THE NATURAL NUCLEAR REACTOR AT OKLO: A COMPARISON WITH MODERN NUCLEAR REACTORS

By Dr. Andrew Karam paksbi@rit.edu

Editors note: Despite some claims, there is no evidence or even credible theory that the Oklo nuclear reactor was anything but a natural phenomenon. The 6 reactor zones are spread over a huge area that was a uranium mine during the time it was first discovered. The reactor zones were the result of natural physical processes, active for thousands of years. It should also be noted that the possibility of natural nuclear reactors was first postulated by P. K. Kuroda (1956).

Abstract

Uranium contains only one naturally occurring isotope, ^{235}U, which will sustain a nuclear chain reaction using normal water to moderate and reflect neutrons. At present, this isotope is present in low abundance (0.72%), requiring enrichment to 3% or greater for effective use in commercial nuclear reactors. Two billion years ago, however, the natural abundance of ^{235}U was approximately 3%. Evidence indicates that a rich uranium deposit in Gabon, West Africa achieved nuclear criticality and operated for tens of thousands of years or longer. Comparing the geometric and nuclear characteristics of the Gabon reactor with those of modern, artificial nuclear reactors supports this possibility. An examination of rare earth elements and ^{235}U abundance in the rocks that comprise the reactor zone confirm that a nuclear reactor did operate at this site about 2 billion year ago (Ga), using surface and ground waters to moderate and reflect fission neutrons in order to sustain the chain reaction. Finally, it is apparent that ^{239}Pu was produced in measurable quantities, suggesting that uranium is not the heaviest naturally occurring element known.

Introduction

Natural uranium is composed of three major isotopes, ^{238}U (abundance = 99.2745%), ^{235}U (abundance = 0.7200%), and ^{234}U (abundance = 0.0055%). The isotopic composition of uranium is thought to be homogeneous globally (Faure, 1986). However, uranium in a rich deposit located in Gabon, West Africa, was found to have ^{235}U abundances as low as 0.440%.

Subsequent investigations indicated the presence of isotopes of neodymium and other elements which, in conjunction with the lower ^{235}U abundance, suggest that a natural nuclear reactor existed in the past. Other zones contained slightly elevated abundances of ^{235}U. These are thought to represent the decay products of ^{239}Pu, formed by neutron capture of ^{238}U during reactor operation. Similar reactions occur in modern nuclear reactors and, indeed, form the basis of plutonium production. This reaction (including the subsequent decay of ^{239}Pu, to = 2.411×10^4 a) is:

$$^{238}U + n \rightarrow {}^{239}U \rightarrow {}^{239}Np + \beta^- \rightarrow {}^{239}Pu + \beta^- \rightarrow {}^{235}U + \alpha$$

The absence of ^{236}U ($t_{1/2}$ = 2.342 x l0^{7}a) indicates that induced fission stopped at least 10^{8} years ago. Dating of the strata in which the reactor is found indicates an age of approximately 1.8 Gy (Cowan, 1976). At that time, ^{235}U had an abundance of approximately 3%, owing to its shorter half-life relative to ^{233}U ($t_{1/2}$=7.04 x l0^{8} yrs and 4.468 x l0^{9} yrs, respectively). This paper discusses briefly the geologic setting of the Gabon reactor and compares its nuclear characteristics with those of modern, manmade nuclear reactors in terms of fuel loading, geometry, neutron flux, power, and uranium enrichment.

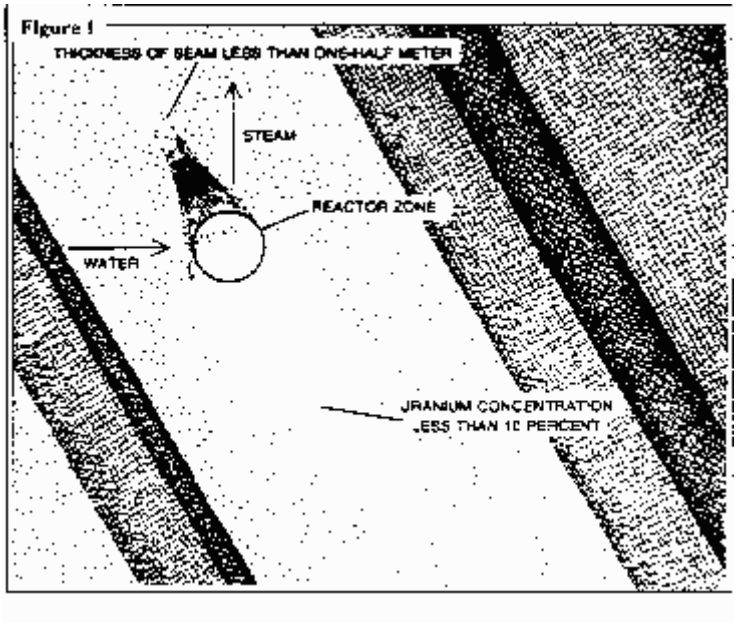
The Gabon Reactor

The Gabon reactor consists of several mineralized zones of uranium minerals in sandstone and conglomerate. The uranium probably originated in nearby igneous deposits, dissolved in oxygenated surface waters, and was deposited at an oxidation-reduction front (similar to the roll-front deposits in Texas). The mobilization, therefore, probably did not commence until there was sufficient free oxygen in the earth's atmosphere to allow oxygenation of surface waters (about 2 Ga). The formation in which the deposits reside was deposited about 1.74 (+0.20) billion years ago (Ga). At this time, ^{235}U had a relative abundance of approximately 3%.

According to Neuilly et al, (1972), "the deposit is stratiform. It is located within the sandstone which forms the basis of the Francevillian (sedimentary basin). The sedimentological characteristics of these sandstones suggest fluviodeltaic deposit conditions. Formed essentially of detrital quartz with some accessory feldspars, they have a cement consisting of secondary silica, phyllite, and organic matter (of asphaltic type). The uraniferous mineralization consists primarily of oxides (uraninite, pitchblende). It occurs in the cement which it may almost totally replace in zones bearing the highest grade uranium ore. It is associated with a few sulfides (pyrite, galena, etc).

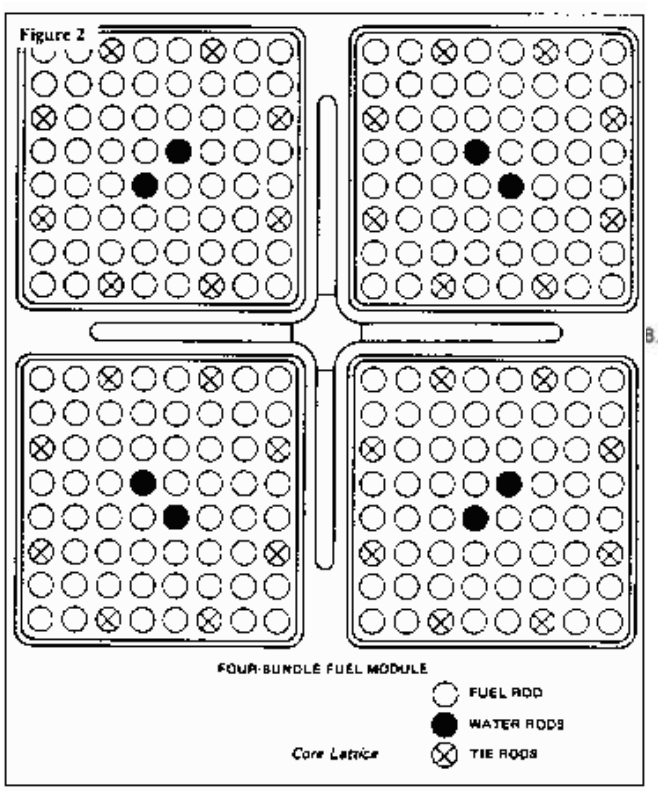
"The thickness of the mineralized layer ranges from 5 to 8 meters. The layer is locally affected by a tectonic movement whose activity served to uplift the sandstone formations bordering on an elongated depression in the sole. The uraniferous mineralization and any redistributions it may have undergone as a result of groundwater flow, appears almost contemporaneous with the sedimentation of the Francevillian."

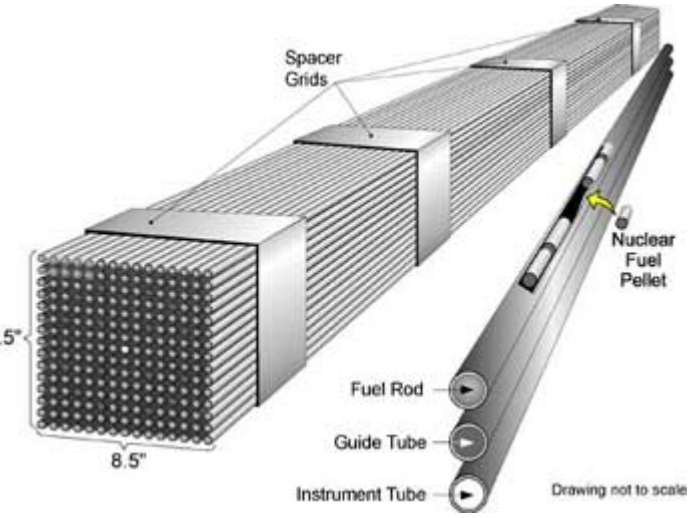
A total of 16 reactor zones have been found in the primary location, and another reactor zone identified about 20 km away (the Bangombé reactor). Current thinking is that a zone of petroleum formation produced an oxygen-free zone because oxygen in groundwater would have been removed by chemical reactions with the petroleum. Uranium, which is fairly highly soluble in oxygenated waters, is insoluble in anoxic waters, so the uranium precipitated out of solution as it crossed the oxidation/reduction front near the petroleum deposits (Janeszek 1999). This is what formed the reactor zones.



An illustration of the reactor is shown in Figure 1 (from Cowan, 1976).

Artificial Nuclear Reactors





A diagram of a typical nuclear reactor fuel assembly is shown in Figure 2 (from Knief, 1990).

Nuclear reactors produce power by controlled induced splitting (fission) of fissionable atoms. Typically these are ^{235}U or ^{239}Pu (although the latter is forbidden for use as reactor fuel in the US). The neutron capture process, however, creates other fissionable nuclides such as ^{241}Pu and ^{243}Pu which, by the end of core life, can contribute significantly to a reactor's power production (Knief, 1992). The nuclear fission process is more efficient with low energy (thermal) neutrons, although fast fission of both ^{235}U and ^{238}U does occur in the fuel.

Since fission neutrons are produced with high energies, they must be slowed down (thermalized) in a moderator in order to be effective at inducing fission. This moderator is typically normal water, although graphite is also used, particularly in reactors intended for plutonium production. Submerging the fuel in the moderator increases reactor efficiency (and decreases required fuel loading) by reflecting escaping neutrons back into the core, allowing them to participate in the fission process.

Due to the low abundance of ^{235}U at present, natural uranium does not undergo a sustained nuclear chain reaction with natural water as a moderator. This is due to the relatively high thermal neutron cross section of hydrogen, which causes the absorption of neutrons that would otherwise cause fissions. Light water reactors, such as the Russian RBMK (graphite moderated) or the heavy water moderated CANDU (CANadian Deuterium Uranium) reactor can maintain self-sustaining chain reactions with natural uranium. Most nuclear reactors operate with uranium fuel which has been enriched in ^{235}U, typically between 3% and 5% (Knief, 1992), although research reactors have ^{235}U enrichments of up to 20%. Military reactors may have enrichments in excess of 90% in order to achieve the power densities necessary for their effective use.

Reactor Physics

Nuclear reactors operate and produce power by maintaining a nuclear fission chain reaction. In order for the reactor to maintain a constant power level, as many neutrons must be produced in each generation as are absorbed or escape from the core.

Of those neutrons which do not escape the core, many are absorbed, but do not cause fissions. Absorbers can be metal non-fuel components of the core, hydrogen in the moderator, fission products that have a high neutron capture cross section, "control poisons" which are placed into the core to shape the neutron flux and to control reactor power, or uranium atoms which do not fission. When the neutron population of the core remains constant the reactor is said to be critical. A growing neutron population is characteristic of a supercritical reactor while a shrinking neutron population makes a reactor subcritical. Therefore, all reactors, when operating at constant power, are critical.

Water is used to cool the reactor core and also to help control reactor power. Neutrons from fission will collide with the atoms in water, losing energy with each collision – this is called “moderation”. This slows the neutrons to the point at which they can cause fission more efficiently. Water will also reflect escaping neutrons back into the reactor fuel, reducing the loss of neutrons from the fuel-bearing region. This effectively increases the neutron population in the core and helps to reduce the amount of fuel needed for a critical assembly. If water is lost from the core, the amount of moderation and reflection is greatly reduced and the reactor shuts down.

The term k_{eff} refers to the relative size of subsequent neutron populations. In a critical reactor, k_{eff} is equal to 1. The value of k_{eff} is determined by the "six factor formula" given below:

$$k_{eff} = \varepsilon L_f P L_{th} f \eta$$

in which:

- ε (the fast fission factor) refers to the ratio of total fissions to those produced by thermal neutrons,
- L_f (the fast nonleakage factor) is the fraction of fast neutrons that do not leak from the core,
- P (the resonance escape probability) gives the fraction of neutrons which are not resonantly absorbed in the core,
- L_{th} (the thermal nonleakage probability) is the fraction of thermal neutron which do not leak from the core,
- f (thermal utilization factor) is the fraction of neutrons which are absorbed by the fuel, and
- η (the fission yield) is the average number of neutrons produced for each thermal neutron captured by fuel material.

The factors L_f and L_{th}, are included in the "buckling" term (B^2); a factor based on core geometry which describes the loss of neutrons from the core through core boundaries. A sphere has the smallest buckling term which is given by:

$$B^2 = \frac{\pi}{2}$$

with R being the radius of the sphere. The presence of a reflector reduces the geometric buckling, as does increasing the size of the fuel bearing region.

As shown by the six factor formula and the buckling equation, there are several ways to increase the probability that a reactor will be able to "go critical". These are:

- Increase the size of the fuel bearing region in order to reduce the buckling (L_f and L_{th}, terms)
- Increase the "fuel to water" ratio to increase the probability that thermal neutrons will be absorbed by fissionable material
- Minimize the number of non-fissionable atoms with high neutron capture cross sections (poisons)
- Provide a reflector to further reduce buckling
- Use a moderator which is close to the mass of a neutron in order to have the maximum energy transfer per collision, reducing the chance for resonance absorption of the neutrons

Reactor Characteristics

Nuclear power reactors typically operate at pressures and temperatures in excess of 1500 psi and 500 °F, although most research reactors operate at atmospheric temperatures and pressures. Fuel loading varies greatly, from tens of kilograms to hundreds of metric tons. Neutron fluxes, too, vary based on the core characteristics, but are typically between 10^{13} and 10^{15} n cm^{-2} sec^{-1}. A commercial nuclear reactor typically has several hundred fuel assemblies, each containing 50 or 60 fuel pins that are approximately 20 feet long and arranged in a circle approximately 20 feet in diameter (Nero, 1979). Military reactors, due to their higher fuel enrichment, are much smaller, as are research reactors, due to their higher ^{235}U enrichment and smaller power output.

Critical mass experiments have shown that a uranium sphere with a 3% enrichment that is fully water reflected will have a minimum critical mass of approximately 2 kg in a heterogeneous reactor and nearly 3 kg in a homogeneous reactor. This minimum critical volume will be just large enough to go critical one time and will not sustain a chain reaction for a prolonged period of time. A larger reactor will have either a higher power output, will be able to operate longer, or both.

Fission Products

Each fission produces approximately 200 MeV in the form of gamma radiation and the kinetic energy of two fission fragments that are produced. This energy is dissipated in the fuel material as the fission fragments interact with surrounding atoms and slow down. The fission fragments are nearly always of uneven mass with varying probabilities of production. Fission yields are highest for nuclides with masses of 95 and 140 amu (when fissioning ^{235}U). The distribution of fission products is characteristic for each fissionable nuclide and can be used to identify the fuel.

Some fission products have high thermal neutron capture cross sections. These can accumulate and poison the chain reaction resulting in either lower power or increased neutron flux to compensate. They are included in the thermal utilization factor in the six factor formula. The final "mix" of fission products, therefore, will reflect the fission yield of each atomic mass number, the half-life and neutron absorption cross section of members of each fission product chain, and the neutron flux in the reactor.

Nuclear Characteristics of the Gabon Reactor

At the time that the Gabon reactor went critical, the abundance of ^{235}U was 3%, similar to that in current commercial nuclear reactors. The approximate shape of the reactor zones is that of a compact mass of uranium oxide surrounded by porous rocks, which were presumably hydraulically connected to surface or ground water, allowing moderation and reflection of the neutrons produced by spontaneous fission or cosmic ray induced fission.

The relatively large size and spherical shape of the uranium bearing region reduced buckling. When the surrounding porous rocks were saturated with water, the subsequent moderation and reflection allowed the reactor to achieve criticality. It is likely that criticality was not continuous. As the reactor power increased, the water moderator would heat, reducing its density and its

effectiveness as a moderator and reflector. This process, known as a negative temperature coefficient, helps to control power during transient conditions in manmade nuclear reactors.

If sufficient power was produced the reactor would have lost moderation and reflection, resulting in a shutdown. Until short lived fission product poisons decayed away, even immediate re-saturation with water may not have resulted in restarting the nuclear chain reactions. Therefore, the reactor probably did not operate continuously, but at discrete intervals with the operating time determined by the power output, water supply pressure and temperature, and water flow through the reactor. The duration of the shutdown periods would have been determined by the buildup of fission product poisons and the length of time required to replace the moderator (if it boiled away) or to cool it sufficiently to resume the reaction.

In fact, a recent paper (Meshik et al, 2004) looked at the operation of the Oklo reactor. The authors deduced that the reactor likely operated cyclically, operating for a half hour until accumulated heat boiled away the water, then shutting down for up to 2.5 hours until the rocks cooled sufficiently to allow water saturation again. The authors also note that the majority of fission products from these nuclear reactions have remained in place for nearly 2 billion years, in spite of their location in fractured, porous, and water-saturated sandstone for most of that time.

In all, the Oklo reactor is thought to have operated for a period in excess of 150,000 years, based on the quantity of fission products present. The total neutron fluence is thought to have been about 10^{21} neutrons per square cm over the life of the reactor, producing a total of about 15 GW yr of thermal energy. During this time it consumed an estimated 5-6 metric tons of ^{235}U, and producing an equal mass of fission products (de Laeter, et al, 1980). Meshick estimates an average operating power of about 100 kW, similar to that of modern research reactors.

Large core size, a low fuel/water ratio, and the presence of fission product poisons would require a large neutron flux with respect to reactor power in order to maintain reactor criticality. Neuilly et al (1972) estimated that the Gabon reactor had a thermal neutron flux of at least 10^{9} neutrons cm^{-2} sec^{-1} and a total fluence of 10^{21} neutrons per square cm. By comparison, the complete fission of one kg of ^{235}U in a nuclear detonation would release approximately 10^{26} neutrons in approximately one microsecond through an area of approximately 200 cm^{2} (Serber, 1992), giving a neutron flux of about 10^{30} neutrons cm^{-2} sec^{-1}. Current nuclear reactors have neutron fluxes on the order of 10^{13} to 10^{14} neutrons cm^{-2} sec^{-1}. The estimates for neutron flux are, therefore, not unreasonable, given the reactor characteristics of low power and large core size.

The rocks of the Gabon reactor indicate the presence of fission product nuclides in abundances which roughly tally with those expected of ^{235}U induced fission. Most interesting are the isotopes of neodymium, which show enrichment in those mass numbers that are characteristic of uranium fission and depletion in those mass numbers which have the highest neutron capture cross sections. The distribution of these nuclides is described in Cowan (1976) and in Neuilly et al (1972). Also convincing is the enrichment in ^{235}U and the presence of ^{232}Th noted in some sections of the reactor zone. The ^{235}U is thought to represent areas in which neutron capture by ^{238}U produced ^{239}Pu which subsequently decayed to ^{235}U. Thorium is thought to result from the reaction:

$$^{235}U + n \rightarrow ^{236}U \rightarrow ^{232}Th + \alpha$$

Other compelling evidence of induced uranium fission is the negative correlation of uranium content versus ^{235}U abundance through this deposit. This indicates that the areas of highest uranium content underwent the greatest depletion of fissionable material, which would be expected in a nuclear reactor.

Conclusions

The reactor zones found in Gabon have the requisite physical and nuclear characteristics to form a self-sustaining chain reaction, given the abundance of ^{235}U present nearly 2 Ga. The compact mass of the reactor zones would have been conducive to minimizing buckling and maximizing thermal neutron utilization in the uranium while the surrounding sandstone and conglomerate would provide ample water to moderate and reflect neutrons, as is the case with artificial reactors today. The inventory of fission products and the slight excess of ^{235}U noted in some zones support the conclusion that induced fission of ^{235}U and neutron capture by ^{238}U occurred in this area.

It also seems likely that other natural reactors were operational in the past. Other parts of the world have large, high assay deposits of uranium mineralization in sedimentary strata, so the circumstances which led to the formation of the Gabon reactor may not have been unique. It seems safe to assume that this process may have taken place throughout the history of the earth. Indeed, there are hints that a natural reactor was operational in the Colorado Plateau, based on a slight depletion of ^{235}U in ore specimens there (Cowan, 1976). It may be that our knowledge of natural nuclear reactors is limited primarily by our explorations to date.

Finally, although the existence of transuranic elements was predicted by Goldschmidt, the common conception is that uranium is the heaviest natural element. Any nuclear reactor, however, produces transuranic elements via the neutron capture reactions mentioned above. Therefore, uranium may need to relinquish this position to plutonium or heavier elements.

Acknowledgements

Many thanks to the following persons:

- Gunter Faure, my MS advisor for whose class this paper was originally written
- Bruce Busby, for translating this into HTML and maintaining the posting on-line
- Jaroslav Franta, for providing feedback, suggestions, and corrections to the original (as well as the drawing of an actual fuel assembly)
- Graham Cowan, for helpful comments and corrections to the original

References

Cowan, A *Natural Fission Reactor,* Scientific American, 7/76: 3647

de Laeter, J.R., Rosman, K.J.R, Smith, C.L., *The Oklo Natural Reactor: Cumulative Fission Yields and Retentivity of the Symmetric Mass Region Fission Products,* Earth and Planetary Science Letters, 50(1980) 238246

Faure, G., *Principles of Isotope Geology*, John Wiley and Sons, 1986

Janueczek, J. *Mineralogy and Geochemistry of Natural Fission Reactors in Gabon*; Chapter 7 in Reviews in Mineralogy vol. 38, Uranium: Mineralogy, Geochemistry, and the Environment, Burns PC and Finch R (eds); Mineralogical Society of America; pp 321-392. 1999

Knief, R. A., *Nuclear Engineering: Theory and Technology of Commercial Nuclear Power,* Hemisphere Publishing Corporation, 1992

Kuroda PK; *On the Nuclear Physical Stability of the Uranium Minerals*. Journal of Chemical Physics vol 25, pp 781-782. 1956

Meshik, A.P. et al., *Record of Cycling of the Natural Nuclear Reactor in the Oklo/Okelobondo Area in Gabon*. Physical Review Letters vol 93, No. 18. 2004

Meshik AP et al., *Anomalous xenon in zone 13 Okelobondo*. Geochemica et Cosmochimica Acta vol. 64 no. 9, p 1651-1661. 2000

Nero, A.V., A *Guidebook to Nuclear Reactors,* University of California Press, 1979

Neuilly, M., et al, *Evidence of Early Spontaneous Chain Reaction found in Gabon Mine,* excerpts from press conference regarding Geological and Mineral Documentation published by Commissariat a l'Energie Atomique, 1972

Paxton, H.C. and Pruvost, N.L., *Critical Dimensions of Systems Containing* ^{235}U, ^{239}PU, *and* ^{233}U, Los Alamos National Laboratory, 1986

Serber, R., *The Los Alamos Primer: The First Lectures on how to Build and Atomic Bomb,* University of California Press, 1992

This document was originally written in 1996 as a paper for a graduate class in environmental isotope geology, taught by Gunter Faure. Several e-mail correspondents have noted errors in the original paper; this revision accounts for their comments as well as for research papers published between 1996 and April 2005.