C. R. Geoscience 343 (2011) 738–748

Contents lists available at SciVerse ScienceDirect

Comptes Rendus Geoscience

www.sciencedirect.com

Petrology, geochemistry (Isotopic geochemistry)

Inception and evolution of Oklo natural nuclear reactors

Génèse et évolution des réacteurs nucléaires fossiles d'Oklo

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ARTICLE INFO

Article history:
Received 11 August 2011
Accepted after revision 30 September 2011

Presented by René Blanchet

Keywords:
Oklo
Natural reactor
Criticality
Reactivity balance
Uranium deposit
Monte-Carlo simulation
Geological Repository

ABSTRACT

The occurrence of more than 15 natural nuclear Reactor Zones (RZ) in a geological environment remains a mystery even 40 years after their discovery. The present work gives for the first time an explanation of the chemical and physical processes that caused the start-up of the fission reactions with two opposite processes, uranium enrichments and progressive impoverishment in ^{235}U. Based on Monte-Carlo neutronics simulations, a solution space was defined taking into account realistic combinations of relevant parameters acting on geological conditions and neutron transport physics. This study explains criticality occurrence, operation, expansion and end of life conditions of Oklo natural nuclear reactors, from the smallest to the biggest ones.

Mots clés :
Oklo
Réacteurs naturels
Criticité
Bilan de réactivité
Gisement d'uranium
Simulation Monte-Carlo
Stockage géologique

RÉSUMÉ

La présence d'une quinzaine de zones de réaction nucléaires (RZ) dans un milieu géologique reste un mystère 40 ans après leur découverte. Le présent travail donne pour la première fois une explication des évolutions ayant conduit au démarrage de ces cœurs au croisement de deux processus antagonistes, la concentration en uranium et l'appauvrissement progressif en son isotope 235. À partir de simulations Monte-Carlo, il a été possible de définir un espace de solutions réalistes — fonction des principaux paramètres tant physiques que géologiques — justifiant la criticité et expliquant l'expansion, le fonctionnement et les conditions d'arrêt des plus petits aux plus gros réacteurs d'Oklo.

Version française abrégée

La zone de réaction 9 (RZ9) représentée sur la Fig. 1 fait partie de la quinzaine de réacteurs nucléaires naturels découverts depuis 1972 dans les mines d'uranium d'Oklo et Okelobondo au Gabon. Plus connu sous le nom

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1631-0713/$ – see front matter
doi:10.1016/j.crte.2011.09.008

« Phénomène Oklo », le démarrage, le fonctionnement et l'arrêt de ces réacteurs sans intervention humaine ont eu lieu il y a 1950 millions d'années (Bodu et al., 1972 ; Gauthier-Lafaye, 1997, 2006 ; Neuilly et al., 1972). La géométrie actuelle du réacteur, étendue sur 12 m de long et 7 m de large, peut être assimilée à une forme lenticulaire de quelques dizaines de centimètres d'épaisseur (Gauthier-Lafaye, 1979). L'intérêt particulier pour ce réacteur provident de la difficulté à expliquer l'occurrence de la criticité dans un tel système. En effet, des simulations plus anciennes des réacteurs d'Oklo (Naudet, 1991) n'ont pas pu expliquer la criticité d'un si petit réacteur naturel.

Les possibilités offertes actuellement par le code de Monte-Carlo MCNP (Harmon et al., 2004 ; X-5 Monte-Carlo Team, 2003) nous ont ouvert la voie de simulations réalistes et détaillées de ce système dans son environnement. La taille minimale nécessaire à la criticité a ainsi été étudiée en fonction de gammes réalistes de porosité, teneur en matière organique (OM), eau, uranium, et en poisons neutroniques. Plusieurs géometries ont été définies (Fig. 2), de manière à prendre en compte le cœur initial, ainsi que son environnement immédiat (roches, fractures). Les configurations critiques ont été étudiées (Fig. 3) en fonction des paramètres physiques pertinents au travers du concept de ligne isocritique (Bentridi, 2011 ; Bentridi et al., 2011).

La criticité de petits réacteurs tels que le RZ9 a pu être expliquée pour la première fois dans le présent travail. Le démarrage, le fonctionnement, et les phases d'extension et d'extinction sont également discutées en terme de variation de la réactivité du système avec l'évolution temporelle des paramètres physiques et géologiques (Fig. 4). Basée sur des simulations statiques, cette étude contribue de façon significative à l'explication de la naissance et de l'évolution du « Phénomène Oklo ». Une de ses principales perspectives est la prise en compte explicite de la dynamique dans ces simulations, afin de vérifier les scénarios proposés. Outre un meilleur suivi de l'évolution des poisons, cela permettrait de prendre en compte explicitement de l'influence de l'évolution de la composition du modérateur, un des points clé notamment pour la fin de vie des cœurs d'Oklo. À la lumière de cette approche, une étude dédiée à l'évolution spatiale vers des structures de grande taille, telle que RZ2, permettrait également de faire le lien avec l'étude initiale de R. Naudet.

1. Introduction

The "Oklo phenomenon", is one of the most amazing and surprising discovery of the 20th century that was made in the domains of geosciences and nuclear physics (Bodu et al., 1972; Naudet, 1991; Neuilly et al., 1972). From 1972 to 1988, fifteen natural nuclear reaction zones (RZ) in the 2 billion years old uranium deposits in Gabon, namely Oklo and Bangombé have been discovered. In order to understand under which geological and physical conditions natural fission reactions could spontaneously start in a natural environment, to determine the effect of such reactions on the surrounding rocks (Pourcelot and Gauthier-Lafaye, 1999) and finally to get information on the behavior of actinides (Bros et al., 1993; Hidaka and Holliger, 1998; Kikuchi et al., 2007) and fission products that were stored for a long period of time in a geological environment, advanced geological, geochemical and physical studies were conducted (AIEA, 1975, 1977; Blanc, 1996; Gauthier-Lafaye et al., 2000; Naudet, 1991; Stille et al., 2003). Due to the state of conservation of those fossilized reactors and their high retention capabilities of actinides, fissiogenic Rare-Earth Elements (REE) and many fission products descendants, that site became a unique example of natural analog of geological repository of high radioactive waste (Gauthier-Lafaye, 1997, 2002; Holliger and Gauthier-Lafaye, 1996).

The geochemical analyses performed on different samples from Oklo reactors (Hidaka and Gauthier-Lafaye, 2000, 2001; Loss et al., 1988; Ruffenach, 1979; Ruffenach et al., 1976) provided anchor points for the neutron physics studies of these nuclear reactors. Even though the occurrence of such natural events has been discussed theoretically in terms of the necessary starting conditions fifteen years before its discovery (Kuroda, 1956), a detailed study about the neutron physics at Oklo was necessary to understand the physicochemical processes that allowed such a phenomenon. These data were exploited during the physical study developed by Naudet (Naudet, 1991), in parallel with the geological and geochemical studies of this phenomenon (Gauthier-Lafaye, 2006). That initial work was performed using calculation tools available at that time. Results obtained by R. Naudet on RZ2, the biggest natural reactor at Oklo, shed light on the physical conditions necessary for such an event to occur, but an explanation for criticality conditions could only be given for RZ1-2 and partially for RZ3-6. He could not extend this explanation to the other natural reactors since they are apparently too small to meet those conditions. R. Naudet could, nevertheless, show that this was not due to the late arrival of uranium in the reactor.

The present work is based on the use of MCNP, a worldwide known neutron transport Monte-Carlo code (X-5 Monte-Carlo Team, 2003). The aim of this study is to bring a physical explanation to the start-up of the small Oklo reactors taking into account their geological and geochemical environments. The main part of this study is

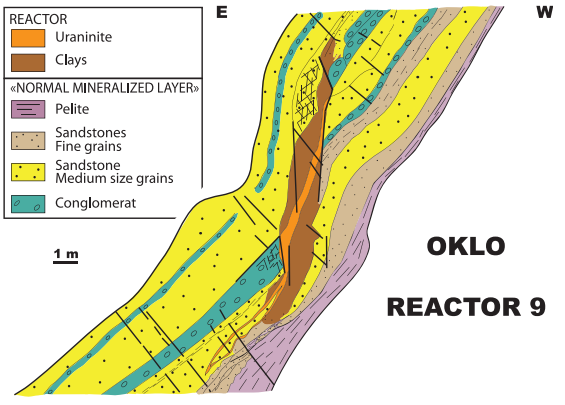

Fig. 1. East-west outcrop of the RZ9 reactor area.

Fig. 1. Coupe est-ouest du réacteur RZ9 et de son environnement.

built on the registered observations about the reaction zone #9 (Fig. 1), which presents with RZ7 and RZ8 some particularities compared to the other reactors. Discovered by Gauthier-Lafaye (at the end of 1978), the RZ9 is located 300 m from the northern sector containing the reactors numbered 1 to 6 (RZ1-6). At present, RZ9 can be viewed as a lens-shaped volume, being a few centimeters thick and exhibiting a 45° dip, which spreads over 7 m length and 12 m width (Gauthier-Lafaye, 1979).

This reactor is situated in a dense fracturation zone that certainly had an important role in the dynamics of the U enrichment process (Gauthier-Lafaye, 1986; Gauthier-Lafaye and Weber, 1988). The present U content in this reactor is relatively low compared to RZ2, most often lower than 30% wgt (considered on dehydrated ore) (Gauthier-Lafaye, 1979; Loss et al., 1988; Naudet, 1991). In contrast to RZ1-6 where desilicification was always observed to be complete, only partial desilicification was registered inside the core itself. In addition, an important abundance of degraded organic matter in form of bitumen was observed in the core, with the weight ratio O/C usually higher than 0.5 (Cortial et al., 1990; Gauthier-Lafaye, 1979).

After describing the geological context, the proposed model will be presented. Out of all the possible parameters that may influence evolution of a natural reactor, the major ones are discussed, as well as their realistic range and influence. Criticality is then discussed through the isocritical line concept. The results of realistic simulations obtained here and their possible time evolution is discussed with scenarios for natural reactor start-up and evolution that could be extended to other situations at Oklo and to other comparable geological cases when they occurred.

This work does not pretend to give an exhaustive and unique description of natural reactors, but it has allowed us to explain for the first time the start-up of those reactors, from smallest to biggest ones and to open new perspectives for the understanding of the Oklo phenomenon.

2. Geological context and reactor implementation process

The Oklo Uranium deposit is located in a 2.1 billion year old sedimentary series which belongs to the Franceville basin in the southeastern part of the Gabon Republic (Bros et al., 1992; Gauthier-Lafaye et al., 1996a; Horie et al., 2005). The stratigraphic column in the basin is identified as a succession of five main formations, labeled from A to E (bottom to top) which are composed of detrital sediments, sandstones and conglomerates (FA) and black shales (FB) at the bottom, and more chemical and volcanoclastic materials at the top (FC to FE) (Gauthier-Lafaye and Weber, 2003; Weber, 1968). The FB black shales with very high organic carbon content (up to 15%) had been the source rocks for hydrocarbons (HC) giving the Franceville basin a very high petroleum potential. It results form the occurrence of oil fields in tectonic traps, which are the oldest-ever discovered (Gauthier-Lafaye et al., 1989, 1996b). The recent discovery of multicellular organisms of large size in the FD black shales suggests in this bassin the occurrence of a very intense and diversified life at that time (El Albani et al., 2010) which is probably responsible for the high petroleum potential of the basin (Mossmann et al., 2005).

The circulation of uranium in the basin was induced by the rise of the atmospheric oxygen content that occurred at that period (Holland, 2009). That allowed a better solubilisation of uranium in diagenetic waters circulating in a network of fractures and connected porosities specific to the sandstone-conglomeratic FA Formation. The zone of interest called "C1" (Gauthier-Lafaye, 1979) is the mineralized layer located at the top of the FA formation. It is made of sandstones and conglomerates and has been mineralized at a depth of 2000 m below the earth's surface (Gauthier-Lafaye and Weber, 1989; Gauthier-Lafaye et al., 1989, 1996b). The uranium ore is mainly made of quartz, clays (chlorite and illite) and organic matter (OM) as solid bitumen. Heavy minerals (zircon, thorite, monazite) represent minor phases mainly located in conglomerate layers. The uranium content averaged in the whole deposit is estimated to 0.4% in weight with some rich zones ranging from 2 to 15% in weight.

Uranium, as uraninite inclusions, is closely associated with organic matter, which played a major reducing role for uranium precipitation. The Oklo deposit is, in fact, the oldest high grade uranium deposit located in a sedimentary sequence and it can be considered as an ancient hydrocarbon field prior being an uranium deposit (Gauthier-Lafaye and Weber, 1981; Gauthier-Lafaye et al., 1989). Realistic rock porosity in such a sandstone rock reservoir may range from 20% to 40% (Nelson and Kibler, 2003). This can clearly be observed in the zones 7 to 9 which show important quantities of degraded OM (Gauthier-Lafaye, 1979; Naudet, 1991). The uranium content in the C1 layer depends on the OM distribution and on the porosity which actually depends on the sandstone grain size and the state of fracturing. Due to the inhomogeneous character of such a natural system, the deposit has very heterogeneous distribution of uranium with places showing up to 15% uranium.

At the present time, the uranium has two main isotopes of masses 235 (0.7204%) and 238 (99.2742%). Both of them are radioactive with very long half-life periods: 7.038×10^8 and 4.468×10^9 years, respectively. The Oklo phenomenon occurred 1.95 b.y ago, when these relative proportions were different than the present ones, due to the difference in radioactive decay period of these isotopes. We can consider the natural abundance of uranium 235 (U5) at that time to be about 3.5%. This value corresponds to the enrichment process for industrial nuclear power plants fuel. This higher concentration in U5, more sensitive to slow neutrons, reduced the uranium concentration needed to reach criticality. However, during the very slow physical and chemical enrichment process of uranium, time is acting against U5 that disappears faster than the isotope 238 (U8). Fortunately, these two antagonistic phenomena led to a time window, when the natural nuclear reactor occurrence became possible. If Nature cared to gather, at Oklo, the conditions for a natural nuclear reactor start-up, the explanation of such a phenomenon is, however, not straightforward.

3. Physical model for the criticality study

3.1. Initial ore of a reaction zone

The realistic model proposed here is based on a study of the neutron mean free paths in geological materials, where it was shown that they are generally larger than the inhomogeneities that are expected inside the Oklo reactor cores (Bentridi, 2011). It combines fast computation and the ability to reproduce observations. The typical ore sample used in this study is defined as a homogeneous initial matrix, which is numerically defined by two main volumes: (i) a solid volume (clay, UO_2, silica); and (ii) a fluid volume fraction located in all forms of porosity even the intrinsic porosity of clays (moderator: water and/or HC) designed by ϕ_T (Bentridi et al., 2011). The barren gangue used here (90% silica + 10% clay) is defined according to observations made in the field and to chemical analyses performed on various samples from the RZ9 core.

During the fission reaction operation and the resulting heating of the environment, there was an important migration of silica out of the reactors and a supply of uranium inside. During this process, the fracture networks played an important role as pathways for the heated fluids. As a result, the uranium concentration in the reactor core increased, due to the mass balance effect and volume compaction of the mineralized layer (Gauthier-Lafaye, 1979; Gauthier-Lafaye et al., 1989; Tchebina-Makosso, 1982).

A convenient volume approach is adopted for the quantification of the two following influential physical parameters:

(1) uranium content V_{UO_2} is defined by the uraninite fraction per volume unit;
(2) the total apparent porosity ϕ_T is defined also as the fraction of free volume that can in principle be filled with any form of fluids. In the present work, only water and organic matter are considered.

In a first approach, the initial ore is supposed to be free of initial neutron poisons since the concentration of heavy elements with large neutron absorption capability may be insignificant in some parts of the considered uranium deposit. We can consider that the major part of the initial natural REE is located in heavy minerals and thus are concentrated essentially in conglomerates while sandstones are almost REE free.

The initial content of neutron poisons such as samarium and gadolinium in the reactors are considered in a second phase of this study. Their neutronic influence is introduced through an equivalent of ^{10}B concentration taking into account macroscopic neutron capture cross-section. This equivalent ^{10}B content (t_{10_B}) expressed in ppm of weight is calculated with respect to the total ore mass including fluids. When compared to measured poisons content to a dehydrated ore weight (Hidaka et al., 1988; Ruffenach et al., 1976; Ruffenach, 1979), it needs to be multiplied by a 1.2 correction factor.

Since Oklo reactors were buried under about 2000 m of rocks, we can consider that they were operated under an hydrostatic pressure of 200 bars and ambient temperature around 150 °C (Oppenshaw et al., 1977). Water density of 0.9232 g.cm^{-3} is deduced from density tables under these pressure and temperature conditions (Lide and Frederiske, 2004). Organic matter density is more difficult to be set down since its precise composition in terms of major elements: H, C and O is not firmly established. However, complementary investigations made for OM at normal temperature and pressure have shown that aliphatic OM has interesting moderation properties (Bentridi, 2011; Bentridi et al., 2011).

3.2. Neutron multiplication factor, reactivity and criticality

A nuclear reactor is operated through the control of the neutron population in the core. Neutrons are multiplied through the fission process and some of them are captured, leading to an effective multiplication factor k_{eff} from one neutron generation to the following, depending on the reactor geometry and state. To start a reactor or to increase its power, a supercritical core is needed ($k_{eff} > 1$), inducing an exponential neutron population time evolution. To get a constant operation, a critical core ($k_{eff} = 1$) is required. Instead of using k_{eff}, it is usual to use deviation to criticality which is expressed by the reactivity ρ, defined as follows:

$$\rho = \frac{(k_{eff} - 1)}{k_{eff}}$$

The total reactivity "ρ_{tot}" of a system is the sum of the positive contributions and negative contributions (anti-reactivity). Reactivity control insures fission chains not to go infinite. That is possible only if the whole reactor is only critical thanks to an additional contribution brought by delayed neutrons. On the contrary, the multiplying system is out of control when it is already critical with only the prompt neutron contribution.

The present work is based on the MCNP code (Harmon et al., 2004; X-5 Monte-Carlo Team, 2003) using probabilistic numerical Monte-Carlo methods, which is capable of, among other things, running criticality calculations for rather complex systems. Once the geometry of the reactor and ore composition are defined, each element or isotope proportion is introduced in the MCNP input file for the different considered volumes. The aim of these series of static computations is to calculate the neutron multiplication factor evolution with respect to influent parameters such as ϕ_T, V_{UO_2} in order to establish minimal size for the starting of natural reactor, 2 billion years ago.

3.3. Geometrical model

The reactors were observed only in their final state. Indeed, the mineralized layer where they started has been altered and deformed during and after reactor operation. Therefore, their initial geometry needs to be established on the basis of field observations and physical parameters imposed by the neutronic laws.

In agreement with observations made in the field, it is stated here that the reactors have the shape of a lens almost parallel to the sedimentary layers. Thus, cylindrical geometry is chosen to represent the studied configuration

volume subject to the present parametrical study. This geometry was also chosen in all previous simulations used to study these reactors (Naudet, 1991; Petrov et al., 2006). Such a simple geometry allows us to consider the geological structure made of a stack of layers by fixing a realist minimum thickness of the initial core and extending reactor ore radius, until we get an initial critical configuration.

From the observations made on RZ9 it is likely to be that the initial thickness was around 70 cm. This value is used as a fixed parameter in the present study. The realistic critical radius R_c is estimated to be slightly less than 1 m.

Two geometrical models are used:

- a reflector less core, being a simple cylinder representing a nude core reactor;
- a core with reflectors represented by the cylindrical assembly shown in Fig. 2. It includes the cylindrical core surrounded in the same plane by an annular reflector, the whole cylinder being covered and sustained by two cylindrical reflectors. All the reflectors are chosen to have the same thickness whose value is taken equal to the core reactor due to the neutron mean free path in this system.

The first model, is not realistic, but gives an upper limit to the critical sizes. In the second model, the effect of the reactor vicinity on neutron population is considered. The core is surrounded by reflectors made of a relatively uranium-poor matrix, corresponding to ore with low total porosity (Gauthier-Lafaye, 1979; Naudet, 1991). One can note such reflectors return to the core neutrons produced by fission inside the reflectors and contribute to neutron moderation, which leads to reactivity increase, by scattering back some neutrons outgoing from the core region.

Given an estimated uranium quantity and with suitable choice of thickness "*e*" with respect to neutron free path, a critical radius R_c can be determined through numerical parametric computations, by varying porosity within realistic geological range.

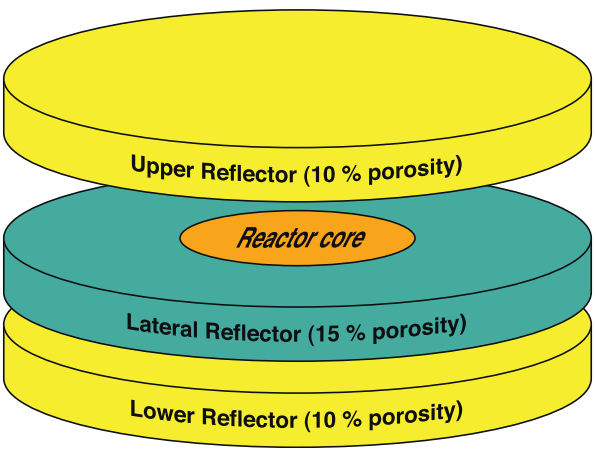

Fig. 2. Representation of reactor model with reflectors used in this work.

Fig. 2. Vue éclatée de la géométrie utilisée pour la modélisation des réacteurs d'Oklo.

4. Physical and geological relevant parameters

From a neutron physics point of view, and considering the geological context of Oklo, at least four parameters can be listed, having a direct impact on the criticality of such a nuclear system:

- the uranium content is defined here by a uraninite volume fraction V_{UO_2} taken equal to 5%. It corresponds to a limit, around 20% in weight in terms of uranium content (evaluated according to total mass for one gram of dehydrated ore) under which no reaction zone were observed;
- the total porosity noted ϕ_T defines a free volume that could be occupied by any fluid present in rocks. Calculations were performed with a porosity range — from 20 to 40% of rocks volume — typical for rocks reservoirs in hydrocarbons deposit. For calculation convenience the fluid considered here is water with density equal to 0.9232 g.cm^{-3} at the P and T conditions considered;
- the initial reactor thickness "*e*" is the major geometrical parameter. It is fixed at 70 cm according to a first set of simulations based on a homogenous and spherical mixture of uraninite and water, without reflectors. This initial approach allowed getting a critical solution for a sphere radius equal to 35 cm with a uranium content of 20% (V_{UO_2} = 5%) (Bentridi, 2011). By considering now a cylindrical geometry, the sphere diameter is replaced by a thickness "*e*" and a cylinder radius "*R*", so the system could be extended only in the radial direction. This thickness is in agreement with the thickness of the conglomerate that frames the reactor RZ9 (cf. Fig. 1);
- the critical cylinder radius noted R_c is considered as a principal parameter of this study and as a variable at the same time. Simulations are performed in order to determine the critical radius evolution by varying one of the former parameters, the others being fixed.

In the last step of this study and in order to be as close as possible to the real situation, we have taken into account in the model the occurrence of neutron poisons, with various concentrations, inside the core and reflectors. Sudden geometry variations were also considered through the modeling of reactivated fractures inside the reactor core (Bentridi, 2011).

It is noteworthy that the initial poison content, expressed in ppm boron-10 equivalent, cannot be defined precisely because of the high inhomogeneity in such a natural system. In this study, the mean value of initial poison content, suitable to the starting of the reactor, has been defined, according to the criticality occurrence.

5. Critical solutions and isocritical lines

To start a natural nuclear reactor, the uraninite concentration should be high enough to compensate initial neutron absorbent elements in order to get a slightly positive reactivity balance. Once diverged, a slight decrease in the total reactivity could easily bring the system to converge again, whereas an increase will

speed-up the divergence. The reactivity balance gives a powerful tool for the reactor condition evaluation, and for tracking in detail the evolution of this dynamical system.

The human-control of a reactor is based on reactivity control with restricting operation procedures to stay in safe operating mode. Since both the neutron flux and the produced power of a diverged reactor follow an exponential time evolution, a strong increase of reactivity could imply major effects on the core. In an extreme case, when the heating exceeds the cooling ability, core content may fuse, generating what is called "corium". The corresponding temperature conditions will drive the moderator out of the active core and that will lead to the definitive shutdown of the reactor. Such a scenario does not match the geological observations.

The most likely scenario is that, after divergence, the power of natural reactors gradually increases up to a long-term steady operating point where it will be stabilized due to thermal effects. The uranium is gradually depleted until the definitive shutdown, which occurred after a long operating period estimated at around 150,000 years (Hidaka and Holliger, 1998; Loss et al., 1988).

Criticality in the case of a finite media is given by the effective multiplication factor k_{eff}. The simulations performed have estimated that factor for the model configuration considered. In the present parametric study, k_{eff} was searched within a satisfactory confidence interval of standard deviation smaller than 0.001, according to a calculation protocol recommendation (Harmon et al., 2004). Even if the exact values obtained can be discussed, they are representative of a realistic and geologically plausible situation.

Compared to time-dependent simulations, the results of MCNP computations on criticality are "snapshots" of an evolving situation. Thus, these results are representative only for a time interval shorter than characteristic time of the system evolution. Given the operating time scale of natural reactors, those conditions could be considered relatively quiet.

The simulations applied to the RZ9 model led to determine a succession of critical situations, by varying individual parameters (variables), while keeping the others constant (parameters). In order to have a clear view in this multi parametric study, the "isocritical line" concept has been defined (Bentridi, 2011; Bentridi et al., 2011). Such a line connects, for a defined set of parameters, the critical solutions in the variable configuration. The corresponding (isocritical line) can be defined in formal way by:

$$k_{\text{eff}}|_{parameters}(variables) = 1$$

For a given configuration with a minimal uranium content, these lines show a set of solutions. They were used to enlighten the possible critical radius as a function of either various realistic uranium concentrations or porosity intervals (Fig. 3), the thickness being fixed at some minimal value below which no critical solution exists. It is important to note that the isocritical lines separate the representation space between possible critical situations: above the line are zones able to start the nuclear chain reaction and below the sterile domains for the model

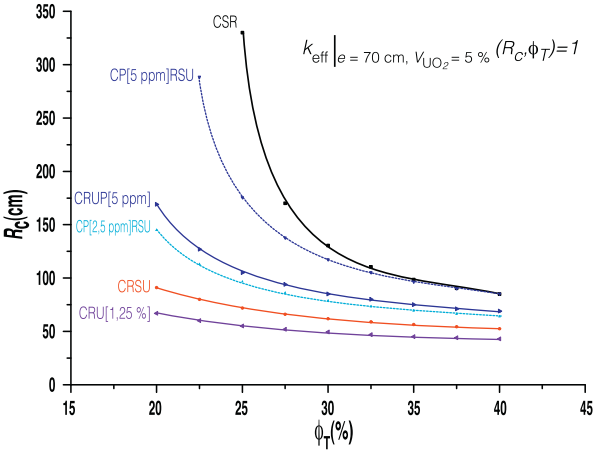

Fig. 3. Isocritical lines for several RZ9 Fresh core studied configurations.

Fig. 3. Lignes isocritiques pour différentes configurations étudiées du réacteur RZ9.

considered. For example, the zone situated above the isocritical line:

$$k_{\text{eff}}|_{e=70\ \text{cm},V_{UO_2}=5\%}(R_c,\ \phi_T) = 1$$

include all possible reactors that could be operated with a uraninite fraction volume V_{UO_2} = 5% and a starting thickness "e = 70 cm". Each geometrical hypothesis led to a curve (Fig. 3). These curves are studied within a porosity interval ranging between 20 and 40%, with a simulation interval $\Delta\phi_T$ = 2.5%.

6. Realistic simulations results

Following the first numerical results and according to the geological data from reactor RZ9, the proposed model has evolved progressively to a more and more realistic situation. Fig. 3 summarizes these results. The first and simplest model used here for a poison-free nude cylindrical core configuration gives the "CSR" (Core without Reflectors) curve. It results that the critical radius ranges between 320 and 100 cm over the whole porosity interval (ϕ_T ranging from 20 to 40%). With such a geometry, configurations located above that isocritical line could start sustained fission reactions, whatever the core environment.

In a second approach, the close environment of the core is also considered even though it may not constitute a high-grade quality neutron reflector. First, we consider the case of cores with barren reflectors (without uranium) represented here by the isocritical line "CRSU". The contribution of such natural reflectors reduces significantly the critical size needed for the starting of the reaction zone. These reflectors were sized according to a prior study showing that there is no improvement of neutron reflection for volumes with thickness greater than 60 cm (Bentridi, 2011). The critical radius represented by the isocritical line "CRSU" varies from 90 to 50 cm in the porosity range, reducing significantly the barren domain.

For situations even closer to geological reality, two steps are still necessary. First, the initial poison content present at reactor start-up is considered in the

CP[Xppm]RSU (Core with Poisons and Reflectors without Uranium) configuration. Secondly, the influence of uranium presence inside reflectors, even with low content, is considered in the CRU[1.25%] (Core with Reflectors with 1.25% of Uranium) configuration. In Fig. 3, both curves "CP[2.5 ppm]RSU" and "CP[5 ppm]RSU" show a strong sensitivity to initial poison content, estimated with ^{10}B equivalence. They stay in the space limited by two lines CSR and CRSU. Adding 1.25% of uranium in reflectors without poisons leads to the poison-free isocritical line CRU[1.25%] situated slightly under the CRSU one.

Addition of 1.25% of uranium in those reflectors with 5 ppm of poisons leads to the CRUP[5 ppm] line which is sharply shifted down from the CP[5 ppm]RSU one. This shows a stronger sensitivity of the reactivity when uranium is added in a poisoned reflector than in a poison-free one. Indeed, in the case given by CRUP[5 ppm] curve, the small uranium presence in reflectors comes to partially counteract the effect of the poisons in the core.

Finally, the occurrence of uranium and poisons inside the core or the reflectors, according to the nature of sandstone, seems to be the most realistic situation (CRUP[5 ppm]). We have shown here that there is strong sensitivity to the presence of poisons depending to their location. The realistic space of solutions for Oklo site is included between the isocritical lines CRUP[5 ppm] and CRU[1.25%] devoid of initial poisons. These curves give reasonable intervals for critical radius between 160 and 70 cm for CRUP[5 ppm] and between 70 and 50 cm for CRU[1.25%]. These results show that in the geological context of the Oklo situation, the occurrence of fission reaction zones can be explained, even for initially small cores. The different isocritical lines seem to be in a physically and geologically realistic critical solution space.

This work can be considered as a major complement to the previous synthesis made by R. Naudet (Naudet, 1991), who could give an explanation for RZ2, the biggest and uranium-rich reactor, but could not explain the smaller reaction zones, such as RZ9. According to the present study two hypotheses may be proposed for the start-up of the largest reactor RZ2. It may have started, like the other small reactors, on a uranium fluctuation of small size located in a zone convenient for an important extension, or it started in a very big, favorable zone. In the latter case, less enrichment in U5 would have been necessary for criticality and this reactor could have been ignited earlier than the other Oklo reactors. In the case of reactor 9, once the start-up was insured, the evolution of its geometry and duration are much less important than RZ2, according to the geochemical data. This could be due to its reduced initial geometry situated in a closed environment presenting less reserve of nuclear reactivity, which means less possibility of expansion.

7. Discussion

In order to get a configuration leading to the occurrence of a critical reactor in the mineralized layer C1, the total reactivity provided by the fuel and modulated by the moderator, must compensate the anti-reactivity due to the presence of initial poisons, which are more or less important, as the ore is mainly made of sandstone or conglomerate. The influence of initial core composition on the starting reactivity equilibrum gives rise to differences in critical radius R_c from one configuration to another, as shown in the critical line drawings (Fig. 3). Furthermore, the balance of these conditions and its evolution appear even clearly on a reactivity balance scheme such as that presented in Fig. 4. In this figure, two evolution cases are considered:

- progressive evolution without fracture, as shown in Fig. 4a;
- induced evolution by fracture reactivation, as shown in Fig. 4b.

During a first phase of their genesis, the reactors will approach criticality when the uranium concentration progressively compensates the ore poisoning due to the initial REE presence that fluctuates depending on zone considered. Due to the geological timescale, the reactor will reach criticality very slowly. A non-linear timescale is used on Fig. 4 in order to get a better insight of the evolution of the balance between the different contributions to reactivity. It slowly evolves in a first divergent phase, called here the "thermal phase".

An increasing of power and of neutrons flux will be registered during this thermal phase, implying a progressive heating of the reactor core. As soon as the power generated by the core induces a direct temperature dependence on neutron flux, negative feedback from thermal effects will start to impact on the reactor equilibrium. This will drive the total reactivity to zero bringing the reactor back to a critical state.

Those thermal effects result from two combined phenomena: the temperature dependence of the moderation due to the volume dilatation, and the temperature dependence of neutrons capture by the ^{238}U resonances which is directly proportional to the temperature variation. The last effect is usually known as "the Doppler" effect in reactor physics, the power level where this effect starts being called the "Doppler threshold". In some specific situations of industrial reactors the moderator contribution to the thermal effect may be slightly positive. Due to the composition of the moderator and the presence of solid initial poisons, that contribution was probably always negative in Oklo.

Once the reactor passed this threshold, leading to a stable operation, its power level is naturally adjusted to the heat dissipation ability of its environment. Such an operation is very similar to industrial reactors driven by the same effect, where the nuclear power will be controlled by the quantity of steam requested by the turbines in order to follow electrical power demand of the network.

The fission produces more than 200 fission fragments, a few of which show a very important neutron capture cross-section, as it is the case for the two major ones ^{135}Xe ($T_{1/2} = 9.2$ h) and ^{149}Sm ($T_{1/2} = 2 \times 10^5$ y). When the neutrons flux becomes stable, different effects which are directly related, such as the fission fragment production rate will also be stabilized. Thus, the concentration of fissiogenic poisons having short half-lives compared to

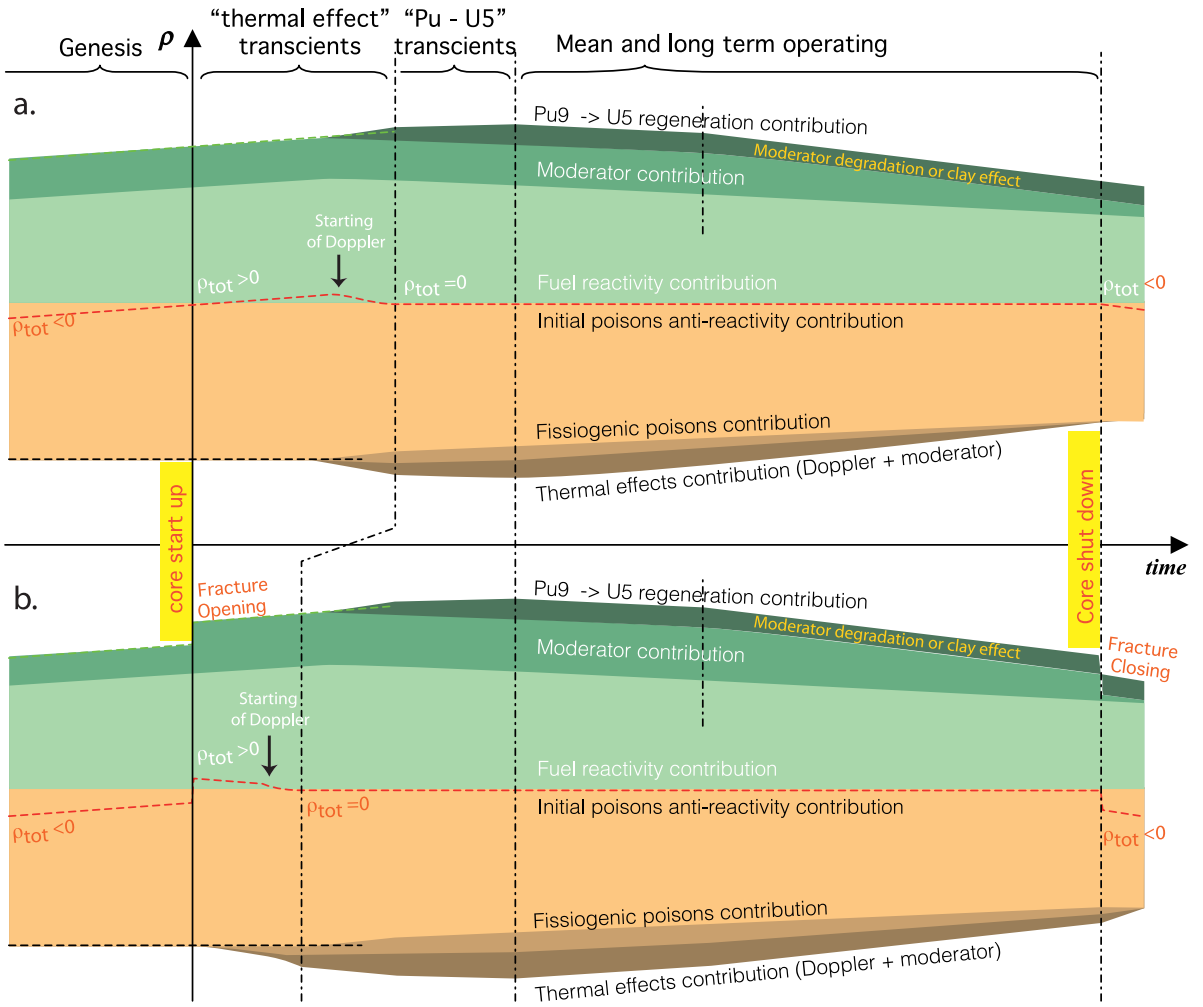

Fig. 4. Illustration of time evolution of the reactivity content with the case of (a) progressive start-up and (b) start-up induced by a fracturation.

Fig. 4. Illustration de l'évolution temporelle du bilan de réactivité des réacteurs d'Oklo avec les cas (a) du démarrage progressif et (b) du démarrage induit par l'apparition d'une fracture.

the evolution time scale of Oklo reactors will directly follow the neutron flux evolution after a transient period. As a result, the corresponding anti-reactivity is following the neutron flux (Fig. 4).

Other effects are more progressive, like the disappearing of initial poisons and the evolution of uranium concentration. So, while uranium is fissioning, its concentration process continues to bring in uranium with the incoming charged solutions (Gauthier-Lafaye, 1979). The mean uranium content of the core is a function of these two mechanisms and will evolve according to their relative importance. Besides, the slow evolution of the core geometry can also induce some variations of uranium concentration.

At the end of that thermal phase, we can consider the regeneration of ^{235}U by ^{239}Pu α-decay ($T_{1/2} = 24\ 100$ y), which is indirectly produced by neutron capture on ^{238}U. This process progressively evolves until it becomes stable and able to partially compensate the ^{235}U consumption. Indeed, considering the half-life decays implied in such a process, the regeneration process of ^{235}U needs at least a few thousand years to have a perceptible contribution and finally to be stabilized.

At the end of that second phase called "Pu-^{235}U conversion", all the principal contributors to criticality will be balanced. After that, the reactor evolves within a final phase referred to as "average and long term", which lasts hundred of thousands of years. During that phase, the nuclear fuel will decrease proportionally to the power level reached by the reactor, the latter depending on the start-up conditions. Logically, these also affect the reactivity effects, since these are essentially due to the neutron flux generated by ^{235}U fission. Thus, poisons decrease in parallel to uranium consumption.

If one admits that the moderator contribution is not affected, one can expect this nuclear system to continue operating until the system reaches the poison-less critical mass.

This is precisely the crucial moment in the reactor's life, where two shutdown scenarios could be considered. The first one is based on the observations of the geological context and RZ's morphology. The reactors and their close environment are strongly altered by physicochemical processes responsible for a progressive dissolution of silica leading to a progressive compaction of the reactor core. The latter becomes more and more concentrated in uranium and contains less and less efficient moderator. From a neutronics viewpoint, this represents two antagonist effects.

Furthermore, those processes lead to a formation of a clayey envelope more and more impervious to fluid circulation. This may condemn the core to go subcritical (converge) as soon as the moderator contribution no longer permits sustaining the chain reaction, and then the Doppler becomes unable to compensate this effect.

The second possible scenario, which can shutdown these reactors, may occur if the core temperature induces a progressive but massive transformation of OM, that will migrate to peripheral colder zones where it solidifies. The core, being gradually deprived of its moderator, will be driven to shutdown or will continue to operate at significantly weaker power.

The scheme of the whole scenario discussed above is shown on Fig. 4a with an arbitrary nonlinear time scale, used only to enlighten the different phases of evolution. Fig. 4b represents a variant of the former scenario by taking into account the possible influence of the fractures that can be observed on Fig. 1. A special interest goes here to their possible dynamical influences (opening and closing of a fracture) on the reactors' operation. That variant corresponds to some initial situations which could not start up with the first scenario. This is particularly the case for reactors with small sizes or with important poison concentrations. Such systems need necessarily a higher input of reactivity in order to diverge in the appropriate space-time window of natural reactors' start-up. For that scenario, it is imperative that a geomechanical incident occurs. Thus, this could directly be related to a presence of one or more fractures inside the core which lead to quasi-immediate and significant reactivity increase. As we could expect, the present work indicates that this contribution is especially favorable to configurations with low porosity (Bentridi, 2011). For reactors which could not startup without such a geomechanical incident, that event leads those systems to operate with a low reactivity and to evolve in the same way as in the first scenario.

On the other hand, if such an event occurs for a reactor quasi-critical or already started, it will suddenly generate a large positive reactivity inducing faster evolution of flux and power as shown in Fig. 4b.

Besides the impact on reactivity brought by that fracture, we must consider the fact that, from a thermo-hydraulics viewpoint, those fractures constitute a pipe for the circulation of fluids favoring the heat dissipation by convection, the departure of silica outside the reactor and the entry of uranium inside. This may explain the structure of the east part of RZ9, where the fission chain reaction could extend along two fractures showing important uranium enrichments that can be observed at the bottom left part of Fig. 1. Unlike the other reactors mainly balanced by thermal conduction, these systems are deeply influenced by convective phenomenon. The heat quantity extracted is more important in such a reactor, leading to a higher stable operating point and a probable significant reactor lifetime reduction.

The operation of such cores at a highest temperature will induce some geomechanical modifications inside these structures. This is likely to induce or accelerate the closing of the fracture which is responsible for the reactor start-up. As is illustrated on Fig. 4b, the closing of the fracture leads the reactor to shutdown immediately, and probably in definitive way. However, it could be envisaged that such structures did not experience fractures closing and in this way, the fission chain reaction stopped, as is shown in scenario one.

8. Conclusion and perspectives

Through a realistic modeling based on Monte-Carlo calculations using the code MCNP applied to the geological conditions of the natural reactors of Oklo, the present study has examined the initial conditions of their start-up. The "isocritical line" was introduced to represent the critical configurations in a suitable parametrical space. The criticality computations performed within this study have been dedicated first to the reaction zone RZ9. They have allowed the identification of the main parameters influent on the criticality and their influence on different systems.

Since those systems had undergone an expected compaction due to the lithostatic pressures to which they were subject, especially after considerable mass departure of silica due to thermo-hydraulic processes induced by the natural reactors in operation, it is now relatively difficult to determine the initial conditions from the present-day observations. The start-up of the reactions in the core may have taken place between two limits: one is related to favorable zones with very large dimensions (as is the case for RZ2) and the other case is related to a little favorable zone with smaller extension, which experienced a relatively low operating rate and a shorter period of operating. Until now start-up of these latter reactors has not yet been explained.

The present study is applied to the geological context of the Oklo site, taking into account the realistic variations of the physical and geological parameters. It allowed for the first time to shed light on the possible start-up conditions for the small sized natural reactors. More generally, any considered configuration between the ideal isocritical line corresponding to a poison-less core surrounded with enriched reflectors with uranium CRU[1.25%] and the less favorable isocritical line for reactors involving 5 ppm of ^{10}B equivalent with barren reflectors CP[5 ppm]RSU could be critical under Oklo conditions with realistic critical radius. Once start-up of those natural reactors is explained, this study described the probable neutronic evolutions of those reactors and their spatial evolutions within stratigraphic layers. By considering all influent parameters and their probable evolution, it was possible to establish descriptive scenarios of various life phases of those reactors from their divergence until their final shutdown.

The present work has also underlined the influence of geological and physical phenomena such as fracture opening on the dynamics of start-up and evolution of a reactor core. The end of reactor life has been also discussed, in particular through the influence of the clay formation accelerated by alterations of the ore around reactor cores, making the neutrons moderation more and more difficult, even impossible when the maximal impermeability was reached.

Besides its importance in the last step of the reactor life, the clayey envelope formed turned out to be very efficient for the preservation of those fossilized reactors and especially for the very high capability of fission product retention, usually highly radioactive. Under many aspects, those reactors could be considered as natural analogues for a radioactive waste repository in a geological environment, which has been frequently argued in several publications and communications dedicated to this phenomenon (Gauthier-Lafaye, 1997; Nagy et al., 1992); reactor physics has much to learn from this exceptional phenomenon.

This study is far from being finished even if it has already significantly contributed to the explanation of the start-up and evolution of the "Oklo" phenomenon. The static simulations performed give snapshots of the reactor situation in a chosen configuration. Thus a major perspective of this study would be to run fuel depletion simulations in order to monitor reactor evolution during the time of their operation, and follow realistic scenarios.

This would allow us to take into account more accurately the time evolution of neutron poison effects. The moderator composition and its distribution during reactor operation is also a key point to be studied in detail, especially for a better explanation of the end of life of the Oklo natural reactors.

In addition to this description it would be also very interesting to study spatial evolution and structural extension of big size reactors like RZ2 with the same approach. Extremes cases from the point of view of the initial poison contents may be very interesting to study. This method could be also applied to reaction zones presenting some ^{238}U fission traces (Hidaka and Holliger, 1998) in order to determine their starting conditions.

Acknowledgements

Authors would like to address a special thanks to A. Clément, F. Lehmann for technical support, G. Schäfer for the discussions around density tables and O. Dorvaux, J. Piot, D. Rouvel, J. Rubert and P. Stille for careful reading and fruitful discussions. Authors would like to thank F. Chabaux, A. Younes and all the GICE team of LHyGeS for fruitful discussions. Authors would like to address a special acknowledgement to C. Jammes and F. Weber for wise and helpful comments reviewing this article.

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