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SEARCH FOR OTHER NATURAL FISSION REACTORS

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ABSTRACT

Precambrian uranium ores have been surveyed for evidence of other natural fission reactors. The requirements for formation of a natural reactor direct investigations to uranium deposits with large, high-grade ore zones. Massive zones with volumes $\gtrsim$ 1 m^3 and concentrations $\gtrsim$ 20% uranium are likely places for a fossil reactor if they are $\gtrsim$ 0.6 b.a. old and if they contained sufficient water but lacked neutron-absorbing impurities. While uranium deposits of northern Canada and northern Australia have received most attention, ore samples have been obtained from the following worldwide locations: the Shinkolobwe and Katanga regions of Zaire; Southwest Africa; Rio Grande do Norte, Brazil; the Jabiluka, Nabarlek, Koongarra, Ranger, and El Sharana ore bodies of the Northern Territory, Australia; the Beaverlodge, Maurice Bay, Key Lake, Cluff Lake, and Rabbit Lake ore bodies and the Great Bear Lake region, Canada. The ore samples were tested for isotopic variations in uranium, neodymium, samarium, and ruthenium which would indicate natural fission. Isotopic anomalies were not detected. Criticality was not achieved in these deposits because they did not have sufficient ^{235}U content (a function of age and total uranium content) and/or because they had significant impurities and insufficient moderation. A uranium mill monitoring technique has been considered where the "yellowcake" output from appropriate mills would be monitored for isotopic alterations indicative of the exhumation and processing of a natural reactor.

INTRODUCTION

In all likelihood, the Oklo natural fission reactor was not a singular occurrence in the earth's history. The conditions of the Precambrian earth were such that there were probably other reactors of this type. The combined probabilities of a reactor forming, of its surviving from Precambrian times, and of its discovery during mining suggest that Oklo was one of several natural fission reactors. If this is so, we should be able to find evidence for other occurrences. Subsequent study of other natural fission reactors would add to our knowledge of the history of the primordial earth and to such timely topics as the geochemistry of uranium ore deposits and the long-term behavior of buried reactor products. At Oklo, readioactive products were remarkably immobile, but their behavior in other environments could be an important consideration in the geologic disposal of contemporary nuclear wastes.

THE OCCURRENCE OF NATURAL REACTORS

The geochemical circumstances which led to the formation of Oklo were very special; a unique sequence of events assembled a critical mass of ^{235}U--the single most important ingredient of natural criticality. When the earth formed $\sim$ 4.5 b.a. ago, ^{235}U constituted about 23% of terrestrial uranium. But there were no known mechanisms to assemble a critical mass, even though a relatively small mass would have sufficed. Ore-forming processes were not likely as the core, mantle, and crust had not differentiated. By the end of the Hedean period 3.4 b.a. ago, uranium, being lithophilic, had concentrated in the mantle and crust and probably occurred as disseminated UO_2 in crystalline rocks. During the Archean period (about 3.4 to 2.6 b.a. ago), procaryotic life forms evolved; but there were no significant amounts of free oxygen during this time, and uranium remained as the unoxidized UO_2. Uranium did concentrate toward the end of this period, however, by means of gravitational sorting of heavy stream sediments, which included UO_2. These unoxidized ores (known as paleoplacer or quartz-pebble conglomerate ores) rarely contained more than a few tenths percent uranium, and even though the relative ^{235}U content was about 8%, there was insufficent total ^{235}U concentration for criticality.

Then, during the Lower Proterozoic (about 2.6 to 2.0 b.a. ago) important changes occurred. This was a period of intense mountain building and subsequent weathering. Also, photosynthetic blue-green algae evolved, and gradually the earth's surface waters and atmosphere became oxidizing. Around 2 b.a. ago, then, an extensive surface "reservoir" of unoxidized uranium-bearing sediments and orogeny-related exposures were attacked by oxygen, and relatively large quantities of complexed uranyl ion were hydrochemically released. When uranyl-bearing ground waters achieved sufficient depth or intersected zones of high organic concentration, the chemical environment became reducing, and uranium precipitated as uraninite, pitchblende, or coffinite. Where this process occurred in porous rock or cavities, large, massive veins of uranium often resulted. This cavity filling (or structural control) seems to be an important characteristic of Proterozoic "vein-type" deposits. Such vein deposits are almost exclusively limited to the Proterozoic, and they represent some of the world's richest uranium reserves. The relative ^{235}U content during the Proterozoic went from about 4 to 1%. This is about the minimum concentration required for any natural assemblage to attain criticality. Hence, nature seems to have cooperated in providing a mechanism for massive uranium accumulation at a very critical time for the formation of natural reactors.

The search for other fossilized natural reactors is guided by the physical requirements [1,2] for formation of a natural critical assembly. The natural reactor requirement of high ^{235}U content restricts investigations to uranium deposits that are very old and very rich. Neutronic arguments require that the reactor zone be relatively large. Thus, any deposits having zones of uranium mineralization that are (1) $\gtrsim$ 0.6 b.a. old, (2) $\gtrsim$ 20% uranium, and $\gtrsim$ 1 m^3 in size are worthy of investigation, regardless of the present day content of neutron-absorbing "poisons" (such as boron and the rare earths) or neutron moderators (such as carbon and water).

SURVEY OF POTENTIAL SITES

Proterozoic, vein-type uranium deposits are the most appropriate locations to search for other natural reactors. These deposits are found in exposures of Precambrian continental shields, which cover large areas of central and southern Africa, northern Canada, eastern South America, and portions of Australia and Eurasia. Not surprisingly, most of the important uranium deposits of the world are in these shield areas, but this investigation is concerned mainly with northern Canada and northern Australia--the two most promising locations. The extensive exploration and mining activities in these regions have provided an excellent opportunity to search for possible reactor zones.

In Canada, massive vein-type uranium deposits have been discovered in the Northwest Territories and northern Saskatchewan. At the depleted Eldorado mine at Port Radium, Northwest Territories, mineralized uranium ore veins have been dated at $\sim$ 1450 m.a. [3] and 1200-1400 m.a. [4]. Major minerals of the veins, which average $\sim$ 0.5 m thick, include quartz, dolomite, hematite, native silver, and chalcopyrite, with lesser amounts of pitchblende. However, massive specimens with $\gtrsim$ 40% uranium have been documented.

At the Ace-Fay-Verna mine [5,6] in the Beaverlodge district, Saskatchewan, epigenetic uranium occurs as disseminated or colloform pitchblende that fills brecciated and hematized fracture zones. These subsidiary zones are adjacent to the major St. Louis fault in the host metamorphics. High-grade mineralized veins up to $\sim$ 1 m thick with tens of percent uranium have been observed. Initial epigenetic mineralization occurred [7] at about 1780 ± 15 m.a. ago, but at least two major periods of pitchblende reworking [8] (or perhaps lead loss [9]) have been proposed: 1100 m.a. and 270 m.a. ago.

Likewise at the Rabbit Lake open-pit mine, Saskatchewan, massive and colloform pitchblende occur in brecciated regions of metasediments associated with a synclinal structure. Primary uranium emplacement took place about 1100 m.a. ago [10] but subsequent periods of remobilization are indicated by younger ages of sooty pitchblende [11]. Large, rich lenses of $\sim$ 30% uranium have been encountered at the Rabbit Lake mine.

The Key Lake, Maurice Bay, and Cluff Lake uranium deposits of the Athabascan region, Saskatchewan, are being delineated and assessed. For Key Lake, fault-controlled uranium mineralization is found at the unconformity of Proterozoic sediments and basement gneisses [12]. The initial mineralization is thought to be Middle Proterozoic ($\sim$ 1 to 2 b.a. ago), and large, high-grade regions have been encountered. High-grade nickel mineralization is associated with the uranium, but the paragenetic sequence of these species is not clear.

Descriptive geological or mineralogical information has not been reported for the Maurice Bay deposit. However, structurally-controlled veins of high-grade uranium mineralization occur, and the age of mineralization is probably Middle Proterozoic also.

At the Cluff Lake "D" ore body [10], structural control contributed to the formation of a very concentrated occurrence of pitchblende-uraninite. The complex uranium mineralization is associated with gold, tellurium, selenium, cobalt, bismuth, lead, and nickel. The ore body has an average grade of several percent uranium, and very high-grade zones in excess of 50% have been observed. Initial mineralization has been dated at 1050 m.a. ago [13] and 1150 m.a. ago [14].

In the regions of the Alligator Rivers and the South Alligator Valley of the Australian Northern Territory, recently discovered Proterozoic uranium deposits have mineralogy and formation controls remarkably similar to the northern Saskatchewan deposits [15,16]. At the Australian Nabarlek deposit, diamond drilling has intersected very rich zones, which have vertical thicknesses up to ∿ 1 m and concentrations to ∿ 60% uranium [17]. "Original" deposition of uranium has been dated at 850-900 m.a. ago [17,18] and 710-815 m.a. ago [19]. However at least one period of uranium redistribution may have occurred: at 450-500 m.a. ago [18,19].

The Koongarra deposit occurs in Lower Proterozoic schists above a reverse fault contact with Middle Proterozoic metasediments [20]. The uranium mineralization has been dated at 850 m.a. [21] and 870 m.a. old [16], and rich regions within the ore body have been observed. Koongarra uranium was probably redistributed around 450-500 m.a. ago [18].

The Jabiluka deposit is perhaps the largest high-grade uranium accumulation ever discovered. The uranium mineralization, which has been reported as 900 m.a. old [21], is thought to have resulted from hydrothermal remobilization and concentration of pre-existing uranium. Large mineralization zones with $\gtrsim$ 50% uranium have been identified in this ore body.

At the Ranger deposit, vein-type pitchblende is found in fracture zones in a Lower Proterozoic schist. But unlike the other ore deposits in the Alligator Rivers region, the structurally-controlled uranium veins of Ranger are generally less than 1 cm thick [22] and reported as being 1600 m.a. old [21].

For these Australian deposits, information on trace element mineralization is scarce. In general, however, uranium is the only economic mineral species [23], but significant accumulations of gold were observed at Jabiluka [21].

According to one theory of uranium ore genesis [21,24], the uranium of the Alligator Rivers region is syngenetic with the Lower Proterozoic sediments; this deposition would have occurred before 1880 m.a. ago. The uranium then underwent concentration at ∿ 1700 m.a. (possibly by mobilization and deposition of pre-existing uranium accumulations) and experienced periods of lead loss at ∿ 900 m.a. and ∿ 500 m.a. ago. Accordingly, the relatively recent dates given for uranium mineralization at Nabarlek, Koongarra, and Jabiluka may not represent the age of the first major concentrations of uranium at these deposits. The existing structurally-controlled accumulations may have been derived from similar, parent accumulations. Or, alternatively, the existing accumulations may be nearly twice as old as their uranium-lead ages suggest. In any event, these deposits have appropriate ages for this investigation.

Ore samples from the previously described Canadian and Australian deposits, together with samples obtained from the Frondel mineral collection of Harvard, were analyzed for isotopic variations in uranium, neodymium, samarium, and ruthenium (some analyses have been previously reported [25]). The samples from the Canadian and Australian deposits were collected from large, high-grade zones in the deposits, but detailed descriptions of these ore configurations are not presented for proprietary reasons. For the Harvard samples, descriptions of the specific sample locations were unavailable; but the samples, in general, were from massive hand specimens.

If the samples were from natural reactor zones, ^{235}U would exhibit a relative depletion,[1] whereas neodymium, samarium, and ruthenium in the samples would have relative increases in their respective fission-product isotopes. Samples from Nabarlek, Key Lake, and Maurice Bay were screened for ^{235}U/^{238}U variations with a field measurement system described elsewhere [26].

Results of the isotopic measurements are shown in Table I. The ^{235}U concentrations were measured by thermionic mass spectrometry, with a relative precision of about 0.2%. The relative ^{235}U content for these ores is normal to within the precision of the technique. The ^{143}Nd/^{142}Nd ratio could indicate an increment of fission-produced ^{143}Nd compared to the nonfission product ^{142}Nd. The normalized values given in Table I suggest that there are systematic tendencies in the ^{143}Nd/^{142}Nd ratio for different ore bodies, but fission produced ^{143}Nd (in excess of ∿ 1% of the natural ^{143}Nd) is not indicated. The ^{150}Sm/^{149}Sm ratio is also a sensitive indicator of a natural reactor, but an increment in this ratio would result from the high neutron fluence associated with a chain reaction. The measured samarium ratios likewise give no indication of a natural reactor.

Ruthenium is perhaps the most sensitive element for detecting natural fission. It has a very low natural abundance of about 1 ng/g, yet it has a high fission yield, in excess of 15%. Recent measurements [27] have shown that for Precambrian uranium ores, the component of fission-product ruthenium resulting from neutron-induced ^{235}U fission can be distinguished from the component resulting from ^{238}U spontaneous fission. Ruthenium isotope ratios for 26 Precambrian uranium specimens were thus used to determine if excessive ^{235}U fission had occurred [28]. The samples were from possible natural reactor sites at Shinkolobwe and Kasolo, Zaire; Rio Grande do Norte,

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[1] In theory, natural reactor uranium could exhibit a slight increment in ^{235}U if the conversion factor exceeded unity. However, the physical requirements for such a "breeder" reactor are very stringent, and they are not likely to have been met compared to the requirements for an Oklo-type reactor.

TABLE I

ANALYSIS OF SELECTED URANIUM ORES

Sample Number	Location	Isotopic Analysis U (wt%)	^{235}U (rel wt%)	$^{143}Nd/^{142}Nd$[a]	$^{150}Sm/^{149}Sm$[b]
8.01	Jabiluka, Australia	63	0.711	1.012	
8.02	Jabiluka, Australia	56	0.710	1.012	
8.03	Jabiluka, Australia	57	0.711	1.011	
8.04	Jabiluka, Australia		0.710	1.017	
8.10	Koongarra, Australia		0.710	1.017	
8.14	Nabarlek, Australia	59	0.712	1.020	
8.15	El Sherana, Australia		0.711		
9.05	Key Lake, Canada		0.713	1.001	
9.13	Key Lake, Canada			1.022	
9.16	Key Lake, Canada	29	0.711	1.014	
9.20	Key Lake, Canada			1.016	
9.23	Key Lake, Canada			1.014	
9.27	Key Lake, Canada			1.017	
9.68	Fay Mine, Canada		0.711	1.012	
9.71	Fay Mine, Canada	12		1.007	
9.77	Port Radium, Canada	41		1.010	
9.80	Port Radium, Canada			1.019	
10.08	Rabbit Lake, Canada	7		1.003	1.002
10.11	Rabbit Lake, Canada	57		1.002	1.000
10.13	Rabbit Lake, Canada	53		1.004	1.000
10.14	Rabbit Lake, Canada	19		1.003	1.000
10.29	Rabbit Lake, Canada	32		1.003	1.000
10.30	Rabbit Lake, Canada	14		1.005	1.001
10.31	Rabbit Lake, Canada	27	0.712	1.006	0.999
10.32	Rabbit Lake, Canada			1.003	1.001
10.33	Cluff Lake, Canada	57		1.013	1.001

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[a] The ratio $^{143}Nd/^{142}Nd$ measured for a sample has been normalized by dividing by the ratio measured for standard material from the National Bureau of Standards.

[b] The ratio $^{150}Sm/^{149}Sm$ measured for a sample has been normalized by dividing by the ratio measured for standard material from the National Bureau of Standards.

Brazil; Gordonia, Southwest Africa; Theano Point, Wilberforce, Rabbit Lake, Key Lake, Cluff Lake, Fay Mine, and Port Radium, Canada; and Jabiluka and Nabarlek, Australia.

For these high-grade samples, the amount of ruthenium resulting from neutron-induced ^{235}U fission was less than 75% of the ruthenium from ^{238}U spontaneous fission. (The total fission-product ruthenium ranged from 1.4 to 23 ng/g.) Precambrian uranium ores are known to have amounts of ^{235}U fission that range from about 3 to 47% of the ^{238}U spontaneous fission [29]; whereas at the Oklo reactor zones, the amount of ^{235}U fission was many orders of magnitude greater than ^{238}U spontaneous fission. Thus, the ore samples do not represent natural reactors.

There is a possibility that other natural fission reactors may have been redistributed or dispersed by geochemical or erosional processes. If this is so, there may be evidence for them in subtle ^{235}U isotopic depletions in present-day uranium ore bodies. Ore samples from various locations have been tested for slight variations in the ^{235}U/^{238}U ratio using precise gas (UF_6) mass spectrometry. These data are presented in Table II. The relative precision of this technique is ∿ 0.01%, which is lower than the natural variability in the ^{235}U/^{238}U ratio [30]. The analyses presented here are an extension of a previous study [30] that identified a bimodal distribution of ^{235}U concentrations in natural ores: magmatic and vein-type ores had values around 0.7108 rel wt%, whereas the values of sandstone-type ores of the Colorado Plateau clustered around 0.7106 rel wt%. It was speculated that the lowered ^{235}U content in Colorado Plateau uranium could have resulted from isotopic fractionation brought about by the repeated oxidation-reduction depositional process of sandstone-type ores, or from ^{235}U-depleted uranium that had been geochemically redistributed from a Precambrian natural reactor.

Certain samples of Table II (3.13, 3.44, 8.04, 10.14, and possibly 9.16) have anomalously high ^{235}U values, which lie well outside the accepted range of ^{235}U content. Duplicate analyses of samples 8.04, 9.16, and 10.14 were conducted by thermionic mass spectrometry, and the anomalous values were not confirmed. It is believed that the high values resulted from some systematic measurement error or sample contamination, and the samples are being reanalyzed.

The data of Table II indicate that these samples are not from natural reactors. However, if a natural reactor had existed and had been redistributed within the ore body, a subtle ^{235}U depletion would result. This generalized depletion would be detectable only if the average ^{235}U depletion (designated as B, in %) in the reactor were sufficiently high and if the subsequent dilution were not too great. The 3σ detection threshold for detecting anomalously low ^{235}U content by gas mass spectrometry is about 0.7100 rel wt% (normal uranium has about 0.7108 rel wt% [30]); so a redistributed reactor system could possibly be detected if the product $Bf \gtrsim 0.1\%$, where f is the fraction of uranium in the deposit that is from the reactor zone. This qualitative limit assumes the reactor system is homogeneously redistributed within the deposit. In reality, a reactor could have experienced partial redistribution or major mobilization away from the deposit.

The lowest value of Table II, 0.7107 ± 0.0001 rel wt%, is virtually indistinguishable from the expected value of ∿ 0.7108 ± 0.0002 rel wt%.

TABLE II

PRECISE URANIUM ANALYSIS OF WORLDWIDE ORES

Sample Number	Location	U (wt%)	^{235}U (rel wt%)[a]	^{235}U (rel wt%)[b]
3.1	Alligator Gorge, Australia	66	0.7110	
3.13	Rio Grande do Norte, Brazil		0.7139	
3.42	S. Alligator Gorge, Australia	70	0.7110	
3.61	Wilberforce, Canada		0.7108	
3.14	Shinkolobwe, Zaire	74	0.7111	
3.44	Shinkolobwe, Zaire		0.7134	
3.46	Katanga, Zaire		0.7110	
3.55	Kasolo, Zaire	69	0.7107	
6.1	Weddington Pit, Texas		>0.711 [c]	
8.01	Jabiluka, Australia	63	0.7112	0.711
8.04	Jabiluka, Australia		0.7128	0.710
8.14	Nabarlek, Australia	59	0.7112	0.712
9.16	Key Lake, Canada	34	0.7115	0.711
10.14	Rabbit Lake, Canada	22	0.7144	0.711
11.02	Witwatersand, SA		[c]	
12.01	Elliot Lake, Canada		0.7110	

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[a] Gas mass spectrometric analysis, 2σ precision ∿ 0.0001 rel wt%.

[b] Thermionic mass spectrometric analysis, 2σ precision ∿ 0.002 rel wt%.

[c] Insufficient uranium in sample for gas mass spectrometry analysis.

Hence, if a natural reactor had been redistributed within this deposit, we can conclude that Bf $< 10^{-2}$%. For example, a redistributed reactor system with a 10% average ^{235}U depletion must have comprised less than ~ 0.1% of the total uranium of the deposit. Deposits typically contain between 10^4 and 10^5 tons of uranium, and 0.1% represents a relatively small natural reactor system. Thus uranium gas mass spectrometry would appear to be effective in detecting a redistributed reactor, but it would be inadequate if there had been a low fission density, a high conversion factor (^{238}U to ^{235}U via ^{239}Pu), or a major loss of the reactor uranium.

MONITORING OF URANIUM MILLS

Isotopically analyzing rich ore specimens from drill cores or from exposures in mines is an obvious way to search for other natural reactors. But such specimens represent only a small fraction of an ore body, and a fossilized reactor zone could easily be missed and subsequently exhumed. While this sampling should be continued, a more systematic and thorough search capability could be obtained by monitoring the output of mills servicing likely Precambrian deposits. Such a plan [31] has been proposed where "yellowcake" (U_3O_8) samples from appropriate mills would be routinely collected and analyzed for subtle decreases in the $^{235}U/^{238}U$ ratio.

In this plan, the effectiveness of mill monitoring is determined for situations where reactor uranium is introduced into the mill episodically or continuously, and both continuous and "grab" sampling of yellowcake are considered. The uranium mill is assumed to act as a large dilution vessel with a comparatively small throughput of uranium (i.e., uranium is refined by continuous rather than batch processing, and mills generally contain several days of uranium output at any one time). Thus, after introduction of reactor ore into the mill, the concentration of anomalous uranium will decrease approximately exponentially[2] with some characteristic "half-time"--the time required for the anomaly concentration to be reduced by one-half. For a given amount of refined reactor ore, the sensitivity for detection is limited by the sensitivity of the isotopic analysis, the extent of ^{235}U depletion in the reactor ore, the frequency of grab or continuous sampling, and the dilution half-time of the mill.

For a natural reactor system the size of Oklo, many months would no doubt be required for processing of the affected ore. A rich ore zone such as this would be deliberately mined, stockpiled, and blended over a long time (compared to the mill half-time) in order to keep the average grade of

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[2] In reality, a mill has many stages of dilution, but usually only one or two locations contain the majority of the uranium. Where there are successive dilutions, the anomaly concentration can be described by rate equations analogous to those for successive radioactive decay. However, the assumption of a single dilution stage is adequate because it yields a detection limit that is conservative for a grab-sampling scenario. And when continuous sampling is employed or when the reactor ore is introduced continuously into the mill, the assumption of single or successive dilution has little effect on the detection limit.

the mill feed at an optimum concentration. Thus, reactor ore would eventually be introduced into the mill approximately continuously, but the isotopic anomaly in the yellowcake could fluctuate considerably. Nevertheless, one would expect that initially there would be erratic "pulses" of anomalous ore being processed. Detection of these events could be crucial to the early detection and preservation of a reactor site. To detect single episodes of anomalous ore throughput, mills with short half-times would require continuous sampling, while those with longer half-times could safely use grab sampling without sacrificing sensitivity. The sample frequency, while important to reactor ore detection, would probably be determined by convenience of collection and shipment and by sample analysis limitations.

For most mill monitoring scenarios, the detection of the first amounts of processed reactor ore would require that $C_oB \gtrsim 10$ to 100 $(\%)^2$, where C_o is the initial percent of total mill uranium that is from the reactor, and B is again the percent ^{235}U depletion in the reactor uranium. For reactor ore like that of Oklo with $\sim$ 30% depletion, an amount of reactor uranium between $\sim$ 0.3% and 3% of the total mill uranium would have to be introduced in order to insure early detection; i.e., between about ten and a few hundred kilograms of reactor uranium, depending on the mill capacity. Once anomalous ore was being continuously processed, however, the requirement for detection would be $BC_{SS} \gtrsim 8$ $(\%)^2$, where C_{SS} is the steady-state percent of total mill-feed uranium that is from the natural reactor. For reactor uranium with $\sim$ 30% depletion and a typical mill operation, only $\sim$ 10 kg/d of reactor uranium would have to be processed to insure detection. It is apparent that the first throughput of reactor ore may not be easily detected, but as the anomalous uranium concentration in the mill increases as a result of continuous input, the anomaly would almost certainly be detected.

DISCUSSION

This investigation has found no evidence for other natural fission reactors. Perhaps this is because the samples analyzed were not from the right locations in the uranium deposits. For most of the Precambrian vein-type deposits studied, there are many potential reactor zones. The samples, however, represent only a fraction of such zones: only those exposed during mining or intersected by drilling. The negative findings do not rule out the possibility that critical assemblages will be found (or have been exhumed, for that matter) in these deposits. As additional potential reactor zones are encountered at Precambrian deposits, samples should be collected and analyzed for isotopic anomalies.

The data do indicate, though, that the specific zones samples were not fossilized reactors--at least not of the type found at Oklo. It is possible that some of the zones may have experienced considerable neutron multiplication, but at least for those samples that received ruthenium or uranium gas mass spectrometric analyses, this was not indicated.

Natural criticality appears to have been prevented in the ore zones because the combined criteria of ^{235}U content, moderator content, and low neutron poison content were not met. In most cases, the samples came from regions where the size requirement was satisfied; drill-core samples, for example, often came from high-grade zones between $\sim$ 0.5 and $\sim$ 2 m thick. Neutron poisons were probably a major deterrent to criticality for the Key Lake and Cluff Lake deposits, and they may have precluded criticality at other locations as well.[3] The ^{235}U content of the ore zones during the Proterozoic was apparently insufficient to overcome the deterrents of excess poisons and insufficient moderation, even though the total uranium concentration ranged from $\sim$ 10 to $\sim$ 60%. Many of the Canadian samples are from post-Athabascan deposits about 1100 m.a. old, and some of the Australian samples may be of a comparable age. At this time, the ^{235}U content was 1.8 rel wt%--nearly twice as depleted as the uranium during the Oklo event.

For older samples, e.g., those from the Beaverlodge and Range deposits, criticality may have been presented, in part, because of the metallogenic process involved. At Oklo, the uranium mineralization and nuclear reaction were interrelated. Organic material, which had accumulated in tectonic structural traps, provided a reducing zone where uranium precipitated. The hydrocarbons, believed to have been present at the outset of criticality, provided additional neutron moderation that may have been an essential factor in initiating the nuclear reaction. Once the reactor was operating, a period of "autometamorphism" followed where the hydrocarbons were removed and the gangue and reactor zones altered to their present state. The hydrocarbons would not have been so crucial later in the period of criticality since the neutron fluence reduced the initial concentration of neutron poisons. Thus, the environment created by the organic material was very favorable to both uranium mineralization and nuclear criticality. Metallogenic processes operating at other Precambrian, vein-type uranium deposits may not have had this added benefit of hydrocarbon moderation to initiate a chain reaction.

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[3] There is no data available on amounts of neutron poisons in these samples, but it would be difficult to establish whether such impurities were present in the Proterozoic configuration. Elemental analyses of the samples were therefore not conducted.

REFERENCES

[1] BRYANT, E. A., COWAN, G. A., DANIELS, W. R., MAECK, W. J., "Oklo, an experiment in long-term geologic storage", Actinides in the Environment (FRIEDMAN, A. M., Ed.) Amer. Chem. Soc. Symp. Series, No. 35, New York (1976) 89.

[2] APT, K. E., BRYANT, E. A., "Criteria for identifying fossil nuclear reactors and their application to Saskatchewan", Uranium in Saskatchewan (DUNN, C. E., Ed.) Saskatchewan Geol. Soc. Special Pub. No. 3, Regina (1976) 193.

[3] ROBINSON, B. W., OHMOTO, H., Mineralogy, fluid inclusions, and stable isotopes of the Echo Bay U-Ni-Ag-Cu deposits, N.W.T., Canada, Econ. Geology 68 (1973) 635.

[4] Geochronology of North America, NAS-NRC Publication 1276 (1965) 149.

[5] LANG, A. H., GRIFFITH, J. W., STEACY, H. R., Canadian deposits of uranium and thorium, Can Geol. Survey Econ. Geol. Series No. 16, 2nd ed. (1962).

[6] TREMBLAY, L. P., Geology of the Beaverlodge mining area, Saskatchewan (revised), Can. Geol. Survey Mem. 367 (1972).

[7] KOEPPEL, V., Age and history of the uranium mineralization of the Beaverlodge area, Saskatchewan, Can. Geol. Survey Paper 67-31 (1968).

[8] SASSANO, G. P., FRITZ, P., MORTON, R. D., Paragenesis and isotopic composition of some gangue minerals from the uranium deposits of Eldorado, Saskatchewan, Can. J. Earth Sci. 9 (1972) 141.

[9] RICH, R. A., HOLLAND, H. D., PETERSON, U., Hydrothermal Uranium Deposits, Elsevier Scientific Publishing Co., New York (1977) 77.

[10] SIBBALD, T. I. I., MUNDAY, R. J. C., LEWRY, J. F., "The geological setting of uranium mineralization in northern Saskatchewan", Uranium in Saskatchewan (DUNN, C. E., Ed.) Saskatchewan Geol. Soc. Special Pub. No. 3, Regina (1976) 51.

[11] KNIPPING, H. D., "The concepts of supergene versus hypogene emplacement of uranium at Rabbit Lake, Saskatchewan, Canada", Formation of Uranium Ore Deposits, IAEA-SM-183, Athens (1974) 531.

[12] TAN, B., "Geochemical case history in the Key Lake area", Uranium in Saskatchewan (DUNN, C. E., Ed.) Saskatchewan Geol. Soc. Special Pub. No. 3, Regina (1976) 323.

[13] HERRING, B. G., The metamorphism and alteration of the basement rocks in the Carswell circular structure, Saskatchewan, M. Sc. thesis (unpublished) Univ. British Columbia (1976).

(14) PAGEL, M., La diagenèse des grés et les gisements d'uranium du bassin Athabasca (Canada), Centre de Recherches Pétrographiques et Géochimiques, Rapport Annuel (1976) 50.

[15] ROBERTSON, D. S., LATTANZI, C. R., Uranium deposits of Canada, Geoscience Canada 1 (1974) 8.

[16] MORTON, R. D., "The western and northern Australian uranium deposits--exploration guides or exploration deterrents for Saskatchewan?", Uranium in Saskatchewan (DUNN, C. E., Ed.) Saskatchewan Geol. Soc. Special Pub. No. 3., Regina (1976) 211.

[17] ANTHONY, P. J., "Nabarlek uranium deposit", Economic Geology of Australia and Papau New Guinea, Vol. 1, Metals (KNIGHT, C. L., Ed.) Australian Inst. Min. Metall. Monograph No. 5 (1975) 304.

[18] HILLS, J. H., RICHARDS, J. R., The age of uranium mineralization in Northern Territory, Search 3 (1972) 382.

[19] COOPER, J. A., On the age of uranium mineralization at Nabarlek, N.T., Australia, J. Geol. Soc. Australia 19 (1973) 483.

[20] FOY, M. F., PEDERSON, C. P., "Koongarra uranium deposit", Economic Geology of Australia and Papau New Guinea, Vol. 1, Metals (KNIGHT, C. L., Ed.) Australian Inst. Min. Metall. Monograph No. 5 (1975) 317.

[21] ROWNTREE, J. C., MOSHER, D. V., "Jabiluka uranium deposits", ibid., 321.

[22] EUPENE, G. S., FEE, P. H., COLVILLE, R. G., "Ranger ore uranium deposits", ibid., 308.

[23] SMART, P. G., WILKES, P. G., NEEDHAM, R. S., WATCHMAN, A. L., "Geology of geophysics of the Alligator Rivers region", ibid., 285.

[24] DODSON, R. G., NEEDHAM, R. S., WILKES, P. G., PAGE, R. W., SMART, P. G., WATCHMAN, A. L., "Uranium mineralization in the Rum Jungle-Alligator Rivers province, Northern Territory, Australia", Formation of Uranium Ore Deposits, IAEA-SM-183, Athens (1974) 55.

[25] APT, K. E., Ed., Investigations of the Oklo Natural Fission Reactor Program: July 1975 through June 1976, Los Alamos Scientific Laboratory Rep. LA-6575-PR (1976).

[26] BALAGNA, J. P., COWAN, G. A., "A non-destructive field measurement of the ratio $^{235}U/^{238}U$ in depleted to moderately enriched uranium", IAEA-TC-119/34, these Proceedings.

[27] MAECK, W. J., DELMORE, J. E., EGGLESTON, R. L., SPRAKTES, F. W., "The measurement of ruthenium in uranium ores and ^{238}U spontaneous fission yields, "IAEA-TC-119/35, ibid.

[28] MAECK, W. J., APT, K. E., COWAN, G. A., "A possible uranium-ruthenium method for measurement of ore age", IAEA-TC-119/32, ibid.

[29] ATTREP, JR., M., TASA, K. S., SHERWOOD, J. D., Estimations of the ratio of induced fission to spontaneous fission in uranium ores, Abs. 167th Natl. Mtg. Amer. Chem. Soc., Los Angeles (1974); and Texas Journal of Science, in press

[30] COWAN, G. A., ADLER, H. H., The variability of the natural abundance of ^{235}U, Geochim. Cosmochim. Acta 40 (1976) 1487.

[31] APT, K. E., Uranium Mill Monitoring for Natural Fission Reactors, Los Alamos Scientific Laboratory Rep., to be issued.