The Oklo Phenomenon

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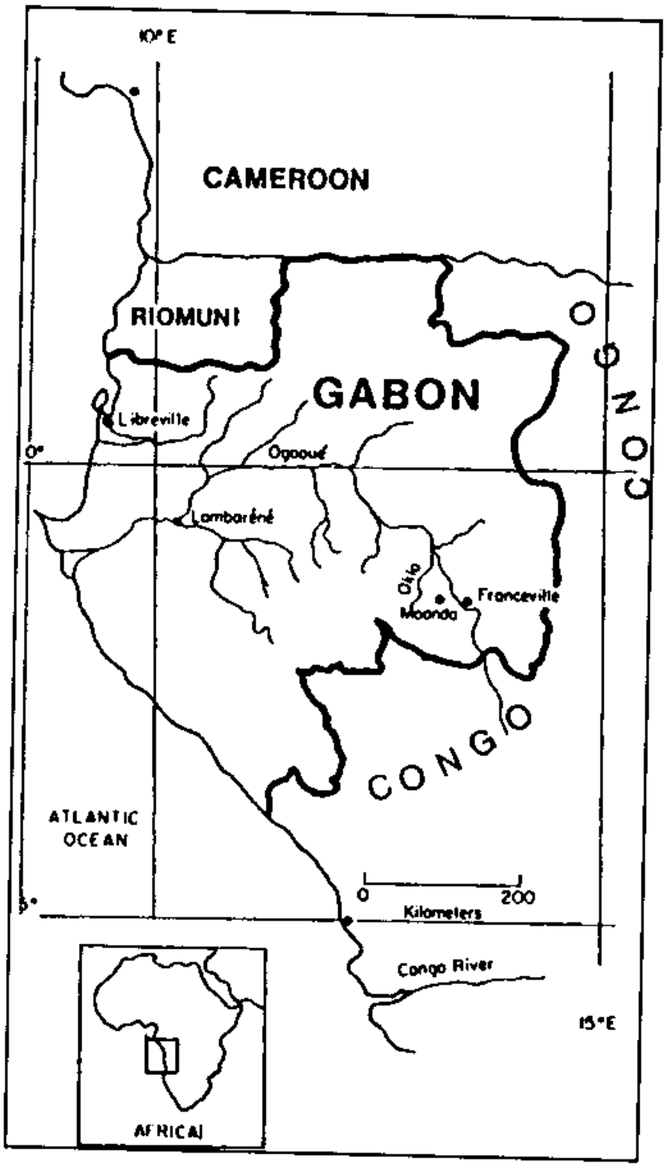
I. Discovery

A. Postulated as early as 1939, the existence of naturally occurring nuclear reactors was mere speculation until 1972.

B. In May 1972, H. Bouzigues, a staff member at the uranium fuel enrichment plant in Pierrelatte, France, was measuring very carefully the ratio of fissile ^{235}U to non-fissile ^{238}U in the UF_6 gas processed at the plant. Instead of the usual 7.20 atoms ^{235}U per 1000 atoms total uranium he found 7.17. Since all the uranium in our solar system was believed to have come from the same source, it all should have the same isotopic ratio, namely 0.720% ^{235}U. To account for this anomaly they checked:

1. Contamination at the processing plant by uranium tailings depleted in ^{235}U,
2. Contamination with ^{238}U, or ^{235}U depletion somewhere between the uranium mine at Oklo and the plant,
3. Actual ^{235}U depletion in the ore itself.

C. Bouzigues did not make a mistake. The ore itself was depleted in ^{235}U. A careful check of records showed that from 1970-1972, the 700 tons of uranium mined from the Oklo mine in the Republic of Gabon, Africa was short about 450 pounds of ^{235}U.



Figures 1 and 2: Show a map of Gabon (left) and a photograph of the mine (right).

D. Checks of the geologic survey cores previously drilled at the mine site showed ^{235}U concentrations as low as 0.440%. Later samples from Oklo gave ^{235}U isotopic abundances as low as 0.296%.

E. If all the uranium in the solar system initially had the same ^{235}U/^{238}U ratio, why was the ratio at the Oklo mine different? The only explanation was that nuclear reactions had changed the uranium into different elements. Fission, like that in man-made nuclear reactors, was the most likely reaction. Uranium-235 nuclei may be split, or fissioned, by low energy (slow or thermal) neutrons. Uranium-238, however, is fissioned best by high energy (fast) neutrons. If most of the neutrons present in the reactor were thermal neutrons, a much larger percentage of the ^{235}U would be transmuted into other elements, compared to the transmutation of ^{238}U, and the ore would be depleted in ^{235}U. If the neutrons were primarily high energy, the ore would have an anomalously high ^{235}U concentration since at higher energies the fission probability is more nearly equal and there is 140 times more ^{238}U in the ore. Thus, more ^{238}U would be fissioned.

When the necessary measurements were made at Oklo in the summer of 1972, the ore was found to contain numerous other distinctive isotopic and elemental fingerprints of nuclear fission.

Moreover, from 10^5 to 10^6 times too many fission products were present in the ore to be accounted for by spontaneous nuclear fission of uranium nuclei.

The conclusion was that nuclear reactors had been created by natural forces in the uranium ore deposits at Oklo.

II. Geologic Formation of the Uranium Deposits

A. The Oklo stratum, including the uranium ores, was deposited from Pre-Cambrian rocks about two billion years ago. (The Cambrian period began 600 million years ago).

B. Concentration of uranium by rivers:

1. The present Oklo mine site was, two billion years ago, part of a coastal river delta in

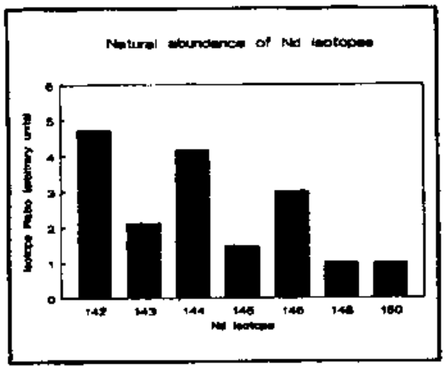


Figure 3

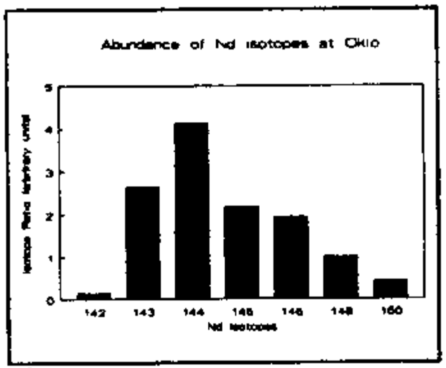


Figure 4

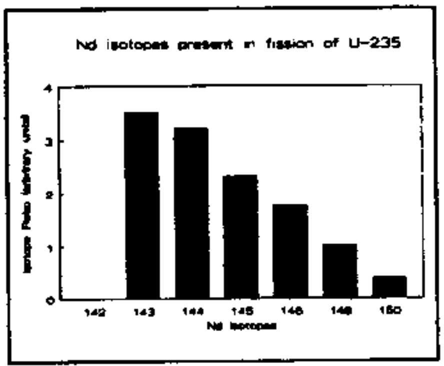


Figure 5

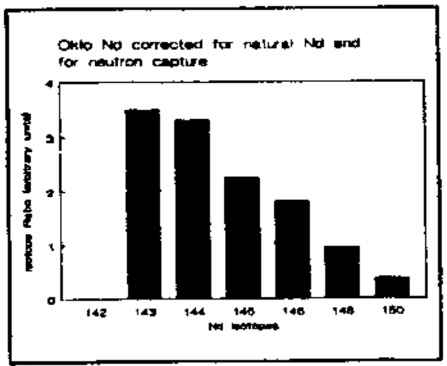


Figure 6

Figures 3-6: Relatively scarce in nature (0.028 g Nd/kg earth's crust), Nd has 7 stable isotopes which are present in the relative concentrations shown in Fig. 3. Not only is Nd more plentiful at Oklo, but as seen in Fig. 4, the Nd at Oklo has a entirely different isotopic distribution. In fact, when corrected for the isotopic composition of natural Nd and for neutron capture, the Oklo Nd (Fig. 6) has the same isotopic distribution as Nd produced in ^{235}U fission reactions (Fig. 5).

West Africa. As silt was deposited in the delta, the shore grew outward, and today the mine site is about 400 km inland.

2. The uranium is believed to have been deposited by the following mechanism:

   a. Geologic material originally spread over large areas in primarily igneous rock formations was weathered.

   b. The uranium then was concentrated as placer deposits in stream and river beds like gold in California.

   c. About two billion years ago, algae capable of photosynthesis evolved and began to produce oxygen. The uranium placer deposits were oxidized by the O_2 to more water soluble U(VI) compounds.

   d. The oxidized uranium compounds were dissolved and carried downstream to the river delta where they encountered strongly reducing conditions in the oxygen depleted organic slime previously deposited in the delta.

   e. The reduced uranium precipitated in the delta.

C. Ore deposition and uplift:

1. Reduced uranium minerals were deposited uniformly in a sandstone sandwich on a granite plate.

2. The rivers continued to deposit more silt which compacted the uranium under increasing amounts of sediment.

3. Uplift began in the granite bed below the sediment to the west of the current mine.

4. Cracks developed as the uplift continued until the ore vein was tilted at 45°, as it is now.

5. Water filled the cracks and concentrated the uranium in small pockets within the ore bed. Nuclear chain reactions began in local uranium pockets when the uranium became sufficiently concentrated.

6. Over the last two billion years, the surface above the uranium deposit eroded until the ore became easily accessible from the surface.

III. Sustainable Nuclear Chain Reactions:

A. The neutron is the key to nuclear chain reactions. A fission chain reaction can only be sustained when, on average, every fission causes at least one other nucleus to fission. When a uranium nucleus splits, two fragments usually of unequal size, two or three neutrons, and energy are released. If an average of one neutron from every fission causes another nucleus to fission, a chain reaction occurs. If the likelihood of a fission causing another fission, which can be called k_{eff}, is less than one, then the reaction dies out; the lower k_{eff} is, the faster the chain reaction stops. A k_{eff} of unity is the ideal for sustaining nuclear reactions. However, when $k_{eff} > 1$, there is an exponential increase in the number of fissions and a nuclear explosion can follow if k_{eff} remains >1.

B. Any natural reactor should meet the following requirements in order to have $k_{eff} \geq 1$.

1. A moderator (H_2O) must be present to slow down the energetic neutrons generated in fission. The fast neutrons are slowed down, or thermalized, in collisions with the moderator. The hydrogen atoms in H_2O are good moderators because they have about the same mass as neutrons.

   Thermal neutrons are 500 times more likely to fission an atom of ^{235}U than fast neutrons are. Also, taking the isotopic ratio of uranium two billion years ago into account, thermal neutrons fissioned 1000 ^{236}U atoms for every ^{238}U atom fissioned.

   At 3% ^{235}U, the conditions that existed two billion years ago (see Fig. 7), at least 6% H_2O by weight was required to moderate the Oklo reactors.

2. The total uranium concentration must exceed 10% to have enough uranium to sustain the chain reaction.

3. In water moderated reactors, ^{235}U must be present in at least 1% isotopic abundance to have enough fission by the moderated neutrons. Because deuterium is much less likely to absorb neutrons, D_2O moderated reactors can operate with isotopic concentrations below 1%.

4. The uranium vein must be at least 1/2 meter thick to ensure that few neutrons escape before being used in fission reactions.

5. Reactor poisons, or elements that very readily absorb neutrons, must be present, but in rather low concentrations. The neutron flux would never have been high enough if there were too many poisons. However, if there were too few poisons present, then k_{eff} would be greater than one.

   Usually, poisons are rendered harmless once they absorb a neutron. Thus, poisons may be "burnt-up" in a reactor. At Oklo it appears as though poisons, especially boron, kept the neutron flux in check by absorbing neutrons until enough uranium had been used up in fission so that neutron production was lowered. Poisons are also formed in the fission process.

IV. The Oklo Reactors

A. Nine reactor zones have been identified. (Only six were initially discovered in 1972.)

B. Physical data:

1. Uranium minerals, mostly uranite UO_2, are present in veins approximately 1 m thick and 10–20 m across in reactor zones.

2. Uranium concentrations in reactor zones typically vary from 25–40%, but can be concentrated in lenses of 50–60% uranium with the concentration rapidly falling off to <10% at the edge of lenses. (A lens is simply a lens shaped deposit.) These concentrations are at least 50 times higher than in the uranium containing minerals surrounding the zones.

C. Isotopic depletion is a function of uranium concentration; the greater the uranium concentration, the greater the percentage ^{235}U depletion. In other words, these natural reactors ran more efficiently the greater the uranium concentration.

D. Two billion years ago, ^{235}U was present in approximately 3% isotopic abundance (see Fig. 7) which allowed chain reactions with H_2O moderation.

E. Water of crystallization in the minerals and water in the fractures created by uplift moderated the energetic neutrons produced in ^{235}U fission and made nuclear chain reactions possible. When the temperature became too high, the water boiled away, and shut down the reactor. This helped to control k_{eff}, since the reactor temperature is proportional to k_{eff}.

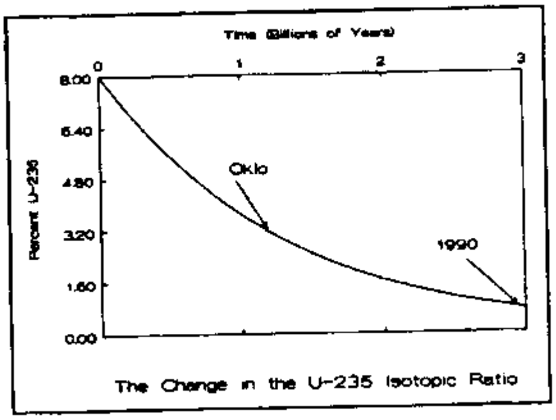


Figure 7: The ^{235}U/^{238}U ratio over the last 4 billion years. Because the much more easily fissioned isotope, ^{235}U, has been present in less than 1% abundance for the last 400 million years, natural nuclear reactors must be more than 400 million years old.

F. The neutron flux was fairly low:

1. Only 50–65% of the fission reactions at Oklo came from ^{235}U originally present in the ore bed.

2. About 6–10% of the fissions came from ^{238}U and ^{239}Pu created by neutron capture in ^{238}U. (Individually, each of the nuclides accounted for 2–8% of the fissions).

3. The remaining fissions (25-45%) came from ^{235}U produced in the cores by neutron capture in ^{238}U and subsequent radioactive decay according to the scheme:

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

4. Some ^{235}U became ^{232}Th by neutron capture and alpha decay as follows:

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

   With a 26 million year half-life, all the ^{236}U has decayed to ^{232}Th. (Ten half-lives is taken as the time required for the complete decay of a nuclide. At this time 0.1% of the original nuclide remains.) Since ^{232}Th has a 14 billion year half-life, nearly all the ^{232}Th produced at Oklo is still present.

Table 1. A Comparison of Oklo and TRIGA Research Reactor

Oklo Natural Reactors	Man-made Reactors (TRIGA)
1. Operated 2x10^{9} years ago when ^{235}U was present in 3% abundance.	Use uranium enriched to 3% ^{235}U
2. Moderated by H_2O	Moderated by H_2O
3. Possible core temperature as high as 300°C, but probably lower	Operate under atmospheric pressure at temperatures less than 100°C
4. Neutron flux (Zone 9): 5.2x10^{7} n/cm^{2}/s	Flux in a 10kW reactor: 4x10^{11} n/cm^{2}/s
5. Power output (Zones 1–6): between 10 and 100 kW	Power output ranges up to 100 kW
6. k_{eff} initially controlled by poisons present in the ore	In controlled amounts, poisons, like Sm in TRIGA, steady the neutron flux

5. If the neutron fluxes were high, much more ^{239}Pu would have been fissioned. Instead, it had time to β decay with a 24,110 year half life.

G. Statistics for the six reactor zones initially discovered:

1. Nuclear reactions started 1.7–1.9 billion years ago and ran off and on for 100–800 million years. (This figure is uncertain because of the uncertainty in the amount of water that was present in the ore).

2. It used six tons of ^{235}U (which left behind six tons of fission products), and produced 2.5 tons of ^{239}Pu.

3. The power output was between 10 and 100 kW, and 15,000 MW-year.

4. The temperature during operation could have been as high as 200–300°C. The pressure exerted on the ore bed by the sediment above it would have kept the H_2O from vaporizing at 100°C.

5. The neutron flux in reactor zone 9 was 5.2x10^{7} n/cm^{2}/s.

V. Oklo Provides Insight into the Disposal of Nuclear Waste

A. Many nations, including the U.S., are considering the disposal of wastes from nuclear reactors in stable and dry geologic formations such as salt beds, clays, and granite.

B. Remember that Oklo is a sandstone deposit with water present.

C. Table 2: Nuclides of Major Concern in Reactor Wastes

Nuclide	10 Half-lives (yrs)	Time of Concern (yrs)
^{90}Sr	290	0 – 300
^{137}Cs	302	0 – 300
^{99}Tc	2,130,000	0 – 1,500,000
^{241}Am	4320	0 –5000
^{243}Am	73,700	0 – 90,000
^{244}Cm	181	0 – 200
^{239}Pu	241,000	0 – 400,000
^{240}Pu	65,600	0 – 80,000
^{237}Np	20,140,000	0 – >20,000,000
^{229}Th	73,000	1000 – >10,000,000
^{226}Ra	16,000	5000 – 2,000,000

D. Two and a half tons ^{239}Pu and six tons of fission products were created in the six reactor zones originally discovered at Oklo.

E. In two billion years the ^{239}Pu, now detected as ^{235}U, did not leave the Oklo reactor cores. (The Pu never migrated more than 10 cm from its formation site).

F. In fact, no actinide (Th, U, Np, Pu, Am, Cm, etc.) and none of the chemically similar lanthanides migrated out of the cores. (Actinides pose the long term radiation hazard in reactor wastes).

G. Strontium-90, one of the major short term radiation hazards, was also retained for at least 300 years, by which time most of the ^{90}Sr had β– decayed to ^{90}Zr. Numerous other radionuclides were simply redistributed in the cores with no appreciable migration out of the reactor cores.

H. There was slight migration and significant redistribution within the cores themselves of Tc, Pb, and Zr.

I. Unfortunately, the alkali elements (which includes ^{137}Cs), Ba, I, Kr and Xe did leave the cores. (The good news is that most of these are short-lived nuclides and the canisters the waste is buried in should contain the waste without leakage for at least 3000 years).

VI. Recent Developments

A. In 1987, Hishita and Masuda (*Naturwissenschaften*, 74, 241-242 [1987]) suggested, based on the 1000 fold variations in the ^{235}U/^{238}U ratio over very small distances (0.01 mm), that ^{239}Pu did move about quite readily, but only over small distances within the reactor cores, not outside, and, on decaying, formed microscopic areas of ^{235}U enrichment.

B. The same data, however, was interpreted by Harms (*Naturwissenschaften*, 75, 47-49 [1988]) as an indication that the Oklo reactors were highly dynamic, and perhaps even chaotic systems with widely varying neutron fluxes. Wildly varying spatial and temporal neutron fluxes would account for Hishita and Masuda's observations without plutonium migration.

VII. For Further Reading

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