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AUTHOR(S): George A. Cowan

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los alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87545

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The Retention of Reactor Products in the Oklo Natural Fission Reactor

George A. Cowan

Within the past few years it has been clearly demonstrated that a series of natural fission reactors operated nearly 2 x 10^9 years ago in rich uranium ore deposits located in the Oklo mine in the southeast part of the Republic of Gabon, West Africa.(1)(2)(3)(4) Four separate reactor zones have been identified, all in the same part of the open-pit mine. They operated over a period of [illegible] 600,000 years. During this time 15000 MW-years of energy w[illegible]d at an average level of 25 KW. Approximately six tons of U [illegible] ere consumed, creating an equivalent amount of fission products deposited directly in the ground. Nearly three tons of Pu-239 were created by neutron capture on U-238 which eventually decayed quantitatively to U-235. Approximately one thousand tons of uranium were involved in the reaction zones.

These zones were highly enriched lenses of uranium ore, averaging over 20% in uranium content within an ore body of about 0.5% average concentration. At the time of criticality the initial atomic ratio of the fissionable U-235 isotope was 3%. The rich lenses were highly irregular in form but tended to be tabular, about one meter thick, and several hundred square meters in area. Following initial deposition in fluvial sediments and subsidence, the formation was tilted about 45° and faulted. The rich ore zones then formed in structural traps and, presumably, achieved criticality as soon as a high enough uranium concentration was reached in the presence of the transporting and neutron-moderating medium, water.

The fact that nature and not man had designed the first terrestrial fission reactor was, at least initially, surprising. Perhaps even more surprising was the remarkable state of preservation of the reactor. Quantitative and semi-quantitative analyses for the various reactor products showed that the majority of them had been retained. It was also possible to demonstrate that the reactor zones had preserved their initial geometries, i.e., the ores had not undergone physical or chemical mixing. The arguments which support these conclusions are presented in detail in the Proceedings of the Oklo Symposium and only the conclusions are summarized here. The degree of retention of the principal reactor products is summarized in Table I.$^{(5)}$

From microscopic and heavy-ion stimulated ion-emission studies$^{(6)(7)}$ a great deal has been learned of the composition of the ore and the location of the reactor products. Although the uranium outside the reactor zones in low concentration ores is largely pitchblende, the mineral in the highly enriched reaction zones is generally uraninite in the form of grains, of the order of 40-200 μm in diameter, distributed in a phyllitic gangue (chlorite plus illite). These analyses show that there is always associated with the uraninite grains some calcium, fluorine, sodium, titanium, rare earth fission products, zirconium, ruthenium, palladium, yttrium, lead, and thorium. In the gangue one finds boron, lithium, potassium, much of the radiogenic lead, barium, strontium, and some of the fission product zirconium and ruthenium. The ratio of U-235 to U-238 appears to be constant in the uraninite grains. The fact that none of the U-235 is outside the grains leads to the conclusion that the large amounts of Pu-239 formed by neutron capture on U-238 decayed within the uraninite grains to U-235.

Additional analytical results are available from samples taken as closely as every 2.5 cm in traverses across the reactor zones.$^{(8)(9)}$ These show generally consistent agreement between depletions in the U-235 abundance, neutron fluences evaluated from second-order neutron capture effects in the rare earths, absolute rare earth fission product concentrations, and uranium concentrations. Although in some traverses there is evidence that a part of the uranium has been lost from regions near the outside of the reactor zones (or that rare earth fission products have been gained), in general the ratio of neodymium to uranium has been quantitatively preserved even on a scale of centimeters. These observations are consistent with the microscopic observations that the neodymium has been quantitatively retained within the original uraninite grains. Taken together, they support the conclusion that there has been no extensive geochemical or geophysical mixing of the uraninite grains.

The apparent stability of the grains of uraninite demonstrated by these observations seems particularly remarkable because even rather small changes in pH and Eh, especially if accompanied by an increase in dissolved carbonates, can lead to rapid oxidation and dissolution in the form of a uranyl double carbonate or, at least, to the formation of secondary, oxidized minerals of uranium.(10) The observed stability can not be attributed to the absence of exposure of the ore to moving ground-water. The reactor ore was deposited by water and the neutron reaction required water to achieve criticality. The most soluble fission products have been almost quantitatively removed or replaced. Age measurements show that two-thirds of the total radiogenic lead has been lost which means that exposure to water occurred over a large part of the life of the ore-body or reoccurred in relatively recent times. Accordingly, it appears evident that water was present but must have been buffered within a narrow pH and Eh range.

Self-buffering of ground water, even at rather shallow depths in a reasonably large volume of a given soil type, is probably the rule rather than the exception.(11) Thus, the Oklo observations support the conclusions that, despite the extreme vulnerability of a mineral such as uraninite to geochemical attack and dissolution, it can retain its integrity and composition to a remarkable degree over billions of years while immersed in natural ground-water. Given the variation in measurements within the reactor zones, we estimate that the transport of uranium out of the principal reactor zones averaged no more than 20% in the most affected regions. This means that the fractional loss of uranium was no more than 10^{-10} per year. The hydrological and geological conditions which were responsible for this degree of stability are probably not uncommon.

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TABLE 1. Migration of fission products and actinides in the "Zone 2" reactor at the Oklo mine

Element	Migration Behavior	References
Kr	Measurable traces, 0.01 to 1% remaining	12,13,15
Rb	Measurable traces, less than 1% remaining	13,14,16,17
Sr	Measurable traces, less than 10% remaining	13,16,17
^{90}Sr (Now ^{90}Zr)	Small migration, most decayed in place	15,16
Zr	Redistributed, mostly in place	7,15,16
Nb	Mostly retained	13,15
Mo	∿ 90% missing	13,15,16
^{99}Tc (Now ^{99}Ru)	Redistributed, migrated as Tc	15,16
Ru	Large fraction retained, redistributed	13,15,16,19
Pd, Ag	Mostly retained	13,15
Cd	∿ 90% missing	13,18
Te	Mostly retained	13,15
I	Mostly gone	13,15
Xe	Measurable traces, 0.01 to 1% remaining	13,15,16
Cs	Measurable traces (as Ba), mostly missing	13,15,17
Ba	Obscured by natural but mostly gone	13,15,16
Ce, Nd, Sm, Gd	Very little migration	9,13,15,16,19,22
Pb	Redistributed, ∿ 2/3 missing from core	7,13,20,21
^{232}Th(^{236}U, ^{240}Pu)	Mostly retained	9,15
^{209}Bi(^{237}Np, ^{241}Pu)	Mostly retained	9,15
^{235}U(^{239}Pu)	No ^{239}Pu separation from ^{238}U	7,13
U	Probably no major migration some redistribution, (see text)	9,13,15,22