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Progress Report

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Oklo—Natural Fission Reactor Program

July 1—September 30, 1979

LOS ALAMOS SCIENTIFIC LABORATORY
Post Office Box 1663 Los Alamos, New Mexico 87545

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Andrew A. E. Norris

CONTENTS

Department of Energy
Mr. C. R. Cooley
Bld. Code GTN
MS B107
Washington, D. C. 20545

Department of Energy
Dr. John A. Patterson
Bld. Code FED
RA-224, MS-6521
Washington, D.C. 20461

Dr. Wayne A. Carbiener (6)
Battelle Office of Nuclear Waste Isolation
505 King Avenue
Columbus, OH 43201

Mr. R. Y. Lowrey
Weapons Production Division
Albuquerque Operations Office
P. O. Box 5400
Albuquerque, NM 87115

Prof. Paul Kruger
Department of Civil Engineering
Stanford University
Stanford, California 94305

Prof. Douglas G. Brookins
Department of Geology
University of New Mexico
Albuquerque, New Mexico 87131

Mr. W. J. Maeck
Exxon Nuclear Idaho Company, Inc.
Radiochemistry Subsection
Post Office Box 2800
Idaho Falls, Idaho 83401

Dr. J. E. Delmore
Exxon Nuclear Idaho Company, Inc.
Radiochemistry Subsection
Post Office Box 2800
Idaho Falls, Idaho 83401

Los Alamos Scientific Laboratory
G. A. Cowan, ADCEL, MS-102
D. C. Hoffman, CNC-DO, MS-760
J. E. Sattizahn, CNC-11
E. A. Bryant, CNC-11
D. B. Curtis, CNC-11
C. J. Duffy, CNC-11
A. J. Gancarz, CNC-11
A. E. Norris, CNC-11
R. J. Vidale, CNC-11

OKLO-NATURAL FISSION REACTOR PROGRAM

July 1 — September 30, 1979

by

A. E. Norris

ABSTRACT

Nearly 200 samples were collected at the Oklo mine in Gabon this quarter for shipment to the United States to continue studies of lead, ruthenium, and technetium migration around natural fission reactors. The first analyses of samples collected near a rich uranium ore body in Canada show the presence of radiogenic lead in pyrite and sandstone materials. Analyses of additional samples are underway to permit the interpretation of the data in terms of transport paths. A technique was developed this quarter to eliminate the interference of organic materials during the mass spectrometric analyses of ruthenium in Oklo samples with high asphaltic contents. A proposal was drafted for a study of naturally occurring radionuclide migration at rich uranium ore bodies in Australia to be performed jointly by the U. S. Department of Energy and the Australian Atomic Energy Commission.

I. PERSONNEL

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

Los Alamos Scientific Laboratory

E. A. Bryant
G. A. Cowan
D. B. Curtis
A. J. Gancarz
A. E. Norris

Idaho National Engineering Laboratory

J. E. Delmore
F. A. Duce
W. J. Maeck
R. A. Nielson

University of New Mexico

D. G. Brookins

II. PROGRAM OBJECTIVE

The goal of this program is the determination of rates of reactor product migration and the mechanisms of transport in geologic media that include natural fission reactors or rich uranium ore bodies.

III. PROGRESS REPORTED PREVIOUSLY

Funding for this program, which includes 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, West Africa. The uranium formed critical masses about 2×10^9 years ago. The duration of criticality was several hundred thousand years. During this time, approximately 6 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. Principal objectives of our current studies of the Oklo phenomenon are identification of the migration paths of some of the mobile fission products and reconstruction of the paleohydrology and transport history of the Oklo site.

Lead was chosen as the first element to be investigated for tracing transport paths. Lead is not a fission product, but it is formed from the radioactive decay of uranium. Previous work has shown that ~70% of the radiogenic lead is missing from the Oklo reactor site. The Oklo setting appears to be favorable for tracing lead transport, because the common lead background is low and the quantity of radiogenic lead that was produced can be calculated. However, when this work was begun, we did not know whether we would be able to obtain requisite samples from the Oklo location. Therefore, we undertook the measurement of radiogenic lead transport in the vicinity of a rich uranium ore body in Canada. This undertaking is providing valuable, because the geologic setting differs from the Oklo provenance and should provide useful information concerning lead transport in a high grade metamorphic rock.

The results of analyses of the Oklo samples that were shipped to us for lead transport studies have been given in a previous quarterly report. Analyses of the data indicated that a major transport path and repository involved the conglomerate material underlying the Oklo natural reactor zones. The data could not be used to infer the method of lead transport from the uraninite grains to the basal conglomerate, but diffusion appeared to be the mechanism by which lead was removed from the uraninite grains, where it was formed. Water flow, we presume, was the means of transport.

Ruthenium was chosen as the second element to be investigated for tracing transport paths. Measurements of ruthenium isotope abundances in conjunction with uranium analyses permit the determination of the gain or loss of fissiogenic ruthenium relative to uranium. Furthermore, because ^{99}Ru is formed by the decay of 2.13×10^5 year ^{99}Tc, it is possible to determine from ^{99}Ru measurements the gain or loss of ^{99}Tc relative to uranium and ruthenium during the time when the ^{99}Tc was decaying. Last quarter we reported the detection of isotopic anomalies at distances as far as 10 meters from a natural fission reactor. These anomalies indicated the redistribution of fissiogenic ruthenium and technetium relative to each other and to uranium. This observation is a significant step toward determining element transport paths and migration rates in a natural environment. Additional work in these ruthenium migration studies has involved development of a uranium-ruthenium age dating technique and isotopic analyses of ruthenium in nanogram quantities.

IV. PROGRESS DURING THE CURRENT QUARTER

A. Additional Oklo Samples

Dr. A. J. Gancarz travelled to the Oklo mine in Gabon, Africa, this quarter to collect samples for additional studies of element migration in the vicinity of the natural fission reactors. The collected samples, approximately 200 in number, can be categorized as follows:

1. Samples from reactor zone 9. The samples were collected from traverses spaced at 3 to 5 meters. The intervals between samples ranged down to 10 cm.
2. Core library samples of rock from a stratigraphic unit containing the natural fission reactors. These samples contain pyrite. They were collected at distances ranging to ~1 km from the reactor zones.
3. Samples collected in an underground mine. These samples are from locations physically lower than the natural fission reactors, but from the stratigraphic unit that contains the reactor zones.
4. Samples containing galena and pyrite from fault and fracture zones in the Oklo mine.
5. Samples of powdered ore. Each sample is an aliquot of a batch of ore that weighed 40 to 60 tons.

When these samples are received at the Los Alamos Scientific Laboratory, an extensive analytical program will begin for the purpose of determining the distances to which radiogenic lead and fissiogenic ruthenium and technetium were transported from the Oklo natural fission reactor zones.

B. Lead Migration Studies at Key Lake

Work is underway to measure uranium and lead concentrations and lead isotopic abundances in samples selected from the 402 that were collected at the Key Lake, Canada, uranium ore deposit. The purpose of this work is to measure the rate of radiogenic lead transport in a geologic regime distinctly different from that at the Oklo location. To date, uranium concentrations have been measured in 21 samples. Lead contentrations and isotopic abundances have been measured in ten of these 21 samples. The data indicate that samples from the Athabasca sandstone and a pyrite rich rock from the contact between the sandstone and the crystalline basement are enriched in radiogenic lead relative to uranium. A pyrite rich sample of basement rock underlying the uranium ore is

relatively deficient in radiogenic lead. Additional work is necessary before statements can be made about likely transport paths for this radiogenic lead, particularly in regard to its transport in the metamorphic basement rock. Our analytical work on the suite of 402 samples is continuing.

C. Techniques for Isotopic Analyses of Ruthenium

The chemical procedure developed for mass spectrometric analyses of nanogram amounts of ruthenium in uranium ore gave poor results when certain Oklo samples were analyzed. The asphaltic content of these ores was sufficiently high that organic material carried through not only one RuO_4 distillation, but even through a second distillation. It was not possible to oxidize the organic material to effect a separation from ruthenium, because any oxidation step vigorous enough to oxidize the organic material would oxidize ruthenium to RuO_4, which would distill, too.

A successful solution to this problem was found by modifying the procedure to include a precipitation that carries most of the ruthenium, while leaving the organic material behind. In this modification, ruthenium is distilled from the solution of ore, then ferric iron is added in sufficient quantity to permit its good recovery as the hydroxide. Sodium hydroxide is added to the solution to precipitate $Fe(OH)_3$, which carries the ruthenium. The solid phase is separated, dissolved, and redistilled. Ruthenium yields as high as 98% have been obtained with ths procedure. Furthermore, the results of mass spectrometric analyses are free from organic interferences, even when ruthenium is separated from ores with high asphaltic content.

D. Joint Proposal for Migration Studies in Australia

Mr. Phillip J. Shirvington of the Australian Atomic Energy Commission visited the Los Alamos Scientific Laboratory from July 16 through July 18 to discuss a possible collaboration between the U.S.A. and Australia to measure the migration of naturally occurring radionuclides at selected Australian uranium ore bodies. A three-year collaborative effort has been proposed to measure the migration distances and transport rates of ^{234}U, ^{230}Th, and ^{226}Ra at the Jabiluka Two uranium ore body; to compare radionuclide migration behavior in zones where uranium is in the 6+ oxidation state *versus* migration in zones where uranium is in the 4+ valence state; and to compare measurements of uranium sorption and desorption in laboratory experiments with information obtained

about sorption and desorption when uranium has been in contact with the sorbant material in nature for times of the order of 10^5 years. The ^{234}U and ^{238}U analyses required for this work would be performed with mass spectrometric techniques at the Los Alamos Scientific Laboratory, while the ^{230}Th and ^{226}Ra analyses would be done by the Australian Atomic Energy Commission research establishment. The sorption-desorption experiments would be done jointly.

The proposal for this collaboration was completed at the end of this quarter. It is being submitted for approval and authorization to the appropriate Australian and United States agencies.

E. Conclusions

The trip to the Oklo mine in Gabon was successful in its goal of collecting additional samples for our studies of element migration in the vicinity of natural fission reactors. While the samples are in transit, we are analyzing the Key Lake ore samples to study radiogenic lead migration in a high grade, metamorphosed rock environment. Finally, a chemical problem that hindered measurements of ruthenium isotopic abundances in some Oklo ore samples with high asphaltic content appears to be solved.

F. Communications

The dissemination of the results of this work is a continuing part of our program. This quarter the following talks given by Prof. D. G. Brookins included aspects of this work.

July 12, 1979, Lawrence Livermore Laboratory, "Argillaceous Rocks as Radioactive Waste Repositories: Evidence from Oklo."

July 31, 1979, Institute for Behavior Research, University of New Mexico, "The Ends of the Nuclear Fuel Cycle: Uranium Resources and the Disposal of Radioactive Waste."

August 22, 1979, East Mesa Civitan Club, Albuquerque, New Mexico, "Geological Aspects of Radioactive Waste Disposal: Oklo, WIPP, and other Examples."

September 20, 1979, Lawrence Berkeley Laboratory, "Oklo and Radioactive Waste."

V. PROBLEMS

No problems are hindering the progress of this work within its funding and manpower levels.

VI. ACTIVITIES PLANNED FOR NEXT QUARTER

The activities planned for next quarter include continuation of the analytical work on the Key Lake samples to measure radiogenic lead migration and presentation of a paper entitled "^{99}Tc, Pb, and Ru Migration Around the Oklo Natural Fission Reactors" at the International Symposium on the Scientific Basis for Nuclear Waste Management, to be held in Boston November 26 through 29.

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