

**DU COMBUSTIBLE NUCLÉAIRE AUX DÉCHETS :
RECHERCHES ACTUELLES**

FROM NUCLEAR FUELS TO WASTE: CURRENT RESEARCH

2 billion year old natural analogs for nuclear waste disposal: the natural nuclear fission reactors in Gabon (Africa)

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Abstract Two billion years ago, the increase of oxygen in atmosphere and the high $^{235}\text{U}/^{238}\text{U}$ uranium ratio ($> 3\%$) made possible the occurrence of natural nuclear reactors on Earth. These reactors are considered to be a good natural analogue for nuclear waste disposal. Their preservation during such a long period of time is mainly due to the geological stability of the site, the occurrence of clays surrounding the reactors and acting as an impermeable shield, and the occurrence of organic matter that maintained the environment in reducing conditions, favourable for the stability of uraninite. Hydrogeochemical studies and modelling have shown the complexity of the geochemical system at Oklo and Bangombé (Gabon) and the lack of precise data about uranium and fission products retention and migration mechanisms in geological environments. *To cite this article: F. Gauthier-Lafaye, C. R. Physique 3 (2002) 839–849.*

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natural nuclear reactors / natural analogue / Oklo / Bangombé / uranium / precambrian

Des analogues naturels de sites de stockage de déchets nucléaires vieux de 2 milliards d'années : les réacteurs de fission nucléaire naturels du Gabon (Afrique)

Résumé Il y a 2 milliard d'années, les réacteurs nucléaires naturels du Gabon ont fonctionné grâce à l'augmentation de l'oxygène dans l'atmosphère et à un rapport $^{235}\text{U}/^{238}\text{U}$ de l'uranium élevé ($> 3\%$) à cette époque. Ces réacteurs sont considérés comme de bon analogues naturels de site de stockage de déchets nucléaires. Leur préservation est due en grande partie à la stabilité géologique du site, la présence d'argiles entourant les réacteurs et constituant un bouclier imperméable et à la présence de matières organiques qui a maintenu un milieu réducteur favorable à la stabilité de l'uraninite. Les études hydrogéochimiques et les modélisations géochimiques mettent en évidence la complexité des systèmes géochimiques à Oklo et Bangombé et le manque de données précises sur les mécanismes de rétention

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et de migration de U et des produits de fission dans des environnements géologiques.

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réacteurs nucléaires naturels / analogues naturels / Oklo / Bangombé / uranium / précambrien

1. Introduction

Natural nuclear fission reactors (hereafter just called reactors) are unique places in the world where fission reactions took place spontaneously 2000 million years ago and have been sustained for quite a long time (100 000 to 500 000 years) in a natural environment. The reactors now contain high-grade uranium ores with a high content of fission products and their end members' decay. This makes these geological objects similar to places where spent nuclear fuels have been stored in a geological environment over a very long period of time i.e. 2.0 b.y.

Since 1972, the year when the first reactor was discovered in the uranium deposit of Oklo [1], many programs have been conducted in order to understand the processes by which fission reactions could occur and the effects of these nuclear reactions on the surrounding rocks. The reactors are considered as good natural analogues for nuclear waste disposal and research is now mainly focused on the behaviour of actinides and fission products in a geological environment. This paper tries to briefly synthesise our knowledge of the geological history of the reactors and their hosted uranium deposits and to give an overview of what we have learned on the geological and geochemical conditions that led to the preservation and the conservation of the reactors.

2. Geological background

2.1. Uranium deposits

The Franceville basin contained five uranium deposits (Oklo, Mounana, Boyindzi, Mikouloungou and Bangombé) which have been now mined out with the exception of the Bangombé deposit that contains the last of the natural nuclear fission reactors (Fig. 1). All together, these deposits contained 28 000 ton of uranium that was mined from 1961 to 1999.

The uranium deposits of the Franceville basin are the oldest known high-grade uranium deposits in sedimentary rocks that were formed by dissolution–precipitation processes [2]. Franceville uranium deposits are younger than those of Witwatersrand, South Africa (2.7–2.9 GA Ma) and Blind River–Elliot

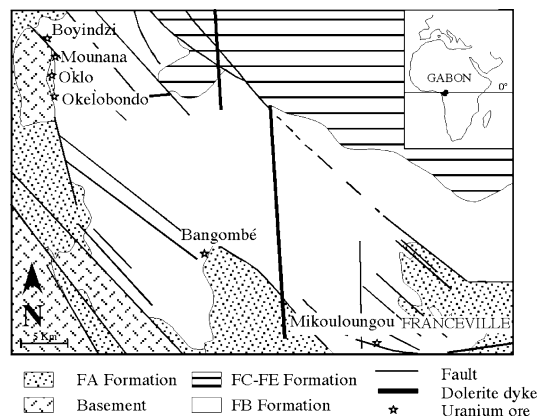
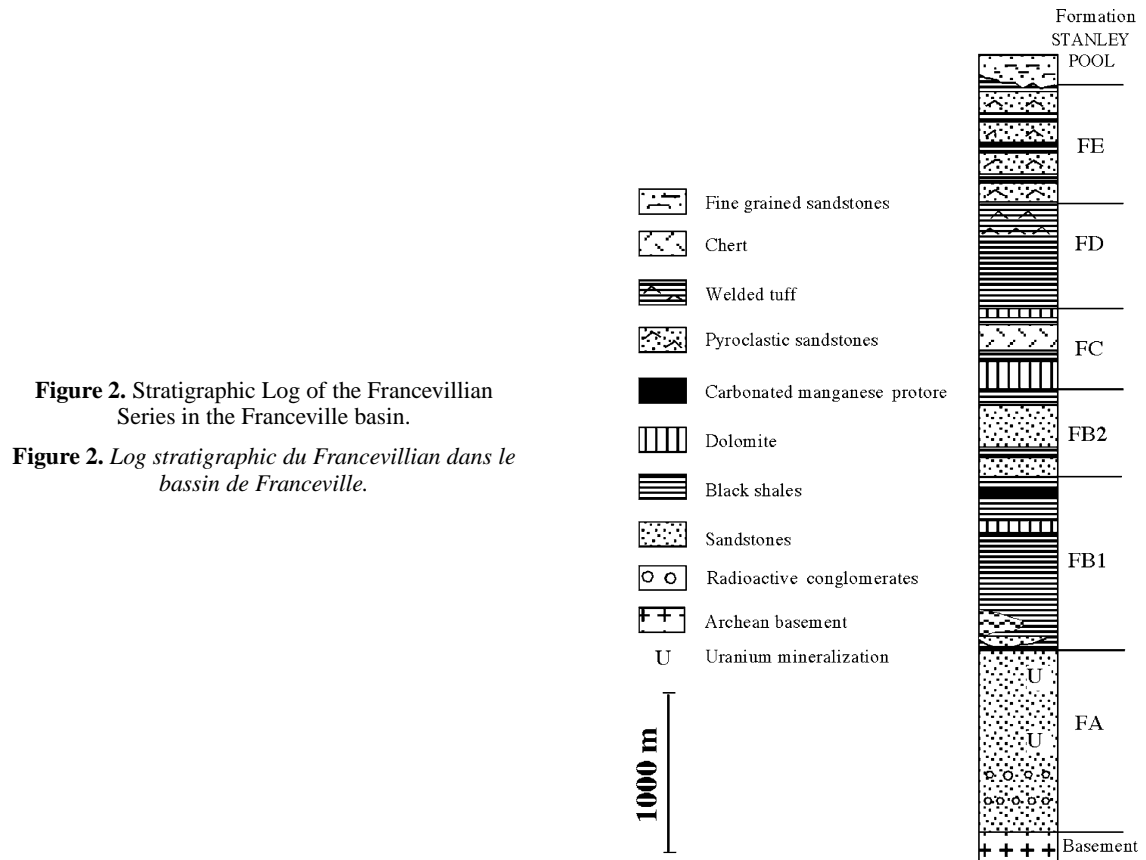


Figure 1. Schematic geological map of the Franceville basin showing the location of the uranium ore deposits.

Figure 1. Carte géologique schématique du bassin de Franceville montrant la situation des gisements d'uranium.



Lake, Canada (2.4 Ga) where most of the uranium has been concentrated by sedimentary (mechanical) processes [3]. The high grade of uranium in the Franceville deposits implies uranium oxidation–reduction processes during their formation and therefore a high oxygen concentration in the contemporaneous hydrosphere and atmosphere. The age of the uranium deposit is 1950 ± 40 Ma as determined using the classical U–Pb method on uraninites [4] and by dating the fission reactions themselves using the fissiogenic REE (Rare Earth elements) and uranium system [5–7].

Uranium deposits are located at the top of a 400 to 1000 thick FA formation mainly made of sandstones and conglomerates (Fig. 2). This formation is underlain by the FB formation made of black shales with organic carbon content ranging between 1 to 15% and few carbonate (dolomite) layers. In the central part of the basin the FB formation is 1000 meters thick [8].

Most of the uranium is of the form of uraninite and is closely associated with migrated hydrocarbons that fill the secondary porosity of the sandstone [9]. Uranium mineralisation occurred during diagenesis, when the mineralised FA formation was 3000 m deep and when oxidised uranium bearing fluids met reduced fluids associated with hydrocarbons in tectonic traps [9]. Uranium bearing fluids are hot (100–170 °C), saline waters with concentrations of Ca up to 30% eq. wt% and Na up to 30 eq. wt.% [10,11]. The sources of uranium are believed to be detrital uranium bearing minerals such as monazite and thorite which are concentrated in several 100 m thick conglomerates in the lower FA formation [2,8].

2.2. Uniqueness of the reactors of the Franceville basin

The conditions for the occurrence of nuclear fission reactions in a geological system have been discussed by Naudet [7]. Fission reactions occur spontaneously in ^{238}U resulting in the production of fast neutrons. If these neutrons are slowed down and thermalised, they may induce fission in ^{235}U or ^{239}Pu . The nuclear reaction can then be sustained. This can happen in a geological system if three conditions are met:

1. The uranium ore must have high uranium content.
2. The fast neutrons produced by the spontaneous fission of ^{238}U must be thermalised by migration through water or graphite.

Naudet [7] has shown that water should have been the main moderator for neutrons in the Oklo reactors. Water is present in the porosity of the sandstones and in clays that were important mineralogical phases in the reactors. Naudet suggests that carbon may also have played an important role in some organic matter rich reactors.

3. The ^{235}U content of the uranium should be high.

At Oklo this condition was met because the radioactive decay constant for ^{235}U ($99.8485 \cdot 10^{-10}$) is smaller than that for ^{238}U ($1.55125 \cdot 10^{-10}$). Fig. 3 shows the variation of the $^{235}\text{U}/^{238}\text{U}$ ratio versus time. 2.0 b.y. ago the ratio was similar to that of fuel in PWR (Pressured Water Reactor) nuclear plants. There is little chance of finding natural fission reactors younger than 1.9 Ma because the amount of fissile ^{235}U was then too small to sustain the fission reaction. At ages > 2.0 Ga, the $^{235}\text{U}/^{238}\text{U}$ was sufficient to sustain fission in natural reactors and even very high at ages > 2.5 Ga. This is the main argument for predicting that fission reactors should have been very common prior to 2.0 b.y. However, no traces of nuclear fission reactions have been found in pre-2.0 Ga uranium deposits such as those in the Witwatersrand or at Elliot lake. This means that other conditions must be important as well. Of course, the concentration of uranium is of particular importance and this may be achieved only in certain geochemical conditions.

Fulfillment of conditions 1 and 3 allows a uranium deposit to reach the critical mass required to sustain a fission reaction. If one takes into account the various physical (porosity) and chemical (content of various minerals and neutron poisons such as REE, B, and V) properties of the Oklo sandstones, Naudet [7] has shown that a sustained fission reaction would have occurred at Oklo where the uranium content was 10% in one cubic meter. Such high uranium concentrations in a sedimentary rock imply that the uranium had been subjected to one or more dissolution–precipitation episodes and implies that enough oxygen was available in water to allow oxidation–reduction reactions of U to occur. Such a condition was not fulfilled before 2.0 Ga, the oxygen content of the atmosphere being low. Holland [12] has suggested that the P_{O_2} level

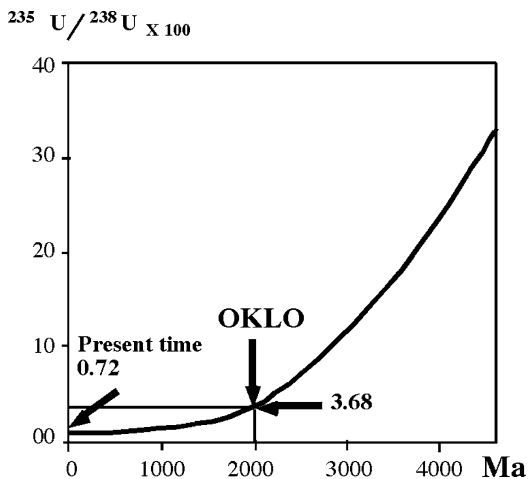


Figure 3. Evolution of the $^{235}\text{U}/^{238}\text{U}$ ration versus time.

Figure 3. Évolution du rapport $^{235}\text{U}/^{238}\text{U}$ en fonction du temps.

in the atmosphere increased rapidly from < 1% PAL to > 15% PAL during the early Proterozoic (2.2–< 1.9 Ga).

Therefore, it appears that 2.0 Ga is a crucial age that allows, at the same time, a high $^{235}\text{U}/^{238}\text{U}$ ratio and, for the first time in the Earth's history, a high uranium content in the rocks.

3. Description of the reactors

Fifteen natural nuclear fission reactors were discovered in two deposits of the Franceville basin. Fourteen are located in the Oklo deposit and one in the small deposit of Bangombé which is 30 km from Oklo. The reactors of Oklo have been totally or partially mined out. Because the open pit and the underground mines have been flooded, the remaining parts of the reactors are no longer accessible. The reactor of Bangombé, which is the smallest, has been preserved, together with the overall uranium deposit for further scientific study on the behavior of fission products and actinides in a geological environment. The Bangombé reactor is located very close to the surface (12 m for Bangombé and 100 to 260 m for the Oklo–Okelonbondo reactors) and is therefore much more affected by weathering than the other one. Extensive geological, mineralogical and geochemical descriptions of the reactors are given by [8,13,14] and in a European report that synthesised the results obtained during the last 3-year European program on the behavior of fission products in the water–rock systems of the various reactors [15].

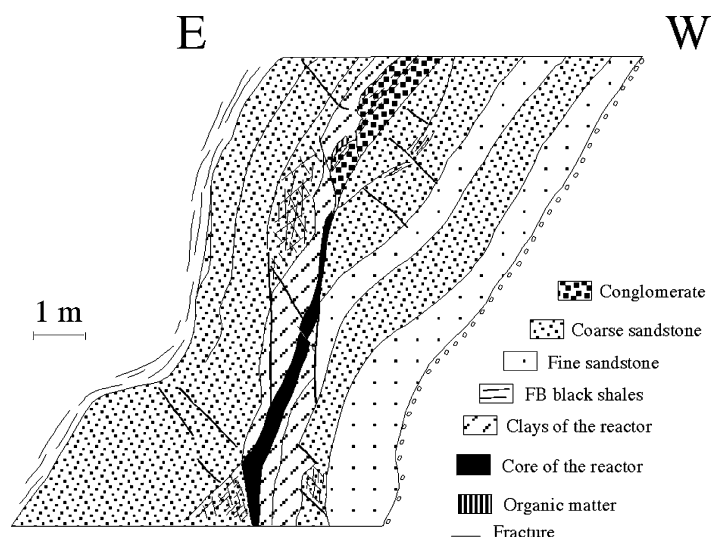
A typical reactor is shown in Fig. 4 (reactor 9). The size of the reactors is quite variable. The biggest reactor (reactor 2) is a 12 m long, 18 m deep and 20 to 50 cm thick lens. At Bangombé the reactor is only 5 m long, 1 m wide and a few centimeters thick. Recent weathering has dissolved a large part of this reactor [15].

The core of the reactors consists of a 5 to 20 cm thick layer of uraninite embedded in clays (illite and chlorites). The uranium content of the core ranges between 40 and 60%. Accessory minerals are mainly sulfides (pyrite and galena), hematite and phosphates (mainly hydroxyapatite). The core of the reactors is usually located in a clayey layer. The newly crystallised clay minerals reflect the thermal gradient during the operation of the reactors [16].

Fission reactions started in high-grade uranium ore, when the uranium content of the sandstone reached 10%. When fission reactions started, the temperature in the core of the reactor increased and a convective hydrothermal system around the reactor started. The temperature in the core reached 400 °C and decreased rapidly at the edges (geothermal gradient around $100\text{ °C}\cdot\text{m}^{-1}$). Heat was transferred largely

Figure 4. Cross section of the reactor 9, in the Oklo deposit. From Gauthier-Lafaye et al. [14].

Figure 4. Coupe du réacteur 9 d'Oklo.



by conduction [17]. The hydrothermal circulations led to the dissolution and migration of 80% of the silica from the mineralised sandstone. This decreased the volume of the mineralised sandstone and increased the uranium content of the residual layer which then consisted mainly of clays and uranium dioxide. Such processes allow the formation of a reactor core containing 40 to 60% uranium.

In order to start and sustain a fission reaction, the concentration of ‘neutron’ poisons in the system must be low. In geological systems B, V and REE are the most important poisons. Differences in the V concentration of the various deposits in the Franceville basin may explain the distribution of the reactors. The deposits of Mounana, Boyindzi and Mikouloungou have no reactors and a V content ranging from > 100 ppm to 1%. Vanadium has even been mined at Mounana. Oklo has a V content that is always below 100 ppm. The concentration of B and REE is low in all of the uranium deposits. For B, this is due to the meteoric origin of the mineralising fluids, which did not migrate through the basement, a common source of this element. If we consider that uranium was released by the leaching of detrital monazites then REE and U fractionated during the migration of the fluids from the conglomerates to the deposits due to varying pH–Eh conditions [11].

4. Behaviour of fission products and actinides

Two main episodes of migration of actinides and fission products must be considered. The first episode, responsible for the so-called ‘old migrations’, took place during the fission reactions, or soon after, as the reactors were still deeply buried (3000 m). The second episode of migration occurred more recently and is still active. It took place when the reactors rose to the surface and went into contact with groundwater. These migrations are dominant in the reactor of Bangombé whereas they are less important in the deeper reactors of Oklo.

4.1. Old migrations

From the early studies on the Oklo reactors it has been recognised that some fission products or their decay products had been retained almost completely, while others had been almost totally lost from the vicinity of the reactors (Table 1) [14]. Elements that have been retained essentially within the core of the reactors were Pu, Th, REE, Zr, Ru, Rh and Pd. All of these elements are soluble in the uraninite crystals and have low solubility in natural groundwater, especially under reducing conditions that pertained in the uranium deposits of Oklo. Other fission product elements, such as rare gases, I, Cd, alkali metals and alkaline earths (e.g. Sr), appeared to have been totally removed from the reactors. These elements either are gaseous or have high solubility in natural groundwater, even under reducing conditions.

Detailed analyses of specific isotopes allowed one to obtain information on the behaviour of their precursors which were formed during the fission reactions. Analyses of isotopes such as ^{235}U [18], ^{99}Ru [19–23], ^{90}Zr [24] and Bi [14], whose precursors are ^{239}Pu , ^{99}Tc , ^{90}Sr and ^{237}Np , respectively, allowed one to show that these fission elements and actinides migrated during the fission reaction or soon after. The processes that allowed their retention in the core of the reactors or its vicinity are highly variable and have different efficiency.

In the reactor it has been recognised that the most efficient matrix for the retention of actinides and fission products is the uraninite mineral. However, it has been shown that other matrices such as apatite, clay minerals, zirconium silicates and oxides (Fe, Mn) may have played a role in the retention of fission products and actinides. As an example, Pu was stored in apatites [25] and chlorites [26] in the core of the reactor 10. In the core of the reactors, between uraninites grains, 20 to 200 μm size metallic aggregates containing fissionogenic Ru, Rh and Te associated with As, Pb and S were found. These aggregates also exist in artificial spent fuels of PWR plant suggesting their analogy with spent fuels.

It appears therefore that the reactors are similar to spent commercial reactor fuels in that for both uraninites, the $^{235}\text{U}/^{238}\text{U}$ ratio at the time of reactor operation was similar and that there are solid solutions

Table 1. Synthesis of the behavior of the fission products and actinides at Oklo; from Gauthier-Lafaye et al. [14]

Tableau 1. Inventaire des actinides et produits de fission à Oklo

Element	Retention				
	Core of the reactor			Clays of the reactor	
	Uraninites	Inclusions	Migration	Clays	Others
Cs			+		
Rb			+		
Sr			+		
Ba			+		
Mo		+	+		
Tc		+		+	
Ru		+		+	?
Rh		+		+	?
Pd		+		+	?
Y	+			+	
Nb	?				
Zr	+			+	heavy.miner/oxide
Te					
REE light	+			+	apatite
Ce					
Pb	+				galena
Bi					
Th	+			+	heavy miner.
U	+			+	apatite
Np	?				
Pu	+			+	apatite

+
+

 Close relation to no relation between the minerals and the chemical elements.

between UO_2 and the original fission products or the isotopes produced after decay. In all cases, and before the ascent of the reactors, the redox conditions have been such that uraninite remained stable.

Uraninites at Oklo and Bangombé have a very constant chemical composition [27]. Their ‘impurities’ contents, when normalised to lead content are: SiO_2 : 0.7–1%; FeO : 0.3–0.7%; CaO : 1.5–1.7%. Lead content, however, ranges between 0 in the very altered uraninites (affected by recent alterations) to 25% in well-preserved uraninite of the deepest reactor (reactor 10). Most of uraninites at Oklo and Bangombé indicate that they have been subjected to coffinitisation, that is, the transformation of $U(IV)O_{2-x}$ to $U(IV)$ -silicate. It has been shown that in old uraninites, this phenomenon did not change drastically the chemical composition of uraninites [27]. It is possible that coffinitisation occurred under anoxic conditions and is related to the process of oxidative alteration of uraninites by water radiolysis and the later precipitation of $U(IV)$ -silicate as the reducing conditions are restored.

4.2. Recent migrations

Recent migrations of actinides and fission products were mainly studied in the Bangombé reactor which is located close to the surface. The Bangombé reactor is located at a depth of around 12 m within an area of regional discharge and is subjected therefore to weathering and chemical exchange with groundwater. Present time equatorial climate originates the formation of a laterite profile affecting the FB black shales

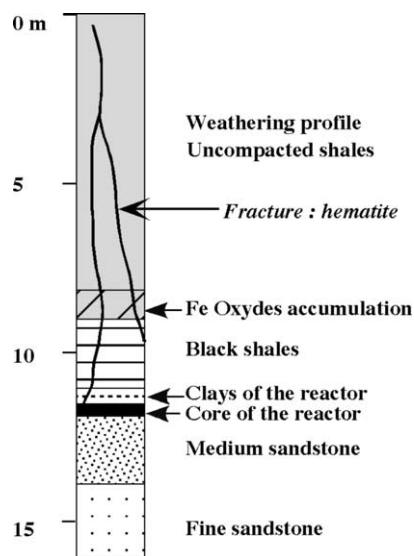


Figure 5. Schematic Log of the bore hole BAX 3 at Bangombé. From Pourcelot and Gauthier-Lafaye [16].

Figure 5. Log schématique du sondage BAX 3 à Bangombé.

underlying the reactor and the oxidation of the mineralised sandstone that host the reactor. An idealised cross-section of the reactor is given in Fig. 5.

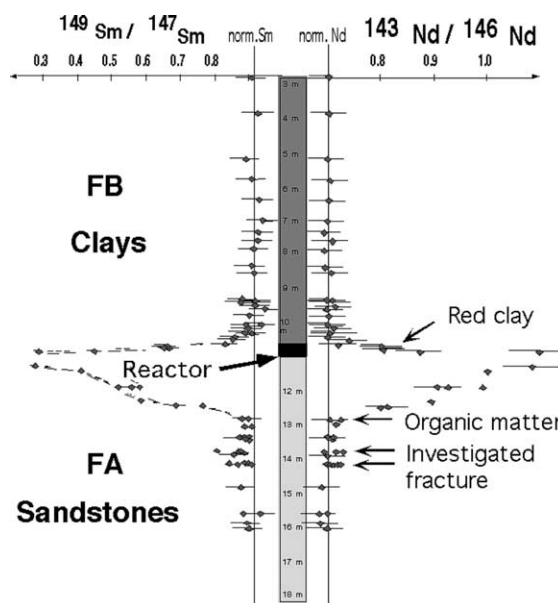
The main effect of migration of the surficial oxidising fluids into the reactor is the dissolution of uraninite crystals and the resulting liberation of fission products in the environment. Therefore one of the main questions that has been studied during the last European programme concerned the behaviour of these elements in the geological environment and their possibilities to be retained in the core of the reactor or its close vicinity.

Dissolution of uraninites has been observed either in the reactor core where secondary U(VI) minerals (idiomorphic U, CA, Fe, Al, LREE phosphates, U (Al, Pb) and U (Fe, Ti) silicates) were observed [28,29] and in the groundwater where uranium with $^{235}\text{U}/^{238}\text{U}$ ratio lower than 0.72% has been measured [30]. It is assumed that the present time shape and size of the reactor were greatly modified due to such alteration processes. However, the fact that uraninite is still present after a long period of time suggests that some geochemical processes should maintain the dissolution phenomenon at a very low rate. It has been shown [31] that the stability of uraninite results from the buffer effects due to FeII/FeIII equilibrium controlled by the iron bearing paragenesis and by the hydrolysis of graphite which is encountered within the 'argiles de pile' in the close vicinity of the reactor. The reducing environment, which is generally maintained inside the reactor can explain the conservation of the uranium in spite of the leaching by oxidising groundwater. These apparent redox-buffering conditions happen to vary with time, i.e. changes in the water table level, explaining the occurrence in the same samples of minerals representative of reducing (uraninite, sulphides) and oxidising (uranophosphates, Fe-oxides, P-rich coffinites) conditions.

Mechanisms of retention of uranium during weathering include the precipitation of new U-bearing minerals and adsorption processes [29,32]. The vertical and lateral flow of groundwater is responsible for the oxidation and dissolution of primary sulphides originating acidic solution which facilitated the oxidation and dissolution of uraninite. The uranyl cations so formed migrated and precipitated as uranyl minerals, mainly phosphates, silicates, silico-phosphates and some of them, especially coffinite, with high content of P and LREE. Adsorption of uranium, together with P, mainly occurs on Fe-oxyhydroxides, but this uranium retention process seems less efficient than the precipitation one, at least in the close vicinity of the reactor. On the other hand, in the weathered FB formation, U adsorption on clays and (Fe–Mn)-oxyhydroxides minerals may be the dominant process [32].

Figure 6. Distribution of fissionogenic Sm and Nd in the bore hole BAX 8 which cross-cut the reactor of Bangombé. Norm.Sm and norm.Nd refer to Sm and Nd having normal isotopic composition, not influenced by the fission products. From Bracke et al. [33].

Figure 6. Distribution de Sm et Nd de fission dans le sondage BAX 8 qui recoupe le réacteur de Bangombé. Norm.Sm et norm.Nd correspondent aux compositions isotopiques normales de Sm et Nd, non perturbées par les produits de fission.



Fissionogenic REE were liberated during the dissolution of uraninite and have migrated in the vicinity of the reactor (Fig. 6) as it is suggested by their distribution along the bore-hole BAX8 [33] and the occurrence in groundwater samples of Sm and Nd having isotopic composition reflecting fissionogenic contribution [34]. Detailed studies on the mineralogy of the reactor and on the elemental and isotopic compositions of U, Sm and Nd in groundwater samples show that new precipitation of P-LREE-coffinites in the core of the reactor allows the retention of most of the dissolved uranium and fissionogenic LREE that were liberated by the dissolution of primary uraninites. This observation is in agreement with the experimental results that have shown that the UO_2 stability in the solid phase and U(VI) predominance in the aqueous phase are thermodynamically compatible, particularly in slightly acidic to neutral pH ranges and in transitional redox waters [35].

5. Conclusions

Natural nuclear fission reactors occurrence is restricted to a very short period of time during the history of the Earth, i.e. around 2000 Ma, when the oxygen content in the atmosphere and hydrosphere allowed the oxidation of U(IV) to U(VI) whereas the $^{235}\text{U}/^{238}\text{U}$ ratio was still high, slightly higher than 3%. Other fission reactors may have existed at that time. Recently, Meshik et al. [36] suggested that the ^{136}Xe content (part of which is of fissionogenic origin) of the atmosphere was enhanced by precambrian nuclear reactors similar to those of Oklo and which have involved 1% of the precambrian crustal uranium. However, these reactors should have been destroyed during the various orogenesis that affected most of the ancient sedimentary basins. The very unique preservation of the reactors of the Franceville basin is due to the location of the basin in the centre part of the very stable western African craton. The occurrence of clays around the reactor created an impermeable shield that protected the core of the reactor against the aggressive diagenetic and weathering fluids. The reduced potential around the reactor was maintained both by Fe^{2+} minerals (chlorite, pyrite, siderite) and by organic matter that played an important role in the preservation of the reactors.

Uraninite crystals have shown good capacity to retain most of the actinides produced by the fission reactions and most of the fission products that have ionic radii close to that of uranium. When uraninite has been dissolved, i.e. in the weathered zone of the Bangombé reactor, the reduced conditions in the close

vicinity of the reactor allow its precipitation in newly formed Si–P–REE–uranium minerals. Around the reactor, when more oxidising conditions prevail, adsorption of U on clays and Fe-oxyhydroxides represent an important mechanism for its retention. Hydrogeochemical studies have shown that at the metric scale, the chemical conditions are highly variable around the reactor of Bangombé involving therefore many different mechanisms for the retention of uranium and REE. It is of great importance to be able to detect each of these processes and estimate their importance in the overall uranium and other elements (REE, platinum metals, etc.) migrations.

The geochemical models that study and predict the behaviour of uranium around the reactors at Bangombé and Oklo are good examples which illustrate our capability to understand such a complicated natural system and our knowledge of the mechanism of retention and migration of uranium and fission products in a natural system. It appears that the basic information regarding the kinetics and thermodynamics of these processes are scarce [37] and in the future, a great effort must be put in that direction in order to make the models more precise and to increase our confidence in them.

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Discussion

Question de É. Brézin

Est-ce qu'aux époques encore plus reculées où la proportion d' ^{235}U était encore bien supérieure on peut imaginer qu'il y a eu des phénomènes de nature explosive ?

Réponse de F. Gauthier-Lafay

On pense que avant 2000 Ma d'années il n'y avait pas de processus géochimiques permettant d'obtenir de fortes teneurs en uranium dans la nature. Aussi, même si les proportions en ^{235}U par rapport ^{238}U étaient très fortes avant 2000 Ma il y a peu de chance d'avoir pu obtenir une masse critique. De plus, Naudet a montré que dans les terrains géologiques et en particulier à Oklo, la chaleur produite par les réactions de fission s'évacue très mal ce qui assure la stabilité des réactions nucléaires et interdit que les réacteurs puissent « s'emballer ».