Plutonium fission in the Oklo natural reactor

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Plutonium fission in the Oklo natural reactor

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Abstract*

The Oklo natural reactor was discovered in 1972. Experimental evidence has indicated that the fuel source was primarily $^{235}\text{U}$ with a small contribution of 7-9% from the fission of $^{239}\text{Pu}$. This article's re-evaluation of data indicates that $^{239}\text{Pu}$ was an important source of fuel in some areas of the reactor: A small portion of xenon and krypton released from Oklo sample 1348 appears to have originated from a source enriched in $^{239}\text{Pu}$. That fuel source may have been the core of a natural breeder reactor.

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Introduction

The possibility of fission chain reactions occurring in natural ore deposits was recognized as early as 1956\textsuperscript{1,2}. It was not until 1972 that proof of this hypothesis was discovered in the Oklo ore deposits in Gabon (Central Africa)\textsuperscript{3}. Numerous studies have since confirmed the existence of the reactor and have provided much information about its characteristics. Previous reports\textsuperscript{4-7} have concluded that the reactor operated more than a billion years ago and was fueled almost entirely by $^{235}\text{U}$. In determining the characteristics of the reactor, the fission products produced in the fission process have been an important source of information. Many of these fission products were originally radioactive but have since decayed to stable elements. These elements serve as a "fingerprint" which yields information about the nuclear processes involved at Oklo. In the case of isotopes of the same element, partial loss of the element after the reactor ceased operating does not destroy the "fingerprint." Some of the characteristics which can be determined from the fission fragments include the time-integrated neutron flux, the type of fuel, and the duration of the process. As indicated by Naudet, et al.\textsuperscript{10}, the Oklo site is not a single reactor, but a number of reactors connected in series. This allows a variety of situations.
Baudin, et al.\textsuperscript{5} reported no evidence of plutogenic xenon in Oklo sample 310. In contrast, Drozd, et al.\textsuperscript{7} calculated the contribution to $^{136}$Xe from plutonium in Oklo sample 321 as being $7.3 \pm 0.4\%$ with the remainder being from $^{235}$U.

In this report, I have evaluated previously published xenon and krypton data from the Oklo mine to obtain additional information about plutonium as a fuel source.

RESULTS AND DISCUSSION

The rare gas isotope data obtained by Shulkolyukov, et al.\textsuperscript{8} are given in Tables I, II and III. They obtained sample 1348 from the reactor zone. The other samples were taken at measured intervals outside the zone of richest uranium concentration. The xenon isotope data are normalized to $^{136}$Xe to permit comparison with xenon from other sources. The isotope ratios are similar to those of xenon formed from the thermal-neutron fission of $^{235}$U. There are some substantial differences, however. These differences are most marked in the sample taken from the reactor zone (1348) and in samples obtained by stepwise heating of sample 1348. The isotope ratios showing the greatest difference from the ratios expected from $^{235}$U fission came off at low temperatures by the stepwise heating experiment.
Shulkolyukov, et al.\textsuperscript{8,17} realized that the anomalous xenon might be significant and considered the following processes as possible mechanisms: (1) The anomaly is due to fission processes. They considered the high $^{132}\text{Xe}/^{136}\text{Xe}$ ratio and noted that there was no known nuclide that gives a peak at that mass number, either for thermal neutron-induced fission or spontaneous fission. (2) They considered neutron-induced reactions on Ba and Te isotopes as a possible means of producing Xe. They concluded that reaction on Te was unlikely and that irradiation of Ba in a thermal neutron flux produced Xe of an entirely different isotopic composition from that formed at Oklo. (3) Finally, they considered whether the anomalies might be due to migration in the crystal structure. They noted the excess of lighter isotopes (in addition to heavy ones) compared to the amounts expected from $^{235}\text{U}$ fission, and concluded that separation by classical diffusion could not explain the observed effect. They also noted that the effect might be due to separation of those xenon isotopes with relatively long-lived precursors such as $^{131}\text{I}$ ($T_{1/2} = 8$ days) and $^{132}\text{Te}$ (78 hours). Shulkolyukov, et al.\textsuperscript{8} found this idea unconvincing since $^{134}\text{Xe}$ also shows the enrichment (Table II) and has no long-lived precursors. Shulkolyukov, et al.\textsuperscript{8} concluded that it was not possible to determine the mechanism which produced the anomalies without additional information. Since they thought that the anomalous xenon was present in all of the samples, they did not calculate the time-integrated neutron flux (fluence) based on xenon or attempt
to determine the contribution of $^{239}$Pu fission relative to total fission. This had been done previously by Drozd, et al.\textsuperscript{7} using xenon from the Oklo site which did not have isotope ratios substantially different from those expected from the fission of $^{235}$U. Drozd, et al.\textsuperscript{7} did observe minor ratio variations at mass numbers 134, 132 and 130 which they attributed to a small contribution from plutonium fission and to neutron capture on $^{129}$I. They calculated a fluence of $4 \times 10^{20}$ n/cm$^2$ and a plutonium contribution of 7% based on the slight deviations of the xenon isotope ratios from the values expected in thermal fission of $^{235}$U.

The interpretation presented here assumes that sample 1348 contains xenon from some process other than thermal fission of $^{235}$U. However, it seems likely that this unusual xenon did not contaminate the xenon from outside the reaction zone and that the xenon from the outlying samples can be used to calculate the neutron fluence and the plutonium fission contribution.

There are several reasons for believing that the unidentified process which produced the anomalous xenon did not extend to the samples taken from outside the reactor zone (1361-1371). The conditions in the reactor zone should have been quite different from the conditions in the outlying area in terms of neutron flux and neutron energy spectrum. Fast neutrons are produced in the reactor zone and are absorbed or moderated as they diffuse outward. In the presence of moderating material such as carbon or water, the neutrons which reach the outlying areas should have a much
lower energy distribution than the neutron flux in the reactor zone. Secondly, the composition of the mineral in the reactor zone is quite different from that in the adjacent areas. This is obviously true for uranium and is probably true for other elements as well. Neutron capture reactions which might produce the anomalous xenon would not necessarily take place outside the reactor zone. Finally, it appears that the anomalous xenon was associated with a higher neutron fluence than existed in the outlying areas.

**Fluence calculations**

$^{130}\text{Xe}$ can be used as a measure of the neutron fluence since it is not produced directly in fission and is shielded from other fission products in the mass 130 decay chain by stable $^{130}\text{Te}$. That the abundance of $^{130}\text{Xe}$ is low in all the samples seems to be sufficient proof that it was produced only by neutron capture. The most likely target in this case was $^{129}\text{I}$ which has a half life of 17 million years. The product of that reaction, $^{130}\text{I}$, would then decay to stable $^{130}\text{Xe}$, while the remaining $^{129}\text{I}$ would decay to stable $^{129}\text{Xe}$.

Since the $^{130}\text{Xe}$ was produced by neutron capture, it is possible to calculate the neutron fluence based on the amounts of $^{129}\text{Xe}$ and $^{130}\text{Xe}$ present now. The $^{129}\text{Xe}$-$^{130}\text{Xe}$ pair was used to
calculate the fluence for each sample according to the equation given by Drozd, et al.\textsuperscript{7}:

\[
\frac{[130\text{Xe}]_n}{[129\text{Xe}]} = \frac{1}{2} \sigma_c \tau \tag{1}
\]

where \([130\text{Xe}]_n\) is the amount of the isotope produced by neutron capture and \([129\text{Xe}]\) represents the abundance of the target material. \(\sigma_c\) is the effective neutron capture cross section which is defined as the differential cross section (including resonances) integrated over a reactor neutron energy distribution and normalized to thermal energy\textsuperscript{7}. \(\tau\) is neutron fluence which is sought. In the above case, all of the \(130\text{Xe}\) should have been created by neutron capture. The effective neutron capture cross section is the 30 barn cross section of \(129\text{I}\), since that was the target material during the active period.

The fluences calculated by the above method should be valid regardless of the source of the xenon anomalies found at mass units other than 130, so this method can be used for sample 1348.

A similar equation holds for the \(131\text{Xe}\) and \(132\text{Xe}\) pair and provides a totally independent means of calculating the neutron fluence. In this case, however, the amount of \(132\text{Xe}\) present due to neutron capture is only a fraction of the \(132\text{Xe}\) present. The amount of \(132\text{Xe}\) produced by neutron capture is found by subtracting the expected value due to \(235\text{U}\) fission from the total
normalized $^{132}\text{Xe}$ abundance. If xenon from $^{239}\text{Pu}$ is present, this will make little difference in the results since the yields for $^{239}\text{Pu}$ and $^{235}\text{Pu}$ are very similar at mass number 132. An effective neutron-capture cross section of 165 barns was used for $^{131}\text{Xe}$. For sample 1348, both $^{131}\text{Xe}$ and $^{132}\text{Xe}$ were influenced by the strange xenon component, and it was not possible to calculate a valid fluence using the 131–132 isotope pair. Accordingly, the $^{131}\text{Xe}$–$^{132}\text{Xe}$ method was used only for samples from outside the reactor zone. The calculated fluences are shown in Table III and in Figure 1.

**Plutonium contribution**

Xenon and krypton, from plutonium and uranium fission, sometimes have different isotope ratios; this difference can be used to determine the importance of plutonium as a fuel source. As shown in Table I, the normalized (to 136) xenon ratios are significantly different at mass numbers 134, 131 and 129 for uranium and plutonium fission. The plutonium contribution can be determined by use of the following methods.

A. Partition on $^{136}\text{Xe}$. For example, if the normalized $^{129}\text{Xe}$ abundance in the Oklo sample is halfway between 0.105 and 0.2576, then $^{239}\text{Pu}$ contributed 50% of the $^{136}\text{Xe}$ in that sample. For $^{129}\text{Xe}$, little addition is expected by neutron capture since the likely target, $^{128}\text{Te}$ (stable), has a cross section of less than one barn. In these samples, there is an indication of small losses of $^{129}\text{Xe}$ to mass number 130. The losses were added back
to $^{129}$Xe to obtain the contribution of plutonium fission which is listed in column (a) of Table IV.

B. The partition of $^{136}$Xe can also be calculated using mass number 131; there is a possibility of some loss to mass 132 but minimal gain from the mass number 130. The likely target for such a gain is $^{130}$Te with a thermal cross section of less than one barn. The values derived from $^{131}$Xe are computed without the addition of lost material to $^{132}$Xe and should be conservative estimates of the plutonium fission contribution.

C. The two methods given above assume the $^{136}$Xe is not altered by the neutron flux since all the values are normalized to $^{136}$Xe. This is most likely true but $^{135}$Xe (9 hour half-life) has a capture cross section of $2.7 \times 10^6$ barns which could increase the relative $^{136}$Xe abundance if the reactor operated for a few days at fluxes in excess of about $10^{12}$ n/cm$^2$. Accordingly, the plutonium contribution can be evaluated in a way that is independent of $^{136}$Xe.
The following equations given by Drozd, et al. give the fraction of $^{134}$Xe produced by plutonium fission and take into account the depletion of $^{131}$Xe and enrichment of $^{132}$Xe by neutron capture:

\[
A\left(\frac{^{131}}{^{134}}\right)_{\text{Pu}} + (1-A)\left(\frac{^{131}}{^{134}}\right)_{\text{U}} = \left(\frac{^{131}}{^{134}}\right)_{\text{m}} + \frac{[^{131}]_{\text{n}}}{[^{134}]_{\text{m}}}
\]

\[
A\left(\frac{^{132}}{^{134}}\right)_{\text{Pu}} + (1-A)\left(\frac{^{132}}{^{134}}\right)_{\text{U}} = \left(\frac{^{132}}{^{134}}\right)_{\text{m}} - \frac{[^{132}]_{\text{n}}}{[^{134}]_{\text{m}}}
\]

(2)

where the numbers refer to the xenon isotopes and the subscripts Pu, U, m and n refer to isotopic ratios produced in $^{239}$Pu and $^{235}$U neutron fission, the measured isotopic ratios in the Oklo samples and the concentrations from neutron capture reactions, respectively. The coupled equations can be solved simultaneously for A which is the fraction of $^{134}$Xe which is due to $^{239}$Pu neutron fission. $[^{131}]_{\text{n}}$, the amount of $^{131}$Xe which has been removed by neutron capture, equals $[^{132}]_{\text{n}}$, the amount of $^{132}$Xe produced by neutron capture. Solution of these equations gives values for the contribution of plutonium which are listed in column (c) of Table IV.

The results shown in Table IV show general agreement of the three methods used to calculate the plutonium contribution outside the reactor zone.

D. The krypton isotope data were used to obtain the plutonium contribution in sample 1348 (the reactor zone). The results are given in column (d) of Table IV. A comparison of the krypton isotope ratios
in the 400° fraction with the fission yields of 239Pu and 235U is shown in Fig. 2. 83Kr has a 200 barn thermal neutron capture cross section which suggested that some of the 83Kr would have been converted to 84Kr. To resolve this problem, coupled equations were used as was done for the xenon isotope ratios. The plutonium and uranium mass yields for the krypton isotopes were obtained from Meek and Rider. They reported an error for these yields of not more than 1% for uranium and not more than 2% for plutonium. The plutonium fission contribution to 86Kr calculated by this method ranges from 5% in the 1350° fraction to 61% in the 400° fraction.

It is significant that the xenon fraction associated with 61% plutonium contribution to 86Kr also has the highest neutron fluence. The number of 239Pu atoms produced when 238U is subjected to a neutron flux is as follows:

\[ N_{239} = \phi N_{238} \sigma_c \]  

(3)

where the number of plutonium atoms formed is \( N_{239} \), \( \phi \) is the neutron flux in resonance absorption region and \( \sigma_c \) is the probability of absorption by 238U. The number of atoms of 239Pu which undergo fission is:

\[ Pu_f = N_{239} \sigma_f(239) \phi_{th} \]  

(4)

The fissions due to 235U are:

\[ U_f = N_{235} \sigma_f(235) \phi_{th} \]  

(5)
The ratio of plutonium fissions to uranium fissions then becomes:

\[
\frac{P_{\text{u}}}{U} = \frac{\phi N_{238} \sigma_c \sigma_{f(239)}}{N_{235} \sigma_{f(235)}}
\]  \hspace{1cm} (6)

The neutron capture probability \( (\sigma_c) \) for \( ^{238}\text{U} \) is difficult to evaluate and requires detailed knowledge of the moderator. The important point here is that the ratio of plutonium fissions to uranium fissions is directly proportional to the flux, assuming that the other factors in equation (6) are constant. The regions of high neutron fluence (integrated flux) ought to show enhanced plutonium fission compared to zones of lower fluence. That seems to be the case for the 400° fraction of sample 1348-a. Since the fluence and plutonium fission percent were obtained independently, that indicates that the plutonium fission contribution is not unreasonable.

There is perhaps some chance that the fast fission of \( ^{238}\text{U} \) might be interpreted as \( ^{239}\text{Pu} \) fission. However, the krypton yields given by Meek and Rider\(^{12} \) give a \( ^{83}\text{Kr}/^{86}\text{Kr} \) ratio of 0.30 for the fast fission of \( ^{238}\text{U} \). This is lower than the observed ratio of 0.331 in the 400° fraction, and tends to rule out fast fission as the cause of the excess \( ^{83}\text{Kr} \). The ratio expected for fast fission at mass number 84 is about the same as for plutonium thermal fission. For the uncontaminated xenon outside the reactor zone, the results are more consistent with plutonium fission rather than fast fission of \( ^{238}\text{U} \).
It should be noted that the plutonium fission contribution in Table IV is the contribution to a specific isotope. For $^{134}\text{Xe}$ and $^{136}\text{Xe}$, this is about the same as the percent of plutonium fissions. For krypton, $^{235}\text{U}$ fission produces 2.6 times as much $^{86}\text{Kr}$ as $^{239}\text{Pu}$ fission. For the 400° fraction this implies that 80% of the fissions are from $^{239}\text{Pu}$.

The amount of rare gas present in the low temperature fractions of sample 1348 is less than 20% of the total gas content. It is obvious that plutonium fission was not a major fuel source for the sample as a whole. Shulkolyukov, et al. concluded that up to 99% of the rare gases had been lost from the mineral. Since that is probably typical of the Oklo reactors, it seems likely that the gas fractions containing the anomalous krypton and xenon ratios originated in a region of high neutron fluence and later migrated to sample 1348 where the gas become loosely bound to the mineral.

One mechanism which might produce an excess of plutonium fissions compared to $^{235}\text{U}$ in nature, is the operation of a breeder reactor where $^{238}\text{U}$ is converted to fissionable $^{239}\text{Pu}$ at a rate slightly greater than the burnup of $^{235}\text{U}$. For thermal neutron fission of $^{235}\text{U}$, $n$, the number of neutrons produced for each neutron absorbed in $^{235}\text{U}$ is 2.07. Of these, one neutron is required to maintain the chain reaction. For a breeder reactor, at least one neutron must be used to convert $^{238}\text{U}$ to $^{239}\text{Pu}$ via the reaction; $^{238}\text{U} + n \rightarrow ^{239}\text{U} \rightarrow ^{239}\text{Pu}$. That leaves only 0.07 neutrons that can be lost by absorption in impurities.
and to leakage. Thus it would seem unlikely that a thermal breeder reactor could exist in natural uranium ores. But there is another factor which serves to increase the number of available neutrons. It has been noted\textsuperscript{15} that the relevant quantity with respect to breeding is not $\eta$ but $\eta \epsilon - 1$, where $\epsilon$ is the fast fission factor and accounts for a slight enhancement due to neutrons produced in the fast fission of $^{238}$U. Weinberg and Wigner\textsuperscript{15} also pointed out that the fast fission factor could be as high as 1.10 for slightly enriched, ordinary water lattices if the lattice elements are close enough together. Widely spaced fuel rods tend to reduce the fast effect since the neutrons are reduced to thermal energy before entering a region of high uranium concentration. If we assume that $\epsilon$ was 1.10 for certain regions of the Oklo reactor, then the quantity, $\eta \epsilon - 1$, which is relevant for breeding is 1.28. This provides some additional margin for neutron losses and in cases where neutron absorbing elements were removed by previous irradiation, it might be possible for a natural reactor to shift to a plutonium fuel regime.

Because of the borderline nature of thermal breeders, the $^{235}$U-fueled natural reactors should be much more probable than a breeder type. Equation 6 suggests that a high percentage of plutonium fissions would be produced simply by irradiating uranium in a sufficiently high neutron flux apart from the question of breeding. Perhaps that was the original of the unusual krypton in the 400° fraction.
Although the Oklo reactors occurred about 1.7 billion years ago, there is some possibility that such natural reactors may exist at the present. Since the $^{235}\text{U}$ content of uranium ores is less than it was in Oklo process (due to the decay of $^{235}\text{U}$), a natural reactor is less likely at the present time. It is possible; however, to have operating reactors with uranium of the present isotopic composition (the first man-made reactor in the 1940s was fueled by non-enriched uranium).

In recent years, anomalies have been found in the natural occurrence of $^{239}\text{Pu}$ which may be related to relatively recent operation of natural reactors. Meier, et al.\textsuperscript{13} confirmed earlier work by Soviet workers\textsuperscript{14}, and found small amounts of $^{239}\text{Pu}$ in lava and other volcanic rocks. The 24,000 year half-life of $^{239}\text{Pu}$ requires that the process which produced it must have taken place recently in terms of geologic time. They noted that they were unable to find $^{239}\text{Pu}$ in magmatic or extrusive rocks, whose age exceeded $10^7$ years. $^{239}\text{Pu}$ does occur to a small extent in uranium minerals\textsuperscript{16}, where the ratio of $^{239}\text{Pu}/^{238}\text{U}$ is on the order of $10^{-11}$ to $10^{-12}$. In contrast, the volcanic material contained uranium to the extent of less than three parts per million, but had a $^{239}\text{Pu}$ to $^{238}\text{U}$ ratio of $10^{-7}$ to $10^{-9}$. In essence, the $^{239}\text{Pu}/^{238}\text{U}$ ratio exceeded that of uranium materials by a factor of $10^3$ to $10^5$. It is difficult to account for the $^{239}\text{Pu}/^{238}\text{U}$ ratio in volcanic specimens unless one assumes a neutron fluence found in or near a natural reactor of the Oklo type.
Sample 1371 in Fig. 1 has a uranium composition in the parts per million range similar to the uranium content of volcanic material. The neutron fluence calculated for this sample would produce an initial $^{239}\text{Pu}/^{238}\text{U}$ ratio of about $10^{-2}$. If the reactor then ceased operation, the $^{239}\text{Pu}/^{238}\text{U}$ ratio would decline to $10^{-6}$ after 300,000 years. It would also still be detectable after that length of time, even though almost all of the fission products would have decayed to stable elements. $^{239}\text{Pu}$ then represents one of the most likely isotopes that would be accidently found from natural reactor zones which operated tens of thousands of years ago. If there is a connection between natural reactors and volcanism, then it might be possible to detect more recent events with fission products as well as with $^{239}\text{Pu}$.

Conclusions

Most of the krypton and xenon in Oklo sample 1348 was produced by the fission of $^{235}\text{U}$. An anomalous component, extracted at low temperatures, seems to have been produced in an environment where $^{239}\text{Pu}$ was the major fuel source. This component was formed in a fluence of at least $3.5 \times 10^{21}$ neutrons/cm$^2$, and the calculated $^{239}\text{Pu}$ contribution to $^{86}\text{Kr}$ is 61%. Since these values are much higher than the normal rare gas component of sample 1348, the anomalous component probably formed at another location and later migrated to sample 1348. The source of this component may have been a zone which produced fissionable material more rapidly than it was consumed.
REFERENCES

5. Baudin, G., C. Blain, R. Hagemann, M. Kremer, M. Lucas, L.
   Merlivat, R. Molina, G. Nief, F. Prost-Marehal, F. Regnaud &
7. Drozd, R. J., C. M. Hohenbert & C. J. Morgan. Earth and
8. Shulkolyukov, Y. A., Ashkinadze, A. B. Verkovskiy, Dang Va
   Min, A. N. Komarov, O. A. Levchenkov, G. V. Ovchinnikova,
   V. V. Pavshukov, V. D. Sprintson & S. Z. Yakovleva, Geo.
9. Sherrill, R. D. "Rare Gas Isotopic Abundances in Meteoritic
   and Terrestrial Samples." Ph.D. Thesis (University of
   Arkansas, 1977).
11. Hyde, E. K. The Nuclear Properties of the Heavy Elements,


Table 1 Normalized to $^{136}\text{Xe}$

<table>
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<tr>
<th>Source</th>
<th>$^{136}\text{Xe}$ (cm$^3$/g·10$^{-9}$)</th>
<th>$^{136}\text{Xe}$</th>
<th>$^{134}\text{Xe}$</th>
<th>$^{132}\text{Xe}$</th>
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Yields**

| $^{235}$U (Thermal) | | | | | | | |
|-------------------|-------------|-------------|-------------|-------------|-------------|-------------|
| 1.00 | 1.25 | 0.672 | 0.411 | 0.105 |

| $^{239}$Pu (Thermal) | | | | | | | |
|-------------------|-------------|-------------|-------------|-------------|-------------|-------------|
| 1.00 | 1.102 | 0.7875 | 0.5935 | 0.2576 |

* Taken from the report of Shulkolyukov, et al.\textsuperscript{8,17}, they report an error of 0.5–2.0%.

** Uranium fission yields are from Shulkolyukov et al.\textsuperscript{8,17}, plutonium fission yields are from Meek and Rider\textsuperscript{12}. 
Table 2* Normalized to $^{136}$Xe

<table>
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<th>Sample</th>
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<th>$^{134}$Xe</th>
<th>$^{132}$Xe</th>
<th>$^{131}$Xe</th>
<th>$^{130}$Xe</th>
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<td>334</td>
<td>1.00</td>
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<td>1.39</td>
<td>0.875</td>
<td>0.609</td>
<td>0.0033</td>
<td>0.182</td>
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</table>

* Taken from the report of Shulkolyukov, et al.®.
<table>
<thead>
<tr>
<th>Sample</th>
<th>Temp. °C</th>
<th>$^{136}$Xe $(\text{cm}^3/\text{g} \cdot 10^{-9})$</th>
<th>$^{86}$Kr</th>
<th>$^{84}$Kr</th>
<th>$^{83}$Kr</th>
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</thead>
<tbody>
<tr>
<td>1348-a</td>
<td>400</td>
<td>334</td>
<td>1.00</td>
<td>0.602</td>
<td>0.331</td>
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<tr>
<td>(2.9 mg)</td>
<td>750</td>
<td>1000</td>
<td>1.00</td>
<td>0.568</td>
<td>0.304</td>
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<tr>
<td></td>
<td>1000</td>
<td>-</td>
<td>1.00</td>
<td>0.526</td>
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<td>6790</td>
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</table>

**Yields**

|          | $^{235}$U (thermal) | 1.00 | 0.5151 | 0.2762 |
|          | $^{239}$Pu (thermal) | 1.00 | 0.6304 | 0.3933 |

* Taken from the report of Shulkolyukov, et al.\(^8\). The data are normalized to $^{86}$Kr.

** From Meek and Rider\(^{12}\).
Table 4

<table>
<thead>
<tr>
<th>Sample</th>
<th>Fluence (n/cm)</th>
<th>Plutonium fission contribution* (percent)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>τ(129-130)</td>
<td>τ(131-132) (a)</td>
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<tr>
<td>1348 (400°)</td>
<td>3.5 x 10²¹</td>
<td>61 ± 11</td>
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<tr>
<td>1348 (750°)</td>
<td>4.4 x 10²⁰</td>
<td>35 ± 10</td>
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<tr>
<td>1348 (1150°)</td>
<td>6.6 x 10²⁰</td>
<td>9 ± 9</td>
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<tr>
<td>1348 (1350°)</td>
<td>5.5 x 10²⁰</td>
<td>5 ± 8</td>
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<tr>
<td>1361</td>
<td>1.1 x 10²¹</td>
<td>1.2 x 10²¹</td>
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<tr>
<td>1364</td>
<td>8.0 x 10²⁰</td>
<td>1.5 x 10²¹</td>
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<tr>
<td>1368</td>
<td>8.7 x 10²⁰</td>
<td>1.0 x 10²¹</td>
</tr>
<tr>
<td>1371</td>
<td>2.1 x 10²¹</td>
<td>7.8 x 10²⁰</td>
</tr>
<tr>
<td>321**</td>
<td>4.0 x 10²⁰</td>
<td>4.0 x 10²⁰</td>
</tr>
</tbody>
</table>

* Plutonium fission contribution was calculated by the following methods:
  (a) Partition method on $^{136}$Xe abundance using $^{129}$Xe.
  (b) Partition method on $^{136}$Xe abundance using $^{131}$Xe.
  (c) Partition method on $^{134}$Xe using coupled equations for xenon.
  (d) Partition method using coupled equations for krypton.

The plutonium fission contribution was calculated using an error of 2% for the Oklo xenon and krypton. The actual error of some of the above values may be less than indicated since Shulkolyukov et al.\textsuperscript{17} reported an error range of 0.5-2.0%, depending on the isotope.

** From Drozd, et al.\textsuperscript{7}.
Fig. 1 Neutron fluence for Oklo samples calculated from xenon isotope ratios. Sample 1348 is calculated from the 1350° fraction as being most representative of the location without the influence of the anomalous component.
Fig. 2 Krypton isotope ratios in 400° fraction of Oklo sample 1348. The horizontal lines are the yields expected from pure $^{235}$U fission or pure $^{239}$Pu fission.
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T. V. Crawford, Research Manager
Environmental Transport Division
Savannah River Laboratory