Session 1 : Special Olko Anniversary

From Natural core breeding to the Oklo discovery

Chair

Benoît GALL

(Strasbourg, France)

Talk Ok 1.1

The Oklo phenomenon, discovery, first questions, first answers

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The discovery in 1972 of the natural nuclear reactors at Oklo provoked many questions among scientists, physicists and geologists. Under the leadership of R. Naudet, the CEA launched the "Franceville Project" which brought together researchers from around the world to answer the many questions raised by this discovery. The initial answers were compiled in two proceedings of two symposia held in Libreville and Paris in 1976 and 1978. This project was then followed by two European projects led by the CEA and the IRSN in 1990 and 1996.

My presentation will quickly recall the scientific adventure of the first years of work on the Oklo site: what were the hypotheses and misunderstandings to finally propose a first coherent but still incomplete scenario in 1978.

The reactors are described with emphasis on the only reactor that has been preserved in its natural state, the other 15 having been exploited. Information is given on the thousands of samples taken.

The history of the reactors is reconstructed in time and the geological context on the basis of the most recent data [1]. In particular, we show that the conditions that led to the functioning of nuclear reactions in a geological series more than 2100 million years old and having had turbulent history are quite exceptional and why they never occurred again [2].

But the great advantage of the Oklo reactors is that they can teach us a lot about the behaviour of fission products and actinides buried for two billion years in a clay environment rich in bitumen. Oklo has already shown that it is a remarkable project in this respect. Much more can be learned, but Oklo's first lesson is to show that we still have a lot of work to do to understand and master a system for trapping nuclear waste in a natural environment.

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Talk Ok 1.2

From routine sample measurements in CEA to the Oklo phenomenon

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In 1972, before shipping natural uranium to the USSR for enrichment operations in ^{235}U, the analysts at Pierrelatte plant noted a slight deficit in ^{235}U: 0.7171 instead of 0.7202. The Direction des Productions of the CEA launched a vast campaign of analyses for the different mines exploited, at all stages of the elaboration of uranium: analyses on the ore, then on the yellow cake, on the uranium oxides issued of the yellow cake transformation, then on the UF_4 and on the UF_6.

For this analysis campaign, the Direction des Productions relied on the analytical laboratory of the Pierrelatte plant and on the Central Analytical Laboratory of the CEA, managed by Michele Neuilly, where I was in charge of analyses by mass spectrometry.

The numerous chemical and isotopic analyses of uranium lead to Gabon and more precisely to the Oklo mine. Indeed, the closer one gets to this site, the higher the uranium content and the higher the ^{235}U depletion.

At a meeting held at the CEA headquarters, it was decided that the laboratories at Pierrelatte and Cadarache would continue to carry out the analysis campaign, and that the laboratory at Cadarache would analyze the samples from Oklo.

COMUF, the company that operated the mines in Gabon, sent two samples of magnesium uranate and two samples of ore to Cadarache. After the analyses of the U and ^{235}U content, there were enough samples left that I decided to analyze them on a spark mass spectrometer, which provides a panoramic analysis of all the isotopes present in the analyzed product. I discovered on the photo plate of the mass spectrometer, isotopic anomalies, in particular the absence of ^{149}Sm, whereas ^{147}Sm was present. The next step was isotopic analyses of some elements, including neodymium and samarium after chemical separation. They revealed that the isotopic composition of these two elements was completely different from that of the natural elements. The results of these analyses were transmitted to the CEA Directorate which sent them to the neutron specialists at Saclay. Their conclusion was as follows, the isotopic compositions of the Oklo ores are identical to those of uranium having undergone a chain reaction of fission.
It was the first time we discovered a natural fission reactor.

The discovery of ^{235}U depletion and the chain fission reaction in the Oklo ore will be the subject of two communications to the Academy of Sciences.

Talk Ok 1.3

Historical interpretations of isotope measurements and applications

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The French PWR power reactors were using uranium enriched to about 3.5% in U235. Prior to enrichment, a routine check at Pierrelatte revealed abnormalities in U235/U238 isotope ratios. Such anomalies made UF6 inappropriate for enrichment, which was essential. An investigation requested by the General Manager of the CEA has made it possible to clarify the origin of the ore treated and the causes of these anomalies: chemical, nuclear or other? Interpretation of isotope measurements of various fission products (particularly Nd then Sm, Eu...) demonstrated the occurrence of sustained fissions in the extracted ore: once the small contribution of natural elements to this ore was deducted, the resulting isotopic vectors corresponded well to the isotopic vectors of the fission products. In the autumn of 1972, the use depletion codes to calculate the concentrations of 600 PF made it possible to better characterize the "operation" of these reactors: some reaction zones were indeed fast-breeder reactors, since the existence of Pu239 and "rapid fast neutron" fissions on U238 had occurred. The possibility of a critical state has been demonstrated with a simple reactor model. This possibility results from the combination of two particularities: a minimal presence of water with an enrichment in U235 of the order of 3.5% due to the age of formation of the uranium deposit (2Gy). Forty years after NAUDET's first Oklo ore criticality study, more complete studies of inception condition were carried out at the University of Strasbourg with realistic models, explaining the startup condition of these cores.

In 2010, Professor EL ALBANI highlighted the presence of fossils dating back to the same period and located in the vicinity of the reactors. This event was be the object of a bomb in the scientific world since the appearance of such an elaborate life form would have appeared only 0.6Gy before our era.

The joint occurrence of the two separately improbable phenomena (reactors and fossils) can only draw attention; this was the origin of the conference/debate organized in autumn 2018 by SFEN/PACA: "Chance hazard or causal relationship?".

In order to stimulate discussion with radiobiologists, we proposed, as a first step, to calculate the deposition of energy in matter over versus the ages. For this purpose, we use isotopic measurements made nowadays. The case of the Oklo reactors is the first application. The second case, the "*La Crouzille*" uranium mine, is complementary to the first in terms of the nuclear conditions (intensity and age of formation). The third case is the situation of a lagoon environment, looking for possible explanation of the start of Life on Earth. For all these cases we determine neutron spectra and concentrations of about 1800 isotopes (fissile with their progeny filiation products, fission and activation products). These concentration values can be used to discuss the storage of natural or non-natural radioactive waste.

Talk Ok 1.4

Oklo natural analogue of radioactive waste disposal, summary of European Commission projects'results (1991-1999)

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At the dawn of the 1990s, the question of the feasibility of the geological disposal of radioactive waste became significant. Among the tools for studying this feasibility, natural analogues had a special place, making it possible to obtain both quantitative and qualitative demonstration elements. Among natural analogue, the Oklo site, in Gabon (equatorial Africa), represents a unique geological environment where nuclear reaction products have been naturally introduced and can still be detected in fossil reaction zones.

From 1991 to 1999, two international research projects conducted under the auspices of the European Commission, studied radionuclide mass transfer processes to the surface, focusing on quantitative assessment of radionuclide migration/retention within the Oklo hydrogeological basin. These projects compiled useful information and tools for the safety assessment of radwaste disposal. The most relevant part included new data on the long-term evolution of spent fuel, suggesting the possible occurrence of a coffinitisation process and measurement of short term leaching rates of Oklo uraninite; modelling tools and data to quantitatively describe the interaction of uranium and rare earth elements with a complex clayey material; the demonstration of radionuclide trapping in generic mineral phases such as Mn and Fe oxides, chlorite, illite and specific secondary minerals such as phosphates and Zr-silicates; and a consistent understanding of redox buffering in a clayey environment, from deep to surface conditions, generic enough to be adapted or applied to other sites.

Tal Ok 1.5

Influence of Initial poisons and clays on the criticality of Oklo natural nuclear reactors.

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In the aim to assess more the Oklo phenomenon through the numerical modelling and simulation, within the geological context based on field observations and measurements, a Python-based code is developed to automate the criticality research for a given configuration among a specific parameter, considered as the main variable of investigation. The home-made python program interacts with dedicated code for nuclear reactor criticality calculation, namely MCNP. This allowed us to investigate the asymptotical criticality occurrence, which corresponds to infinite multiplication factor k_∞ as a function of Uraninite fraction volume and total saturated porosity: k_∞ (V_UO2,Φc)≆ for different situation defined with relevant parameters, namely: Initial Poisons (Gd, Sm and Nd) and Clay fraction in the Gangue part of the U-rich ore. Indeed, in the first step of the present work, a generic U-rich ore was simulated over a given interval of Uraninite volume fraction and the corresponding porosities needed (Critical Porosity Φc) to reach criticality were obtained. It shows that an optimal point can be defined as the minimal one on the isocritical curve k_∞ (V_UO2,Φc)≆. This point, called "Inception point", is the most likely configuration to occur with low Uranium and less water defined by totally saturated porosity.

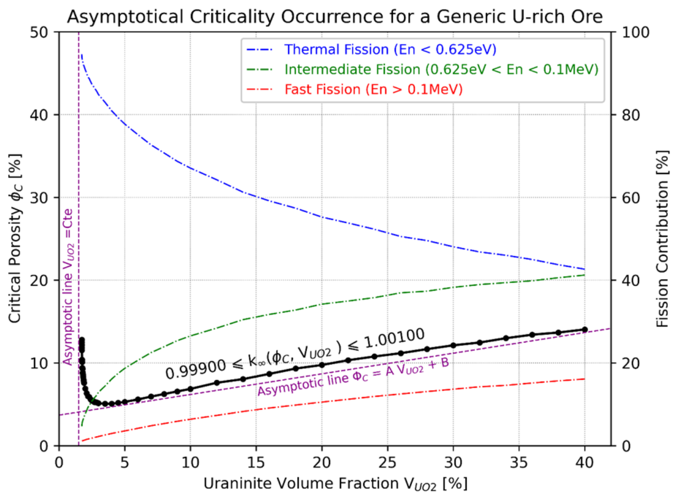

Figure 1. *Typical results of criticality research for a generic U-rich ore obtained with Python-based code with MCNP*

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Session 2 : Special Olko Anniversary

From Oklo samples to natural core simulations

Chair

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Talk Ok 2.1

The Gabonionta: Great Oxidation Event, reactors and Life...

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The emergence of complex life more than five hundred million years ago marked the beginning of change in the Earth's biosphere. This evolutionary change is associated with numerous events including increasing predation, burrowing, and animal diversity during the so called "Cambrian Explosion" at the Ediacaran-Cambrian boundary. However, several studies have reported that scattered fossils of large individual multicellular macro-organisms that use cells as building blocks existed during most of the Proterozoic Eon, and some of these early lineages (such as red or green algae) still exist. The recent discovery of centimetre-sized fossils of more than 1500 specimens from the 2.1 Ga Paleoproterozoic black shales in Gabon reveals growth of macro-organisms in a coordinated manner. The biogenicity of these remains was investigated using a multi-approach studies. On the surface, the fossils resemble irregularly shaped cookies with split edges and a lumpy interior. High-resolution X-ray tomography revealed their varying elongate, lobate, string, circular and a sheet-like structure with a pervading radial fabric and a neat pattern of central fold shapes and sizes. These structures are too complex to be simple products of inorganic processes. Geochemical data confirmed that the carbon contents in the fossilized tissues were assembled by biological processes. Moreover, the iron-sulfide (pyrite) mineral replacing most of the tissue were formed by bacteria "breathing" sulfate, rather than oxygen, during decomposition of the organisms in sediments. Some of these species showed evidence of organism motility in shallow marine waters with free oxygen and provide support for presence of multicellular life to a minimum of 2.1 billion years ago, almost at the beginning of the Proterozoic Eon. Large size generally signifies an energy-demanding way of life. Breathing oxygen, as we do, is a much more efficient way of obtaining energy than other physiological processes. The Proterozoic Eon saw two major events of oxygen build-up in the atmosphere (and, thereby, in the oceans); the first near the beginning of the Eon, 2.45–2.2 billion years ago, and the second at the end, 0.8–0.54 billion years ago. The evolution of the Gabonese biota, representing an early step toward large-sized multicellularity, may have become possible by the first boost in oxygen, whereas the "Ediacara biota" could have been fuelled by the second. Why it took around 1.4 billion years for the multicellular organisms to take over is currently one of the great unsolved mysteries in the history of the biosphere.

Talk Ok 2.2

Natural Nuclear Reactors: prediction, search, discovery, operation and implications

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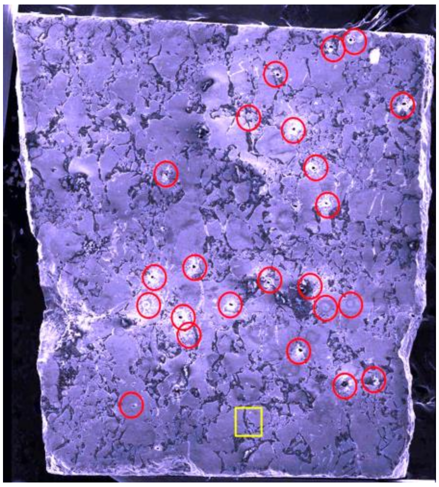
Natural nuclear reactors operated in Oklo (Gabon) about 2.0 billion years ago [1]. This phenomenon was predicted and led to a systematic search for uranium deposits that went critical. Fifty years ago, just a few years after this search was abandonded, the Oklo phenome-non was discovered by chance. Many elements extracted from the Oklo reactor material still carry clear isotopic signatures of ^{235}U fission, ^{239}Pu production and neutron capture reactions. Isotopic compositions of these elements provided reconstruction of neutron fluence, amount of consumed ^{235}U, and an effective duration of nuclear fission chain reaction that was estimated to last for hundreds of thousands of years [2]. It was not clear, however, whether the reactor was operating continuously or in pulses. One proposed mechanism was based on burning up highly neutron absorbing impurities (RRE and/or boron) [3]. As the strong absorbers were burned up at one edge of the reactor zone and uranium at the other one, the active zone could have shifted along the U-vein making different parts of the natural reactor been active at different times [3]. Another potential self-regulation mechanism could have involved water acted as a neutron moderator. As the temperature of the reactor increases, all unbounded water was converted into steam, reducing neutron thermalisation and shuting down the chain reaction. Only when the reactor cooled down and the water concentration increases again, could the chain reaction resume. A tiny sample from reactor zone RZ-13 kindly provided by Maurice Pagel, Philippe Holliger and François Gauthier Lafaye carried the answer to this and several other questions.



Novel analytical techniques LENGA and NAUTILUS used for analyses of this 4×3 mm slab revealed:

* the highest concentration (~8´10^{17} atom/g) of fission Xe ever observed in natural materials [4]
* cycling operation of RZ-13 of Oklo and self-regulating mechanism [5],
* lowest ^{235}U/^{238}U = 0.3655 and capture of fission Cs and Ba 5 yr. after the shutdown [6] ,
* Al-phosphates and metallic aggregates preserve certain fission products over a geologic time [4, 7].

(Red circles show craters made by LENGA = Laser Extraction Noble Gas Analyses. Yellow square shows area studied by NAUTILUS = NAval Ultra-Trace Isotope Laboratory's Universal Spectrometer, the US Naval Research Laboratory).

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Talk Ok 2.3

Evidence of fast neutron operating in Oklo

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The fission processes for thermal neutron induced ^{235}U and ^{239}Pu fission and for fast neutron induced ^{238}U fission produce fragments with a wide range of mass (72<A<162; A=mass number), and neutrons. As the results of fission events, many elements in the Oklo reactor zones (hereafter, RZs) and the related samples show the variations in the isotopic compositions caused by a combination of reactions of nuclear fission, neutron capture and radioactive decay. Isotopic measurements by mass spectrometry provide useful information of geochemical behavior of fissiogenic radioisotopes and nuclear characteristics of the reactors. Since the discovery of the first RZ in 1972, many isotopic studies have been performed to understand the mechanism of the operation as fission reactors and to trace the migration behaviors of fissiogenic isotopes produced in the Oklo RZs [1-2]. In this talk, I will show some typical examples of the isotopic data, and explain the interpretation how and why the fission reactions occurred in the Oklo RZs. In particular, one of the Oklo RZs, RZ 13, is several specific features in the view point of nucleonic characteristics. As representative parameters to characterize the operating conditions of RZs, neutron fluence as the time integration of a neutron flux generated in RZ, duration of RZ operation, restitution factor of ^{235}U from decay of ^{239}Pu produced by neutron capture of ^{238}U, and the proportion of fission events due to ^{235}U, ^{238}U and ^{239}Pu, are listed in Table 1.

By comparison of the data between RZ13 and other RZs, fission contribution of ^{238}U for RZ13 is found to be significantly higher than those of other RZs. Considering that ^{238}U reacts with fast neutron rather than thermal neutron, RZ13 might have located at less moisture area, and had reacted with fast neutrons. Furthermore, the lower restitution factor of RZ13 may also support the fast neutron operating in RZ13 because of the preference of fission reactions of ^{238}U instead of neutron capture reaction caused by the generation of fast neutrons.

Table 1. Nuclear parameters to characterize the operation conditions of the Oklo RZs [3-5]

RZ No. sample No.	3 SC52-1472	5 KN267-2194	9 mean (n=8)	10 SF84-1485	13 SD37
neutron fluence($\times 10^{20}$ n/cm^2)	2.28	4.37	3.62	6.22	7.80
operating duration($\times 10^5$ years)	3.00	0.95	2.24	1.56	0.242
Restitution factor of ^{235}U	0.41	0.47	0.48	0.38	0.11
fission contribution					
^{235}U (%)	95.0	96.0	87.7	92.4	74.9
^{238}U (%)	2.0	3.0	7.8	3.8	17.9
^{239}Pu (%)	3.0	1.0	4.5	3.8	7.2

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Talk Ok 2.4

The Oklo natural fission reactors and dynamical models of dark energy

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Paul Dirac is credited with being the first physicist to speculate in print that fundamental constants like the Newtonian gravitational constant G may change over cosmological intervals. Incongruous though this notion may be, it has gained traction among physicists. Modern theoretical frameworks, which attempt to unify all the fundamental forces, accommodate the possibility of varying fundamental constants, as do dynamical models of dark energy. There are intriguing hints, from absorption spectra of interstellar matter, that the fine structure constant α, which determines the strength of electromagnetic interactions, may vary spatially across the cosmos, the changes found being at the level of about 10 parts per million (ppm). This astrophysical result is in stark contrast to a seminal study (conducted by Thibault Damour and Freeman Dyson) of Oklo neutron capture data which concludes that, in the time since the Oklo natural fission reactors were active (about 1.95 billion years ago), α has changed by less than 0.1 ppm. There is a tendency to take this Oklo bound *cum grano salis* because of the perception that the nuclear physics invoked in its derivation is fraught with substantial unquantifiable uncertainties. I discuss excitation, Coulomb, and deformation corrections, using deformed Fermi density distributions fitted to the output of Hartree-Fock + BCS calculations (with both the SLy4 and SkM* Skyrme functionals), the energetics of the surface diffuseness of nuclei, and thermal properties of their deformation. Although the net correction is uncertain to a factor of 2 or so, it constitutes no more than 25% of the Damour-Dyson estimate. Making allowance for additional uncertainties in the modelling of the Oklo reactors, I conclude that, subject to a weak and testable restriction on the change in light quark masses, the relative change in α over the last 1.9 billion years is less than 0.01 ppm (95% C.L.). This bound reinforces the idea that, of the many dark energy models which predict that fundamental constants do change, only those which suppress the variation of α in the presence of matter are phenomenologically acceptable.

Talk Ok 2.5

Parallel between natural Oklo cores and industrial reactors operating

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The first man-made nuclear reactor was developed by Fermi at the University of Chicago and was first started in December 1942. This was the confirmation that one is able to use sustained fission reactions to produce energy. Following this success, the many types of nuclear reactors studied have given rise to several families of reactors corresponding to different orientations and technical choices. They are linked mainly to the choice of fuel (natural uranium, enriched uranium, plutonium), coolant (water, carbon dioxide, helium, sodium), fast or slow of neutrons and moderator for slow neutron reactors (graphite, light water, heavy water).

Out of all these choices the Pressurized Water Reactor (PWR) family is the closest to the Oklo natural reactors. Many intriguing similarities are observed and discussed in the present Supplementary Information C. Our present-day understanding of the PWR operating conditions has been a great help for understanding the Oklo reactors. On the other hand, the fast neutron reactors can also be put in parallel to Oklo cores since they did breed significant amount of plutonium-239 and since some zone are known to be operated as fast neutrons. The presentation will set a parallel between what Nature offered us with Oklo cores and the optimized cores we are able to build and operate.

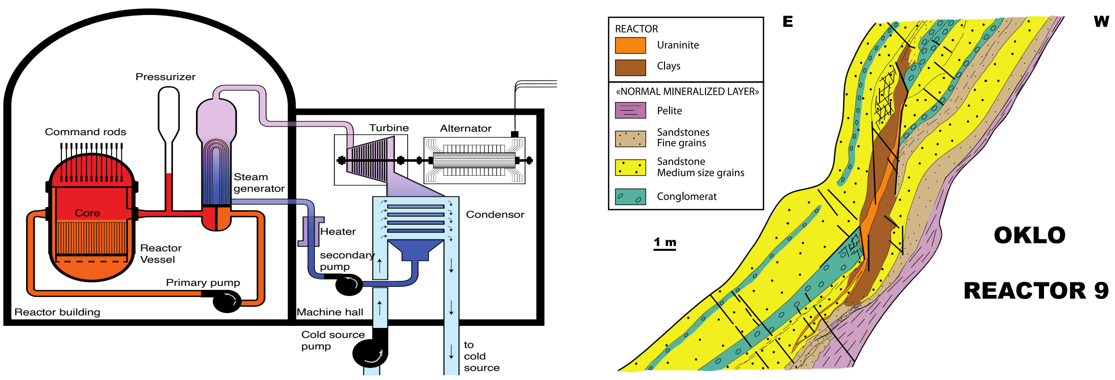

Figure 1 | **Illustration of PWR circuits and Oklo core.** The primary circuit is a forced heat conduction loop. The 155 bar pressure is regulated by the pressurizer and prevents water from boiling in primary circuit. The secondary circuit is a water/vapour circuit with a pressure at full power of 71 bar. Steam is produced in the steam generator runs the turbine/alternator before being again transformed into liquid water in the condenser. Approximately one-third of the thermal power is transformed into electrical power and the other two-thirds is exhausted to the cold source (river or refrigerating tower). The Oklo core (figure from [1]) was 2000 m below surface with rather sililar pressure and temperature as the PWR reactors !

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