'Oklo phenomenon' of natural nuclear reactors – recent insights into its evolution and extinction

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In an ancient microcontinent called Atlantica[1], an assembly of six Archaean cratonic segments around which more landmasses accreted later and finally separated to form the independent continents of Africa and South America, an interesting natural phenomenon had occurred in the Palaeoproterozoic period, about 2 billion years (b.y.) ago. This event, which came to the notice of scientists only during the 1970s, happened in Gabon, which during this Palaeoproterozoic was a small corner in one of these ancient segments called the West African craton. Here, in a mine located at place called Oklo, nature had pioneered the splitting of uranium and initiated a well-controlled chain reaction and operated a veritable nuclear reactor, which ran for a few thousand years, and this came to be called the 'Oklo Phenomenon'. The Oklo natural reactor produced enormous energy and released nuclear wastes, which nature even managed to lock up within mineral structures, yet another of its early demonstration of waste immobilization technique. How these ancient uranium deposits achieved the critical enrichment of the fissile ^{235}U for initiating nuclear chain reaction and how the reactor operated for such a long time without causing explosion had remained unexplained till recent years.

The development of nuclear energy may be considered to have begun in 1938, when Otto Hahn and Fritz Strassman, little aware of earth's accomplishment hundreds of millions of years earlier in splitting uranium, announced their triumph in doing the same by bombarding uranium with neutrons. Soon, this was followed in 1942 by the historic squash court experiment at the University of Chicago by Enrico Fermi who succeeded in initiating self-sustaining nuclear chain reaction, which soon paved the way for the utilization of nuclear energy. In the wake of these scientific breakthroughs, scientists wondered whether such nuclear reactions might have occurred in earth's past resulting in release of nuclear energy. Prompted by these speculations, Kuroda worked out in 1956 conditions required for initiation of such fission reactions in nature based on theoretical calculations for the nuclear physical stability of uranium minerals[2]. However, these predictions for such occurrences in the past remained a theoretical possibility only until another 16 years when scientists while pursuing certain clues that came up during routine examination of uranium ores, were led to the recognition of sites where nature, in its geological past, had performed such nuclear chain reactions.

In 1972, French scientists while examining enriched uranium ore samples mined from Oklo in Franceville Basin, Gabon (west central Africa), were baffled by certain anomalies – a small depletion in the amounts of ^{235}U and excess of certain fission product isotopes (e.g. Nd and Ru). Since these disproportionate amounts can result only through nuclear chain reactions, this discovery of isotope abnormalities led them to conclude that nuclear reactions must have taken place in this uranium mine, possibly around 2 b.y. ago. Initially, only two such natural reactors at Oklo and neighbouring Okelobondo mines were identified but soon 17 more such reactor zones, the largest among them with a core measuring $12 \times 18 \times 0.2$–0.5 m were discovered[3]. These U-deposits occur intermixed along with the deltaic sediments, which overlie a sequence of coarse sandstone and conglomerate beds[3].

Uranium minerals, chiefly uraninites (UO_2), usually occur disseminated in granitic rocks, pegmatites and as hydrothermal veins. Weathering of these rocks releases the grains of uraninites and other rock-forming minerals which collectively get transported and deposited in basins like shallow seas or other terrestrial lakes. Uraninite grains being relatively heavier (sp. gr. 8–10) than other minerals in the detritus, they get separated from the rest by gravitational sorting during the transport and deposition in the basins and thus a limited concentration is achieved while being laid down in these basins. However, this concentration is not sufficient to initiate nuclear reaction as the fissile component ^{235}U in them may have decreased subsequent to their crystallization around 4.3 b.y. ago, the decrease arising due to the faster decay of ^{235}U (half-life 0.70 b.y.) relative to ^{238}U (half-life 4.5 b.y.). To obtain the critical proportion of ^{235}U, the uranium ores have to be further concentrated by removal of the intermixed gangue minerals as well as rare earth elements (REE) and other elements occurring in the uraninites in minor and trace amounts through selective dissolution. But, uraninites are normally insoluble except in the presence of oxygen and this vital component is considered absent in the Hadean and Archaean atmosphere. Nonetheless, early earth appears to have overcome this problem and had dissolved and enriched these deposits sufficiently to initiate chain reactions. According to recently advanced views, nature achieved this task by exploiting the emergence of oxygenic photosynthesis, presumably during the preceding Archaean era[3,4].

Oxygenation of the anoxic ancient basins is thought to have taken place under certain favourable chain reactions perceived to have occurred in the Archaean atmosphere and this apparently had promoted the proliferation of oxygenic photosynthesizing bacteria like the cyanobacteria[4]. These organisms, called the photoautotrophs, are inhabitants of the photic zone or the shallow depths of the basins penetrated by sunlight, and they use water as the electron donor in the presence of this light and produce oxygen as a waste product of their cellular processes. According to the established belief, these organisms were supposed to have arrived between 2.25 and 2.05 b.y. ago, a period considered globally eventful heralding the rise of oxygen in the atmosphere as well as evolution of multicelled life forms and hence often referred as the great oxidation event (GOE). However, some scientists believe that these bacteria had in fact flourished even earlier during the pre-3 b.y. period[5–8], and though several biological evidences in support of this view were also reported, most of them were subsequently either discredited or their biogenecity doubted or their antiquity disputed[9]. However, unlike these much questioned

biological proofs, later reports of Mo–Re abundance in kerogenic shales as well as C-isotopic evidences from pelagic shales and mineralogical clues, all recorded from the Archaean period, have revived the views supporting the existence of oxygenic photosynthesizing bacteria, long before the GOE[6–8]. Currently, it is believed that during the Archaean era, isolated oxygenated basins (oases) may have existed or possibly the oxygenation of the basins exhibited ups and downs in these times.

In recent years, views have been advanced[3,4] that post-depositional oxidation of the Archaean water basins dissolved the insoluble detrital uraninites (U^{4+}) and carried them in solution as hexavalent uranium (U^{6+}). On entering zones in the basins still in the anoxic state, all the latter remobilized (oxidized) uranium (U^{6+}), being highly redox sensitive, were supposed to have been reduced to insoluble form of uraninites (U^{4+}) and redeposited in structurally favourable traps in a very highly purified state, free from neutron absorbers like boron and rare earth[4]. The fissile ^{235}U is now present at about 3%, almost equalling the amounts required in modern day reactors (presently, the natural abundance of ^{235}U in uraninite is <1%, this depletion arising from the ageing of the mineral since its primary crystallization). A few other factors were also favourable in this region to achieve the nuclear chain reaction. For example, the deposits were of sufficient length and thickness, and these prevented escape of neutrons released from the uranium ore. Also, the presence of a moderator in the form of abundant intergranular porewater (groundwater) slowed down the neutrons and facilitated splitting of more U nuclei. Taking into consideration the temporal decay of ^{235}U expected in the uraninites since they first formed, and also the shape of the ore-bodies, their mineralogical and chemical composition, the study has calculated that the critical mass of uranium (combined ^{235}U and ^{238}U) required during most of the Archaean must have been less than 1000 kg or 0.1 cubic metre[4]. This requirement is expected to increase with exponential decrease of ^{235}U with passage of geological time. The calculations have also indicated that the deposits had adequate amounts to sustain reaction for prolonged duration.

Isotope dating and studies on the amounts of fission products released have revealed that the self-sustaining nuclear reactions were operating for 2×10^4 to 2×10^5 years and produced a steady power output of ~100 kilowatts[4]. Surprisingly, the reactors carefully regulated themselves throughout their life span and did not explode and this feature had remained an enigma for a long time. Answers to this have recently come up from mass spectrometric studies carried out on xenon isotopes, which are products of nuclear fission reactions[10]. Five of the longer lived isotopes of this noble gas – ^{136}Xe, ^{134}Xe, ^{132}Xe, ^{131}Xe and ^{129}Xe, which are known to be produced sequentially in varying amounts at specific stages of the natural reactor operation, were extracted selectively from gaseous inclusions, present in unchanged state (Xe is chemically inert) within apatites, a Ca, Al phosphate mineral formed by the interaction between heated groundwater and the surrounding rocks. Interpretations based on the production sequence of the Xe isotopes indicated that the heat from the reactor gradually boiled away the groundwater serving as neutron moderator, and this had resulted in the slowing down or stopping of the nuclear reaction. This stoppage led to cooling of the reactor zones and enabled groundwater to recharge the region once again, thus promoting resumption of reactor operation[10]. This process of heating and cooling at regular intervals is supposed to have regulated the nuclear reactions and saved these reactors from self-destruction.

The nuclear reactions which operated for quite a few thousand years have since died, probably due to accumulation of fission product neutron poisons like the REEs in the uraninites, with progress of time[4,10]. Today, Franceville Basin, Gabon, is the only known site hosting world's fossil reactors, and all that is left here are the signatures of the bygone nuclear reactions in the form of depleted ^{235}U relative to ^{238}U and enriched fission product isotopes, mostly non-volatile REEs, Y and Zr. These have all been well contained in clays and phosphate minerals (apatites), particularly those of REEs, Ra and Pu which had migrated and redistributed away from the radioactive zone due to supergene weathering[10,11]. The latter weathering, however, is geologically a slow process and therefore it is likely that ionizing radiation from some of the short- and long-lived isotopes from these reactor zones may have adversely affected the survival of organisms in the vicinity, particularly the oxygen producing photoautotrophs though the latter scenario is unlikely to have delayed the rise of oxygen in the atmosphere and basins globally[4].

The Oklo phenomenon occurred in early Proterozoic, but there could have existed similar natural reactors even during earlier geological periods, considering the fact that the amounts of fissile component ^{235}U to initiate chain reaction must have been in much higher proportion, possibly around 10% or more during the preceding Archaean and Hadean eras. Perhaps future searches for appropriate isotopic signatures to such past events in the few surviving crustal areas of these periods around the world may perhaps lead to the discovery of such sites of extinct natural reactors predating the Gabon occurrences.

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