The Oklo natural nuclear reactor

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1. INTRODUCTION

The discovery of the natural nuclear reactor in Gabon (West Africa) is, perhaps, one of the most striking events in reactor physics since Enrico Fermi and his collaborators succeeded in 1942 in producing a self-sustaining fission chain reaction by artificial means.

At present, the probability of a random formation of a nuclear reactor under natural conditions is negligible. The development of a chain reaction requires that the multiplication factor k, i. e., the ratio of the secondary neutrons to primary neutrons, should be greater than or equal to unity. For large reactors, $k = k_{\infty}P$, where P is the probability that the neutron will not succeed in escaping from the reaction in the course of slowing down and diffusion, and k_∞ is the multiplication factor in an infinite medium, i. e., it is independent of the size of the system. It follows from the condition $k_\infty \geq 1$ that at least $k_\infty > 1$, which is not easy to achieve even if we deliberately set out to do so.

The point is that natural uranium contains only 0. 72% of the fissile isotope ^{235}U. The rest is ^{238}U, which is capable of only radiative capture of the slowing down fission neutrons. On the other hand, at thermal neutron energies, the small concentration of ^{235}U is compensated by the high fission cross section (see Table I). If we calculate the number η of secondary neutrons produced as a result of the capture of one thermal neutron in natural uranium

$$\eta = \bar{\nu}\,\sigma_f/\sigma_a = (1-\xi)\sigma_{f5} + \xi(\sigma_{f5}/\sigma_{a5}), \qquad (1.1)$$

we find that $\eta = 1.34$, i. e., it is greater than unity ($\xi = \xi_0$ is the contemporary fraction of ^{235}U in the natural isotopic mixture and σ_{a5} is the uranium absorption cross section averaged over the isotopic composition). However, to reach thermal energies as a result of slowing down from fission energy ($E_f = 2$ MeV), the neutron must also avoid capture by the large resonances in the ^{238}U absorption cross section. The fraction of fission neutrons that is slowed down to thermal energies should not be small. This can be achieved by mixing natural uranium with a moderating material assisting the neutrons in avoiding the resonance capture region. However, by increasing the probability φ of avoiding resonance capture through the use of the moderator, one reduces the fraction θ of thermal neutrons captured directly by uranium, because of the absorption of thermal neutrons by the moderator itself (σ_a^*). There is an optimum relationship between the concentration N_{3l} of the moderator and the concentration N_v of the uranium for which k_∞ is a maximum. This is given by

$$k_\infty = \eta\varepsilon\varphi\theta, \qquad \theta = \sigma_{a5}N_v/(\sigma_{a5}N_v + \sigma_{a3}N_{3l}) < 1 \qquad (1.2)$$

where ε takes into account multiplication by fast fission neutrons in ^{238}U and, at best, tends to increase k_∞ by a few percent. To reduce resonance capture in ^{238}U, the uranium is made into blocks whose size and separation are optimized (see, for example, Weinberg and Wigner[12:1]). It is only weakly absorbing moderators such as D_2O, C, Be, and BeO that can be used to obtain $k_\infty > 1$. Concentrated heavy water is not found in nature, and a random formation of natural uranium with pure graphite or beryllium of the quality necessary for reactors in the necessary block geometry is hardly possible.

For the optimum concentration of natural uranium in ordinary water, the value of k_∞ is less than unity, although it is quite close to it. This is why ^{235}U-enriched uranium is used in power-producing reactors in which ordinary water is both the moderator and the coolant. According to (1), an increase in ξ from 0.72 to 3% results in an increase in η from 1.34 to 1.83. There is an accompanying increase in k_∞ because σ_a" increases by a factor of three, namely, from 7. 6 to 23 b. Since φ remains practically constant, the result is that k_∞ increases to the extent that it is possible to use zirconium, or even the moderately neutron absorbing stainless steel,[13:1] as the construction material in the interior of the pile.

In 1953, Wetherill and Inghram[1143] drew attention to the fact that, in the distant past, the concentration of ^{235}U in natural uranium was much higher because it has a shorter half-life than ^{238}U (Table I). Using the contemporary concentration of uranium in pitchblende, they concluded that about two billion years ago when the fraction of ^{235}U was 3%, the prevailing conditions may have

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TABLE I.

Element	$^{235}U_{92}$	« o .	$^{239}Pu_{94}$
*i. br	1.03· 10»	6.50-10»	3.5240*
σ_{γ}^{t}, b¹	98.6±1,5	2.70±0.02	268.8±3
«J. b,	582.2±1,3	—	742.5±3
I'_{r}, bi	144±6	275±5	200±20
I_{f}, b¹	275±5	—	301±10
v,	2.412±0.008	—	2.871±0.006

Notation: τ,→half-life; $a^{\wedge}$ and $Of-^{1}$ radiative capture and fission cross sections for neutrons of 0.0253 eV (velocity $v = 2200$ m/sec); ij and /^—resonance integrals for radiative capture and fission, evaluated over the epithermal-neutron spectrum: $/ o'(E)dE/E$; v_{i}—number of secondary neutrons per fission by thermal neutrons.

been close to the critical conditions. Three years later, Kuroda[c5] showed that, if in the distant past, deposits of this kind contained water, then even the condition fe. > 1 may have been satisfied, and a spontaneous nuclear reaction may have taken place. However, until quite recently, no traces of a natural reactor of this kind had been found.

2. HISTORY OF THE DISCOVERY OF THE NATURAL REACTOR

On June 7, 1972, in the course of routine mass-spectrometric analysis at a factory at Pierrelatte (France), which manufactures enriched fuel, Bouzigues *et al.*[c6> 7] found that the primary uranium hexafluoride gas contained 0. 717% ^{235}U atoms instead of the 0. 720% which was usual for terrestrial rocks, meteorites, and lunar specimens. The French Atomic Energy Commission (CEA) then initiated a study of the reasons for this anomaly.

The simplest hypothesis, namely, that the uranium was contaminated with "tailings" (the waste products of the enrichment cycle), was found to be incorrect. The anomaly was traced through the numerous stages of the manufacturing process right back to the ore-enriching plant at Mounana near Franceville in Gabon. The original ore with mean uranium concentration of 0.4-0.5% was mined at Oklo.

Isotopic analysis of specimens rich in uranium demonstrated a substantial depletion in the ^{235}U isotope and a deviation from the natural distribution of the rare-earth isotopes that are the products of the fission process.[c618:] This, in fact, served as proof of the existence of a spontaneous chain reaction in the distanct past. The proof itself took less than three months.

Retrospective analysis of documents and samples at the enriching plant at Mounana showed that, in 1970-1972, the plant received ore containing at times up to 20% uranium depleted down to 0. 64% in the ^{235}U isotope.[m] Since the ore was mixed in the course of mining operations, the concentration of uranium in individual natural samples may have been even higher and the depletion even greater. More than 700 tons of the depleted uranium, which participated in the chain reaction, was mined and the deficit (initially unnoticed) amounted to about 200 kg of ^{235}U.

As a result of an agreement with the Government of Gabon, the company concerned with the mining of uranium ores in Franceville (COMUF) agreed to halt their mining operations in the region of the natural reactor. A Franco-Gabon group headed by R. Naudet began a systematic study of the Oklo phenomenon. Numerous specimens obtained by drilling were sent for analysis to many laboratories across the globe, including those in the USSR.[C10J] The data thus obtained were reviewed together at an IAEA symposium held in Libreville (Gabon) in the summer of 1975. The result was a reconstruction of the operation of the reactor during the precambrian epoch.

According to data obtained by the U/Pb method, the Oklo deposits were formed $(1.81\pm0.05)\times10^{9}$ years ago.[c11] A sedimentary layer of sandstone, enriched with uranium, 4-10 m thick and 600-900 m wide, was formed in the delta of the ancient river.[c12:] The layer rested on a basalt bed which descended to a depth of several kilometers as a result of tectonic processes. This was accompanied by the splintering of the uranium vein and by its penetration by ground water, which gave rise to a situation favorable for the appearance of a natural reactor. The final uplift of the deposit to the contemporary level occurred some tens of millions of years ago.

An ore with a mean uranium concentration by weight of 0.5% contains clay lenses in which the uranium concentration is up to 20-40% or even more. A lens is a compact ore body with dimensions of 10 to 20 m and a thickness of the order of 1 m. The formation of such lenses with ultrahigh concentration of uranium is explained by the action of filtration water, although a detailed picture of this process is not altogether clear.[c13] The chain reaction occurred precisely in these lenses. Altogether 6 reaction foci (Figs. 1 and 2) were found over a distance of a few tens of meters.[c9] Two of them were partially mined, but the other four were in their

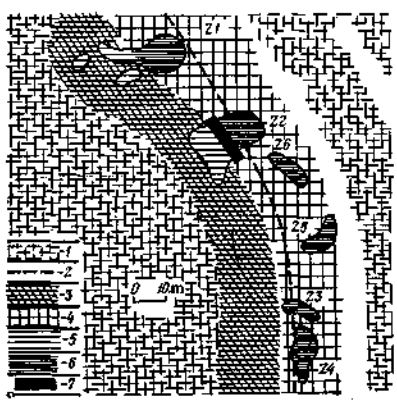

FIG. 1. Disposition of the active zones in the Oklo reactor: 1—sandstone; 2—boundary of mined ore; 3—sandstone wall; 4—floor of pit; 5—mined part of reactor; 6—explored part of reactor; 7—area to be preserved.

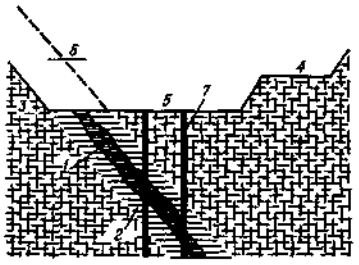

FIG. 2. Schematic section through the pit: 1—uranium-rich vein; 2—ore body; 3—sandstone wall; 4—profile of quarry; 5—floor of pit; 6—mined ore; 7—sample cores.[17]

natural state. It was suggested that part of the Z2 area should be retained as a natural memorial.[8, 13]

3. REACTOR PARAMETERS

A. Age

The age t_0 of the reactor can be determined if one knows the number $N\%_5(-t_0)$ of ^{235}U atoms that underwent fission and the number $N_{v8}(0)$ that have survived to this day.

The following balance equation must have held at the time of the chain reaction:

$$\frac{dN_{U5}}{dt} = -\left(\langle\sigma_a^{U5}\Phi\rangle + \frac{1}{\tau_5}\right) N_{U5} + \frac{N_{Pu9}}{\tau_9}, \qquad (3.1)$$

where $(\sigma_o^{\ddot{u}5}\Phi>$ is the product of the ^{235}U capture cross section and the neutron flux Φ, averaged over the neutron spectrum, and r_5 and τ_9 are the half-lives of ^{235}U and ^{239}Pu. The last term in (3.1) describes the creation of ^{235}U through a decay of ^{239}Pu, formed in the reaction ^{238}U(«, y)~^{239}U(2/r)-^{239}Pu.

Assuming that the life of the reactor, d, was much greater than r_9 and that the neutron flux changed very little during the time d, we may suppose that the plutonium concentration succeeded in reaching equilibrium:

$$N_{Pu5} = (\sigma_\gamma^{U8}\Phi)\, T_{9i} V_{U8}\, (1 + (\sigma^{\wedge\prime}\Phi)\,\tau,)^{\prime\prime\prime}, \qquad \tau_9 < d. \qquad (3.2)$$

It then follows from (3.1) that the relative concentration ζ of ^{235}U at the present time is

$$\zeta = \frac{N_{U5}(t)}{N_{U8}(t)} = \zeta_0 e^{-\langle\sigma_a^{U5}\psi\rangle}\,[1 + C\,(e^{\langle\sigma_a^{U5}\psi\rangle} - 1)],$$
$$C = \frac{\langle\sigma_\gamma^{U8}\Phi\rangle\, e^{-t_0/\tau_9}}{\langle\sigma_a^{U5}\Phi\rangle\,\zeta_0\,[1 + \langle\sigma_a^{Pu5}\Phi\rangle\,\tau_9]}, \qquad J_- = \frac{1}{\tau_5} - \frac{1}{\tau_8}, \qquad (3.3)$$

where $£_0$ is the contemporary concentration of ^{235}U in natural uranium and $\psi = /\!\!b\Phi(i)<i£$ is the fluence (integrated neutron flux). The fraction of consumed ^{235}U atoms is given by

$$\delta = \frac{N_{U5}^a(-t_0)}{N_{U5}(\cdot)} = e^{t_0/\tau_5} \int_0^{\psi} \zeta(\psi')\,\langle\sigma_a^{U5}\rangle\, d\psi'. \qquad (3.4)$$

If we measure the three quantities ζ, δ, and ψ at a given point in the reactor, we can use (3.3) and (3.4) to calculate two unknowns, namely, the age i_0 and the coefficient of conversion of ^{238}U into ^{235}U, which we have denoted by C. The above formulas are only illustrative in character because, in reality, the experimental data were analyzed by solving the equations on a computer.

The concentration ζ was determined with good accuracy by isotopic analysis of the burnt-up uranium. Several independent methods were used to determine δ. For example, δ can be found by measuring the relative concentration of ^{232}Th, formed as a result of the reaction ^{235}U(n, $_y$)_$23_{6u(Q!)}$_23_{2Th}-C14.15]

Another method of determining δ is to measure the relative concentration of one of the fission fragments. In the case of slow burning of an isotope i, the relative concentration is equal to $\gamma_i\delta\ (\sigma/^5>/(\sigma_\alpha^{\ddot{u}5})$, where γ_i is the fission yield. Neodymium is particularly convenient for this purpose since the natural distribution of its isotopes is very different from their yield in the fission of ^{235}U by thermal neutrons (Table n[c8b:]). In particular, the fission products do not contain ^{142}Nd, so that its amount can be used to determine the cencentration of natural neodymium in the ore, and to eliminate it. After introducing corrections for the isotopic distribution, there is excellent agreement with the fission yield (the last two columns in Table Π), and this serves as a convincing argument supporting the fission chain reaction at Oklo.[c6] In addition to Nd/U ratios, measurements were carried out of the Sm/U, Eu/U, Rb/U, etc. ratios.[c8,14-161] As a result of all these measurements, the fraction δ was obtained to an accuracy of between one and a few percent.

Equations (3.3) and (3.4) contain the fluence φ. This can be measured by using a pair of isotopes, one of which gives rise to strong neutron absorption and the other to weak neutron absorption. The accumulation of the stable isotope i is described by an equation such as (3.1):

$$\frac{dN_i}{dt} = -\langle\sigma_a^i\Phi\rangle N_i + \langle\sigma_a^{i-1}\Phi\rangle N_{i-1} + \gamma_i\langle\sigma_f^{U5}\Phi\rangle N_{U5}. \qquad (3.5)$$

TABLE II.

Isotopes of Nd	142	143	144
	18,7±0,7	325±10·)	3,6±0,3
4- b[1]	9±1	140±30	5,0±0,5
Natural concentration, %i	27,11	36,02	
Fission of "V, %	0	55,18	
95% of fissions of "' U + 5% of ^{111}U, %	0	54,78	
Oklo samples	0	54,95	

Isotopes of Nd	IIS	1i t	148	150
4	42±2	1,4±0,1	2,5±0,2	1,2±0,2
4, <b[1]	240±35	3,2±0,5	19±1	14±2
Natural concentration, %	25,52		5,73	5,62
Fission of "' U, %	33,53		8,16	3,13
95% of fissions of "' U + 5% of "• u, %	33,65		8,28	3,29
Oklo samples, %	33,46		8,25	3,34

*)The experimental value averaged over the neutron spectrum of a typical light-water reactor is $\langle o_a^{Nd}\rangle$ = 266 b.[16]

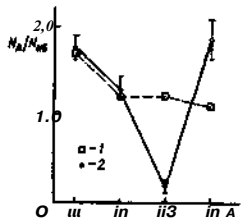

FIG. 3. Burn-up of ^{113}Cd in the Oklo reactor: 1—yield of Cd Isotope relative to ^{116}Cd during fission; 2—ditto for the Oklo sample.[1tr1]

When $(\sigma[\varphi) \ll 1$, the burning of the isotope i can be neglected. In the other opposite case, $(\sigma'_\alpha \varphi) \gg 1$, we can neglect the derivative in (3.5) and establish the equilibrium concentration which then follows the time variation of N_{v5}. It follows that, when $\{\sigma^*_\alpha \varphi) \ll 1 \ll (\sigma\pounds\varphi)$, we have

$$\frac{N_k}{N_i} = \frac{\gamma_{ik}\xi}{\gamma_{i\bar{\xi}}(\sigma_\alpha^{v5}\psi)}, \qquad (3.6)$$

where $\bar{\xi}$ is the concentration of ^{235}U, averaged over the time of the reaction. It turns out that, in the active zone, $(\sigma_\alpha^{v5}\psi > \sim 1$ (which corresponds to ψ-IO^{21} neutrons/cm^2) and the condition $(\sigma_\alpha^*\psi) \gg 1$ can be replaced by $\sigma_\alpha^* \gg \sigma^{v5}$. This condition is satisfied by the isotopes ^{113}Cd, ^{149}Sm, ^{151}Eu, ^{155}Gd, and ^{m}Gd, which are strong absorbers of thermal neutrons. Since the numerator in (3.6) is large, their yield is reduced (by an order of magnitude or more) as compared with the others. At the same time, the concentration of the next $(k+1)$ isotope will, according to (3.5), increase. This phenomenon has been observed experimentally[6,17a] (Fig. 3). This in itself immediately shows that a chain reaction initiated by thermal neutrons has taken place.

Isotopes with cross sections comparable with σ_α^{v5}, for example, ^{143}Nd[t16] (see Table Π) were used to determine φ. Analysis of the data obtained for a large number of samples yielded the following value for the age of the reactor: $i_0 = (1.84 \pm 0.07) \times 10^9$ years.[C15]

The uncertainty in the fraction of ^{235}U formed as a result of the ^{239}Pu decays [the term containing C in (3.3)] introduces a substantial uncertainty in the calculation of f_0. This fraction was about 50% for many of the samples and depended on the total time of operation of the reactor. In general, formulas such as (3.3) and (3.4) are valid provided: 1) the ratio of resonance neutrons to thermal neutrons does not vary with time; 2) there is no additional supply of uranium during the reaction and no loss of uranium after the reaction, and 3) fission fragments have not appreciably migrated over the last two billion years. The value of t_0 may also be affected by improved values of the temperature of different parts of the active zone. A group of scientists in the USA consider that the most probable value of f_0 lies between 1. 7 and 1.9 billion years.[c15>1G:1] The French group also quotes a value of 2 billion years.[14] This figure is reduced to 1.8 billion years if the analysis of the experimental data is based on the ^{143}Nd absorption cross section adopted by the American group, i. e., $(\sigma^{TM^3}) = 2661$ instead of the 325 b (see footnote to Table II).[a]

B. Duration of the chain reaction

The duration d of the operation of the reactor can be determined from the amount of ^{239}Pu produced. For $d \gg T_s$, the equilibrium concentration of plutonium is determined by the neutron flux Φ [see (3.2)]. If, for a given sample, we know both φ and Φ, the mean time of operation of the reactor is $d = \psi/\Phi$. The fission plutonium can be separated from uranium by using the relative yield of neodymium isotopes: 150/(143 +144). For ^{239}Pu, it amounts to 0.1175 and for ^{235}U it is 0.0566.[c16] Unfortunately, the fraction of ^{239}Pu fissions among the total number of fissions is only a few percent and is comparable with the fraction of ^{238}U fissions by fast neutrons, for which the above isotopic ratio is 0.1336.[c16118] The result of all this is that d is subject to much greater uncertainty than t_0. A least-squares fit, carried out for d together with t_0, yields for the simplest model (without supply or loss of uranium): $d = (0.23 \pm 0.07) \times 10^6$ years.[c16] However, the authors of this report themselves regard this figure as the lower limit because fast-neutron ^{238}U fission was not taken into account sufficiently accurately. When this is done, the result is $d \sim 0.6$ billion years[c18] or even 0.8 billion years.[c17b:1]

C. Neutron flux

Figure 4 shows one of the numerous distributions of the integrated neutron flux over the active zone obtained as a result of a composite analysis of the samples (mainly determinations of the ^{144}Nd/^{m}Nd ratio). The vertical bars show the range of values of φ, depending on the different assumptions about the time of contamination by natural neodymium (ranging from the beginning of the chain reaction to after its end).[c19] As expected, the spatial distribution of uranium depletion shown on this figure is related to the distribution of φ.

The dashed curve in Fig. 4 shows the distribution of the uranium concentration by weight in the ore.[c20] It varies rapidly over a distance of 1. 4 m. Consequently, the thickness of the active zone (corrected for the 45° inclination of the deposit) is roughly 1 m.

In the 400-401 region, there is a burst in the neutron flux. This is due to the fact that the uranium concentration and, consequently, the macroscopic thermal-neutron cross section $\Sigma_\alpha^{\wedge} = \sigma_\alpha^{"} N_{\text{Д}}$ are lower in this re-

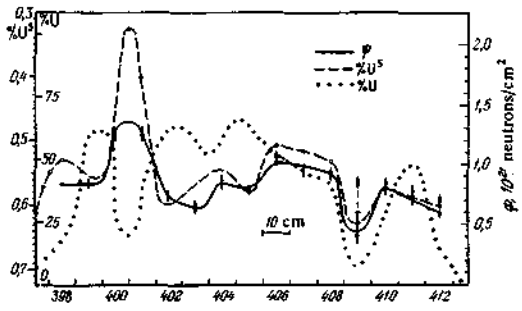

FIG. 4. Distribution of the integrated neutron flux φ, ^{235}U concentration in uranium (£), and the concentration of uranium in the ore. Sample numbers adopted for the Oklo reactor[19,20,21] are plotted along the abscissa axis.

TABLE III.

	SiO_2	Al_2O_3	FeO	Fe_2O_3	MgO
%	41	24	13	4	10

gion. Moreover, chemical analysis shows[12] that it is precisely in this region that the concentration of water is higher by a factor of 2-3, and this increases the density of the slowed-down neutrons q. Since spatial diffusion of thermal neutrons is small in comparison with their capture, it follows that $\$ \sim q/\sim L_a$, and there is a local burst in Φ as q/Σ_a increases. The result is the formation of a neutron trap very similar to that specially constructed in powerful modern research reactors (such as SM and PIK), designed to produce a high thermal-neutron flux.[c23] Judging by the concentration of water in uranium, a similar trap should be present in the 409 region, but the substantial contamination by natural neodymium (greater by a factor of 16 than at 400)[2i,22] means that the data referring to this region are unreliable.

D. Power

The total amount of energy produced by the reactor is estimated as 1.5×10^4 MW-year.[C7] The two blocks of the Leningrad Atomic Power Station generate this amount of energy in 2.3 years under 100% load. If we suppose that the Oklo reactor was operative for d=Q. 6 million years, its mean power output was only 25 kW. Therefore, despite the fact that the fluence at individual points in the hot zone was $\varphi = 1.5 \times 10^{21}$ neutrons/cm^2, the thermal neutron flux $\Phi = \varphi/d$ did not exceed 10^8 neutrons/cm^2-sec.

E. Neutron balance and control

Although studies of the physics of the Oklo reactor are still at an initial stage, the overall picture of the course of the chain reaction is now clear in general outline. Calculation of the neutron balance shows that the criticality conditions should have been satisfied for the Oklo reactor.[c24]

It is clear from Fig. 4 that the concentration of uranium in the active zone is very substantial, and that it was higher by 40% 2 billion years ago. The uranium in the ore appears in the form of uranium dioxide, UO_2. The main components of the remaining rock, excluding water, are shown in Table ΠI.

The elements listed in Table ΠI are moderate neutron absorbers. Iron has the highest absorption cross section among them ($\sigma_{\beta} = 2.55$ b).[C13]

Water is represented in the ores in two ways. More than half of it is bonded water, which was driven off by laboratory heating of the minerals above 500-600°. The remainder is free water which penetrates the ore and fills the voids. The density of the water molecules varies in the range 0.3-0.5 g/cm^3. For the contemporary concentration by weight of uranium (30%), the H_2O/UO_2 weight ratio in the past was 0.2 or more. This figure corresponds precisely to the water-moderated reactor which also uses UO_2 as the fuel with roughly the same ^{235}U enrichment. Since the size of the active zone of the Oklo reactor is comparable with the size of the artificial reactor, one would expect, in advance of any particular calculations, that the condition $k = 1$ will be satisfied.

Direct calculations of k^ for different parts of the active zone yield values of 0.9-1.1, depending on the concentration of water in uranium.[12ii] The relatively high temperature and the relatively hard neutron spectrum reduced η from 1.83 (Chap. 1) down to 1.74-1.78. The magnitude of ε in (1.2) is greater than unity by 1. 5-5%. The probability that resonance capture will be avoided varies from 0.85 to 0.7 when the weight concentration of uranium is varied from 20 to 50%.

The probability 1 - P of leakage from the reactor is determined by its size, and the migration area M^2 is determined by the mean square of the distance between the point at which a fast neutron is created and the point at which it is captured (by which it becomes a thermal neutron). The active zone of the Oklo reactor may be looked upon as a flat pancake of thickness $H \sim 1$ m. If the distribution of the fuel and water over the active zone is constant, the probability of avoiding escape from the reactor is approximately given by $P = (1 + B^2M^2)^{mi}$, where $B = irH^{n}$. The value of M^2 is largely determined by slowing down through interaction with the hydrogen in water and amounts to 50-60 cm^2, so that $P \sim 0.95$. It follows that criticality calculations show that $k = kj^{>} {}^{a} 1$, and the Oklo reactor did, in fact, work.

We now have the question: why did the reactor not explode as soon as it went critical? The answer is that reactors of this type have a negative temperature coefficient and the same self-regulation mechanism that ensures stable and safe operation of nuclear power stations is operative. The temperature in the Oklo reactor may have reached 300 °C or more, so that the entire unbonded water was converted into steam. If, at the same time, the condition $k > 1$ was satisfied, the reactor power and the temperature increased and, under constant external pressure, there was a reduction in the concentration of water-vapor molecules in the active zone. This reduced k because of the reduction in φ and the increase in M^2. The result of all this was the reestablishment of the condition $k = 1$ and the stabilization of the chain reaction, but now at a higher power level.

The increase in the fraction of water in the active zone may have also partially compensated the burning-up of uranium and ensured stable operation of the reactor for a long time. Another method of compensation^ is to burn up the initial highly absorbing impurities. This could have been boron which has been detected in the Oklo ore.[c24] Rare-earth isotopes have also contributed during the initial stage because they have much higher themal-neutron absorption cross sections than boron. R. Naudet has therefore suggested an interesting model for the operation of the reactor: as the strong

absorbers are burned up at one edge of the active zone and uranium at the opposite edge, the active zone shifts and the chain reaction moves along the uranium vein like a flame over a wet log. If this were the case, different parts of the active zone were working at different times and, possibly, continued to do so for different periods of time.

Thus, all the data on the Oklo phenomenon can be naturally explained within the framework of reactor physics.

4. CONSEQUENCES OF THE DISCOVERY OF THE OKLO PHENOMENA

A. Storage of radioactive waste

The surprising survival to this day of the traces of operation of a natural reactor is of major interest in connection with the problem of storage of radioactive waste in nuclear power engineering. Research carried out from this point of view[c26,273] has shown that the reactor itself was an excellent store of the radioactive waste. There has been exceedingly little migration of the heavy elements (thorium, uranium, and plutonium) over 2 billion years. Rare-earth elements and many heavier fission products have remained in place. They were found in the same grains of uraninite as the burnt-up uranium, and did not escape into the surrounding mineral clay.[28] It is precisely this fact that has made possible the analyses summarized in the previous section.

Many of the light fragments such as zirconium, ruthenium, rhodium, palladium, niobium, and silver, have also remained in place almost completely. On the other hand, inert gases, krypton and xenon, have almost completely escaped and, judging by the deficit in their decay products, they left the reactor during its operation. Metals that are readily soluble in water have remained only partially. However, analysis of the distribution of ^{90}Sr ($T_{90} = 41$ years) shows that about 95% of this isotope decayed into ^{90}Zr inside the reactor, and there was very little migration over the period of the order of τ_{90}.[c29] Radiogenic lead has survived only partially because of the considerable change in the chemical properties in the chain of successive radioactive transformations.

Of maximum interest for the study of the migration of fragments is the "contamination zone" on the boundary between the reactor and the remaining ore. The final conclusion is still a matter for the future, but even preliminary studies of the results of the natural experiment performed over almost 2 billion years suggest that it may be possible to achieve prolonged safe storage of radioactive waste from nuclear power stations.[1,30,1]

B. Connection with geology and biology

The chain reaction occurred soon after, or almost simultaneously with, the formation of the Oklo deposit. The results of dating of the surrounding ore by traditional U/Pb, Rb/Sr, and K/Ar methods can therefore be compared with the independently calculated age of the natural reactor, and this is of considerable interest for geochronology.

Another obvious consequence of the discovery of the Oklo reactor is the necessity of a search for further such reactors. Experience shows that this must be done by carefully analyzing the isotopic composition of uranium with the mass spectrometer.

The natural reactor was active during the Precambrian epoch (6×10^8 years), which is important from the point of view of biological evolution. The age of the reactor coincides with the period of transformation of prokaryotes, i.e., cells without nuclei, into the more complex single-cell eukaryotes which have a nucleus ($1.7 - 2.1 \times 10^9$ years ago). The Oklo reactor produced an enhanced level of radiation and may have had a local influence on the mutation rate of the single-cell organisms. Preliminary results have shown the presence of a higher degree of evolution of organic matter as one approaches the reactor.[c31]

C. Possible variation of fundamental constants with time

The Oklo reactor is an instrument sensitive to the magnitude of neutron cross sections in the distant past. By comparing them with contemporary values, one can estimate the extent to which they (and hence the fundamental constants) and are, in fact, constant in time."[23]

In 1937, Dirac suggested that the gravitational constant might be a function of time.[c33] The reason for the weakness of the gravitational interaction between two protons as compared with the electromagnetic interaction was thought by him to be due to the reduction in the gravitational constant in inverse proportion to the lifetime T of the Universe. George Gamow replaced the Dirac hypothesis by a linear increase in the electromagnetic constant: $a = e^2/Kc \sim T$.[C34] Without going into the details of this discussion (see the review given by Dyson[c351]), it must be admitted that the conventional belief in the time-independence of the dimensionless constants of the different fundamental interactions does require experimental confirmation.[t36]

The absence of anomalies in the fine splitting of the spectral lines of oxygen and magnesium ions in the radiation arriving from distant galaxies with red shifts of 0.17-0.5 leads to the restriction $\dot{a}/a < 2 \times 10^{-12}$ year^{-1}.[C37:i] Dyson obtained a more stringent limit by analyzing the β decay of the long-lived nuclei ^{187}Re - ^{187}Os and ^{40}K-^{40}Ar: $\dot{b}t/a < 2 \times 10^{-14}$ years.[35] The lifetime of ^{187}Re ($T = 6 \times 10^{10}$ year) is long because of the low transition energy ($\Delta\mathcal{E} = 2.5$ keV), and a small change in a, producing a different difference between the total Coulomb energies of the daughter and parent nuclei, would have a strong effect on AE and τ. This analysis has resulted in the following restriction on the weak interaction constant ($\beta = Gm_p^2 = 1(\Gamma^5)$: $\dot{\beta}/\beta < 1(\Gamma^{10}$ year^{-1}. Davis considered the additional possibility that ΔE might vary because the density of nuclear matter might be affected by a change in the strong interaction constant g\ (the strong interaction is conventionally characterized by one constant), and has obtained a restriction on its variation with time which is of the same order as the restriction for a.[c38]

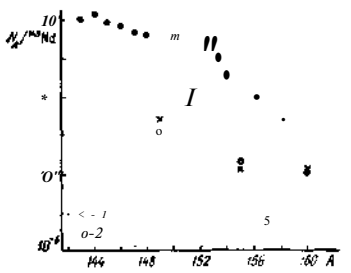

FIG. 5. Comparison of calculated (1) and measured (2) concentrations of fission fragments relative to the ^{U3}Nd concentration for one of the Oklo samples.[11761]

Recently, A. I. Shlyakhter has noted that the sensitivity to changes in the nuclear potential increases by sensitivity to changes in the nuclear potential increases by a few percent if we consider neutron capture.[C32] Because of the sharp resonances in its absorption cross section, the nucleus is a highly tuned detector of neutrons. Resonances along the energy scale will shift if there is a change in the nuclear potential by analogy with the shift in the reception frequency in an ordinary radio receiver when there is a change in the parameters of the resonance circuit.

In the strong absorbers listed above, the resonance is located near zero neutron energy, and the resonance energy itself is of the order of the capture width: $E_r \sim r_r \sim 0.1$ eV. The thermal-neutron capture cross section of these isotopes will change sharply when the resonance is shifted by an amount of the order of $\Gamma_,$. Analysis of the experimental data for ^{149}Sm and ^{151}Eu, based on (3. 6), taking into account three standard errors and the uncertainty in temperature, shows that the shift $\Delta\mathcal{E}_\Gamma$ of the resonances since the operation of the Oklo reactor does not exceed 0. 05 eV.[c32:i] We note, by the way, that one could avoid the determination of the fluence ψ in (3. 6) if the values of N_K/N_O measured at the same point in the reactor were available for two strong absorbers. Instead of (3. 6), one could then use the formula

$$\frac{\langle\sigma_a^{K1}\rangle}{\langle\sigma_a^{K2}\rangle} = \frac{\gamma_{K2} N_{K2}}{\gamma_{K1} N_{K1}}, \quad \langle\sigma_a^{K}\psi\rangle d \gg 1, \tag{4.1}$$

which follows from (3. 5). Comparison of the ratio $(\sigma_a^{*1})/\sigma_a^{*2})$, obtained from (4.1), with the contemporary value can be used to verify directly the absence of a shift in the resonance of one absorber relative to the other.

The absence of an appreciable shift of near-threshold resonances will also follow qualitatively from the fact that all the contemporary strong absorbers were strongly burnt up in the Oklo reactor whereas the weak absorbers were weakly burnt up. To augment the cadmium data (Fig. 3), we reproduce the results of measurements of the concentration of rare-earth elements relative to ^{143}Nd (the right-hand branch of the mass distribution of the fission fragments) in one of the Oklo specimens (Fig. 5).[c17c] The valleys in the distribution correspond to strong absorbers: ^{149}Sm, ^{151}Eu, ^{155}Gd, and ^{157}Gd. The burn-up depth, calculated with the aid of contemporary values of the absorption cross section, is in excellent agreement with experiment, especially if we recall that the neutron spectrum over which the cross section has to be averaged is not all that well known. We therefore conclude that, over the 1.8 billion years since the operation of the Oklo reactor, the resonances or, in other words, the compound-nucleus levels, shifted by less than $\Gamma,/2$, i. e., the mean rate of shift did not exceed $3 \chi 10'^{11}$ eV/year. This is less by three orders of magnitude than the experimental limit on the rate of change in the transition energy in the decay of ^{187}Re.[35:

There are no theoretical calculations at present that would reliably connect the position of each neutron resonance with the nuclear potential parameters. However, even preliminary qualitative estimates can be used to reduce substantially the limits on g_v/g_s and a/a.[c32] These estimates confirm the absence of a power law or a logarithmic asymptotic dependence of the strong and electromagnetic interaction constants on the lifetime of the Universe. As far as the weak and, especially, CP-odd constants are concerned, the possibility of a variation with T has not as yet been experimentally excluded. In this connection, it would be interesting to carry out special measurements at different points in the active zone of the Oklo reactor, and to improve the limit on AE_r for different isotopes. However, it will also be necessary to have more accurate theoretical calculations of the influence of variations in the fundamental constants on the parameters of the neutron resonances.

The discovery of the Oklo reactor is a considerable contribution to different branches of modern science. It seems appropriate to emphasize in conclusion one further aspect of this striking discovery: "In the design of fission reactors man was not an innovator but an unwitting imitator of nature."[Ω]

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