THE OKLO PHENOMENON AND THE ROLE OF NUCLEAR DATA IN ITS STUDY

The study of the OKLO phenomenon requires, but also provides, in some cases better nuclear data than were available at the time of its discovery. Examples of this situation are given particularly for rare earth's fission yields and neutron capture cross sections.

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Introduction

The story of the discovery of the OKLO phenomenon has been told at other occasions (1). In this paper I wish to emphasize how important, for the understanding of the processes that led to the occurrence of natural fission chain reactions, are precise nuclear data, such as fission yields and neutron cross sections.

Small uncertainties on nuclear quantities make it impossible to choose qualitatively between various geophysical hypothesis.

But, more important perhaps to this audience, is the fact that the study of these natural reactors contributes to improve the knowledge of such quantities.

General pictures of the OKLO quarry and of the "nuclear reactors"

It is situated in the south-eastern part of Gabon. The Gabonese Republic lies at the equator in western Africa, (Fig. 1)

The geologic formation where the uranium deposit is found is known as Francevillian. It has been thoroughly studied.

It is a sedimentary, precambrian series, that rests upon a crystalline (granitic) base.

In one of the sandstone layers, close to an upsurge of the crystalline bed, and covered by pelitic layers, uraniumiferous material is found.

Within an area several hundred meters long and at depths varying by 50 meters, nine reaction zones have been discovered. A tenth has been reported about 1.5 km south from the quarry. (Fig. 2)

The characteristics that enabled chain reactions to take place were:

a) the age of the deposit. It is roughly two billions years old, and at that time natural uranium was about 3% rich in \(^{235}\)U.

b) The chemical concentration of uranium. It is high, often more than 20% now.

c) The thickness of the layers. It exceeds often several decimeters.

d) Enough water, and little nuclear poison present at the time of deposition.

These necessary conditions for divergence were met at OKLO.

In the regions explored up to now about 800 tons of uranium were involved in nuclear reactions, and a total quantity of about 6 tons of uranium \(^{235}\) underwent fission.

However, due to a conversion factor of .4 to .5 and to decay, the present deficit in \(^{235}\)U at OKLO is only about 600 kilograms.

A total energy of 500 billions megajoules was evolved, corresponding to the continuous operation of a 1000 M\(\text{Wth}\) reactor for about 15 years, or rather of a 15 kilowatt source for one million years, as we will see.

As soon as the proof of chain reactions was offered, by both the chemical and the isotopic compositions of rare earths present in the uranium ore, the following questions were raised.

How did the "reactors" start, how were the reactions sustained and finally brought to an end?

To establish these, in turn we wanted to know quantitatively, the "burn up" of the ore, i.e the integrated fluence it underwent. Together with the fluence the instantaneous flux, or power level, provides the duration of the reaction.

Finally an evaluation of temperatures reached is important to understand moderation and cooling mechanisms. A knowledge of the proportions of fissions that occurred in \(^{238}\)U (\(\text{ox}\)) and \(^{238}\)U (\(\beta\)) also contributes to the total picture. Isotopic analysis of fission products and uranium provide many of the answers, when the necessary nuclear data are at hand, as can be seen from the following section, and 9.

Use of nuclear data to evaluate physical parameters characterising the chain reactions

Measurement of total fluence \(\Phi\)

From observed variations in isotopic composition of uranium one can calculate the fluence \(\Phi\).

As first approximation (without considering the formation of \(^{237}\)Np and it's decay to \(^{233}\)Th),

\[ \Phi = \frac{1}{\sigma_s - \sigma_C} \ln \frac{N_N}{N_N^0} \]  

where \(N_N = \frac{(N_8/N_8^0)n}{(N_8/N_8)} \) (2)

\(N_N^0\) are the number of atoms of \(^{235}\)U and \(^{238}\)U present now in the sample.

\(N_N\) is the "normal" ratio usually observed.

\(\sigma_s\) is the total neutron cross section of \(^{238}\)U (fission plus capture)

\(\sigma_C\) is the capture cross section of \(^{238}\)U.
When the "restitution factor" $C$ is taken into account the formula becomes:

$$\tau = \frac{1}{(1-C)\sigma_c - \sigma_c}$$  \hspace{1cm} (3)

WHERE $C = N_8 \cdot \sigma_c / N_5 \sigma_c$

From an isotope of a fission product
if $\sigma_c$ is the fission yield of this isotope
$\sigma_c$ its capture cross section
$N_8$ the total number of atoms of this isotope that were formed
$N_5$ the remaining number of atoms, after neutron capture.
then:

$$N_8 - N_5 = \frac{1}{ln \frac{N_8}{N_5} \sigma_c} \cdot \frac{exp(-\sigma_c \tau) - exp(-\sigma_c \tau)}{1 - C}$$

from this formula $\sigma_c \tau$ is derived.

Spectral index and resonance integral

From the well known formula

$$\sigma_c = \sigma_c + A \cdot \tau$$ \hspace{1cm} (4)

one also derives $\lambda$, the "spectral index" from $\sigma_c$ measured, $\sigma_c (2200$ m/s cross section), and $\lambda$ (resonance integral) tabulated.

Proportion of $^{235}U$, ($<\lambda$) and $^{239}Pu$ ($\lambda$) fissions.

One uses two isotopes showing fission yields different for the three nuclides $^{235}U$, $^{238}U$, $^{239}Pu$ (table I).

From every measured isotope yield one derives a linear relation between $\lambda$ and $\beta$ that can be represented by a straight line. The use of two isotopes supplies two linear relations, or graphically two straight lines, whose intersection in the $\lambda$, $\beta$ plane has the desired values of $\lambda$ and $\beta$ as coordinates. (Fig. 3 from 5)

Duration of the reaction ($\Delta \tau$)

It is calculated from nuclides decaying both by neutron capture and natural decay. This method is analogous to that used in geophysics to evaluate exposure times of meteorites.

$$\Delta \tau \text{ from Plutonium } 239$$

(239Pu formation obeys the following equation:

$$\frac{dN_p}{dt} = N_p \cdot (\sigma_c + \lambda \cdot \sigma_c - \sigma_c \cdot \lambda)$$

Equilibrium is reached when $\frac{dN_p}{dt} = 0$

then $N_p = \frac{N_0 \cdot \sigma_c \cdot \lambda}{\sigma_c + \lambda \cdot \sigma_c}$

As $^{239}Pu$ is formed from $^{239}Pu$:

$$\Delta \tau = \frac{5 \cdot \sigma_c \cdot \lambda}{N_p \cdot \sigma_c \cdot \lambda}$$

finally $N_p = \frac{N_0 \cdot \sigma_c \cdot \lambda}{\sigma_c + \lambda \cdot \sigma_c}$

and as $\beta = \frac{\sigma_c}{\sigma_c + \lambda \cdot \sigma_c}$

It is seen that one must calculate $\sigma_c$, $\lambda$, and $\beta$ in order to obtain $\Delta \tau$.

$\Delta \tau$ from $^{99}Tc$

Fig (6) shows the disintegration scheme of $^{99}Tc$ to $^{99}Ru$ (T $1/2 : 2.10^5$ y).

$^{99}Tc$ is produced with a high fission yield, and produces also $^{100}Ru$ by neutron capture.

When such competition arises between two different modes of decay of a nucleus, leading to two discernible end-product, and when one of them has a constant relaxation time the duration of the other can be calculated.

Age of the reaction

The age of the uranium deposit can be calculated from conventional isotope geochemistry, applied to elements unperturbed by the fissions. Thus, from isotopic lead analysis and/or uranium-lead ratios, in normal zones of the quarry, or from Rubidium-Strontium studies on the pelite coverages one can be calculated.

However, in addition, an important method of cross-checking ages has been developed.

From the above mentioned nuclear data, namely total fluence and conversion factor, the number of fissions that took place in a given volume of ore can be calculated, and expressed as a proportion of the number of atoms of Uranium undergoing fission. Under the assumption that the system has behaved as a closed box the present ratio of a given stable fission nuclide to the remaining Uranium isotope provides a "chronometer":

The comparison of ages calculated by such time-keepers (they are as many, in principle, as stable fission nuclides) with those resulting from well-established methods leads to two different kinds of results:
1. a check of the validity of the closed box model. This is of primary geochemical importance, and in the present case leads also, element by element to an evaluation of the stability of fission products and transuranium elements buried in the same environment as Uranium.
2. a check of the values of fission yields and capture cross sections for some nuclides.

When the abundance of an isotope of an element can be interpreted according to the closed box model, one should be able to calculate the abundances of the other isotopes of the same element in the same way, in cases where they are also formed directly in fission. Discrepancies must lead to check nuclear data. Several examples of this situation have been quoted in the literature (8 to 11) and are illustrated by the attached tables (fables 2 to 4) of sensitivity of $\lambda$ and $\Delta \tau$ to $\sigma_c$, and of isotopic composition to $\lambda$.

Temperature measurements

A situation similar to that arising from the comparison of different methods of measuring ages develops in the case of evaluation of temperatures at which the chain reactions took place.
Non-nuclear methods are founded on thermal conduction estimates, observation of fluid inclusions, mineralogical observations (nature of crystalline species, reflecting power of graphitized deposits, etc.).

Nuclear determinations are based on the measurement of isotopic ratios in Lutetium. Lutetium is not a fission product, is present during the reaction and has two isotopes: $^{175}\text{Lu}$ and $^{176}\text{Lu}$. The neutron cross section of the latter is particularly sensitive to temperature. In our laboratories a model has been developed to calculate this temperature from Lutetium isotopic ratios (see 3). Gadolinium 155 can be used also for neutron temperature measurement in a similar way, though (fig. 1) shows that its variation with temperature is in the opposite direction of that of $^{176}\text{Lu}$. (Fig. 5) shows the parametric variations of $\frac{\Delta}{\Delta}^{176}\text{Lu}$ with $\Delta H$

The validity of the results is again sharply dependent on the relative stability in the deposit of Uranium and rare earths and the coherence of evaluations from different methods provide both geochemical evidence of this stability, and again needs to rest on well established nuclear data. They may therefore lead to reconsider the existing literature, specially when data arising from two different sets of isotopes are compared.

Conclusions

The study of the OKLO phenomenon has already led to the reinvestigation of many fission yields and neutron cross sections. The necessity that geochemical deductions be coherent when obtained by using different independent nuclear groups of data is a particularly sensitive test on some of these.

Preservation of part of a "reactor" in zone two of the OKLO quarry has been undertaken in order, to keep the road open to further sampling for further investigations.

Acknowledgements

Dr DEVLILERS and Dr HOLLIGER worked out the temperature measurement methods. Many ideas developed have benefited by discussions with Dr NAUDET.

References


2. In the OKLO phenomenon Proceedings of a symposium held in Libreville (1975) ed. by International Atomic Agency (IAEA) Vienna (1975) see P. MOLINA and J.C. BESOMBES, and references quoted.

3. R. NAUDET - private communications, and in 2 and 4.


12. G. COWAN in 2 and 4


15. B.F. RIDER and M.E. MEEK - Nedo 12154-2 (D) (1977)
FIG (1) Location of the OKLO quarry.

FIG (2) From 3
View of the "reactor's zone.

FIG (3) From 5
Determination of $\alpha$ and $\beta$.

FIG (4) From 6
Factors for 155Gd, 176Lu, 177Lu.
Effective cross section-temperature relations for different spectral indexers.

\[ \sigma_{\text{eff}}(\text{barns}) \]

\[ T_{\text{neutro}} \text{ (°C)} \]

**FIG (5) from 8**

Desintegration schema of $^{99}\text{Tc}$

- **Fission $^{235}\text{U}$**
  - $^{99}\text{Tc}$
    - (1.8 s)
  - $^{99}\text{Tc}$
    - (2.5 mn)
  - $^{99}\text{Tc}$
    - (67 h)
  - 87% stable
  - 13%

$^{99}\text{Tc}$

- (6 h)
  - (2.1 x $10^5$ s)
  - $^{99}\text{Ru}$ stable

($\rho_{\text{gg}} = 6.13 \%$)
### Table 1

<table>
<thead>
<tr>
<th>GAS NUMBER</th>
<th>F.Y.</th>
<th>I.P.</th>
<th>F.Y.</th>
<th>I.P.</th>
<th>F.Y.</th>
<th>I.P.</th>
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<tbody>
<tr>
<td>144</td>
<td>-</td>
<td>-</td>
<td>2.229</td>
<td>61.14</td>
<td>-</td>
<td>-</td>
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<tr>
<td>147</td>
<td>1.025</td>
<td>29.49</td>
<td>1.601</td>
<td>53.65</td>
<td>1.504</td>
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<tr>
<td>148</td>
<td>2.66</td>
<td>13.75</td>
<td>.519</td>
<td>10.71</td>
<td>.591</td>
<td>14.21</td>
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<tr>
<td>150</td>
<td>-</td>
<td>-</td>
<td>.213</td>
<td>4.39</td>
<td>.273</td>
<td>5.56</td>
</tr>
<tr>
<td>152</td>
<td>2.02</td>
<td>2.02</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>TOTAL</td>
<td>3.546</td>
<td>4.007</td>
<td>4.213</td>
<td>4.213</td>
<td>4.152</td>
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### Table 2

<table>
<thead>
<tr>
<th>$\sigma^*$ in barns</th>
<th>$\sigma^{141} = 325$</th>
<th>$\sigma^{141} = 714$</th>
<th>$\sigma^{141} = 714$</th>
<th>$\sigma^{141} = 325 + 100 \lambda$</th>
<th>$\sigma^{141} = 325 + 100 \lambda$</th>
<th>$\sigma^{141} = 52 + 669 \lambda$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\bar{\Gamma}$</td>
<td>1.33 $10^{21}$</td>
<td>1.63 $10^{21}$</td>
<td>1.36 $10^{21}$</td>
<td>$338 + 100 (\lambda + 600 \lambda)$</td>
<td>$338 + 100 (\lambda + 600 \lambda)$</td>
<td>$52 + 669 \lambda$</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>0.13</td>
<td>0.13</td>
<td>0.17</td>
<td>0.17</td>
<td>0.17</td>
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<tr>
<td>$\xi$</td>
<td>0.47</td>
<td>0.57</td>
<td>0.49</td>
<td>0.49</td>
<td>0.49</td>
<td>0.49</td>
</tr>
<tr>
<td>$\tau$ in $10^3$ years</td>
<td>1.98</td>
<td>1.78</td>
<td>1.79</td>
<td>1.79</td>
<td>1.79</td>
<td>1.79</td>
</tr>
</tbody>
</table>

**References:**
- From Ref. 3,7
- From Ref. 2
- From Ref. 12
### TABLE III
Sensitivity of Calculated Duration to $^{299}T_e$ and $T_l$

**SAMPLE** 50-323 $T_e = 1.12 \times 10^{21}$ n/cm$^2$

<table>
<thead>
<tr>
<th>$\lambda$</th>
<th>$\sigma_{299}T_e = 19 \times 340$ n</th>
<th>$\sigma_{299}T_e = 20 \times 118$ n</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.15</td>
<td>$\Delta t = 5 \times 10^6$</td>
<td>$\Delta t = 1 \times 10^6$</td>
</tr>
<tr>
<td>0.17</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.18</td>
<td></td>
<td></td>
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**REFERENCES:**
- FROM BNL 525
- FROM HANFORD 12

### TABLE IV
Comparison of Observed and Calculated Isotopic Composition for Neptunium

<table>
<thead>
<tr>
<th>ISOTOPES</th>
<th>MEASURED</th>
<th>CALCULATED ASSUMING $\beta = 2.7$</th>
<th>CALCULATED ASSUMING $\beta = 5.2$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>SAMPLE 50-3560</td>
<td>FISSION YIELDS FROM 15</td>
<td>FROM 16</td>
</tr>
<tr>
<td>$^{243}$Pm</td>
<td>55.78</td>
<td>55.32</td>
<td>54.81</td>
</tr>
<tr>
<td>$^{244}$Pu</td>
<td>33.95</td>
<td>33.59</td>
<td>33.94</td>
</tr>
<tr>
<td>$^{245}$Pu</td>
<td>8.17</td>
<td>8.28</td>
<td>8.29</td>
</tr>
<tr>
<td>$^{246}$Pu</td>
<td>3.30</td>
<td>3.31</td>
<td>3.31</td>
</tr>
</tbody>
</table>

The set of fission yields used for the last column provides the best agreement.