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SEARCH FOR AN "OKLO PHENOMENON" IN THE NORTHEASTERN REGIONS OF BRAZIL. A TENTATIVE CONTRIBUTION TO THE CONTINENTAL-DRIFT THEORY

F. W. LIMA,* MARINA B. A. VASCONCELLOS,* MARIA JOSÉ A. ARMELIN,*
R. FULFARO,* V. J. FULFARO,** B. B. BRITO NEVES***

**Instituto de Pesquisas Energéticas e Nucleares (IPEN), C.P. 11049 (Pinheiros), São Paulo, SP, 01000 (Brazil)*
***Instituto de Geociências, University of São Paulo (Brazil)*
****Departamento de Geologia, Federal University of Pernambuco (Brazil)*

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Rock samples from the Northeastern region of Brazil were analysed for their ^{235}U isotopic abundance, in search for the occurrence of an "Oklo Phenomenon" in this part of the Brazilian territory. The samples were collected in locations that could have been connected to the African continent, according to the continental-drift theory, in accordance to the Francevillian formations in the Gabon Republic, in which place the Oklo natural fossil reactor is situated. Two methods were used for the determination of the ^{235}U abundance: activation analysis followed by high resolution gamma-ray spectrometry and activation analysis by delayed neutron counting. No evidence of ^{235}U depletion was found in the rock samples analysed.

Introduction

The spectacular discovery, in 1972 [1], of the occurrence of a natural nuclear fission reactor , in which a spontaneous fission chain reaction took place, about two billion years ago, in an open-pit uranium mine at Oklo, in the southeastern part of the Gabon Republic, near the equator on the coast of West Africa, opened the possibility of scientific research in many fields of study.

The phenomenon was recognized by means of significant anomalies found in the isotopic composition of uranium [2] and by the presence of stable products of the nuclear fission of uranium in the depleted vein of the ore.

Apt et al.[3] carried out a search for other natural fission reactors in ore samples from Zaire(South West Africa), Rio Grande do Norte(Brazil), Northern Territory(Australia) and some ore bodies in Canada, but no evidence was found of the occurrence of a natural fission chain reaction.

In respect to similar researches in Brazilian territory, another aspect of the question must be taken into consideration. In the last few years, evidence has accumulated to support Alfred Wegener's original continental-drift theory[4] indicating that many of the actual continents were united for a long period in the Proterozoic and that a supercontinent must have existed during this period of time[5-7]. If correct, this conclusion has a great importance regarding the History of the Earth as a whole.

Paleomagnetic data suggest that, during the Proterozoic, Africa and South America were part of a single continent and that North America also belonged to this continent up to about 1000 million years ago[8].

J.D.A.Piper[5-7] found,initially, that the models of migration of the magnetic poles of Africa and North America could be strictly superimposed through all their extension since 1000 to 2000 million years and possibly 2700 million years ago. When this occurred, the afro-arabic and north-american regions formed a single continent and the continent of Gondwana occupied its position in the Pangeae during the Proterozoic at least as close as 750 million years ago, according to McElhinny and Embleton[9]. The combination of these results is the configuration shown in Figures 1 and 2 : a stable supercontinent, since at least 2200 million years up to the minimum close period of 1000 million years before the present one.

The postulated supercontinent suffered a process of rifting and drifting in the superior Proterozoic at about 1200 to 1000 million years up to 850 million years ago.

It is difficult to assign which litho-structural units of the Precambrian region of the Brazilian northeastern territory could correspond to the Oklo contingent. In the first place, an incontestable pre-drift reconstitution map is not available. Secondly, an integrated map of Northwestern Africa,in the 1/100 000 or 1/500 000 scale, for instance, with an intercontinental validity in Africa, and which can serve as a support to a safe geotechnical and lithostructural comparison between the two continents is not available.

Fig. 1. Distribution of the diamictites of the Upper Pre-Cambrian plotted on the (Upper Proterozoic) Supercontinent of Piper[5-8]

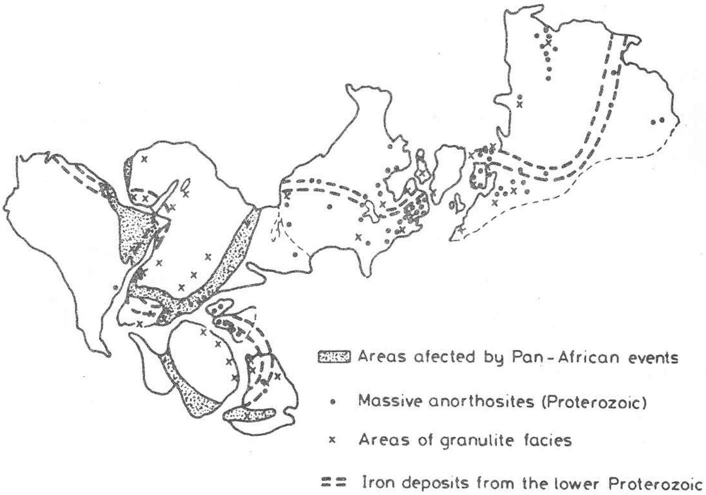

Fig. 2. Proterozoic Supercontinent of Piper,[5-8] showing the distribution of massive anorthosites, Pan-African Belt Areas of Pre-Cambrian Granulite Facies and iron layered deposites from the Lower Proterozoic

Another difficulty arises from the position of the Francevillian system,away from the coast line, where the comparisons are less complex and where there are certain reference marks.

Among the regions of the Brazilian territory that stand a chance of having been connected to the African continent in correspondence to the so-called Francevillian formations in the Gabon Republic, in which the Oklo natural fossil reactor is situated, are the ones of the Precambrian rocks of the Estancia Area(definition according to Silva Filho et al.[10] in the state of Sergipe and the ones of the Precambrian rocks of the Garanhuns and Venturosa Area, situated in the state of Pernambuco[11]. These possibilities, drawn from a very broad view of the regional gology, carry a considerable amount of inferences and hypothesis which should not be discussed in this text.

If the presence could be detected on Brazilian coasts of vestiges of a natural fission reactor, in the regions corresponding to Gabon, this would also come in support of the continental-drift theory, since the Oklo reactor dates to the Precambrian Era, which is anterior to the occurrence of this phenomenon.

In fact, S.Eklund[12], Director General of the IAEA, has already pointed out, on making comments about the Oklo phenomenon, that it would be interesting to see whether a corresponding discovery would be made in future in some of Brazil's uranium mines.

Samples were obtained from the mentioned regions of the Brazilian territory. In Figure 3, the locations of collection of the samples are shown. In the Gneissic-Migmatitic complex region, samples 1,2,3 and 6 (chalco-silicatic rocks) were collected, in the Venturosa Area. From the Garanhuns Area, samples 4,5 and 7 belonging to the "Quartzitic Unit of Garanhuns"[13] were taken. In the Estancia Area, six other groups of samples were collected, as shown in the map.

The concentration of uranium in these rocks varied from a few parts per million to about 2%.

Determinations were made of the ^{235}U isotopic abundance of some of these samples, with the aim of detecting possible anomalies that could indicate some correlation with the Oklo natural fossil reactor.

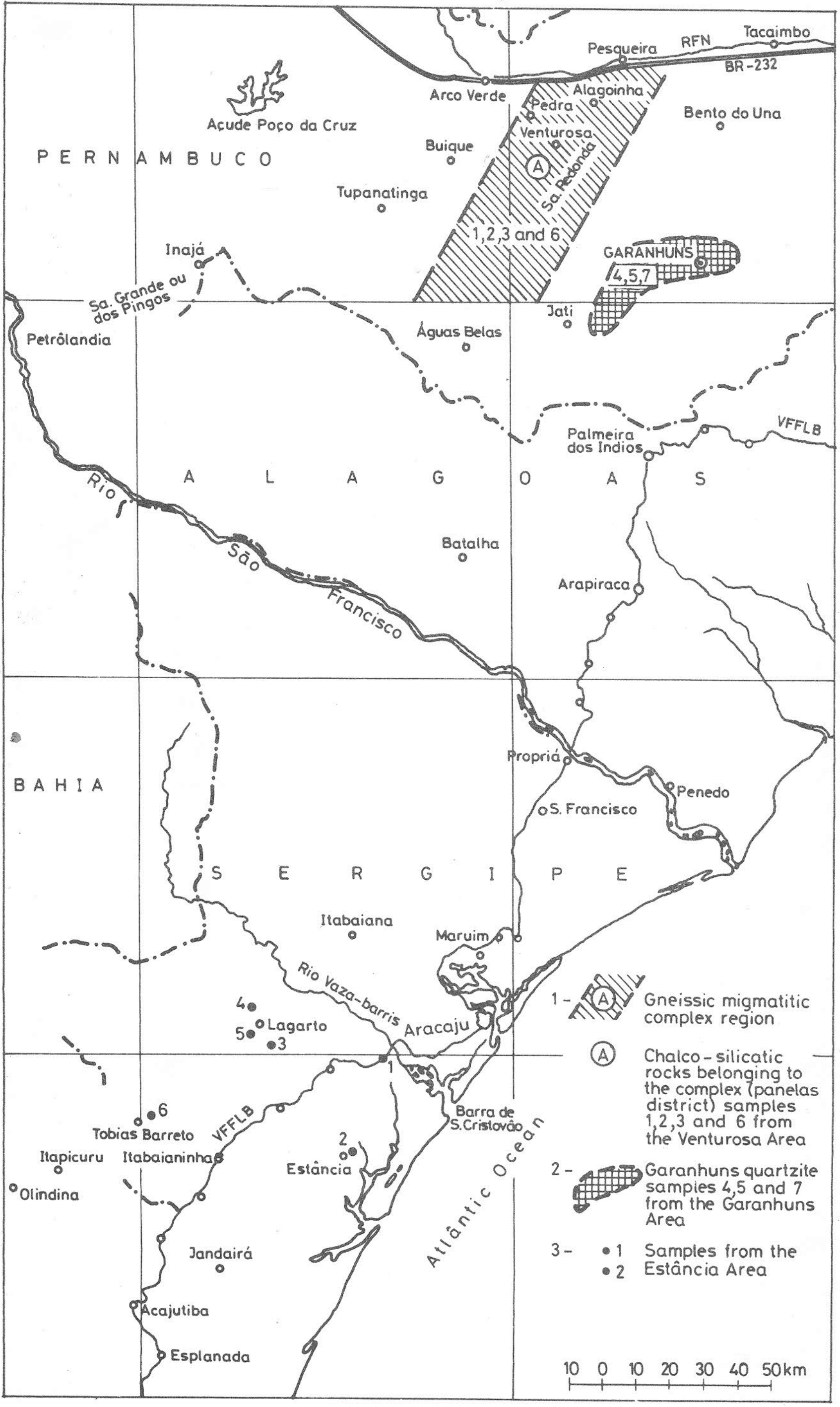

Fig. 3. Sites of collection of samples

Two methods were employed for the determination of the percentage of ^{235}U: activation analysis utilizing high resolution gamma-ray spectrometry and activation analysis by delayed neutron counting. The results obtained by these techniques, although less accurate than those of mass spectrometry, could be adequate to the purpose of the present work, in the case of ocurrence of considerable anomalies in the isotopic composition of the uranium found in these samples. Some samples of the Oklo reactor presented isotopic compositions down to 0.29% in ^{235}U[2].

Experimental

Sample preparation

The rock samples were broken in fragments of about 0.5 cm, first by using a hammer and then in a porcelain mortar. The final grinding was carried out in a SPEX mixer-mill with steel balls, in which the material was reduced to a fine powder, of granulometry lower than 150 mesh.

Determination of the isotopic percentage of ^{235}U by delayed neutron counting activation analysis

Neutron activation analysis followed by delayed neutron counting has been applied by Amiel[14] for analysis of the isotopic composition of uranium in NBS isotopic standards.

Performing irradiations with and without a cadmium shielding it is possible to determine the ^{235}U isotopic abundance, taking into consideration the fact that ^{235}U is fissioned mainly by thermal neutrons and ^{238}U by fast neutrons. For the calculations, the amount of ^{234}U was considered as negligible.

Irradiation and counting

The samples and standards (IAEA reference uranium ores[15], of natural uranium isotopic composition) were sent to the irradiation position by means of a pneumatic system and the countings were performed in a neutron counting facility consisting of six BF_3 neutron detectors embedded in a paraffin cylinder and coupled to an electronic counting system. The detection and counting system has been described with more details elsewhere[16].

The irradiation time was of 1 minute, the delay time between the end of irradiation and the beginning of counting was of 20 seconds and the counting time of 1 minute. The thermal neutron flux in the irradiation position was of 4.3×10^{11} neutrons/cm^2.s.

Determination of the isotopic abundance of ^{235}U by high resolution gamma-ray spectrometry

Thermal neutron activation analysis followed by high resolution gamma-ray spectrometry allows the determination of isotopic ratios of some elements, provided that two or more of the isotopes of these elements give origin to well identifiable species.

In the case of uranium activation, ^{239}Np, originated from ^{238}U and several of the fission products of ^{235}U present many easily identifiable peaks in the gamma-ray spectrum obtained with a Ge(Li) solid state detector.

De Wet and Turkstra[17] and Mantel et al.[18] have already applied this method to the determination of $^{235}U/^{238}U$ isotopic ratios, in NBS isotopic standards.

The ratios between the areas of two photopeaks corresponding to one of the fission products of ^{235}U and the other corresponding to ^{239}Np will be proportional to the $^{235}U/^{238}U$ atom ratio in the sample.

In order to improve the precision of the method, several ratios between peaks of the fission products and of ^{239}Np can be used instead of only one, and the averages of the various ^{235}U abundances obtained through this procedure can be computed.

For the calculation of the peak ratios used in the present work, the areas of the 106, 118, 210, 228 and 278 keV peaks of ^{239}Np were chosen. As to the fission products, the following peaks were employed: 140 keV ($^{99}Mo-^{99m}Tc$), 668 and 773 keV($^{132}Te-^{132}I$), 293 keV(^{143}Ce), 530 keV(^{133}I) , 743 keV($^{97}Zr-^{97}Nb$). These peaks were considered to be the most convenient, since the corresponding activities were high enough for the purposes.

Preparation of samples and standards for irradiation

Since the determination of the ^{235}U isotopic percentage was carried out in rocks, which generally present a very complex composition, it was decided to perform a simple chemical separation of uranium, prior to irradiation , to avoid too many interferences in the gamma spectra. Uranium was extracted with TBP and then back-extracted into an aqueous phase, of 5% $(NH_4)_2CO_3$ solution, which after evaporation was pipetted on filter paper for irradiation.

Irradiation and counting

Each group of two samples and one standard processed was irradiated under a thermal neutron flux of 5×10^{12} neutron/cm^2.s, for an 8 - hour period.

After a delay time which varied between 2 and 5 days after the end of irradiation, the countings were carried out, using a solid state ORTEC Ge(Li) detector, with a resolution of 2.1 keV for the 1332 keV peak of ^{60}Co. The detector was coupled to a 4096-channel Hewlett-Packard analyser. Counting time varied between 20 and 50 minutes for each sample.

The assignment of the energies of the several photopeaks in the gamma spectrum and the calculation of the corresponding areas was performed by a Hewlett-Packard minicomputer, using a program in Basic-Language,specially developed for the analysis of gamma-ray spectra.

Results and discussion

The results of the determinations of the isotopic abundance of ^{235}U by delayed neutron counting activation analysis are presented in Table 1.

In Table 2,the results obtained for the determination of the ^{235}U isotopic percentage by the method of thermal neutron activation analysis and high resolution gamma-ray spectrometry, are presented.

Table 1
Determination of the ^{235}U isotopic abundance in rocks by delayed-neutron counting

ROCK SAMPLE	^{235}U (%)	STANDARD DEVIATION (s)	RELATIVE STANDARD DEVIATION (%)	RELATIVE ERROR (%)
Sample nº 1 (Estancia Area)	$0.705 \pm 0.051^{*}$	0.715^{**}	10.1^{***}	2.1
Sample nº 1 (Gneissic-Migmatitic Complex Region)	0.722 ± 0.027	0.0471	6.5	0.25
Sample nº 2 (Gneissic-Migmatitic Complex Region)	0.723 ± 0.027	0.0473	6.5	0.39

* $\bar{x} \pm \frac{ts}{\sqrt{n}}$ (95% confidence level)

** $\frac{s}{\bar{x}}$ $\bar{x}$ = found mean

*** $\frac{\bar{x} - \mu}{\mu}$ μ = true value = 0.7202 [1]

Obs.: The results presented for the % ^{235}U are means of at least ten determinations.

The results presented in the Tables show that the means obtained for the isotopic abundance of ^{235}U in the rocks analysed, by both methods employed, can be considered as equal to the value of 0.7202%, taken as "natural". This conclusion was drawn by applying a t-test, on a 95% confidence level.

Table 2

Determination of the ^{235}U isotopic abundance in rocks by high-resolution gamma-ray spectrometry

ROCK SAMPLE	^{235}U (%)	STANDARD DEVIATION (s)	RELATIVE STANDARD DEVIATION (%)	RELATIVE ERROR (%)
Sample nº 1 (Estancia Area)	0.714±0.016*	0.0155	2.2	0.90
Sample nº 2 (Estancia Area)	0.721	-	-	0.11
Sample nº 1 (Gneissic-Migmatitic Complex Region)	0.725±0.009*	0.00848	1.2	0.67
Sample nº 2 (Gneissic-Migmatitic Complex Region)	0.719±0.021*	0.0198	2.7	0.17
Sample nº 6 (Gneissic-Migmatitic Complex Region)	0.705	-	-	2.1
Quartzite of Garanhuns	0.710	-	-	1.4

* Means of six determinations.

The low relative errors attained show that the methods are adequate for the analysis of the isotopic abundance of ^{235}U, considering the purpose of the present work, although not attaining the precision and accuracy of mass spectrometry.

On the other hand, if we compare the methods of delayed neutron counting and gamma-ray spectrometry, it is obvious that the second presented a precision, of about 2%, better than the first method, whose reproducibility was between 6 and 10%. This fact can be attributed to the low delayed neutron countings obtained, due to the not very high neutron flux (4.3×10^{11} neutrons/cm^2.s) available in the irradiation position used. Amiel[14] obtained a reproducibility of 0.3% based on counting statistics only, but the level of neutron counting was much higher.

In the case of the method of high resolution gamma-ray spectrometry , counting statistics is also the limiting factor of the precision attainable. By using the same sample for comparing the activities corresponding to fission products of ^{235}U and the activity due to ^{239}Np, many of the other common causes of errors in activation analysis, such as flux variations , self-absorption, weighing errors, different counting geometry of sample and standard, etc are eliminated and high precision should be attainable. It was observed that the fission products peaks which presented the lowest counting levels and, consequently, poorest counting statistics, lead to the less accurate results.

The analysis of the samples chosen indicate that, in the regions where they have been taken, no fossil nuclear reactor existed. Additional collection and analysis of rocks should proceed in order to gain more knowledge about the possible areas for a fossil reactor.

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