The Oklo Phenomenon

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Until recently, scientists believed that the chemical elements were synthesized only in stars. The discovery of the Oklo phenomenon in the Republic of Gabon in 1972 has revealed, however, that a nuclear “fire” had existed on the earth and large-scale transmutations of the elements were occurring on our planet 1.7 \times 10^9 years ago. The formation of natural (or Pre-Fermi) reactors is closely related to the appearance of life on our planet. The Pre-Fermi reactors were probably never formed until about 2 \times 10^9 years ago, when oxygen was injected into the earth’s atmosphere by a new generation of living organisms carrying out photosynthesis.

Early Ideas

About a half century ago, it was becoming increasingly evident that all the elements existing on the earth had already been discovered. Scientists were soon to begin discovering “new” elements by means of the artificial synthesis. In 1925, Walter and Ida Noddack [1] published a paper entitled „Die Ekamangane“ in this journal and in 1930, Ida and Walter Noddack [2] published another paper entitled „Die Häufigkeit der chemischen Elemente“. They reported element 43 (which they called “Masurium”, Ma) and element 75 (rhenium, Re) to be the least abundant of all the stable elements. The Noddacks were trying to isolate element 43, which was one of the last remaining ‘missing’ elements in the 1930’s.

A paper entitled „Über den Nachweis und das Verhalten der bei der Bestrahlung des Urans mittels Neutronen entstehenden Erdalkalimetalle“ by O. Hahn and F. Strassmann [3] appeared in this journal in 1939. S. Flügge [4], then published a paper entitled „Kann der Energieinhalt der Atomkerne technisch nutzbar gemacht werden?“ in the same year 1939. In this paper, he considered the possibility that a self-sustaining uranium chain reaction might have taken place under natural conditions sometime in the past, possibly in large pitchblende ore deposit at St. Joachimsthal in Bohemia or in carnotite deposits in the Colorado region of the United States, but he noted that: „Im ganzen kann man sagen, daß das Auftreten einer Explosion in der Natur ein sehr unwahrscheinlicher Vorgang ist, da wir nirgends Anhäufungen von hinreichender Mächtigkeit bei zugleich hinreichenden Stoffen vorfinden“.

The 1938 Nobel Prize in Physics was awarded to Enrico Fermi in Italy and three years later he was leading a team of American scientists at the University of Chicago to build the first nuclear reactor. On December 2, 1942, man achieved here the first self-sustaining chain reaction and thereby initiated the controlled release of nuclear energy — so reads the plaque at the football stadium of the University of Chicago.

It seems that there were scientists who doubted the success of Fermi’s experiment. In 1957, Hughes [5] wrote: “— the argument was advanced by some scientists in the early days (of WW II) that the chain reaction would be impossible. They reached this conclusion on the basis that if it really were possible it would have already taken place naturally somewhere in the earth’s surface, with disastrous results. To these people it just did not seem possible that the large-scale release of nuclear energy, so tremendous in comprehension, could be accomplished by the limited efforts of men“.

The spectacular success of Fermi’s experiment was interpreted, however, by many scientists as an unequivocal proof against the occurrence of a large-scale uranium chain reaction in nature. The opin-
ion of the scientists during the post-WWII period was best described by Cowan [6], who wrote in 1976: "— the announcement of the Oklo reactor was received by American nuclear scientists with skepticism. Some of the world's best physicists had constructed the Stagg Field reactor with careful attention to mechanical detail, to the purity of the materials and to the geometry of the assembly. Could nature have achieved the same result so casually? We now know that the answer is yes".

The possibility of the occurrence of large-scale nuclear processes in the earth's crust was discussed by a number of investigators during the period between 1939 and 1950, but the arguments presented by these investigators were either vague or inconclusive. In 1956, Burkhardt [7] published an article in this journal, in which he discussed the resonance capture reactions of neutrons occurring in uranium ore deposits.

**The Theory of Natural (or Pre-Fermi) Reactors**

In a short article entitled "On the Nuclear Physical Stability of the Uranium Minerals", which was published in the same year 1956, the writer [8] stated: "The infinite multiplication constant, $k_{\infty}$, may be considered as an indicator of the stability of the uranium minerals, which are the natural assemblages of uranium, moderator, and impurities. We may consider a system to be quite 'stable', when the infinite multiplication constant of the assemblages is less than unity. The system will be nuclear physically 'unstable' when the infinite multiplication constant is greater than unity".

According to Fermi's pile theory [9],

$$k_{\infty} = \varepsilon p f \eta,$$  \hspace{1cm} (1)

where $\varepsilon$ is the fast fission factor, $p$ is the resonance escape probability, $f$ is the thermal utilization factor, and $\eta$ is the number of fast neutrons available per neutron absorbed by uranium. When dealing with geological events, the change of the uranium enrichment as a function of geological time should also be taken into consideration. The major neutron sources in minerals are the spontaneous fission of $^{238}$U and the $(n, \alpha)$ reactions.

The values of $p$ and $f$ can be calculated if the chemical composition of the ore deposit (or the mineral) is given, $\varepsilon$ is always close to unity, and $\eta$ as a function of the uranium enrichment is known. The uranium enrichment is a function of the geological time: for example, the $^{235}$U enrichment at the present time is 0.72%, but it was 1.3% 700 million years ago, 2.3% 1400 million years ago and 4.0% 2100 million years ago and so on. Hence the value of $k_{\infty}$ of a uranium ore deposit at any geological time can be calculated by choosing a suitable model for the process of formation of large uranium ore deposits.

The simplest model is to imagine that a large ore deposit has suddenly appeared on the surface of the earth essentially in its present form at a certain geological time (Model I). The calculations based on Model I, however, invariably lead to the conclusion that the values of $k_{\infty}$ have never exceeded unity at any geological time, which means that the chain reaction could not have become self-sustaining at any time during the geological history of the earth.

A somewhat more complicated, but geochemically reasonable, model is to imagine that uranium was transported by water, precipitated and dehydrated to form a large ore deposit (Model II). The calculations based on Model II lead to the conclusion that a high-grade uranium ore deposit could have easily become an operating pile about $2 \cdot 10^9$ years ago, if the size of the ore deposit was such that its thickness was greater than, say, about 30 cm.

Of the two models considered here, Model I is obviously an oversimplification and one must accept the conclusion reached from the calculations based on Model II; Pre-Fermi reactors should have existed on the earth about $2 \cdot 10^9$ years ago. At the time the paper was written in 1956, it appeared to be highly unlikely that a natural reactor would be actually discovered within a foreseeable future, but it so happened that the Oklo phenomenon was discovered 16 years later.

**Discovery of the Oklo Reactor**

On June 7, 1972, an anomaly in the uranium isotopic ratio of a sample of natural uranium was observed by H. Bouzigne, R.J.M. Boyer, C. Seyve, and P. Teulieres [10] at the French Atomic Energy Establishment at Pierrelatte. The $^{235}$U abundance in this uranium was 0.7171 atom-%, as against 0.7202 $\pm$ 0.0010 for normal natural uranium. During the months of June to August 1972, the following new facts emerged: (a) the anomalous ore was traced to Oklo in the Republic of Gabon, Africa, and it was found that, between December 1970 and May 1972, the ore which originated from Oklo was deficient in $^{235}$U by a total amounting to 200 kg of $^{235}$U; (b) uranium with an unbelievably low isotopic abundance of 0.440% $^{235}$U was discovered; and (c) fission-produced neodymium and samarium isotopes were found in the ore.

These developments led to the special announcement by the French Atomic Energy Commission...