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From routine sample measurements in CEA to the Oklo phenomenon

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In 1972, before shipping natural uranium to the USSR for enrichment operations in ^{235}U, the analysts at Pierrelatte plant noted a slight deficit in ^{235}U: 0.7171 instead of 0.7202. The Direction des Productions of the CEA launched a vast campaign of analyses for the different mines exploited, at all stages of the elaboration of uranium: analyses on the ore, then on the yellow cake, on the uranium oxides issued of the yellow cake transformation, then on the UF_4 and on the UF_6.

For this analysis campaign, the Direction des Productions relied on the analytical laboratory of the Pierrelatte plant and on the Central Analytical Laboratory of the CEA, managed by Michele Neuilly, where I was in charge of analyses by mass spectrometry.

The numerous chemical and isotopic analyses of uranium lead to Gabon and more precisely to the Oklo mine. Indeed, the closer one gets to this site, the higher the uranium content and the higher the ^{235}U depletion.

At a meeting held at the CEA headquarters, it was decided that the laboratories at Pierrelatte and Cadarache would continue to carry out the analysis campaign, and that the laboratory at Cadarache would analyze the samples from Oklo.

COMUF, the company that operated the mines in Gabon, sent two samples of magnesium uranate and two samples of ore to Cadarache. After the analyses of the U and ^{235}U content, there were enough samples left that I decided to analyze them on a spark mass spectrometer, which provides a panoramic analysis of all the isotopes present in the analyzed product. I discovered on the photo plate of the mass spectrometer, isotopic anomalies, in particular the absence of ^{149}Sm, whereas ^{147}Sm was present. The next step was isotopic analyses of some elements, including neodymium and samarium after chemical separation. They revealed that the isotopic composition of these two elements was completely different from that of the natural elements. The results of these analyses were transmitted to the CEA Directorate which sent them to the neutron specialists at Saclay. Their conclusion was as follows, the isotopic compositions of the Oklo ores are identical to those of uranium having undergone a chain reaction of fission.
It was the first time we discovered a natural fission reactor.

The discovery of ^{235}U depletion and the chain fission reaction in the Oklo ore will be the subject of two communications to the Academy of Sciences.