SUPPLEMENTARY INFORMATION A

Natural nuclear reactors

A1. Evidence of criticality in Nature

Fourteen Nuclear Reaction Zones were observed in the Oklo deposit and a fifteenth one in the Bangombé deposit located 30 km southeast of Oklo[GAU86,GAU96] (see Fig. 1). Uranium from these 2.0 Ga old deposits is concentrated in the lower 10 m of the sandstone reservoir (red "C1 layer" in Fig. 1) which lies 100 to 1000 m above organic-rich black shales (blue in Fig. 1).

The uranium deposit was formed 2.0 Ga ago, during the diagenetic history of the basin, at a depth of 2 km and a temperature of 200 °C[GAU96,GAU89,NAU91]. Migration of uranium in the basin was due to its dissolution through oxidation by diagenetic fluids migrating in the fracture network and the porosity specific to the sandstone of a petroleum rock reservoir[GAU96,NEL03]. Mineralization as uranium oxides (uraninite) occurred when oxygen-bearing fluids met reducing environments, i.e., the petroleum traps. Due to the inhomogeneous character of such a natural system, the very heterogeneous uranium deposit averages 0.4% by weight over the whole deposit with some rich zones showing up to 15% by weight[GAU96,GAU89].

The reaction zones (RZ) are numbered in the order of their discovery. RZ1 and 2 owe their discovery on June 6, 1972 to the an anomalous ^{235}U isotope concentration found in a routine analysis performed in the Cadarache laboratory of the French Atomic Energy Agency (CEA): a sample of uranium contained 0.7171%[BOD72,NEU72] of ^{235}U whereas the present-day natural proportion is 0.7202%.

Figure 1 | **Localisation of the OKLO reactors.** Geological map of the Oklo area of the Franceville basin (left). Uranium deposits are indicated by red stars on the map (from ref. BEN13). The drawing on the right locates reactors 1 to 9 on a 3D perspective of the Oklo deposit.

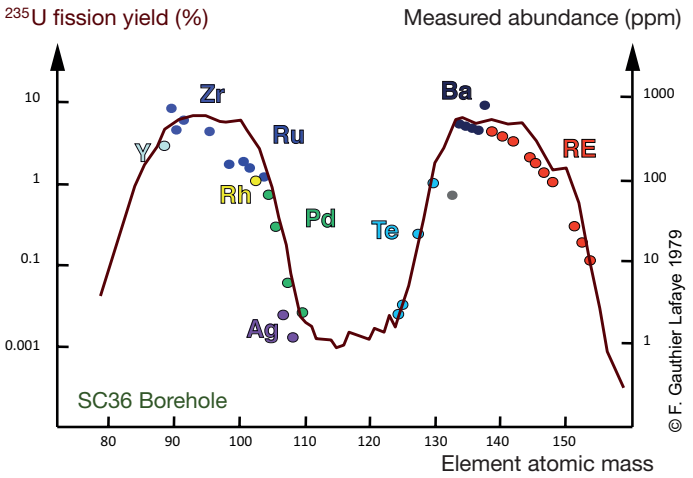

Figure 2 | **Evidence of sustained fission in Nature**. Elemental analyses (dots) from borehole SC36 (RZ2) compared to the ^{235}U fission fragment yields (solid curve) show a nice correlation[GAU79].

Even a lower concentration was observed a few days later. The evidence for sustained fission came from the good correlation between the fission-derived elements present in the Oklo rocks and the known ^{235}U slow neutron-induced fission fragment yields[NEU72] (Fig. 2). It is worth nothing that due to the age of these reactors all fission fragments have been transformed through radioactive decay to their ultimate stable descendants.

The Oklo reactors were discovered at depths ranging between 20 and 200 m below the current surface[GAU89,TOU96]. Fig. SA 3 shows the Oklo mine soon after discovery of the "Oklo phenomenon", when the first exploration work started. The top of RZ2 can be seen in Fig. 3 by the arrow in a delimited

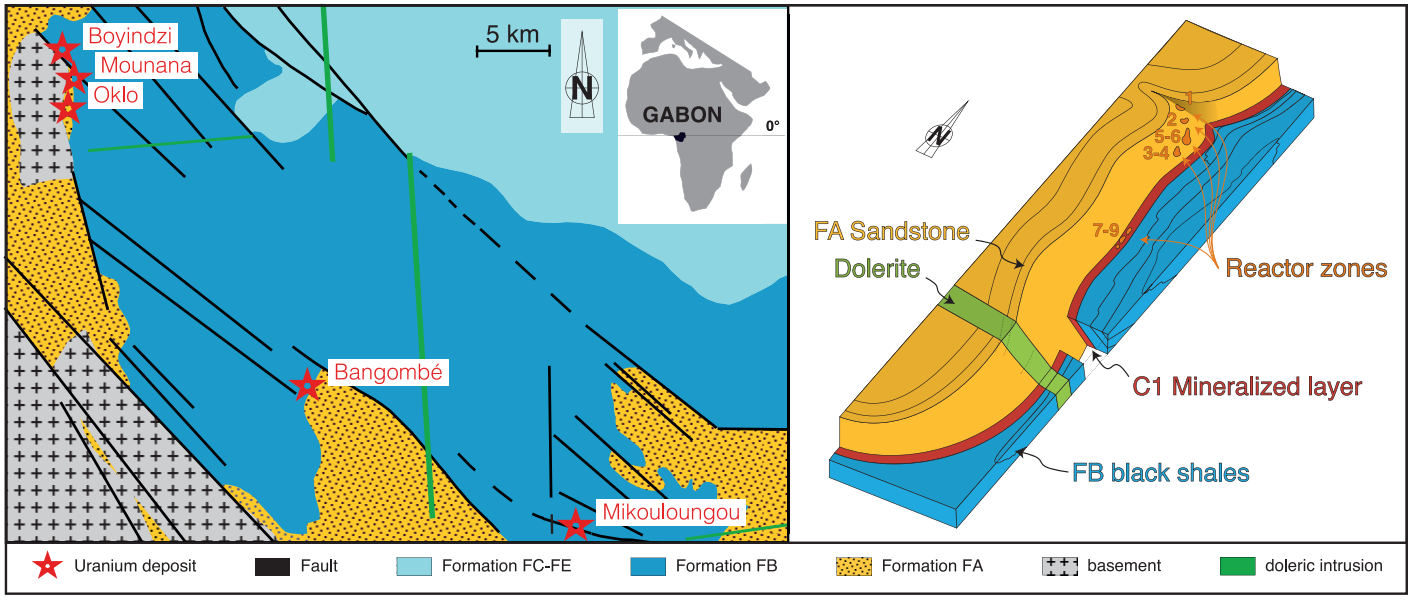

Figure 3 | View of the open pit of Oklo during exploration of the reactors (1973). The top-left limit of the C1 uranium-rich layer is indicated by the yellow dashed line. An arrow points to the top of Reactor 2 to the right of the person standing in the area cleared for scientific studies.

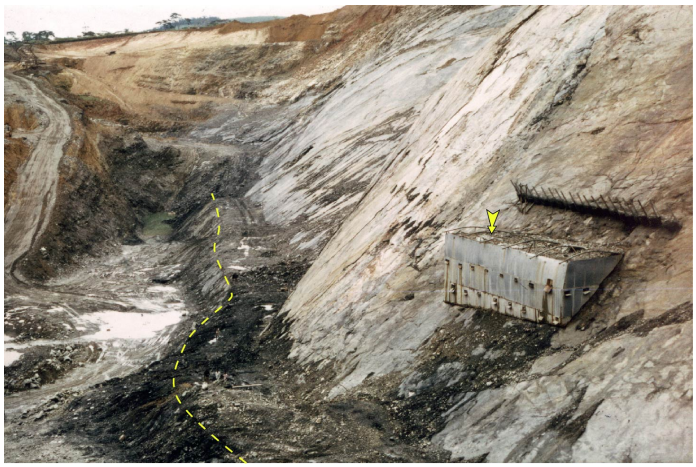

Figure 4 | Same view after mining out reactor 2. The metallic container pointed to by the yellow arrow hosts a slice of reactor 2 for further scientific studies. The top-left limit of the C1 layer ready for commercial exploitation is marked by a dashed line.

Figure 5 | Impact of reactor operation on underlying sandstone of reactor 1 (left) and reactors 7-9 (right). A major effect of hydrothermal alteration on the surrounding rocks during fission operation is dissolution of quatz. The shape of the dissolution zone mimics perfectly the shape of reactor 1 which was situated just above the sandstone layer's upper surface. For reactors 7-9, the dissolution zone was controlled by fractures crossing the reactors

cleared area. On the right part of this picture, one see the sandstone of the FA formation dipping 50° eastward. This layer, together with the overlying C1 mineralized layer and the reactors, were less deformed at the time the reactors were operating. Their present orientation is due to a late tectonic event.

Fig. 4 shows the same mine a few months later. One clearly sees the 8 m thick black C1 mineralized layer ready for exploitation. The metallic container, called "the bunker" was built to preserve part of RZ2 for further scientific studies. It was mined out in the 90s, before the flooding of the open pit when mining work ended. The small irregularities seen in the sandstone above the container are due to hydrothermal dissolution of silica during the reactor operation. Such thermal-induced dissolution effects on underlying sandstone layer can also be seen on Fig. 5.

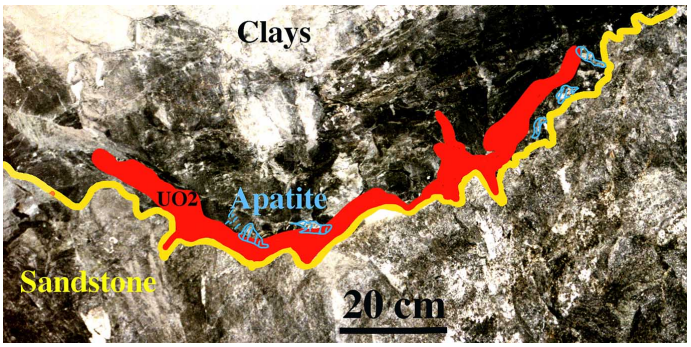

Figure 6 | Vertical structure of reactor 10 seen in the mine of Oklo. The core of the reactor is depicted in red. It is surrounded by sandstone (yellow) and clays (above). Note the occurrence of aluminum phosphate (in blue) which trapped plutonium during or soon after fission operation[BRO93].

In the field, reaction zones present specific features relative to the surrounding rocks. They were observed as 5 to 20 cm thick layers with up to 60% uranium by weight surrounded by clays[GAU86,GAU96,GAU89]. The clay envelopes result from the hydrothermal alteration of the surrounding rock by fluids heated by the fission reactions. Such clays never occur in a "normal" mineralized ore with uranium showing its normal isotopic composition. Fig. 6 shows the lower end of RZ10 that had by far the biggest core. The uranium-rich zone became rather thin after the compaction process induced by core operation. Above it, a quite thick clay layer can be observed.

The reactor of Bangombé has been preserved together with the deposit for future scientific studies[GAU86,GAU96,GAU89]. It is today only 12 m under ground level. Fig. 7 shows a view of the Bangombé deposit during the drilling exploration of the reactor. It is of primary importance to continue preserving this site as the unique remaining natural nuclear reactor on earth.

Figure 7 | View of the Bangombé reaction zone during the drilling exploration (1996). It is the last natural nuclear reactor known. There are no pictures of this reactor since it was preserved. These boreholes produced the evidence for the existence of this reactor.

A2. Illustration of the different reactor sizes

To obtain a good understanding of the Oklo phenomenon, first of all it is important to observe that the reaction zones show quite different sizes. Table 1 gives the size of the reactors and various nuclear parameters as they were observed. Of the fourteen reactors at Oklo, RZ1, RZ2 and RZ10 are the largest ones. They involved a much larger volume of uranium than typical present-day nuclear power plants. Most of the other reactors are quite small, initially making it difficult to explain how they were able to achieve ignition. The observed average thickness of the uranium-rich layer of these cores is typically 7 cm with variations ranging between 2 to 20 cm. Including the clay generated by the core operation, these thicknesses can go up to a few meters for the biggest cores.

Extrapolation of the amount of ^{235}U back 1.95 Ga ago has to consider the influence of its radioactive decay as well as the ^{235}U breeding effect in the core. By considering only the half-life of ^{235}U (0.703 Ga), the present-day ^{235}U concentration in natural uranium should be multiplied by 5.1 to get its value at the start-up time of the Oklo phenomenon (see Fig. 8). On the other hand, estimation of the total ^{235}U burnt by the sustained nuclear fission reactions is obtained by considering a mean ^{235}U restitution factor of 0.48[NAU91,RUF76]. This factor includes the radioactive decay of ^{239}Pu (24110 y) produced by neutron capture of ^{238}U during operation of the reactor cores. Almost 90% of the ^{235}U was restited after ten half-lives of the ^{239}Pu.

Measurement of the isotopic distribution of fission products, shows that about 50% of the ^{235}U burnt was generated in the core through ^{238}U activation and subsequent radioactive decay of ^{239}Pu[RUF76,LOS84]. This scenario is justified by the fact that the low flux of neutrons generated at the Oklo reactors produces less fission of ^{239}Pu.

The estimated total released energy is calculated by considering that the fission of 1g of ^{235}U produces 0.95 MW·d, since one fission of uranium releases approximately 200 MeV. Equivalent power can also be found by dividing the total released energy by the estimated operating duration. This deduced power ranges between a few kW and a little more than 10 kW. This is at least 10^7 times less than generated by present-day nuclear power plants. Nevertheless, fuel burn-up went up to 15 GW·d/ton at Oklo. It is intriguing that neither the fractional amount of ^{235}U burnt nor the observed mean power are correlated with reactor size!

When the reactors were first observed, ignition could be explained only for the biggest reactors like RZ1, RZ2 and RZ10[NAU91]. The question of ignition of the smallest reactors remained open until recent work on RZ9 that finally gave an explanation for the occurrence of ignition in such small reactors[BEN13,BEN11a,BEN11b].

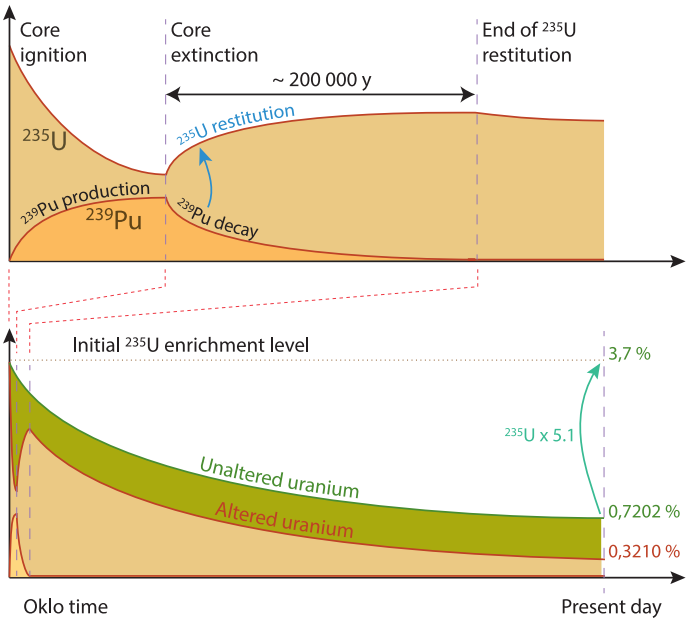

Figure 8 | ^{235}U timeline of Oklo natural reactors. One can see the ^{239}Pu contribution to the core and to the restitution of some ^{235}U during and after reactor operation.

Table 1 | Geometry and physical characteristics as observed in 1975 for some Oklo reaction zones.

Reaction Zone	U content[NAU91]		^{235}U amount		Reaction Zone extension	Estimated power			Burn-up (GW·d/ton)
	Mass[NAU91] (tons)	Mass fraction (% wt.)	Missing ^{235}U (kg)	Burnt ^{235}U (in kg 1.95 Ga ago[NAU91])	Maximum ellipsoidal Width[GAU76,GAU79] X(m) x Y(m)	Released Energy[NAU91] (GW·d)	Operating Duration[GAU96, BLA96] (10^3 y)	Mean power[NAU91] (kW)	
RZ1	230-240	40	200[NAU91]	2012	12 x 40	$1.9x10^3$	-		8.1
RZ2	200	50-60	200[NAU91]	2012	12 x 30	$2.0x10^3$	~800	6.5	9.5
RZ9	44	25-30	26.4[NAU91]	265	7 x 12	$2.5x10^2$	~220	3.2	2.8
RZ10	340	50-60	90[BLA96]	905	20 x 50[BLA96]	$8.6x10^2$	~200	11.8	2.5

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