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2.05 X 10^9 YEAR U-Pb AGE OF THE OKLO URANIUM DEPOSIT*

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ABSTRACT

U and Pb isotopic data on samples (10 to 100 gram) 2 to 10 m away from the borders of the Oklo reactor zones indicate a primary age of 2.05 x 10^9 years for the Oklo deposit and a secondary age of 0.375 x 10^9 years. All samples show effects of Pb loss; the average loss is 50%. Both the U-Pb and Pb isotopic data are consistent with a model of a primary 2.05 x 10^9 year age of the deposit, continuous volume diffusion of Pb from uraninite, and either continuous or recent loss of this Pb. In this case the 0.375 x 10^9 year age is an artifact without time significance. Using an average value of D/a^2 3.5 x 10^{-11} a^{-1} (Cowan, this conference) this model explains the apparent 1.8 x 10^9 year Pb age observed by other workers. From the $^{208}Pb/^{206}Pb$ data our average U/Th value calculated for the Oklo deposit is ∿ 100.

*Work performed under the auspices of the U.S. Energy Research and Development Administration.

INTRODUCTION

The determination of age of the Oklo uranium deposit and the time when portions of this deposit behaved as a natural uranium fission reactor both have been the subject of a number of investigations. Weber and Bonhomme [1] summarized the Rb-Sr and K-Ar isotopic data and the geologic relationships, many of which appear to remain poorly defined. In general, isotopic data on rocks of the Franceville series to which the Oklo uranium deposit belongs yield ages of $\sim 1.8 \times 10^9$ years, whereas isotopic data on igneous and metamorphic rocks marginal to the sedimentary Franceville basin indicate that the age of deposition could be as great as 2.2×10^9 years. The K-Ar data on dolerite samples [1] yield ages of $\sim 0.85 \times 10^9$ years which they suggest is the time of intrusion of these dolerites into the Franceville series and also yield ages of $\sim 0.50 \times 10^9$ years which they suggest is the time of faulting and folding of the series.

U and Pb isotopic data reported by Lancelot et al. [2] do not yield a well-defined age, although these authors suggest an age of 1.75×10^9 years. Similarly, Devillers et al. [3] reported U and Pb isotopic data which do not yield a well-defined age and conclude that the age could be from 1.8 to 2.05×10^9 years.

Using data on the abundances of Nd, Sm, Ru, and Th isotopes produced in the reactor, Devillers et al. [3] deduced an age of 2×10^9 years for the reactor. Cowan et al. [4] modeled the reactor by assuming the reactor operated during the period of U deposition and by varying the accretion rate calculated ages ranging from 1.8 to 2.1×10^9 years. Similarly, Bryant et al. [5] using the different published thermal neutron capture cross sections on ^{143}Nd calculated ages of the reactor of 1.8 to 2.1×10^9 years. Ruffenach et al. [6] have calculated from U fission data an age of $2.0 \pm 0.1 \times 10^9$ years for the reactor.

In this report U and Pb isotopic data are presented on U ore samples peripheral to the reactor zones. These samples are from 2 to 8 meters from the borders of the reactor zones and were analyzed in an attempt to clarify the age of the deposit and to see to what extent Pb of anomalous isotopic composition produced from the U of anomalous isotopic composition has migrated from within the reactor zones and deposited in these peripheral regions.

ANALYTICAL TECHNIQUES

Samples weighing from 10 to 100 g along with analyses of U concentrations and U isotopic abundances were supplied through the courtesy of R. Naudet of the French Atomic Energy Commission. 2 to 10 mg aliquots were taken for the Pb isotopic analyses. The samples were dissolved in a mixture of HCl and HNO_3. This procedure does not dissolve silicate minerals and in all cases the silicate residue consisted primarily of quartz. Pb was separated using conventional ion exchange techniques. U was separated from the sample using Dowex 1-X8 (100-200 mesh) and eluting Pb with 10 N HCl, followed by an anion exchange column Dowex 1-X8 (100-200 mesh) to separate Pb from the alkalis and alkaline earths, and finally by a cation exchange column Dowex 50-X8 (100-200 mesh) incorporating the techniques described by Tera and Wasserburg [7]. Pb was analyzed on an AVCO model 9100 mass spectrometer using the silica gel - thermal ionization technique.

Pb concentrations were determined by X-ray fluorescence on 5 g aliquots of the samples.

ANALYTICAL RESULTS

The U and Pb concentration and isotopic data are presented in Table I. The U concentrations vary by a factor of 10 as do the Pb concentrations, and the Pb/U values range from 0.1 to 0.3. All of the samples have "normal" U isotopic abundances.

The Pb data are plotted on Figure 1. We have chosen to plot the data on this $^{207}Pb/^{206}Pb$ versus $^{204}Pb/^{206}Pb$ representation rather than the more conventional $^{206}Pb/^{204}Pb$ versus $^{207}Pb/^{204}Pb$ diagram because of the large $^{206}Pb/^{204}Pb$ values. For reference we have included an insert on this figure on which modern and primordial Pb isotopic abundances are plotted. The bar labelled "Figure" represents the region in which the data plot. In general the data define a linear array; the line has a very steep slope indicating the presence of extremely radiogenic "initial" Pb and it intersects the $^{207}Pb/^{206}Pb$ axis at a value corresponding to a time of 0.375×10^9 years. There is deviation of the data about the line significantly outside of error, a point to which we will subsequently return. We note that these data are quite different from those reported by Lancelot <u>et al</u>. [2] and Devillers <u>et al</u>. [3]. Our measured $^{206}Pb/^{204}Pb$ values range from 4000 to 9000 whereas those reported by Lancelot <u>et al</u>. [2] on samples with normal U isotopic abundances range from 1000 to 5000 and those reported by Devillers <u>et al</u>. [3] range from only 1000 to 2000. It is not clear whether the relatively low $^{206}Pb/^{204}Pb$ values are due to large laboratory contamination of the samples by these workers or if there are large amounts of ^{204}Pb in the silicate minerals which these workers dissolved and we did not.

On a plot of $^{208}Pb/^{206}Pb$ versus $^{204}Pb/^{206}Pb$ (not illustrated) the data also define a good linear array. The colinearity of the data indicate that <u>all</u> the samples have the same Th/U value. The steep slope of the line again indicates the presence of a very radiogenic "initial" Pb and the intersection of the line at the $^{208}Pb/^{206}Pb$ axis is consistent with an age of 0.375×10^9 years.

The data corrected for common Pb using $^{206}Pb/^{204}Pb = 15.34$ and $^{207}Pb/^{204}Pb = 15.57$ are listed in Table II. The * symbol indicates the corrected data. These data are plotted on Figure 2 and with one exception form a precise linear array whose lower intersection with concordia is 0.375×10^9 years. The upper intersection is 2.05×10^9 years which we interpret as the age of formation of the Oklo uranium deposit. This age of 2.05×10^9 years is greater than the $\sim 1.8 \times 10^9$ year age of the Franceville series determined from Rb-Sr isotopic data. It is also greater than the U-Pb age presented by Lancelot <u>et al</u>. [2], but as noted earlier their data did not precisely define an age. The age is, however, in agreement with the ages calculated by modeling the reactor using rare earth element isotopic data [3,4,5].

The age of 0.375×10^9 years determined by the U-Pb data (Figure 2) is the same as the age determined by the ^{207}Pb-^{206}Pb-^{204}Pb data (Figure 1) and is consistent with the ^{208}Pb-^{206}Pb-^{204}Pb data. The usual interpretation of these U-Pb data is that 2.05×10^9 years is the primary age of the uranium deposit and 0.375×10^9 years is the time of an episodic disturbance with Pb isotopic homogenization and redistribution. All of the samples we analyzed plot to the right of concordia indicating only Pb loss during the 0.375 x 10^9 year "event". In Table II the fraction of Pb lost at this time ($\mathcal{L}$) is tabulated for each sample. The losses range from 20% to 80%; the average loss is 50%.

From the U-Pb and Pb isotopic systematics we calculate for the Oklo deposit at 0.375 x 10^9 years average values of $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{206}Pb/^{204}Pb$ of 46, 500, and 3450, respectively. Assuming normal common Pb isotopic abundances as the initial Pb at 2.05 x 10^9 years, these Pb isotopic abundances at 0.375 x 10^9 years correspond to an average $^{238}U/^{204}Pb$ value (calculated as today's equivalent) in the interval 2.05 to 0.375 x 10^9 years of 11,000 (equivalent to U/Pb = 1500) and an average U/Th value of 60 which corresponds to an enrichment of U relative to Th by a factor of $\sim$ 250 compared to the average U and Th abundances in rocks.

None of the isotopic data, heretofore, presented has indicated geologic activity at 0.375 x 10^9 years which could be associated with Pb redistribution. Cowan [8] suggests that the extensive Pb loss observed throughout the Oklo deposit is due to diffusion of Pb out of uraninite and subsequent transport out of the deposit. Following the conventions established by Tilton [9] the locus of points for values of D/a^2 (i.e. a time and temperature independent value of the diffusion coefficient, D, and an effective grain radius, a) and an initial age of 2.05 x 10^9 years intersects the concordia at 2.05 x 10^9 years, passes through the colinear array of data points on Figure 2, and curves to a sub-horizontal line at $^{207}Pb^*/^{206}Pb^*$ = 0.09. Thus, 2.05 x 10^9 years is the primary age and the age of 0.375 x 10^9 years is an artifact of diffusion as a consequence of extending the linear portion of the diffusion locus from $^{207}Pb^*/^{206}Pb^*$ = 0.13 to 0.09 to intersect concordia. This age, therefore, has no time significance.

The linear array which we observe could be produced if Pb diffused from uraninite grains from 2.05 x 10^9 years until recently and recently various amounts of this Pb were removed from the system. The value of the $^{207}Pb^*/^{206}Pb^*$ removed would be 0.15 (see Figure 2) and thus the maximum $^{207}Pb^*/^{206}Pb^*$ observable age (assuming one had a pure Pb sample) would be 2.2 x 10^9 years and using the value of $D/a^2 = 3.5 \times 10^{-11}\ a^{-1}$ (Cowan [8]) the average $^{207}Pb^*/^{206}Pb^*$ age would be 1.8 x 10^9 years which is that observed by Lancelot <u>et al</u>. [2] and Devillers <u>et al</u>. [3].

We also note that samples which at 2.05 x 10^9 years had the same value of $^{238}U/^{204}Pb$ will as a consequence of diffusion form a linear array on a $^{207}Pb/^{206}Pb$ versus $^{204}Pb/^{206}Pb$ plot which extrapolates to a $^{207}Pb/^{206}Pb$ value corresponding to an age of 0.375 x 10^9 years. The mixture then of ^{204}Pb-rich Pb from silicate minerals with the Pb remaining in the uraninite grains would produce a scatter of data points which on the $^{207}Pb/^{206}Pb$ versus $^{204}Pb/^{206}Pb$ diagram would converge to τ = 0.375 x 10^9 years. This may explain the scatter in our data array (Figure 1) and the even greater scatter when we plot the Pb isotopic data of Lancelot <u>et al</u>. [2] and Devillers <u>et al</u>. [3] and also explain the relatively ^{204}Pb-rich <u>nature</u> of the samples <u>analyzed</u> by these coworkers.

From our samples which exhibit the least Pb loss we calculate an average U/Th for the Oklo deposit of $\sim$ 100. This value is consistent with the estimate of Naudet [10].

In summary it appears that the explanation of our data in terms of a volume diffusion model explains why 0.375×10^9 years is not an age indicated by other isotopic data, explains the $\sim 1.8 \times 10^9$ year age observed by other workers and explains how Pb is lost from the deposit.

ACKNOWLEDGMENTS

Stimulating discussions with my colleagues George Cowan and Ernest Bryant were critical in developing the ideas presented in this paper.

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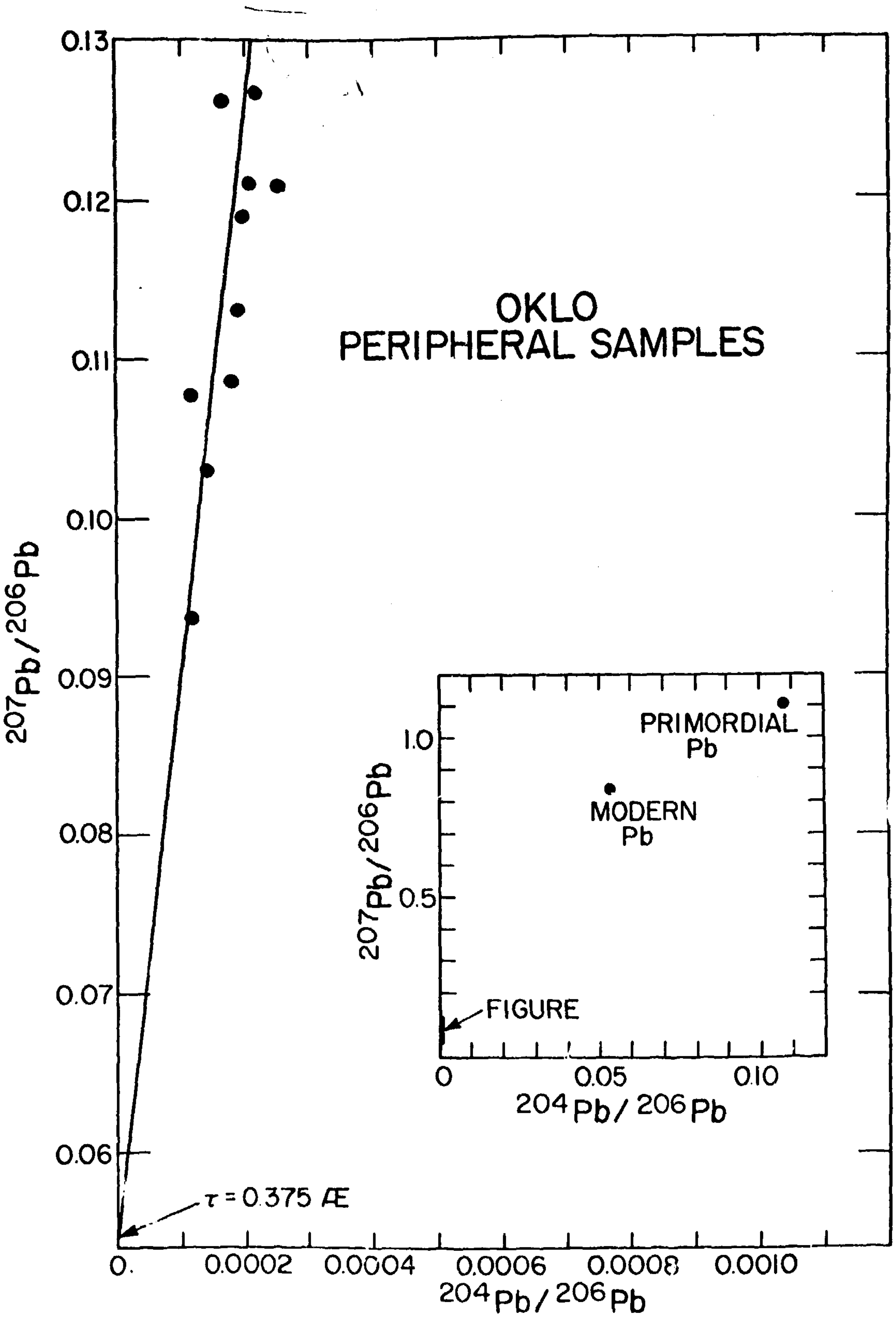
OKLO
PERIPHERAL SAMPLES
$^{207}Pb/^{206}Pb$
$^{204}Pb/^{206}Pb$
0.13
0.12
0.11
0.10
0.09
0.08
0.07
0.06
0.
0.0002
0.0004
0.0006
0.0008
0.0010
$\tau = 0.375$ Æ
PRIMORDIAL
Pb
MODERN
Pb
FIGURE
1.0
0.5
0
0.05
0.10

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FIGURE 1 to accompany manuscript by GANCARZ-- 2.05 x 10^9 year U-Pb age of the Oklo Uranium Deposit.

12/12/77

FIGURE 1 : The data for samples peripheral to the Oklo reactor zones are plotted on this $^{207}Pb/^{206}Pb$ versus $^{204}Pb/^{206}Pb$ diagram. The linear array extrapolates to the $^{207}Pb/^{206}Pb$ axis at a point corresponding to 0.375 x 10^9 years. The age is calculated using $\lambda(^{238}U)=0.155125 \times 10^{-9}a^{-1}$ and $\lambda(^{235}U)=0.98485 \times 10^{-9}a^{-1}$.

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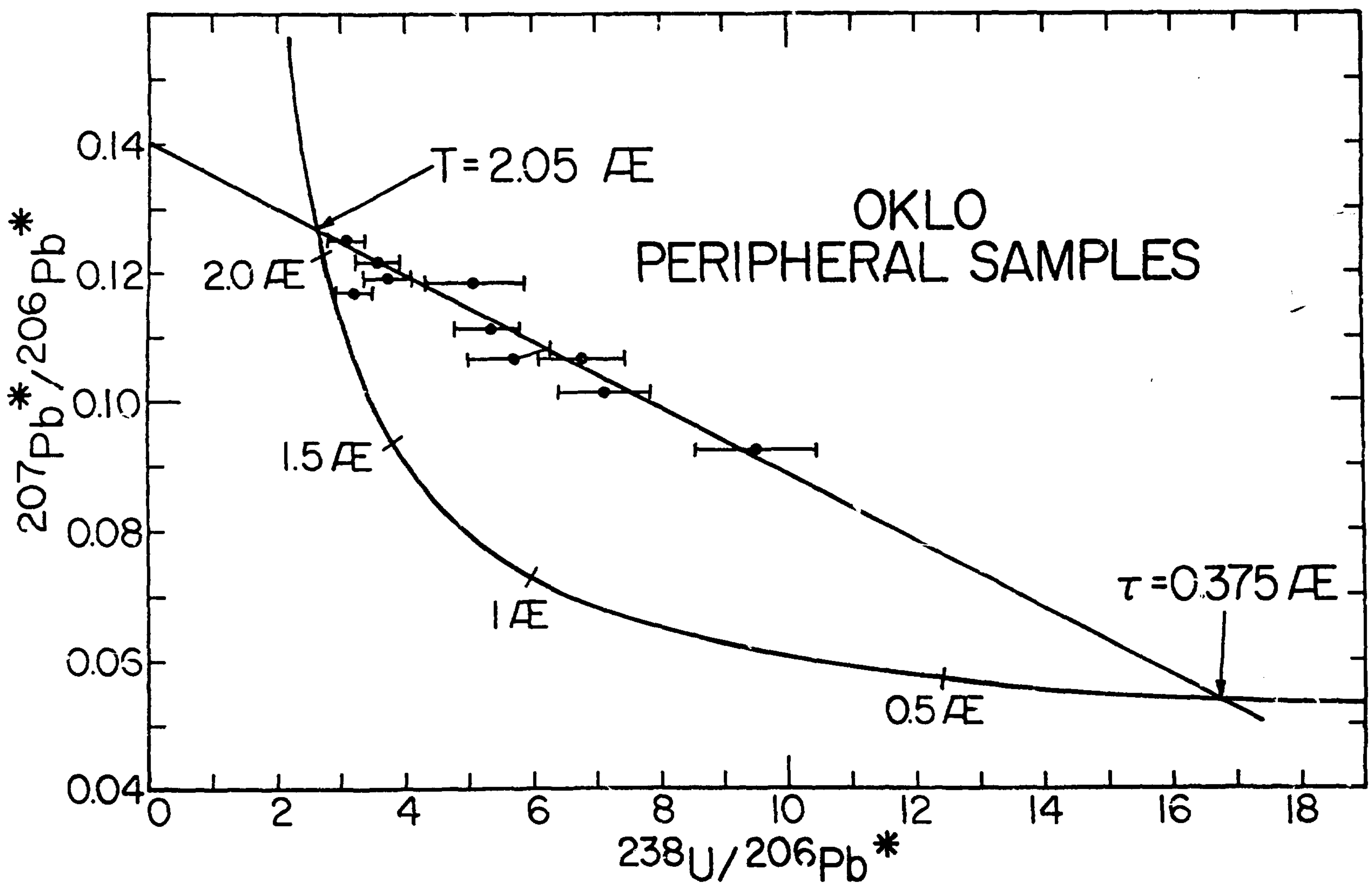

OKLO
PERIPHERAL SAMPLES
T = 2.05 Æ
2.0 Æ
1.5 Æ
1 Æ
0.5 Æ
τ = 0.375 Æ
0.14
0.12
0.10
0.08
0.06
0.04
0
2
4
6
8
10
12
14
16
18
207Pb*/206Pb*
238U/206Pb*

'IGURE 2

FIGURE 2 to accompany manuscript by GANCARZ-- 2.05 x 10^9 year U-Pb age of the Oklo Uranium Deposit 12/12/77

FIGURE 2: U and Pb isotopic data for samples peripheral to the Oklo reactor zones. The concordia curve is calculated using $\lambda(^{238}U) = 0.155125 \times 10^{-9} a^{-1}$ and $\lambda(^{235}U) = 0.98485 \times 10^{-9} a^{-1}$. The data define a linear array whose upper intersection with concordia is 2.05 x 10^9 years which we interpret as the primary age of the Oklo deposit.

TABLE I

OKLO PERIPHERAL SAMPLES: U-Pb DATA

Sample		Wt.% U	Wt.% Pb	Pb/U	$^{238}U/^{235}U$	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$
N 270	between Zones 1 & 2	13.3	1.9±0.2	.14	138.1	0.00859±1	0.10851±4	0.000180±1
CO 2252		~3.2	0.62±0.06	.19	137.9	0.01270±1	0.12099±2	0.000256±1
N 258	peripheral to	~2	0.27±0.03	.14	138.0	0.00773±12	0.10291±5	0.000140±3
N 266	Zones 5 & 6	8.5	2.7±0.3	.32	137.8	0.00769±2	0.12645±3	0.000167±3
N 244		2.7	0.83±0.08	.31	137.8	0.00877±1	0.11887±2	0.000198±1
N 255		5.3	0.98±0.10	.18	137.9	0.01030±1	0.11310±6	0.000219±2
N 268		~1.3	0.34±0.03	.26	138.0	0.01006±1	0.12117±3	0.000206±1
N 224	peripheral to	3.0	0.30±0.03	.10	138.1	0.00522±1	0.09369±2	0.000112±1
N 236	Zones 3 & 4	1.5	0.41±0.04	.27	138.9	0.01061±1	0.12630±2	0.000179±1
N 241	15 m S. Zone 4	8.3	1.4±0.1	.19	137.9	0.00591±3	0.10760±3	0.000115±1

TABLE II

SAMPLE	$^{238}U/^{206}Pb^*$	$^{207}Pb^*/^{206}Pb^*$	£
KN 270	6.74	0.10600	0.72
SCO 2252	5.05	0.11747	0.56
KN 258	7.09	0.10095	0.74
KN 266	3.15	0.12417	0.18
KN 244	3.16	0.11614	0.18
KN 255	5.24	0.11006	0.58
KN 268	3.84	0.11834	0.36
KN 224	9.43	0.09210	0.85
KN 236	3.59	0.12336	0.30
KN 241	5.69	0.10600	0.63

FIGURE CAPTIONS

FIGURE 1:

The data for samples peripheral to the Oklo reactor zones are plotted on this $^{207}Pb/^{206}Pb$ versus $^{204}Pb/^{206}Pb$ diagram. The linear array extrapolates to the $^{207}Pb/^{206}Pb$ axis at a point corresponding to 0.375×10^9 years. The age is calculated using $\lambda(^{238}U)=0.155125 \times 10^{-9}\ a^{-1}$ and $\lambda(^{235}U)=0.98485 \times 10^{-9}\ a^{-1}$.

FIGURE 2:

U and Pb isotopic data for samples peripheral to the Oklo reactor zones. The concordia curve is calculated using $\lambda(^{238}U)=0.155125 \times 10^{-9}\ a^{-1}$ and $\lambda(^{235}U)=0.98485 \times 10^{-9}\ a^{-1}$. The data define a linear array whose upper intersection with concordia is 2.05×10^9 years which we interpret as the primary age of the deposit.