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LA-9377-PR
Progress Report

dr. 708

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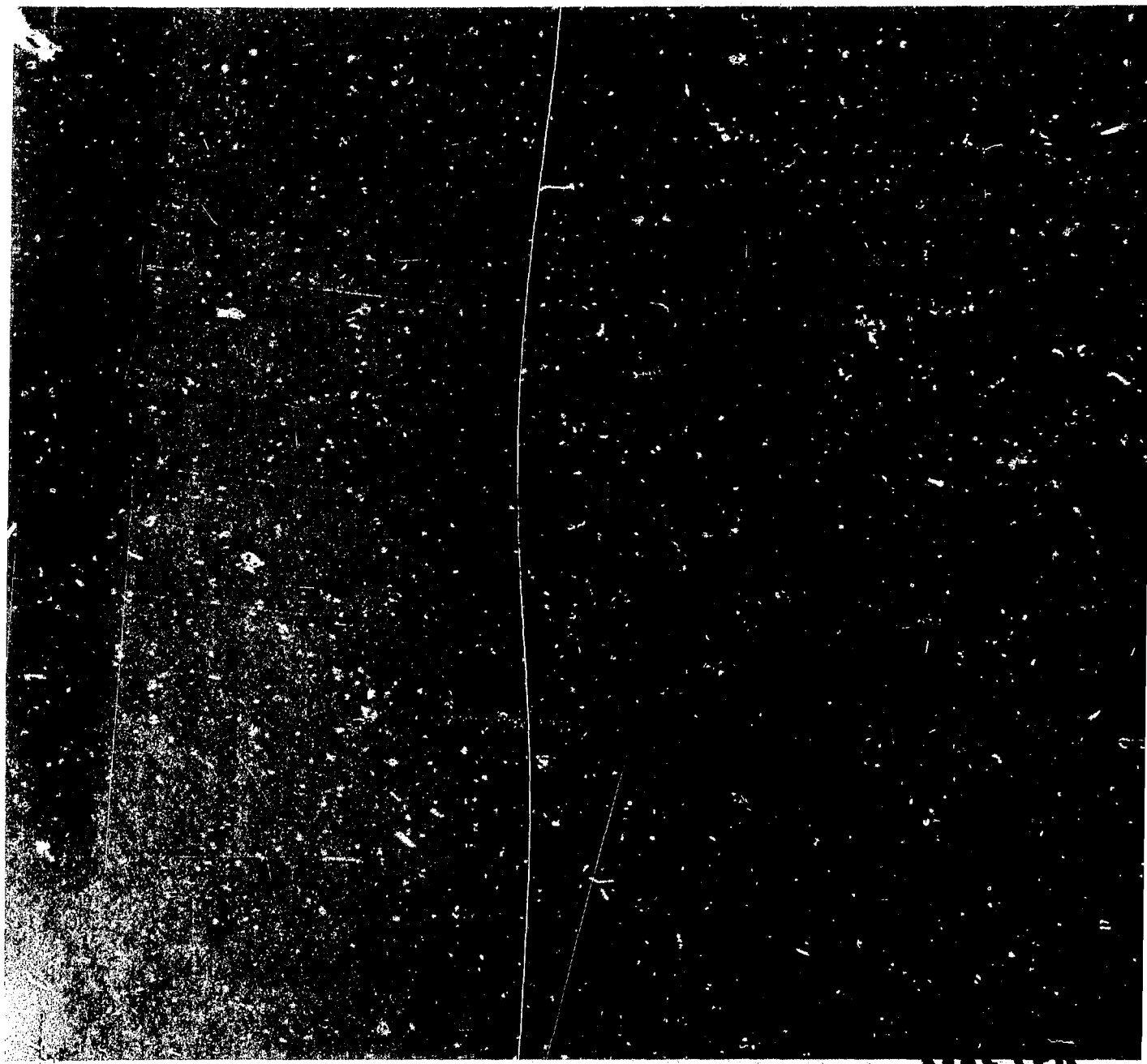

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Natural Repository Analogue Program

January 1—March 30, 1982

Compiled by
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NATURAL REPOSITORY ANALOGUE PROGRAM

January 1--March 30, 1982

Compiled by

David B. Curtis

ABSTRACT

Lead and uranium isotopic abundances in rocks from the Oklo mine show large deficiencies of radiogenic lead in the mineralized regions and enormous excesses of this element outside the uraniferous zones. A fracture lined with secondary minerals and its host rock from distances as far as ~13 meters away contain lead that was deposited contemporaneously. The isotopic composition of lead in these samples varies systematically as a function of distance from the fracture. This regularity may reflect the nature of the processes that transported lead from the ores and deposited it in the surrounding rocks.

I. PERSONNEL

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

Los Alamos National Laboratory

T. M. Benjamin

E. A. Bryant

J. H. Cappis

D. B. Curtis

D. J. Frank

A. J. Gancarz

A. E. Norris

D. J. Rokop

II. PROGRAM OBJECTIVE

Geologic burial is the favored method being considered for the disposal of commercially generated radioactive wastes. Appropriately chosen sites are thought to be relatively inert environments with respect to the stability of engineered waste containers. In addition, the rock itself will provide the ultimate containment, assuring that the wastes will remain isolated from the biosphere for adequate periods of time. However, efforts to evaluate the effectiveness of the geologic environment in this regard are limited by our understanding of the effects of natural, physical, and chemical processes acting over periods of time much longer than that recorded by human experiences. The Natural Repository Analogue program is a research effort to study the chemical processes and conditions that affect the retention and transport of elements in the earth's crust. The work examines the stability of elements in the geologic record and their response to natural conditions over appropriately long 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 a Precambrian pitchblende deposit in Gabon, Africa. The uranium formed critical masses 2×10^9 years ago. During criticality $\sim 10^4$ kg of ^{235}U were fissioned. Early work in the program determined the inventory of elements produced and destroyed by the nuclear reactions. Comparisons between the abundances of nuclear products produced in the natural reactors and those measured in them today provided a quantitative measure of the loss of elements from these rocks in the two-billion years since the reactors were critical. Uranium, the principal nuclear fuel, appears to have been extremely stable during this time. Individual grains of uraninite have remained intact since the time of nuclear criticality. Despite the apparent stability of the minerals that contained the nuclear fuel, many of the nuclear products have been lost from these mineral grains and redistributed into the geologic surroundings. Individual samples from the reactor zones have lost as much as

70% of the mass 99 (^{99}Tc + ^{99}Ru), 57% of the ruthenium, and 28% of the neodymium. It is estimated that about 100 kg of each of these chemical species was transported away from the zones of criticality. Proportional losses vary systematically with the intensity of nuclear activity, which suggests that the rate of loss was dependent on some property that varied with the degree of criticality (temperature, radiation dose, etc.). Abundances in rocks outside the reactor zones suggest that most of the ^{99}Tc and ruthenium was retained within a few tens of meters of the reactors. Redistribution of ruthenium and ^{99}Tc occurred two billion years ago, beginning with the initiation of nuclear criticality and spanning a period of time of less than one million years. Technetium was preferentially removed from the reactor zone relative to ruthenium. The observed geochemical properties suggest that loss of nuclear products was controlled by the rate of diffusion from uraninite, the mineral form of the primary nuclear fuel. Transport of ruthenium and technetium away from the rocks that sustained the nuclear fission and their redeposition in the surrounding rocks may have been controlled by temperature-dependent redox reactions.

To examine the redistribution of elements at Oklo under conditions less severe than those produced by sustained nuclear fission, the geochemical behavior of lead resulting from the decay of uranium is being studied. The rate of production of radiogenic lead (as controlled by the half-life of the uranium isotopes) is sufficiently slow that fractionation of lead and uranium during the critical period at Oklo would not perturb the isotopic composition of lead produced since that time.

IV. ACTIVITIES DURING THE PERIOD

Lead and uranium isotopic analyses have been made on 75 samples from the Oklo mines. Roughly half of these were done by French scientists and the other half at the Los Alamos laboratory. About 75% of these have been reported in the literature. The rest, which are mostly samples from outside the uranium mineralized region, have not yet been reported. There is a great consistency in the data: the uraniferous mineralized rocks are almost without exception deficient in radiogenic lead. Losses are as small as 10% and as large as 90% of the radiogenic ^{206}Pb that has been produced by radioactive decay in these rocks over the last 2×10^9 years. These losses represent many tons of lead that has been transported away from the Oklo

ores during this time. Conversely, virtually all of the rocks away from the mineralized zone contain enormous excesses of radiogenic ^{206}Pb. These excesses represent as little as a few percent and as much as five orders of magnitude more lead than could have been produced by uranium decay in these rocks.

One section of an unmineralized core contains a centimeter-size fracture lined with secondary minerals, the most obvious being quartz and galena (PbS). Isotopic analyses of the galena showed it to be composed of lead produced by the radioactive decay of uranium in an environment with a large $^{238}U/^{204}Pb$: the lead that was transported to and deposited in the fracture was from the nearby uranium ores. This fracture is encompassed by recrystallized sandstone, which typifies local strata of the Francevillian basin. Samples of the sandstone taken at distances of 2 cm and ~13 m from the fracture contain lead that is isotopically related to that in the walls of the fracture. Variations of $^{207}Pb/^{206}Pb$ vary linearly with changes in $^{204}Pb/^{206}Pb$ in these three samples. Such a relationship indicates that lead in the three samples is temporally related and will permit an evaluation of the time when the transport occurred. Two observations may be made with respect to the time of emplacement: it was instantaneous within the time resolution of the lead isotope chronometer, and it did not occur in recent geologic history. The ratio $^{206}Pb/^{204}Pb$ decreases as a function of distance from the fracture, which suggests a decreasing proportion of lead from the fracture as the distance from it increases, a functional relationship that might suggest the nature of the processes responsible for the element redistribution.

V. PRESENTATIONS DURING THE REPORTING PERIOD

David Curtis presented a seminar to the Department of Geological and Planetary Sciences at the California Institute of Technology on March 9, 1982. The title of the presentation was "The Geochemistry of Nuclear Products - Studies of the Oklo Natural Reactors."

David Curtis presented the Earth Sciences Colloquium at the University of California at Los Angeles on March 11, 1982. The title of the talk was "The Gechemistry of Nuclear Products - Studies of the Oklo Natural Reactors."

T. M. Benjamin made a presentation at the Symposium on Geochemistry of Nuclear Waste Disposal held in conjunction with the 183rd National Meeting

of the American Chemical Society in Las Vegas, Nevada, on March 3, 1982. The title of the presentation was "The Geochemistry of Tc, Ru, and Nd at the Oklo Natural Reactors."

David Curtis was co-organizer and co-chairperson of the Symposium on Geochemistry of Nuclear Waste Disposal held in conjunction with the 183rd National Meeting of the American Chemical Society in Las Vegas, Nevada, March 29-30, 1982.

VI. FUTURE ACTIVITIES

Our plans for future activities include completing the measurement of barium isotopic abundances in rocks from Oklo reactor zone 9 and the surrounding sandstone. We also plan to continue writing a manuscript on the geochemistry of lead, uranium, plutonium, and thorium at the Oklo uranium deposit and begin preparing a manuscript on the geochemistry of ruthenium, technetium, molybdenum, and neodymium at Oklo reactor zone 9.