LA-8644-PR

Progress Report

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Oklo Natural Fission Reactor Program

April 1—August 31, 1980

LOS ALAMOS SCIENTIFIC LABORATORY

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Oklo Natural Fission Reactor Program

April 1—August 31, 1980

Compiled by

David B. Curtis

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OKLO NATURAL FISSION REACTOR PROGRAM

April 1--August 31, 1980

Compiled by

David B. Curtis

ABSTRACT

An interim report has been published on the redistribution of uranium, thorium, and lead in samples representing several million cubic meters of sandstone and metamorphosed sediments in the Athabasca Basin which is located in the northwest corner of the Canadian province of Saskatchewan. The region of study includes zones of uranium mineralization at Key Lake. Mineralization occurs at the unconformity between the Athabasca sandstone and the underlying metasediments and in fault zones within the metasediments.

Lead isotopes record a radiometric age of 1300 ± 150 m.y. in samples from above and below the unconformity. This age probably reflects the time of deposition of the sandstones and an associated redistribution of uranium and/or lead in the underlying rocks. Many of the samples have been fractionated with respect to radiogenic lead and the actinide parent elements since that time. Sandstones and altered rocks from the region above the unconformity have been a transport path and are a repository for lead. In contrast, mineralized rocks are deficient in radiogenic lead and must be an important source of lead in the local geologic environment.

Samples from Oklo reactor zone 9 and nearby host rocks have been prepared for isotopic analyses of ruthenium, molybdenum, uranium and lead.

I. PERSONNEL

This report covers some of the activities of the following individuals.

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II. PROGRAM OBJECTIVE

Geologic burial is the favored method being considered for the disposal of commercially generated radioactive wastes. Efforts to evaluate the effectiveness of geologic media in isolating the wastes from the biosphere are limited by the necessity to consider periods of time much greater than that recorded by human experiences. The Oklo Natural Fission Reactor Program is a research effort to study the retention and migration of elements in the Earth's crust by examining the geologic record which does extend over appropriate periods of time.

III. PROGRESS REPORTED PREVIOUSLY

Funding for the United States' participation in the international investigation of the Oklo natural fission reactor phenomenon commenced in fiscal year 1975. The first formal report of progress in this program was the annual report of fiscal year 1976, issued in November 1976. The Oklo phenomenon refers to the occurrence of self-sustaining fission chain reactions in a series of very rich uranium ore pockets located in an extensive Precambrian

pitchblende deposit in Gabon, Africa. The uranium formed critical masses about 2 x 10^9 years ago. The duration of criticality was several hundred thousand years. During criticality, approximately 10 tons of uranium were fissioned. Studies of the Oklo phenomenon have shown that many fission products were retained at the sites where they were generated. Other products from uranium decay or fission have been shown to be missing in whole or in part from the fossil reactor cores. Our studies have examined the spatial, temporal and chemical features of the processes that fractionated and redistributed the fission products and radioactive decay products. Previous reports have discussed the fractionation and redistribution of ^{99}Tc relative to Ru, Ru relative to U and Pb relative to U.

These studies have indentified continous volume diffusion of radiogenic lead from uraninite grains as the process by which lead is fractionated from uranium. Transport paths have been identified in the ores and in the rocks which confine the ores. Limits have been placed on the times when the elemental redistributions occurred.

Elemental and isotopic analyses primarily using mass spectrometric methods are the principal analytical methods used in these studies. The development of appropriate analytical techniques have comprised an important part of the efforts. The results of this development work has been reported in previous programmatic reports.

IV. PROGRESS DURING THE HALF YEAR APRIL 1, 1980 - AUGUST 31, 1980

A. Fractionation and Redistribution of Lead, Uranium and Thorium at the Key Lake Uranium Deposit, Saskatchewan, Canada

To extend our studies to physical, chemical and geologic environments different than those found at Oklo, samples were obtained from rich Precambrian uranium deposits at Key Lake. The major effort during this half year has been the analysis of these samples and the interpretation of the results. An interim report on the Key Lake studies has been issued (Curtis and Gancarz, 1980). A synopsis of the report follows:

1. Introduction

The Key Lake deposit is one of several rich uranium deposits discovered in the Athabasca Basin in the northwest corner of the Canadian province of Saskatchewan. It offers an excellent set of circumstances for the study of

the retention and migration of uranium, thorium, and lead over geologic periods of time. Ores containing large quantities of uranium and lead are confined in host rocks that have "normal" quantities of these elements. Because of the extraordinary composition of the ores, the lead isotopic composition is significantly different than that of lead formed in situ in the country rocks. This disparity provides a means of identifying mixtures of lead that originated in different sources.

Drill cores, taken as part of the exploration program, have provided well documented samples from the mineralized region. These samples, which include ores and unmineralized host rocks have been analyzed for lead, uranium, and thorium concentrations and lead isotopic abundances. The goal of the study is to understand the chronology of the rocks, assess the extent of uranium and lead migration, identify sources, transport paths and repositories for these elements and try to understand the mechanisms involved in the processes.

2. Geology of Key Lake

Key Lake is located on the southeastern edge of the Athabasca Formation, a fluviatile, quartz sandstone deposit. The age of this deposit has been determined to be 1350 ± 50 m.y. by Rb/Sr dating (Raemaekers and Dunn, 1976) and 1330 ± 30 m.y. from Pb isotopic data (Gancarz, 1978). The Athabasca sandstone unconformably overlays rocks of the Wollaston Domain, a major structural province of the Canadian Shield. The highly deformed basement complex consists of synforms and antiforms with granitoid Archean cores rimmed by Proterozoic metasediments. (Hoeve and Sibbald, 1978). Radiometric dating of the granitoid basement show it to be at least 2.5×10^9 years old. Multiple periods of metamorphism are observed in the metasediments. The major metamorphic period occurred about 1.7×10^9 years ago with episodes as recently as 1.57×10^9 years (Cummings and Scott, 1976; Money, 1968). At the unconformity there is a transition zone that lithologically resembles the Athabasca sandstone. However, there is a decrease in quartz and an increase in phyllosilicates with depth, grading into the crystalline basement. The entire region is overlain by Pleistocene glacial debris.

Pre and post Athabasca faults are important structural features at Key Lake. They trend northeast-southwest and dip at 50° to 70° to the northwest, roughly parallel to the stratigraphic dip of the basement metasediments. The Key Lake ore bodies are localized in the fault zones which appear to impose a

structural control on the mineralization. Uranium and nickel are the major metallic elements in the ores, with smaller abundances of lead, zinc, and copper. Concentrations of the two major elements often run into the tens of percent. Three uranium minerals have been identified (Dahlkamp, 1978). An oxide called pitchblende I, a more oxidized variety of pitchblende, called sooty pitchblende or pitchblende II, and the silicate mineral coffinite. The arsenosulfide, gersdorfite is the most abundant nickel mineral with nickel arsenides and sulfides also being prominent phases. Galena is ubiquitous throughout the ores.

3. Sample Descriptions and Locations

Table I contains brief descriptions of the samples that have been analyzed. Also included in this Table are approximate distances from the nearest ore-bearing regions. Locations of material that resided at or above the unconformity are oriented relative to the contact between the sandstone and the basement and to the nearest ore that occurs at the contact. Basement rocks are oriented by distance downdip from the nearest ore bearing basement rocks and through the stratigraphic section relative to the principal ore bearing strata.

Samples taken from above the uncomformity represent a maximum horizontal separation of 300 meters in a north-south sense and 500 meters in an east-west sense. Samples KL-12 and KL-1 are the samples of sandstone furthest removed from uranium rich regions. Samples KL-10 and KL-15, and KL-11 and KL-13, are each a pair of samples separated from one another vertically in the same core. The sample identified as KL-25 was taken from the basement near Zimmer Lake, a region well removed from known mineralization, about 5 km to the southwest of Key Lake. It was analyzed to provide reference data for basement rocks.

4. Experimental

Approximately ten grams of rock were extracted from the interior of core sections chosen for analysis. These interiors were crushed and sieved through 100 mesh screens. One gram aliquots were taken for determination of uranium and thorium concentrations. Concentrations of these two elements were determined by neutron activation analysis techniques using the automated analysis system at the Los Alamos Omega West Reactor. Lead isotope abundances are measured by isotope dilution mass spectrometry on 200 to 500 mg aliquots of the powdered samples. Analytical results are presented in Table II.

TABLE I

DESCRIPTION OF KEY LAKE SAMPLES

SAMPLES AT OR ABOVE THE UNCONFORMITY

Sample No.	Description	Distance Above Unconformity (Meters)	Horizontal Distance From Nearest Ore (Meters)
KL-1	Sandstone	40	180
KL-7	Sandstone	10	10
KL-8	Sandstone	5	35
KL-9	Sandstone	55	10
KL-10	Sandstone	45	70
KL-11	Sandstone	25	10
KL-12	Sandstone	2	150
KL-16	Sandstone	2	80
KL-13	Pyrite bearing contact	0	10
KL-15	Oxidized contact	0	70

SAMPLES BELOW UNCONFORMITY

	Description	Stratiographic Distance From Ore Bearing Zone (Meters)	Horizontal Distance from Nearest Ore (Meters)
KL-19	Graphic gneiss	+45	0
KL-20	Quartz from quartz vein	+45	0
KL-21	Contact between KL-20 and basement rock	+40	0
KL-14	Pyrite bearing contact between KL-17 and KL-18	-15	0
KL-17	Quartz from quartz vein	-15	0
KL-18	Basement rock 10 cm from KL-17	-15	0
KL-2	Quartz-biotite-feldspar gneiss	-30	140
KL-25	Pyrite bearing basement	Sample Taken at Zimmer Lake 5.5km from Key Lake Deposits	

SAMPLES OF ORE

Sample No.	Description
KL-3	Friable ore from contact zone
KL-6	Massive pitchblende from contact zone

TABLE II

U-Th-Pb DATA FROM KEY LAKE SAMPLES

Sample	Pb g/g rock[a]	U g/g rock[b]	Th g/g rock[b]	^{206}Pb nm/g rock	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{204}Pb/^{206}Pb$	α^c	β^c	γ	μ
KL-1	7.99×10^{-6}	9.5×10^{-7}	2.8×10^{-6}	10.65	1.880 ± 1	0.6934 ± 7	0.0431 ± 2	23.20	16.09	43.62	8.69
KL-2	1.17×10^{-5}	7.3×10^{-7}	3.4×10^{-6}	13.83	2.133 ± 6	0.8919 ± 17	0.0568 ± 3	17.61	15.70	37.55	3.90
KL-3	3.9×10^{-3}	1.5×10^{-1}	—	17400.	0.031 ± 3	0.049 ± 4	<0.0002	—	—	—	—
KL-6	6.0×10^{-2}	4.9×10^{-1}	—	256000.	0.0613 ± 7	0.0740 ± 8	<0.0003	—	—	—	—
KL-7	7.64×10^{-5}	5.0×10^{-6}	4.7×10^{-6}	107.7	1.750 ± 1	0.6309 ± 4	0.0387 ± 1	25.87	16.32	45.28	5.04
KL-8	7.42×10^{-6}	2.02×10^{-6}	2.4×10^{-6}	10.53	1.713 ± 1	0.6480 ± 5	0.0398 ± 1	25.14	16.29	43.06	20.2
KL-9	3.00×10^{-5}	2.33×10^{-6}	3.9×10^{-6}	40.78	1.818 ± 3	0.6863 ± 10	0.0425 ± 2	23.53	16.15	42.78	5.65
KL-10	7.56×10^{-6}	1.12×10^{-6}	2.2×10^{-6}	10.24	1.834 ± 2	0.6878 ± 7	0.0427 ± 1	23.42	16.11	42.95	10.8
KL-11	1.16×10^{-4}	8.8×10^{-6}	1.2×10^{-5}	168.3	1.667 ± 2	0.6272 ± 6	0.0382 ± 1	26.18	16.42	43.64	5.78
KL-12	1.46×10^{-6}	5.8×10^{-7}	3.8×10^{-6}	1.815	2.143 ± 2	0.6954 ± 7	0.0436 ± 1	22.93	15.95	49.14	30.8
KL-13	2.03×10^{-4}	1.28×10^{-5}	$<3 \times 10^{-6}$	330.1	1.436 ± 1	0.5081 ± 5	0.02974 ± 7	33.62	17.08	48.29	5.48
KL-14	1.89×10^{-6}	1.11×10^{-5}	$<2.5 \times 10^{-6}$	4.65	0.688 ± 2	0.2624 ± 8	0.01260 ± 6	79.37	20.83	54.63	790
KL-15	1.04×10^{-5}	5.0×10^{-6}	2.84×10^{-5}	13.9	1.93 ± 1	0.63 ± 3	0.0398 ± 4	25.77	16.32	49.8	38.9
KL-16	9.44×10^{-5}	8.6×10^{-6}	1.03×10^{-5}	148.7	1.460 ± 3	0.5699 ± 6	0.0340 ± 2	29.41	16.76	42.94	7.1
KL-17	7.21×10^{-7}	1.07×10^{-6}	$<2 \times 10^{-6}$	1.44	1.038 ± 12	0.3512 ± 6	0.0193 ± 3	51.87	18.22	53.81	161
KL-18	3.57×10^{-6}	1.11×10^{-5}	4.2×10^{-6}	9.68	0.5550 ± 56	0.2187 ± 22	0.0097 ± 49	103.1	22.55	57.2	493
KL-19	1.07×10^{-5}	9.6×10^{-6}	1.77×10^{-4}	8.88	4.472 ± 7	0.3267 ± 7	0.01680 ± 9	59.52	19.45	266.2	268
KL-20	3.06×10^{-7}	7.7×10^{-7}	$<4 \times 10^{-6}$	0.54	1.32 ± 1	0.401 ± 10	0.019 ± 1	52.3	20.0	69.5	312
KL-21	2.78×10^{-6}	2.4×10^{-6}	8.4×10^{-6}	5.10	1.24 ± 1	0.377 ± 3	0.0203 ± 2	49.3	18.65	61.1	97
KL-25	1.18×10^{-5}	3.8×10^{-6}	—	16.5	1.70 ± 1	0.698 ± 3	0.0435 ± 4	22.98	16.05	39.1	22.0

[a] Isotope dilution

[b] Delayed neutron counting technique

[c] $\alpha = {}^{206}Pb/{}^{204}Pb$; $\beta = {}^{207}Pb/{}^{204}Pb$; $\gamma = {}^{208}Pb/{}^{206}Pb$; $\mu = {}^{238}U/{}^{204}Pb$

5. Results and Discussion

a. Chronology. With a single exception, sample KL-20, non-ore samples form a linear array on a lead isotope diagram that define an age of 1.3×10^9 years. Data in the linear array include those from Athabasca sandstones, highly altered rocks from the contact zone and metasediments from the basement. This age is the same as that determined for the Athabasca by Raemaekers and Dunn (1976). Presumably it reflects the time of deposition of the Athabasca sediments. Metasediments from deeper in the Wollaston Domain than those found in the basement complex at Key Lake, all have radiometric ages older than 1.56×10^9 years (Cummings and Scott, 1976). It is likely that deposition of the sedimentary overburden established chemical conditions that reset the uranium-lead radiometric clock in the underlying rock.

Although the 1.3×10^9 year age is clearly imprinted on the rocks, several of the data fall significantly off the best linear fit to all the data. These deviations reflect lead-uranium fractionation in the last 1.3×10^9 years or the isotopic signature of a previous geologic history.

Two samples of ore contain relative abundances of lead isotopes that fall significantly below the best linear fit to the non-ore data. The lead in these ores is almost entirely radiogenic and have isotopic compositions that could only have evolved from the decay of uranium in closed systems over periods of time significantly less than 1.3×10^9 years.

b. Element Fractionation. If the rocks at Key Lake had a uniform lead isotopic composition 1.3×10^9 years ago and there had been no uranium-lead fractionation in the meantime, the abundances of ^{206}Pb relative of ^{238}U are simply defined by equation (1).

$$\left(^{206}Pb/^{204}Pb\right)_o = \left(^{206}Pb/^{204}Pb\right)_I + \left(^{238}U/^{204}Pb\right)_o \left(e^{\lambda T} - 1\right). \qquad (1)$$

The subscript o refers to present ratios, and I refers to the isotopic ratio in the rock when it was formed. Using measured values of $(^{238}U/^{204}Pb)$, and a value of $(^{206}Pb/^{204}Pb)_I$ and $T = 1.3 \times 10^9$ years inferred from the linear array of the lead isotope ratios, values of $(^{206}Pb/^{204}Pb)_o$ can be calculated for each sample. Agreement between this calculated value and the measured one indicate rocks that have neither lost nor gained lead or uranium for 1.3×10^9 years. Two samples from above the unconformity are of this type and appear to have been closed chemical systems during this time. A similar conclusion is

reached by examining the relative abundances of ^{232}Th and its radiogenic progeny ^{208}Pb. The remaining eight samples from above the unconformity contain large excesses of ^{206}Pb and ^{208}Pb relative to the predicted quantities.

Three of the seven basement rocks contain relative abundances that do not vary from those predicted within the uncertainties of the measurements. Two of the rocks from 15 meters below the ore are significantly depleted in lead relative to uranium. Another two samples, one taken 5 km from Key Lake, have relative lead excesses that constitute 20-30% of the total lead in the rocks. Both of the ore samples that have been analyzed are deficient in lead. Again using the 1.3×10^9 years age, the ore sample KL-6 is depleted in lead by a factor of two, and KL-3 by a factor of seven.

c. Element Migration. There is a consistent pattern of element fractionation in rocks from above the Athabasca unconformity. The majority of samples from this region are enriched in radiogenic lead or depleted in uranium and thorium. Only two do not show this pattern, these are unfractionated with respect to lead and its parent elements. The isotopic systematics cannot distinguish between lead excesses and uranium and thorium deficiencies. However, a comparison of lead abundances in the sandstones suggests that lead was transported to, and deposited in, these rocks: Sample KL-12, the only sandstone that does not contain a relative excess of radiogenic lead, has an absolute abundance of lead that is one to two orders of magnitude less than in any of the other sandstones. We conclude that large volumes of rock above the unconformity have been a transport path for a mobile phase and are a repository for lead.

Work is in progress to establish a relationship between the excess lead found in the sandstone and the lead missing from the ores. If this relationship can be established, transport paths from the ores into the overlying rocks can be identified by the presence of excess radiogenic lead.

B. Multielement Fractionation and Redistribution at Oklo Reactor Zone 9

A cross section of reactor zone 9 and the rocks enclosing it were exposed at the bottom of the Oklo mine during the sampling trip by A. J. Gancarz in September 1979. This provided a unique opportunity to obtain samples at well defined locations in and around an area that had sustained nuclear criticality. These samples will be analyzed for a chemically diverse suite of fission and

decay products to identify chemical fractionation patterns associated with the transport of elements over distances of less than 10 meters.

The exposed cross section of the reactor zone was a narrow band extending about four meters in the N-S direction and only a few cm in the E-W direction. Seven samples were taken from the reactor zone. These represent a fairly comprehensive sampling through the entire exposed section of the zone. The samples were sent to Bill Maeck at Exxon Nuclear Corporation where they were pulverized and prepared for isotopic analysis. Analyses of these samples will assess the extent to which nuclear products have been redistributed within the reactor as opposed to being lost from the zone of reaction and redistributed in the surrounding rocks.

Fourteen samples from the nearby host rocks were selected and pulverized in preparation for isotopic analyses. These samples consist of a traverse across 6.4 meters to the east of the reactor zone; three E-W traverses at distances of 2.5, 4.4 and 6.5 meters to the south of the reactor zone; two samples from near the basal conglomerate 2.5 meters west of the reactor and a sample from the F_B strata that overlies the ore-bearing section. Four composite samples were prepared from 1) the traverse to the east; 2) the E-W traverse at 2.5 meters south; 3) the E-W traverse 4.4 meters south and 4) the E-W traverse 6.5 meters to the south. These composites and the three uncomposited samples were split. Half was sent to Bill Maeck for Mo, Ru and U isotopic analyses and half was retained at LASL for lead isotopic analysis.

C. Problems

Shifts in the responsibilities of various individuals in the program delayed the submittal of progress reports. This report covers the work of the last two quarters of FY-80. In the future every attempt will be made to meet the reporting requirements for the program.

D. Activities Planned for Next Quarter

The isotopic analyses of rhuthenium, molybdenum, uranium and lead in the samples from around Oklo Reactor Zone 9 will be completed in the next quarter. Samples representing a traverse of several hundred meters away from the ore bearing regions at Oklo will be prepared for lead and uranium analysis and these analyses will begin. Procedures to measure the isotopic composition of barium will be tested. These procedures will be used as part of the study of multi-element redistribution around reactor zone 9.

Minerals in samples from the Athabasca sandstone and from the transition region at the unconformity with the basement will be selectively dissolved. The various portions will be analyzed for lead isotopic composition to determine the relative abundances of excess lead in the rocks and the lead that is indigeneous to them.

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