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ABSTRACT

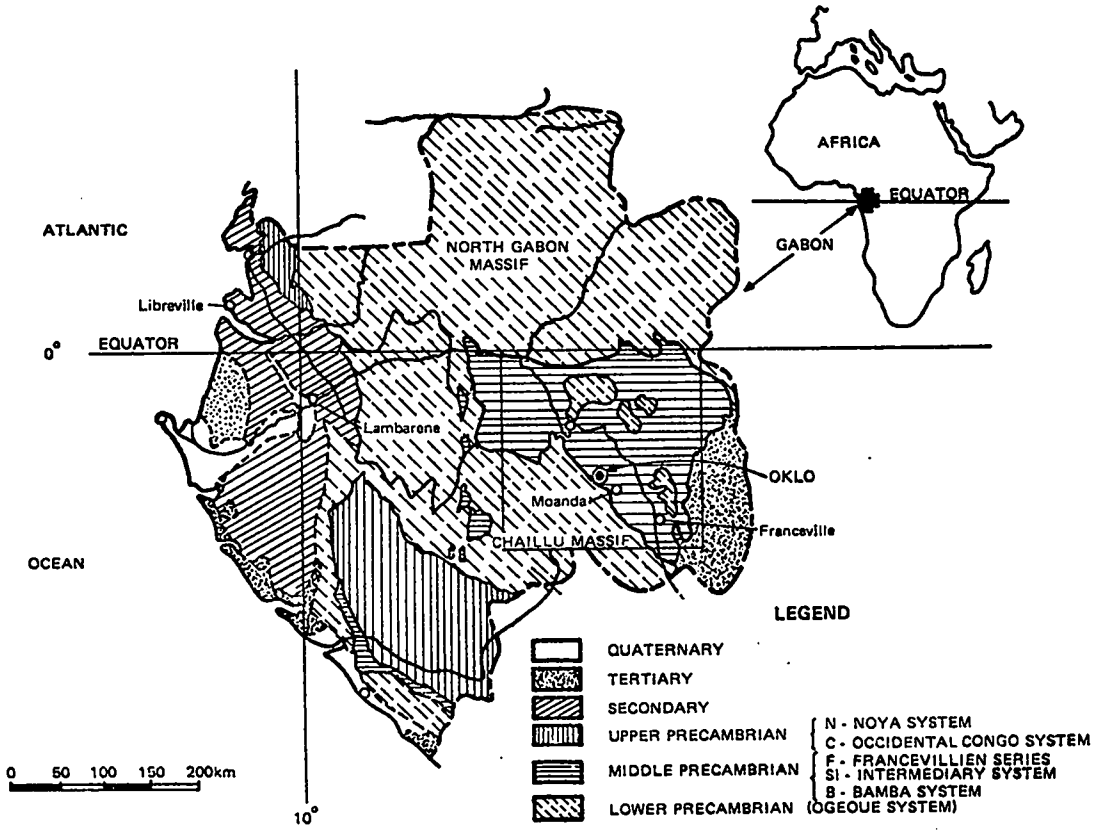
In 1972 scientists associated with the French Atomic Energy Commission announced the discovery of a "fossil" fission reactor in the Oklo mine, a rich uranium ore deposit located in southeast Gabon, West Africa. Further investigations by scientists in several countries have helped to confirm this discovery. The age of the reactor is 1.8 billion years. About 15 000 megawatt-years of fission energy was produced over a period of several hundred thousand years equivalent to the operation of a large 1 500-MW power reactor for ten years.

The six separate reactor zones identified to date are remarkably undisturbed, both in geometry and in retention of the initial reactor products (approximately six tons) deposited in the ground. Detailed examination of the extent of dispersion of Oklo products and a search for other natural reactors in rich uranium ore deposits are continuing. Information derived from fossil reactors appears to be particularly relevant to the technological problem of terminal storage of reactor products in geologic formations.

In September 1972 two unusual communications appeared in the annals of the French Academy of Sciences. Almost on the thirtieth anniversary of the start-up of the first manmade fission reactor at Stagg Field in Chicago, scientists associated with the Commissariat a l'Energie Atomique announced the discovery of the fossil remains of a prehistoric natural reactor. The setting was as esoteric as the event, an open-pit uranium mine located in the southeast part of the Republic of Gabon, just below the equator on the coast of West Africa. The disclosure climaxed an investigative tour-de-force reminiscent of Agatha Christie's detective Poirot at his best.

It began inauspiciously enough in May 1972 with the observation by mass spectroscopist H. Bouzigues of a slight depletion in the isotopic ratio of uranium-235 to uranium-238 in a standard prepared at Pierrelatte, the French nuclear fuel production plant. Terrestrial uranium contains 0.7110 weight percent or 0.7202 atomic percent of the easily fissionable isotope mass 235. The freshly prepared standard was found to contain 0.7171 atomic percent of mass 235, a relative decrease of 0.4% compared to the accepted, presumably constant natural value. The difference might have been attributed to contamination of the sample with a small amount of production "tails," uranium depleted in mass 235 in the gaseous diffusion plant. But the low values persisted on repeated analyses. Puzzled scientists traced the source of the uranium back through a processing plant in France to the Mounana mill near Franceville in Gabon. The ore originated in the nearby Oklo mine operated by C.O.M.U.F., the Compagnie des Mines d'Uranium de Franceville. Analyses of scrupulously preserved samples taken from each batch of the Mounana mill output showed that shipments of slightly depleted uranium to France had begun in 1970 and were still continuing. By mid-1972 the total shortfall of uranium-235 in the French purchases was of the order of 200 kilograms involving a total of 700 tons of processed uranium.

The ore body at Oklo had been defined by core sampling on a closely spaced grid. Some of the cores were stored in France and it was possible to reanalyze portions of them individually. Several were found to be strongly depleted in the light



GEOLOGIC MAP OF GABON

The Oklo mine is located in southeast Gabon at the edge of a middle Precambrian sedimentary basin bordering on a large expanse of lower Precambrian crystalline rock. The ore is in the Francevillian series, just above a sandstone formation, and contains facies varying from coarse-grained sandstones to fine-grained clay minerals. The ore body dips downward at an angle of 45°.

isotope including one, containing only 0.44% mass 235, which came from a zone in the ore body currently being mined.

The size and complexity of the gaseous diffusion plants at Oak Ridge and Pierrelatte testify to the difficulty of separating isotopes of the heavy elements. There seemed to be no plausible mechanisms for achieving such separations naturally. But the investigators persisted. Could the missing mass 235 have been destroyed by a copious flux of neutrons? The key analyses were quickly carried out and suddenly the explanation was obvious. Fission product elements were present in abundance in the depleted uranium vein. They were almost absent elsewhere. Their isotopic abundances were quite different than those in the natural elements. In fact, they corresponded to the yields expected from fission. Three months after the investigation had begun in earnest, the main mystery was solved. Nature, not man, had constructed the world's first fissioning chain reactor.

The evidence for a fission reactor is convincing. It is both quantitative and redundant. Although all of the radioactive products of the reactor have long since decayed to stable, nonradioactive isotopes, both the absolute amounts and the isotopic ratios of the elemental species can be explained only by their origin in fission. Remarkably, at least half of the thirty-odd elements in the fission product distribution remained immobilized in the ore and are still there after 1.8 billion years, the age of the ore deposit and the reactor. These include the rare earth elements lanthanum, cerium, praseodymium, neodymium, europium, samarium,

gadolinium, and the chemically similar yttrium. Most or all of the zirconium, ruthenium, rhodium, palladium, niobium, and silver, some of the molybdenum and iodine, and even traces of the rare gases krypton and xenon, remain. As might be expected the relatively soluble monovalent and divalent metals, rubidium, cesium, strontium, barium, and probably cadmium have largely migrated away. However, there is no appreciable deficit of zirconium-90 indicating that strontium-90, with a 30-year half-life, decayed to zirconium before very much strontium had migrated. Lead, a product of uranium decay, has partly migrated. Other fission products are made in low yields compared to their natural backgrounds and their fate has not yet been determined.

When immobile fission product elements with more than one isotope are separated from the ore and analyzed mass spectrometrically, the observed isotopic ratios can be interpreted in terms of two sources, natural background and fission. Neodymium is particularly suitable as a measure of fission because it is not abundant in nature and, of its seven isotopes ranging from mass 142 to 150, only six are made in fission. The mass 142 chain in fission does not decay to neodymium but stops at cerium and therefore neodymium-142 serves to measure the very small amount of natural element in the ore. When the six other isotopes are corrected for background and neutron capture effects, they are found to be present in ratios which correspond precisely to the well-known fission yield distribution for thermal neutron-induced fission in uranium-235.

Apparent discrepancies occur at masses 143 and 144 because the neutron capture cross section of neodymium-143 is very large and a significant fraction of it is transformed to neodymium-144. A much

ANALYSIS OF Nd ISOTOPES
Sample Oklo M (KN-50)

$$\frac{^{235}U}{^{235}U + ^{238}U} = 0.4400 \pm 0.0005 \text{ at.\%}$$

(1)	(2)	(3)	(4)	(5)	(6)	(7)
Nd Isotopes	Natural Nd (at.%)	Measured Sample (at.%)	(3) Corrected for Natural Element	(4) Corrected for Neutron Capture $\phi t = 1.5.10^{21}$ n/cm^2	Nd from ^{235}U Fission (at.%)	(5) / (6)
142	27.11	1.4	0	0	0	
143	12.17	22.1	22.6	28.6	28.84	0.99
144	23.85	32.0	32.1	26.4	26.50	1.00
145	8.30	17.5	18.0	19.0	18.97	1.00
146	17.22	15.6	15.6	14.6	14.42	1.01
148	5.73	8.0	8.1	8.1	8.26	0.98
150	5.62	3.4	3.3	3.3	3.12	1.06
Relative Accuracy	0.2%	2 to 3%			0.5%	

smaller but similar effect exists at masses 145 and 146. If the appropriate neutron capture cross sections are accurately known, the size of the discrepancy is a measure of the total neutron exposure in each sample of ore. Given the neutron exposure, the depletion of uranium-235 can be calculated but it is always considerably higher than the observed value. The relative discrepancy is generally about 40%. Most of this difference is due to the fact that uranium-238 captures neutrons and decays quickly by beta emission through neptunium to plutonium-239 which then decays with a 24 400-year half-life to additional uranium-235. The observed depletion in this isotope is less than the calculated depletion by a factor which is related to (1-C) where C is the "conversion" factor, the ratio of capture in mass 238 to fission in mass 235. The conversion factor increases wherever the neutron spectrum is not fully thermalized. In some parts of the reactor it reaches a value of nearly 0.8.

Further evidence for a fossil reactor is found in the thorium content of the ore. For every six atoms of uranium-235 which undergo fission with thermal neutrons, one atom simply captures a neutron and becomes uranium-236, an alpha emitter with a half-life of 23 400 000 years. All of the mass 236 made in the reactor has by now decayed to nearly stable thorium-232. Thorium is, indeed, found in the reactor zones in the expected amounts and is nearly absent outside. Similarly bismuth, which is the stable end product of the decay of reactor product uranium-237, is present in quantity only inside the reactors.

No other reasonable hypothesis can account for all of these observed facts: multiplying chain reactions occurred.

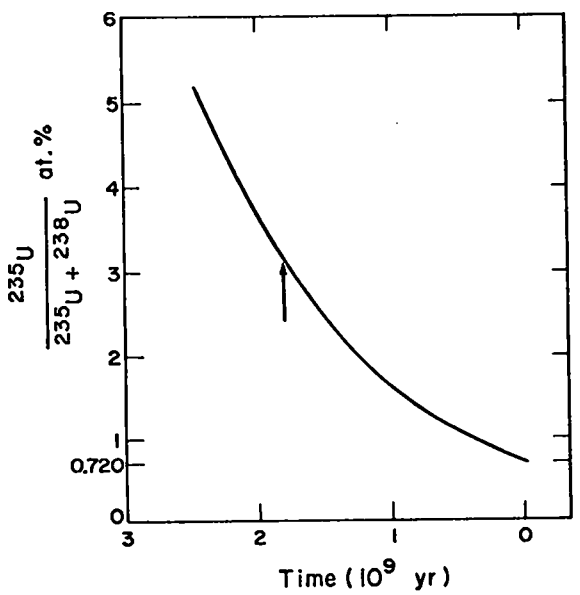
What conditions were necessary to achieve a nuclear reaction at Oklo? The list of requirements is far from trivial. The number of neutrons made per fission in uranium-235 is 2.5. In order to sustain a chain reaction, one of these must be absorbed to produce fission; 1.5 can be absorbed elsewhere or escape. To satisfy this condition, the following are the minimum requirements for a chain-reacting ore deposit:

1. We know that chain reactions are possible in present-day natural uranium containing only 0.72 atomic percent uranium-235 but the moderator must

be nonnatural heavy water, enriched in deuterium, or the reactor must be constructed in a geometric lattice of uranium and moderator with carefully specified dimensions. We can assume that such a lattice will not occur naturally. Nature is more likely to assemble an essentially infinite mass of relatively pure uranium oxide with an optimum amount of admixed water moderator. Under these conditions an absolute minimum abundance for criticality is 1% which means that natural reactors can no longer occur. In fact they must be greater than 400 000 000 years old. In the much older Oklo deposit the mass 235 ratio was 3%.

2. The uranium concentration in the ore must average 10% or more in seams at least 0.5 m thick. In a thin deposit neutrons escape and do not sustain the chain reaction. The reactor zones at Oklo meet the thickness requirement.

3. The ore must contain sufficient moderator to slow the neutrons to a thermal distribution. Otherwise the uranium-238 captures too many neutrons and "poisons" the reaction. By far the best naturally occurring moderator is water. The optimum ratio of atoms of hydrogen to uranium varies with



The relative atomic abundance of uranium-235, which decays by alpha emission with a half-life of 7.04 x 10^8 years to thorium-231, decreases with age. The most abundant isotope, uranium-238, decays much more slowly.. Uranium-235 was 17% abundant in uranium when the earth was formed 4.6 x 10^9 years ago; 3.1% abundant when the Oklo phenomenon occurred; and is 0.72% abundant now. The arrow points to the abundance at the time the Oklo reactor operated.

the uranium concentration and the amount of mass 235. It is 10 at 15 weight percent uranium in a two-billion-year-old ore and 5 at 75 weight percent uranium in a 500-million-year-old ore. This requirement means, for instance, that the older ore must contain 6% by weight water. The water of crystallization in a sedimentary ore should more than satisfy this requirement. In addition, at Oklo the medium was probably saturated with water which would have further overmoderated the neutrons. If the ore became chain-reacting in this condition, it would boil off water until it became slightly undermoderated, eventually moving back to optimum moderation. Thus, the initial moderator requirement has rather broad limits.

4. The concentration of neutron poisons must not be too great. Large quantities of heavily neutron-capturing elements such as lithium, boron, or the rare earths can prevent criticality. Based on present-day analyses, there is no evidence to indicate the presence of excessive amounts of neutron-capturing "poison" in the Oklo ore prior to initiation of the chain reaction.

It should be noted that these conditions refer to parameters which varied during the course of the reaction, changing its rate and eventually stopping it entirely. They are responsible for both short-term and long-term control of the power level and functioned in such a way as to make the reactor incapable of achieving very high power levels.

The relative abundance of mass 235 in the Oklo reactor decreased with increasing neutron exposure. (Under very special conditions which will be discussed later, the conversion factor can exceed one and make the reactor a "breeder.") This isotopic depletion effect, with a consequent decrease in reactivity, can be compensated by an increase in uranium concentration or deposit thickness during the reaction. Uranium accretion or loss can also occur after the reaction.

Some increase in reactivity probably occurred due to the burnout of neutron-capturing "poisons" at a faster rate than the fissile atoms were destroyed. If the initial amounts of elements such as lithium and boron were large enough, this effect could have been a major form of control.

The amount of water varied during the reaction, serving as a short-term control mechanism. If the

reactor were buried deeply enough, the boiling point of water may have been as high as 300°C. When the reactor reached this temperature, water boiled off until a critical hydrogen-to-uranium ratio was reached at which point the power leveled off and remained approximately constant, just high enough to equal the rate of heat transfer to the region outside the critical zone. An initially overmoderated reactor has a positive coefficient of reactivity as water boils off and moderation becomes optimum. However, the process continues until the reactor is undermoderated and the temperature coefficient of reactivity becomes negative. The excess reactivity attributable to its initial overmoderation serves, in the end, as a form of long-term reactor control.

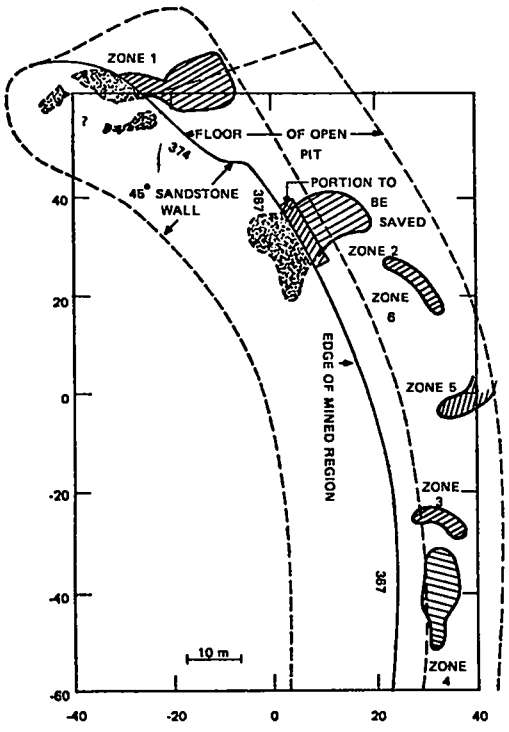
The possibility of a sustained nuclear reaction in a natural uranium ore deposit was considered in the 1940's and '50's by a number of scientists. In 1953 Wetherill and Inghram stated at a Wisconsin conference on nuclear processes in geologic settings: "It is interesting to calculate the degree to which (this) pitchblende deposit was an operating pile. (Our) calculation shows that 10% of the neutrons produced are absorbed to produce fission. Thus, the deposit was 25% of the way to becoming a pile. It is also interesting to extrapolate back 2 000 million years where the uranium-235 abundance was (4%) instead of 0.7. Certainly such a deposit would be closer to being an operating pile." Three years later Professor Paul K. Kuroda of the University of Arkansas described the requirements for a natural reactor in a terrestrial uranium deposit in more detail. His description of an "unstable" ore mass comes very close to fitting the conditions at Oklo. Despite such predictions, the announcement of the Oklo reactor was received with some scepticism by American nuclear scientists. Some of the world's best physicists had assembled the Stagg Field reactor with the most exquisite attention to mechanical detail, purity, and geometry. Could nature have achieved the same result so casually?

We know now that the answer is "Yes." Remember again the difference in time. In the 1.8 billion years between Oklo and Stagg Field, uranium-235, with a 704 000 000-year half-life for alpha decay, fell to one-fifth of its initial abundance in the ore deposit. The more stable uranium-238 decayed only 30% in the same time. Thus a Precambrian nuclear physicist would have found it almost easy to build a nuclear reactor. In retrospect, it seems inevitable that nature did it accidentally.

There appeared to be few reservations concerning the reality of the Oklo reactor among an international group of 70 investigators from 20 countries which met in Libreville, the capital of Gabon, late last June to discuss "The Oklo Phenomenon." The meeting was sponsored by the International Atomic Energy Agency, the French C.E.A., and the government of Gabon. Its business was to present and review all of the information available from Oklo analyses carried out largely in French laboratories but also in the United States, England, Russia, and Australia.

A much more detailed picture of the phenomenon emerged in the course of this week-long meeting. It opened with a spectacular expedition, a flight over the tropical rain forest to Franceville in southeast Gabon and from there some kilometers by car to the floor of the huge Oklo pit. From a rostrum decorated with palm fronds, representatives of the I.A.E.A., C.E.A., and ministers of the Gabonese government welcomed us. We sat for an hour on benches set at the edge of reactor 2, one of the six reactor zones which have been discovered so far. It was marked out by pegs driven into the ground connected by strings which defined the sampling lines. The chief geologist of C.O.M.U.F., J. P. Pfiffelmann, took his place at the edge of this zone and lectured us on the geology of the formations surrounding us. Behind him a sandstone wall slanted up at a 45° angle, ripple marks on the face attesting to its beach-like origin. The exposed reactor zone, about a meter wide, extended along the pit floor to the north for 10 meters at the interface with the wall. An unmined ore bench rose immediately beyond it, displaying a continuation of the uranium vein. Atmospheric oxygen and water combined to convert traces of black reduced uranium to an oxidized slurry which formed bright yellow plaques on the buffy sandstone-clay ore.

We were looking at what had once been a river delta. Eons ago the crystalline, igneous rocks were eroded by running streams. Minute traces of heavy metals and oxides accumulated in bottom sediments and pebble conglomerates. Swampy waters were rich then with single-celled organisms, the highest



The six reactor zones located to date in the open-pit Oklo mine are indicated by the crosshatched outlines. The speckled areas in zones 1 and 2 represent the presumed borders of reactor zone ore which had already been mined and processed when the phenomenon was discovered in 1972. The ore body slants downward at 45° so that reactor zones 3, 4, 5, and 6, as shown in this plan, are not exposed but have been found by core drilling. The floor of the pit is 50 meters below the surface as it existed at the start of mining.

The reactor zones are of the order of 10 to 20 meters on a side and about one meter thick. They occur in rich lenses of ore, averaging 20-30% uranium by weight which is more than 50 times the average concentration in the ore.

The segment of zone 2 indicated as "portion to be saved" will be suspended in mid-air, pinned to the sloping sandstone wall, as the floor of the pit continues to be lowered by mining operations.

existing form of terrestrial life. Here the sediments became loaded with organic material. As a result of photosynthesis induced by proliferating blue-green algae, the oxygen content of the water became sufficiently high in some regions to convert reduced uranium to more soluble oxidized compounds. The uranium remained in solution until it reached the delta sediments, rich in organic ooze. There it was immobilized again in a reduced form and compacted into the sandstone ore which now surrounded us. The basement granite slowly sank and the sedimentary layer deepened. But relatively soon the granite to the west rose and tilted the sedimentary overburden to its present 45° angle. The ore layer, which averaged 0.5% uranium by weight, was fractured and water circulated through the new channels, producing pockets of rich ore in a clay-sandstone which in some places became nearly pure uranium oxide. It was in these rich pockets that fission chain reactions were initiated as soon as a critical mass of uranium was formed.

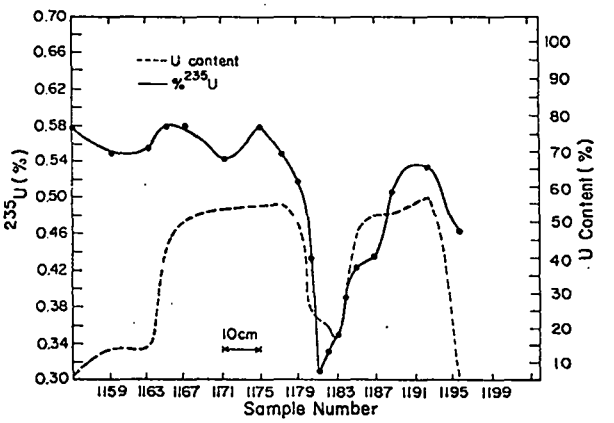
If uranium can be so easily dissolved in oxygenated ground water, how can the Oklo uranium deposit have survived intact for almost two billion years? The geologists told us that the basin containing the Oklo sediments apparently sank to a sufficient depth to protect the uranium ore from redissolution over most of its geological history. Only in recent times, within the past few million years, has the ore horizon approached the surface where normal prospecting efforts could succeed in identifying it. Thus the series of special circumstances necessary to the discovery of the Oklo phenomenon includes not only the processes which produced a natural critical mass of uranium but the unusual geophysical and geochemical conditions which preserved the ore body for nearly half the lifetime of the planet and finally brought it to the surface.

Back in Libreville we learned from R. Naudet, the director of the French Oklo study of "Project Franceville," that their most recent estimate of the total fission energy produced in those reactor zones identified to date was 15 000 megawatt years corresponding to the operation of a major nuclear power plant for 10 years and the fissioning of six tons of uranium. Only about two-thirds of these fissions occurred in the uranium-235 initially present. Most of the remainder occurred in additional mass 235 made by neutron capture in uranium-238 followed by alpha particle decay of the resultant plutonium-239. A few percent occurred by fast neutron fission in uranium-238 and another few percent occurred in plutonium-239 before it decayed. The fact that there was so little fission in plutonium-239 indicates that the reaction went on much longer than the 24 400-year half-life of this isotope, which means that the duration was of the order of hundreds of thousands of years. The same conclusion can be drawn from a consideration of the heat transfer rate. The reactor could not have continued to

operate at a temperature so high that the necessary water moderator was completely boiled off. Simple transfer calculations show that if the power level exceeded a few tens of kilowatts, the chain reaction would stop. A major unknown in such a calculation is the amount of water which might have been actually flowing through channels in the formation. In any case, if the power level were as high as 100 kilowatts, the operating time would be 150 000 years. The total elapsed time from start-up to final shutdown was probably longer because the reactor did not necessarily operate continuously.

The unusual physical and chemical stability of the ore deposit and its reactor products was confirmed in considerable detail by the analysis of samples taken every 2.5 centimeters along several cross sections of the reactor zones. On this scale there was generally good correspondence between the extent of depletion in the uranium-235 and neutron fluences calculated from neodymium analyses. Clearly, relatively little uranium migration occurred inside the reactor zone. Some puzzling anomalies were found at the borders. However, the geometry of the reactors was well-preserved. The distribution of neutron fluences corresponds to that which is calculated for a somewhat undermoderated reactor. If random mixing had occurred, the measured distribution would also be random. Excursions in the fluence occur at faults in the zone which were probably water-filled channels at the time of criticality. The water served to trap neutrons and increase their density even though the uranium concentration went down in this region. Such "neutron traps" are a much used design feature in modern-day, high-flux reactors.

Scientists at the University of Paris used an ion probe to obtain information on the distribution of isotopes within individual grains. On this scale partition of several of the fission products could be seen between grains of uraninite and clay. However, the rare earths and uranium were in the same grains. Even more striking was the fact that, within the resolution limit of 0.0001 centimeters, there was complete correspondence between the uranium-235 and uranium-238. Because nearly half of the residual mass 235 was the daughter product of plutonium-239 synthesized in the nuclear reactor, the fact that no region enriched in mass 235 could be found meant that



Samples taken every 2.5 to 5 centimeters through the reactor zones generally show evidence of correlation between higher neutron fluences (and, therefore, greater depletion in uranium-235) with higher uranium concentration in the ore. However, in the case shown here, a crack is visible in the ore between samples 1179 and 1183. Apparently it existed at the time of reactor operation and was filled with water. Although the uranium concentration goes down sharply in the vicinity of the crack, the neutron fluence and uranium-235 depletion increased. This was probably due to the effect of the water in thermalizing and trapping an undermoderated neutron spectrum. Such "neutron traps" are a common feature of modern-day, high-flux reactors.

the precursor plutonium had remained completely fixed for times comparable to its lifetime of 24 400 years.

Both Frejaques of France and Walton of the United States called attention to the relevance of Oklo to the problem of long-term geologic storage of nuclear wastes, especially to the stability of the long-lived, troublesome plutonium. Walton said "We must regard the survival of the Oklo deposit as an unusual but not a unique phenomenon ... in a considerable number of places, the original structures of the earliest eras have been remarkably well preserved ... However, not many geological structures are so little metamorphosed as the uranium vein at Oklo ... There has never before been an opportunity to document the geochemical stability of a mineral formation in such remarkable detail as in the Oklo case ... Apparently, the major radioactive products that might have been measured in the surrounding environment at the time of the reaction would have been due to krypton-85 and, possibly, to cesium-137 and strontium-90 ... Relevance of these tentative conclusions (regarding the migration of radionuclides) to other geologic formations must await further study of comparative chemistry and geology."

Among the results presented by the French and U.S. participants was an evaluation of the age of the reactor obtained from analyses of a number of Oklo ore samples. If the uranium and neodymium were both present in their correct ratios, then we can use the observed absolute uranium concentrations, the mass-235 depletions, the absolute neodymium concentrations, and the neodymium isotope ratios to calculate the date of initiation of the nuclear reaction. A best fit to the U.S. data occurred in the age interval 1.7-1.9 billion years in good agreement with dates for the host geological formation which had been determined by rubidium-strontium, uranium-lead, and potassium-argon measurements.

The symposium closed with round-table discussions on three topics, the geology, isotopic geochemistry, and reactor physics of Oklo. These subjects represent the principal disciplines which are relevant to the study of the Oklo phenomenon. One more subject intruded and that was economics. The company which helped sponsor the meeting, C.O.M.U.F., had patiently suspended mining the very rich ore in the reactor zones for three years. But now it was time to deepen the pit and mine out the uranium in the protected regions. Presumably ore from the highly depleted portions would be separately processed or segregated. It was proposed to wall off and pin a portion of reactor zone 2 to the 45° sandstone face, leaving the ore suspended over the new pit floor, an elevated monument to the first discovery of a natural reactor.

Outside the conference there was some free-wheeling scientific speculation. Did procaryotic, nonmitotic cells evolve to eucaryotic cells as early as 1.8 billion years ago? Was it possible that Africa was not only the cradle of man but that Oklo was responsible for the first evolution to mitotic cells? There was general agreement that the plot should be worked into a science-fiction story but that the movie rights would be worthless. It would be much better to introduce a spaceship from another planet which had dumped its used reactors on the site, replenished its fuel supply, and departed. Unfortunately, no one had thought to invite Stanley Kubrick to the meeting.

The end of the symposium did not signal a conclusion to Oklo studies. They may, in fact, have only begun. There remains an extensive collection of ore samples at Saclay under the stewardship of M. Roger Naudet. They are available to other qualified investigators. Large numbers of precise analyses on a wide variety of reactor products and associated minerals can lead to a much more detailed description of all the important parameters of the phenomenon including uranium accretion and loss, reactor control, and final shutdown. It is a unique event in geology to have a quantitative inventory of 30 elements inserted into a given environment at a known moment in Precambrian time and to have the whole assembly available for study in an intact geometry. A very large amount of effort will be required for the various investigations of interest. More measurements are being planned but it is possible that the scale of the investigation, however impressive, will never fully exploit the opportunity for new information. There was some discussion of the possibility of broadening the investigation to an international search for other natural reactors with planning provided by a steering or advisory group sponsored by the I.A.E.A. However, to date the efforts have been largely unilateral.

What do we wish to do as a follow-on to the analysis of Oklo samples? Clearly, it would be interesting to know if other Oklo-type reactors exist. Apart from the possible significance of finding new Oklos to questions of purely scientific interest, authorities who are concerned with the long-term storage of nuclear wastes want to know if Oklo is unique or whether additional stable reactor deposits exist in other geological settings. There is also a question important to producers and buyers of uranium: how much valuable mass 235 has been burned out of ore reserves? It is a question of equal interest to monitoring authorities who are concerned with accounting for all fissile material and are expected to notice mysterious shortages of a few hundred kilograms of uranium-235. In addition, in these days of intense prospecting for new sources of uranium, does the Oklo phenomenon tell us anything about the way these ore deposits were accreted and, possibly, where to find them?

It was pointed out earlier that as the oxygen content of the atmosphere rose in Precambrian times, uranium in low-concentration deposits which were exposed to oxygenated water could become mobile as hexavalent uranium and then reconcentrate in richer

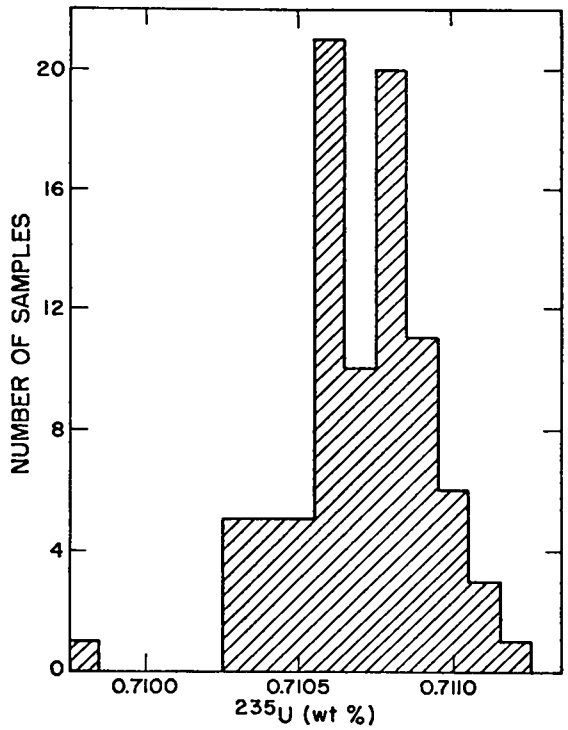
ore deposits wherever a highly reducing environment existed. Rich uranium ore deposits are found in several parts of the world in geologic formations of approximately the same age, about 1.6 to 1.8 billion years, not only in Africa but also, particularly, in Canada and northern Australia. As of now, none have been identified as reactor sites. It seems quite possible that more such deposits were formed, that they supported nuclear chain reactions, and that they then disappeared. They may have been buried under younger sediments where they are unlikely to be discovered. They may also have been dispersed by geophysical instabilities or geochemical mobility. How probable is such dispersion? Is it likely or unlikely?

These questions bear not only on the probability of finding new Oklos but, obviously, on the evaluation of the life expectancy of long-term geologic storage sites. Their importance is even more general. The number of significant deposits of valuable minerals is determined by the difference between their rates of formation and rates of dispersion. Although the problem of dispersion rates is generally overlooked by mineral exploration geologists and is not discussed in most geochemistry texts, scientists responsible for the long-term storage of reactor products cannot ignore it. It is a question which deserves to be examined with respect to ore deposits of many individual elements in a variety of geological and geochemical settings. The Oklo deposit, therefore, is not a unique source of relevant information. The particular importance that attaches to it so far as the long-term storage problem is concerned is that it contains all of the elements of interest in one place and demonstrates that, at least in this one environment which may be unique, the deposit was geophysically and geochemically stable over an enormously long period of time.

The evidence for geochemical stability probably comes as no surprise to a number of waste storage scientists who, for many years before the discovery of Oklo, had been investigating the adsorptive properties of shales and clays and the extreme stability of many bound elements in such matrices. Suggestions for reactor product storage in adsorptive geological formations have been under active study for years in the United States and elsewhere. It is obviously difficult to devise an experiment which will evaluate the stability of an adsorbed cation for a million years or more under field conditions. Because Oklo resembles such an experiment, the data have been examined with great care to determine what part is relevant to the current technological problem.

Some puzzling information exists in ERDA files. It has not been widely publicized but is now being reexamined in light of Oklo. Scientists have widely accepted the fact that the ratio of uranium-235 to uranium-238 is a constant in our solar system. Most of the mass spectroscopic analyses of many terrestrial ores, meteorites, and moon rocks on which this conclusion is based were obtained with thermal ionization sources and have a characteristic error, at the 95% confidence level, of the order of 0.1% relative. Within this error, the isotope rate does, indeed, appear to be constant. However, at the uranium hexafluoride gaseous diffusion plants which produce uranium enriched in mass 235, very precise gas mass spectrometry is performed routinely, not only on the product and "tails" but on the feed material. The characteristic error in samples analyzed at production plants such as Pierrelatte, Oak Ridge, Paducah, and Portsmouth, is 0.01%. When isotopic abundance data from a set of nearly 100 analyses of ore carried out in the U.S. plants are examined, it becomes evident that the 235/238 ratio in uranium from sedimentary deposits in the Colorado Plateau region is approximately 0.03% lower, relatively, than the same ratio in uranium mined from deposits located mostly in "magmatic"-type ores outside the United States. The difference is small but significant enough to reopen the question of the possible effect of dispersed fossil nuclear reactors on the abundance of uranium-235 in natural ores. More than 10 years ago the U.S. Atomic Energy Commission recognized a variability in the 235/238 ratio which might be as high as 0.1%. However, it was generally believed without being proved that it was due to chemical differentiation in sandstone rocks. At the present time we do not have enough information to decide which explanation of this variability is most likely to be correct.

If it were possible to disperse an Oklo-type reactor which had undergone a net depletion of five tons of mass 235, as at Oklo, into a uranium reservoir the size of the total reserves in the Colorado Plateau, which are of the order of 400 000 tons of



The bimodal distribution of the ratio of uranium-235 to uranium-238 in 88 ore samples taken from various parts of the world is largely due to slightly lower values which are consistently found in samples of uranium ore taken from the Colorado Plateau region in the United States. The decrease from the slightly higher valued mode representing samples taken chiefly in Canada, Europe, and Australia is 0.03% relative. It may be due to chemical differentiation of uranium isotopes in the Colorado Plateau or to dispersion of uranium from a Precambrian deposit which was depleted in a nuclear reactor similar to the Oklo reactor.

uranium, the net overall depletion in the uranium-235 abundance would be about 0.03% relative. Thus we cannot dismiss the dispersion and mixing hypothesis on the basis that the effect would be negligible.

If dispersion was not the general fate of natural reactors, we can hope to find additional reactors by surveying core samples from rich uranium deposits which are greater than one billion years old. One way to do this is to use field instruments which can measure the 235/238 ratio with sufficient accuracy to reveal depletions of 10% or more without processing the ore. The cooperation of industrial and government authorities will be required to identify those core samples from blocks of ore which meet most of the criteria for criticality identified earlier in this report. We are, of course, eager to hear from those who are aware of deposits which merit further investigation.

Why should a mining corporation cooperate in this search when a successful result is likely to produce a lower selling price for some of its richest ore? We believe that cooperation will be forthcoming for two reasons: we have found that an appeal to scientific curiosity can be very strong even in the most profit-oriented enterprises; in addition, the prospect that identification of depleted uranium may occur after mining and processing rather than before, with an even greater risk of monetary loss, should help convince conscientious managers of the need for more information.

It was also mentioned earlier that massive deposits of pure pitchblende can become critical at ages considerably less than one billion years. However, the younger ores have the peculiar property of approaching or even exceeding the "breeding" ratio in which all the burned-up fissile material is replaced by neutron capture in uranium-238. Thus, large deposits of pure pitchblende of, say, 800 000 years of age such as existed in the mined-out Belgian Congo would not necessarily be discovered by a field examination for depletion in mass 235. Such deposits may, in fact, be very slightly enriched. It is provocative to observe that the rather few samples of old Belgian Congo ore for which we now have precise gas mass spectrometric analysis do appear to be slightly enriched in mass 235 (0.03% relative) with respect to the world average of "magmatic"-type ores.

An even more difficult fossil reactor to identify will be one in which most of the uranium has been dissolved away, leaving intact the less mobile products such as the rare earths. If a field geologist were to find an ore pocket containing rare earths, yttrium, zirconium, niobium, ruthenium, and rhodium, all in abundances of the order of 0.01-0.1%, perhaps he will remember having read about Oklo and arrange to have these elements analyzed mass spectrometrically. Quite probably he will find abnormal isotopic distributions. Otherwise such a phenomenon, if it exists, is unlikely to be discovered.

On the whole, there is reason to believe that we shall find more fossil reactors. If there had been only Oklo, the statistical probability of finding it must approach zero. When we consider

the almost accidental way in which it was discovered, we are tempted to conclude that other such reactors have already been mined out without discovery. The search for uranium is intensifying. Unless all the Precambrian uranium lodes have now been found, which is most unlikely, we can probably safely assume that soon an Oklo Prime will be announced. In due time we shall learn whether to regard the survival of the Oklo deposit as a unique phenomenon in natural history or as a particularly valuable experiment in long-term geological storage from which we can draw useful, general conclusions. It does not seem too far-fetched to anticipate that natural reactors will tell us under what conditions reactor products, particularly plutonium, should remain fixed indefinitely. In any case, one message from the past is already clear. In the design of fission reactors man has unwittingly imitated rather than anticipated nature.