

Comparison of a post-closure transient criticality model with the Oklo natural reactors

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ABSTRACT

In support of the disposal system safety case for a geological disposal facility (GDF) there is a requirement to consider ‘what-if’ hypothetical scenarios for post-closure nuclear criticality. Although all such scenarios are considered very unlikely, one ‘what-if’ scenario is the mobilization of fissile material from a number of waste packages and its slow accumulation within the GDF or the immediate surroundings. Should sufficient fissile material accumulate a quasi-steady-state (QSS) transient criticality event could result. A computer model has been developed to understand the evolution and consequences of such an event.

Since a postulated QSS criticality could persist for many millennia, building confidence in the modelling approach is difficult. However, the Oklo natural reactors in Africa operated for similar durations around two billion years ago, providing a natural analogue for comparison. This paper describes the modelling approach, its application to hypothetical criticality events for a GDF, and how the model can be compared to Oklo. The model results are found to be in agreement with the observational evidence from Oklo, building confidence in the use of the QSS model to simulate postulated post-closure criticality events in GDFs.

KEYWORDS: disposal, post-closure, criticality.

Introduction

THE Nuclear Decommissioning Authority (NDA) have been charged with implementing the UK Government’s policy for the long-term management of higher activity radioactive waste by planning, building and operating a geological disposal facility (GDF). The Radioactive Waste Management Directorate (RWMD) is in the early stages of planning for implementation (Nuclear Decommissioning Authority, 2010a). At present a site for a GDF has not been identified, and RWMD has produced an initial ‘generic’ disposal system safety case (DSSC) (Nuclear Decommissioning Authority, 2010b), to put forward the safety arguments for geological

disposal using a range of illustrative disposal concepts and host geologies.

Given that a GDF will include the disposal of fissile nuclides, and such nuclides could, under certain conditions, lead to an unplanned nuclear chain reaction (‘criticality’), the demonstration of criticality safety forms an important part of the DSSC. In particular, the guidance given in the Environment Agency and Northern Ireland Environment Agency (2009) *Guidance on Requirements for Authorisation (GRA)* for a GDF requires a demonstration that “the possibility of a local accumulation of fissile material such as to produce a neutron chain reaction is not a significant concern.” In addition, the guidance states that the “environmental safety case should also investigate, as a ‘what-if’ scenario, the impact of a postulated criticality event on the performance of the disposal system.” The environmental safety case is a key document in the DSSC.

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Waste packages can be designed to ensure that criticality is not possible during the transport and operational phases of a GDF, and for a significant period post-closure (Nuclear Decommissioning Authority, 2010c). Over longer times, however, packages may degrade, and groundwater flows could lead to a localized accumulation of fissile material, so that, although it is considered very unlikely, criticality cannot be ruled out completely.

The Nuclear Decommissioning Authority (2010c) document provides an overview of how the localized accumulation of fissile material could, hypothetically, lead to a critical system within a GDF, and how the critical system could develop as either a quasi-steady-state (QSS) or rapid transient (RT) event under certain conditions. Mason *et al.* (2012) also present an overview of, and example results from, the mathematical models developed to understand the consequences of hypothetical post-closure criticality. Further details of the models are available in Smith *et al.* (2007a,b).

This paper focuses on the QSS model, developed to understand postulated QSS transient criticality events under GDF conditions (Smith *et al.*, 2007a). In particular, research undertaken to build confidence in the modelling approach is described, through a comparison exercise of the mathematical model with the available data for the Oklo natural reactors which occurred in Africa approximately two billion years ago.

The QSS model: application for GDF conditions

The Nuclear Decommissioning Authority (2010c) document describes how, in a GDF containing fissile nuclides, such as uranium-235 (^{235}U) and plutonium-239 (^{239}Pu), a self-sustaining chain reaction of fission could, hypothetically, be established. In a fissile system, the ratio of the rate of neutron production (by fission) to the rate of neutron losses in the system is termed $k_{\text{effective}}$. At the point of criticality $k_{\text{effective}}$ is equal to unity. For super-critical systems $k_{\text{effective}}$ is greater than 1, and it is less than 1 in sub-critical systems. The emplacement of any waste materials within a GDF will be significantly sub-critical, so that the initial distribution of fissile materials will need to change substantially for a critical configuration to arise. This gives confidence that the criticality event is low probability.

Should fissile material accumulate into a critical configuration, and continue to accumulate, a QSS

transient criticality could arise (Smith *et al.*, 2007a; Nuclear Decommissioning Authority, 2010c; Mason *et al.*, 2012). In such a system, the accumulation of fissile material would act to increase the value of $k_{\text{effective}}$ for the system. However, the heat produced by fission, and the associated temperature rise, would act to reduce the value of $k_{\text{effective}}$. In addition, the nuclear burn-up of fissile materials (the transmutation of fissile material through the absorption of neutrons) typically acts to reduce the value of $k_{\text{effective}}$, although in some cases the capture of free neutrons might act to increase $k_{\text{effective}}$ by changing a non-fissile nuclide to a fissile one. For example, neutron capture in the non-fissile uranium-238 (^{238}U) isotope will produce the fissile isotope ^{239}Pu . In a quasi-steady-state the physical processes acting to increase $k_{\text{effective}}$ balance those acting to reduce it, so that, even though the temperature and composition of the volume undergoing fission will evolve, the system will remain at the just critical point where $k_{\text{effective}} = 1$.

Figure 1 shows a schematic diagram of the key physical processes in a QSS transient criticality under GDF conditions, as used to construct a mathematical model. Firstly a mechanism such as groundwater flow, must exist to transport fissile materials. Within this flow the chemical conditions must undergo some change, so that fissile nuclides leached from part of a GDF can be deposited in a region which could become critical, given sufficient fissile mass in the right configuration (Cummings *et al.*, 2007; Nuclear Decommissioning Authority 2010c; Mason *et al.*, 2012). Assuming a critical system is established, and that the accumulation process continues, then a QSS transient criticality can result. This leads to sustained fission and changes to the nuclide composition in the critical region, heat production and thus heat transfer to a region surrounding the critical volume.

The lower part of Fig. 1 illustrates the key nuclide transformations, and is termed the burn-up model. For example, in the presence of free neutrons some of the ^{235}U will fission, ultimately leading to the production of inert fission products (inert meaning that although the fission products occupy volume, they do not have any other contribution to the value of $k_{\text{effective}}$), and some of the ^{235}U will capture a free neutron to produce uranium-236 (^{236}U). It is noted that, in reality, a range of fission products would be produced. However, most of these are short-lived compared to the timescales of interest and sensitivity

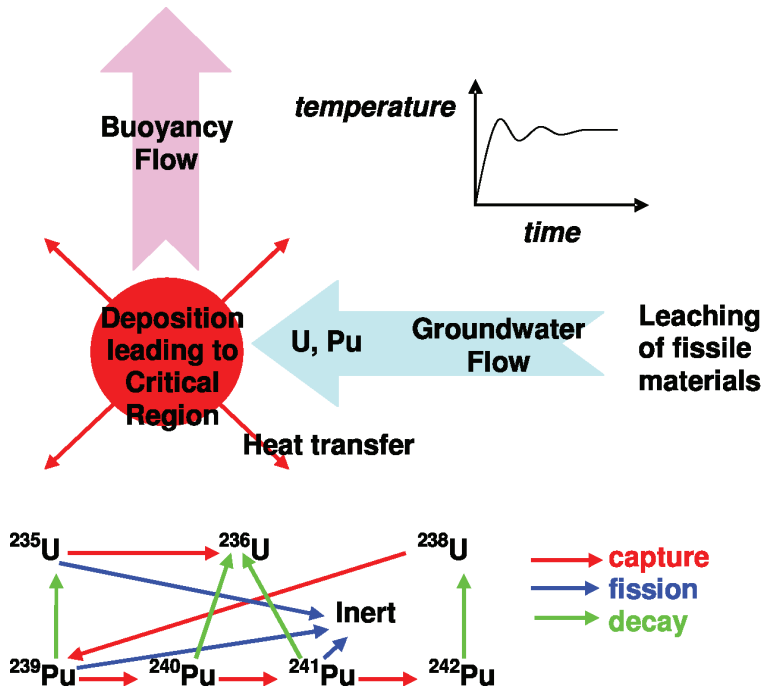


FIG. 1. Conceptual model of a QSS transient criticality event under GDF conditions. Note that in the burn-up model (lower part of the figure) the green ‘decay’ arrows are a simplification of a multi-step process, where short lived intermediate nuclides have been neglected. The blue ‘fission’ arrows are also a simplification of the fission process, where it is assumed that fission leads to an ‘inert’ set of final fission products.

calculations have shown that inclusion of representative fission products in the burn-up model do not have any significant effects (Mason *et al.*, 2007). Over long timescales the natural decay of fissile nuclides is also important. For example, with a half-life of approximately 24,000 years, ^{239}Pu will decay to ^{235}U .

The *QSS* model solves a system of equations, carefully constructed to capture the key physical processes. For a system with a given number of nuclides, N , in the burn-up model (e.g. $N = 8$ in Fig. 1, where there are three uranium isotopes, four plutonium isotopes and the inert nuclide), a mathematical system has $N + 1$ values that can evolve; the amount (or concentration) of each nuclide present in the critical region, and the temperature. Mathematically these are formulated as follows (Smith *et al.*, 2007a): (1) one ordinary differential equation which states that the rate of change of $k_{\text{effective}}$ is zero (i.e. if $k_{\text{effective}}$ is initially unity it will stay at unity); and (2) N ordinary differential equations (one for each nuclide) which show how the rate of change of each

nuclide is determined by either accumulating material or gains/losses from nuclear fission, capture or decay.

Within these equations, the neutron flux (a measure of how much fission is occurring) is eliminated through an energy conservation equation which states that the energy released from fission must equal that used in heating the critical region and that conducted away to the surroundings. The key inputs to the model to enable solution are: the nuclear burn-up model; an interpolation function for $k_{\text{effective}}$ as a function of temperature and composition; the fissile material accumulation rate; and heat transfer parameters.

Using this approach the *QSS* model has been successfully applied to a wide range of hypothetical accumulation scenarios under GDF conditions (Mason *et al.*, 2009a,b), providing insight into the predicted consequences of this ‘what-if’ scenario. In particular, the calculations have demonstrated a ‘rule of thumb’ to scale the results for different accumulation rates which is

consistent with the understanding of the physical processes involved (e.g. Nuclear Decommissioning Authority, 2010c), and have supported analysis of the wider consequence assessment of a transient criticality (Cummings *et al.*, 2008).

A detailed review of the QSS model by Mason *et al.* (2009c) has shown it to be a powerful tool in understanding transient criticality, although the difficulty in validating the modelling approach was noted. Given the durations involved (many millennia) and the difficulty of undertaking analogue experiments, this is not unexpected. Although not offering formal validation, information from the Oklo natural reactors provides an excellent opportunity for comparison with the QSS model. These natural reactors are believed to have operated under similar conditions to postulated QSS criticality transients, about two billion years ago. A successful comparison of the QSS modelling approach with the data from these reactors is therefore an important step in building confidence in the modelling approach taken for 'what-if' GDF criticality scenarios.

The Oklo natural reactors: an overview

The Oklo natural reactors were discovered in Gabon, Africa during the mining of uranium in the twentieth century. The key evidence for the reactors was that uranium mined from elsewhere on earth has a consistent enrichment (the weight percentage of ^{235}U in uranium) of 0.711%. In some of the Oklo mines the percentage was less than 0.3%, and this can only be explained by the uranium having undergone a nuclear reaction at some time in the past (e.g. Naudet, 1991).

At the time of the Oklo reactors all uranium on earth would have had an enrichment of 3.7%, which is much higher than that today. The reason for this is that ^{235}U decays with a half-life of approximately 700 million years. At this enrichment level it is possible to achieve a nuclear reaction in the presence of a suitable amount of water, and a suitable geometry. This is believed to have occurred within several reaction zones at Oklo (Naudet, 1991).

An understanding of the functioning of the 'fossil reactors' was developed by a French team from the Commissariat à l'Énergie Atomique (CEA) (Naudet, 1991). At the time ten reactor zones had been identified, although a few more were identified later. Reviews of the Oklo phenomena have been prepared by Oversby

(1996) and Zetterström (2000) for the Swedish geological disposal programme, SKB, and by Smellie (2006) for Nirex¹.

Naudet (1991) focussed his attention initially on a reaction volume termed zone 2 which was subjected to detailed measurements. Figure 2 presents an overview of the natural processes and phenomena that are believed to have led to the operation of the natural reactors, including that for zone 2. During the first stage, a number of physical and geological processes are believed to have caused the oxidation of uranium, its mobilization, and precipitation, leading to a much higher than usual concentration of uranium within certain volumes (of order 10% by mass compared to about 1% in the rest of the deposit). During the second stage, the concentration of uranium is believed to have caused an initial criticality (i.e. $k_{\text{effective}} = 1$), resulting in increased temperature, desilification of the host sandstone, and an increased porosity.

During the third stage the system entered a quasi-steady-state where, with a stable rock porosity, the nuclear reaction was sustained through the burn-out of neutron poisons (nuclides which can easily capture free neutrons), until no longer able to sustain the nuclear reaction. In the fourth stage, subsequent geological activity transformed the reaction zones into the form discovered when they were mined.

Although they are not identical to the GDF application of the QSS model, the Oklo natural reactors share several similarities during the third stage (as identified in Fig. 2). In particular the critical region undergoes changes in nuclide composition, and an increase in temperature, and remains just critical at $k_{\text{effective}} = 1$. The difference is that under GDF conditions the mechanism sustaining the criticality transient is the continued accumulation of fissile material, whereas it is the burn-out of neutron poisons for Oklo. Mathematically, however, the governing equations are very similar, making the Oklo reactors an ideal natural analogue for comparison with the QSS model.

The following sections summarize the comparison undertaken, the conclusions drawn, and how they help to build confidence in the modelling

¹ The RWMD undertakes work formerly undertaken by United Kingdom Nirex Limited; Nirex was subsumed into the NDA in 2007.

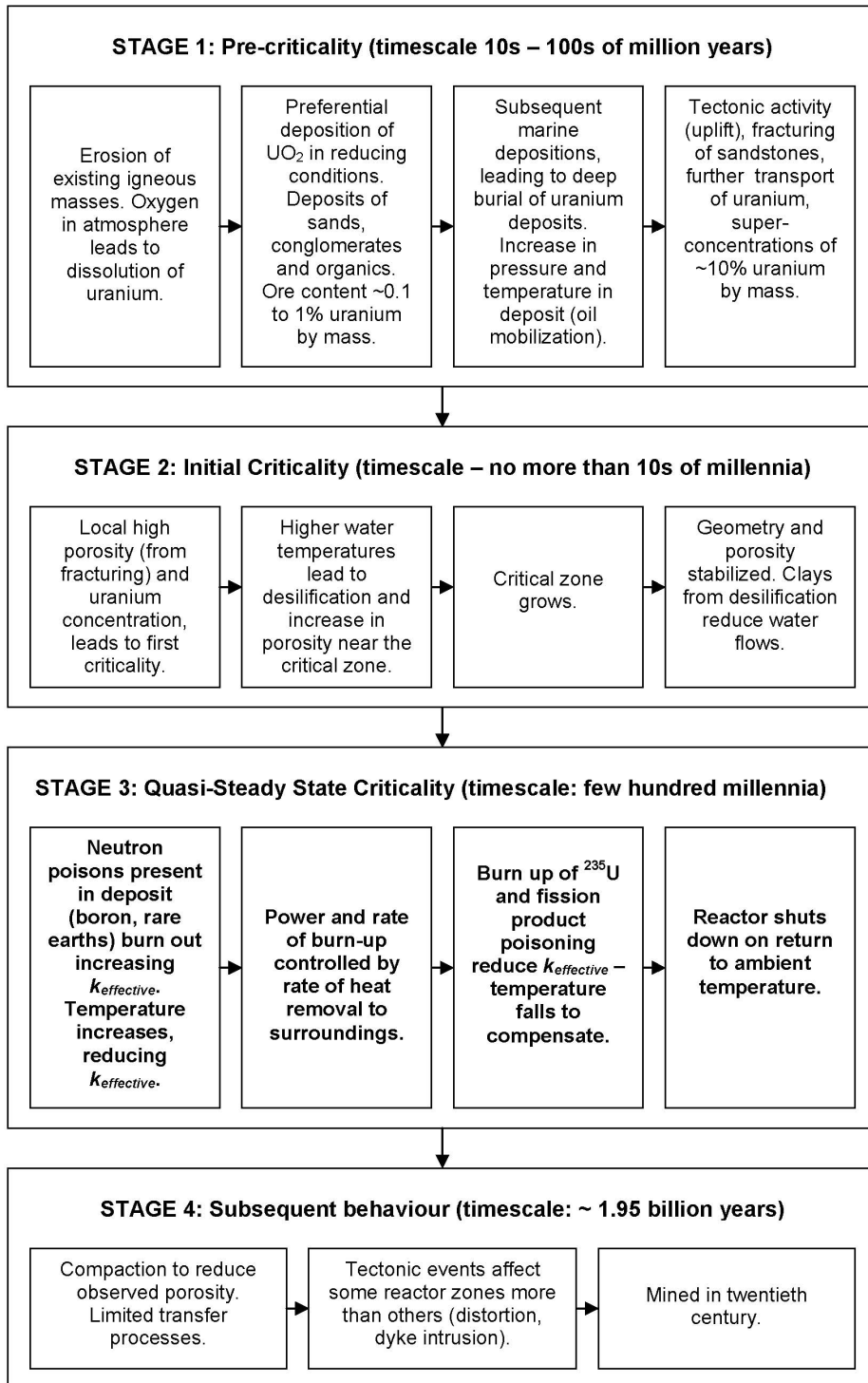


FIG. 2. Schematic diagram of the production and operation of the Oklo zone 2 natural reactor.

approach. Further detailed discussion of the modelling is presented by Mason *et al.* (2011).

The QSS model: application to an Oklo reactor

Given the passage of about two billion years since the Oklo natural reactors occurred, understanding the processes, and taking measurements have limitations. As zone 2 has a relatively large amount of measured data available, this zone was chosen for comparison with QSS model calculations. In terms of undertaking a QSS simulation of Oklo the following were taken from the conclusions of analyses by Naudet (1991): (1) a water-filled porosity during operation of 37% in a host rock of sandstone, constant for the duration of the nuclear reaction; (2) a lenticular reaction (critical) zone about 1 m thick and 10 m in diameter; (3) an initial uranium enrichment of 3.68%; (4) an initial temperature of 160°C; and (5) an initial presence of four neutron poisons in quantities as deduced by Naudet. The neutron poisons are boron-10 (¹⁰B), samarium-149 (¹⁴⁹Sm), gadolinium-155 (¹⁵⁵Gd) and gadolinium-157 (¹⁵⁷Gd).

An initial analysis of the burn-up processes for 3.68% enriched uranium (Mason *et al.*, 2011) showed that the important nuclides (i.e. those with the potential to affect *k_{effective}*) were three uranium isotopes, two plutonium isotopes, the neutron

poisons listed above, and three fission products; rhodium-103 (¹⁰³Rh), neodymium-143 (¹⁴³Nd) and samarium-149 (¹⁴⁹Sm). Together with two inert nuclides to represent the stable end points of fission products and captures, these nuclides form the burn-up model for a QSS calculation, as shown in Fig. 3.

With the burn-up model defined, identification of a suitable just critical initial condition, an interpolation function for *k_{effective}* (dependent on the temperature and the concentration of each of the non-inert nuclides in Fig. 3 and obtained from calculations using detailed neutronic software), and heat transfer parameters for sandstone, a calculation can be undertaken with the QSS model to simulate the Oklo zone 2 reactor.

To establish whether the results of the QSS simulation are reasonable, data are clearly required for a comparison. For Oklo zone 2, there are a number of independent measurements that can be used. From the concentrations and isotopic compositions of fission products the burn-up can be deduced. For instance, neodymium from fission of ²³⁵U contains little neodymium-142 (¹⁴²Nd), whereas this isotope comprises more than a quarter of naturally occurring neodymium. The difference allows the amount of fission that occurred to be estimated (Naudet, 1991), which in turn allows the neutron fluence (the total number of neutrons passing

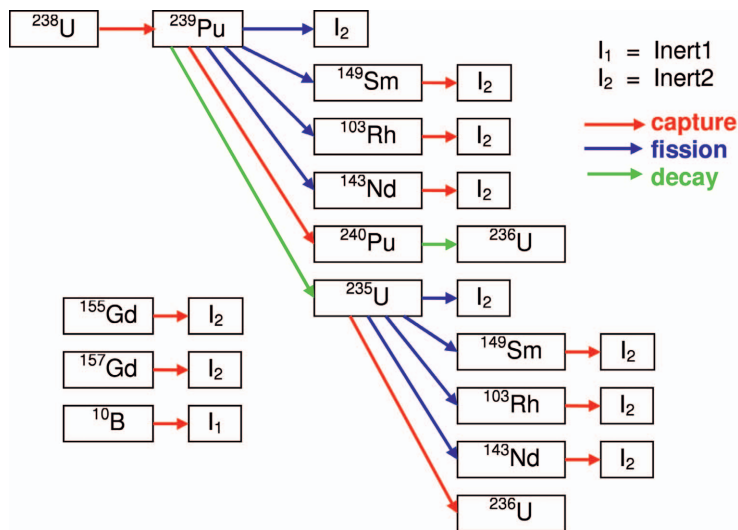


FIG. 3. Fissile material nuclear burn-up model for the application of the QSS model to the Oklo zone 2 natural reactor.

through a unit area during the criticality) to be estimated. Naudet (1991) concluded that the fluence was about 8.7×10^{20} neutrons cm^{-2} .

The burn-up of ^{235}U can also be estimated from the measured uranium enrichment, which is less than that found in naturally occurring uranium. However, the difference is less than that required to explain the estimated burn-up based on the fission product measurements. The reason is that ^{239}Pu is created by neutron capture in ^{238}U . Some of the ^{239}Pu would have undergone fission but some would have decayed to form ^{235}U . The ratio of fission to decay depends on the power and duration of the reactor. It was deduced by Naudet (1991) that a reactor lifetime of about 620,000 years is consistent with the measured uranium enrichment. The analysis also allows the burn-up of uranium, which is the fraction of uranium mass (both ^{235}U and ^{238}U) which has been converted to other nuclides through fission and capture, to be estimated; giving an average of 0.7% over the reactor.

The presence of uranium oxide in the form of uraninite indicates operating temperature in the range 300–350°C. The ambient temperature at the depth at which the zone 2 reactor is believed to have operated is 160°C. It is concluded that the temperature rise produced by the zone 2 reactor was about 140°C.

The above values, deduced independently from measurements are a sufficient set to undertake a meaningful comparison with a *QSS* model simulation of the Oklo zone 2 reactor. Even before doing so, the evidence suggested that a comparison should be reasonable, as calculations

for GDF scenarios with 3% enriched uranium (Mason *et al.*, 2009a) show similar behaviour, even though the sustaining mechanism and geometry are different.

A further useful comparison is with analysis undertaken by Naudet (1991) to understand the contributions to maintaining $k_{\text{effective}}$ at unity. This simple analysis compared the dominant effects acting to increase $k_{\text{effective}}$ with those acting to reduce it. This analysis produced a simple estimate of the reactor temperature as a function of neutron fluence, which can be compared directly to the results of the *QSS* model simulation for Oklo zone 2. Figure 4 shows a comparison of the *QSS* model results when used to simulate Oklo zone 2 against the $k_{\text{effective}}$ balance model in Naudet (1991). Given the fact that both models are approximations, and that no parameters were tuned in the *QSS* model to attempt a ‘fit’ with Naudet’s curve, this is considered a good indication that the *QSS* model produces sensible results.

Table 1 shows a comparison of the *QSS* model results with each of the estimates from field measurements as discussed above. Again, no parameters were tuned in the *QSS* model to attempt a ‘fit’ to any of the measured values. The table shows good agreement between the measured data and the results from the *QSS* model simulation, with the agreement being within a factor of two for each of the quantities compared.

One effect thought to be missing from the *QSS* model, as applied to Oklo zone 2, is the spread of the fission region to the surrounding material. This is expected to occur due to the leakage of

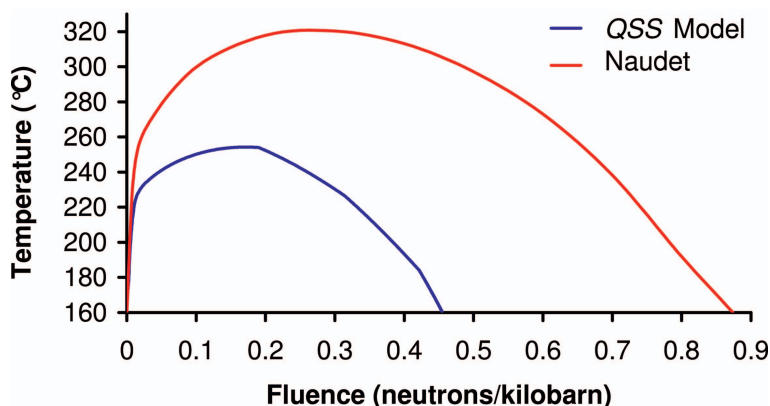


FIG. 4. Comparison of temperature rise as a function of neutron fluence for different models of the Oklo zone 2 natural reactor (1 kilobarn = 10^{-25} m^2).

TABLE 1. Comparison of estimated (Naudet, 1991) and calculated (*QSS* model) values for Oklo zone 2.

Parameter	Units	Estimated	<i>QSS</i> model
Duration	millennia	620	500
Burn-up of uranium	%	0.7	0.9
Fluence	neutrons cm ⁻²	~8.7 × 10 ²⁰	4.5 × 10 ²⁰
Temperature rise	°C	~140	100

neutrons from the initially critical central region, causing burn-out of poisons from the surrounding region. Naudet (1991) suggested that this resulted in the growth of the critical region and a consequent lengthening of the duration of the operation of the reactor. The growth of the critical region is not included in the *QSS* model and therefore it is to be expected that the simulation will underestimate the duration. This is evident from Table 1, which shows a 20% underestimate of the duration. The measured burn-up is 22% lower than calculated with the *QSS* model and may be linked to the growth of the critical region leading to lower burn-up in the extended reaction zone. Finally the *QSS* model temperature rise is about 30% lower than suggested by the observations. The calculated value will depend on the composition of the fissile region and its thermal properties (particularly the thermal conductivity). No attempt was made to tune these parameters to obtain a better fit.

Conclusions

The *QSS* model has been developed to understand the study of hypothetical post-closure criticality events in a GDF. The model has been successfully applied to simulate the Oklo zone 2 natural reactor and the results of the calculation are in good agreement with values inferred for this reactor by Naudet (1991). In particular, the duration, fluence, temperature rise and burn-up of the uranium were all found to agree with values deduced from the Oklo observations to within a factor of two, without the need to tune any parameters in the *QSS* model. Through this successful comparison exercise with a natural analogue, it is concluded that important evidence to support the *QSS* modelling approach has been presented, and builds confidence in the model application to GDF scenarios.

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