

## CHAPTER 6

# NATURAL NUCLEAR REACTORS, THE OKLO PHENOMENON

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### 6.1 INTRODUCTION

In 1972, natural nuclear reactors were discovered in West Africa in the Gabon Republic, at the adjacent Oklo and Okelobondo mines, and at Bangombe 35 kilometers away.

The natural reactors became critical about 1.8 eons or billion of years ago. One of the reactors released 15,000 MWth.years of energy and consumed six metric tonnes of uranium. It operated over several hundred thousand years at a low average power of 100 kWth. Sixteen reacting zones where these natural reactors occurred have been identified.

From 1942 until 1972, humans thought that they have achieved a feat that nature did not, when the first sustained nuclear chain reaction was initiated at the Chicago Pile number one, CP1. In CP1, graphite, which is a form of carbon, was used as a moderator containing spheres of natural uranium metal and oxide,  $UO_2$ .

Scientists believed for a long time that the elements were synthesized only in the stars. The chemist Francis W. Aston in 1922 speculated that the process of nucleosynthesis could also occur on Earth. He suggested a situation in which: “In this event the whole of the hydrogen on the Earth might be transformed at once and the success of the experiment published at large to the universe as a new star.”



Figure 1. The Oklo uranium mine in the Gabon Republic, West Africa.



Figure 2. View of the Zone 2 at the Oklo mining site.



Figure 3. The ancient super continent Pangea. In about 100 million years from present time, the Pacific Ocean will be half as wide as it is today, the Atlantic Ocean will be twice as wide or more, the Mediterranean will be gone and replaced by a mountain range, the Rocky mountain would have been eroded, so will the Alps and many other mountain ranges and the continents will well be on their way to rejoining into a New Super Pangean continent.



Figure 4. Past location of the Oklo site in the Pangean supercontinent. Presently, it is located at the Gabon Republic, Africa.

In 1953, George W. Wetherill of The University of California at Los Angeles, and Mark G. Inghram of the University of Chicago suggested that some uranium deposits may have once operated as natural fission reactors.

Paul K. Kuroda, a chemist from the University of Arkansas in 1956 suggested that an assembly of uranium and water could become a self-sustained chain reacting system in the early history of the Earth. He calculated that the size of the deposit should exceed the average diffusion length of a fission neutron at about 2/3 of a meter. Paul K. Kuroda was proven right in May 1972, when it was discovered that humans were not first in achieving a self-sustained fission chain reaction. Nature had preempted them about 1.8 eons or billions years earlier. Aston was also proven right to a certain extent, where the process of transmutations of the elements had existed naturally on Earth billions of years ago in the form of fission rather than in the form of fusion as he speculated.

The fissile isotope  $U^{235}$  has a natural atomic abundance of 0.7202 percent, but some samples from the Oklo site have a reduced  $U^{235}$  natural abundance of 0.7171 percent. Other samples are depleted down to 0.44 percent. This difference could only be explained on the basis that some of the  $U^{235}$  had been consumed in some nuclear, possibly fission reaction.

Like contemporary power reactors, a natural reactor would require the existence of several special conditions, namely fuel, a moderator, a reflector, lack of neutron absorbing poisons and some way to remove the heat generated. At Oklo, the area was naturally loaded with uranium by water transport and deposition. The concentration of  $U^{235}$  is artificially enriched for most modern reactors to a level of 3-5 percent, but at the time of the Oklo

reactor it was naturally enriched with an abundance of approximately 4 percent. The 4 percent abundance of  $U^{235}$  with water and clay as moderators was enough for a self-sustained critical fission nuclear reaction. The Oklo site was saturated with ground water, which served as a moderator, reflector and coolant for the fission reaction. There was a lack of neutron poisons such as boron or lithium, before the reaction began, and fission products like xenon and neodymium served as neutron absorbing poisons, absorbing neutrons and acting to moderate the power level.

## 6.2 URANIUM CONCENTRATION

According to James Lovelock in: “The Ages of Gaia;” a unique consequence of the appearance of oxygen in the early history of the Earth was the advent of the world's first nuclear fission reactor.

When the Earth formed 4.6 eons or billion years ago as a result of a star super-nova, uranium was much more enriched in the fissile isotope  $U^{235}$  to about 17 percent rather than the present 0.72 percent because it decays more rapidly than the more common isotope  $U^{238}$ . The Earth’s atmosphere was not oxidizing in the early Archean era, otherwise many critical reactor occurrences would have occurred.

The element uranium is insoluble in water under oxygen-free conditions, but is readily soluble in water in the presence of oxygen. When enough oxygen appeared in the Proterozoic era as a result of the photosynthesis process to render the ground water oxidizing, uranium in the rocks began to dissolve. In the form of the uranyl ion, it became one of the many elements present in trace quantities in flowing streams.

The strength of the uranium solution would have been at most no more than a few parts per million (ppm), and uranium would have been but one of many ions in solution. In the place that is now Oklo, a stream flowed into an algal mat that included microorganisms with a strange capacity to collect and specifically concentrate uranium. They performed their task and eventually enough uranium oxide was deposited in the pure state for a nuclear reaction to occur.

## 6.3 URANIUM MINING AT OKLO

The Pierrelatte uranium enrichment plant in France used the uranium ore mined from the Oklo mine located in the southeastern part of the Gabon Republic in Africa (Figs. 1, 2). Figures 3, 4 show the past location of the Oklo site. The continents at that time formed a single global Pangean continent. The tectonic plates have since moved apart to the present separated positions.

The ore was milled at the Mounana mill near the mine, and then shipped to a processing plant in France, and then to an enrichment plant. At the Pierrelatte enrichment plant, it is a routine procedure to conduct mass spectroscopic analyses of the uranium hexafluoride gas ( $UF_6$ ) used in the enrichment process. In May of 1972, a discrepancy was noticed in the proportion of  $U^{235}$  relative to the  $U^{238}$  contained in the samples.

Natural uranium is found in nature today in the form of the isotopes shown in Table 1.

Table 1. Half lives and natural abundances of the naturally occurring uranium isotopes.

Isotope	Natural Abundance a/o (atomic percentage)	Half-Life $T_{1/2}$ (years)
$U^{234}$	0.0054	$2.440 \times 10^5$
$U^{235}$	0.7200	$7.040 \times 10^8$
$U^{238}$	99.2746	$4.488 \times 10^9$

The most abundant of the uranium isotopes is the one with the longer half-life:  $U^{238}$ . The  $U^{234}$  isotope is primarily a radioactive decay product of  $U^{238}$ . Being an even atomic number (Z) and an even mass number (A) isotope,  $U^{238}$  is fissionable, but not fissile, that is; it cannot fission with low energy neutrons. On the other hand, the even atomic number and odd mass number isotope  $U^{235}$  is fissile with a less stable nuclear shell structure and can fission with practically zero energy neutrons.

The natural abundance at 0.72 percent or about 1/140 of the  $U^{235}$  isotope has not been constant since the creation of the Earth, 4.6 billion years ago. At this time, the natural abundance of  $U^{235}$  was 17 percent in natural uranium. It has since transformed through the process of radioactive transformation into other isotopes with a half-life of 704 million years, eventually into stable lead isotopes such as  $Pb^{207}$  for  $U^{235}$  and  $Pb^{206}$  for  $U^{238}$ . Since this is less than the half-life of the  $U^{238}$  isotope, its natural abundance will decrease as a function of time. About two billion years ago, the natural abundance of  $U^{235}$  in U was about 4 percent.

The mass spectroscopic discrepancy that was noticed at the Pierrelatte plant was a subtle one. In the samples that were analyzed, the abundance of  $U^{235}$  in the sample was 0.7171 percent instead of the 0.7202 percent found in nature. The discrepancy needed to be urgently accounted for, since  $U^{235}$  has to be meticulously assessed by the manufacturing as well as the regulatory agencies, to assure that none of it would be diverted to weapons proliferation.

There were also some samples that were found slightly enriched in the  $U^{235}$  isotope. A possible explanation is that it is due to the formation of  $Pu^{239}$  with subsequent decay into  $U^{235}$ .

Let us go back to those samples that were depleted in  $U^{235}$ . At first, it was thought that the  $U^{235}$  was contaminated by depleted uranium that is left over after the enrichment process. A detailed accounting process showed that no depleted uranium was missing at the plant. Detailed sampling showed that all the samples originating from the Oklo mine had the discrepancy. Some samples even showed a lower abundance of  $U^{235}$  at the level of 0.44 percent. Mining of richer ore zones in the mine did not affect the discrepancies shown in Fig. 5. Figure 6 displays the change in the abundance of  $U^{235}$  in the ore as well as the uranium ore concentration over the period of the investigation.

A hypothesis was advanced suggesting that induced fast fission has occurred in the ore reducing the abundance of the  $U^{235}$  isotope. Vlasov, a Russian scientist who had advanced a theory explaining the Tunguska event in Siberia as being caused by an antimatter meteorite impacting the Earth, came with the hypothesis that the  $U^{235}$  at the mine site may have been partially fissioned by fast neutrons released by such an impact.

## 6.4 NATURAL FISSION REACTORS

The tectonic hypothesis suggests that the reactors occurred in an aquatic environment that eventually was raised through plate and fault movements, as shown in Fig. 7. Concentration of the uranium is surmised to have happened through algal or density concentration. The reactors would have occurred in a mixture of uranium, clay and water as shown in Fig. 8.

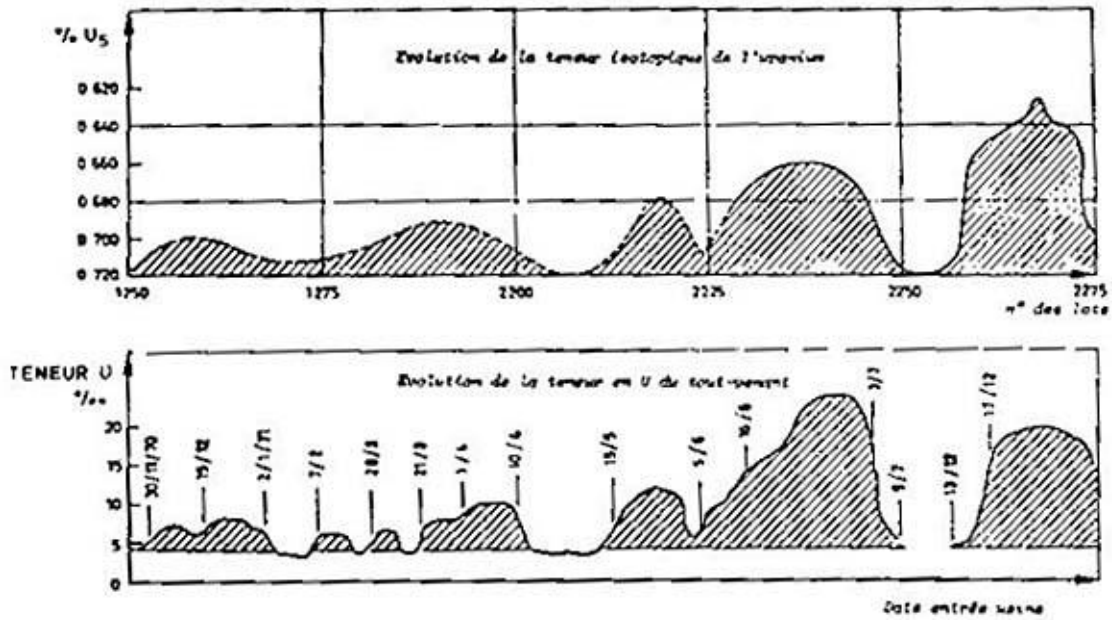


Figure 5. Uranium concentration and U<sup>235</sup> concentration in the uranium ore.

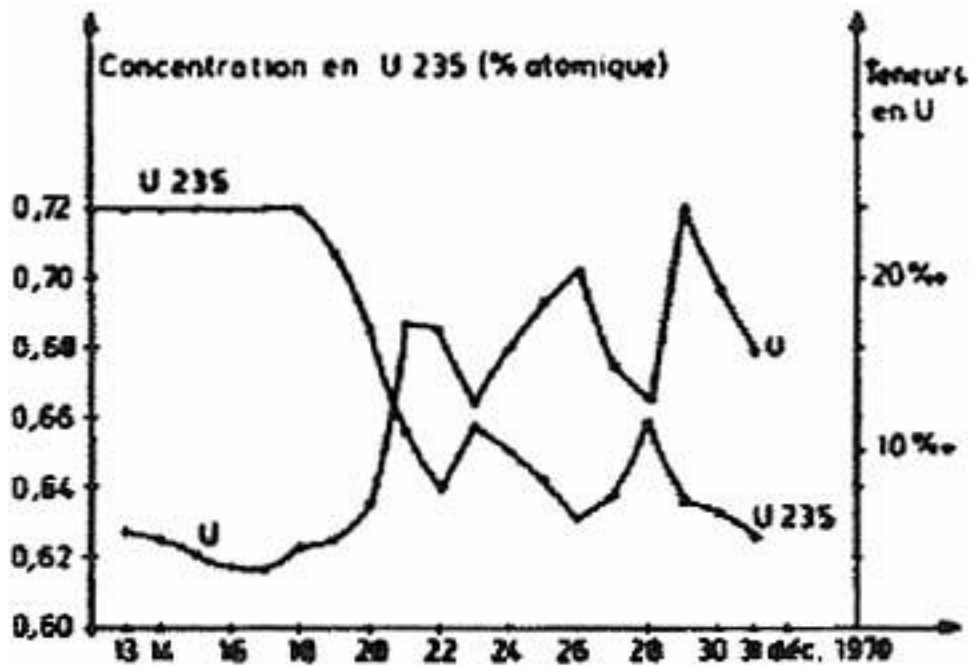


Figure 6. The change in the abundance of  $U^{235}$  in the ore as well as the uranium ore concentration over the period of the investigation.

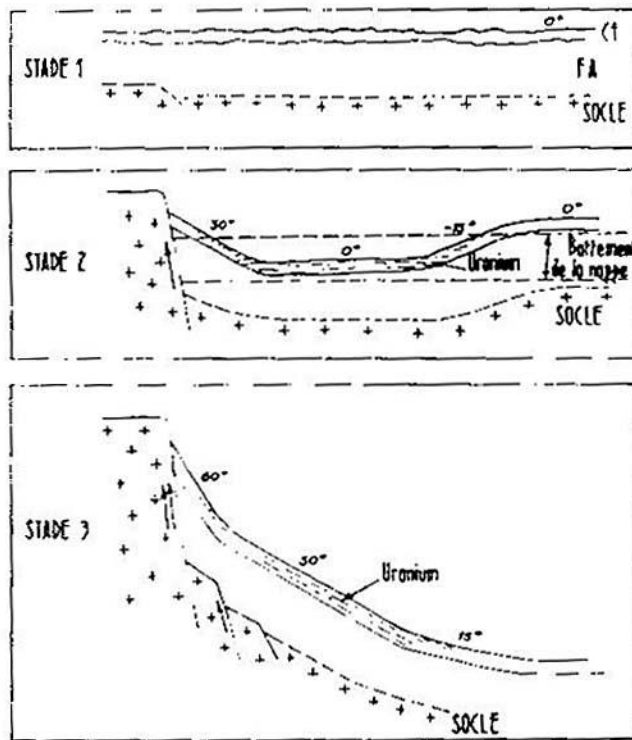


Figure 7. Tectonic movement raised the Oklo site from its original shallow aquatic stage.



Figure 8. Sample of helical ore in a kaolinite matrix suggesting thermal effects in a clay rich in water with convective movement. Magnification is 100 times.

Scientist started investigating the presence of fission products at the mine site. It was discovered that indeed fission products were present, not just at one site, but at sixteen different reactors zones. Figure 9 shows the locations of six natural reactors zones at the Oklo site. The simultaneous impact of several antimatter meteorites at the same location being quite improbable, this questions the validity of Vlasov's theory of an antimatter meteorite impact.

When the samples were analyzed for their different isotopes content, the abundances of the different elements in the samples did not agree with the normal natural abundances of these elements in nature. The distribution of certain isotopes that were found in the samples, such as the rare earth metal neodymium, did not correspond to their natural abundances. After corrections for neutron capture and the natural background, they were found to correspond to the distribution of these isotopes that result from the fission of  $U^{235}$ . Neodymium has several stable isotopes at the end of short fission product chains, which have significant fission yields.

Neodymium as a fission product gave strong indications that the natural reactors had indeed operated. The analysis of the distribution of the neodymium isotopes is used in safeguards and nonproliferation enforcement activities to detect clandestine fission reactor, reprocessing, or nuclear testing activities. Table 2 shows the abundance of the neodymium isotopes in nature, from the fission process, and from samples from the Oklo site. The distribution of these isotopes is consistent with a fission process rather than natural abundance.

Table 2. Percentage Atomic Abundance of the Neodymium isotopes in nature, fission and the Oklo samples.

Isotope	Natural Abundance a/o	Fission of $U^{235}$ a/o	Oklo Sample M a/o	Oklo sample M, corrected for the natural element a/o
$U^{235}$	0.7202	-	0.4400	-
$Nd^{142}$	27.11	0.0	1.38	0.0
$Nd^{143}$	12.17	28.8	22.1	22.6
$Nd^{144}$	23.85	26.5	32.0	32.4
$Nd^{145}$	8.30	18.9	17.5	18.05
$Nd^{146}$	17.22	14.4	15.6	15.55
$Nd^{148}$	5.73	8.26	8.01	8.13
$Nd^{150}$	5.62	3.12	3.40	3.28
$Eu^{151}/Eu^{153}$	0.916	2.580	0.145	-
$Ce^{140}/Ce^{142}$	7.990	1.060	1.5700	-
$Sm^{149}/Sm^{147}$	0.924	0.475	0.0030	-

Neodymium has seven stable isotopes, but only six are fission products. The abundance of the neodymium at Oklo sites was compared to other areas and to the neodymium found in fission reactors. Once the samples were compared, the abundance of



the neodymium isotopes was found to correlate positively with that found in present-day fission reactors. The fission products studied matched what would have been the result of a self-sustained nuclear fission reaction. There is even evidence that the reactor bred its own fuel, bombarding the  $U^{238}$  with neutrons, creating another fissile isotope  $Pu^{239}$ , which in turn fissioned and decayed through alpha particles emission over time into  $U^{235}$ .

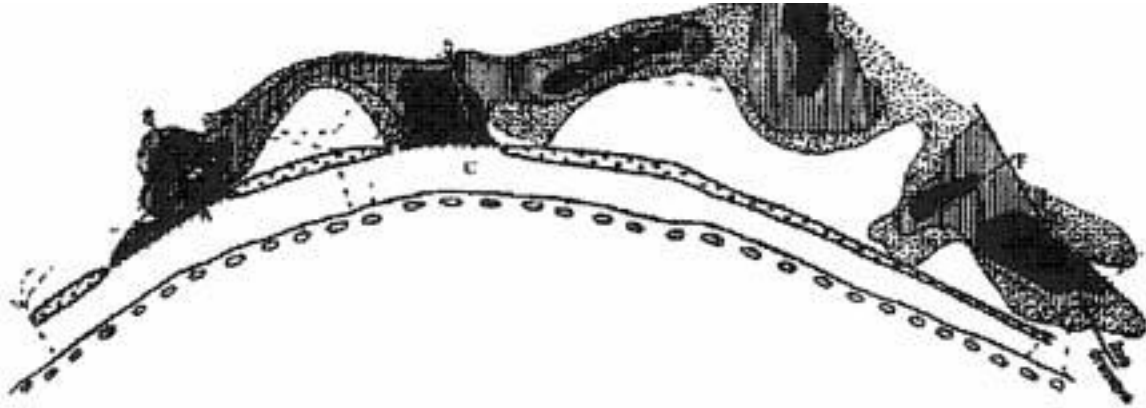


Figure 9. Locations of six of the sixteen natural reactors zones at the Oklo site.

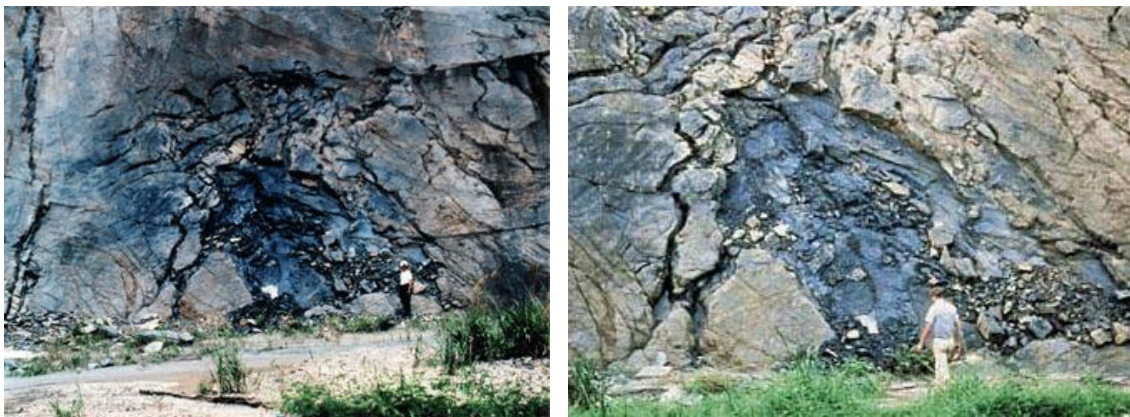
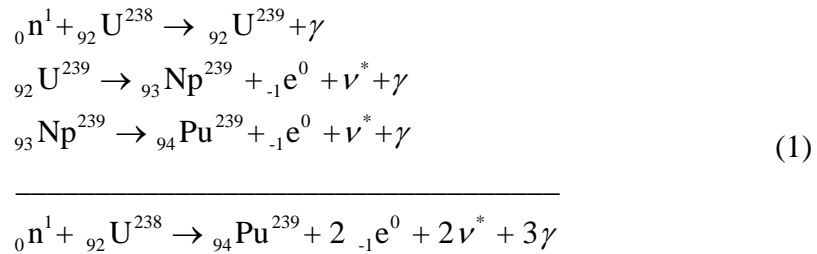


Figure 10. Outcropping of one of the Oklo natural reactors.

Using radioactive dating methods, the samples were found to be about 1.7 billion years old. The  $U^{235}$  abundance at this time in the past would have been in the range of 4 percent, rather than the present 0.72 percent.

The geological evidence suggests a marshy environment where uranium would have been concentrated and mixed with a kaolinite clay background in the soil, with water present and providing moderation to fast fission neutrons into thermal neutrons (Fig. 10).

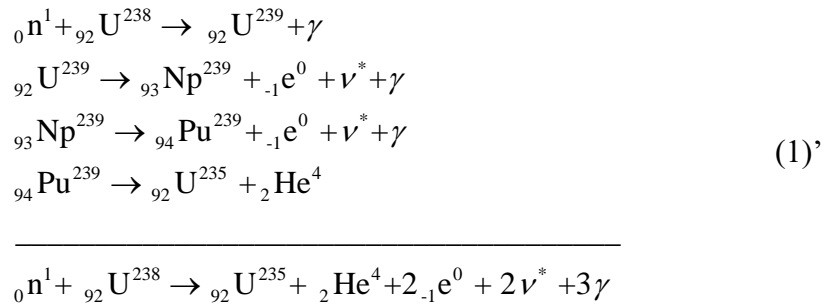
The anomaly could be explained on the basis that in the distant past, conditions at the site of the mine were such that sixteen “natural fission reactors” went critical and operated at the site of the Oklo mine, long before humans achieved a self-sustained fission chain reaction. The fissioning process would have depleted the amount of  $U^{235}$ . The enriched samples would have contained  $Pu^{239}$  bred from  $U^{238}$  through the nuclear capture and breeding reactions:



It then fissioned or transformed into  $\text{U}^{235}$  through the radioactive decay reaction:



Adding Eqns. 1 and 2 yields the overall reaction:



The last equation shows that  ${}_{92}\text{U}^{239}$ ,  ${}_{93}\text{Np}^{239}$  as well as  ${}_{94}\text{Pu}^{239}$  act as catalysts, in chemistry's parlance, in the process of conversion of the  ${}_{92}\text{U}^{238}$  isotope into the  ${}_{92}\text{U}^{235}$  isotope and an alpha particle or  ${}_2\text{He}^4$  nucleus, while releasing nuclear energy in the process.

This reaction would have also fed the reactor with the  ${}_{92}\text{U}^{235}$  fissile isotope. It seems that 2/3 of the fission process in the reactor zone was caused by the  ${}_{92}\text{U}^{235}$  initially present in the ore. The remaining 1/3 would have been contributed by the bred  ${}_{94}\text{Pu}^{239}$  and  ${}_{92}\text{U}^{235}$ .

The presence of 30 elements generated as fission products at the reaction zones is an unmistakable proof that this event did in fact occur at a situation where the conditions for a self-sustained chain reaction were available in nature. These conditions can be described as follows.

## 6.5 THE $\text{U}^{235}/\text{U}^{238}$ OR FISSILE TO FISSIONABLE FUEL RATIO

Nuclear reactors depend on an intricate balance between neutron creation from the fission process, their absorption in the materials of the reactor, and their leakage from the surface of the reactor volume.

It is possible to maintain a self-sustained chain reaction if, after a fission process has released on the average three and half neutrons, at least one neutron is still available to

cause fission and continue the chain reaction. This is after the two and a half other neutrons on-average have been absorbed, or have leaked from the system.

One can use existing uranium with its natural abundance of 0.72 percent in the  $U^{235}$  isotope, only if materials with a low absorption for neutrons, such as heavy water ( $D_2O$ ) or pure graphite (carbon) are surrounding the uranium fuel as cooling media or moderators. In fact, a successful reactor concept designated as the CANDU concept, for Canadian Deuterium Uranium, uses natural uranium for a self-sustained chain reaction.

If light water ( $H_2O$ ) surrounds the uranium fuel, the neutron absorption in hydrogen is too high for a self-sustained chain reaction to occur, unless the proportion of  $U^{235}$  in uranium is increased beyond the 0.72 percent natural abundance. This is achieved in contemporary Pressurized Water Reactors (PWRs), and Boiling Water Reactors (BWRs), by “enriching” the fuel in increasing its content in  $U^{235}$  from the natural 0.72 percent to a level of 3 to 5 percent. This is accomplished in specialized enrichment plants using physical processes such as gaseous diffusion or centrifugation in the enrichment process.

Heavy water did not exist at the site of the Oklo reactors. However, the ratio of  $U^{235}$  to  $U^{238}$  has not been constant over the passage of time. Since the half-life of  $U^{235}$  (704 million years) is shorter than the half-life of  $U^{238}$  (4.488 billion years), it is being transformed into other isotopes at a faster rate than the  $U^{238}$  isotope.

Neglecting the amount of  $U^{234}$  present in natural uranium, for a simple analysis, one can write at time  $\tau$  for the natural enrichment of  $U^{235}$ :

$$\varepsilon(\tau) = \frac{N^{235}(\tau)}{N^{235}(\tau) + N^{238}(\tau)} \approx 0.007202 = 0.72\% \approx \frac{1}{140} \quad (3)$$

According to the law of radioactive decay:

$$N(t) = N_0 e^{-\lambda t} \quad (4)$$

where  $N_0$  is the number of nuclei present at time  $t=0$ , and  $\lambda$  is the decay constant for the particular isotope, relate to the half-life  $T_{1/2}$  as:

$$\lambda = \frac{\ln 2}{T_{1/2}} \quad (5)$$

Substituting from Eqn. 4 for  $U^{235}$  and  $U^{238}$  into Eqn. 3, we get:

$$\varepsilon(\tau) = \frac{N_0^{235} e^{-\lambda^{235} \cdot \tau}}{N_0^{238} e^{-\lambda^{238} \cdot \tau} + N_0^{235} e^{-\lambda^{235} \cdot \tau}} \quad (6)$$

Inverting Eqn. 6, we get:

$$1 + \frac{N_0^{238} e^{-\lambda^{238} \cdot \tau}}{N_0^{235} e^{-\lambda^{235} \cdot \tau}} = \frac{1}{\varepsilon(\tau)} \quad (7)$$

The initial ratio of the number of  $U^{238}$  to  $U^{235}$  nuclei can be obtained from Eqn. 7 as:

$$\frac{N_0^{238}}{N_0^{235}} = \frac{(1-\varepsilon)}{\varepsilon} \cdot e^{(\lambda^{238}-\lambda^{235}) \cdot \tau} \quad (8)$$

Adding unity to both sides of Eqn. 7 and inverting it, yields the initial abundance of  $U^{235}$  at time  $\tau$  in the past, as:

$$\begin{aligned} \frac{N_0^{235}}{N_0^{235} + N_0^{238}} &= \frac{1}{1 + \frac{(1-\varepsilon)}{\varepsilon} e^{(\lambda^{238}-\lambda^{235}) \cdot \tau}} \\ &= \left[ 1 + \frac{(1-\varepsilon)}{\varepsilon} e^{(\lambda^{238}-\lambda^{235}) \cdot \tau} \right]^{-1} \end{aligned} \quad (9)$$

Thus, over time, the  $U^{235}$  to  $U^{238}$  ratio has steadily decreased. In the past, it was larger than it is now. About 2.5 billion years ago this ratio was at about 4 percent, at the same level as in present day light water cooled reactors. Table 3 shows the variation of this ratio over the time before present.

It is interesting to notice that the conditions existing in present day reactor concepts using an enrichment of 3-5 percent, existed at the time of the Oklo reactors 2.5 billion years ago.

Table 3. Variation of  $U^{235}$  enrichment in years before present (bp).

Percent $U^{235}$ enrichment a/o	Geological Time billion ( $10^9$ ) of years bp.
0.72	0.00
1.30	0.70
1.60	1.00
2.30	1.40
4.00	2.10
7.00	2.80

## 6.6 THE $H_2O/U^{235}$ OR MODERATOR TO FUEL RATIO:

Neutrons emanating from the fission process have an average energy of 2 MeV. At this energy, the neutrons act in a wave like manner, and due to quantum mechanical considerations, are less capable at being grabbed by a nucleus and causing it to fission than slower energy neutrons.

Slow neutrons, particularly those in thermal equilibrium with the moderator, or “thermal neutrons”, are more efficient at causing fissions to occur. In technical terms, they have a higher cross section for fission than fast neutrons. These thermal neutrons at room temperature have a kinetic energy around 0.025 eV corresponding to a neutron speed of 2,200 meters/sec. This factor of  $2.0 \times 10^6 / 0.025$  or about 100 million times reduction in

neutron energy is achieved by collisions by the neutrons, without being absorbed, with the moderator nuclei.

A chain reaction can easier be achieved with the presence of a moderating material when the ratio of fissile material atoms to moderator atoms is at an optimal value. This allows the neutrons released from fission to lose their energy in the moderator before encountering another fuel nucleus and being absorbed by it. Table 4 shows the criticality parameter designated as the infinite medium multiplication factor. This pertains to a medium theoretically infinite in size, hence not accounting for the leakage of neutrons from the surface of a finite fission reacting system. The infinite medium multiplication factor is defined for a medium sufficiently large that neutron leakage is negligible as:

$$k_{\infty} = \frac{\text{Number of neutrons in a given fission generation}}{\text{Number of neutrons in the previous fission generation}} \quad (10)$$

A value equal to unity suggests the possibility, ignoring neutron leakage, of a self-sustained fission chain reaction.

The infinite medium multiplication factor is itself a product of four factors and the relationship is expressed as the four-factor formula:

$$k_{\infty} = \eta \varepsilon p f \quad (11)$$

where:  $\eta$  is the regeneration factor,  
 $\varepsilon$  is the fast fission factor,  
 $p$  is the resonance escape probability,  
 $f$  is the fuel utilization factor.

With an increase in the moderator to fuel ratio, the infinite medium multiplication factor can be made to exceed unity, hence a self-sustained chain reaction can be achieved, as shown in Table 4. Notice that an increase in the moderator to fuel ratio results in a decrease in the fuel utilization factor  $f$ , and an increase in the resonance escape probability  $p$ . Since the infinite medium multiplication factor is a product of an increasing and a decreasing function, it reaches a maximum value for a given moderator to fuel ratio.

Table 4. Infinite medium multiplication factor as a function of the moderator to fuel ratio.

Infinite medium multiplication factor* $k_{\infty}$	Moderator to fuel ratio $H_2O/U^{235}$	Resonance escape probability $p$	Fuel utilization factor $f$
0.55	0.25	0.29	0.99
0.88	0.50	0.47	0.98
1.15	1.00	0.62	0.97
1.34	2.00	0.74	0.95
1.40	3.00	0.79	0.93
1.42	4.00	0.82	0.91

1.43	5.00	0.84	0.89
1.33	10.00	0.86	0.81

\*The fast fission factor  $\epsilon$  is taken as unity.

The regeneration factor  $\eta$  is taken as 1.91.

The Oklo deposit was formed in a marine marsh environment where the algae concentrated the heavy metal uranium in this case. This uranium was mixed with clay, and was saturated with water. The saturation with water may have originally over-moderated the neutrons leading to an explosive energy release. The heat generated may then have evaporated enough water to lead to optimal operating conditions. In present day light water reactors this ratio is about ten percent, the same as existed at the time of the Oklo event.

## 6.7 CRITICAL SIZE OF ORE AND MODERATOR MIXTURE

To achieve a self-sustained chain reaction the leakage from the surface of the fuel and moderator mixture must be at level that would not shut down the chain reaction. Since the leakage is proportional to the surface area, and the fission process occurs in the volume of the deposit, the surface to volume ratio becomes an important consideration in reaching a critical size and a critical mass as shown in Fig. 11. For instance, if we consider a sphere of radius R, the surface to volume ratio is given by:

$$\left(\frac{S}{V}\right)_{sphere} = \frac{4\pi R^2}{\frac{4\pi R^3}{3}} = \frac{3}{R} \quad (12)$$

Similarly, for the case of a cube of side length a, one gets:

$$\left(\frac{S}{V}\right)_{cube} = \frac{6a^2}{a^3} = \frac{6}{a} \quad (12)'$$

The surface to volume ratio is inversely proportional to the sphere's radius R or to the cube's side length a. This is the same as saying that the surface leakage to volume reactions ratio would decrease if the radius of the sphere R, or the side length of the cube a, are increased.

In more formal terms, the finite size of the geometry modified the value of the infinite medium multiplication factor into a smaller value designated as the effective multiplication factor, given by:

$$k_{eff} = k_{\infty} \cdot P_f \cdot P_{th} \quad (13)$$

where  $P_f$  is the fast neutrons non-leakage probability,  
 $P_{th}$  is the thermal neutrons non-leakage probability.

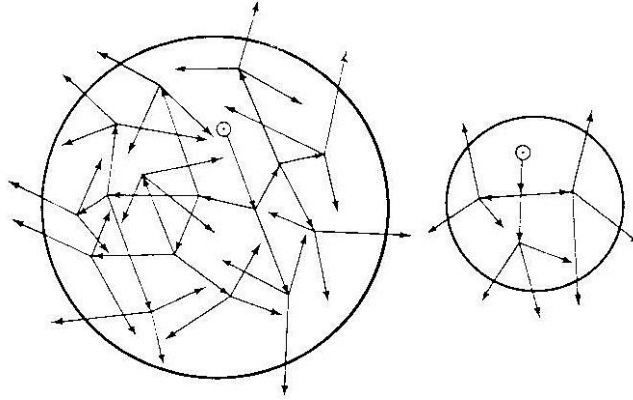


Figure 11. Large size sphere intercepts more neutrons causing fissions within its volume, with a smaller number leaking from its surface, than a smaller size sphere.

For a small size sphere, the neutrons leaking from its surface are not balanced by the fissions occurring in its volume. As  $R$  increases, the surface to volume ratio  $S/V$  decreases, and the leakage of neutrons relative to those capable of causing fissions, decreases accordingly. As we start from a sphere of fuel and moderator, the system is subcritical and no chain reaction can be sustained. As the size is increased, we reach a critical size at which the chain reaction can be sustained. In the case of the Oklo phenomenon, the uranium had to be sealed in seams at least half a meter thick. In fact this occurred in not just one, but in sixteen reaction zones.

## 6.8 NEUTRON POISONS EFFECT

Some elements like cadmium, boron, lithium, indium, and many of the rare earths elements strongly absorb neutrons, and are in fact used in modern reactors in the control rods. There is no evidence at the Oklo site of excessive amounts of these neutron absorbing elements in the ore, which is the conditions ideal for a self-sustained chain reaction in nature.

The conditions at the Oklo site varied over time, which led eventually to its shutdown. It is estimated that the reactor operated intermittently over a period of about 150,000 years. As the reactors were in operation, the content in  $U^{235}$ , decreased, or was depleted, as was observed at the Pierrelatte uranium enrichment plant in France.

## 6.9 RADIOACTIVE FISSION PRODUCTS FATE

The fission products from the chain reaction have mostly transformed into stable isotopes, and were found to have remained localized at the site of the reaction zones. This stability of the Oklo ore deposit is supported by the fact that the uranium isotopes were confined within the grains of the clay that is mixed with the ore. Since about half the remaining  $U^{235}$  resulted from the decay of plutonium, this means that the plutonium remained confined to the ore zone for a time period comparable to its 24,110 years half-life and longer. About 1/2 of the thirty fission products also remained confined to the fission zones.

The strontium<sup>90</sup> fission product was mainly confined to the local zones, with some release to the environment estimated from the krypton<sup>85</sup> noble fission gas and cesium<sup>137</sup>. Engineers and scientists working on the technical aspects of the long-term storage of radioactive fuel and fission products consider these observations with interest.

The distribution of the various nuclides in the ore yielded information about the neutron flux level, the conversion ratio, the fluence or time-integrated neutron flux, the fraction of epithermal neutrons, and the number of fissions caused by the fissile U<sup>235</sup> and Pu<sup>239</sup> and fissionable isotope U<sup>238</sup>.

The total released energy is estimated at 15,000 MWth.years. An existing Nuclear Power Reactor at a power level of 3,000 MWth, would have generated this amount of energy in 5 years. Humans, in the existing Light Water Reactors (LWRs) designs were unknowing imitator of nature.

## 6.10 OTHER NATURAL FISSION REACTORS

The conditions at the Oklo site were probably not unique. Extra reactors locations were discovered a few miles away from the Oklo site.

The isotopic composition mined in the Colorado plateau in the USA shows a lower level of U<sup>235</sup> abundance than uranium mined elsewhere on the American Continent and worldwide. This is shown in Table 5 and suggests that some other natural fission reactors occurrences as well as uranium deposits surrounding them remain to be discovered in the future.

Table 5. Isotopic composition of natural uranium in different parts of the world.

Ore source and location	U <sup>238</sup> /U <sup>235</sup> ratio
Uraninite, Joachimsthal	137.8 +/- 0.28
Uraninite, Great Bear Lake, Canada	137.8 +/- 0.25
Mineral Joe mine, Colorado	137.1 +/- 0.35

## 6.11 DISCUSSION

According to James Lovelock:

“Bacteria could not have debated the costs and benefits of nuclear power. The fact that the reactors ran so long and that there was more than one of them suggests that replenishment must have occurred and that the radiation and nuclear waste from the reactor was not a deterrent to that ancient bacterial ecosystem. The distribution of stable fission products around the reactor site is also valuable evidence to suggest that the problems of nuclear waste disposal now are nowhere near so difficult and dangerous as the feverish pronouncements of the antinuclear movement would suggest. The Oklo reactors are a splendid example of geophysical homeostasis. They illustrate how specific materials can be segregated and concentrated in the pure state, an act of profound negentropy in itself, but



also an invaluable subsystem of numerous geophysical processes. The separation of silica by the diatoms and of calcium carbonate by coccolithophoridons and other living organisms, both in nearly pure form, are such processes and have had a profound effect on the evolution of the Earth.”

## REFERENCES

1. Paul Kazuo Kuroda, “On the Nuclear Physical Stability of the Uranium Minerals,” *J. Chem. Phys.*, Vol. 25, No. 4, pp. 781-782, 1956.
2. Neuilly et. al., “Analysis of Ore Samples from the Oklo Mines in Gabon, Africa,” *C. R. Acad. Sci. Paris*, 275, Ser. D: 1847, 1972.
3. George A. Cowan, “A Natural Fission Reactor,” *Scientific American*, Vol. 235, No.1, pp. 36-47, July 1976.
4. W. Walker, G. J. Kirouac and F. M. Rourke, “Chart of the Nuclides, Knolls Atomic Power Laboratory, The General Electric Company, 1977.
5. James Lovelock, “The Ages of Gaia,” 1988.
6. S. K. Lamoreaux and J. R. Torgensen, “Neutron Moderation in the Oklo Natural Reactor and the Time Variation of Alpha,” *Physical review D*, Vol. 69, Paper 121701(R); June 2004.
7. A. P. Meshik, C. M. Hohenberg and O. V. Pravdivtseva, “Record of Cycling Operation of the Natural Nuclear Reactor in the Oklo/Oklebondo Area in Gabon,” *Physical review Letters*, Vol. 93, No. 18, Paper 182302; October 29, 2004.
8. Alex P. Meshik, “The Workings of an Ancient Nuclear Reactor,” *Scientific American*, pp.83-91, Nov. 2005.

## EXERCISES

1. Today, the natural abundance of  $U^{235}$  in natural uranium is 0.72 percent. Calculate the natural abundance or enrichment in atomic percent of  $U^{235}$  in natural uranium ( $U^{235} + U^{238}$ ), ignoring  $U^{234}$ , at the time of occurrence of the Oklo natural reactors phenomenon, about  $2.5 \times 10^9$  years ago. Compare the enrichment at the time of the Oklo phenomenon to that used in present days Light Water Reactors (LWRs) at 3-5 percent.
2. The concentration of  $U^{234}$  is small nowadays in natural uranium, since it occurs as a decay product of  $U^{238}$ . A more exact expression for the  $U^{235}$  enrichment can be written as:

$$\varepsilon(\tau) = \frac{N^{235}(\tau)}{N^{234}(\tau) + N^{235}(\tau) + N^{238}(\tau)} = 0.007202 = 0.72\% \approx \frac{1}{140}.$$

Calculate a more correct value of the enrichment of  $U^{235}$  at the time of the Oklo phenomenon, accounting for  $U^{234}$ :

$$\frac{N_0^{235}}{N_0^{234} + N_0^{235} + N_0^{238}}.$$

Consider that  $U^{234}$  is a radioactive decay product of  $U^{238}$ .