The natural Reactors at Oklo (Gabon): 2 billion Years before Fermi!

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(Synopsis)

1. The Italian Navigator has landed in the New World.

On December 2, 1942, this cryptic message announced that the team gathered around Enrico Fermi in Chicago had managed to sustain a fission chain reaction in the first ever man made nuclear reactor, CP1. This was the climax of a decade long search, starting with the discovery by Chadwick in 1932 of the neutron, a particle able to interact with the nuclei without being hampered by their electric charges, the series of experiments by Fermi sending "moderated" neutrons against every nucleus of the Mendeleyev Table, the discovery of the fission of uranium by Otto Hahn and Lise Meitner in 1938. When the team led by Joliot discovered, a few months later, that 2 to 3 new neutrons were emitted during the fission, they were able to conceptually design a nuclear reactor, a facility using a sustained fission chain reaction to generate vast amounts of energy, but World War 2 shifted the research efforts to America.

And for three decades, it was believed that CP1 was not only the first man made reactor, but the first nuclear reactor ever – full stop.

2. Radioactive Earth.

Not everybody realizes that geothermal energy is just another name to describe the radioactivity of our planet. Among the heavier elements retained during the formation of Earth (most of the lightest elements escaped its too small gravity), a number have only radioactive isotopes. Potassium[1], Thorium and Uranium are the most abundant remaining today. The energy they keep releasing during their radioactive decay is the central heating system which supplements what we receive from the Sun.

Natural uranium is (today) composed of three major isotopes, ^{238}U (abundance 99.2744%), ^{235}U (abundance 0.7202%) and ^{234}U (abundance 0.0054%). This very precise composition is the same – almost – everywhere on Earth. All these isotopes are radioactive and decay with time, but not with the same speed. The half-life of ^{238}U is 4.51 billion years while ^{235}U decays by half in "only" 710 million years. Therefore, the relative abundance of ^{235}U increases if we go back in time: at the creation of the solar system, it was close to 17%, and about 3.58% two billion years ago. 3.5% is the level to which we painfully enrich the uranium today to fuel our Light Water Reactors… In the 50s, some authors played with the idea that fission chain reactions could have occurred naturally when the enrichment was so high, but so many conditions would have been required that it seemed far fetched, and there was no evidence left anyway.

3. A Nuclear Detective Story.

In June 1972, at the Pierrelatte enrichment plant devoted to Defense Applications, a routine mass spectrometry analysis of UF_6 feed material exhibited a discrepancy: only 0.7171% of the uranium in the samples ^{235}U, instead of the magic 0.7202! Even though the discrepancy was small, it was so unusual that the French Atomic Energy commission

[1] ^{40}K in our bones is responsible for half of the radioactivity of our own body, which amounts to about 8000 Bq for an adult.

CEA, operator of the plant, started a thorough investigation. First, it was not an artifact: the anomaly was confirmed on several measurements on other samples. Accidental contamination by depleted uranium from the plant itself was then eliminated and so was the use of reprocessed uranium as there was no ^{236}U in the samples. The investigators then traced the anomaly back through all the stages of uranium processing, from Pierrelatte to Malvesi to Gueugnon where the concentrates exhibited the same low ^{235}U concentrations. These concentrates all came from COMUF which operated two uranium mines in Gabon, at Mounana and Oklo, the mill being located at Mounana. Very soon it appeared that all the anomalous ore came from the northern part of the – very rich – Oklo deposit. In some shipments, the level of ^{235}U was as low as 0.44%. Between 1970 and 1972, in the 700 tons of uranium delivered by the Mounana mill, the deficit of ^{235}U exceeded 200 kg, hardly a trifle!

Oklo mine uranium was indeed different from natural uranium everywhere else. Why?

"Natural" isotopic separation was excluded: if it had produced depleted uranium, where was the enriched fraction? As soon as August, the hypothesis of very ancient fission chain reactions was formulated, and investigators started to search for fission products (or, rather, the granddaughters of hypothetical fission products). The spectrum of fission products is so distinctive that it constitutes an unmistakable marker that fission reactions have taken place. The presence of such fission products was clearly identified: at some point in the uranium deposit history, it had become a "natural" nuclear reactor. The discovery was duly heralded [1, 2] but many questions remained. When did the reactor "started"? How long did it "operate"? How was it "controlled"? The detective story was not finished.

Later on, it was found that there were actually 15 reactor sites in Oklo, and another one in Bangombé, 30 kilometers away from the main deposit.

4. Current answers to some questions about Oklo.

To run a nuclear reactor, you need a high concentration of uranium with a minimum percentage of ^{235}U[2], you need water to slow down the neutrons[3] and evacuate the calories and you must avoid those elements which absorb neutrons greedily like boron, cadmium, hafnium, gadolinium and other "poisons". You need also a minimum size (in the case of a deposit, a minimum thickness of the seam) to prevent too many neutrons from escaping from the reaction zone.

It is only around 2.2 billion years ago that the patient work of photosynthesis accomplished by the first algae released enough oxygen in our atmosphere for the surface waters and ground water to become oxidizing. Only then could the uranium diluted in granite be leached out and concentrated before mineralization in places where oxido-reduction would occur. Rich deposits cannot be older. On the other hand, since 1.5 billion years, ^{235}U abundance has decayed below a level which makes spontaneous fission workable. It took a lot of studies, in geology, chemistry and reactor physics to narrow the bracket of time to the present estimated value : the reactions must have started **1 950 ± 30 million years ago**.

The deposits were located in very porous sandstone where the ground water concentration may have been as high as 40%, probably due to the partial leaching of the silica (quartz particles) by the hot groundwater, at a time where, the radioactivity of

[2] You can operate reactors with natural uranium but only if you use heavy water D_2O or very pure graphite as moderator and a specific "heterogeneous" fuel/moderator pattern, like in CANDU and Magnox types. It would be very unlikely to find such pattern in nature.

[3] Neutrons emitted during fission move too fast to split easily other nuclei, but if the neutrons can "bounce" off the nuclei of a moderator, this will slow them down and make further fission more likely.

Earth being higher than today, the thermal gradient underground was probably higher too. During the reactors operation, the water temperature rose significantly, accelerating this "de-silicication" process and, by difference, increasing the concentration in uranium, therefore compensating for its depletion by fission. As a matter of fact, the concentration of uranium in the reaction zones is extremely high, sometimes above 50%, and the higher the uranium concentration, the lower its ^{235}U content. Furthermore, losing its silica, the surrounding sandstone became clay and thus prevented an excessive migration of groundwater and keeping the uranium in place.

From the fine analysis of the spectrum of fission products, we know that a number of the fissions occurred in plutonium, bred by neutron capture in ^{238}U and now fully decayed to ^{235}U since its half-life is only 24 000 years (By the way, so much for the notion that plutonium is "artificial"). This allowed the physicists to calculate that, varying from one zone to another, reactions did take place during an enormous period of time ranging **from 150 000 to 850 000 years**!

The reactors where "controlled" by several mechanisms, the main one being temperature: as the fission power was released, the temperature rose. Higher temperature means both an increase in absorption of neutrons (without fission) by ^{238}U and a decrease in the efficiency of water as a moderator: at a given temperature level, a level varying with time and the progressive depletion of fissile uranium, the reactions stabilize, as they do in our reactors[4].

By combining geology and temperature considerations, it is now believed that the reactors in the northern part of the deposit operated at **a depth of several thousand meters**, under deltaic then marine sediments. At such depth, the conditions of pressure and temperature were close to those of the Pressurized Water Reactors of today (350 to 400°C, 15 to 25 Mpa), while the southern zones operated at roughly 500 meters deep, with conditions resembling more to those of a Boiling Water Reactor (250°C, 5 Mpa)[5]: even the Oklo designers did not choose between the present fierce competitors!

Even though significant alteration occurred in recent times when the tectonic uprising and erosion brought the reactors close to the surface, and especially when the Okolo Néné River gouged the valley, the heavy elements thorium, uranium and plutonium did not move at all, nor did the rare earths fission products, as well as zirconium, ruthenium, palladium, rhodium and a few others. On the other hand, krypton, xenon, iodine, barium and strontium have moved, but maybe only after a few million years.

5. Oklo as a "natural analogue" of a radioactive Waste Disposal Site?

Soon after the discovery, and beyond the pure scientific thrill, the nuclear community was very excited by its implications, notably as a "natural analogue" for the geologic disposal of High Level radioactive Waste (HLW).

There is more and more an international consensus that the best way to dispose of HLW issued from the production of electricity by nuclear reactors is to install them, with a proper conditioning and packaging and additional engineered barriers, in a stable underground geologic stratum where the radioactive decay will progressively reduce their toxicity to a harmless level. But this decay takes a long time, and it is quite a challenge to demonstrate the containment of the radioactive products over such a long period of time, ranging from tens to hundreds of thousands of years. It can only be done through

[4] Radioactive decay of some absorbing fission product also played a role over such long periods.

[5] If the operating time was immense, the power density in the « core » was only **one millionth** of its value in a commercial reactor today.

physico-mathematical modeling, with the inherent uncertainties associated with the completeness and accuracy of the models and their propagation along the calculations.

There, in Oklo, Mother Nature had contained *precisely the same radioactive elements* not for hundreds of thousands, not for millions, but for a couple of billion years, and without engineered barriers or special packaging.

So much is true, especially for the heavier elements which constitute most of the radiotoxicity of the HLW packages[6]. But the comparison cannot be pushed too far. To use a teenager's expression, the Oklo reactors are "too much"… If we could find a similar phenomenon one million years old, that would be perfect, but we have seen this is physically hopeless. For instance, most of the migration occurred during the reactions themselves, over close to a million years, when the conditions were far more troubled than what we expect in a steady and cozy disposal facility: the site has been deeply modified, losing by de-silicication three quarters of its substance, minerals have been altered by irradiation, temperature have run high and significant water convection did occur! Let us say Oklo provides a good presumption, but not a demonstration.

6. Conclusion : A unique Phenomenon?

Let me borrow my conclusion from the foreword by the late Jules Horowitz to the book by Roger Naudet [3] which I have used extensively for this paper: "*It is after all plausible that fission chain reactions might have spontaneously occurred about two billion years ago, during a period of time long enough to provoke locally significant anomalies in the isotopic composition of some elements, notably uranium. What constitutes a miracle is that, despite the upheavals that the Earth surface has undergone since this ancient era, the evidence did survive to our time, in Oklo, to be discovered owing to the watchfulness of the CEA analysts*".

There is no reason to believe that what occurred at lest 16 times near Oklo did not happen anywhere else on the Earth, especially in old and rich deposits like exist in Australia or Canada… but more than three decades after its discovery Oklo remains unique. It remains unique as a geologic curiosity, and it remains unique as a nuclear detective story.

[6] They have been retained within the UO2 crystallites themselves

A few References

The Discovery (September 1972)

[1] R. Bodu et al. Sur l'existence d'anomalies isotopiques rencontrées dans l'uranium du Gabon. CR Académie des Sciences Paris 275 D p.1731

[2] M. Neuilly et al. Sur l'existence dans un passé reculé d'une réaction en chaîne naturelle de fissions dans le gisement d'uranium d'Oklo (Gabon) ibid. p.1847

Synthesis

[3] R. Naudet OKLO: Des réacteurs nucléaires fossiles. Etude physique. Eyrolles, Paris, 1991

Selected Websites

www.wonuc.org/nucwaste/oklo.htm
(with many interesting links !)

www.ans.org/pi/np/oklo/

www.energethique.com/notions/oklo.htm
(in French)

www.ocrwm.doe.gov/factsheets/doeymp0010.shtml
(Oklo and HLW disposal)