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Progress Report

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Oklo—Natural Fission Reactor Program

October 1—December 31, 1978

University of California



LOS ALAMOS SCIENTIFIC LABORATORY

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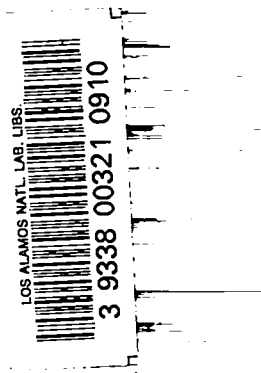
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A. E. Norris



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OKLO-NATURAL FISSION REACTOR PROGRAM

October 1--December 31, 1978

by

A. E. Norris

ABSTRACT

Journeys to France and to Canada this quarter succeeded in obtaining 421 samples for use in developing techniques to determine the migration paths of radiogenic lead away from rich uranium ore bodies. Work continues in the effort to develop quantitative elemental analyses of ruthenium and reliable measurements of ruthenium isotopic abundances, both at the nanogram level, to permit uranium-ruthenium age determinations of uranium ores. For this purpose, an additional 37 samples were collected in Canada.

I. INTRODUCTION

This report covers activities of the following individuals.

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II. PROGRAM OBJECTIVE

The goal of this program is the determination of mechanisms of reactor product 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 for 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 tonnes 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 the present investigation are identification of the migration paths of some of the mobile fission products and reconstruction of the paleohydrology and transport history.

Lead was chosen as the first element to be investigated for tracing transport paths because previous work has shown that ~ 70% of the radiogenic lead is missing from the Oklo reactor site, the common lead background is low, and procedures for both elemental and isotopic analyses are relatively easy to perform. The second element to be used for tracing transport paths is ruthenium. However, a reliable isotopic analysis procedure for ruthenium has not been developed yet. Developmental efforts were described in the previous quarterly report.

IV. ACTIVITIES DURING THE CURRENT QUARTER

A. Lead Samples

Radiogenic lead was chosen for the initial attempt to trace migration paths away from the Oklo reactors for the reasons given above. To accomplish the goal of this program, it is necessary both to collect lead samples to analyze for their radiogenic content and to learn about the geologic setting from which the samples come. These purposes were accomplished by means of journeys to France to collect Oklo samples, and to Canada to collect samples from the very rich Dielman uranium ore body at Key Lake, Saskatchewan. The Key Lake samples will be used to develop the required techniques for this work, to minimize the use of the relatively scarce Oklo samples.

From the French sources Oklo samples were collected that included six powdered rocks containing excess lead relative to uranium, plus one sample of authigenic pyrite and five samples containing PbS from the base of the uranium-bearing stratigraphic unit. In addition, the trip resulted in the gathering of much useful information concerning the geological setting of Oklo, the possibility that samples pertinent to this work will be collected in Gabon and sent here, and an invitation to participate in the collection of additional samples at Oklo next May.

From the Dielman ore body in Canada, 409 samples were collected, which are being shipped to Los Alamos. The samples are well documented in terms of stratigraphic and structural position relative to the ore body.

B. Development of a U-Ru Dating Technique

Prior studies have indicated that the measurement of ruthenium produced by the spontaneous fission of ^{238}U shows promise as a means of dating rich uranium ore deposits. Development of this technique would permit inferences to be made concerning ruthenium and technetium migration.

The procedure being used is that of determining U-Ru dates for comparison with U-Pb dates from the same samples. The difficulty lies in the quantitative assay of ruthenium and the reliable measurement of ruthenium isotopic abundances, both at the nanogram level. Efforts this quarter were concentrated on increasing the sensitivity of the mass spectrometric measurements of the ruthenium isotopic abundances.

The results of work on this problem during the quarter have shown that the sensitivity increases a factor of 10 to 20 when ruthenium is collected on an ion exchange resin bead before being mounted on the mass spectrometer filament, compared to evaporation directly onto the filament of a solution containing ruthenium of comparable concentration. The reason for this enhanced sensitivity is not understood. The resin bead technique may result in a concentrated point source of ruthenium on the filament or in some form of ruthenium occurring during decomposition of the resin bead that is more amenable to ionization than that produced by direct evaporation.

The distributions of ruthenium between various solutions and Dowex 1 or 2 resin beads are being measured to determine the maximum amount of ruthenium that can be incorporated onto a single 100-mesh resin bead. As much as 20 ng of ruthenium has been exchanged onto a single resin bead, when CH_3OH is added to the 8M HBr solution that is used to collect the distilled RuO_4 .

One problem is the undesirable interference of molybdenum in the ruthenium spectrum. The primary source of molybdenum is from impurities in the rhenium filament material. Intense pre-baking of the filament reduces the intensity of the molybdenum interference, but an amount sufficient to cause problems at mass 100 remains. Tantalum filaments were tried, because pre-baked tantalum is free of molybdenum, but the ionization was so small that the sensitivity per nanogram of ruthenium was considerably less than that from rhenium filaments. A new filament post assembly was designed for use with rhenium, in which only 2 to 3 mm of filament is heated. When the ion exchange bead is placed in the center of a short rhenium filament, the molybdenum interference is reduced by a factor of 5 to 10. However, this interference is still more than is desirable for precise results on small samples.

Another facet of the work to develop a U-Ru dating technique is the requirement for appropriate ore samples to compare U-Pb and U-Ru dates. This requirement was fulfilled through a trip to the Cluff Lake uranium ore body in Saskatchewan,

where 37 samples were collected for this work. These samples are being shipped to Los Alamos.

C. Conclusions

The work done this quarter has resulted in a supply of ore samples that significantly advance the progress of this program. However, the problem of reliable measurement of ruthenium for the U-Ru dating technique has not been solved.

D. Communications and Publications

An important aspect of this work is the dissemination of results. This quarter two papers were presented at scientific meetings.

1. D. G. Brookins, "Thermodynamic Considerations Underlying the Migration of Radionuclides in Geomedia: Oklo and Other Examples," Symposium on Science Underlying Radioactive Waste Management, Nov. 30, 1978, Boston. Abstract in Mat. Res. Soc. Ann. Mtg. Abs. 52-53 (1978).
2. D. G. Brookins, "Lanthanide and Actinide Migration from Oxidized Uranium Deposits," invited paper presented at the western annual meeting of the American Geophysical Union, Dec. 7, 1978, San Francisco. Abstract in EOS 59, 1224 (1978).

V. PROBLEMS

Further progress in development of a U-Ru dating technique depends on resolving the technical problems associated with the reliable analyses of ruthenium at the nanogram level.

VI. ACTIVITIES PLANNED FOR NEXT QUARTER

Lead isotopic analyses will be performed on some of the Oklo samples just brought here from France. The uranium ore samples collected in Canada should arrive shortly in Los Alamos. Work will begin then to analyze the Key Lake samples for elemental lead.