

APPLICATION OF A DISPOSAL FACILITY POST-CLOSURE CRITICALITY MODEL TO OKLO ZONE 2

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ABSTRACT

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. At this stage, without a site or geology, the generic safety case is based on understanding the scientific and engineering principles supporting geological disposal. For post-closure safety RWMD has undertaken research to understand the features and processes that ensure a low likelihood of criticality, and 'what-if' post-closure criticality scenarios.

A 'what-if' scenario for GDF criticality is the mobilisation of fissile material from a number of waste packages and its subsequent localised accumulation. Should sufficient fissile material accumulate, a quasi-steady-state (QSS) criticality could result, where an increase in reactivity from continued accumulation is balanced by a temperature increase. The QSS model has been developed to simulate such a criticality.

A postulated QSS criticality could continue for many millennia so building confidence in the model is difficult. However, the Oklo reactors operated for similar durations around two billion years ago, so a study has been undertaken to compare the available information with a QSS model simulation of this natural analogue.

The paper describes the QSS model approach and its application to an Oklo reactor. The results are found to be in agreement with observational evidence for burn-up, temperature and duration, building confidence in applying the QSS model to the 'what-if' scenario for GDF criticality.

Key Words: geological disposal, transient, criticality, Oklo

1 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 [1]. The licensing process will include close examination of detailed safety cases covering all aspects of the disposal system. It will not be possible to produce a full safety case until there is a site and detailed design for a GDF. At this stage the safety case is based on an understanding of the scientific and engineering principles supporting geological disposal and as it is not specific to a site or geology, it is called a 'generic' safety case. A suite of documents form the generic Disposal System Safety Case (DSSC) [2]. In support of the DSSC the RWMD has undertaken a programme of research [3] to obtain a better understanding of the features and processes that ensure that the likelihood of a criticality is low, and of 'what-if' scenarios of sufficient accumulation of fissile materials occurring post-closure to give rise to a criticality.

A 'what-if' scenario for a criticality under GDF conditions is the mobilisation of fissile material from a number of waste packages and its subsequent slow accumulation at some location within the

GDF or its immediate vicinity. In the unlikely event that sufficient fissile material accumulated, a quasi-steady-state (QSS) criticality could result. This is a system with negative temperature feedback, where the increase in reactivity from the addition of further fissile material is balanced by the reduction in reactivity from a gradual rise in temperature, thus maintaining the reactivity close to zero. The QSS computer model has been developed to calculate the evolution of such a criticality [4,5,6].

Such postulated criticalities could have durations of many millennia. This raises the question of how to build confidence in such a model, given that it is impractical to conduct an experiment over such a large timeframe. Since the Oklo natural reactors in Africa operated for similar durations around two billion years ago [7], a study has been undertaken to compare the available information for Oklo zone 2 with a QSS model simulation of this natural analogue. The choice of zone 2 is described in Section 3.

The QSS model contains a reactivity function which has a non-linear dependence on the composition and temperature (see Section 2.2). In order to model the zone 2 reactor at Oklo a reactivity function was constructed that includes the effects of the poisons thought to be important [7]: boron, gadolinium and samarium. FISPIN [8] calculations of burn-up revealed that isotopes of the fission products neodymium and rhodium also have a significant effect on reactivity and so their effects were included in the reactivity function. With this reactivity function it was possible to simulate the operation of the Oklo zone 2 reactor.

The paper describes the QSS model approach in Section 2, and a summary of information that is available for the Oklo natural reactors in Section 3. Section 4 describes the application of the QSS model to the Oklo zone 2 natural reactor and a comparison with the available information. Conclusions are drawn in Section 5. The results are shown to be in agreement with the observational evidence on burn-up, temperature and duration, thus building confidence in the use of the QSS model to simulate postulated post-closure criticalities in Geological Disposal Facilities.

2 DESCRIPTION OF THE QSS MODEL

2.1 Basic Equations

In the QSS model the system is assumed to remain in a state of zero reactivity, $\rho = 0$, (equivalently a $k_{effective}$ value of unity, where $\rho = 10^5(k_{effective} - 1)/k_{effective}$ is the reactivity in mNiles), in which increases in reactivity due to change in composition (e.g. arrival of fresh fissile material or burnout of poisons) are balanced by reductions in reactivity due to temperature rises or burnout of fuel [4,5]. This is expressed in terms of the reactivity feedback coefficients: ρ_i , the number density feedback coefficient of the i^{th} nuclide (mNiles kmole⁻¹ m³) and ρ_T , the temperature feedback coefficient (mNiles K⁻¹). The system is initialized with zero reactivity and the rate of change of reactivity is set to zero:

$$\sum_{i=1}^N \rho_i \dot{n}_i + \rho_T \dot{T} = 0 \quad (1)$$

The change in concentration of the i^{th} nuclide ($i=1 \dots N$), n_i (kmoles m⁻³), is due to:

1. A source of the nuclide, S_i (kmole m⁻³ s⁻¹);
2. Loss due to decay with half life λ_i (s);
3. Gain due to decay of another nuclide;

4. Loss due to capture, with cross-section σ_{ci} (m^2);
5. Gain due to capture in another nuclide;
6. Loss due to fission, with cross-section σ_{fi} (m^2);
7. Gain due to fission of another nuclide.

The conservation equations for the nuclides can be written as:

$$\dot{n}_i = S_i + \sum_j \varepsilon_{ij} \frac{\ln(2)}{\lambda_j} n_j + \phi \sum_j \chi_{ij} n_j \quad (2)$$

where ϕ is the neutron flux ($\text{neutrons m}^{-2} \text{s}^{-1}$), and, for each nuclide the constants ε_{ij} and χ_{ij} are defined by:

$$\varepsilon_{ij} = \begin{cases} -1 & j = i \\ +1 & j \text{ decays to } i \\ 0 & \text{otherwise} \end{cases}, \quad \chi_{ij} = \begin{cases} -(\sigma_{fi} + \sigma_{ci}) & j = i \\ v_{ij} \sigma_{fi} & i \text{ is formed by fission of } j \\ \sigma_{ci} & i \text{ is formed by capture in } j \\ 0 & \text{otherwise} \end{cases}, \quad (3)$$

and v_{ij} = number of atoms of nuclide i resulting from the fission of one atom of nuclide j .

The power can be expressed in terms of the neutron flux, fission cross-section and energy per fission. The power must also equal the sum of the rate of increase of sensible heat of the reactor and the rate of heat loss to the surroundings. Equating these two expressions yields:

$$P = \phi E_f V_f N_A \sum_i \sigma_{fi} n_i = MC \Delta \dot{T} + \bar{H} \Delta T \quad (4)$$

where

$$\begin{aligned} V_f &= \text{volume of Fissile Material (FM) region} \\ E_f &= \text{energy per fission } (3.20435 \times 10^{-11} \text{ J}) \\ N_A &= \text{Avagadro's number } (6.022 \times 10^{26} \text{ molecules kmole}^{-1}) \\ M &= \text{mass of reactor material (kg)} \\ C &= \text{specific heat capacity of reactor material (J kg}^{-1} \text{ K)} \\ \Delta T &= \text{temperature rise above ambient, } T - T_0, \text{ (K)} \\ \bar{H} &= \text{heat transfer coefficient (W K}^{-1}\text{)}. \end{aligned} \quad (5)$$

Equation 4 is used to eliminate the neutron flux from Equation 2, in favour of the temperature rise. Equations 1 and 2 then form a complete set of $N+1$ ordinary differential equations (ODEs) for the $N+1$ unknowns ($n_1, \dots, n_N, \Delta T$). The Mathematica code [9] is used to numerically integrate the ODEs, using user-supplied reactivity feedback coefficients, which are a function of composition and temperature.

2.2 The Reactivity Function

In order to calculate the reactivity feedback coefficients, a reactivity function is constructed and differentiated with respect to the nuclide concentrations and temperature. The reactivity function consists of a set of calculated reactivities at specified compositions and temperatures. The value of the reactivity at any point within the specified range of compositions and temperatures is obtained by interpolation between the neighbouring evaluated reactivities. For restricted ranges of

temperature and composition sufficient reactivities may be evaluated for linear interpolation to provide adequate accuracy. This was the case for the Oklo zone 2 simulations, for which several thousand reactivities were calculated [10] using the MONK code [11].

On the other hand, the ‘what-if’ scenarios for a hypothetical GDF criticality were evaluated for a wide range of sizes, temperatures and compositions. In order to cover such a wide range of scenarios with a practicable number of reactivity calculations, non-linear interpolation was used, in order to accommodate wider separation of the evaluation points in parameter space. The interpolation scheme uses the buckling formula, relating the system multiplication factor, $k_{effective}$, to the multiplication factor for an infinite medium, k_{∞} , the geometric buckling, B^2 , and the migration area, M^2 , [12]:

$$k_{effective} = \frac{k_{\infty}}{1 + B^2 M^2}. \quad (6)$$

Analytic expressions are available for the geometric buckling for a range of geometries. For a cylinder of radius R and height H the geometric buckling is [12]:

$$B^2 = \left(\frac{2.405}{R + l_R} \right)^2 + \left(\frac{\pi}{H + l_H} \right)^2 \quad (7)$$

where l_R/l_H is the ratio of the radial to axial extrapolation lengths.

The extrapolation length for water moderated systems is typically a few centimetres, so for reactors that are significantly larger than this the extrapolation length can be ignored, with little loss of accuracy [12]. The values of $k_{effective}$ and k_{∞} are calculated using computer codes such as MONK [11] or WIMS [13]. The migration area can then be evaluated using Equations 6 and 7.

For the reactivity function interpolation scheme k_{∞} is expressed in terms of the four factor formula [14]:

$$k_{\infty} = \epsilon p f \eta \quad (8)$$

where,

- ϵ is the fast fission factor (ratio of the total number of fission neutrons to the number of fission neutrons from just thermal fissions)
- p is the resonance escape probability (fraction of fission neutrons that manage to slow down from fission to thermal energies without being absorbed)
- f is the thermal utilization factor (the probability that a neutron that gets absorbed does so in the fuel)
- η is the reproduction factor (the number of fission neutrons produced per absorption in the fuel).

It is then broken down into the product of a thermal component, k_{th} , and a fast and resonance region component, k_{fr} :

$$k_{\infty} = k_{fr} k_{th} \text{ where } k_{fr} = \epsilon p \text{ and } k_{th} = f \eta. \quad (9)$$

The thermal component is calculated from thermal averaged cross-sections for the nuclides present in the critical region. The value of k_{fr} is obtained as the ratio of k_{∞} to k_{th} and is stored at each of the chosen compositions.

The calculated values of k_{fr} and M^2 at the chosen sizes and compositions form the basis of the reactivity function. In order to obtain $k_{effective}$ at any point the values of k_{fr} and M^2 are obtained by

linear interpolation in composition, using the stored values, and then $k_{effective}$ is reconstructed using equations 6, 7 and 9.

2.3 Burnup Schemes

Burnup schemes and decay chains are required for the QSS model to determine the contributions in Equation 3 to Equation 2. For example, in a critical event, the capture of a neutron in ^{235}U will form ^{236}U , while decay of ^{239}Pu will form ^{235}U .

When the QSS model is used to simulate a hypothetical post-closure criticality in a GDF the criticality is assumed to be sustained by the continued arrival of fresh fissile material. The nuclides in the fissile material (FM) region on which the reactivity depends are ^{235}U , ^{236}U , ^{238}U , ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{242}Pu [5]. In early calculations, materials were used to represent the fission products resulting from the burn-up of the fissile U and Pu nuclides, but were found to have little effect on the reactivity and are therefore neglected. Similarly, lead and molybdenum have been used to represent the stable endpoints of the decay chains of the unstable actinides and fission products, but again were found to have little effect on the reactivity and are therefore neglected [5]. The simplified burn-up and decay chains used for GDF calculations are displayed in Figure 1.

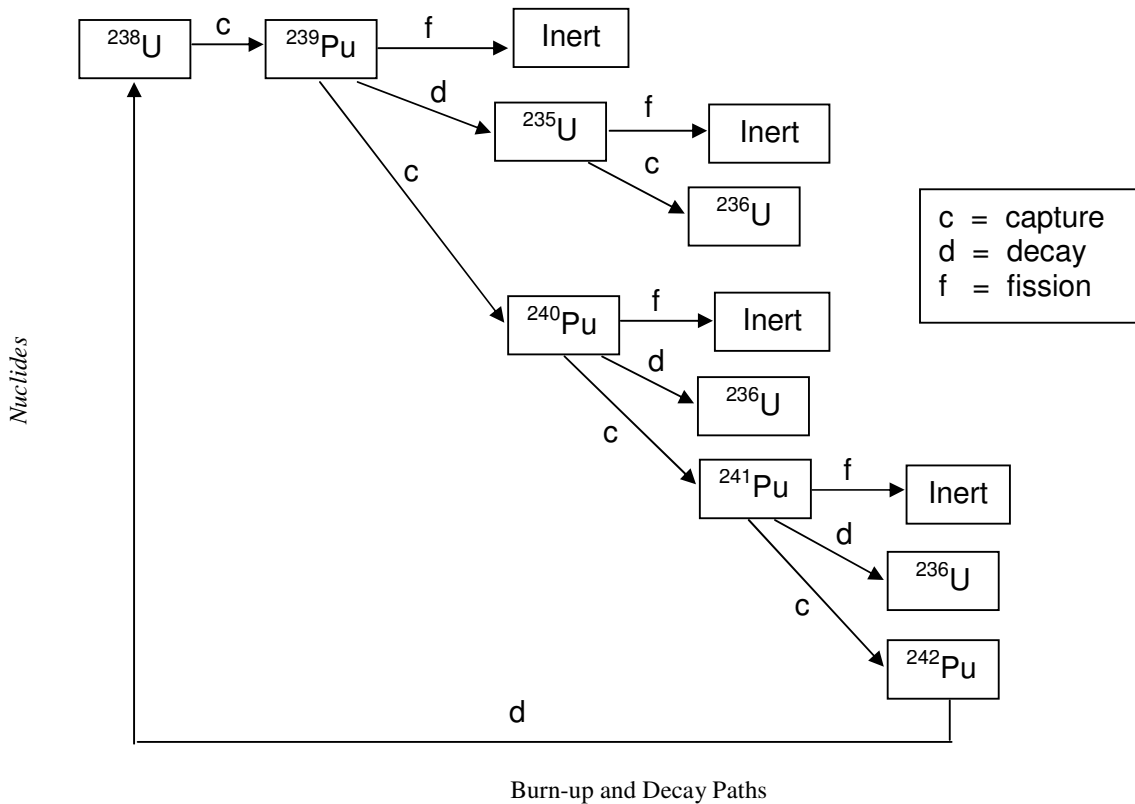


Figure 1. Burnup Scheme for GDF ‘what-if’ Calculations

The Oklo reactors, described in more detail in the following section, were fuelled by natural uranium. The plutonium isotopes present would have been created by neutron capture in the uranium nuclides and subsequent decay and only ^{239}Pu is created in significant quantities by this process. Therefore the uranium and plutonium nuclides considered are ^{235}U , ^{236}U , ^{238}U , and ^{239}Pu . In addition the poisons ^{10}B , ^{149}Sm , ^{155}Gd and ^{157}Gd are included in order to simulate the sustainability of the criticality due to the burn-up of poisons [7]. Furthermore, FISPIN [8]

calculations indicate that the fission products ^{103}Rh and ^{143}Nd have an effect on reactivity of similar magnitude to the above poisons and are therefore included in the reactivity function [10]. The resulting decay chain used for the Oklo calculations is displayed in Figure 2. Two inert species are included to allow for a small molecular weight inert product produced from capture in ^{10}B (Inert 1) and all other inert products with larger molecular weights (Inert 2).

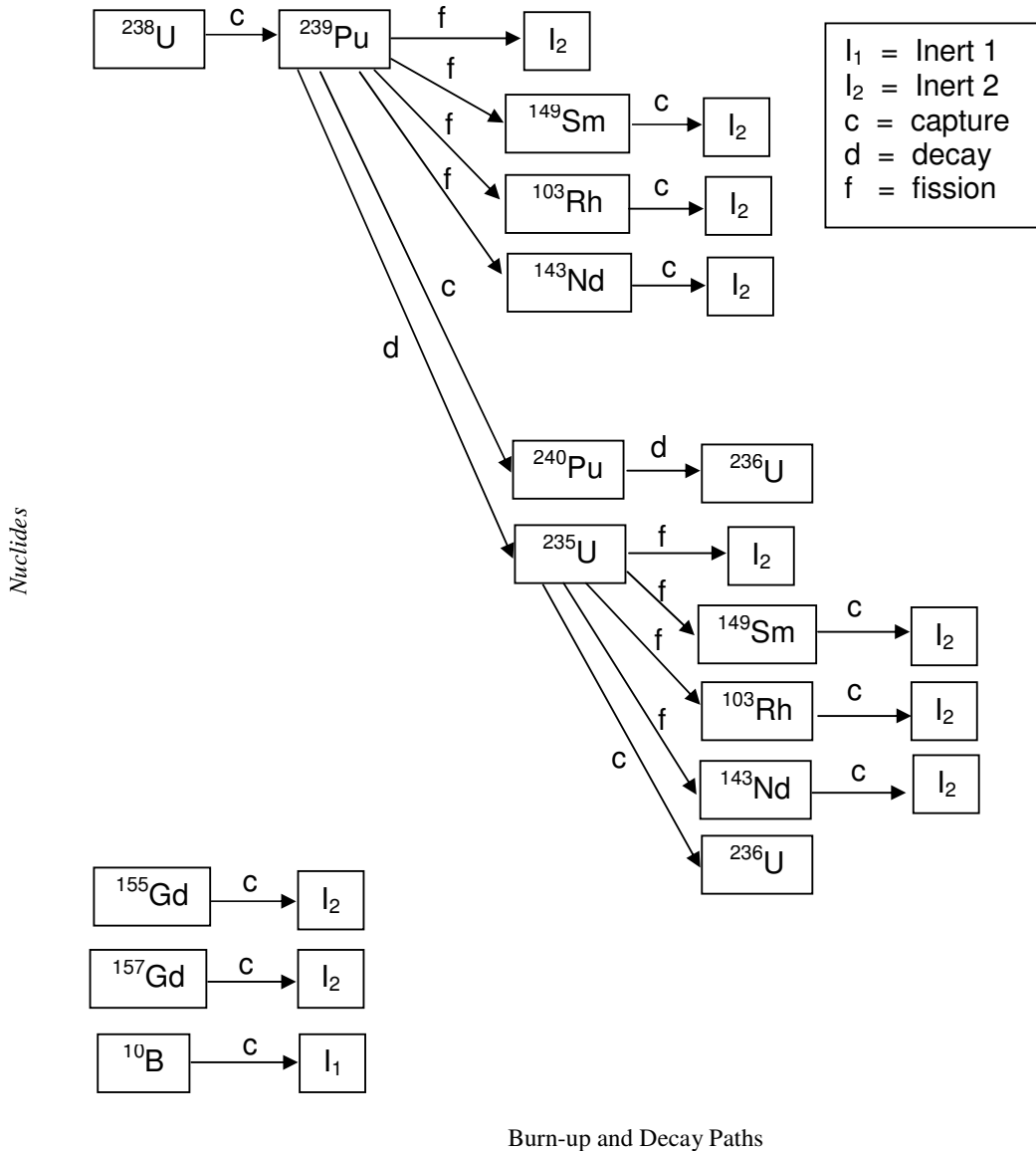


Figure 2. Burnup Scheme for Oklo Calculations

2.4 Using the QSS Model

To undertake calculations with the QSS model the following steps are required:

1. Establish the initial conditions for a just critical ($\rho=0$ or $k_{effective}=1$) system. This will be for a given geometry for the reactor region, and will determine the initial temperature and nuclide concentrations – i.e. the initial conditions for the $N+1$ variables ($n_1, \dots, n_N, \Delta T$).

2. Based on estimates of the likely variation in temperature and nuclide concentrations, construct a reactivity (interpolation) function for the fixed reactor geometry which gives the reactivity as a function of temperature and the required nuclide concentrations, $\rho = \rho(n_1, \dots, n_N, T)$. For the ‘what-if’ GDF scenarios this can be taken from the more general function described in section 2.4, using the function at fixed geometry, R and H .
3. From the reactivity function calculate the feedback coefficients as functions of temperature and nuclear concentrations. i.e.

$$\rho_T(n_1, \dots, n_N, T) = \frac{\partial \rho}{\partial T} \text{ and } \rho_i(n_1, \dots, n_N, T) = \frac{\partial \rho}{\partial n_i} \text{ for } i = 1, \dots, N.$$

4. With suitable burnup and decay chains (including cross-sections and half-lives), source terms (where applicable), a heat transfer coefficient and specific heat capacities, the system of $N+1$ ODE’s can be integrated to give temperature and nuclide concentrations as functions of time.

3 THE OKLO NATURAL REACTORS

Understanding of the functioning of the ‘fossil reactors’ was developed by a French team from the Commissariat à l’Energie Atomique (CEA) led by Roger Naudet and published 1991 [7]. At the time ten reactor zones had been identified and subsequently a few more reactor zones were identified at Oklo and Okelobondo and one at the Bangombé site also in the same geological basin. Additional studies were performed in the EC-funded project: Oklo –Natural Analogue. Some of this work is reported by Gauthier-Lafaye et al [15]. Reviews of the Oklo phenomena have been prepared by Oversby [16] and Zetterström [17] for SKB and by Smellie [18] for Nirex¹.

Naudet [7] focussed his attention initially on zone 2 which was subjected to detailed measurements. In view of the relatively large amount of measured data available, this zone was chosen for comparison with QSS model calculations.

Naudet reached the conclusion that the porosity in the core of the reactor must have been much higher than it was when discovered, and that this porosity was filled with water, to act as a moderator. The neutron spectral index was determined based on isotopic compositions and found to be consistent with a water-filled porosity during operation of ~37%. This value was also consistent with the inferred porosity at the end of operation of the reactor, based on the need to obtain a neutron multiplication factor of unity. From these two pieces of information, Naudet drew the conclusion that, at least for zone 2, the reactor zone structure was nearly constant for most of the operating period of the reactor.

It is a feature of zone 2, and some of the other high burn-up zones, that the core reactor contains no silica. The mechanism for removing the silica is assumed to be thermal convection induced by the heat from the fission process dissolving the silica and carrying it away from the heated core. Naudet argued that this desilification period was short compared with the overall lifetime of the reactor (again referring to the spectral index data). He further suggested that, in the later operating period, convection was suppressed because the removal of the permeable sandstone particles left the much less permeable clay residuals behind. Thus Naudet was able to come up with a consistent picture for the operation of zone 2, based on the assumption that the materials were in their original locations, except that the porosity had been reduced by subsequent events.

Naudet argues that continued accumulation of uranium did not occur during the majority of the time of the reactor because it would have changed the composition of the uranium deposits in the

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

reactor zone. He concluded that the burn-up was controlled by the balance between the burn-up of poisons adding reactivity and the temperature increase and burn-up of fissile material in the reactor core which reduced the reactivity. Hence modelling zone 2 required modification to the reactivity function in QSS to include the effect of the burn-up of poisons on the reactivity, as discussed in Section 2.

A number of parameters pertaining to the Oklo reactors can be determined from experimental evidence. From the concentrations and isotopic compositions of fission products the burn-up can be deduced. For instance, neodymium from fission of ^{235}U contains little ^{142}Nd , whereas this isotope comprises more than a quarter of naturally occurring neodymium. This allows the naturally occurring neodymium to be subtracted from the neodymium vector in the spent reactor core. The resulting neodymium vector has the same spectrum as that from ^{235}U and allows the amount of fission that occurred to be estimated. This allows the energy released or the fluence to be estimated. It is concluded in [7] that the fluence was about $8.7 \times 10^{20} \text{ n cm}^{-2}$.

The burn-up of ^{235}U can also be estimated from the measured ^{235}U enrichment, which is much 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 the beta decay of ^{239}U produced by neutron capture in ^{238}U (the whole process taking less than two and a half days). Some of the ^{239}Pu will fission due to the neutron flux and some will undergo alpha decay to form ^{235}U . The ratio of fission to decay depends on the power and duration of the reactor. It is found that a reactor lifetime of about 620,000 years is consistent with the production of the required amount of ^{235}U from this route to account for the difference in burn-up estimated from the ^{235}U enrichment to that deduced from the fission product concentrations. The above analysis also allows the burn-up of ^{235}U to be estimated, giving an average of 0.7% over the reactor.

The presence of UO_2 in the form of uraninite indicates operating temperature in the range 300-350°C, which is the range over which uraninite forms. Other evidence is available from isotopic analysis of noble fission gases. 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.

4 COMPARISON OF QSS RESULTS WITH OKLO DATA

With the concentrations of poisons suggested by Naudet [7], the Boron worth is 6 Niles, the Gadolinium 1 Nile and the Samarium 0.4 Niles. When the system becomes critical the poisons start to burn out, increasing the reactivity. The chain reaction produces power from the fissions, which raises the temperature, thus restoring the reactivity to zero.

By constructing a reactivity function for Oklo zone 2 as described in Section 2.2, and the burn-up and decay chains from Figure 2, a QSS calculation was undertaken. Figure 3 shows the contributions of different nuclides and temperature to the rate of change of reactivity (the different contributions to Equation 1, rescaled to a timescale of millennia). The contributions of ^{236}U and ^{238}U are not shown, since they are too small to be seen. It can be seen that for the first ~30,000 years the burnout of Gadolinium gives rise to the largest increase in reactivity (with contributions from Boron and Samarium). This is counteracted by the rise in temperature. After this most of the Gadolinium has burned out and by roughly 100,000 years the Samarium has reached an equilibrium concentration, at which the rate of burnout is equal to the rate of production from fission (as ^{149}Sm is a fission product).

From approximately 30,000 years to 300,000 years the main increase in reactivity is due to the burnout of Boron, which is counterbalanced by the reduction in reactivity due to the burn-up of the ^{235}U fuel, with a contribution due to temperature variations. Beyond about 150,000 years the

temperature is falling, thus producing an increase in reactivity. Beyond roughly 350,000 years the increase in reactivity due to cooling roughly balances the reduction due to the burn-up of fuel, with some contribution from the burnout of Boron. By the time the system becomes sub critical approximately 80% of the Boron has burned out.

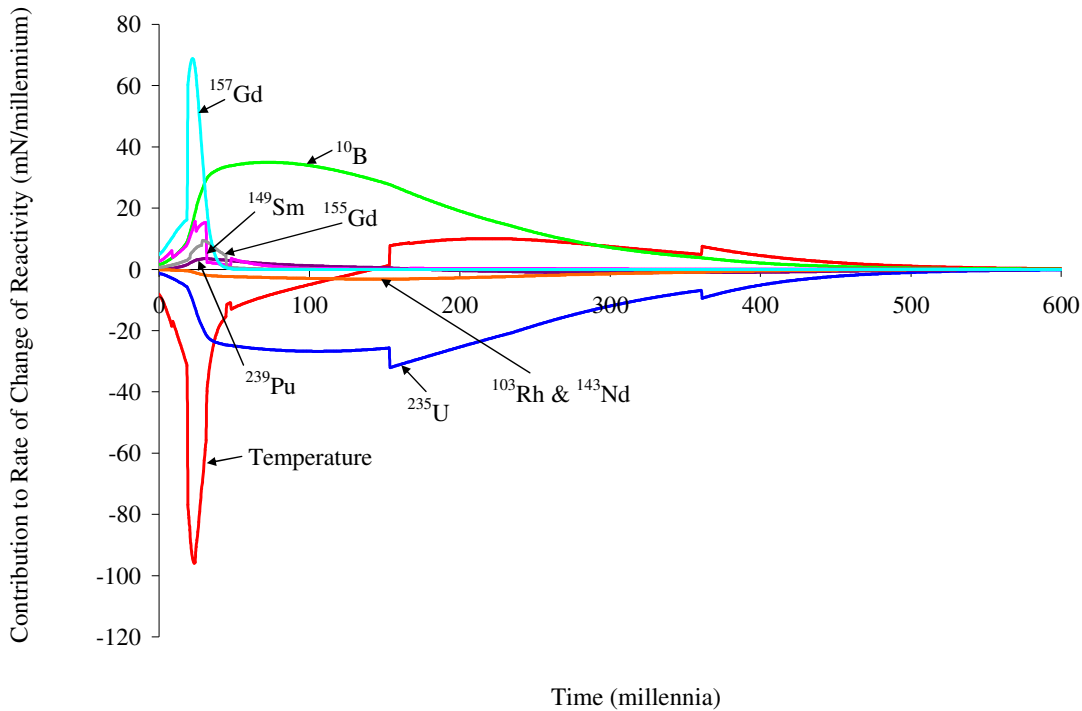


Figure 3. Effects on Rate of Change of Reactivity

As noted in Section 3, a number of parameters relating to the operation of the zone 2 reactor have been estimated, without recourse to the use of neutron transport models. These include the duration, burn-up of fuel, fluence and temperature rise. The experimentally estimated values are displayed in Table I and compared with the calculated values using the QSS model.

Table I. Comparison of Measured and Calculated Values for Oklo Zone 2

Parameter	Units	Measured	QSS
Duration	millennia	620	500
Burnup	%	0.7	0.9
Fluence	n cm ⁻²	<14.5x10 ²⁰	4.5x10 ²⁰
Temperature Rise	°C	~140	100

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 neutrons from the initially critical central region, causing burnout of poisons from the surrounding region. This spreading of the chain reaction from the initially critical central region is believed to have resulted in the growth of the critical region and a consequent lengthening of the duration of the operation of the reactor [7]. The growth of the critical region is not included in the QSS model and therefore it is to be expected that the QSS simulation will underestimate the duration and fluence. This is evident from Table I, which shows a 20% underestimate of the duration. The fluence

displayed in the table is an upper bound and Naudet's modelling yields an estimated fluence of 8.7×10^{20} n cm⁻² [7], which is within a factor of two of the QSS calculation.

The measured burn-up is 22% lower than the QSS calculation 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 has been made to tune these parameters to obtain a better fit.

5 CONCLUSIONS

The QSS model, developed for the study of hypothetical post-closure criticality events in a GDF, has been successfully applied to simulate the Oklo zone 2 natural reactor. The results of the QSS calculation are found to be in good agreement with values inferred for the Oklo zone 2 reactor by Naudet. 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. It is concluded that the application of the QSS model to the Oklo natural reactors provides important evidence to support the QSS model, and builds confidence in its application to GDF scenarios, such as those presented at ICNC 2007 [4].

6 ACKNOWLEDGEMENTS

The authors would like to thank NDA for funding this work and in particular Peter Wood for his continued support of the modelling. They would also like to thank Jim Gulliford (OECD/NEA) for many stimulating discussions and contributions on this topic. Thanks also to Julie Martin (Serco) for undertaking the MONK calculations in support of the Oklo modelling.

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