

The Ranger uranium mine in northern

When Enrico Fermi and his team constructed their nuclear fission reactor at Stagg Field in Chicago in 1942 they would have assumed that it was the first time a fission reaction had taken place on the planet. But, as Mike Follows explains, Nature beat them to it - and by almost two billion years.

There are three principal isotopes of uranium, summarised in the table. U-235 is the most valuable as it is the only one that can sustain a nuclear chain reaction, useful for generating power. But everywhere you look - the Earth's crust, the Moon, even meteorites - its natural abundance is only 0.7202%. This means that only seven in every 1000 uranium nuclei dug out of the ground are U-235. Samples of uranium are routinely tested. This is to ensure that none of the mined U-235 has already been used (e.g. in a power station) or been illegally diverted to make nuclear weapons.

isotope	natural abundance (%)	half-life (million years)
U-238	99.2743	4468
U-235	0.7202	703
U-234	0.0055	0.245

Three important isotopes of uranium. The natural abundance shows the fractions of each isotope found in samples from around the world, and beyond.

In May 1972 a scientist at the nuclear fuelprocessing plant at Pierrelatte in France performed a routine mass spectrometry analysis on a uranium ore sample. It had been sent from the Franceville uranium ore deposits near Oklo in Gabon, a former French colony in West Africa. It contained 0.7171% of U-235. The slight shortfall in the expected proportion of U-235 sparked a thorough investigation. Other samples were taken. About 500 tonnes of uranium had an average U-235 concentration of 0.62%, with one sample as low as 0.44%. This meant that about 500 kg of U-235 was missing - enough to make at least half a dozen nuclear weapons with the destructive power of the one dropped on Hiroshima in August 1945.

In 1956 Paul Kuroda had published calculations of what it would take for uranium ore to spontaneously undergo self-sustained fission. Did the Oklo ore prove him right?



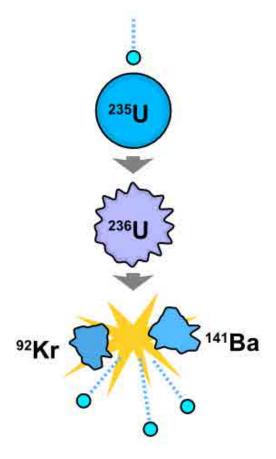
The location of Gabon in western central Africa.

Australia

**Key words** 

### What is fission?

Nuclear fission is a nuclear reaction where an unstable 'parent' nucleus, such as U-235, splits into smaller 'daughter' nuclei. Free neutrons are released, and these can induce further fission events as part of a self-sustaining chain reaction. Energy is released in each fission event.



A representation of nuclear fission. Krypton and barium are just two of the elements which may be the result of a fission event.

Water is important in a natural fission reactor. It slows down neutrons and increases the likelihood of further fissions – it acts as a moderator. (Water also has this role in a pressurised water reactor or PWR, the most common reactor in operation.) Nuclear power stations use control rods to absorb neutrons in order to slow down or stop the fission process. Without coolant or control rods there is a danger that man-made reactors can undergo meltdown as happened at Chernobyl in 1987 and Fukushima in 2011. With no control rods what is there to stop a natural reactor going into meltdown and exploding?

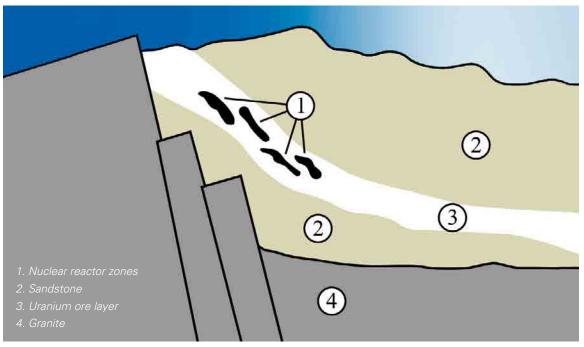
Paul Kuroda calculated that certain conditions had to be met for the operation of a natural nuclear reactor.

- A uranium deposit must exceed two thirds of a metre, the average distance that a fissioninducing neutron travels. Smaller deposits risk too many neutrons escaping before they can induce fission in another U-235 nucleus.
- The uranium ore must contain at least 10% uranium, of which at least 3% has to be U-235.
- Water must be present to act as a moderator.

There should be no significant boron, lithium or other 'poisons', which absorb neutrons and would prevent a natural reactor even starting.

## The Oklo discovery

Sixteen natural nuclear fission reactors were discovered in the Franceville basin – 15 clustered together and one about 30 km away (the Bangombé reactor). The biggest is 12 m long, 18 m deep and between 0.2 and 0.5 m wide. Because the surrounding sandstones and clays were porous, there was plenty of water present to act as a moderator.



The geological structure which made natural fission reactors possible.



The dark area on this cliff face shows the site of one of Oklo's natural reactors. Notice the small figure on the right.

But how did the uranium end up where it did? The landscape around Oklo was once a river delta. Streams would have been eroding the igneous rocks from across the catchment and heavy metals like gold would have accumulated in so-called 'placer deposits' upstream. The uranium would have remained there, quite dispersed, if it were not for photosynthesis and the Great Oxygen Event 2.3 billion years ago. This is when micro-organisms like cyanobacteria used the energy in sunlight to drive the reaction between carbon dioxide and water to make carbohydrates. This emitted oxygen gas as a waste product, which dissolved in the streams and oxidised the uranium. Uranium oxide is soluble so it was flushed down all the streams and collected at the delta where it met sediments impregnated with petroleum. This oxygen-free zone meant that the uranium oxide was reduced to uranium, which is insoluble, so it precipitated out of solution and formed lodes in the sandstone.

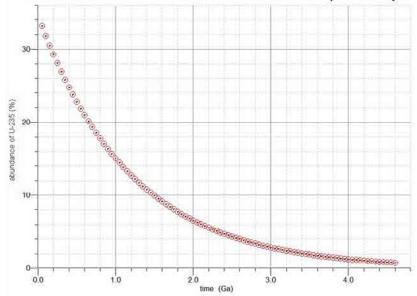
# The smoking gun

Using the relative abundances of daughter nuclei it is possible to work backwards to the parent nucleus. Scientists found stable isotopes of more than 30 elements, including neodymium, serving as a 'smoking gun' which proved that a nuclear fission chain reaction had taken place. Some zones even contained slightly elevated abundances of U-235 and this is thought to be the result of neutron (n) capture by U-238. Two subsequent beta ( $\beta$ ) decays produce Pu-239. Followed by an alpha ( $\beta$ ) decay, this produces U-235:

$$\begin{array}{c} ^{238}_{92} U \, + \, ^{1}_{0} \, n \, \longrightarrow \, ^{239}_{92} U \, \longrightarrow \, ^{239}_{94} Pu \, + \, 2 \left( ^{0}_{-1} \, \beta \right) \\ ^{239}_{94} Pu \, \longrightarrow \, ^{235}_{92} U \, + \, ^{4}_{2} \alpha \end{array}$$

But how could fission have taken place when U-235 normally needs to be enriched to account for at least 3% of the uranium? From the table on page 13 you can see that U-235 has a half-life seven times shorter than U-238 so that two billion years ago (when the Earth was 2.6 billion years old), the abundance of U-235 would have been 4%. Note that the unit of time on the graph is Ga. This stands for billions of years.





The pattern of radioactive decay of U-235. Because it decays faster than U-238, its abundance in natural uranium ores was much higher in the distant past.

## Modelling the reactor

In 2004 Alex Meshik and his team developed a mathematical model to show how the reactor behaved. It generated  $5 \times 10^{17}$  J of energy, equivalent to one 40 GW power station running for four years. But the Oklo reactors ran for 150 thousand years with a power output of 100 kW. (For comparison, 3 kW is the power output of a typical fan heater.)

The Oklo reactor would have been 'switched on' for half an hour and 'off' for at least two and a half hours. This explains why meltdown did not occur. As heat from the reactors increased, the moderator would expand, reducing the density of water molecules that could slow down the neutrons. Fewer neutrons would be moderated and this would naturally reduce the rate of fission. This process helps to control power in man-made nuclear power stations.

#### The future

The concentration of xenon (Xe-136) in the Earth's atmosphere is higher than expected because of natural nuclear reactors, suggesting that Oklo is not unique. Plate tectonics separated the African and South American continents about one hundred million years ago so we may yet find the 'fossilised remains' of natural reactors in Brazil.

Since the advent of nuclear power generation, huge amounts of radioactive gases like xenon-135 and krypton-85 have been released into the atmosphere. These gases have been found trapped in aluminium phosphate minerals at Oklo, safely locked away for at least two billion years. Perhaps, as the result of the Oklo research, we have found a way to capture radioactive gases. Perhaps we also need to re-write those textbooks which say that uranium is the heaviest naturally-occurring element; that honour belongs to plutonium.

Mike Follows is 100% Physics teacher.