A Natural Fission Reactor

Two billion years ago in west Africa a rich deposit of uranium ore began operating as a nuclear reactor. Long dormant but preserved intact, the reactor came to light during the mining of the deposit

by George A. Cowan

In 1942, when Enrico Fermi and his associates started up their nuclear-fission reactor at Stagg Field in Chicago, there was every reason to believe it was the first such reactor on the earth. The record book must now be corrected. In an open-pit uranium mine in the southeastern part of the Gabon Republic, near the Equator on the coast of West Africa, are the dormant remains of a natural fission reactor. Within a rich vein of uranium ore the natural reactor once "went critical," consumed a portion of its fuel and then shut down, all in Precambrian times. The experiment at Stagg Field had been anticipated by almost two billion years.

The history of the natural reactor is an extraordinary sequence of seemingly improbable events. First, uranium from an entire watershed accumulated in concentrated local deposits, including one at a place now called Oklo. Then the conditions necessary to sustain the fission chain reaction were established; these included constraints on the concentration of uranium in the ore, on the size and shape of the lode and on the amount of water and other minerals present. After the reactor had shut down, the evidence of its activity was preserved virtually undisturbed through the succeeding ages of geological activity. Finally, the discovery of the reactor involved an investigative tour de force worthy of the best sleuths in detective fiction.

The first clue was found by H. Bouzigues, who is on the staff of the nuclear-fuel-processing plant at Pierrelatte in France. In May, 1972, he obtained a curious result during a routine analysis of a standard sample prepared at Pierrelatte from uranium ore. Natural uranium consists mainly of the isotope of atomic mass 238; only .7202 percent of the atoms are the easily fissionable isotope of mass 235. In the freshly prepared standard Bouzigues found the proportion of U-235 to be even smaller than the usual value: it was .7171 percent.

Bouzigues's analysis was performed by mass spectrometry, in which molecules are ionized and accelerated, then deflected by a magnetic field. The mass of the molecule is revealed by the extent to which it is deflected. The technique is a delicate and precise one, and when it is applied to the gaseous feed materials employed in the enrichment of uranium, it can achieve even greater accuracy than is usual under most other circumstances. The discrepancy Bouzigues had found was a small one, but it was considered significant. The French Commissariat à l'Énergie Atomique (C.E.A.) began an investigation to discover its cause.

The raw material for the isotopic analysis was not the uranium ore itself but uranium hexafluoride gas, the form in which uranium is processed for enrichment. One plausible explanation was that the gas had become contaminated with "tailings," the waste product of the enrichment cycle. The peculiar isotopic composition persisted, however, in the results of repeated analyses of other samples. The possibility of contamination was excluded from further consideration when the anomaly was traced back through the various stages in the manufacture of the uranium hexafluoride gas: through a processing plant in France to the Mounana mill near Franceville in southeastern Gabon. The ore had come from the nearby Oklo mine, operated by the Compagnie des Mines d'Uranium de Franceville (C.O.M.U.F.). Samples had been preserved from each batch of ore processed at the Mounana mill; they showed that shipments of uranium slightly depleted in U-235 had begun in 1970 and were still continuing. By mid-1972 the affected shipments from the Oklo mine involved ore that yielded about 700 tons of uranium; the deficiency of U-235 amounted to roughly 200 kilograms.

The ore body at Oklo had been defined by drilling sample cores on a closely spaced grid. Some of the cores were stored in France, and it was possible to analyze portions of them individually. Several were found to be strongly depleted in U-235. One core, removed from a region of the ore body that was then being mined, contained only .44 percent U-235.

The isotopic composition of uranium is thought to be a constant of the solar system in any one era. (It has been measured not only for many terrestrial ores but also for moon rocks and meteorites.) Chemical processes can make one region rich in uranium and leave another region poor; that is how the deposit at Oklo was formed. U-235 and U-238, however, are virtually indistinguishable chemically, so that any process that affects one of them must affect the other in the same way. Indeed, the difficulty of separating the isotopes is attested to by the size and complexity of uranium-enrichment plants such as those at Pierrelatte and at Oak Ridge, Tenn. There seemed to be no plausible mechanism in nature that might selectively remove one isotope to the extent observed in the depleted ore.

As the investigation continued, the possibility emerged that the missing U-235 had not been displaced but had simply been destroyed in situ. When the required tests were made, the explanation was suddenly obvious. Elements that are characteristic products of nuclear fission were abundant in the depleted vein, but they were almost absent elsewhere in the ore body. Their isotopic composition was quite unlike that of the natural elements, and it corresponded to the composition expected from fission. Three months after the investigation had begun in earnest the mystery was solved. Nature, not man, had constructed the world's first nuclear-fission reactor. Eventually six reactor zones were identified in the Oklo pit, four of them in strata that had not yet been mined.

The fissioning of a U-235 nucleus begins when it absorbs a neutron. The absorption of the neutron excites the nucleus and changes its shape so that about 85 percent of the time it becomes unstable and splits into two fragments and typically two or three neutrons. If at least one of the neutrons is absorbed by another fissionable nucleus and leads to fission, the reaction is

OPEN-PIT URANIUM MINE at Oklo in the Gabon Republic contains scattered pockets of ore that in Precambrian times achieved all the conditions necessary for a fission chain reaction. Six of these "reactor zones" have been identified; part of one, designated Reactor Zone 2, is visible at the left, at the base of the nearer wall of the pit, opposite a stack of core samples draped with a blue tarpaulin. The benches and rostrum, decorated with palm fronds, were set up on the floor of the pit mine for an international meeting convened a year ago to discuss scientific aspects of the reactor.

self-sustaining. The fragments produced by fission are almost always unequal in size—a pair might have masses of 99 and 133—and they are themselves unstable, or in other words radioactive. They decay with half-lives ranging from a few seconds to many years and yield a varied spectrum of daughter fission products. The final products include numerous stable isotopes of more than 30 elements.

The spectrum of fission products is so distinctive that it serves as an unmistakable sign that a chain reaction has taken place. In the Oklo uranium deposit the presence of these elements is convincing and quantitative evidence that a natural reactor once operated there. Both the absolute amounts of the elements and their isotopic composition can be explained only by their origin in fission.

Remarkably, at least half of the 30-odd fission-product elements have remained immobilized in the ore. These include the rare-earth elements lanthanum, cerium, praseodymium, neodymium, europium, samarium and gadolinium and also yttrium, which is not one of the rare earths but is chemically similar to them. Most or all of the zirconium, ruthenium, rhodium, palladium, niobium and silver remains, and so does some of the molybdenum and iodine. There are even remaining traces of the inert gases krypton and xenon.

Metals with a valence of one or two have a relatively high solubility in water and are readily leached away; hence the rubidium, cesium, strontium and barium made in the reactor, and probably the cadmium, have for the most part disappeared. On the other hand, there is no appreciable deficit of zirconium 90; this isotope of zirconium is produced by the decay of strontium 90, which has a half-life of about 30 years. One can conclude that little of the strontium was transported from the vicinity of the reactor

REACTOR ZONE 2 on the floor of the pit at Oklo is marked with pegs and strings, which define the lines along which sample cores were taken. The reactor was discovered in 1972, following the observation in a uranium-processing plant in France of an anomaly in the relative abundance of the two common isotopes of uranium, uranium 235 and uranium 238. The anomaly was traced to ores from the Oklo mine, which were found to be depleted in U-235, the more easily fissionable isotope. Cores from the reactor zones contain the uranium deficient in U-235; in addition they contain elements that are characteristic products of nuclear fission. Elsewhere in the mine uranium has a normal isotopic composition and the fission products are absent. On the rock face above Reactor Zone 2 the vein of uranium ore continues. The black regions are shale containing carbonaceous material; yellow regions are oxidized uranium formed on exposure to the atmosphere.

in that period. Lead, a final product of uranium decay, has migrated to some extent. Other fission products are made in quantities that are small compared with the amounts present naturally, and their fate has not yet been determined.

A particularly suitable element for an analysis of fission products is neodymium. It is not an abundant element and therefore the contribution from its natural background—the amount present from sources other than fission—is small. Moreover, neodymium has seven stable isotopes, ranging in mass from 142 to 150, but only six of them are manufactured as fission products. Fission fragments with a mass of 142 do not decay to neodymium but halt in a stable region of the periodic table at cerium 142. The neodymium 142 present in the depleted Oklo ores is therefore unrelated to fission, and from its amount and the known ratios of neodymium isotopes in natural deposits the background levels of the other six isotopes can be calculated. Subtracting these amounts from the total neodymium in the deposit gives the amount of neodymium made by fission.

For a complete isotopic analysis one more correction is required. Neodymium 143 and neodymium 145 both readily absorb neutrons; they are said to have a large neutron-capture cross section. Neutrons were abundant during the operation of the reactor and as a consequence many atoms of these fission products were altered by neutron capture after their formation. On absorbing a neutron neodymium 143 is converted into neodymium 144; neodymium 145 becomes neodymium 146. This effect perturbs the isotopic ratios, and it is particularly important for the mass-143 and mass-144 pair. Its magnitude can be calculated, however, and its influence can be fully accounted for. When these corrections for natural background level and for neutron capture are made, the abundance ratios of the neodymium isotopes correspond precisely to those measured experimentally in modern reactors of the appropriate type.

Similar analyses can be made of other elements present in the ore. An example is thorium. For every six atoms of U-235 that fission when they absorb a neutron, one simply captures the neutron and becomes an atom of U-236. This nucleus has a half-life of about 24 million years and decays by the emission of an alpha particle (a helium nucleus). When an alpha particle is emitted, the atomic mass of the nucleus is decreased by four and the atomic number is decreased by two. All the U-236 made in the natural reactor has by now decayed into thorium 232, a nearly stable isotope. Thorium is found in the reactor zones and is almost absent elsewhere. Similarly, U-238 in the reactor gives rise to a small quantity of U-237; this decays by a series of alpha-particle emissions to bismuth. At Oklo bismuth is present in quantity only in the reactor zones.

What conditions had to be met to achieve a nuclear reaction in the Oklo deposit? The list of requirements is hardly trivial. The basic requirement is that the flux of neutrons be sustained. On the average, the fissioning of a U-235 nucleus results in the prompt emission of 2.5 neutrons; one of them must be absorbed and must induce fission in another nucleus. The rest can be absorbed elsewhere or escape.

A chain reaction is possible in unenriched uranium containing .72 percent U-235 but only under rather special circumstances. A quantity of deuterium (the isotope of hydrogen with an atomic mass of two) must be present to serve as a "moderator." This is the system employed in the Canadian natural-uranium ("Candu") power reactors. Alternatively, the reactor could be constructed in a geometric lattice of uranium and a moderator with carefully specified dimensions. Neither a deuterium moderator nor a precisely assembled lattice is likely to be found in nature. About the best configuration that has a reasonable probability of being formed accidentally is a mass of relatively pure uranium oxide whose size is large compared with the distance a neutron travels before it is captured. This distance and hence the required size of the deposit are minimized by the presence of an effective moderator. It is reasonable to expect that such a moderator would be present in the form of water saturating the ore.

Given these best plausible conditions, no natural reactor could operate today with uranium containing .72 percent of the mass-235 isotope. The ratio of U-235 to U-238, however, has not been constant throughout the history of the earth. The half-life of U-235 is about 700 million years, that of U-238 about 4.5 billion years. Since the fissionable isotope is decaying faster, it must have been more abundant in the past. Indeed, from the decay rates it is possible to extrapolate into the past to determine the relative abundance of the two isotopes at any time back to the formation of the solar system. When the earth formed, natural uranium was about 17 percent U-235; the isotope has reached its present abundance of .72 percent through an exponential decline. The minimum abundance for the operation of a plausible natural reactor is 1 percent, which means such a reactor could have operated up until about 400 million years ago. In the much older Oklo deposit the relative abundance of U-235 was about 3 percent.

Apart from the isotopic ratio the concentration of uranium in the ore also affects the rate of the reaction; for a chain reaction it must average at least 10 percent. There are also constraints on the shape of the ore body. A sphere is the most efficient shape and requires the smallest quantity of uranium, but it is sufficient that the ore be deposited in seams at least half a meter thick. In a thinner deposit too many neutrons would escape. The reactor zones in the Oklo mine meet the requirements of uranium concentration and seam thickness.

An important requirement for the operation of the reactor is that the neutrons emitted by the fissioning nuclei be slowed down; this is the function of the moderator. The neutrons are emitted with high energy and therefore high velocity; in that state they are readily absorbed by U-238. Ideally the neutrons should be slowed to a "thermal" distribution of energies: their velocities should be those characteristic of random thermal motion at the temperature of the medium. This can be accomplished if each neutron collides with a great many nonabsorptive nuclei. Slow or thermal neutrons are much less likely to be absorbed by U-238, and hence they are more likely to survive to encounter a nucleus of U-235.

By far the best moderator available in a natural reactor is water. The moderation is provided primarily by the hydrogen atoms in the water, and for an ore that is two billion years old the optimum ratio is about 6 percent water by weight. The water of crystallization in a sedimentary ore such as that at Oklo should more than satisfy this requirement. In addition, at Oklo the medium was probably saturated with groundwater, which would have overmoderated the neutrons. If the ore became chain-reacting in this condition, the heat evolved would evaporate some of the water, so that optimum moderation would eventually be attained. For this reason the constraints on the amount of water initially present are not confining.

Finally, the reactor could not operate in the presence of large quantities of elements that strongly absorb neutrons (that is, elements that have a large neutron-capture cross section). Such elements are called neutron "poisons"; among the more potent ones are lithium, boron and many of the rare earths. There is no evidence that excessive amounts of such poisons were present in the ore before the reaction began.

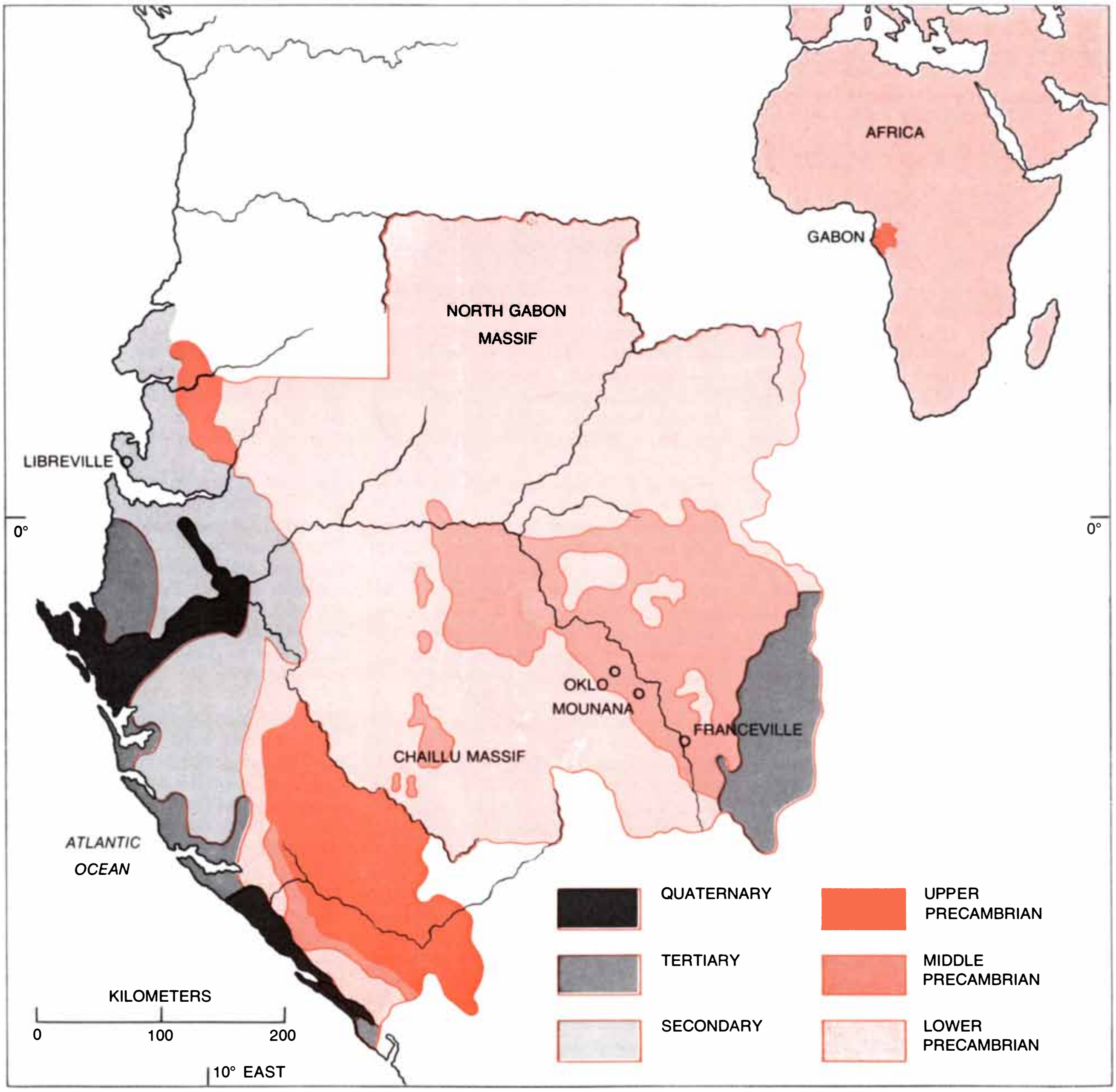
These circumstances varied during the course of the reaction, changing its rate and eventually stopping it entirely. They are responsible for controlling the power generated, both over short periods and over the entire history of the reactor. Together they functioned to limit the reactor to modest power levels.

In the long run the behavior of the Oklo reactor was determined by the continuous decrease in the relative abundance of U-235. Almost all this decrease was a direct consequence of the fission itself—the reactor was consuming its fuel—and it was therefore proportional to the total flux of neutrons in the reactor. As I have mentioned, the conversion of neodymium 143 to neodymium 144 (by neutron capture) is also proportional to the total neutron exposure in the ore, and the present isotopic ratios of neodymium therefore predict the total depletion in U-235. This calculation gives a result about 40 percent greater than the observed depletion; in other words, knowing the amount of U-235 present initially and the amount remaining, we find that the amount consumed was greater than the difference. Several factors could contribute to this discrepancy, including changes in the concentration or shape of the deposit during the reaction, and the accretion of ore afterward. The principal cause, however, is an additional nuclear reaction. As we have seen, U-238 readily captures high-energy neutrons; it is thereby converted to U-239. This nucleus decays by the emission of a beta particle (an electron). In beta decay atomic mass remains essentially unchanged but atomic number increases by one. Hence U-239 becomes neptunium 239. Through a second beta emission the neptunium becomes plutonium 239, which decays by alpha emission with a half-life of 24,400 years. The product of the last decay is U-235. Thus the absorption of fast neutrons by U-238 results in the eventual creation of additional U-235.

The operation of the reactor might also have been modified by a decrease in the quantity of neutron poisons present. As soon as a nucleus captures a neutron it is transformed, usually into a nucleus with a smaller neutron-capture cross section. In this way neutron poisons may have been "burned out" of the ore soon after the reactor began operating. If the initial amounts of elements such as lithium and boron were large enough, this effect could have been a major factor controlling the reactor.

Short-term control of the reaction was almost certainly dominated by the amount of water present. If the stratum containing the reactor was buried deep enough, the boiling point of the water may have been 300 degrees Celsius or higher. When the reactor reached that temperature, the water boiled away until the ratio of hydrogen to uranium reached a critical value where the reactor was undermoderated. The power generated by the reactor then leveled off and remained roughly constant, at a level just sufficient to compensate for the heat transferred to regions outside the reactor. (It is possible that the ore deposit was buried so deep that the pressure was great enough to prevent water from boiling, but that has not been demonstrated.)

The possibility of a sustained nuclear reaction in a natural uranium ore deposit was first considered more than 20 years ago. In 1953 George W. Wetherill of the University



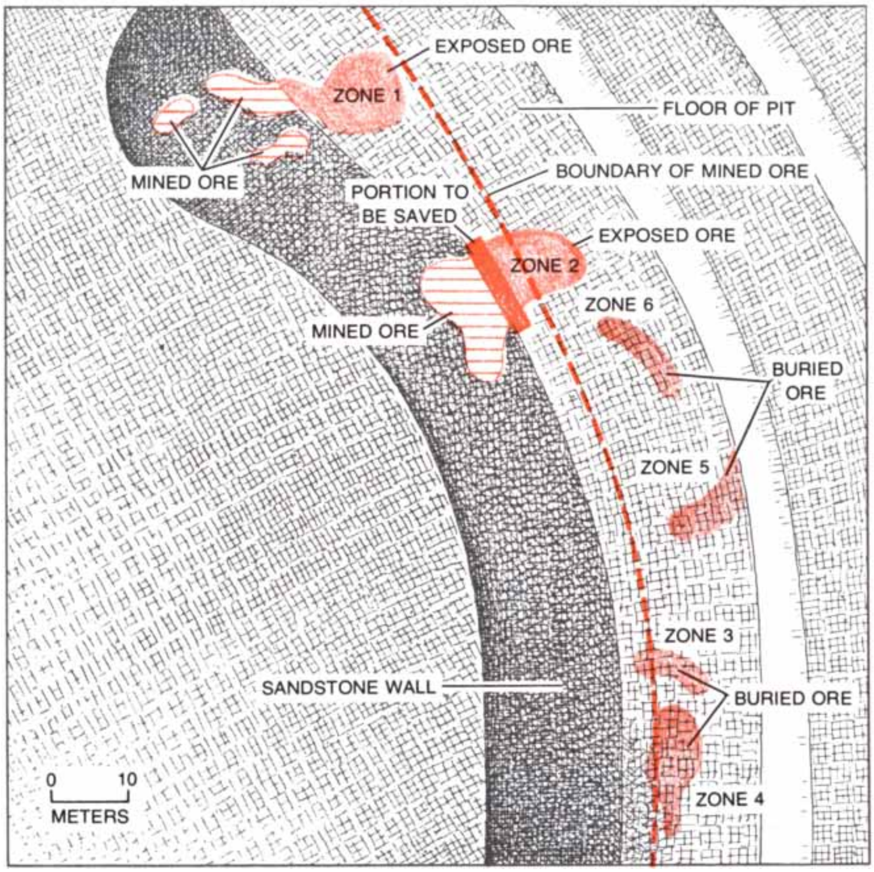
GEOLOGY of the region surrounding the Oklo mine is predominantly Precambrian, that is, older than about 600 million years. The mine itself is at the edge of a basin made up of sedimentary rock from the Middle Precambrian period; it borders an expanse of igneous rock from the Lower (earlier) Precambrian. The analysis of cores from the reactor zones suggests that the fission chain reaction began between 1.7 and 1.9 billion years ago. Independent evidence gives approximately the same age for the strata in which the reactor is embedded.

of California at Los Angeles and Mark G. Inghram of the University of Chicago stated with reference to a pitchblende deposit: "[Our] calculation shows that 10 percent of the neutrons produced are absorbed to produce fission. Thus the deposit is 25 percent of the way to becoming a pile [a reactor]. It is also interesting to extrapolate back 2,000 million years, when the uranium 235 abundance was [3 percent] instead of .7. Certainly such a deposit would be closer to being an operating pile." Three years later 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 describing the conditions at Oklo. In spite of such speculations the announcement of the Oklo reactor was received by American nuclear scientists with skepticism. Some of the world's best physicists had constructed the Stagg Field reactor with careful attention to mechanical detail, to the purity of the materials and to the geometry of the assembly. Could nature have achieved the same result so casually?

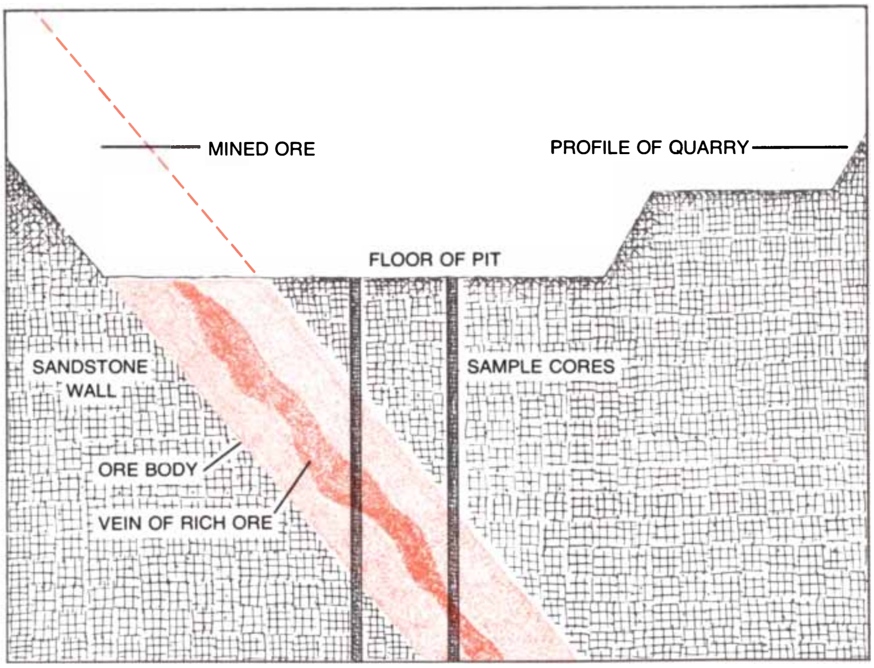
We now know that the answer is yes. Remember again the importance of the difference in date. In the two billion years between Oklo and Stagg Field the relative abundance of U-235 declined from 3 percent to .7 percent. A Precambrian physicist would have found it almost easy to build a nuclear reactor. In retrospect it seems inevitable that it happened accidentally.

Last June 70 investigators from 20 countries met in Libreville, the capital of Gabon, to discuss the "Oklo phenomenon." The meeting was sponsored by the International Atomic Energy Agency (IAEA), the French C.E.A. and the government of Gabon. Its business was to present and review analyses of the Oklo ores, carried out largely in French laboratories but also in the U.S., Britain, the U.S.S.R. and Australia.

The meeting opened with a spectacular expedition: a flight over the tropical rain forest to Franceville in southeastern Gabon and from there some kilometers by car to the floor of the open-pit mine at Oklo. From a rostrum decorated with palm fronds representatives of the sponsoring organizations and ministers of the Gabonese government welcomed us. We sat on benches at the edge of Reactor Zone 2, which was marked out by pegs and strings defining the sampling lines. J. P. Pfiffelmann, chief geologist of the C.O.M.U.F., took up a position at the edge of the reactor zone and lectured on the geology of the formations surrounding us. Behind him a sandstone wall slanted up at an angle of 45 degrees; there were ripple marks on its face attesting to its aquatic origin. The exposed reactor zone, about a meter wide and 10 meters long, extended along the floor of the pit near the sandstone wall. Immediately beyond the reactor zone rose a bench of unmined ore, displaying a continuation of the uranium vein. Water and atmospheric oxygen had converted traces of black, reduced uranium to a slurry of uranium oxide, which formed bright yellow



SIX REACTOR ZONES at Oklo were found in lenses of exceptionally rich ore. Parts of zones 1 and 2 had already been mined when the reactor was discovered, and their boundaries are therefore conjectural. Zones 3, 4, 5 and 6 have not yet been exposed; they lie below the present floor of the pit and were found by drilling cores. A portion of Zone 2 has been pinned to the rock face so that it will not be destroyed when the pit is deepened. In ore from the reactor zones the average concentration of uranium is from 20 to 30 percent, more than 50 times the concentration elsewhere. On the other hand, the uranium in the reactor zones is strongly depleted in U-235.

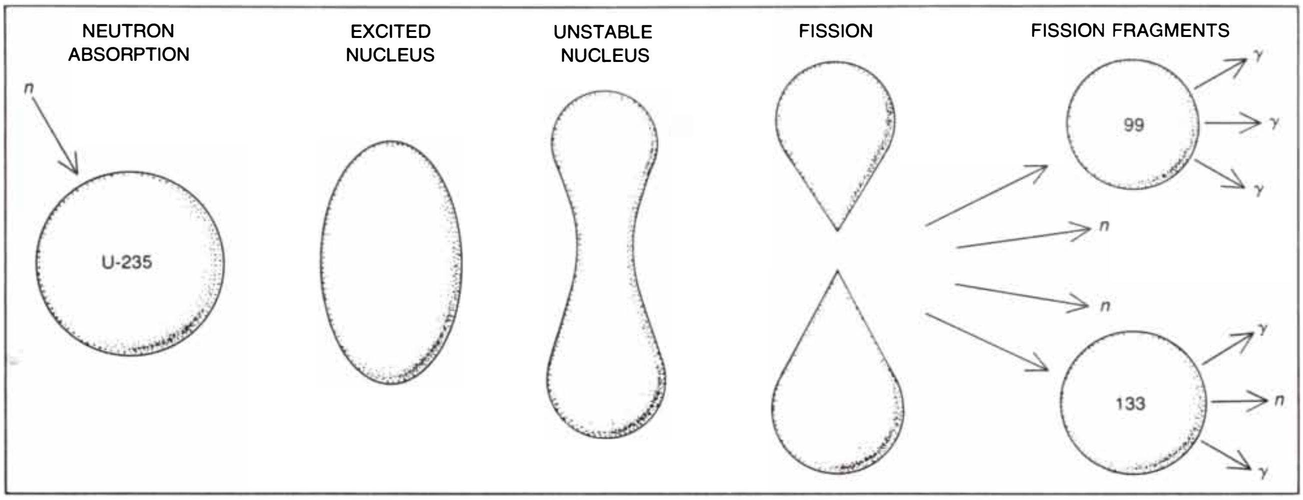


ORE BODY AT OKLO, shown in a schematic profile of the mine, lies above a sandstone wall with an average slope of about 45 degrees. The reactor zones are within the rich vein of ore, which is roughly a meter thick. The zones generally have a lenticular cross section. The extent of the ore body and the location of the reactor zones were revealed by removing core samples.

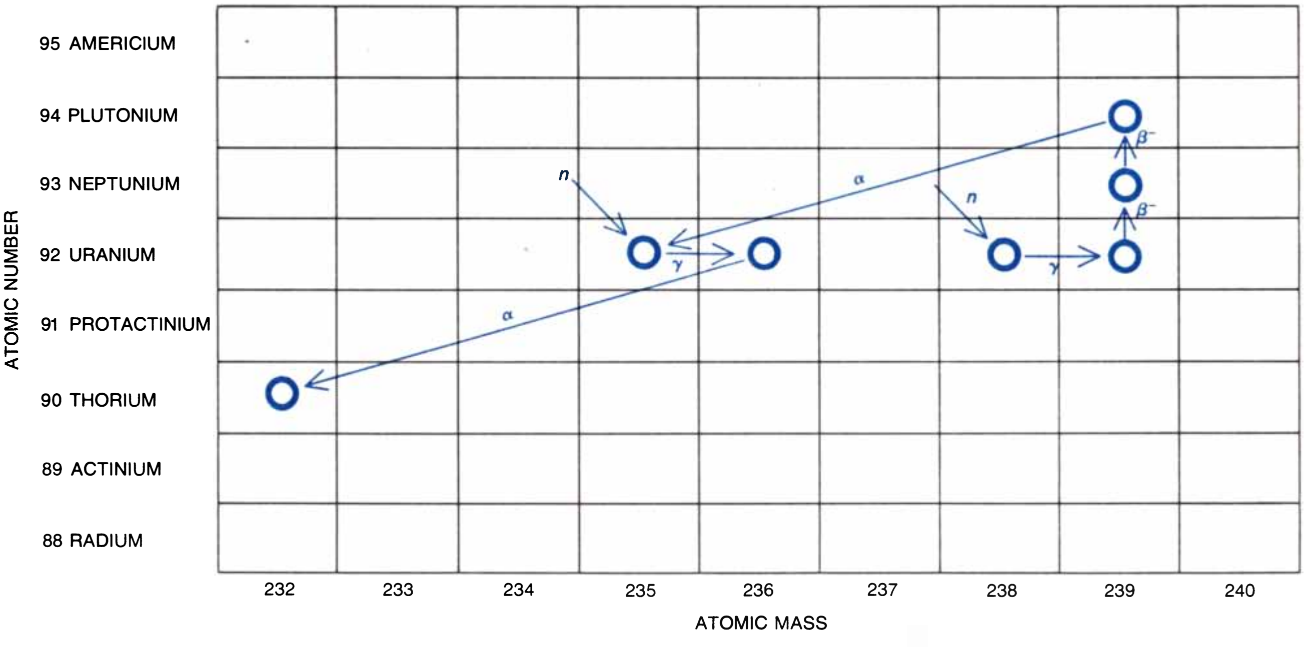
plaques on the buff-colored ore of sandstone and clay.

The landscape around us had once been a river delta. On an ancient African watershed crystalline igneous rocks were eroded by running streams, releasing minute quantities of heavy metals and their oxides. The heavier material accumulated in bottom sediments and pebble conglomerates, much like the placer deposits of gold and other noble metals found in streams today.

About two billion years ago the chemistry of the waters changed abruptly. The change was brought about primarily by the rise of blue-green algae, the first organisms capable of photosynthesis. As a result of their activity the oxygen content of the waters rose, and in some regions it became high enough to convert reduced uranium into more soluble oxidized compounds. The uranium was once again mobilized, and it remained in solution until it reached the river delta. There the bottom sediments were rich in organic ooze and oxygen-poor



FISSION OF A NUCLEUS of U-235 is induced by the absorption of a thermal, or slow, neutron (n), which excites the nucleus and deforms it. About 85 percent of the time the deformed nucleus becomes unstable and splits into two fragments of unequal size. The fission fragments shown have atomic mass numbers of 99 and 133; many other pairs of fragments are possible and each has a well-defined probability. The fragments are themselves unstable and are transformed by their subsequent decay, so that the total spectrum of fission products includes many isotopes of more than 30 elements. At the moment of fission high-energy photons, or gamma rays (γ), are emitted, as are a few neutrons. For a chain reaction to be sustained at least one neutron must be absorbed and must induce fission in another U-235 nucleus.



TRANSFORMATIONS OF HEAVY NUCLEI affect the rate and the nature of the fission chain reaction. Most U-235 nuclei that absorb a slow neutron become unstable and fission, but about 15 percent dissipate their energy of excitation by emitting gamma rays and simply retain the neutron. The addition of a neutron does not change the atomic number or the chemical identity of a nucleus, but it increases the atomic mass by one, so that the U-235 becomes U-236. This nucleus is unstable and decays by the emission of an alpha particle (α): a helium nucleus, made up of two protons and two neutrons. In alpha emission atomic number decreases by two and atomic mass decreases by four, so that U-236 becomes thorium 232. U-238 readily captures neutrons, particularly if they have a somewhat higher energy than thermal neutrons. It is thereby transformed to U-239, a short-lived species that decays by the emission of a beta particle (β^-): an electron. Beta decay can be considered as the conversion of a neutron into a proton; it does not alter atomic mass, but it increases atomic number by one. The U-239 thus decays quickly into neptunium 239, which in turn decays by a second beta emission into plutonium 239. The plutonium decays, with a half-life of 24,400 years, by alpha emission to yield U-235. Plutonium is readily fissionable, but in the Oklo deposits the rate of the chain reaction was low enough so that most of the plutonium decayed to U-235 before it could absorb a neutron and fission.

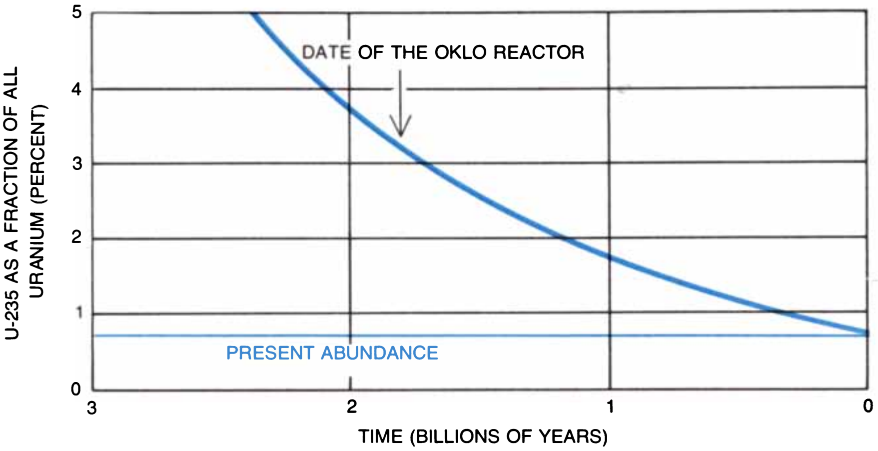
conditions again prevailed. The oxidized uranium was converted into the reduced form and compacted into the sandstone ore that now surrounded us. Subsequently the basement granite sank and the sedimentary layer deepened; then the granite to the west was uplifted, and it tilted the sedimentary overburden to its present angle. The ore layer, which averaged .5 percent uranium by weight, was fractured and water circulated through the new channels, creating pockets of rich ore that in places became almost pure uranium oxide. It was in these rich pockets that fission chain reactions began as soon as a critical mass of uranium had accumulated.

If uranium dissolves so readily in oxygenated water, how has the Oklo deposit survived almost two billion years? The sedimentary basin was apparently buried deep enough to protect the uranium ore from redissolution during most of its history. Only recently (within the past few million years) has the ore horizon approached the surface, where normal prospecting procedures could succeed in identifying it. Thus the series of special circumstances necessary to the discovery of the Oklo phenomenon includes not only those processes that led to the formation of a natural critical mass of uranium but also the unusual geophysical and geochemical conditions that preserved the ore body for almost half the lifetime of the planet and finally brought it to the surface.

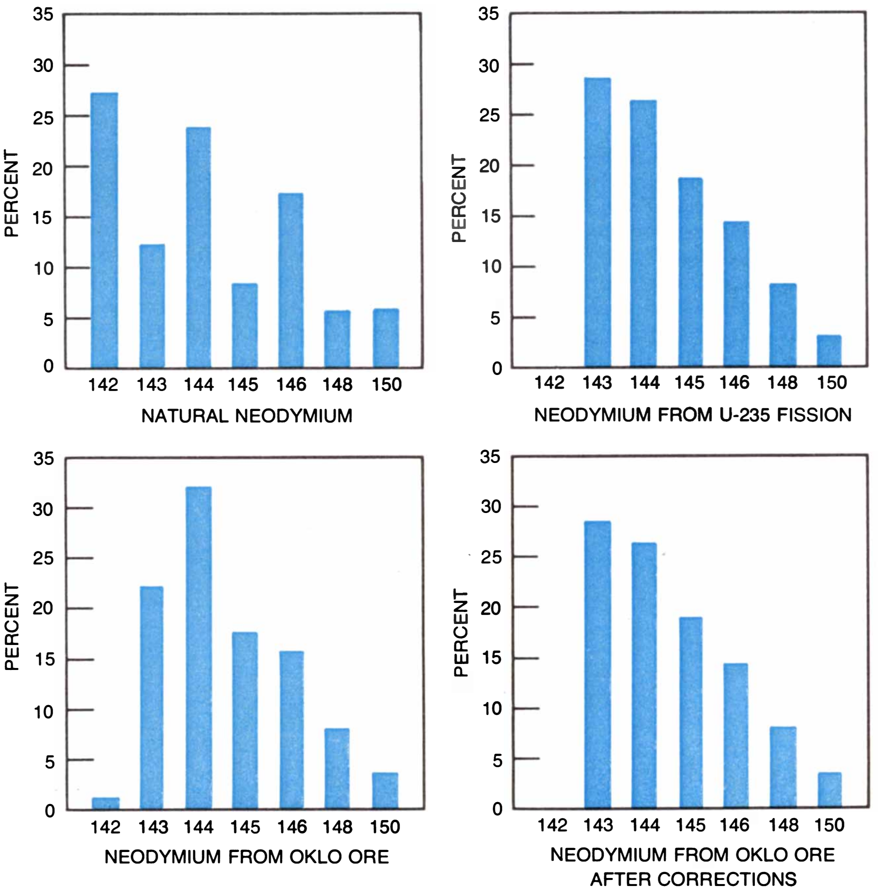
Back in Libreville, Roger Naudet, the director of the C.E.A.'s study of the Oklo phenomenon, reported some of that group's findings. They have estimated that the total energy released in the reactor zones was 15,000 megawatt-years, representing the consumption of six tons of U-235. That is approximately the energy produced by the reactor in a large nuclear power plant in four years.

Only about two-thirds of the fission events involved the U-235 that was originally present. Most of the remainder were in additional U-235 created by neutron capture in U-238 and the subsequent decay through plutonium 239. A few percent of the events were fissions of U-238 induced by fast neutrons, and another few percent were attributed to plutonium 239 that fissioned before it decayed. The modest contribution from plutonium 239 indicates that the reaction lasted much longer than the 24,400-year half-life of that isotope. The duration was on the order of hundreds of thousands of years.

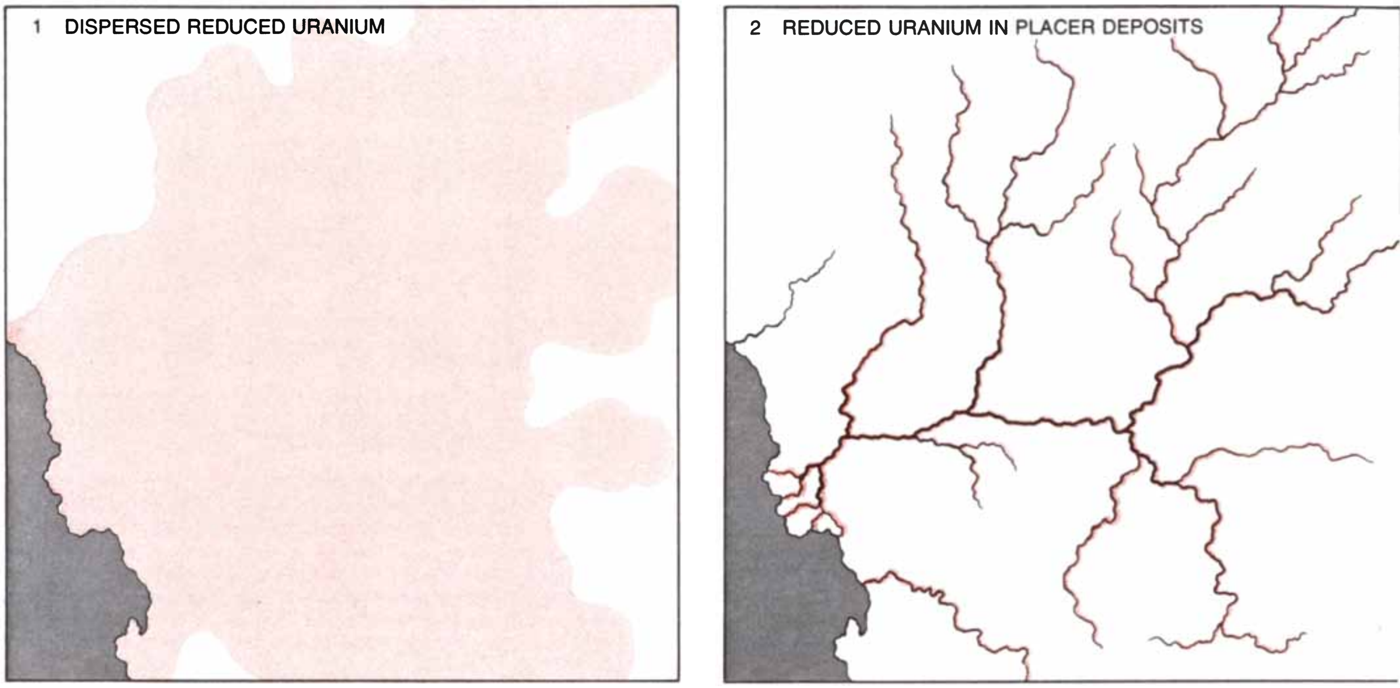
The same conclusion can be deduced from studies of heat transfer in the reactor. Since the reactor could not continue to operate if the temperature much exceeded the boiling point of water, the power level was probably limited to a few tens of kilowatts. This calculation is somewhat uncertain because it is not known how much water was actually flowing through channels in the ore body, but even if the power level was as high as 100 kilowatts, the duration of the reaction would still be 150,000 years. The total



ISOTOPIC COMPOSITION of uranium has changed during the history of the earth. Because U-235 decays about six times faster than U-238 the abundance of U-235 as a percentage of all uranium has declined. It is thought that when the earth formed some 4.6 billion years ago, uranium was about 25 percent U-235; today a commonly accepted value for the relative abundance of U-235 is .7202 percent. When the Oklo reactor was operating, the uranium in the ore contained about 3 percent U-235. The rate of radioactive decay is constant, and at any one moment the isotopic composition of uranium should be essentially the same everywhere in the solar system; it was for this reason the anomalous isotope ratios in ores from Oklo were investigated.



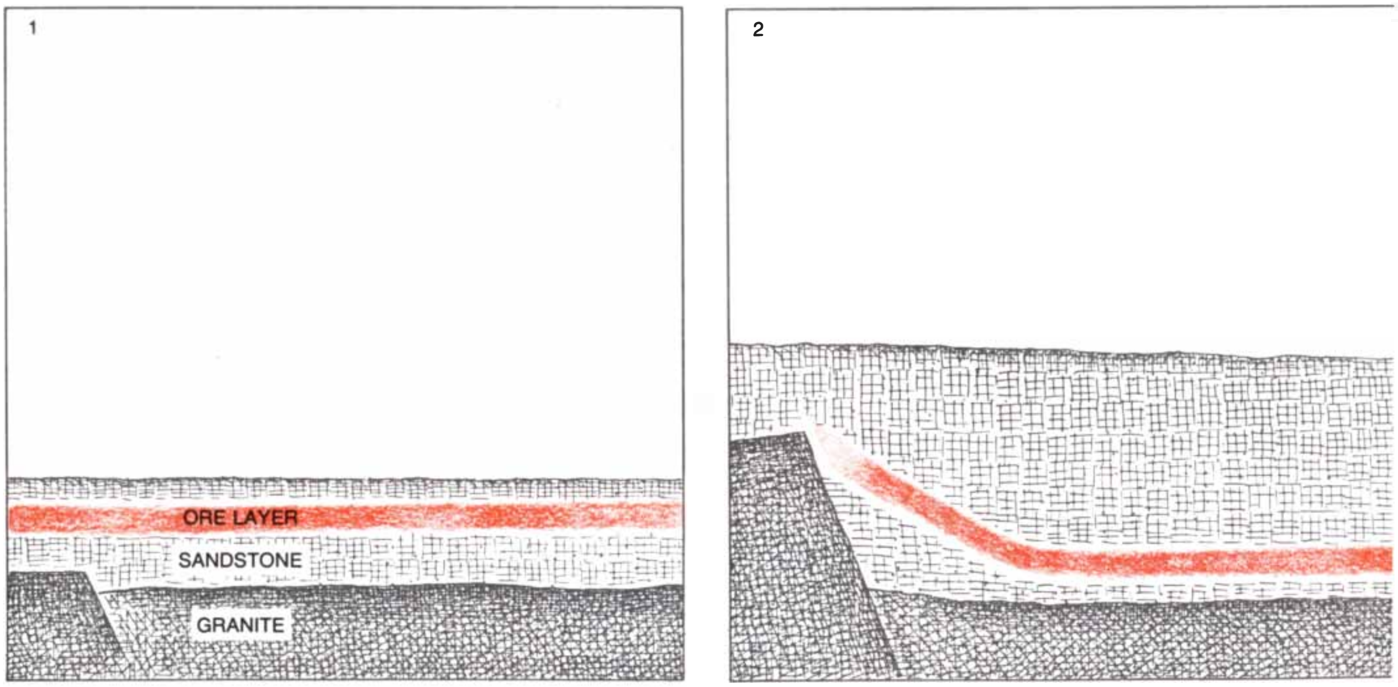
ISOTOPIC ANALYSIS of neodymium, a fission product, provides convincing evidence for a chain reaction at Oklo. Natural neodymium has seven stable isotopes, ranging in mass from 142 to 150, and their relative abundances are a characteristic of the element. Neodymium made by the fission of U-235 has a distinctively different composition; in particular, the isotope of mass 142 is entirely absent. At first the isotopic composition of neodymium from the Oklo reactor zones resembles neither of these distributions, but its origin becomes apparent when two corrections have been made. First, there is a little natural neodymium in the ore, and this must be subtracted. Second, neodymium 143 readily captures neutrons and is thereby converted into neodymium 144; similarly, but to a lesser extent, neodymium 145 is converted into neodymium 146. Since neutrons were abundant in the reactor, the ratios of these isotopes were substantially altered. When the presence of the natural element and neutron capture are taken into account, the composition of the neodymium in the Oklo ores closely matches that produced by fission.



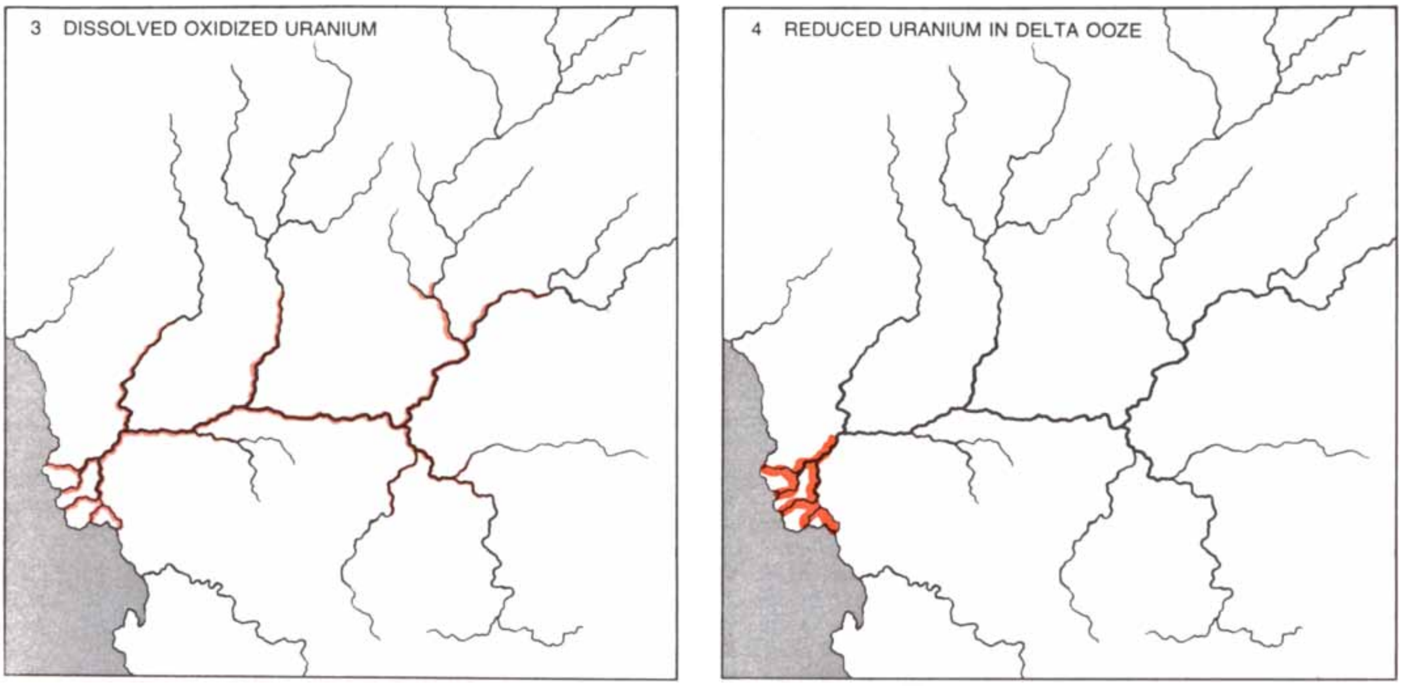
ACCRETION OF THE ORE DEPOSIT at Oklo involved the concentration of uranium derived from an entire watershed. The uranium (*color*) was originally dispersed in igneous rocks over a large area (*1*). As the rocks were eroded by weathering, the uranium accumulated in streambeds in a relatively reduced state (*2*). It may have formed placer deposits like those in which gold and low-grade, Precambrian uranium ores are found today. Some two billion years ago a biological development profoundly altered the disposition of the uranium:

time elapsed from the initiation of the chain reaction to the final shutdown was probably even greater, because the reactor did not necessarily operate continuously.

The unusual stability of the ore deposit was confirmed by 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 U-235 depletion and the total neutron flux over the life of the reactor, as calculated from isotopic ratios in neodymium. Although some puzzling anomalies were found at the borders, there was little migration of uranium inside the zones. Distinct excursions in both the neutron flux and the U-235 depletion were observed in the vicinity of faults in the ore body. When the reactor was functioning, these faults were most likely water-filled channels that trapped neutrons and increased their local density. Such neutron traps are a feature of some modern research reactors.



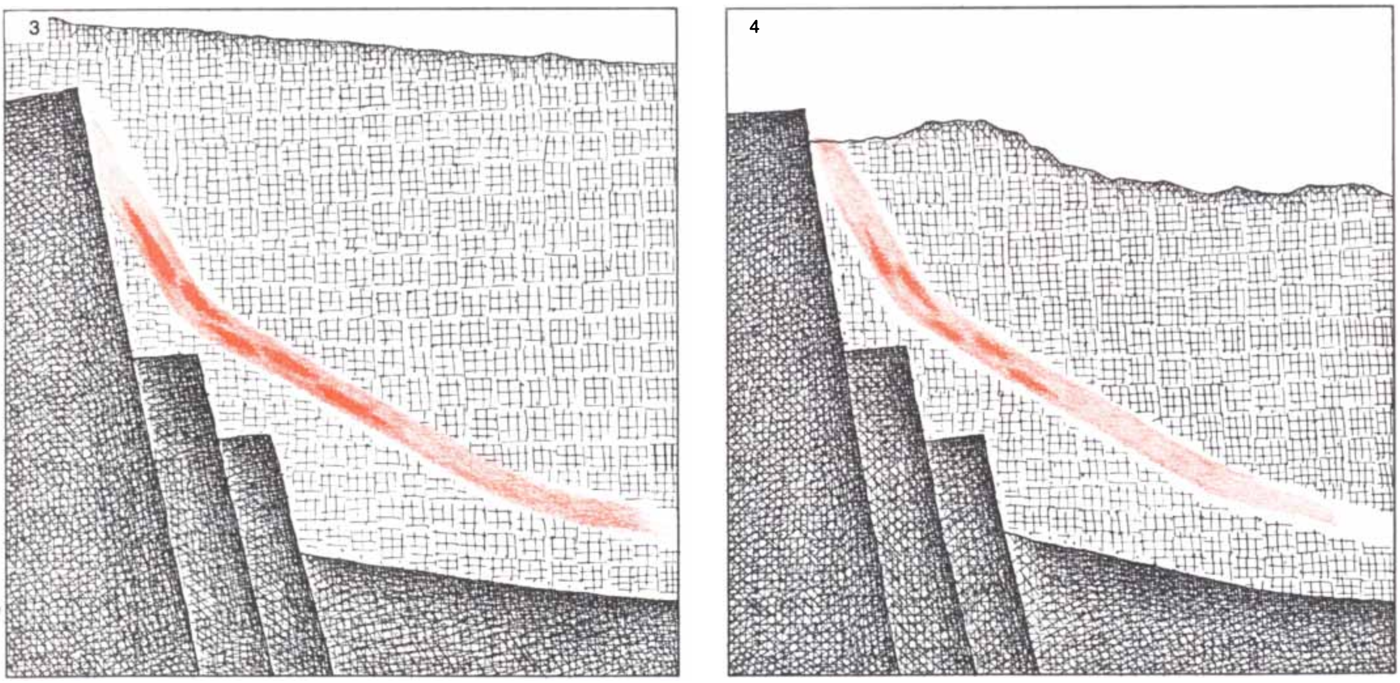
GEOLOGICAL TRANSFORMATION of the ore body created the conditions necessary for the operation of the natural reactor and preserved the evidence of the reaction for almost half the life of the planet. After the ore had been deposited in the river delta it was compacted along with other sediments into a layer of sandstone (*1*) overlying basement rock of granite. Gradually the sedimentary overburden deepened and the ore layer sank (*2*), while the granite to the west of the deposit began to be uplifted. As the uplift continued, the vein of ore



the proliferation of photosynthetic algae increased the concentration of oxygen in the water. Highly oxidized uranium is more soluble than the reduced form, and the uranium was therefore dissolved and carried downstream (*3*). It remained in solution until it reached the delta of the river system, where sediments rich in organic ooze again created a condition of oxygen deficiency. In these sediments the uranium was returned to the reduced form and was precipitated out of the solution (*4*). In the delta the ore was covered by later sediments.

A group of investigators from the University of Paris employed an ion probe to study the distribution of isotopes within individual grains of ore. The partitioning of several fission products between grains of uraninite and clay was noted, but the rare earths and uranium were found in the same grains. More remarkable was the complete confinement of U-235 and U-238 to the same grains. Almost half of the residual U-235 in the reactor zones is a daughter product of plutonium 239. The fact that no regions were enriched in this daughter product indicates that the precursor plutonium was completely immobilized for times comparable to its half-life of 24,400 years. The possible relevance of this observation to proposals for the long-term storage of nuclear wastes was pointed out by investigators from both France and the U.S.

Finally, both French and American participants presented estimates of the age of the reactor based on analyses of the ore samples. The estimates assume that the



was tilted to its present average slope of about 45 degrees. The tilting caused numerous fractures, so that water could percolate through the ore, creating small pockets of very concentrated ore (*3*). In these pockets the chain reaction began as soon as enough uranium had accumulated. For most of its history the ore was probably buried deep under other sedimentary rock, where it was protected from dissolution and dispersal by groundwater. Only much later, in the past few million years, did it approach the surface where it could be detected (*4*).

abundance of uranium and neodymium in the ores has not been extensively altered since the reactor stopped functioning. The absolute abundances of those elements and their isotopic compositions then give a date for the initiation of the reaction. The best fit to the American data is an age of from 1.7 to 1.9 billion years, which is in good agreement with independent estimates of the age of the host geological formation.

Outside the conference, discussion was given over to freewheeling speculation. Did prokaryotes (living cells without nuclei) evolve into eukaryotes (cells with nuclei) as early as 1.8 billion years ago? Was it possible that Africa was not only the cradle of man but also the birthplace of the cells that led to all the higher forms of life? There was general agreement that the plot should be worked into a science-fiction story but that without considerable embellishment the motion-picture rights would be worthless. It would have been far better to introduce a spaceship from another planet that had dumped its used reactors at the site, replenished its fuel supply and departed.

For the three years from the discovery of the depleted uranium to the symposium in Libreville, the C.O.M.U.F. had suspended mining of the very rich ores in the reactor zones. Now it was time to deepen the pit and mine the uranium in the protected regions. Presumably the highly depleted ores would be processed separately. It was proposed to save a portion of Reactor Zone 2 by walling it off and pinning it to the 45-degree sandstone face. The ore will be suspended above the new floor of the pit, an elevated monument to the first discovery of a natural fission reactor.

C there have been other natural reactors of the Oklo type. The final stage of the process that formed the Oklo lode could have been a very common one in Precambrian times: as the oxygen content of the atmosphere rose, reduced uranium in scattered deposits would have become mobile, and it could then have been reconcentrated in richer ore deposits wherever a reducing environment was encountered. Rich uranium ore deposits have been found in other geological formations of approximately the same age, not only in Africa but also in other parts of the world, particularly in Canada and northern Australia. None of these deposits has yet been identified as a reactor site. It is entirely possible that chain-reacting ore lodes formed in these areas and have since disappeared. They may have been buried under younger sediments, where they are unlikely to be discovered, or they may have been dispersed as a result of geophysical instabilities or geochemical mobility.

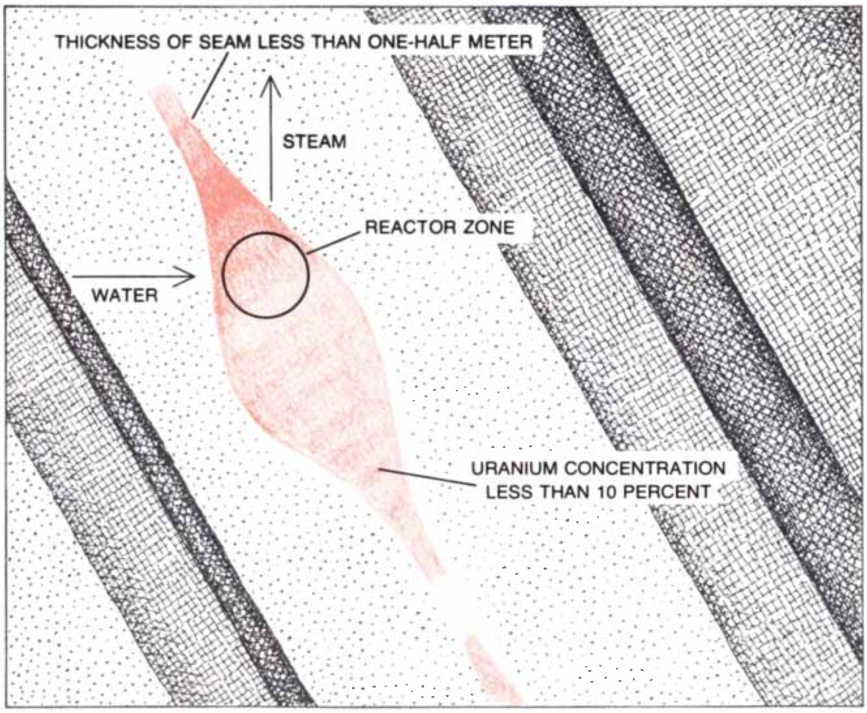
What is the probability of such dispersion? Apart from its obvious scientific interest, the answer to this question has possible economic and technological consequences. It has a direct bearing on the problem of the long-term storage of nuclear wastes. In this regard the stability of the Oklo deposit is plainly encouraging. It contains all the chemical elements of interest and demonstrates that, at least in this one environment, they remained in place for an enormously long time.

At the Libreville symposium Ray D. Walton, Jr., of the U.S. Energy Research and Development Administration (ERDA) presented the results of a preliminary American attempt to determine the significance of the Oklo phenomenon for radioactive-waste storage. He suggested that at the time of the reaction the principal radioactive products that were released into the environment in measurable amounts were krypton 85 and possibly cesium 137 and some strontium 90. Plutonium, the most worrisome reactor product, was efficiently confined.

The adsorptive properties of shales and clays and the extreme stability of many elements bound in such matrixes were known before the discovery of Oklo, and suggestions for the storage of reactor products in such formations have been under investigation for many years. It is obviously difficult to devise an experiment that would evaluate the stability of an adsorbed metal ion for a million years or more under field conditions. Since Oklo resembles such an experiment, the data have been examined with great care to determine what part of them might be relevant to the problem of waste storage.

The possibility that other ore deposits once supported chain reactions also affects the producers and buyers of uranium. It should be of interest as well to those regulatory agencies whose responsibility it is to account for all fissionable materials; they are expected to notice mysterious shortages of a few hundred kilograms of U-235.

In this regard it is interesting to note that not all natural reactors would necessarily give rise to depleted ores. As the isotopic abundance of U-235 diminished over geologic time, the conditions necessary for reactor operation became more restrictive, but at the same time the nature of the chain reaction was subtly altered. In particular, the relative importance of neutron capture in U-238 increased, since that isotope came to form a progressively larger fraction of the total uranium. If a natural reactor was able to form as late as 800 million years ago, when the relative abundance of U-235 was about 1 percent, it might actually have become a breeder reactor. The U-235 consumed in the reaction would have been more than replaced by new U-235 created by the decay of plutonium. The pitchblende deposits in the former Belgian Congo were



RATE OF A CHAIN REACTION is determined by the flux of neutrons, and at Oklo this was influenced by several factors acting in combination. The reaction could begin only in regions of the ore body where the concentration of uranium was greater than about 10 percent. The shape of the deposit was also important: thin seams allow too many neutrons to escape. Elements that act as neutron "poisons," strongly absorbing neutrons, can also prevent a reactor from functioning, but apparently none of these elements was abundant at Oklo when the chain reaction began. A final requirement is a moderator, a substance that slows neutrons so that they are not too readily captured by U-238. At Oklo the moderator was water, and the amount present was presumably the most important factor, over relatively brief periods, controlling the power level of the reactor. Any increase in power would raise the temperature and boil off water, slowing the reaction. Through this mechanism power output was maintained at a modest level. In the long term the reaction was controlled and finally stopped by the diminishing abundance of U-235.

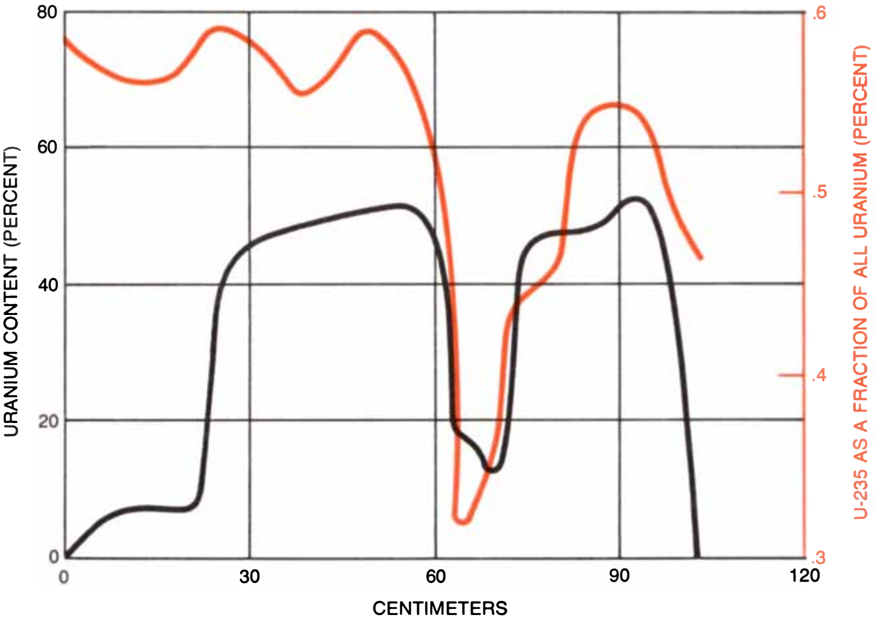
of the appropriate age. They have now been mined out, but precise isotopic analyses are available for a few samples of the ore. It is provocative that these samples appear to be slightly enriched in U-235.

The number of economically significant deposits of any mineral is determined by the difference between the rate at which the deposits form and the rate at which they are dispersed. The second term in this equation—the one dealing with rates of dispersion—has largely been overlooked. Its importance to waste storage is apparent. Ultimately it may also determine the probability of discovering other natural reactors.

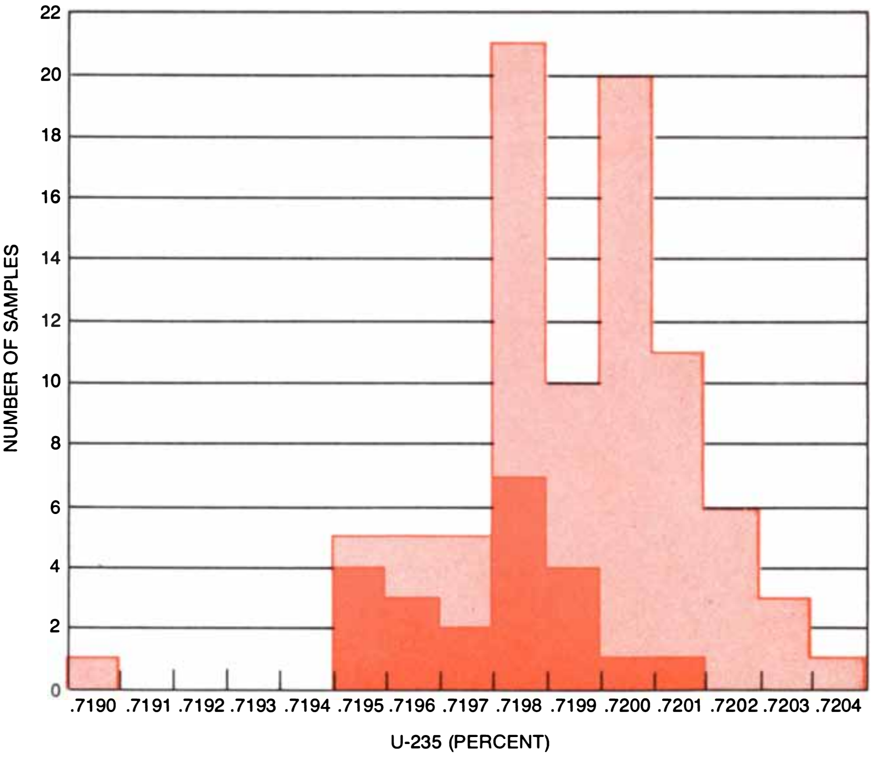
The statement that the isotopic composition of uranium is constant throughout the solar system is based mainly on analyses accurate to about .1 percent. At that level of precision the ratio is indeed constant. The more refined mass spectrometry possible at uranium-enrichment plants gives results accurate to about .01 percent, and at this level variations in the ratio have been discovered. In particular, sedimentary deposits from the Colorado Plateau in the U.S. are depleted in U-235 by about .03 percent. These variations have generally been attributed to chemical differentiation of the isotopes in sandstone rocks. Another hypothesis is possible: the depletion might result from the operation of a reactor of the Oklo type in the Colorado Plateau, the remains of which have since been dispersed throughout the region. The total uranium reserves of the plateau are on the order of 400,000 tons; to explain the observed depletion requires a reactor that caused a net loss of five tons of U-235. We do not yet have enough information to decide which of these explanations is the more likely to be correct.

If dispersion was not the general fate of natural reactors, we can hope to find their dormant remains by surveying rich uranium deposits more than a billion years old. In principle it would even be possible to recognize a reactor zone in which most of the uranium had been dissolved and washed away, since the less mobile rare earths would probably remain in place. Any ore pocket containing rare earths, yttrium, zirconium, niobium, ruthenium and rhodium, all in abundances greater than .01 percent, could be considered a candidate. The origin of these elements in a fission reaction could be demonstrated by isotopic analysis.

On balance, the prospects for finding additional reactors seem good. If Oklo had been a unique event in the history of the earth, the probability of our having discovered it must approach zero. Considering the almost accidental way its existence was revealed, one is tempted to conclude that similar reactors have already been mined out without being noticed. In 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. In any case one message is already clear. In the design of fission reactors man was not an innovator but an unwitting imitator of nature.



STABILITY OF THE OKLO DEPOSIT during the 1.8 billion years since the reactor operated is suggested by a detailed examination of the ores. In some regions samples were analyzed every 2.5 centimeters. (The horizontal axis gives distance along the sampling line.) Generally the concentration of uranium in the ore was closely correlated with the depletion in U-235, as at the extreme left. The sharp dip in uranium concentration is caused by a crack in the ore sample. When the reactor was operating, the crack was apparently filled with water and served as a "neutron trap" that slowed neutrons and greatly increased the efficiency of the reaction in its vicinity. That increased efficiency is reflected in the corresponding decline in the abundance of U-235. The correlation of the two curves suggests there has been little migration of uranium.



ABUNDANCE OF U-235 in 88 samples of uranium ore shows a clearly bimodal distribution. The peak at .7200 percent represents ores mined mainly in Canada, Europe and Australia. The peak at .7198 percent is produced by ores taken from the Colorado Plateau region of the U.S. One group of these ores, mined in New Mexico, is shown separately (*dark color*); it plainly suggests that uranium from the Colorado Plateau has a U-235 content smaller than the world average. One possible explanation of this small discrepancy is that the isotopes were separated by some chemical process in the ore-bearing rocks. Another explanation is that a natural reactor once operated in the region and that its depleted ore has been dispersed throughout the plateau.