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RADIOACTIVE WASTE MANAGEMENT : A GEOSCIENTIFIC ASSESSMENT

"THE OKLO REACTORS : NATURAL ANALOGUES TO NUCLEAR WASTE REPOSITORIES"

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INTRODUCTION

The possibility of long-term storage of radioactive waste originating from nuclear reactors and reprocessing plants in natural geological repositories has been under consideration for some time. However a number of uncertainties still exist about the safety of burying wastes in underground salt or rock formations. Most of the uncertainties involve a lack of knowledge about potential geophysical, geochemical or hydrological interactions between the wastes and the geological structure in which they might be stored.

Efforts to evaluate the effectiveness of geological media in containing radioactive wastes is severely constrained in that the time periods involved are greater than that recorded by human experience. Although many short-lived fission products decay to low levels in tens of years, a number of longer-lived radionuclides like ^{137}Cs and ^{90}Sr take approximately 600 years before they are effectively extinct. However the actinides with their larger half-lives, may take 100,000 years to decay to low levels, so that no artificial repository can hope to provide definitive answers to the long-term problems of nuclear waste storage.

Nature, it would seem, had anticipated man by something like 1800 million years, in bringing about the first self-sustained nuclear chain reaction on the Earth: And, contrary to popular opinion, it was not in the squash courts of the University of Chicago in December 1942, but in

the wilds of what is today the Republic of Gabon in Equatorial West Africa at a place called Oklo, that this event took place.

In 1972 it was found that uranium samples delivered to the French Atomic Energy Commission at Pierrelatte had an anomaly in the isotopic abundance of uranium-235 in relation to uranium-238, (0.7171 ± 0.0010 atomic percent of ^{235}U), compared with the normal value for natural uranium of 0.7202 ± 0.0006 atom percent, (Bouziqes et al., 1975).

During 1972 the following facts emerged:-

- (i) the anomalous uranium ore was traced to Oklo and it was found that between December 1970 and May 1972, the ore which originated from Oklo was deficient in ^{235}U by some 200 kg.
- (ii) uranium with an isotopic abundance of 0.440% ^{235}U was discovered, (since then 0.296% ^{235}U has been found in uranium samples from this source).
- ii) fission product neodymium and samarium isotopes were detected in the ore by spark source mass spectrography.

These developments led to the announcement by the French Atomic Energy Commission in September 1972, that it was believed that self-sustaining nuclear chain reactions had occurred at Oklo nearly two

billion years ago for approximately one million years. Since that time this unique occurrence has been extensively studied, not only to understand how the nuclear reactions occurred, but also to investigate the geological and geochemical parameters involved. The results of many of these studies were presented at two Symposia organised by the International Atomic Energy Agency - at Libreville (in 1975) and in Paris (in 1977).

It is of interest to note that Wetherill and Inghram (1953) raised the possibility of natural nuclear reactors occurring in the past history of the earth some 30 years ago. Subsequently Kuroda (1956), by a detailed examination of Fermi's pile theory, showed that it was theoretically possible for a natural assemblage of uranium, moderator and impurities to behave as a nuclear reactor at some time in the past.

The key to understanding how a nuclear reactor could have occurred in the past is tied to the relative half lives of the two principal isotopes of uranium, ^{235}U and ^{238}U . The fissile isotope ^{235}U decays more rapidly than ^{238}U , with the result that the relative abundance of ^{235}U decreases with time. If one extrapolates back into the past, it can be shown that two billion years ago the ^{235}U abundance was 3.65% as compared to the present-day value of 0.72%. This is a similar concentration to that now used in the fuel of modern pressurised water reactors.

According to Maurette (1976), the high concentration of uranium found at Oklo resulted from a long series of repetitive fractionation processes, in which oxygen played a key role as an oxidising agent. It is assumed that oxygen was injected into the earth's atmosphere approximately two billion years ago by a new generation of living organisms carrying out photosynthesis. Thus high grade uranium ore deposits needed to trigger a nuclear chain reaction were probably never formed before two billion years ago, and the occurrence of "fossil reactors" were therefore limited to a time window some 1 to 2 billion years ago.

A search has been made for other natural reactors by studying the $^{235}\text{U}/^{238}\text{U}$ ratios of uranium ores (Cowan and Adler, 1976). The results were inconclusive, although ore from the Colorado Plateau, U.S.A. show a slightly lower value than the 'norm'. Maurette (1976) has summarised various characteristics of uranium deposits from different parts of the world. He has identified a number of conditions relating to size, age and uranium concentration of the deposits, which suggest that a number of deposits have "Oklo-like" characteristics. However none of these have as yet been identified as natural reactors, including a number from Australia, and thus at this time the Oklo natural reactors are unique.

What is so important about this discovery for us today? The state of preservation of these "fossil reactors" is exceptionally good. Thus the amount of the various fission products in the reactor zones can be

interpreted in terms of neutron physics which enables the geochemical stability of the fission products and the transuranium elements to be evaluated. In the context of our Symposium today, these Oklo reactors offer a unique opportunity to examine the fission products and use them as isotopic tracers to examine the mobility/retentivity of numerous elements in the reactor zones themselves and in the surrounding geological formation. The stability of the fission products produced in the Oklo deposits some 2 billion years ago, is pertinent to the storage of radioactive wastes in natural geological formations, and in the design of artificial repositories.

GEOLOGICAL SETTING

The Oklo uranium open-pit quarry is located in the south-east part of the Republic of Gabon in the Franceville sedimentary basin, which covers an area of 35000 km². Uranium mineralisation is located in a sandstone conglomerate lens which shows a 40° mean slant and which follows the rise of the crystalline basement relative to the sedimentary cover. Each reactor zone consists of a compact accumulation of rich uranium ore, flattened in the direction of the stratum, usually as a single block of material. The length of each block ranges from 10-20 m with a thickness of about 1 m. The size of the uranium rich blocks was sufficiently large to exceed the critical mass and so allow the chain reactions to occur.

These uranium blocks are embedded in a clay gangue. In many of the reactor zones the uraninite is present as a lens of uranium rich ore, surrounded by a transition zone which grades into ordinary ore concentration, where it is present as pitchblende, and finally into the clays and sandstones surrounding the ore (Figure 1).

The uraninite is remarkably deficient in elements possessing high neutron capture cross sections which would have prevented the nuclear chain reactions occurring. A number of reaction zones have been discovered in the Oklo minesite and another at Oklobondo, about 1.5 km south of Oklo (Figure 2). It is estimated that 800 metric tons of uranium participated in the nuclear reactions, the present deficit in ^{235}U being about 600 kg. About 5 metric tons of fission products have been generated in these well-localised zones. The total accumulated heat produced was greater than 500×10^9 megajoules and the temperature in the reactors may have reached 400°C (Naudet, 1978). The energy was probably dissipated mainly by convection currents of water.

FISSION PRODUCT STABILITY

It was soon apparent that the Oklo Phenomenon presented a unique opportunity to investigate the geochemical stability of the fission products which had been produced in the reactor zones. It was important to discover if the fission products had remained in situ; if they had escaped from the reactor zones but were retained in the surrounding geological material, or if they had escaped completely from the minesite.

Detailed mass spectrometric studies have now been carried out on most of the fission products using the stable isotope dilution technique to measure the amounts of the various elements in the reactor core uraninite and the peripheral pelitic sandstone. Calculations of the amount of fission products produced in the period of criticality can be made from a knowledge of the nuclear parameters, and these values can be compared to the measured concentrations to enable the fraction of these isotopes retained in the reactor zones and in the surrounding material to be determined.

The following results summarise our present knowledge of the major elements produced in the reactors:-

- (i) Rare Earths: The rare earth elements have remained stable within the reaction zones themselves. The amounts of fission product rare earths that have migrated outside the reactors is only a few percent of the total amount produced by the fission process (Ruffenach, 1978).

- (ii) Rare Gases: Krypton and xenon produced in fission have been almost completely lost from the reaction zones. It is thought that most of this occurred during the period of criticality itself.

- (iii) Alkali and Alkali Earths: Although it has been shown that essentially all the rubidium, caesium, strontium and barium has been leached from the uranium, there is evidence to suggest that most of the ^{90}Sr remained in situ until it had decayed to ^{90}Zr . Unfortunately the evidence with respect to ^{137}Cs is equivocal because of the massive amounts of terrestrial barium which are associated with the ores in comparison to the amount of ^{137}Ba produced by the decay of ^{137}Cs . Brookins et al. (1975) argues that the Oklo reactors were stable with respect to caesium mobility for some 25-30 million years, but more definitive evidence is required to confirm this hypothesis.
- (iv) Ruthenium and Technetium: The isotopic composition of ruthenium from Oklo reaction zone samples can reveal evidence of migration of ^{99}Tc , which decays to ^{99}Ru with a half life of 2.13×10^6 years. A deficiency in the relative abundance of ^{99}Ru may indicate a preferential depletion of technetium with respect to ruthenium. Hagemann et al. (1978) has shown that ruthenium shows great stability in uraninite whilst technetium, although showing some mobility from the uraninite, is contained within the surrounding material.
- (v) Palladium, Silver, Tin, Cadmium and Tellurium: De Laeter et al. (1980) have shown that tellurium and palladium are retained almost completely in the reaction zones, silver and

tin have been partially removed, whilst cadmium has been almost completely lost from the uraninite. However much of the fission products which have escaped from the reaction zones have been contained in the surrounding material (Loss et al, 1983).

(vi) Zirconium and Molybdenum: Zirconium is retained in the uraninite and the clay surrounding the reaction zones, whilst molybdenum is a mobile element which has been lost almost completely from the reaction zones and surrounding material (Hagemann and Roth, 1978).

(vii) Transuranic Elements: Several metric tons of ^{239}Pu were formed in the reactors by neutron capture on ^{238}U . About 93% of the ^{239}Pu formed decays into ^{235}U during the period the reactors were critical, whilst some fissioned and some were converted to ^{240}Pu by neutron capture. Havette et al.(1975) have shown that plutonium is very stable in the uranium and that even on the micron scale ^{238}U and ^{235}U occupy similar lattice site positions. Similarly Devillers and Mones (1978) have shown that thorium is almost entirely retained in the uraninite.

THE STORAGE OF RADIOACTIVE WASTES

The natural reactors at Oklo offer a unique opportunity to apply isotopic geochemistry to a study of the processes involved in the transport of fission products through geological materials. Nature has performed a long-term experiment and the findings are encouraging. Investigations carried out at Oklo have shown that many fission products, including plutonium, have been contained in the uraninite reaction sites and the surrounding pelitic sandstone for an indefinite period of time. Crystalline matrices like uraninite or thorianite undoubtedly have the ability to retain many elements in their structure.

The inventory at Oklo corresponds to 10-20% of the uranium and 1-2% of the plutonium and fission products of the United States spent fuel inventory as of 1980 (U.S.D.E., 1980). It must be noted that during reactor criticality, the material which comprised the reactors were subjected to a much larger radiation dose than would be expected in a natural repository. The total energy produced is approximately 16 times the total amount of energy which would be released in a repository if it were filled with the entire U.S. inventory of spent fuels (U.S.D.E., 1980). Again during the time the reactors were critical, the average thermal loading within the reactor zones was two to five times greater than the local thermal loadings determined for waste repositories.

It is believed that water was both the moderator and the control agency for the Oklo reactors (Naudet, 1978). Thus during the high temperature phase of reactor criticality, hot aqueous solutions convectively circulated through the rocks within the reactor zones. The reactor zones and surrounding rocks were therefore subjected to circulating hydrothermal fluids driven by the heat generated by the reactors.

As has already been observed, many fission products have not been retained in the reactor zones themselves. There are gross deficiencies of rare gases, halogens, molybdenum, cadmium and the alkali and alkali earths. Some of these elements have been contained in the material surrounding the reaction zones, although it is unlikely that this would be true for the gaseous elements. Fortunately some of these mobile elements do not possess fission products which are particularly hazardous. Again ^{90}Sr seems to have been contained, at least until most of it had decayed to ^{90}Zr . However the evidence for the retention of ^{137}Cs over the period of its life is equivocal.

In the context of the multiple barrier philosophy of nuclear waste disposal, geochemical considerations are of paramount importance in determining the stability of geological environments. The Oklo reactors offer a unique opportunity to examine the mobility/retentivity of a number of fission products, and to understand the geochemical mechanisms responsible for their behaviour in a well-defined geological repository.

Despite high temperatures, large radiation doses and circulating fluids, the Oklo minesite successfully contained large quantities of radioactive waste material for some two billion years. Despite the fact that some radioactive species migrated, the actinides, which pose the really long-term hazard, remained in-situ.

Thus the lesson that Oklo provides is that the aim of achieving artificial storage under defined geological conditions for long periods of time is not, a priori - an impossible task. Although much still remains to be learnt from Oklo, at least the existing evidence is supportive of the concept of geological burial of man-made radioactive waste provided host materials of well-identified characteristics are incorporated in a multiple barrier disposal system.

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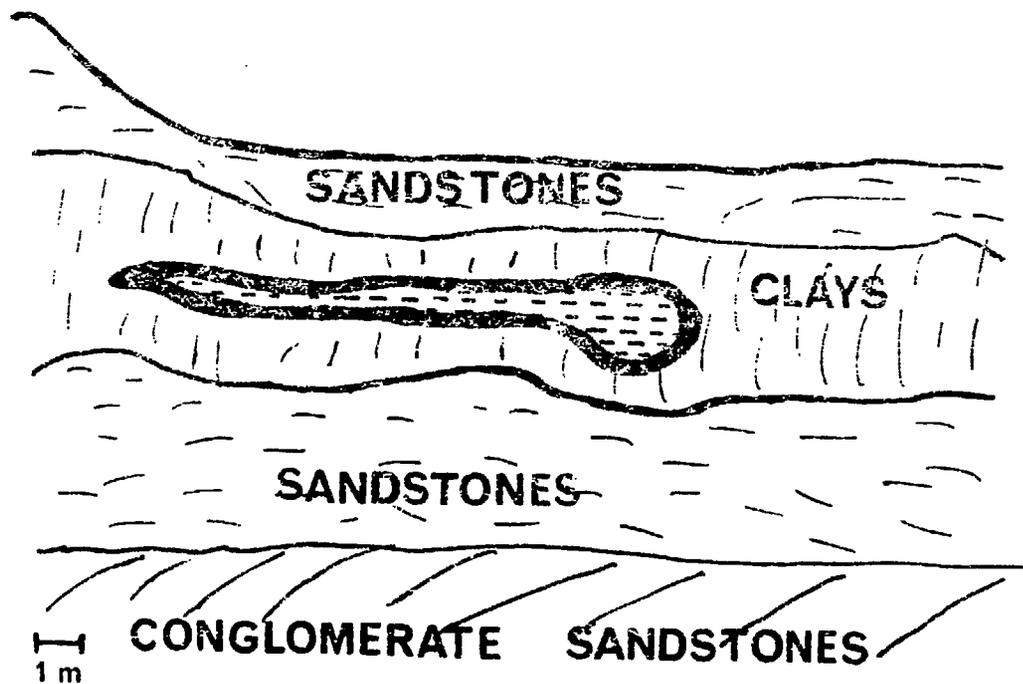
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FIGURE CAPTIONS

Figure 1: Schematic diagram of the geological formation in the neighbourhood of reaction zone 2 at Oklo.

Figure 2: The disposition of the fossil reactor zones at Oklo. Another reaction zone has been found at Oklobondo, approximately 1.5 km to the south of the Oklo reaction zones.



 **U > 10 %**
 **U > 20 %**

