCUSSIONS OF THE POSSIBLE CONSEQUENCES OF CRITICALITY**

Recent calculations by Bowman and Venneri (undated manuscript) claimed that autocriticality and explosive conditions could be reached during disposal of all categories of waste containing fissile actinides. Their calculations were done for a system of plutonium-239 in a mixture of water and silica, with that system embedded in an infinite silica medium to act as a neutron reflector. The authors noted that in realistic cases  $^{240}\text{Pu}$  would also be present. The half life of  $^{240}\text{Pu}$  is  $6.56 \times 10^3$  years, while that of  $^{239}\text{Pu}$  is  $24.1 \times 10^3$  years (General Electric Company, 1984). While  $^{240}\text{Pu}$  is present, it will effectively prevent thermalization of neutrons by capturing them. The case illustrated that gave positive feedback, or autocatalytic, behavior was for accumulation of  $^{239}\text{Pu}$  by redeposition in a silica

plus water sphere of 200 cm radius. This system showed calculated positive feedback for all values of water to silica, including dry (no water); however, the minimum amount of  $^{239}\text{Pu}$  required to be in the sphere was approximately 60 kg dispersed in a 70 ton silica mass. Spherical volumes with smaller dimensions could become critical at lower masses of total  $^{239}\text{Pu}$ , but the region of positive feedback in these cases would be limited to a range of water contents. In these cases, as reactions heated the system, water would be boiled away and the reactions would eventually stop when the region of negative feedback was reached. The authors did not present any mechanisms for how the assumed geometric configuration could be achieved, but called upon dispersion through natural or unnatural processes. In particular, Bowman and Venneri considered that plutonium would become dispersed if criticality events occur.

Bowman and Venneri (undated) claimed that autocatalytic criticality would also be possible for commercial reactor spent fuel. They based this conclusion on the decay of  $^{240}\text{Pu}$  before the  $^{239}\text{Pu}$  had completely disappeared, and on the assumption that the difference in solubility of uranium and plutonium would allow the  $^{238}\text{U}$ , which is a neutron absorber, to be dissolved away and leave the  $^{239}\text{Pu}$  behind. Their argument seems to be that the greater quantities of commercial spent fuel as compared to weapons grade plutonium would make the spent fuel a greater problem. Behrenz and Hannerz (1978) considered the possibility for criticality to occur due to preferential dissolution and removal of the U from Pu. In the real conditions, the transport rates for U through the bentonite buffer would limit the amount of uranium that could be removed from a canister. Removal of Pu by preferential dissolution and transport is inconsistent with the known chemical behavior of Pu and U. Even if the bentonite buffer were assumed to have failed so that unlimited transport of uranium could occur, and that the most favorable condition of a homogeneous mixture of Pu and water inside residual zircaloy cladding tubes was assumed to occur, there was insufficient Pu in the fuel to achieve criticality at any time (Behrenz and Hannerz, 1978, Appendix 2).

Van Konynenburg (1995) reviewed the Bowman and Venneri paper and concluded that the paper blurred important distinctions between and among the various fissile materials under consideration for geologic disposal. Further, he found that the paper contains assumptions and statements that are inconsistent with known properties and behavior of actual waste forms and real rock, and that the authors did not present plausible explanations of a process or processes that could convert actual disposed waste into a configuration close enough to their hypothetical configuration to make the calculations relevant. Van Konynenburg (1995) recommends the use of risk-based analysis of possible scenarios for criticality, taking into account the actual details of proposed designs, rather than the use of hypothetical, idealized configurations. This recommendation is similar to the approach used by Behrenz and Hannerz (1978), who assessed the potential for

processes to occur, but then calculated effects of assumed accumulations of fissile material under possible, but highly improbable, circumstances.

To test the possibility for autocatalytic criticality in spent fuel disposal under conditions similar to the Swedish KBS-3 concept, let us examine the case of BWR spent fuel in a canister that contains 12 BWR fuel assemblies. Twelve assemblies amounts to approximately 2000 kg of original low-enriched uranium (~3%  $^{235}\text{U}$ ). Table 1 shows the total amounts of uranium and plutonium isotopes (excluding  $^{238}\text{U}$  and minor isotopes) in a canister of spent fuel. The total amount of  $^{239}\text{Pu}$  per canister never exceeds 10 kg for a burnup of 35MWd/kgU. This is only 1/6 the amount of  $^{239}\text{Pu}$  needed for the most favorable conditions of pure silica plus plutonium to produce a totally autocatalytic system in a sphere of 200 cm radius. As shown in Table 2,  $^{239}\text{Pu}$  increases with increasing burnup of the fuel, but not fast enough to substantially alter the total amount of  $^{239}\text{Pu}$  in the canister. For a 100 cm radius sphere, which has a limited region in pure silica that showed autocatalytic behavior, the minimum amount of  $^{239}\text{Pu}$  would be approximately 12 kg. The presence of materials that absorb neutrons, which would occur in all real

**Table 1: BWR Fuel composition for a canister with 12 assemblies, burnup of 35MWd/kgM, amount in kg per canister. Total fuel material - approx. 2000kg U. Data from Guenther et al. (1991)**

Isotope	At 20 yrs	At 1000 yrs
$^{235}\text{U}$	11.2	11.5
$^{236}\text{U}$	7.6	8.1
$^{239}\text{Pu}$	9.8	9.6
$^{240}\text{Pu}$	4.8	4.4
$^{242}\text{Pu}$	1.14	1.14

**Table 2: BWR Fuel partial list of actinide content and change with burnup. Fuel age = 20 yrs. Amount in g/kgM**

Isotope	25MWd/kg	30MWd/kg	35MWd/kg
$^{235}\text{U}$	9.70	7.44	5.63
$^{236}\text{U}$	3.33	3.62	3.81
$^{239}\text{Pu}$	4.70	4.85	4.92
$^{240}\text{Pu}$	1.88	2.20	2.43

cases, would increase the minimum amount of  $^{239}\text{Pu}$ . Thus, for the Swedish canister design,  $^{239}\text{Pu}$  from more than one canister would be needed to produce the proposed autocatalytic process. This would require some form of yet-to-be-identified transport process to combine the  $^{239}\text{Pu}$  content of 2 or more canisters in the required geometric configuration.

In the long term,  $^{239}\text{Pu}$  will decay to  $^{235}\text{U}$ , giving a total  $^{235}\text{U}$  content of 1.44% for BWR fuel with 25 MWd/kgM and 1.07% for BWR fuel with 35 MWd/kgM (including a 0.01% addition due to decay of  $^{243}\text{Am}$ ). Behrenz and Hannerz (1978) considered the case of criticality due to accumulation of uranium in the tunnel filling material above the deposition boreholes. The tunnel fill was assumed to be 85% sand and 15% bentonite. For a fuel with 1.66%  $^{235}\text{U}$  after decay of  $^{239}\text{Pu}$ , the minimum critical mass was 4.4 t if the uranium accumulated in a spherical geometry at a concentration of  $0.78\text{g/cm}^3$ . For both the original KBS and the recent SKB (1994) canister concepts, fuel from more than 2 canisters would need to contribute to the uranium accumulation to reach a minimum critical mass. For the lower  $^{235}\text{U}$  contents given above, the minimum critical mass would be even larger. Accumulation of a critical configuration within fractures in the crystalline rocks was considered to be impossible because of neutron leakage from the uranium in narrow fractures and subsequent absorption of the neutrons by components of the rock.

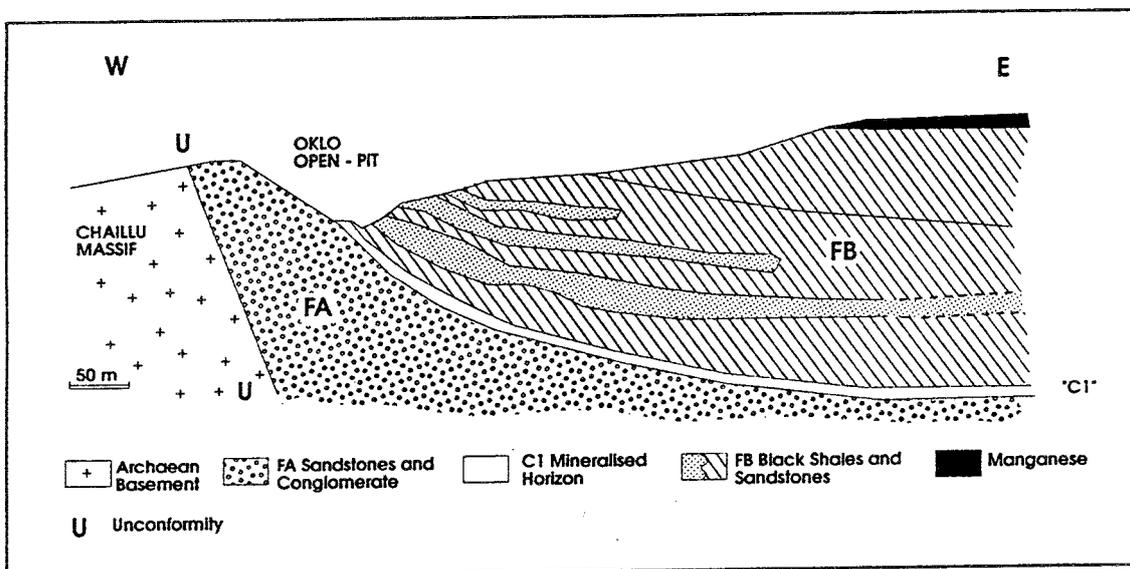
Behrenz and Hannerz (1978) also considered the potential to accumulate a critical mass within a deposition borehole using the content of a single canister. The only type of scenario that could produce this condition would be (1) loss of about 45% of the bentonite from the top of the borehole and replacement with water in the void spaces created, (2) penetration of the canister, (3) diffusion of hexavalent uranium into the upper part of the deposition borehole, (4) diffusion of ferrous iron down from the tunnel into the borehole, and (5) reduction and precipitation of uranium (IV) in the upper part of the borehole containing high-water-content bentonite. The density of the uranium would have to be of the order of  $2\text{g/cm}^3$  or greater in the aqueous portion of the water/bentonite mixture. This scenario would require a number of improbable chemical and hydrologic circumstances to occur in precisely the assumed sequence.

In the sections that follow, we will review the geological, geochemical, and hydrologic features of the Oklo, Gabon, uranium deposit that contains pockets of ore that sustained nuclear reactions approximately 2 billion years ago. We will use these data to draw inferences about the likelihood of transport of uranium and plutonium in a geologic disposal situation, the potential for a critical configuration to be achieved, and the consequences if a criticality event did occur.

## 2

# THE OKLO NATURAL REACTORS

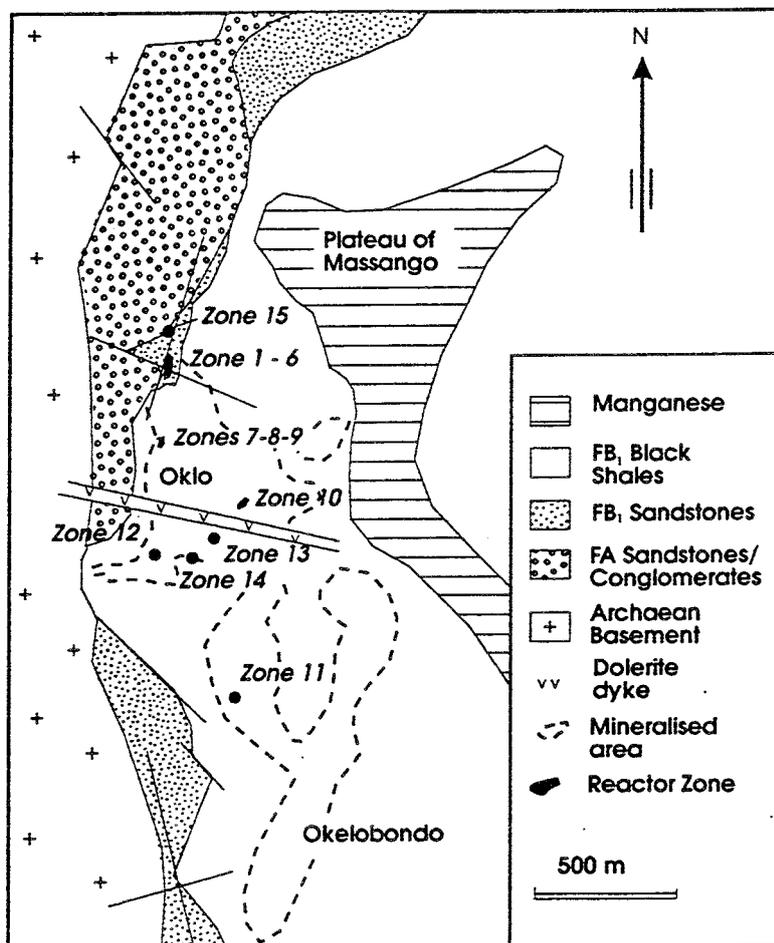
In 1972 a laboratory in France found that the isotopic composition of uranium ore from a mine in Gabon had lower amounts of the 235 isotope than all other natural uranium. The discovery was quickly realized to show the existence in the distant past of fission reactions occurring in a natural setting (Neuilly et al., 1972). A major geological and geochemical undertaking was organized by the French Commissariat à l'Energie Atomique to investigate the ore body and gain further understanding of this unique occurrence. Five years of study were summarized in an IAEA conference held in Paris in 1977 (IAEA 1978). In the late 1980s interest in the Oklo phenomenon was revived and a new study was initiated by CEA with support from the Commission of the European Communities (H. von Maravic, 1993).



*Figure 1. Schematic geological profile across the western margin of the Franceville Basin. Indicated is the Oklo open pit showing the position of the C1 layer of pelitic sediments that host the presently mined uranium ore and the fossil reactor zones (from Smellie, 1995).*

The uranium deposit at Oklo is located near the top of the lowest unit of the Francevillian series, the FA sandstone and conglomerate formation. The ore is found in a narrow region called the C1 layer and is overlain by the black shales of the FB formation. Figure 1 gives a schematic geological cross-section of the ore-bearing region.

Mining began in a region where the C1 bed intersected the ground surface. The mining operation was done in a pit and over time 9 zones were discovered that had isotopic compositions of uranium that were depleted in the 235 isotope. Additional reaction zones have been identified in deeper locations underground at Oklo and Okelobondo and in a shallow location at Bangombé, about 30 km east of Oklo (Gauthier-Lafaye et al., 1989). Figure 2 shows the geographic distribution of the Oklo and Okelobondo uranium deposits and reactor zones.



**Figure 2.** Simplified geological map of the Oklo and Okelobondo uranium deposits showing locations of fossil reaction zones 1-15; reactors 1-9 are located in the open pit and 10-15 are underground. (From Smellie, 1995)

The normal ore at Oklo consists of 0.2 to 1%  $UO_2$  dispersed in the C1 sandstone bed. Where the reactor zones are found, the sandstone has been replaced by a particular clay mineral assemblage not seen in any other location in the Franceville Basin, and the ore is a very rich uraninite, with 20 to 60%  $UO_2$  (Gauthier-Lafaye et al., 1989). The reactor zones are typically lenticular in shape, with average length and width of 10 meters, and a thickness of 10 to 50 cm. Figure 3 shows an idealized cross-section of a fossil reactor.

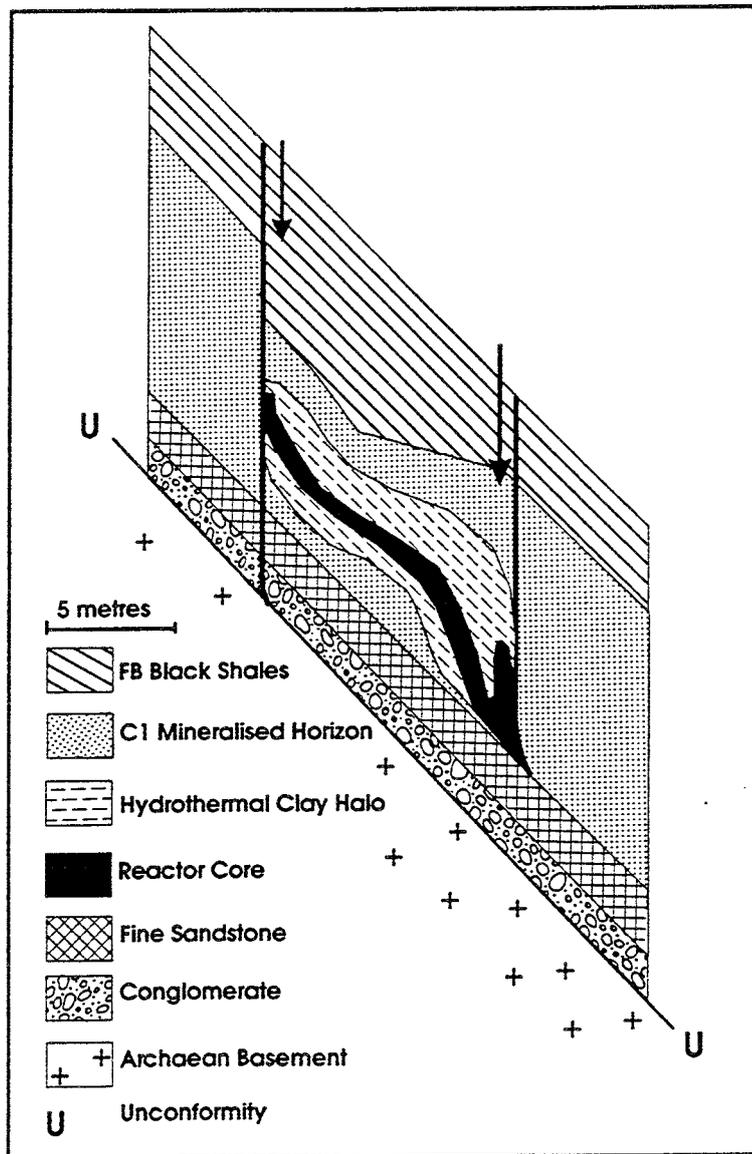
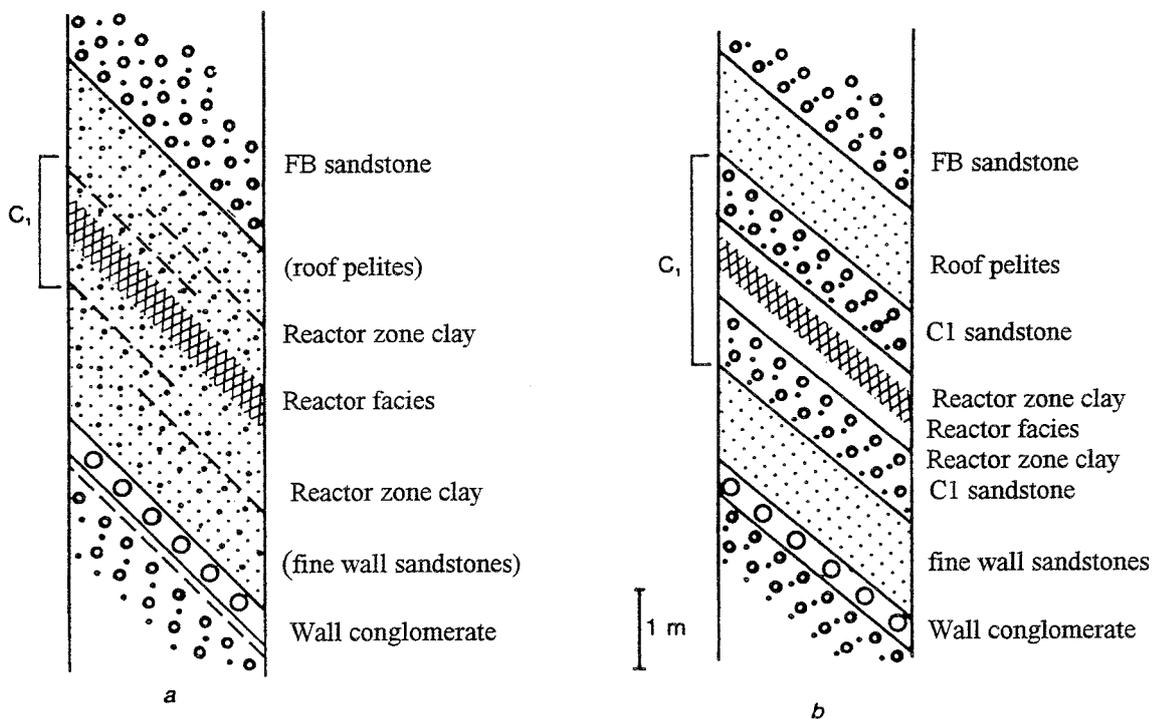


Figure 3. Idealized cross-section of a fossil reactor zone (from Smellie, 1995).

As we shall see in the more detailed discussions of how criticality was sustained at Oklo, the ore became enriched through the dissolution and removal of silica from the sandstones, leaving behind clay materials that originally were only about 10% of the sandstone mass. The clay minerals were altered by the high temperatures during the nuclear reactions, and in some cases were precipitated as the reactors cooled after they ceased to function. For reactor zones 1 and 2, desilicification was complete, while for other zones residual silica remained in the reactor core region at the borders of the core (Naudet, 1991). Figure 4 shows an idealized section for these two cases.



**Figure 4.** Stratigraphy of the reaction zones. *a) Complete desilicification of the C1 sandstone. b) Incomplete desilicification.*

The Franceville Basin was subjected to a burial stage with a maximum ambient temperature for the FA-FB contact region of 175°C, followed by an uplift period when the uranium ore accumulated in a thermal regime of about 130°C. Studies of oxygen isotopes and of clay mineral structures suggest temperatures of 300 to 350°C for the reactor cores, dropping to 200°C at the borders (Gauthier-Lafaye et al., 1989).

The uranium now in the ore deposit was derived from weathering of the Archaean basement complex and deposition of sediments as the basal conglomerate and

sandstone unit of the FA formation (Smellie, 1995). Concentration of the ore was achieved by dissolution in circulating oxidizing fluids that later encountered the reducing conditions in the black shales of the FB formation, giving precipitation of the uranium in a narrow region just below the base of the FB. The FB shales have total organic carbon contents of 5 to 20%, formed from algal matter trapped at deposition (Cortial et al., 1990). This material can reduce U(VI) in aqueous solutions to U(IV), thereby leading to its precipitation as  $UO_2$ . In some of the reactor zones at Oklo, zones 7 to 9 in particular, uraninite grains are found inside graphite nodules. It appears that the organic matter became liquid at reactor operating temperatures and then solidified to a graphitic structure as the reactors cooled. This produced an intimate mixture of uraninite containing fission products in a graphite matrix, which helped preserve the uraninite through time (Nagy et al., 1991).

## 2.1 CONDITIONS CONTROLLING CRITICALITY

There are many factors that control the ability of a system to sustain a nuclear fission chain reaction. The situations we are concerned with involve fission induced by the absorption of a neutron with suitable energy by a nucleus of fissionable material. At the start of the chain reaction, the fissionable material is  $^{235}\text{U}$  and to a much lesser extent  $^{238}\text{U}$ . The first neutrons must be derived from spontaneous fission and then be multiplied and conserved to produce the controlled chain reaction condition.

Fission in  $^{235}\text{U}$  is most probable when the neutrons have low energies, that is comparable to the kinetic energy at room temperature - in this case, the neutrons are "thermalized", becoming so-called thermal neutrons. The number of neutrons emitted during fission depends on the nucleus that splits. For  $^{235}\text{U}$ , the average number of neutrons emitted per thermal neutron absorbed is 2.1 (Friedlander et al., 1964). The neutrons are emitted with high energies and must undergo collisions with other nuclei in order to reduce their energies to the thermal region. Energy loss per collision is highest for masses that are comparable to the neutron mass. Thus, hydrogen is the most efficient moderator of energy for neutrons.

Each interaction of a neutron with a nonfissionable nucleus during the thermalization process carries with it the risk that the neutron will be absorbed without producing fission. The likelihood of absorption depends on the neutron energy at the time of collision and on the nature of the nucleus encountered. Some elements, such as the gadolinium isotopes with odd atomic weights, have extremely high affinity for absorption of neutrons. The measure of this affinity is the neutron capture cross section, given in units of "barns". The cross section for thermal neutron capture for  $^{157}\text{Gd}$  is  $2.42 \times 10^5$  barns, compared to that for Si at 0.16 barn,

and oxygen at less than 0.0002 barn (Friedlander et al., 1964). To evaluate the loss of neutrons through capture during moderation of their energies to the thermal regime, the detailed chemistry of the environment must be considered. Trace elements in the environment can produce more loss of neutrons than the major elements in the medium, even when the trace elements are present in very small amounts.

As fission occurs, fission products accumulate in the uranium minerals. These fission products can also absorb neutrons, so the loss factor for neutrons from absorption changes as the chain reaction proceeds.

For the chain reaction to occur under steady-state conditions, the number of neutrons from each fission event that survive the thermalization process and produce a new fission event must be 1. The ratio of the number of fissions produced by a set of neutrons to the number of fissions giving rise to those neutrons is called the multiplication factor,  $k$ . The multiplication factor that can be calculated from measured physical parameters is that for a uniform homogeneous medium with infinite size,  $k_{\infty}$ . The actual value of  $k$  must then be estimated by accounting for the loss of neutrons from the surface of systems with finite size. For these systems, the concept of critical size is extremely important, because loss of neutrons from the surface of systems with low concentrations of fissionable material can reduce a system with  $k_{\infty} > 1$  to a system with  $k_{\text{eff}} < 1$ . It is this property of concentration of fissile material together with considerations of absolute size and leakage of neutrons from the system that will be very important in the analysis of natural reactors and for the case of geologic disposal of fissile material.

One method to reduce loss of neutrons is to surround the fissile material with a neutron reflector. In nuclear reactors this is done with an engineered element. In the natural reactors at Oklo, the layers above and below rich ore zones could act as neutron reflectors if they did not contain high concentrations of neutron poisons (elements that have a high affinity for neutron capture).

The ratio of uranium isotopes 235/238 is important when considering fission chain reactions. This is because the number of neutrons that are produced per fission of  $^{238}\text{U}$  is only 1.3, compared to 2.1 for  $^{235}\text{U}$  or for  $^{239}\text{Pu}$  (Friedlander et al., 1964). In addition,  $^{238}\text{U}$  requires fast neutrons for fission; with neutrons of thermal or moderate energies,  $^{238}\text{U}$  can capture a neutron and the product nucleus will eventually decay to  $^{239}\text{Pu}$ . In modern commercial reactors using normal water as the moderator, the isotopic composition of the uranium is enriched in 235 isotope to facilitate achieving criticality. The average fuels for these reactors have about 3 to 4%  $^{235}\text{U}$  in the uranium at the start of fuel use.

## 2.2 CRITICALITY AT OKLO - A MODEL FOR CALCULATION

Analysis of criticality in engineered systems can be done using computer models of the system. Even though the systems are complex, they can be modelled as multiple units of subsystems that are themselves homogeneous. The natural systems at Oklo were clearly not so simple. The discussion that follows is a summary of material from the work of Naudet (1991) unless otherwise noted.

In addition to the problems caused by geologic heterogeneity, there are difficulties because important nuclear parameters must be deduced from the reaction products rather than measured directly. Fluence, the time-integrated number of neutrons passing through a sphere of unit cross-section area for the reaction regime, is one example. The best fluence indicator for Oklo is probably  $^{143}\text{Nd}$ . The cross section for thermal neutron capture for  $^{143}\text{Nd}$  is 320 barns and changes as neutron velocity increases in proportion to  $1/v$ . Any mobilization of Nd during or after reactor operation will affect the estimate of fluence.

Another important parameter in the model is the number of fissions per nucleus of  $^{235}\text{U}$ . This will be used to relate fission yield and fluence to the age of the reactions. The duration of the reactions is modelled using a reaction of  $A \rightarrow B$ , where A has a half-life about the same as the duration of the reactions. Reactions used for Oklo were  $^{239}\text{Pu} \rightarrow ^{235}\text{U}$  and  $^{99}\text{Tc} \rightarrow ^{99}\text{Ru}$ . The latter reaction was less useful because some of the Tc migrated away from the reactor sites during operation or after the reactions stopped.

Samples selected from the Oklo cores and boreholes had to be sufficiently large to be representative of the "average" flux, but small enough not to obscure important details of the processes. A sample size of 2-cm cubes was selected, with spacing of 10 cm between samples. Because the fission process occurs best with thermal neutrons, the richest ore does not necessarily show the largest amount of fission. The rich ore generates initial neutrons, but they must be slowed by collisions in the environment, so more fission may be produced on a per-initial-nucleus basis in a poorer ore patch nearby. Thus, modelling that includes heterogeneity will be important in understanding the distribution of isotopic depletions at Oklo.

One of the particularly difficult aspects of modelling the Oklo reactors is that one must assume that no migration of uranium or fission products used in the calculation occurred after significant irradiation had taken place. Then the results of the calculations must be used to check whether this assumption was true. No single set of calculations will allow this to be done. Only with many sets of calculations and the accumulation of consistent sets of information can any conclusions be decisively reached.

Another particular difficulty with modelling of Oklo criticality and reaction progress is that only the final state of the reactors is seen. This state is strongly affected by tectonic events that occurred near and at the end of reactor operations. The clues contained in the rocks must be investigated thoroughly and pieced together to work out a picture of the reactors throughout their functioning.

The conceptual model for reactor operation starts with a sandstone section of the C1 bed that is particularly rich in uranium. Examples of these "rich sandstones" are still present at Oklo. If the sandstone consisted of 80% SiO<sub>2</sub>, 10% clay cement, and 10% UO<sub>2</sub> and was subjected to desilicification because of circulation of hydrothermal fluids during reactor operation, the end product would be 50% uraninite and 50% clay by weight. The volume reduction is a factor of 7, changing an original 5 meter thickness of sandstone into a 70 cm thick reactor zone called *faciès-pile*. Evidence that this mechanism took place comes from the continuity of the sandstones with the *faciès-pile*, the microscopic evidence for silica dissolution, and the interpretation of the faulting patterns in the reactor sections.

The start of reactor operation was probably through response to extension forces in the rocks, possibly occurring during uplift, giving rise to faulting and an increase in porosity in an already existing rich ore zone in the sandstones. Water could then enter, start dissolving quartz, provide a moderator for the neutrons and support the nuclear reactions. Once desilicification was essentially complete, which could have been in a short time compared to the length of time the reactions went on, no significant amount of uranium could be transported in. If it had, the homogeneity of the uranium 235 depletion pattern would have been disturbed.

The final state of the Oklo reactors must form the starting point for detailed modelling of criticality. Porosity and water content as now seen at Oklo may be much less than just before reactions stopped, and the structures may have collapsed, but the chemistry of the reactor zones now reflects the chemistry at the end of the reaction period. Table 3 gives the chemical composition of the two important clay minerals, magnesian chlorite and illite, and a list of minor and trace elements that are important in the neutron balance calculations. The kaolinite that is now seen in some of the reactor zones is interpreted to be a later alteration product, so is not included in the criticality model.

Naudet (1991) and colleagues developed a special computer code, BINOCLE (Bilan Neutronique Oklo - Neutron Balance for Oklo) to analyze the various aspects of criticality in the ore deposits. Neutrons travel about 10 cm after "birth" and undergo 30 to 40 collisions in that path, so in most cases the average environment over 10 cm is what is important. In some cases, however, such as the resonant absorption of a neutron by <sup>238</sup>U, which occurs over a narrow energy band, a much finer scale of modelling is required.

**Table 3. Chemical compositions of materials important to the neutron balance calculations.**

Minerals (in wt %)	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	FeO	Fe <sub>2</sub> O <sub>3</sub>	MgO	K <sub>2</sub> O
Chlorite	41	25	13	4	10	1
Illite	44	32	9	3	3	5

**Minor elements:** CaO: 0.5 to 4% (strongly correlated with U),  
Na<sub>2</sub>O ~ 0.6%, BaO ~ 0.3%, TiO<sub>2</sub> ~ 0.7%,  
MnO ~ 0.5%, Zr ~ 0.1%, trace C, S, P, As

**Minor elements with high neutron capture cross sections:**

V (200-800 ppm)  
Cr, Ni, Co, Cu (20 to 100 ppm each)

**"Poisons":** B (100 to 600 ppm, with K in illite)  
REE: Sm (10 to 30 ppm); Gd ~ 0.6 x Sm (both correlated with U)  
Li < 100 ppm;  
Cd < 0.1 ppm

Mean capture cross section for the above assemblage is 0.75 mm<sup>2</sup>/g (excluding REE) + 0.25 mm<sup>2</sup>/g for REE.

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The model was formulated with 5 components: uranium as UO<sub>2</sub>, argillaceous gangue, water in various forms, neutron poisons, and quartz that had not yet dissolved. Since there are interrelations among these parameters and the chemical compositions, it is useful to set out some definitions and equations.

X = wt. gangue/g UO<sub>2</sub>  
Y = wt. water /g UO<sub>2</sub>  
Q = wt. quartz/wt. gangue  
x = capture by poisons expressed as equivalent ppm <sup>10</sup>B in the gangue.

The value of X is calculated from the present uranium content of the ore by correcting for the ingrowth of radiogenic lead over the 1950 million years since the reaction time and by correcting for the present amount of water. For a Pb/U = 0.1 and a water content of 14.5%, this gives

$$X = (100 - 1.235t)/1.863t$$

where t is the present U content of the ore.

The code uses six explicit oxides in the calculations: Si, Al, Fe (as FeO), Mg, K, and Mn. All trace elements were included in the Mn value to account for their neutron capture effects. Most calculations were done with a standard composition for the gangue, with adjustments for elements that correlated with uranium content,  $t$ . The standard formulation is shown in table 4.

**Table 4. Model chemistry for the gangue used in criticality calculations, in weight percent. Present uranium content in weight percent in the ore =  $t$ .**

---

SiO <sub>2</sub>	44.5 - 0.05 $t$
Al <sub>2</sub> O <sub>3</sub>	30.5 - 0.05 $t$
MgO	11.7 - 0.06 $t$
FeO	10.0 + 0.15 $t$
K <sub>2</sub> O	2.5
MnO	0.8 + 0.01 $t$

---

Two kinds of water were included in the model: "structural" water that was bound up in the clay minerals, and "free water" that occupied open porosity in the rocks. Two new variables are then defined

$$m' = \text{wt. ratio of structural water/clay}$$

$$v = \text{volume of open porosity/ g UO}_2$$

This gives a relationship for total water,

$$Y = m'X + vd_{\text{water}}$$

where the density of water,  $d_{\text{water}}$ , varies with temperature. Hydrocarbons were not explicitly included in the general form of the model, but were approximated by an equivalent amount of water to account for the hydrogen.

BINOCLE did not use elaborate calculations of neutron spectra (energy distributions) since there was little plutonium compared to <sup>235</sup>U and the only resonant cross section for neutron capture that was important was that for <sup>238</sup>U. To check this assumption, Naudet (1991) performed calculations using the CEA APOLLO code, which included elaborate spectral distinctions, and found that for uranium contents between 2 and 60% agreement between the two codes was 0.1% or better.

Other parameters that are used in the calculations are

$e$  = isotopic percentage of  $^{235}\text{U}$  today ( $e = 0.72$  if no irradiation occurred)

$m$  = the volume occupied by free water (ratio to gangue plus ore)

$B^2$  = geometric Laplacian used to characterize the overall exchanges of neutrons in the environment

As an example of the use of the code, Naudet (1991) gives a calculation for nonirradiated uranium with  $t = 40$ , 11% structural water in the gangue, poisons  $x = 50$  (equivalent ppm  $^{10}\text{B}$ ), weight ratio of gangue to  $\text{UO}_2$  in the grains = 0.2, and with  $m = 0.5$ , which means that free water occupies a volume that is half of that occupied by the uranium and the gangue. With this set of parameters, Naudet calculates what is the critical thickness above which neutrons will be retained in the ore to cause criticality. This is done using the Laplacian relationship  $B^2 = (p/\text{He})^2$ , where  $\text{He}$  = critical thickness. The calculation gives the result that  $\text{He} = 63.1$  cm. This means that an ore zone with the parameters used in the calculation would not have reached criticality at the time 1950 My age (with  $T = 160^\circ\text{C}$  and  $P = 500$  bars) if its thickness had been less than 63.1 cm. The lack of criticality was caused by neutron escape at the boundaries of the rich zone.

A series of parametric analyses were done to illustrate how the criticality threshold varies with ore grade, water abundance, and porosity available for water entry. Figure 5 illustrates the change in effective neutron multiplication factor with water quantity for ores ranging from 20 to 60%. The most efficient moderator value is for  $Y = 0.4$  for ores up to 40% uranium. These calculations also clearly show the importance of ore concentrations to the neutron balance. For ores with less than 40% uranium it will be difficult under any conditions to achieve  $k_{\text{eff}} \geq 1$ .

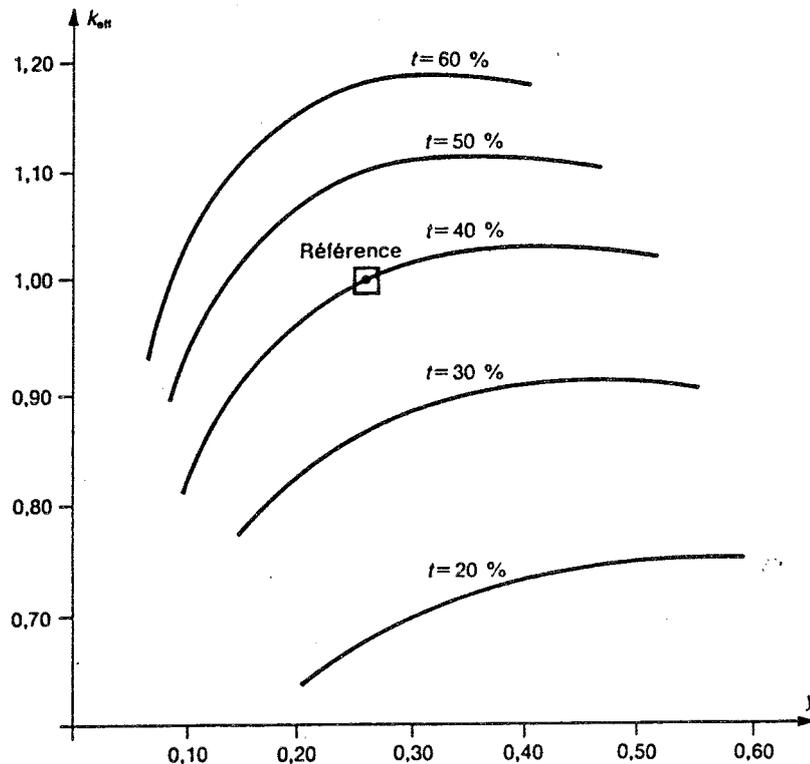
A series of calculations was also made by Naudet (1991) to study the effect of residual quartz in the ore body on the value of  $k_{\text{eff}}$ . The residual silica acts as a diluent, lowering the uranium content of the ore. If quartz dissolves, the ore grade goes up, but if the porosity left by the dissolution is lost, then water entry to the ore is limited and achieving criticality is harder. For moderately high grade ores, with  $t = 35\%$ , dissolution of quartz accompanied by entry of water can result in criticality. For lower grade ores, quartz dissolution accompanied by water entry will not give criticality. (Recall that  $t$  is the grade of the ore as seen today, not the uranium concentration at the time that desilicification started. For  $t = 35\%$ , and an original quartz to clay ratio of 9, the original sandstone uranium concentration would only have been about 6.5%).

Parametric calculations concerning the effects of organic matter showed that for moderate levels of  $(\text{CH}_2)_n$  hydrocarbons the effects were similar to those of a

similar amount of water. If the organic matter was added at the expense of silica, the level of  $t$  needed to achieve criticality dropped to less than 30%.

Temperature affects the reactions in three ways. First, the neutron capture cross sections for fission will vary because "thermal" neutrons are those with energies equilibrated with their surroundings. Second, neutron escapes increase somewhat as temperature increases. Finally, resonant absorption by  $^{238}\text{U}$  increases as temperature goes up. The biggest effect of temperature will come from its effects on the density of water, which also depends on pressure and salt content. The overall effect of increasing temperature from 160°C to 360°C on the reactions is very negative in terms of reactivity.

The progress of the reactions themselves, with all other parameters held constant, affects the reactivity of the system. If we start with  $k_{\text{eff}} = 1.000$  and  $e = 0.72$  and let the system react,  $e$  (the present day abundance of  $^{235}\text{U}$ ) will decrease because of fission, fission products will accumulate, and poisons will burn out through absorption of neutrons. This produces an increase in  $k_{\text{eff}}$  to a maximum of 1.034 when  $e$  is 0.67%. Then  $k_{\text{eff}}$  decreases to 1.023 when  $e$  is 0.62 and to 0.9979 when  $e = 0.57\%$ . In other words, for the system examined, the maximum extent of reaction produces a present day isotopic abundance of  $^{235}\text{U}$  of 0.57%.



**Figure 5.**  $K_{\text{eff}}$  as a function of amount of water for different uranium concentrations. (from Naudet, 1991)

## 2.3

### CRITICALITY AT OKLO - EXAMINATION OF THE REACTOR ZONES

Before modelling of the reactor zones could be done, many simplifications and adjustments were necessary. Naudet (1991, chap.6) began by assuming that the reactors could be considered as essentially a flat cakelike structure, where neutron escape would only be important in one dimension and the medium could be considered to be uniform and large (in the first approximation) in the two lateral dimensions. Samples from both drill cores and from mining faces intersected the reactor zones at an angle because the beds were dipping at about  $45^\circ$ . This produced distortions in the dimensions along the core, making the beds look thicker than they were. The distances were corrected by multiplying the measured distances by the cosine of the dip angle. In many cases, dips were not well known, introducing uncertainty in the actual distances.

Measurements of the neutron spectral index from isotopic data indicated that the environment during the reactions had a much greater porosity than is presently the case. The beds must have compacted after the reactions were finished and this change in geometry must be considered in the modelling. Measurement of actual present porosity in the reactor zones is difficult because the coring device caused breaking up of the rock and increased the porosity by an amount estimated by Naudet (1991) to be 10 to 15%. In the end, Naudet (1991) estimated from the spectral index that the porosity must have been about 37% and, to correct presently seen thicknesses back to the precompacted state, one must multiply the observed lengths after correction for dip by a factor of about 1.2 to restore the precompacted porosity.

Generally the Oklo reactor zones show a sharp decrease in uranium concentration at the edges of the rich zone. To simplify the modelling, the ore is taken to be a region of uniform high concentration bounded by a zone with low uranium values. This is important because the U-poor zone acts as a reflector and sends some neutrons back into the rich zone to cause reaction. This decreases the thickness of ore required to reach criticality in comparison with a calculation that assumes a void surrounding the rich ore. For the case discussed in the previous section, consideration of the reflector gain reduces the thickness required to achieve criticality from 63 cm to about 43 cm.

Combining the effects of dip between  $30$  and  $45^\circ$ , so that  $\cos\alpha = 0.7$  to  $0.85$ , the correction for compaction of 1.2 to 1.25, and the reflector, Naudet (1991) arrived at an equivalent thickness for the reactor zones during the reaction period of 50 to 75 cm for the materials in zones 1 and 2.

The conditions for initial criticality were examined using  $e = 0.72$  (present day ratio for unirradiated uranium) and  $Q = 0$ , being completely desilicified ore. The

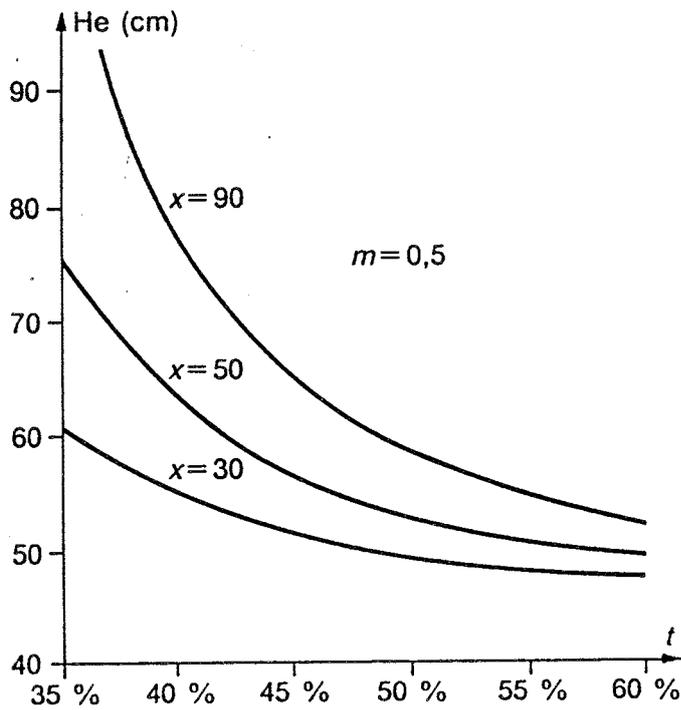
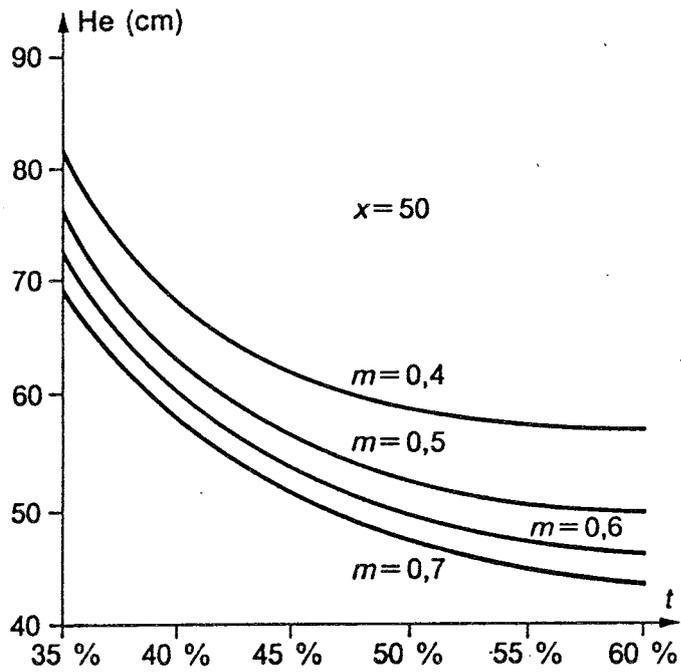
medium has four intrinsic parameters that control criticality:  $t$ , the ore concentration of uranium;  $x$ , the amount of poisons expressed as equivalent  $^{10}\text{B}$ ;  $m$ , the amount of the volume occupied by free water; and  $He$ , the equivalent thickness of the rich zone. If  $m$  and  $x$  are fixed, the condition that criticality be reached provides a relationship between  $t$  and  $He$ . Figure 6 shows this relationship for various values of  $m$  when  $x$  is fixed at 50 ppm equivalent boron, and for various values of  $x$  when  $m$  is fixed at 0.5, which represents free water occupying 1/3 of the volume (i.e., 33% porosity). For ores with uranium less than 45%, the thickness of rich zone needed to achieve criticality increases dramatically as the amount of poisons increases. If we take reference values of  $x = 50$  and  $m = 0.5$ , criticality cannot be reached unless  $t > 28\%$ , even at infinite thickness, and  $He$  must be at least 50 cm, regardless of how rich the ore is.

The role of the neutron poisons can be seen more clearly by calculating the amount of poisons that can be allowed for a fixed amount of water ( $m = 0.5$ ) for different combinations of  $He$  and  $t$ . This relationship is illustrated in Table 5. The amount of rare earth elements presently in the rich ore has a capture section equivalent to about 15 ppm  $^{10}\text{B}$ . The boron-10 is about 20% of total boron, and the boron in the gangue material now is about 50 to 300 ppm, giving reasonable poison values of 25 to 75 for the ore zones.

**Table 5: Critical value of  $x$  (ppm equivalent of  $^{10}\text{B}$ ) for different combinations of the ore grade and thickness of the rich zone. (From Naudet, 1991)**

He (equiv. thickness)	t (U%)			
	35%	40%	45%	50%
50 cm	6.6	16.4	25.8	34.5
60 cm	28.8	43.7	59.5	76.5
70 cm	43.1	61.2	81.1	103.3
80 cm	52.7	73.0	95.7	121.5

All of these examples were calculated for an age of the reactions of 1950 million years. If the reactions were younger, the amount of poisons compatible with maintaining criticality would be less. For reaction ages of 1800 and 2000 million years the difference in allowable poisons is a factor of 2 (Naudet, 1991). Having

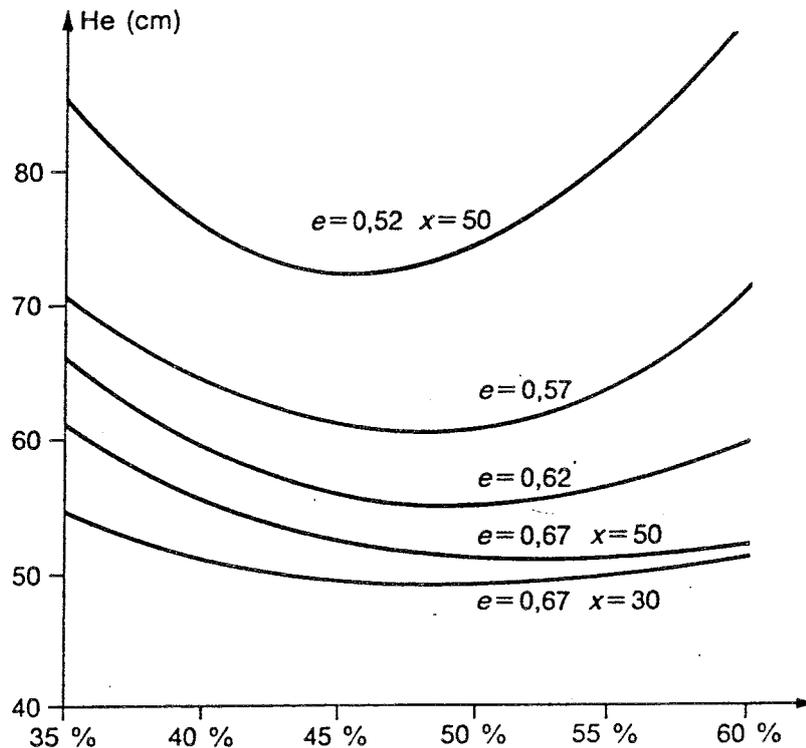


**Figure 6.** Thickness required for criticality for the rich ore as a function of the uranium content,  $t$ , for different values of the amount of water,  $m$ , and poisons,  $x$ . (from Naudet, 1991).

some poisons in the reactor zones at the start of reactions is important because they act as a reserve of reactivity. They are "burned out" by absorbing neutrons as the reactions proceed, but the fission products that grow in can also absorb neutrons. If there were no poisons initially present and if the reactor zones were just critical, then as soon as a little fission occurred, the reactions would stop through self-poisoning.

Modelling of criticality at the moment when the reactions stopped uses the final measured isotopic depletion,  $e$ , as a fixed parameter. The amount of quartz remaining in the reactor zone is also known by observation and is  $Q = 0$  for zones 1 and 2. In addition, the original neutron poisons would have burned out and residual neutron capture equivalents can be calculated based on the rock chemistry and the fission product yield. Since measurements of the spectral index fix the amount of water rather well, and these measurements all indicated  $m$  between 0.5 and 0.65, calculations were done using  $m = 0.5$ .

Figure 7 shows the calculated minimum equivalent thickness to support criticality in the end conditions for several combinations of isotopic composition and initial poison level,  $x$  (now burned out), as a function of the ore grade. In the rich ores with higher burnup (lower  $e$ ) the neutron capture by fission products becomes



**Figure 7.** Critical thickness ( $He$ ) as a function of the ore grade for different values of the isotopic depletion ( $e$ ) and amount of poisons ( $x$ ) (Naudet, 1991).

important, so increasing ore grade ( $t$ ) can increase the minimum ore thickness required to reach a given isotopic depletion. The level of isotopic depletion achieved in the Oklo reactors depended for the normal values of the rich ore concentrations on the thickness of the rich zone. Reactor zones with only slight depletions were originally probably thinner than those in which the greatest depletions are found.

In order to understand how the Oklo reactors first started to function it is necessary to consider the process of desilicification and whether the reactors can begin to function before silica is removed to a significant extent.

Calculations were done by Naudet (1991) to show how the effective neutron multiplication factor,  $k_{\text{eff}}$ , changed as quartz was removed from ore zones with different properties. For a thick region of low uranium concentration,  $k_{\text{eff}}$  always increased as quartz was removed. The same was true for a thick zone of rich ore; however, for a very thin zone of very rich ore the  $k_{\text{eff}}$  passed through a maximum when about half of the quartz had been dissolved. Zones 3 through 5 at Oklo are examples of the very thin, very rich ore case, where it is impossible to explain criticality if the ore had been completely desilicified when the reactions started.

So far, we have considered how the condition of criticality was maintained once the reactions were under way. To understand how the reactions started in the first instance, we must consider sandstones that are at ambient temperature for the burial conditions,  $160^{\circ}\text{C}$ , and that have not yet been desilicified. While the exact conditions of the sandstones prior to the start of reactions are impossible to know precisely, the indications are that the ratio of quartz to clay,  $Q$ , was between 6 and 10. Some C1 sandstones, however, have less quartz. To cover the possibilities, Naudet (1991) calculated cases for  $Q = 8$  and  $Q = 4$ . For the case where  $Q = 8$ , criticality could only be initiated in the intact sandstones when the ore had enough uranium to give a concentration of 45 to 50% after desilicification and, more importantly, when the porosity was on the order of 10% to allow ingress of water.

The normal levels of porosity in the intact sandstones are 1 to 5%, so one must call upon tectonic activity to increase the porosity in order to allow enough water into the rock to efficiently moderate the neutrons. As the level of poisons is increased, the porosity required increases from 10% when  $x = 30$  to 13% when  $x = 50$ .

The conditions for starting the reactions are summarized in Table 6. The calculations were done assuming optimal conditions for moderation, which is a porosity of 15 to 18%. The ore grade prior to desilicification,  $t_g$ , is the average for a 2-meter thickness of original ore with the given  $Q$  value. For the highest  $Q$  values, which are most likely to represent the original sandstones before reactions started, only those that produce very rich final ores, with present ore grades,  $t$ , of 45% or more, can become spontaneously critical. While such ores are

**Table 6: Critical ore grade in the vicinity of optimal moderation.**

Q	x = 30		x = 50		x = 70	
	tg %	t%	tg %	t%	tg%	t%
4	11.8	34.6	13.9	38.5	16.0	42.0
6	10.7	38.8	12.2	32.0	13.8	44.9
8	9.9	42.0	11.1	44.8	12.4	47.2

not unknown at Oklo, they are by no means the rule. Naudet (1991) concludes that only under certain very favorable conditions could the reactions start spontaneously and begin heating the rocks. Once the heat began to raise the temperature of the rocks, silica dissolution could begin and thermal convection could begin to transport silica out of the environment, increasing porosity and improving the neutron balance. The first spontaneous reactor site may have allowed desilicification of nearby ores to begin and then produce somewhat more favorable conditions for starting the reactions in other locations.

## 2.4 THE UNIQUENESS OF OKLO

After Oklo was understood to contain fossil chain reaction sites, searches for other such natural reactors have failed to produce another example. The more  $^{235}\text{U}$  an ore contains, the easier it is for a given mass of uranium to reach criticality. As we go back in time, the relative abundance of  $^{235}\text{U}$  increases greatly with respect to the longer-lived  $^{238}\text{U}$  isotope. At the time the Oklo reactions occurred the amount of fissile  $^{235}\text{U}$  was about 3.5%, similar to the enrichments of  $^{235}\text{U}$  used in modern engineered power reactors. The variation in uranium isotopic abundances with time is shown in Table 7.

**Table 7: Concentration of  $^{235}\text{U}$  as a function of its age. (From Naudet, 1991)**

800 MA - 1.38%	1600 MA - 2.61%	2400 MA - 4.9%
1000 MA - 1.62%	1800 MA - 3.06%	2600 MA - 5.72%
1200 MA - 1.90%	2000 MA - 3.58%	2800 MA - 6.66%
1400 MA - 2.23%	2200 MA - 4.19%	3000 MA - 7.76%

Since older ore bodies could achieve criticality more easily than younger ones, why have we not found another fossil reactor of greater age than Oklo? This apparent paradox was examined by Naudet (1991), who concluded that the reason was tied to the evolution of the earth's atmosphere. Ore deposits with ages greater than about 2000 million years are quartz pebble conglomerate ores with low uranium levels and that accumulated through the action of gravity. This process operated under conditions where there was very little oxygen in the atmosphere.

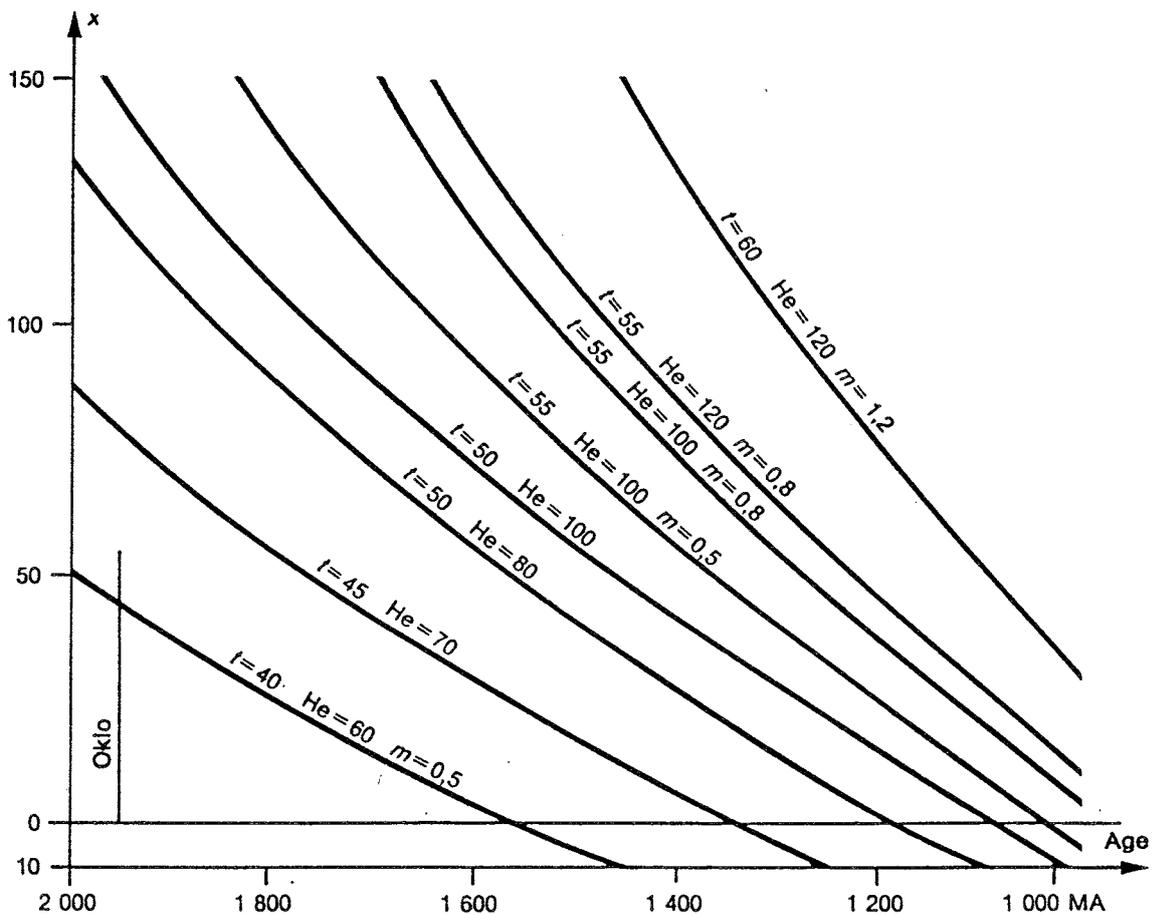
The first significant levels of oxygen appeared in the atmosphere about 2000 million years ago and is evidenced by the first appearance of oxidized iron in sedimentary ores known as "red beds" (Smith, 1981). The oxidation of uranium from U(IV) to U(VI) occurs at about the same redox potential as that for Fe(II) to Fe(III). The lower oxidation state of uranium (IV) has a much lower solubility than uranium (VI). It is likely that the very rich ores needed to produce an Oklo-type deposit could not accumulate until there was enough oxygen available to produce the higher oxidation state of uranium, and its transport and enrichment through processes of dissolution and reprecipitation.

The occurrence of oxygen in the atmosphere is linked to the development of biological activity that led to photosynthesis becoming an important process. This changed the carbon dioxide in the atmosphere into oxygen and organic carbon on the lithosphere. Deposition of the organic carbon then could provide strongly reducing conditions locally in rock zones. The uranium deposit at Oklo is associated with a zone rich in organic matter that could have provided the reducing environment to change dissolved uranium (VI) into uranium (IV), thereby causing it to precipitate. The narrow zone of the C1 bed at the base of the organic rich FB formation argues strongly for this origin for the ore (Cortial et al., 1990).

Naudet (1991) investigated the conditions that would be needed for ores younger than Oklo to reach criticality. In comparison to Oklo conditions, for a younger ore there must be either a richer uranium concentration, a thicker zone of rich ore, fewer poisons present, or more favorable neutron moderation conditions through more water for cases where Oklo was not optimally moderated. Figure 8 shows the increasing demands on conditions to achieve criticality as the age of the ore is decreased. The curves are plotted as the maximum value of poison level,  $x$ , to ensure criticality as a function of age for the fixed values of present day uranium concentration,  $t$ , equivalent thickness of the rich zone,  $H_e$ , and  $m$ , which is the fraction of the total volume taken up by free water.

The curves in Figure 8 show that there are possible combinations of parameters that could produce criticality at ages less than 2000 million years. Naudet (1991) discussed the plausibility of achieving the required conditions. The very low level of poisons at Oklo is possible because vanadium, which is a strong neutron capturer and which usually is concentrated with uranium, is for unknown reasons

virtually absent at Oklo, even though a vanadium deposit occurs nearby at Mounana. Manganese is another neutron capturing element that is low in the Oklo ores, even though it is abundant in nearby strata, and frequently is found in rather high levels in uranium ores. Thus, the level of poisons at Oklo is already rather lower than expected for such a rich ore deposit and the likelihood of finding another ore with similar richness and low poisons must be considered to be low.



**Figure 8.** Critical value of the level of poisons,  $x$ , as a function of age for different combinations of ore grade,  $t$ , critical thickness,  $He$ , and volume ratio occupied by water relative to ore plus gangue,  $m$ . (from Naudet, 1991).

While the curves can be calculated for combinations of parameters such as equivalent thickness of 120 cm and present ore concentration of 60% uranium, this

combination represents an ore that represents a concentration of uranium that is twice as much as found at Oklo. The lateral extent could be reduced somewhat to reduce the constraints, but the two lateral dimensions must be large enough so that they do not result in loss of too many neutrons. Naudet (1991) concludes that as the assumed age of the ore body decreases, the probability of finding the required combination of conditions decreases drastically. For ages less than 1500 million years, the probability of all conditions being satisfied simultaneously is so low that it is unrealistic to think that such an occurrence will be found.

Ages older than 2000 million years provide an easing of conditions, but with the problem of finding a mechanism to cause the enrichment of the ore. Table 8 shows the minimum value of present day uranium content of sandstones of various ages to reach criticality. The ore thicknesses must be large and the concentrations of uranium are very much higher than those found in old ores. The conditions were chosen with high values of quartz and low amounts of clay in order to minimize absorption of neutrons in the gangue. The lowest concentration of uranium at 2800 million years is only about 3%, but it is difficult to see how this could be reached by using only detrital processes.

**Table 8: Critical ore grade in % for sandstones as a function of their age in million years. Q = weight of quartz/weight of other non-ore gangue, x = ppm equivalent <sup>10</sup>B neutron poisons, He = equivalent thickness in cm.**

Q	x	He	Age in millions of years				
			2000	2200	2400	2600	2800
8	50	250	11.35	8.60	6.42	4.97	3.94
8	30	250	9.60	7.24	5.52	4.32	3.42
10	30	250	9.27	6.89	5.20	4.04	3.18
10	30	380	8.50	6.25	4.80	3.78	2.99

## 2.5 VARIABILITY AT OKLO - A COMPARISON OF ZONES 2 AND 9

By the fortunate accident of its discovery time, reactor zone 2 was the most thoroughly studied zone. Zone 2 had a relatively simple stratigraphy, even after the compaction of the desilicified core, and was not greatly affected by sliding of material away from its original reaction location. The completeness of the reactor section allowed Naudet (1991) and his colleagues to develop a detailed picture of the reactor before, during, and at the close of its activities. If the modelling of reactor zone 2 had not been so successful, the difficulties encountered in dealing

with zones 3-5 and 7-9 might have caused such discouragement as to convince those involved in the studies to abandon the effort to develop a complete understanding of these zones.

Zones 1 through 6 form a coherent group of geographically closely related reactor zones at the northern end of the Oklo deposit. The core regions of zones 1 and 2 were completely desilicified, while for zones 3-5 there was a residual C1 sandstone border on either side of the argile de piles. The latter zones contained thin regions of very rich ore, required very high porosities to function, and probably were extinguished prematurely through collapse of their sandstone roofs giving a loss of the needed porosity. This is evidenced by low levels of reaction in these zones. Despite these differences in operating details, the basic temperature, pressure, and tectonic setting for zones 1 through 6 were the same. Another similarity of these zones is the very low abundance of organic matter in them.

Zones 7 to 9, by contrast, have abundant organic matter. Uraninite is found encased in bituminous/graphitic nodules that form a locally highly reducing environment, thought to enhance the geochemical stability of the uranium ore and some of the fission products. In addition, the temperatures recorded in fluid inclusions in zone 7-9 are lower than those for zone 2. Desilicification in these zones did not occur in the core, but large volumes of sandstones above the core have been dissolved away. Naudet (1991) interprets this as an indication of boiling in the core region, with little silica dissolved from there, but with condensation higher up and dissolution and transport of silica then. This is consistent with lower pressures for zones 7 to 9 when they were operating and implies a slightly younger age for them. The age difference could be quite small; what is required is for some continued uplift to have occurred between the operation of zones 1 to 6 and zones 7 to 9, bringing the reactor zones closer to the surface.

Because organic matter was abundant in zones 7 to 9, the entry of water was not so important in causing criticality. In fact, the absence of criticality for these zones at the time (i. e., pressure) that zones 1 to 6 functioned suggested that the limiting feature here was uranium concentration. Naudet (1991) suggests that criticality was achieved when the ore was closer to the surface and when oxidizing fluids could cause dissolution of uranium from other parts of the deposit and that uranium could be trapped in the organic matter.

Loss et al. (1988) studied samples from reactor zone 9 and reconstructed a picture of the reactor conditions using models similar to those used by Naudet (1991). They found that the neutron fluence was  $3.6 \times 10^{20}$  n/cm<sup>2</sup>, somewhat smaller than for zone 2, which had  $1 \times 10^{21}$  n/cm<sup>2</sup>. The duration of the reactions was  $2.2 \times 10^5$  yrs., in comparison with an indicated duration of  $8 \times 10^5$  yrs. for zone 2. The coefficient of restitution, being the fraction of uranium-235 that was restored by alpha decay of plutonium-239 (formed by neutron absorption by uranium-238) for

zone 2 was 0.52 (Naudet, 1991), while that for zone 9 was 0.48 (Loss et al., 1988). The similarity of the coefficients of restitution suggests that the "standing crop" of plutonium in the two reactor zones was similar when they were functioning, and that the rate of functioning was similar for both reactors. This is further indicated by the proportions of fissions due to  $^{239}\text{Pu}$  in the two reactor zones, which are given as 3.8% for zone 2 and 4.5% for zone 9. Thus, despite the differences in pressure, temperature, total time over which function occurred (where functioning may not have been continuous), total fluence, and abundance of organic matter, the reactor zones appear to have remarkable similarities.

## 2.6 CHEMICAL AND PHYSICAL STABILITY OF THE REACTOR ZONES

The original discovery of the Oklo natural reactors was based on the deficiency in the 235 isotope of uranium and the finding of fission products associated with the uranium that were consistent in their isotopic patterns with the products expected from  $^{235}\text{U}$  fission. Initial investigations indicated that to a large extent the reactors had functioned under contained conditions in which the uranium and the fission products remained in the ore. Later studies have led to a revision of this point of view (Naudet, 1991).

Some fission products that are volatile at low temperatures, such as the rare gases and iodine, and those with very high solubilities, such as cesium, have long been known to have escaped from the reactor zone. This probably happened at the time the reactors were functioning. Other fission products, such as technetium, have been partially lost. This is important when one tries to use fission isotope abundances to deduce reactor operating parameters.

Neodymium isotopes are used to estimate the fluence of neutrons in the reactor, and it must be assumed that the uranium and neodymium remained together for the whole time that significant fission was occurring. The stability and retention of uranium can be checked by studying the Th/U ratio in the ore. When  $^{235}\text{U}$  absorbs a neutron and does not fission,  $^{236}\text{U}$  is formed. This subsequently decays to  $^{232}\text{Th}$ . The original ore had very little thorium in it, so the present amount of  $^{232}\text{Th}$  can be corrected for the small initial contribution. Values for the total reaction amount based on thorium are 1.03 times those based on neodymium, indicating a small loss of Nd relative to Th. If uranium had been lost during the reactions, the  $^{236}\text{U}$  that subsequently decays to thorium would have been too low, and the calculations based on thorium would have been low compared to those based on Nd.

When all of the measurements and calculations are considered, the most consistent picture is found when one assumes that uranium and thorium have remained almost completely inside the reactors and in almost the same location relative to each other. That is, movements of uranium and thorium have been on the scale of mm

to cm, not meters. For Nd, about 2.5% seems to have been lost from the core regions and is now found in the borders of the reactor zones, which show an excess of fissionogenic neodymium. About 4.5% of the samarium and 10% of the gadolinium also seem to have been moved out of the reactor core and into the border regions (Naudet, 1991).

The calculated values of the coefficient of restitution showed that half of the  $^{235}\text{U}$  that fissioned in the reactor zones was produced by decay of  $^{239}\text{Pu}$ . Since the  $^{239}\text{Pu}$  was formed by neutron capture by  $^{238}\text{U}$  and the burnup of the  $^{235}\text{U}$  shows a fine-structure that can only be explained by lack of mixing of uranium within the reactor zones even during operation, the plutonium must have remained almost entirely within the uranium that produced it. That is, movements of plutonium within the reactor were confined to mm to cm distances.

Only one instance of plutonium migration out of the reactor core has been found. Bros et al. (1993) report the discovery of a clay mineral approximately 3 meters above reactor zone 10 containing 3.34 ppm U, of which 2.47 ppm was leachable from the surface in dilute acid and was somewhat depleted in  $^{235}\text{U}$ . This depleted uranium would have been sorbed onto the mineral surface from fluids circulating since the mineral formed and indicates a small level of uranium movement in the vicinity of the reactor core. The residue of the clay mineral after leaching contained 0.83 ppm U with an isotopic composition of  $235/238 = 0.0105$ . This is about 30% more  $^{235}\text{U}$  than is present in normal uranium and was interpreted to represent plutonium incorporated into the clay mineral as it crystallized during or shortly after the reactor zone 10 was operating. The chemistry of the clay is Fe,Al chlorite. The highly reducing conditions imposed by the abundant organic matter could lead to selective dissolution of some Pu(III), leaving behind the less soluble U(IV) (Bros et al., 1993). This solution that was relatively high in Pu/U could then have been incorporated into the newly forming clays outside the reactor zone. The low contents of both U and implied Pu in the clay suggest that the concentrations of both of these elements in the solutions circulating around the reactor zone were low.

One region has been identified in reactor zone 1 that shows evidence for the transport and mixing of depleted uranium, probably during reactor operation. In two drill cores, SC 30 and SC 30*bis* somewhat depleted uranium with  $^{235}\text{U} = 0.687\%$  is found uniformly distributed over a distance of 1.2 m in the border region of the reactor zone. Naudet (1991) interprets this to be an indication of circulation of fluids containing uranium and deposition of the uranium in the reducing conditions at the border near the FB formation. Naudet attributes this uranium movement to the importance of tectonic fractures in the border region, instead of the normal condition in zones 1 and 2 where the newly-formed clays effectively sealed the reactor zones and held in the uranium.

The mechanical stability of reactor zone 2 was the reason that a detailed model for the reaction period could be developed. The final compaction of the reactor zone porosity occurred in a relatively gentle and uniform manner, leaving the reacted materials *in situ*. In reactor zones 3-5, there was a more chaotic end to the reactions with more clear evidence for collapse of the roof rocks, but the general geometry of the reactor zones was preserved and criticality of the zones could be understood using the materials in their present location, but with greatly increased porosity to allow water to be present. In zones 7 to 9, there is a very different situation. Naudet (1991) notes that it is impossible to explain criticality in these zones using only the present uranium configuration. Since there is no evidence for dissolution and removal of uranium after the reactions occurred, Naudet argues for tectonic displacement of the reactor zone materials. This was achieved through sliding of part of the reactor zone down-dip during a severe collapse of the zone at the end of the reaction period. Despite the tectonic upheaval, the chemistry of the zones was maintained essentially intact.

### 3

## ASSESSMENT OF THE POTENTIAL FOR CRITICALITY DUE TO $^{235}\text{U}$ IN A GEOLOGIC REPOSITORY

The canister for spent fuel discussed in the 1993 SKB Annual Report (SKB 1994) was designed to hold 12 BWR fuel assemblies. The outer, 50 mm thick copper portion of the canister was to provide the corrosion resistant barrier and the inner 50 mm thick steel portion of the canister was to provide structural strength. For spent fuel with normal burnup of 35 MWd/kgU or more, criticality cannot be achieved inside the canister even if the void space is filled with water (Oversby, 1994). For fuel with lower burnup, the possibility for criticality to occur inside the canister can be avoided by filling the void space with some material that is low in hydrogen.

Any scenario that addresses criticality must first assume that the canister has been breached in a way that allows free movement of water into the canister. The expected performance of the canister is that neither internal nor external corrosion processes would lead to a breach through the entire canister thickness (Werme, 1990). A scenario that might allow water to enter the canister is if there were a flaw in the weld of the canister that left a hole through the entire thickness of the canister. While it is unlikely that such a flaw would go undetected, it is assumed to occur because it is the only scenario currently thought to be possible that would allow water to enter the canister (Widén and Sellin, 1994). The calculations of Behrenz and Hannerz (1978) concerning the transport of uranium out of a failed canister can be used to place an upper limit on the amount of uranium transport. Behrenz and Hannerz (1978) neglected any restriction to transport caused by the finite size of the pathway through the failed canister. Instead, they used the rate of diffusive transport of uranium through the bentonite buffer to determine the limits to transport. They assumed that the uranium had been oxidized to the more soluble U(VI) and was that it was present as uranyl carbonate. Under the reducing conditions expected in a repository in the deep crystalline rocks of the Fennoscandian Shield, there is unlikely to be enough oxygen for this to happen, even if the results of radiolysis are considered. The solubility of U was assumed to be 1070 g/m<sup>3</sup> in the water at the canister surface. Under these conditions, they found that after 100,000 years, 65 kg U would be present in the deposition borehole due to reduction of U by Fe(II) in the bentonite, 310 kg U would have escaped into the host rock through joints, and 200 kg U would be in the tunnel.

Neretnieks (1979) also calculated a bounding case for transport of U from a spent fuel canister using diffusion in clay as the limiting step. He assumed that the canister top was missing, which is equivalent to assuming there is no transport resistance through a hole in the canister. He found that it would take 35 million years to transport all of the uranium from one canister through the bentonite buffer and into the tunnel. No reduction of uranium in the buffer was considered, and no loss of uranium to the rock fractures was considered.

Both the calculations of Behrenz and Hannerz (1978) and Neretnieks (1979) were for the canister with Pb cast filling. They assumed that lead corrosion products would be soluble and carried away. For the Cu-Fe composite canister (SKB 1994), corrosion of the inner iron canister would produce hydrous oxides of iron, which would be expected to remain inside the canister. These iron hydroxides have a high affinity for sorption of uranium from solution, especially if the iron is Fe(III), the oxidation state expected if uranium is in the soluble U(VI) valence state. Thus, transport of uranium from the canister, even if the canister has a large penetration allowing free access of water to the fuel, would be expected to be far less than that calculated previously.

The calculations of Behrenz and Hannerz (1978) assumed 1.66%  $^{235}\text{U}$  after decay of  $^{239}\text{Pu}$ . This is approximately the amount expected from BWR fuel with a burnup of 20 MWd/kgU (Guenther et al., 1991). The amount of  $^{235}\text{U}$  in natural uranium 1000 million years ago was 1.62%. Let us use the data in Figure 8 for criticality as a function of age to estimate the amount of uranium needed to achieve an Oklo-type criticality in a repository. The first parameter to estimate is the amount of neutron absorbers present in various parts of the repository. The spent fuel originally contains about 0.6% rare earth elements. These elements have low solubility, so would be expected to remain inside of the canister even if the uranium dissolved away. The rare earth elements with high capture cross sections are Sm, which would be 500 ppm in the assumed spent fuel, and Gd, which would be 39 ppm. This would translate into an equivalent  $^{10}\text{B}$  (x) of over 40 ppm. Based on the data in figure 8, which shows that the minimum thickness of "ore" for  $^{235}\text{U}=1.62\%$  would be 120 cm, while the canister inner diameter is only 95 cm, we can conclude that the amount of neutron absorbers in the fuel should be sufficient to prevent criticality inside the canister.

As discussed above, transport of uranium out of the canister is likely to be hindered because of sorption of uranium onto iron corrosion products. Nevertheless, it is instructive to calculate the maximum amount of uranium that might be assembled outside the canister under very long times, such as the 35 million year period calculated by Neretnieks (1979). The Cu-Fe canister was designed to contain 12 BWR fuel assemblies, which corresponds to about 2 t of fuel. We will examine the case of migration of uranium from several canisters into the tunnel to see whether there is any possibility, however unlikely, that a critical configuration could be

achieved. The tunnel fill is assumed initially to be a mixture of 85% sand and 15% bentonite. To allow the possibility for reaching the conditions for criticality, enough sand must dissolve and be replaced by water so that the porosity is 54.5% ( $m=1.2$ ). In addition, more sand must dissolve to make room for the inward-migrating uranium. For an ore grade of 60% by weight in the solid phase and solids taking up 45.5% of the volume, the uranium-rich region would have the composition given in Table 9. This assumes that no clay dissolves, so that all increase in porosity and in space for uranium comes from quartz dissolution.

**Table 9. Composition of the assumed uranium-rich region in a disposal tunnel after dissolution of quartz and replacement by uranium and water to achieve the optimal critical configuration. Density =  $2.53 \times 10^3$  kg/m<sup>3</sup>.**

Material	Weight in g per cm <sup>3</sup>
Uranium	1.19
Sand	0.45
Clay	0.34
Water	0.545

Using the results from Table 9 and the minimum thickness of 120 cm for the rich zone from Figure 8, together with the normal lateral dimension for Oklo reactors of 10 m by 10 m, we find that a volume of 120 m<sup>3</sup> would provide a critical assembly. This volume would contain 143 t of uranium, or more than the total uranium from 71 canisters. Naudet (1991) stated that the lateral dimensions might be reduced somewhat, but the reduction is limited so that too many neutrons are not lost. For a reduction to 5 m by 5 m by 1.2 m, the volume would be 30m<sup>3</sup>, with a total amount of uranium of about 36 t, representing the total content of 18 canisters. Even for a large reduction in lateral dimensions, which probably would adversely affect the neutron balance, with 2 m by 2 m by 1.2 m we need 4.8 m<sup>3</sup> volume containing 5.7 t uranium, which is nearly 3 canisters worth. The imagined concentration process, which already contains several assumptions for processes to occur that are unlikely in reality to be possible, now requires that fluid flow in the tunnel be directed toward a central location rather than be in a single direction. Taken together, we can see that assembly of a critical mass of <sup>235</sup>U in the repository for reasonable fuel burnup requires a sequence of highly improbable events, some of which stretch the bounds of credibility. The likelihood that a sequence of such events might occur is so small as to be negligible.

## 4 CONCLUSIONS

The uranium deposits at Oklo contain more than a dozen regions of very rich ore that were the site of spontaneous fission reactions 1950 million years ago. At the time of the reactions, the  $^{235}\text{U}$  isotope was about 3.5% of the total uranium, a value that is similar to enriched uranium used in present day commercial light-water nuclear power reactors.

The ores at Oklo were originally present in sandstones. Elevated temperatures in the vicinity of  $300^{\circ}\text{C}$  caused by the start of nuclear reactions and fracturing in response to tectonic tensile forces initiated hydrothermal circulations that dissolved silica and enriched the ore. The reactor cores became typically 40% uranium dioxide embedded in clay, with or without residual silica. The capture of neutrons by  $^{238}\text{U}$  led to a build up of  $^{239}\text{Pu}$  in the ore during reactions. For typical conditions in the reactor core, the Pu/U would have been 0.001 (Bros et al., 1993), giving a total  $^{239}\text{Pu}$  content of 0.4% when the reactors were operating. This is comparable to the level of  $^{239}\text{Pu}$  in commercial light water reactor fuels destined for geologic disposal (see Table 2 above).

The reactors operated over periods of 200 to 800 thousand years. For reactor zone 9, the total energy released corresponded to a burn-up of about 5 MWd/kg U (Loss et al., 1988). Thus, the remaining level of  $^{235}\text{U}$  at the end of the reactions was still very high - perhaps 3% even in the rich zones - and the ore contained an amount of  $^{239}\text{Pu}$  equivalent to that found in commercial reactor fuels with much lower  $^{235}\text{U}$  values after normal burn-ups of 30 to 35 MWd/kg U.

The Oklo ores were very low in elements with high neutron capture cross sections such as vanadium, manganese, rare earth elements, and boron. The low abundances of these elements in the ores made the reaching of a sustained nuclear fission reaction possible. Higher values of neutron capturing elements or lower levels of  $^{235}\text{U}$  would have made criticality impossible to achieve (Naudet, 1991).

Once the nuclear reactions started at Oklo, the reactors made their own environment through desilicification. The reactions could only be sustained because there was enough initial porosity to allow adequate water to enter the environment and start the desilicification process that increased the porosity even further. When porosity was lost through compaction of the overlying strata, the reactions were prematurely halted (Naudet, 1991). The high initial porosity

(> 10%) was produced as the result of uplift and tectonic extension forces.

Examination of the reactor zones at Oklo shows no evidence for significant amounts of plutonium to have become separated from the uranium ore and concentrated in another area. Only a single case of minor plutonium movement over a distance of a few meters has been found. The reactor zones examined include those where the border regions were undergoing active desilicification with fluid circulation while the reactions were going on and plutonium was present.

Comparison of the Oklo conditions to those expected for geologic disposal in granites of the Fennoscandian shield raises the following considerations.

1. The Fennoscandian shield area is an old and rather stable geologic structure, unlikely to be effected by tectonic forces sufficient to generate 10% open porosity and initiate major hydrothermal circulations.
2. The levels of neutron capturing elements in the repository situation are considerable. The fuel contains  $^{238}\text{U}$  and fission products that will capture neutrons. In addition, the copper from which the corrosion resistant outer canister will be made in the Swedish design has a thermal neutron capture cross section of about 4 barns (Friedlander et al., 1964). This contrasts with the "average rock" value used by Bowman and Venneri (undated) of 0.255 barns.
3. The redox state in the Oklo reactor for which minor plutonium movement has been documented was and is dominated by a high abundance of organic carbon. This controlled the redox state for the uranium and plutonium to a level low enough for plutonium to be dissolved as the more soluble Pu(III) species. The redox conditions in the granite forseen for waste disposal of spent fuel in Sweden will be reducing, but much less so than for Oklo. Under the conditions forseen in Sweden, plutonium would be stable in the Pu(IV) state, which has much lower solubility than either U(IV) or U(VI). Thus, migration of plutonium away from uranium and out of the copper canister seems to be impossible.
4. The levels of plutonium in the spent fuel destined for disposal are comparable to those that were present at Oklo during reactions and lower than those that were needed to produce the supposed autocriticality event by Bowman and Venneri (undated). The uranium-235 present in spent fuel of reasonable burn-up, even after addition of that produced by plutonium decay, does not seem to be sufficient to produce criticality of any kind in the disposal configuration or in any chemically feasible remobilized configurations.

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## **APPENDIX 1:**

### **SUMMARY OF KBS TEKNISK RAPPORT 108 "CRITICALITY IN A SPENT FUEL REPOSITORY IN WET CRYSTALLINE ROCK" BY P. BEHRENZ AND K. HANNERZ AND DISCUSSION OF THE IMPACT OF NEW CANISTER DESIGNS ON THE REPORT'S CONCLUSIONS**

#### **A.1 CANISTER DESIGN DESCRIPTION**

The KBS reference canister used by Behrenz and Hannerz (1978) was assumed to contain either 498 BWR fuel rods or 744 PWR rods. The canister was made of high conductivity copper with a wall thickness of 20 cm. After placing the fuel in the canister, molten lead was to be cast around the fuel to fill the spaces between fuel pins and reduce the risk for dissolution of the fuel or for criticality to occur. Three copper lids were successively added above the fuel, with the final closure by electron beam welding.

The current SKB canister concepts are described in the 1993 SKB annual report (SKB 1994) and in Werme and Eriksson (1995). The first of these concepts uses a double walled canister with the outer wall being a 5 cm thick copper vessel for corrosion resistance and the inner wall being a 5 cm thick steel cylinder to give mechanical strength. Criticality control would be achieved by adding filling material in the form of small particles of metal, glass, or ceramic material to fill the space between the fuel pins. The new canister concept replaces the steel cylinder with a cast solid with rectangular channels to hold the fuel assemblies. The cast solid could be made from iron, steel, or a non-ferrous metal. Since the amount of space available for water in the canister is then very limited, the design would eliminate the possibility of criticality for everything except fuel with rather high enrichment (4.2%  $^{235}\text{U}$  in 4 17x17 PWR assemblies or 3.6%  $^{235}\text{U}$  in 12 SVEA-64 BWR assemblies) that has had less than one normal irradiation cycle (Werme and Eriksson, 1995).

#### **A.2 REPOSITORY DESIGN DESCRIPTION**

The KBS-3 repository concept assumed a disposal site at a depth of about 500 m in crystalline rock of the Fennoscandian Shield region. The rock was assumed to have low permeability, with joints of about 0.1 mm width spaced at one per meter or less. For this size and spacing the joint system could not accommodate accu-

mulation of enough water with dissolved fissile material to constitute a critical volume. Thus, criticality would only be possible in a mined region of the repository. The flow of water in the rock would be low, amounting to less than 1 liter per square meter of rock cross section per year. The repository would be located in an area that is tectonically and seismically stable. Canisters of spent fuel would be placed in holes in the bottom of horizontal tunnels.

The canister in the deposition borehole would be surrounded by a bentonite buffer material, with a highly compacted dry density of 2 tonnes per cubic meter. The tunnel above the deposition boreholes would be backfilled with a mixture of quartz and bentonite. When water enters the deposition borehole, the dry bentonite absorbs the water and swells, filling the void space and eventually being confined by the rock wall at a swelling pressure of 5 to 10 MPa. Water in the tunnel region is also absorbed by the bentonite and fills the space between the quartz grains with a high water content gel. The bentonite limits mass transfer of substances capable of causing copper corrosion so that the canister under normal conditions will outlive the  $^{239}\text{Pu}$ .

The current repository concept for SKB is essentially the same as that used by Behrenz and Hannerz (1978) in their assessment of criticality.

### A.3 CRITICALITY DUE TO PLUTONIUM

The expected lifetime of the canister far exceeds the half-life of  $^{239}\text{Pu}$ , so the only mechanism for criticality due to plutonium is a premature failure of the canister due to a fabrication defect. This was, and is, considered to be a very improbable event. Even if the copper canister corrodes, the corrosion products such as  $\text{Cu}_2\text{S}$  or  $\text{Cu}_2\text{O}$  have low solubility, so will remain in the deposition borehole. Thus, Behrenz and Hannerz (1978) considered the two cases for plutonium criticality to be either inside or outside a canister with a hole in it.

For criticality to occur inside the canister, the canister must remain mechanically intact. If the canister were to collapse, the bentonite would swell to fill the void space and there would be no room for accumulation of enough water to moderate the neutrons. Since the canister was initially filled with lead, in order to allow water to enter the canister either the lead or the fuel matrix must dissolve and be transported out of the canister. The most likely corrosion product of lead would be  $\text{PbSO}_4$ , which would be expected to precipitate and block the hole in the canister, thus stopping corrosion of the Pb. It should be noted that for transport processes available in the repository and for the low solubility of lead salts, removal of a substantial amount of the lead in a relatively short period of time cannot occur in reality. One might, however, consider the absence of Pb in the canister due to failure to include the lead fill during packaging of the fuel. While this, too, is an extremely unlikely event, it is physically possible.

The current SKB canister designs contain either filling materials of low solubility or cast inserts made from low solubility materials. There are no physically reasonable methods to dissolve and transport more than a small fraction of these materials out of the canister. If the filling materials or cast inserts remain in the canister, there will not be enough room to accumulate sufficient water to moderate neutron energies and allow a critical condition to occur except for unirradiated fuel with higher than average initial enrichment of  $^{235}\text{U}$ . This fuel would not contain  $^{239}\text{Pu}$  because it had not been irradiated.

Behrenz and Hannerz (1978) calculated a scenario for criticality due to plutonium inside the canister by assuming the following conditions. First, they assumed that there was a large leak in the canister at one end. Then they assumed that nearby leaks occurred through the lead filling and through the ends of the Zircaloy tubes containing the fuel. To preserve a favorable geometry, they assumed that further corrosion of the lead and Zircaloy did not occur. It is then assumed that radiolysis products oxidize the uranium matrix to the more soluble U(VI) and that the water in the canister contains carbonate ions dissolved from the buffer material. The uranium is then assumed to be instantaneously removed from the canister as the soluble uranyl carbonate complex, even though it is noted that diffusion rate limitations would make this impossible. The configuration for which the calculations were done is then a homogeneous slurry of plutonium in water inside the residual cladding tube configuration. In reality, if plutonium were to be made into a slurry with water, most of the plutonium would settle out rather quickly in colloidal form, further decreasing the possibility for criticality. The calculations showed (Appendix 2 in Behrenz and Hannerz, 1978) that there was never sufficient plutonium in the canister to achieve criticality even under this highly unrealistic set of assumptions. [Note: In the main text, B&H attribute the result to a time dependent dissolution of U, which is not the case stated in the appendix where the data are given. The conclusion drawn in Appendix 2 does not require any limit to the rate of uranium transport, so is stronger than that given in the main text.]

The current SKB canister concept contains more fuel assemblies than that used by Behrenz and Hannerz (1978). The key parameter in comparing the two systems is the amount of plutonium in each canister. For the 1978 calculations, the amount of  $^{239}\text{Pu}$  initially in the canister was 9.37 kg for the PWR case and less for the BWR case. For the current SKB designs, the amount of  $^{239}\text{Pu}$  would be 9.8 kg for BWR fuel with 35 MWd/kgHM burnup (see Table 1 in this document). Using the results from Behrenz and Hannerz (1978) in their Appendix 2, with adjustment for the small increase in plutonium, we can conclude that criticality inside a canister of BWR fuel is not a possible scenario. This conclusion remains valid until the amount of  $^{239}\text{Pu}$  in the canister exceeds 24 kg, so criticality for the PWR canister with somewhat more plutonium is also not a credible scenario.

For criticality outside the canister, plutonium must dissolve, be transported from the canister, and be separated from uranium and other neutron absorbers. One method for this to occur would be for plutonium to preferentially dissolve from the fuel and be transported out of the canister. Given the low solubility of pluto-

niium under repository conditions and the low general mass transfer rates in the repository, this scenario was dismissed as impossible by Behrenz and Hannerz (1978), a conclusion that is still considered to be valid.

The case for criticality due to plutonium outside the canister considered by Behrenz and Hannerz (1978) was to assume highly oxidizing conditions so that both uranium and plutonium would be oxidized to the more soluble (VI) valence state and diffuse in that state out of the canister. For criticality to occur, the uranium must be separated from the plutonium. This was assumed to occur by the plutonium becoming reduced on contact with the bentonite, while the uranium was neither reduced nor sorbed. They then assumed a spherical geometry for PuO<sub>2</sub> in bentonite that had 29.6% water before the plutonium precipitation. If all of the plutonium from a single canister were able to accumulate in the assumed spherical geometry, it would be theoretically possible to have criticality outside the canister up until 50,000 years after disposal. After that time, there is not enough fissile plutonium remaining to assemble a critical mass from the contents of a single canister under any conditions.

The theoretical possibility for criticality, however, was shown by Behrenz and Hannerz (1978) not to be possible when the limitations imposed by diffusive transport from the canister were considered. The amount of plutonium that could diffuse out of the canister, assuming the same diffusion rate as for uranium, was only a fraction of the amount needed for a critical mass. The closest approach to assembly of a critical mass for the assumed conditions was at 30,000 years after disposal, when about 40% of a critical mass could have been released under the assumed highly oxidizing conditions. Behrenz and Hannerz (1978) noted that a smaller amount of plutonium would be needed to form a critical mass if some of the bentonite in the borehole were missing. Even though there is no feasible mechanism for this to occur once the bentonite has been emplaced, they calculated how much bentonite must be lost in order to allow criticality to occur. If approximately 50% of the bentonite is missing, then the critical mass would be reduced to the same amount as that assumed to be outside the canister under the transport assumptions given above. Since this scenario requires two events with vanishingly small probabilities to occur together, it is reasonable to conclude that criticality due to plutonium outside the canister can be dismissed.

The current SKB designs contain more fuel than those used by Behrenz and Hannerz (1978). The assumptions used in their calculations, however, depended on the amount of water available and the solubility of uranium, together with the diffusion coefficient for uranyl ion. Thus, the conclusions remain independent of the amount of fuel per canister.

Behrenz and Hannerz (1978) performed some calculations to investigate what the effects on neighboring canisters would be if a single canister with a defect somehow managed to reach a critical condition. Since the maximum temperature that could be reached by the critical configuration is limited to 265° C by the boiling point of water under the repository hydrostatic pressure of about 5 MPa, the maximum increase in temperature of an adjacent canister would be limited to 45°

C. This is not sufficient to change the corrosion lifetime of the canister by enough to cause serious concern. In other words, criticality in one canister would not trigger events that might cause criticality to occur in another canister.

## A.4 CRITICALITY DUE TO URANIUM

Behrenz and Hannerz (1978) do not explicitly consider the case of criticality due to  $^{235}\text{U}$  inside the canister. This is presumably because of the Pb casting around the fuel pins, which is presumed to remain essentially in place even in the long time frames because of the low corrosion rate of Pb and low solubility of its corrosion products. The presence of the Pb casting prevents sufficient water from entering the canister and thus allowing moderation of the neutrons needed to support a chain reaction with  $^{235}\text{U}$ . For the present SKB designs with either particulate filling materials or a cast insert, the same conclusions would apply.

The uranium content used by Behrenz and Hannerz (1978) was 1.66%  $^{235}\text{U}$ , which was stated to correspond to the case of PWR fuel irradiated to a burnup of 33 MWd/kgU after decay of all  $^{243}\text{Am}$  and  $^{239}\text{Pu}$ . This would imply an initial enrichment of about 3.5%  $^{235}\text{U}$ . They considered two possible sites for accumulation of sufficient uranium to allow criticality: (1) the tunnels, and (2) the deposition holes. Accumulation of a critical mass of uranium in the rock joint system was ruled out on the grounds of restricted geometry. All calculations assumed that uranium diffused out of the canisters and accumulated in a favorable geometry in the prescribed location. Since the scenarios for criticality due to  $^{235}\text{U}$  could only occur in the long term, alpha radiolysis to produce oxidizing conditions is no longer a possible mechanism. Oxidizing conditions to dissolve and transport the uranium must be assumed to come from the ground water system flowing through the repository.

For the tunnel calculations, the uranium was assumed to accumulate as a spherical mass homogeneously distributed in the tunnel mixture of sand, bentonite and water. For this case, the critical mass would be 4.4 t and the resulting sphere would be 3.5 m<sup>3</sup> in volume and radius of 0.9 m. If power were generated due to criticality, a larger mass would be needed. Likewise, if the geometry were not a sphere, the critical mass would be larger. With the 1978 canister design, fuel from 4 canisters would be needed. For present SKB designs, fuel from at least 3 canisters would be needed. The diffusing uranium would need to be attracted to a central location and caused to precipitate; i.e., there must be a radially inward transport of uranium or some cause for precipitation in the tunnel in the downstream direction of water transport. It is very unlikely that this would be possible since the conditions for dissolving the uranium and transporting it out of the canister are opposite to those required for precipitation, yet the water entering the canister will have been conditioned by geochemical interactions with the tunnel material before it can reach the canisters.

The remaining case of potential criticality due to  $^{235}\text{U}$  is for accumulation of a critical mass within a single borehole. With the originally emplaced bentonite in the borehole, more absorption of neutrons occurs, so a larger amount of uranium is needed to reach a critical mass than for the tunnel case. Behrenz and Hannerz (1978) concluded that "It goes without saying that the transport of uranium away from several deposition holes and into one other such hole is so strange a scenario as to hardly merit consideration." To achieve criticality in the borehole, one must assume that a large fraction of the bentonite has escaped from the borehole. Behrenz and Hannerz (1978) considered the following scenario that might allow a critical mass to be assembled.

1. A significant tectonic event occurs that opens large cracks around the deposition borehole.
2. Bentonite squeezes into the cracks until at least 45% of the bentonite has escaped.
3. Corrosion of the canister causes penetration.
4. Uranium becomes oxidized and diffuses out of the canister.
5. Ferrous iron from the tunnel diffuses down into the borehole.
6. The iron in solution reacts with the uranium to produce low solubility uranium (IV), which precipitates into a spherical mass above the canister.
7. The uranium density in the sphere must be 2 to 3 gU/cm<sup>3</sup>.

For a suitably selected repository site, occurrence of a major tectonic event would have very low probability. Nevertheless, if it occurred, cracks would form and the bentonite could be partially lost from the boreholes. As a consequence of the tectonic event, ground water conditions could become oxidizing, allowing accelerated corrosion of the canister and oxidation of the uranium. The step in the proposed scenario that would not be possible, however, is the downward transport of ferrous iron. If the ground water is oxidizing, the iron will be oxidized to ferric iron and be precipitated in situ within the tunnels. In other words, the ground water cannot be assumed to be both oxidizing and reducing at the same time. It should also be noted that the accumulation of a spherical mass of uranium with density as high as assumed would constitute a small ore body with uranium content greater than 60% by weight. It is exceedingly unlikely that this would be possible without invoking unrealistic geochemical conditions such as the intrusion of large amounts of organic matter into the borehole after the bentonite was expelled.

With current SKB canister designs, for those with iron present there is a further restriction on the transport of uranium. Since there is a large mass of iron present and water will contact the iron before the uranium, any oxidizing waters will generate ferrous and/or ferric iron. This will insure that uranium either is limited to

the U (IV) state or is coprecipitated onto ferric hydroxide if it is present as U (VI). Only for the cases of stainless steel or non-ferrous canister inserts is it possible to consider the possibility of oxidation and transport of significant amounts of U (VI).

Despite the unrealistic nature of the scenario that they considered, which Behrenz and Hannerz (1978) themselves emphasized, they considered the consequences of criticality under the most extreme situation imaginable. They noted at the outset that collecting a critical mass would be a slow process, producing a "reactor" with a strongly negative temperature and power coefficient. As such, there would be no possibility for sudden energy release, i.e., explosion. The scenario imagined was to move all of the uranium out of the deposition holes and into the tunnel with the configuration that would give the maximum amount of reaction. This turned out to be a cylinder with the radius of the tunnel and a length of 27 m, with a density of U of 0.78 t/m<sup>3</sup>. This configuration could generate a maximum power of 130 kW from 227 tonnes U. This configuration would release energy very slowly, over about 100,000 years, which is similar to the operating interval for the reactor zones at Oklo. The release of radionuclides to the ground water was considered to be equivalent to those that might be released from one canister failure per 1000 years under normal conditions. Thus, any performance assessment scenario that assumed a canister failure rate of 1 per 1000 years would provide an estimate of the consequences of the imagined criticality scenario, which is an upper limit to the potential consequences of criticality. It must be remembered that the geometry for the criticality calculation was *assumed* and that there are no credible scenarios for achieving that geometry.

## A.5 CONCLUSIONS

The analysis of the potential for and consequences of reaching a critical configuration in the repository carried out by Behrenz and Hannerz (1978) showed that

- (1) Achieving a critical configuration required assumption of conditions that were either of extremely low probability or were impossible.
- (2) Consequences of criticality, if achieved, would be small compared to the overall impacts of repository disposal.

Some of the transport scenarios considered by Behrenz and Hannerz (1978) required the use of internally inconsistent assumptions concerning transport of redox active species, so many of their low probability scenarios are actually impossible scenarios.

Modifications to the spent fuel disposal canister design since 1978 do not produce conditions that significantly increase the possibility to achieve a critical mass in the repository. Some designs (e.g., those containing metallic iron) would reduce

the possibility to oxidize and transport uranium and as such would reduce the already very low probability of accumulating a critical mass at any time.

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