Nature’s reactor: the story of Oklo

43 years ago, at the Pierrelatte uranium enrichment plant in France, a worker carrying out a routine analysis of some samples from an African mine noticed a minor discrepancy in the ratio of the uranium isotopes. This seemingly trivial observation was to lead to a stunning discovery; nature can make nuclear reactors, and had indeed done so, some two billion years before Enrico Fermi did in 1943. This is the fascinating story of the Oklo reactor.

Dude, where’s my uranium 235?

In nature, uranium is made up of three forms (known as isotopes) of differing atomic mass: uranium 234, uranium 235 and uranium 238. Of these, uranium 238 is by far the most abundant, whilst uranium 235, due to its use in nuclear reactors and weapons, is the most coveted (poor uranium 234 is present in such small amounts that it tends to get rather overlooked). Crucially, the ratio of the isotopes of uranium in natural ores is incredibly consistent, and this is used as a check to ensure that none of the precious uranium 235 goes missing.

Thus it was that when an employee at the French uranium plant noticed that there was only 0.717% (rather than the expected 0.720% - yes, that small a difference *was* significant) of uranium 235 in his sample, an investigation was begun. It was initially thought that the sample had somehow become contaminated with depleted uranium (uranium from which most of the uranium 235 has already been removed) from the same plant, but this was quickly ruled out as none was missing. The investigators eventually traced the discrepancy in the ratio all the way back through the uranium processing route to the original ore extracted from a mine called Oklo, in Gabon. They then conducted further analysis which showed that all the ore taken from the Oklo mine contained considerably less uranium 235 than expected, down to values as low as 0.44%. In total, 200 kg of uranium 235 appeared to be missing, sufficient to make half a dozen nuclear bombs! The officials in charge of the investigation at the French Atomic Energy Commission were bemused, to say the least. How could they have lost such a huge amount of such a tightly controlled substance?

Oddly enough, an explanation for the missing uranium 235 in the Oklo mine had actually been published in a scientific paper by a chemist from the University of Arkansas in 1953. His name was Paul Kuroda, and he had described the conditions that would be required to sustain fission in a natural body of uranium ore. However, to fully appreciate his conclusions, and how they relate to the Oklo reactor, we need to take a moment to consider how a fission reactor works.

Nuclear fission 101

In a nuclear fission reactor, uranium nuclei are bombarded with neutrons. This breaks them apart into smaller nuclei, and also releases a vast amount of energy and two more neutrons, which then rampage off and induce further fissions. Uranium 235 is far more susceptible to fission than uranium 238. To generate a sustainable fission reaction, the uranium 235 content must be enriched to 3% (this, indeed, is the role of uranium enrichment plants. The depleted uranium mentioned earlier is that from which this extra uranium 235 has been taken). Also, for efficient fission of uranium 235, the neutrons must have the right amount of energy. Generally the neutrons formed in fission have too much energy, and must be slowed down (or moderated) by a substance (a moderator), such as water or graphite, which is able to absorb some of their energy. These lower energy neutrons are far more likely to induce fission when they collide with uranium 235 nuclei, and thus the moderator is crucial in ensuring the continuation of the nuclear reaction, or in other words, that at least one of the neutrons released in one fission process is able to induce fission of another nucleus.

What if too many neutrons of the correct energy for fission are produced and the nuclear reaction starts threatening to get out of control? To prevent this situation arising, nuclear fission reactors are fitted with control rods. These are sticks of made of elements such as boron or cadmium that are very effective at absorbing neutrons. Insertion of these rods into the core of the reactor core serves to reduce the number of neutrons available for fission and thus slow the reaction. As the neutron absorbing elements effectively kill the reaction, they are known as nuclear poisons. In this way the fission process can be carefully and safely controlled.

A natural reactor!

So now we know how a fission reactor works, let's get back to Paul Kuroda and Oklo. Kuroda set out four conditions that had to be fulfilled for a natural, self sustaining fission reactor to occur. Firstly, there would need to be a sufficiently rich deposit of a fissionable isotope, say 10% of uranium in an ore. Secondly, there would need to be a moderator of some sort to slow down the neutrons sufficiently to induce fission. Thirdly, there would need to be an absence of nuclear poisons which would absorb the neutrons and prevent any nuclear reaction. Finally, the deposit of fissionable material would need to be sufficiently thick to prevent the escape of large numbers of neutrons, or, in other words, be wider than the average length (2/3 m) travelled by a fission-inducing neutron, in order that as many of the neutrons possible released during fission go onto be absorbed by other fissionable nuclei and trigger further fission processes.

The mystery of the missing uranium 235 was solved when physicists realised that two billion years ago, the site of the Oklo mine in modern day Gabon met all of these conditions. At that point in the earth's history, uranium would have contained around 3% of uranium 235. This may seem like an odd statement, given that I said earlier that the ratio of the isotopes of uranium in natural samples is incredibly consistent, to the point where a difference of 0.003% was big news, but actually, the ratio of isotopes of all elements does change due to radioactive decay. Uranium 238 decays more slowly than uranium 235, and thus uranium deposits have gradually, over millions and millions of years, become less and less rich in uranium 235. It was easy to envisage the presence of water as a moderator, and there was no evidence to suggest that there were large quantities of nuclear poisons like boron present in the region around Oklo. Finally, the uranium ore deposits at Oklo were larger than 2/3 m in width and sufficiently rich in uranium, so neutrons would have been easily contained within the belt of fissionable uranium fuel. Oklo thus fulfilled all of Kuroda's conditions for the emergence of a natural fission reactor.

The proof that the Oklo mine had once been the site of a series of natural nuclear reactors came rapidly. Nuclear fission reactions generate a set of products whose identity is so distinctive that they can act as a fingerprint for nuclear activity. This set of more than 30 isotopes was detected in the uranium deposit at Oklo. These initial findings gave rise to a series of intriguing questions. Why did the reactors not explode? How come the nuclear reactions were kept in check? Did the reactors operate continuously or sporadically? And, if so, how did they start and stop? The answer to these questions was provided, perhaps surprisingly, by a gas called xenon.

Trapped xenon

Xenon is extremely rare in nature, and the particular isotopes found at Oklo were a sure indicator that nuclear fission had been occurring. Intriguingly, the xenon at Oklo was not trapped in uranium ores, as would be expected if it was a direct fission product of uranium 235, but rather in grains of aluminium phosphate. Indeed, these aluminium phosphates showed the highest concentration of xenon ever found in any natural material. Another interesting observation was that the trapped xenon contained a rather different ratio of isotopes to that which is typically produced in nuclear reactors. In particular, xenon 134 and 136, known to be formed rapidly by from the uranium fission process, were missing. These findings were to prove crucial in the answering the questions posed in the previous paragraph.

Fission processes generate radioactive isotopes, which in time decay radioactively to produce further radioactive isotopes, and so on, until finally, after a period ranging from minutes to millennia, a stable isotope is formed. The xenon isotopes trapped in the aluminium phosphate were actually formed by the decay of radioactive isotopes of iodine and tellurium, sometime after the initial fission reaction. Researchers worked out that the missing xenon isotopes, 134 and 136, would have been formed about an hour and a minute, respectively, after the onset of self-sustained nuclear fission, and, as gases, would have simply drifted off into the atmosphere. This explains their absence in the aluminium phosphate. On the other hand, the processes to generate xenon 131 and 132, however, would have taken some days, but how did they get into aluminium phosphate?

Nuclear fission is, of course, a highly energetic process, releasing large amounts of heat, and presumably at a certain point this would have been sufficient to evaporate the water. Without the water moderator, the nuclear reaction would stop, unable to start again until the rocks had cooled and more water had accumulated. Some of the precursors to the xenon isotopes found in the aluminium phosphate, such as tellurium and iodine, dissolve readily in water. Scientists thus think the water transported these isotopes away from the uranium where they were originally formed, and then, as the rocks became hotter and the water evaporated, the water soluble fission products were deposited as contaminants in grains of aluminium phosphate. When the xenon was formed later, it found itself trapped within these grains, unable to escape.

By using mathematical modelling to further investigate the process of xenon gas formation within aluminium phosphate, scientists were able to deduce that the Oklo reactor operated in a cycle of 30 minutes ‘on’ followed by an ‘off’ period of at least 2.5 hours, and that this process continued for hundreds of thousands of years. This is a rather similar pattern to the one observed in geysers, which tend to heat up their water slowly before boiling it all off rapidly, and then filling up and repeating the cycle. This ‘on-off’ pattern caused by the boiling of the water moderator is likely what prevented the occurrence of a nuclear meltdown or explosion throughout the operational period of the Oklo reactor.

Not just a curiosity

The story of the Oklo reactor is not just a fascinating curiosity, but is of real interest for environmental physicists today. As we wrestle with the difficult question of how and if we can store the waste from nuclear reactors safely and indefinitely, Oklo provides us with a perfect case study. We can look at the effect of the fission reactors on the environment over a period of 2 billion years.

One of the most exciting findings is the way that nature was able to store its nuclear waste, in particular xenon, inside simple, low cost minerals that we could easily envisage using for a similar purpose.

Oklo, then, is a story worth studying, a triumph of scientific detective work in deducing just how it came about, and a rich source of lessons for the future for those trying to establish how best to store the nuclear waste that we generate today.