

# re-actiONS

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From the American Nuclear Society to teachers interested in the nuclear sciences.

## A nuclear detective story

# The Oklo “reactor”

Twenty years ago, a minor discrepancy at a French uranium enrichment plant led to an astonishing discovery: Enrico Fermi did not create the first nuclear reactor. Nature had preempted him by about 2 billion years.

Some 20 years ago, in May 1972, at the Pierrelatte uranium enrichment plant in France, an employee conducting mass spectrometric analyses of uranium hexafluoride gas ( $UF_6$ ) samples from an African uranium mine noticed a discrepancy in the amount of uranium-235 contained in the samples. In nature, uranium is comprised primarily of uranium-238 atoms; the fissionable uranium-235 atoms make up only 0.7202 percent of natural uranium. In the samples being processed, however, only 0.7171 percent of the uranium was uranium-235. The discrepancy was a tiny one, but it was one that needed to be accounted for, since uranium-handling facilities and regulatory agencies must meticulously account for all uranium-235 to assure that none is diverted for weapons purposes. The French Atomic Energy Commission began an investigation.

At first, investigators thought that the  $UF_6$  had somehow accidentally been contaminated with depleted uranium from the Pierrelatte plant (depleted uranium is uranium from which most of the uranium-235 has already been stripped). But no depleted  $UF_6$  was missing from the Pierrelatte plant, so none could have accidentally contaminated the sample.

Investigators then traced the anomaly back through all stages of uranium processing—from the original ore, mined from the Oklo mine located in southeastern Gabon, through the Mounana mill located near the mine, to a processing plant in France, to the Pierrelatte enrichment plant. The good availability of samples kept for control at every stage of handling enabled investigators to discover that all shipments from the Oklo mine showed a lower than expected level of uranium-235—some as low as 0.44 percent. Oklo mine uranium was different from natural uranium everywhere else. The new question was, why?

To explain the anomaly, several hypotheses were put forth, one of them being the possibility of induced fission. Investigators began a search for fission products—isotopes of more than 30 elements created during nuclear fission. The spectrum of fission products is so distinctive that it can serve as an unmistakable sign that a chain reaction has taken place. The presence of these fission products was detected in the uranium ore deposit at Oklo. Somehow, at some point in the uranium deposit's history, it became a “natural reactor” that went critical, burned up a portion of its nuclear fuel, and then shut down.

The next question to be resolved was, when? Now investigators turned to chemistry to find the solution.

Most nuclear power reactors rely on *enriched* uranium to operate—uranium that has had its uranium-235 content increased from the normal 0.7202 percent to about 3 percent. These reactors use natural (or “light”) water as a moderator to slow down neutrons. (Neutrons given off during fission initially move too fast to split other nuclei, but if the neutrons can bounce off the atoms of a moderator, this will slow them down to a speed at which further fission is possible.) It is possible to run a reactor with natural uranium—the Canadian Candu reactor uses natural uranium—but such a reactor requires a special design and must have heavy water—water high in deuterium (the isotope of hydrogen that has an atomic mass of two)—to act as a moderator. Investigators felt that it was extremely unlikely that nature would have provided both the special design and the heavy water.

The ratio of uranium-235 to uranium-238, however, has not been constant throughout the history of the earth. The half-life of uranium-235 is about 700 million years. The half-life of uranium-238 is about 4.5 billion years. When the earth was formed several billion years ago, natural uranium was about 17 percent uranium-235. But since that isotope decays faster than uranium-238, its ratio will decrease over time. Two billion years ago, the ratio of uranium-235 to uranium-238 was about 3 percent, just right for a light-water-moderated chain reaction.

In addition to a certain ratio of uranium-235, however, a chain reaction needs a certain concentration of uranium in the ore body—at least 10 percent, and the ore itself must be concentrated in seams at least half a meter thick. In a sustained chain reaction, the fissioning of a uranium-235 nucleus results in the prompt emission of 2.5 neutrons; one of these must be absorbed and must induce fission in another nucleus. The rest can be absorbed elsewhere or can escape. If the ore seam is too thin, too many neutrons will escape, and the chain reaction will not be sustained.

Another requirement for a self-sustaining chain reaction is water. The Oklo deposit was probably saturated with groundwater. In fact, this saturation may have originally overmoderated the neutrons, but if the ore became chain-reacting under these conditions, the heat created would have evaporated some of the water, eventually leading to optimum moderating conditions.

The final requirement for a chain reaction is the absence of elements that strongly absorb neutrons—neutron “poisons,” as they are known. These elements include boron, lithium, and many of the rare earths. There is no evidence that excessive amounts of these elements existed in the ore before the reaction began.

These conditions must have varied during the lifetime of the reactor, changing its rate of fission and eventually leading to its final shutdown. And over the years, as the reactor “operated,” the amount of uranium-235 decreased, leading to depleted ore two billion years later when it was mined. Investigators estimate that the reactor must have operated—off and on—for at least 150,000 years, but best estimates suggest that the actual duration was on the order of hundreds of thousands of years.

Were these conditions unique to the Oklo deposit? Investigators do not

think so. Other natural reactors may have operated in the same time frame. In view of the accidental way the existence of the Oklo reactor was discovered, one could conclude that similar natural reactors may have already been mined out without being noticed. Or, one could be discovered tomorrow, to become another case for the world’s nuclear detectives. □