The Francevillian (Lower Proterozoic) Uranium Ore Deposits of Gabon

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Abstract

The lower Proterozoic uranium ore deposits in the Franceville basin (Gabon) are the oldest high-grade uranium accumulations known. They are unique in that they contain evidence for natural nuclear fission reactors.

Sedimentologic, tectonic, petrographic, and geochemical studies have been performed in order to reconstruct the geologic conditions in which uranium mineralization took place. Uranium deposits are located in deltaic sediments overlying fluviatile deposits of coarse sandstone and conglomerates which are the source rocks for uranium. Deltaic sediments are overlain by marine black shales (the FB formation). Petrographic observations, electron microscope studies, and geochemical and carbon isotope data indicate that these FB black shales are source rocks for petroleum trapped in the uranium deposits.

Tectonic studies show that all the uranium deposits are in tectonic structures that served as traps for both petroleum and uranium. Uranium mineralization occurs in this setting when an oxidized uranium-bearing fluid has mixed with a reduced petroleum-bearing fluid. The uranium ores are affected by hydrofracturing which forms a good pathway for the oxidized uranium-bearing fluids and the reduced fluids. Hydrofracturing may be initiated by overpressured fluids coming from undercompacted zones in the FB black shales.

Introduction

THE Francevillian uranium ore deposits at Oklo are well known because they host the only natural fission reactors we know in the world at the present time. These are also the oldest uranium ore deposits in a sedimentary environment having high uranium contents. For these reasons it seemed worthwhile to investigate the geologic conditions, mainly sedimentologic, tectonic, and geochemical, in which such mineralization took place. This paper presents the geologic history and setting of the uranium deposits and their host rocks. A second paper will present the natural fission reactors in more detail.

Geologic Setting

The Francevillian Series is a 1,000- to 4,000-mthick sequence of clastic and volcano-sedimentary sediments of lower Proterozoic age, 2,150 Ma (Bonhomme et al., 1982). The series is widely distributed in southeastern Gabon and rests on the Archean (2,700 Ma) basement rocks of the North Gabon and Chaillu massifs (Fig. 1).

The Francevillian basin consists mainly of two different domains: the mobile zone of Ogooué with a metamorphosed series and the Franceville domain consisting of nonmetamorphosed sediments.

The mobile zone of Ogooué occurs in the western part of the Francevillian basin and has not been well studied to date because the density of the forests makes penetration very difficult. In this area, the Francevillian Series sediments and the underlying Archean basement rocks are affected by three major phases of tectonometamorphic events (Gauthier-Lafaye, 1986). The first two events resulted in mediumgrade pressure metamorphism as defined by Winkler (1976) along a north-south-trending axis in the central part of the mobile zone, the intensity of which decreases toward the eastern and western edges. These two events resulted in isoclinal folds with schistosity parallel to the axial plane and disthene and sillimanite crystallization. The second metamorphic phase has been dated by Vachette (1964) on muscovites from pegmatites using the Rb-Sr method of dating. The results compared with the new constants (Steiger and Jäger, 1977) give an isochrone with an age of $1,970 \pm 30$ Ma. The third metamorphic event is less important, creating only local schistosity in the central part of the mobile zone; large tectonic structures like wide folds correspond with present cartographic structures. Rb-Sr dating of the biotites related to the third tectonic event gives an isochrone at 1,874 \pm 91 Ma (Bonhomme et al., 1982; Gauthier-Lafaye, 1986).

Unmetamorphosed sediments of the Francevillian Series are located in the eastern part of the basin and belong to what is generally called the Francevillian Series sensu stricto. This area is affected by northwestsoutheast-trending faults that separate structurally high domains (e.g., the Plateau des Abeilles and Dambi and Andjogo shoals) where the Francevillian Series is less than 1,000 m thick from the lows (i.e., the Okondja, Lastoursville, and Franceville basins) and where the Francevillian Series is thicker. At the far eastern part of the Francevillian basin these unmetamorphosed sediments are overlain by flat-lying Mesozoic continental sediments of the Congolais pla-



FIG. 1. Geologic map of Gabon. Insert shows the location of Gabon in Africa.

teau, the Stanley Pool, and the Bateke plateau series. All the known uraniferous ore deposits are located in the Franceville basin. In addition to the most famous uranium deposit of Oklo, the Franceville basin contains five other uranium ore deposits: Boyindzi, Mounana, Okelobondo, Bangombé, and Mikouloungou. For this reason only the geology of the Franceville basin will be presented here.

Stratigraphic column in the Franceville basin

Weber (1968) has subdivided the Francevillian Series in the Franceville basin into five formations which are indexed from FA (bottom) to FE (top) (see Fig. 2). The FA formation is 100 to 1,000 m thick and consists of sandstones and conglomerates whereas the upper formations (FB-FD) consist mainly of black shale. Dolomites and banded cherts are also dominant in FC, and tuffs are present in FD and FE.

The FA formation: This formation contains all the uraniferous mineralization and for this reason has been well prospected. Sedimentologic studies of rocks in quarries and boreholes located in the Franceville basin indicate that the sedimentologic record of the FA formation is dominated by a marine transgression from the southeast toward the northwest. This transgression is characterized by deltaic sedimentation overlying fluviatile deposits (Fig. 3). All the uranium ore deposits are located in deltaic plain sediments, at the transition between fluviatile and prodeltaic environments.

In the center of the basin the FA formation has been subdivided into five units corresponding to various sedimentary environments (Fig. 4) and is numbered from the bottom to top into unit 4, the Otobo unit, and units 3 to 1.

At the bottom, unit 4 consists of 200 to 300 m of fluviatile deposits. These sediments consist of coarse, poorly sorted and conglomeratic sandstones containing angular elements. Simple sequences are always a few centimeters thick and fining upward and are often truncated at their top. Bedding consists of oblique stratifications with minor deviation of their orientations. Red radioactive conglomerates with heavy minerals concentrated in thin layers are important deposits (100–200 m thick) in these fluviatile sediments. Petrographic observations reveal that this radioactivity is due to thorite and uraniferous thorite minerals, which appear altered (Table 1).

Unit 4 is overlain by the Otobo unit which consists of two or seven (depending on the area) coarseningupward sequences, each 10 to 50 m thick. Each sequence starts with black marine shales at the bottom and ends with coarse-grained sandstones or conglomerates indicative of progradation of a deltaic plain environment (Haubensack, 1981; Gauthier-Lafaye, 1986). This Otobo unit contains the uranium mineralization at Kiene.



FIG. 2. Stratigraphic section of the Francevillian Series in the Franceville basin.



FIG. 3. Paleogeographic reconstruction of the FA formation in the Franceville basin and location of the uranium ore deposits.

Unit 3 consists of 100 to 150 m of alternating fine sandstones and shales, representing a tidal environment. This is followed by a new cycle of fluviatile sedimentation which contains the uranium mineralization at Mikouloungou (unit 2) and deltaic sedimentation (unit 1). In the deltaic sediments the laminations show wide variation, sometimes with opposite orientations but are well oriented in the fluviatile sandstones. Slump bedding and slump folds are common in the deltaic shales but are absent in the fluviatile sediments. Finally, deltaic unit 1 is overlain by the marine FB formation black shales.

In contrast to the central part of the basin, deltaic sedimentation is less developed in the edge of the basin, resulting in the marine transgression from the southeast toward the northeast (Fig. 4).

At the edge of the basin, most of the FA formation consists of fluviatile sandstones and conglomerates with thickness up to 800 m, the deltaic sediments being restricted to the upper 10 m of the upper FA formation. The Oklo mineralized layer (C_1 layer) is a good example of deltaic environment in the upper FA sedimentation of the western edge of the Franceville basin (Fig. 5). It consists of a 7- to 10-m thick coarsening-upward unit in which beach deposits, prodeltaic and front sediments, and marine shales are seen.

The FB formation: The FB formation lies slightly discordantly over the FA formation and has been subdivided into two units, FB1 and FB2. Unit FB1 is mainly black shale, consisting of fine-grained, silty sediments, more or less calcareous, and rich in organic matter. The basal sediments of the FB1 unit at the edge of the basin contain olistoliths and polygenic breccias which can be correlated with brecciated mud flow and turbidites (banded silty argillite) from the internal part of the basin. These facies reveal a collapse stage of the basin during the transition of the FA facies to the FB. Tectonic activity during FB1 sedimentation is also responsible for the great thickness change of this formation in the Franceville basin. The maximum thickness (>1,000 m) occurs in the center of the basin; at the edge of the basin FB1 sediments are about 400 m thick. At the top of this unit lies the manganese ore deposit of Moanda. This deposit has been formed through the weathering of an horizon rich in manganese carbonates (Weber, 1968). Unit FB2 overlies FB1 and comprises 100 m of thick, well-sorted sandstones with irregular geographic distribution; the sandstones form sedimentary bodies of several square kilometers which are separated by layers of black shale zones.

The FC, FD, and FE formations: The FC formation consists chiefly of massive dolomite and thickly banded cherts, which provide a useful stratigraphic reference. This formation, generally 10 to 40 m thick in the Franceville basin, may reach 150 m thick at the northwestern edge of the Lastoursville basin and is transgressive over the basement in the structural highs. The FD formation consists of black shales, with ignimbrite tuff becoming more dominant at the top of the formation. The FE formation comprises epiclastic sandstones and interlayered shales. These FD and FE formations cover very large areas and are probably more than 1,000 m thick in the Franceville basin.



FIG. 4. Sedimentologic log of the FA formation in the central part of the Franceville basin.

Drill hole 1 Dep	no. O oth 492	Ok 24 492.00 m		Ka 13 358.00 m				Gr 31 593.20 m			
Eo ⁺²	5.01	5 10	0.17	0.39	9 53	4.01	0.24	0.00	9 93	0.13	0.00
re v	2.01	2.20	1.59	1.05	1.53	1.07	9.13	1.84	1.85	1.85	913
I V	0.24	3.20	1.00	1.55	0.04	2 22	2.15	1.04	0.02	0	2.10
⊾ Si	13.94	0 14.50	12.77	16.94	13.66	2.33	17.83	17.77	17.96	17.31	17.43
La	0	0	0.23	0	0.14	0.25	0.80	0.83	2.18	1.09	1.56
Al	0	0.30	0.65	0.90	0.67	8.32	0.35	0.34	0.30	0.32	0.19
Ce	0.45	1.30	1.39	0.57	0.17	0.13	5.50	4.98	5.52	6.04	5.50
Mg	0	0	0	0.01	0.02	0.56	0.01	0.03	0	0	0
Ca	2.35	2.20	3.79	0.82	6.69	0.59	0.98	1.29	1.02	1.19	1.25
U ⁺⁴	0.35	0.80	3.02	3.58	3.46	3.03	1.54	3.23	2.76	1.27	5.89
Th	56.84	51.50	38.58	31.77	28.01	24.69	58.14	54.21	59.08	60.68	54.17
Pb	0	0	0.26	0.46	0	0	0	0	• 0	0	0
Zr	1.59	2.00	22.02	24.46	20.66	15.63	0	0	0	0	0.58
Р	5.03	3.40	1.61	2.58	6.48	1.33	1.75	1.63	1.48	2.13	2.22
Total	88.00	84.40	86.08	84.37	84.06	87.41	89.27	86.13	92.42	92.00	90.93

TABLE 1. Microprobe Analyses (wt %) of the Uraniferous Thorites of the Fa Formation

Tectonic structure of the Franceville basin

The Franceville basin was deformed by collapse along major northwest-southeast normal faults, and to a lesser extent by east-west dip-slip faults with throws of 200 to 1,000 m (Fig. 6). These deformations produced a graben basin of 60 km long and 30 km wide. Reverse faults and strike-slip faults (mainly north-south and northwest-southeast) also occur and are associated with folds characterized by high dip axes and wide northeast anticlinal axes.

A major north-south-trending fault along which basement horsts occur (the Mounana and Boukoussou horsts) separates the Franceville basin from the Las-



FIG. 5. Sedimentologic log a...d interpretation of the C_1 layer at the Oklo ore deposit.

toursville basin (Fig. 7). This fault was active during sedimentation as a normal fault and was reactivated later as a strike-slip fault.

These tectonic structures are interpreted using a shearing dynamic model (Wilcox et al., 1973) in which the major shears are the north-south senestral strikeslip faults, the northwest-southeast faults are the Riedel's synthetic shears (R) counterpart (Riedel, 1929), and the east-west normal faults are the tensional transversal structures or second-order synthetic shears. This model may be applied to the other Francevillian basins and implies an east-west compressive stress. This agrees with the submeridian schistosity trend measured in the Ogooué mobile zone (Gauthier-Lafaye, 1986).

The uranium ore deposits

The uranium ore deposits of the Franceville basin can be separated into two distinct groups according to their economic importance and geologic location and are designated here as deposits of the northwest edge and central part of the basin.

Uranium ore deposits of the northwestern edge of the basin: The deposits of the northwest basin are the most important ones and are located at the eastern edge of the Mounana basement horst (Fig. 7). These deposits include from north to south: (1) Boyindzi, a blind deposit of 3,100 tons of uranium, now mined underground, (2) Mounana, the first deposit to be discovered in Gabon in 1955 and which produced 5,700 tons of uranium until 1975, (3) Oklo, the most important of the uranium ore deposits with 17,300 tons of uranium, and (4) Okelobondo with 7,400 tons of uranium. Oklo was first mined by the open-pit method and is now mined underground; the Okelobondo deposit has not yet been mined. Together these deposits represent 80 percent of the known uranium in the Franceville basin.



FIG. 6. Geologic cross sections in the Franceville basin.



FIG. 7. Structural map of the Franceville and Lastoursville basins. Abbreviations: B = Bangombe, Bo = Boyindzi, K = Kiene ore deposits, M = Mikouloungou, Ok = Okelobondo; 1, 2, 3, 4 = location of cross sections in Figure 4.

1 to 9 eactors

Uraniferous lave

C₁ layer

Olistholith



the natural fission reactors number 1 to 9. Note steep, easterlydipping strata and the occurrences of uraniferous C_1 layer in the upper FA formation.

Uranium is located in the upper part of the FA sandstones and is always associated with tectonic structures. The main structure is a wide monoclinal fold, with a fold axis parallel to the north-south fault of the Mounana basement horst and with its west flank resting against the basement horst. In the Oklo deposit, northwest-southeast faults showing horizontal slip intersect this monoclinal fold producing large drag folds with high dip axes (Fig. 8). The first and main natural fission reactors were found around these folds. At Mounana and Boyindzi, the west flank of the monoclinal fold has a higher dip $(60^\circ - 90^\circ)$ and the deposits are located at the intersection of the northwest-southeast faults and the north-south fault of the Mounana basement. At this intersection intense fracturing occurred and local cataclastic breccias were formed.

Uranium ore deposits in the central part of the basin: Less important deposits are located in the basin center and include the unmined Bangombé deposit (3,000 tons of uranium), Mikouloungou (4,700 tons of uranium) which has been partially mined, and an uraniferous area called Kiene where the economic interest is uncertain. These deposits are located in two types of structures. At Bangombé the uranium ores are concentrated at the top of a large anticline with a northeast-trending axis. As in the deposits in the northwest basin, mineralization at Bangombé is restricted to the upper FA sandstones. In 1986 new natural fission reactors were discovered in this deposit. At Mikouloungou and Kiene, the mineralization is associated with east-west and northwest-southeast normal faults, respectively. The normal faults put the FA sandstones in tectonic contact with the FB black shales. The walls of these normal faults are oxidized and represent higher uranium content relative to the footwall sandstones. At Mikouloungou and Kiene, the uranium ores are located in the lower FA formation. A cross section of Kiene is given in Figure 9.

In the deposits of the Franceville basin, the highest uranium contents are associated with microfracture networks, more or less parallel to the bedding. These microfracture networks are associated with faults and changes in grain size, which are most easily observed in sandstones overlain by the FB black shales or interlayered pelites (see later section).

All the uranium ore deposits of the basin appear to be located in tectonic structures, which illustrate the main characteristics of petroleum traps. The ore deposits are located in the upper levels of the basin, and the uranium-bearing deposits are capped by the impermeable black shales of the FB formation.

Petrographic and Geochemical Data

Main characteristics of the FA sandstones

The main effects of diagenesis in the FA and FB sediments are silicification of the quartzite levels and illitization or chloritization in the argillaceous matrix.



FIG. 9. Cross section of the Kiene ore deposit.

Dolerite

Quartz overgrowths occur in sandstones anywhere in the FA stratigraphic column although they are more important in clean sandstones, which have no argillaceous matrix, and in well-sorted quartz grains of the surrounding deltaic sandstones. In some cases, two stages of silicification can be distinguished when they are separated by sulfide, clay crystallization, or organic particles with shapes like solidified oil products.

Sulfates, mainly anhydrite, and dolomite are also important sandstone cements occurring in deltaic sediments and sometimes in fluviatile unit 4.

X-ray analyses of the clay fraction ($<2 \mu$) of the FA sandstones show that illites have a crystallinity index (measured at half height of the 10 Å XRD peak, as measured by Dunoyer de Segonzac, 1969) ranging between 3.5 and 6 which decreases with stratigraphic depth (Fig. 10). These illites are predominantly of a 1M type; only in the deepest levels of the FA does the 2M1 type assume some importance. These data are characteristic for a diagenetic to very low grade metamorphism.

Rb-Sr and K-Ar isotope analyses have been performed on these diagenetic clay minerals (<2 μ) in order to date this diagenesis (Bonhomme et al., 1982). Results give two Rb-Sr isochrones with ages of 1,867 \pm 78 and 1,875 \pm 30 Ma, respectively, and with values ranging between 1,821 and 1,860 Ma for K-Ar analyses. This age, around 1,870 Ma, is interpreted as the age of the last thermic event affecting the new crystallized clay minerals and thus is the age of the diagenesis of the Francevillian sediments and the maximum point of burial of the Franceville basin.

The uranium ores

Two types of ores are distinguished in the uranium ore deposits: a commonly occurring low-grade ore with a uranium content between 0.1 and 1.0 percent, and a high-grade ore with a uranium content ranging from 1.0 to 10 percent. The high-grade ore is restricted to areas associated with tectonic structures and is located in bodies about 5 to 20 m in length, 5 to 10 m wide and 0.3 to 2 m thick. The economic character of the mineralization is due to the common ore, but the high-grade ore is important because it is the ore in which fission reactions started.



FIG. 10. Cristallinity index (C.I) of illite and 2M illite proportion vs. depth in the FA formation of the Franceville basin (borehole BA2, Bangombe).

Using the U-Pb method of dating, the age of the uranium mineralization ranges between 1,700 and 2,100 Ma, depending on the data of various authors. Lancelot et al. (1975) and Hagemann et al. (1974) gave two ages, 1,700 to 1,800 and 2,000 to 2,100 Ma, respectively, and pointed out the importance of lead migrations. Devillers and Menes (1978) put the age of the uranium mineralization event before 1.970 Ma; they suggest old and recent perturbations and possible enrichment of radiogenic lead in the uranium deposit during the mineralization process. Finally, Gancarz (1978) and Gancarz et al. (1979) interpret their results as suggesting an age of 2,000 Ma for the uranium mineralization event; they suppose that 1,800 Ma should be interpreted as a consequence of a continuous diffusion of lead. In all these studies, samples come from the Oklo deposit and many of these authors do not make clear the difference between samples associated with the nuclear fission reactors and samples which have no relation to them. Such mixing of samples makes the various interpretations doubtful. Furthermore, we will see that two stages of uranium mineralization occurred in these deposits, making it difficult to interpret the U-Pb data because a correction for initial lead is unknown. Ion microprobe analyses on uraninite and galena single crystals should produce better results. Preliminary data seem to agree with a complex history of the uranium mineralization process (Holliger et al., 1989).

The common uranium ore: The common ore consists of black, silicified sandstones. The sandstone matrix consists largely of secondary silica resulting from quartz nourishing and contains chlorite and illite in varying proportions. Sulfides are abundant and are dominated by pyrite and galena with minor marcasite and copper sulfides (chalcopyrite, digenite, and covellite). Examination of polished sections and XRD analyses of ores reveals that the uranium occurs mostly in the form of pitchblende, sometimes with coffinite surrounding pitchblende. Except for the Mounana deposit, uranium minerals containing U⁺⁶ (e.g., uranocircite, renardite, torbenite, and autunite) are rare and are always linked with recent secondary oxidation. At Mounana, five uranium vanadates with U⁺⁶ were described as new species: francevillite, curienite, mounanaite, vanuralite, and metavanuralite. All these minerals were located in the upper part of the deposit, in the oxidized zone, associated with vanadium mineralization. Vanadium oxides such as chervetite, lenoblite, and barianite were described for the first time in this deposit.

In this common ore, pitchblende is always associated with hydrocarbon material of high catagenetic rank. The mineralized organic matter consists of solid bitumen (asphaltite) with a hydrogen/carbon ratio of less than 0.5 atomic percent (Fig. 11), and releases only a little methane when it is pyrolyzed. Petrographic observations show that the bitumen consists

FIG. 11. H/C and O/C diagram of the extracted organic matter from sediments of the Franceville basin.

A Coal

0.2

Black shale FB-C barren bitumen FA barren bitumen

FA mineralized bitumen

0.3

•

Mixar

0.1

O/C atomic

mostly of spherical particles in the secondary porosity of the sandstones. The secondary porosity is created by the microfracture network and by the corrosion of detrital quartz and quartz overgrowths. The microfractures are often concordant with the bedding and affect the detrital and secondary quartz as well as the matrix of the sandstones (Fig. 12). The microfractures are thought to have been created by hydrofracturing (see later section). Calcite, whose fluid inclusions will be described in a later section, occurs in association with the mineralized bitumen. In the bituminous particles, pitchblende is present as microscopic $(1-10 \mu)$ inclusions and may form rims around the organic matter. An intimate association between uranium and organic material has been suggested by Vanderbroucke et al. (1978), Cortial (1985), and Cortial et al. (1988) who have shown by electron microscope examination that bituminous particles have high concentrations of uraninite microinclusions a few angstroms in size. Chemical analysis of such bitumen (Table 2) also reveals their very high uranium grade: greater than 2

TABLE 2. H/C, O/C, and Uranium Contents of Some FA Bitumens

Samples Location, drill hole no., and depth	H/C (atomic)	O/C (atomic)	U (%)
Bangombe			
BA2, 16.30 m	0.40	0.130	1.75
Oklo			
SF 24, 43.60 m	0.33	0.138	1.60
OK 99, 403.55 m	0.45	0.203	1.45
GL 3541	0.39	0.251	>2.00
y = -208	0.38	0.204	>2.00
Reactors			
9. KP 3, 4.50 m	0.43	0.181	6.80
8. KP 13, 1.75 m	0.45	0.128	2.64
8. KP 13, 5.80 m	0.53	0.173	1.35
7. GL 2497	0.38	0.190	-2.00
7. GL 2413	0.46	0.163	10.75
7. GL 2478	0.44	0.213	29.70

percent and even greater than 10 percent in samples related to the fission reactors.

The high-grade uranium ore: High-grade ore generally occurs in more highly fractured sandstones than does the common ore and is interpreted to consist of hydraulic breccias (Fig. 13) associated with tectonic structures such as faults and joints. The high-grade ore is a black sandstone of variable silicification, containing hematitic spots. The uranium occurs mostly as pitchblende and some coffinite but is never included in the organic material. All uranium mineralization is restricted to secondary porosity made by corrosion of detrital quartz and quartz overgrowth related to the microfracture network. Uranium is associated with sulfides (mostly pyrite and galena) and is located in a matrix consisting mainly of chlorite and some illite. The uranium mineralization is located around the hematitic spots from which it is separated by a white rim. The matrix of the hematitic spots contains mainly hematite and illite whereas the white halo



FIG. 12. Typical example of hydrofracturing in a common uranium ore (from Oklo).

H/C

atomic

1

0.0



FIG. 13. Typical example of hydraulic breccia in a high-grade uranium ore (from Oklo).

consists mostly of illite. Formation of high-grade ore is interpreted to be the result of an oxidation-reduction process occurring in strongly hydrofractured common ore. Uranium in the common ore is mobilized by oxidized fluids migrating in the tectonic structures, resulting in their oxidiation, and it precipitates when the uranium-bearing fluids meet more reduced conditions.

Colors of the sediments and their oxidation states

In the Franceville basin, the FA rocks are often red colored and generally occur in a sequence of greenand black-colored sediments. Black sandstones are restricted to higher levels in the basin where uranium ore deposits occur, whereas green sediments are always found between black and red sandstones. Because we know the importance of the oxidation-reduction process for uranium occurrences, we describe these various colored sediments to show the origin of such colors.

The contact between sediments of different colors is clearly discordant with the stratification (Fig. 9) and indicates that the arrangement of the colors is not sedimentary but was acquired during the diagenetic stage.

The main petrographic characteristics of each colored sediment are summarized in Figure 14. It can be seen that these three facies have a particular clay mineral composition and a different type of alteration of the detrital biotites.

The red color is due to hematite impregnation of the argillaceous matrix; hematite also surrounds the quartz grains between their detrital boundary and their overgrowths. This latter observation suggests that oxidation of the sediments started during early diagenesis. The argillaceous matrix of these sandstones mainly consists of illite. Sandstones also contain detrital biotites, and microscopic examination reveals that these biotites are altered, forming opaque minerals with a high hematite content. Microprobe analyses of these altered biotites show that they have a muscovite composition (Table 3). The structural formulas, however, reveal an excess of iron and titanium which probably results from the iron and titanium oxides occur not in the mica network but on the surface of the mineral. These red-colored sediments, especially in the fluviatile environment, also contain carbonates (mainly dolomite) and sulfates (anhydrite, gypsum, and barite). Barite is generally restricted to the vicinity of the uranium ore deposits.

The color of the black sediments is mainly due to organic matter present in pore spaces. Petrographic examination of these rocks permits differentiation into two types of black sediments, depending on the alteration of the detrital biotites and on the clay mineral composition.

The first type consists of rocks with brown biotites and micas showing open flakes. Microprobe analysis of biotites (Table 3) reveals an intermediate compo-



FIG. 14. Main characteristics of the various colored sediments.

	Chlo	rites	Altered detrital biotites			
	Sedin	nents	Sediments			
	Black	Green	Black	Green	Red	
SiO ₂	25.05	24.04	30.17	30.67	43.78	
Al_2O_3	20.80	19.76	23.29	20.25	26.20	
MgO	6.06	5.61	5.08	7.05	1.42	
CaO	0	0.04	0.06	0.02	0.07	
Fe_2O_3					9.30	
FeO	3 3.78	35.75	27.30	28.94		
Mn ₃ O ₄	0.03	0.12	0.06	0.11	0.07	
TiO ₂	0.15	0	0.08	1.72	3.34	
Na ₂ O	0	0.03	0.12	0.03	0.12	
K ₂ O	0.32	0.07	2.13	1.53	9.00	
Total	86.34	85.12	92.45	90.92	92.49	
	Struc	tural form	nulas			
Si	2.795	2.773	3.012	3.373	3.060	
Al(IV)	1.205	1.227	0.988	0.627	0.940	
Al(VI)	1.526	1.455	1.747	1.564	1.214	
Mg	1.014	0.971	0.761	0.973	0.149	
Fe ⁺³					0.487	
Fe ⁺²	3.141	3.437	2.271	2.218		
Mn	0.003	0.011	0.005	0.008	0.004	
Ti	0.013	0	0.006	0.119	0.175	
Total Octahedral	5.696	5.873	4.789	4.881	2.029	
Ca	0	0.005	0.006	0.002	0.005	
Na	0	0.007	0.023	0.005	0.016	
K	0.046	0.010	0.271	0.180	0.803	
Charges	27.78	28.00	26.71	27.13	22.00	

TABLE 3.Microprobe Analyses (wt%) of Chlorites and
Detrital Biotites of the FA Formation

sition between a mica and a chlorite with a depletion in Si, K, and Al. Their structural formulas have been therefore computed using the "chloro-mica" mode, which builds a mica with all the potassium (one K atom in each half unit cell), and a chlorite with the remaining elements. The total iron is expressed as FeO, and the half unit cell possesses between 22 and 28 negative charges. Biotites thus calculated have a composition intermediate to Fe-Al chlorite and muscovite, but closer to Fe-Al chlorite than to muscovite. In these rocks the argillaceous matrix consists mainly of chlorite with a high iron content (Table 3) and some illite.

The second type of black sediment always has a high organic matter content. In this facies, the detrital biotites appear very altered and are like muscovite with a pale color, very low pleochroism, and much titanium oxide on their surfaces (Table 3). The argillaceous matrix consists of illite without chlorite. In barren sandstones, organic matter is located in primary porosity, around detrital quartz boundaries, or trapped in quartz overgrowths. This type of organic matter shows the typical shapes of solidified petroleum drops. The first type of black rock is interpreted as sediment which has never been oxidized, whereas the second type corresponds to originally oxidized sediment which was later reduced by introducing bitumen.

The matrix of the green sediments contains both illite and green chlorite in varying proportions. Detrital biotites are altered to muscovite and green chlorite, both of which crystallized between opened mica flakes. Microprobe analyses of chlorite of the argillaceous matrix and of chlorite from the alteration of the biotites are given in Table 3; both chlorites have similar chemical compositions. In some sandstones, hematite rims persist around the quartz grains and between the boundary and the overgrowth of detrital grains, suggesting that these sediments were originally oxidized. Pyrite accumulations are common in the green sediments. Pyrite corrodes both the detrital and overgrowth quartz and is generally associated with the green chlorite of the matrix. Pyrite accumulations are interpreted as reduced fronts, resulting from the flow of reducing fluids through oxidized sediments. The green chlorite crystallized in this reduced environment.

These data suggest that the oxidation of the FA formation sandstones started during early diagenesis. Oxidized fluids from the sandstones containing no organic matter flowed upward during sediment compaction. In some areas, the flow of oxidized fluids caused the oxidation of the deltaic sediments containing organic matter and even of the bottom of the FB black shales. The oxidized fluid lost its oxidizing potential rapidly through interaction with reduced sediments. At the same time, the burial of the sediments became more important and black shales in the FA deltaic sediments and in the FB formation began to generate oil (see later section). Oil generation resulted in the flow of reduced fluids which became more important as the organic matter matured. The migration of these reduced fluids into the previously oxidized sediments created reduced fronts and the green-colored sediments. It appears therefore that the oxidation state of the sediments is the result of the competition between two fluids which started very early in the diagenetic history of the sediments and continued very late. We will see that such a diagenetic history may have implications for the deposit of uranium ore.

Fluid inclusion data

Fluid inclusion studies have been performed to determine the pressure and temperature conditions of the diagenetic environment and the composition of diagenetic fluids (Openshaw et al., 1978; Gauthier-Lafaye and Weber, 1981; Gauthier-Lafaye, 1986). Microthermometric studies have identified three types of fluid inclusions from variations in salinity and homogenization temperatures (Figs. 15 and 16): type 1 with a medium salinity (3–10 equiv wt % NaCl) and high homogenization temperature ($140^{\circ}-180^{\circ}C$), type 2 with a low salinity (0.7–1.0 equiv wt % NaCl) and low temperature ($100^{\circ}-130^{\circ}C$), and finally type 3 with a high salinity (20 equiv wt % NaCl) and low homogenization temperature ($120^{\circ}-140^{\circ}C$). All fluid inclusions studied here are composed of liquid and gas at room temperature.

Type 1: This type has been found as inclusions hosted by overgrowth quartz, euhedral quartz located in fractures, FB dolomite, and dolomite cement in FA sandstones (Figs. 15 and 16). Some quartz overgrowths include bitumen and are therefore assumed to be contemporary with the oil migration in the FA sandstones. Homogenization temperatures of type 1 fluid inclusions in quartz overgrowths range between 80° to 400°C, and melting temperatures for all type 1 inclusions are relatively constant, ranging between -6.0° to -1.0° C, corresponding to 10 to 3 equiv wt percent NaCl (Fig. 16A). The wide range of homogenization temperature of fluid inclusions in quartz overgrowths is illustrated in Figure 17 for sandstone from the Bangombé uranium ore deposit. This figure also shows that almost all the fluid inclusions fall into lines corresponding to microplanes called Tuttle's planes (Tuttle, 1949) which are associated with hydrofracturing (see later section). The wide scatter of the homogenization temperatures suggests that many of these fluid inclusions have been modified after the fluid was trapped by hydrofracturing (Gauthier-Lafaye and Weber, 1984). However, the histogram of



FIG. 15. Location of the different types of fluid inclusions studied.



A, HIGH TEMPERATURE AND MEDIUM SALINITY FLUID INCLUSIONS



B.LOW TEMPERATURE AND LOW SALINITY FLUID INCLUSIONS



FIG. 16. A, B, and C. Homogenization (T_h) and melting (T_m) temperatures of the three different types of fluid inclusions (see text).



FIG. 17. A. Example of fluid inclusions in quartz overgrowth showing their shape and homogenization temperatures. Most of the fluid inclusions are located in Tuttle's planes (Tuttle, 1949). B. Homogenization $(T_h^{\circ}C)$ and melting $(T_f^{\circ}C)$ temperature histograms of fluid inclusions located in quartz overgrowths (samples from borehole BA2 at 13.30, 16.60, 19.00, 21.60, and 109.60 m).

the homogenization temperatures (Fig. 17) shows that many fluid inclusions homogenize at temperatures between 140° to 180°C.

Openshaw et al. (1978) have analyzed the Na-K ratio of fluid inclusions in euhedral quartz from fractures of the Oklo ore deposit, the fractures contain only type 1 fluid inclusions (4 equiv wt % NaCl and T_h of 160°C). The average of the Na/K atomic ratio for five analyses is 0.054 ± 0.007 . Using extrapolation toward low temperature of the Na/K geothermometer

established by Poty et al. (1974) and Lagache and Weisbrod (1977), Openshaw et al. (1978) determined a temperature of $238 \,^{\circ}C \pm 17$. Nevertheless, in this temperature range it seems better to use the empirical geothermometer of White (1965, 1970) and Ellis (1970), which gives temperatures of 178° and 188°C, respectively, since they are in better agreement with the results of the microthermometric study. The isochore of this fluid has been constructed using data from Hilbert (1979) and shows that a temperature of 183°C (average temperature between White and Ellis geothermometers) corresponds to a fluid pressure of 410 bars and a geothermal gradient of 40°C/100 bars. Assuming that the fluids in the FA sandstones were in the hydrostatic pressure regime, the upper FA formation was overlain by 4,100 m of sediments.

Type 2: Fluid inclusions of this type are observed in dolomite cement of FB1 sandstones rich in organic matter (sample from Okelobondo area), and in quartz crystallized in fractures. These fractures intersect FB dolomites and are also filled with bitumen (sample from the Bangombé area). Openshaw et al. (1978) have also found similar fluid inclusions in quartz from fractures in FA sandstones located under the Bangombé uranium ore deposit. Homogenization and melting temperatures of these fluid inclusions range respectively between 100° to 120°C and -0.03° to -1.5°C, corresponding to 0.7 to 2.0 equiv wt percent NaCl.

Tupe 3: This type of fluid inclusion is found in calcite in microfractures in the uranium ore. These microfractures (the result of hydrofracturing, see later section) are also filled with uranium oxides and bitumen. Similar fluid inclusions have been found in barite cements of FA sandstones surrounding the Oklo ore deposit at the same stratigraphic level as the mineralized layer. Openshaw et al. (1978) also describe similar fluid inclusions in dolomite cements of the FA sandstones. These fluid inclusions are characterized by very low melting temperatures, ranging from -38° to -16° C representing very high salinity, greater than 20 equiv wt percent NaCl. The lowest temperature is lower than the eutectic point in the NaCl-H₂O system, suggesting the presence of divalent ions. The homogenization temperatures are very similar to those of type 2 fluid inclusions, ranging between 100° and 140°C.

Petrologic observations suggest that the three types of fluid inclusions are related to two main events, burial and uplift, which occurred during the sediment diagenesis.

During the burial stage, sediments were buried to a depth of about 4,000 m; the temperature of the diagenetic fluid was about 180°C. During the early phase of this stage, the sediments reached the oil window and petroleum invaded the FA sediments when the sandstones were silicified. The burial temperature is recorded in type 1 fluid inclusions.

During uplift stage, quartz overgrowths are affected by microfractures which are filled with quartz and calcite containing type 2 and 3 fluid inclusions. Fluid inclusion data reveal that microfractures are filled at lower temperatures than the preceding diagenetic silicification. If we consider a geothermal gradient of 40°C/100 m, temperature and capture pressure of these fluid inclusions can be estimated. They are quite similar for these two types of fluid inclusions, ranging between 120° to 140°C and 270 to 320 bars. The low-temperature and high-salinity fluid represented by type 3 inclusions follows paragenetically the fluid characterizing the deepest burial stage of the basin and is associated with the main mineralization stage. The low-salinity fluid related to the bitumen occurrences (type 2) is associated with the oil migration during this uplift event.

Organic matter in the Franceville basin

Because uranium and organic matter are intimately associated and the uranium ore deposits are located in structural oil traps, it is appropriate to discuss the main characteristics of the petroleum deposits in the Franceville basin.

The petroleum potential of the Franceville basin may be estimated by studying the nature of the oil source rocks. The total organic matter content of various facies of the Francevillian formations are illustrated in Figure 18. The FB and FD black shales have high concentrations of organic carbon, up to 10 to 15 percent. Microscopic examination of black shale shows that the organic matter is concentrated in texturally coarse layers concordant to the bedding, suggesting a primary migration of the oil. Reflected light microscopy indicates that this organic matter has a high anisotropy, which has been called anthracitic anisotropy by Cortial (1985). Electron microscope studies using the dark-field method described by Oberlin (1977) indicate that most of the organic matter of the black shale consists of a mixture of particles with small and large molecular structure. Such mi-



FIG. 18. Organic carbon distribution in the stratigraphic series of the Franceville basin (n = number of analyses).

crotextures correspond to a mixture of kerogen and solidified oil (or bitumen), respectively, when compared with the charter of the microtextures of organic matters from various origin (Oberlin et al., 1980). Organic matter also occurs as bitumen occupying primary porosity and fractures in permeable sandstones and dolomites interbedded in the FB black shales. This bitumen has a very high reflectance and a granular anisotropy under reflected light. Electron microscope observations suggest that this bitumen is slightly oxidized and corresponds to a solidified hydrocarbon with a very large molecular structure.

In the FA sandstones, two types of bitumen are classified according to their mineralized or barren state. Barren bitumen is always located in closed primary porosity or in quartz overgrowths and is similar to the organic matter located in the permeable FB sediments. In contrast, the mineralized bitumen occurs in open secondary porosity. Mineralized bitumen has a very low reflectance and anisotropy, except around the pitchblende inclusions, and is generally more oxidized than the barren bitumen as suggested by its high O/C atomic ratio (Fig. 12) and its very small molecular structure, as revealed by electron microscope studies. These two facies of FA bitumen reflect different diagenetic histories. The evolution of the mineralized bitumen stopped when it became oxidized during the mineralization process, while the less oxidized barren bitumen may continue the normal catagenetic evolution. This is supported by petrographic observation of organic particles located at the border of quartz overgrowths. For the same particle, the barren facies (with high reflectance) occurs trapped in the quartz overgrowths while the mineralized facies (with low reflectance) is located in the pore spaces of the overgrowth.

Carbon isotope data of the organic matter suggest that the FB black shales are the potential source rock of FA sandstone bitumen (Fig. 19). Organic matter from the FA and FB formations have similar δ^{13} C values (between -21 and -30 ‰ PDB) whereas the δ^{13} C values become progressively more negative moving up the stratigraphic column to a minimum of -46.2 per mil in the upper part of the Francevillian Series (Weber et al., 1983). In this model the oil source rock (FB black shales) is at a higher stratigraphic level than the oil reservoir (FA). However, the numerous faults which affect the Franceville basin (Fig. 6) may move the FA reservoir rocks at higher levels than the FB oil source rocks in many places and allow migration of the hydrocarbons from the FB to the FA formation.

Hydrofracturing

The association of microfractures with uranium ore has been noted. These microfractures consist of a dense, opened microfracture network which splits quartz grains and their overgrowths, without showing



FIG. 19. δ^{13} C (PDB) of extracted organic matter of various facies in the stratigraphic series of the Franceville basin.

any lateral displacement of the wall. Microfractures are filled by calcite, clays, pitchblende, and bitumen. In high-grade ore, the fracturing resulted in hydraulic breccia with all quartz split and highly corroded. Hydrofracturing of the sandstones is always associated with planes of secondary fluid inclusions, named Tuttle's planes (Tuttle, 1949), which cut quartz grains and their overgrowths. Tuttle's planes are intercrystalline microstructures whose orientations are not dictated by the crystallographic axes of the quartz crystals. Tuttle (1949) has suggested that these planes can be parallel to shearing planes and to tension gashes. The ubiquitous association between microfractures and Tuttle's planes which contain many fluid inclusions suggests that the fluids play a major role in the microfracturing process. Thus these microfractures are interpreted as hydrofracturing; overpressured fluids are a major factor in the control of this type of fracturing (Gauthier-Lafave and Weber, 1988). In the Franceville basin, however, it is possible that in addition to overpressured fluids, tectonic stress occurs and induces hydrofracturing (Gratier, 1984).

Homogenization temperatures of the Tuttle's plane inclusions (type 1) have very scattered values, ranging from 100° to 400°C, which suggests that modification occurred as a result of microfracturing. However, it has been seen that fluid inclusions in calcite (type 3) in hydraulic fractures may correspond to the uplifting stage of the basin. Thus, Tuttle's planes and hydraulic fractures probably formed during the uplift stage of the basin and followed quartz silicification and burial diagenesis.

We propose that hydraulic fracturing results from overpressured fluids in the FB and FD pelites and black shales (Gauthier-Lafave and Weber, 1988). It is not possible to reconstruct definitively the vertical fluid pressure gradients in the Franceville basin prior to its present compaction state. However, using modern examples of oil basins, well constrained by boreholes (e.g., Perrodon, 1980; Magara, 1981), and considering the main lithologic character of the Francevillian stratigraphic column, it is possible to propose a reconstruction of the pressure gradient before the sediments were completely lithified. In modern basins such as the Gulf Coast (Magara, 1978 and 1981), the Mackensie delta (Evans et al., 1975), and the Mahakam delta in Indonesia (Durand and Oudin, 1979), the fluid pressure gradients are very close to the hydrostatic gradient to a depth of about 2,000 m, regardless of the rock lithology. At this depth, pressures can be in excess of hydrostatic pressure. The ratio between the fluid pressure and the lithostatic pressure at this depth (which always lies between 0.41 for hydrostatic pressure and 1 for lithostatic pressure), ranges between 0.6 and 0.7 and even up to 0.9 in some cases (Jones, 1969). Generally, overpressured zones occur in undercompacted zones where burial rates are high, inhibiting fluid escape. Near these zones, which are generally 400 to 1,000 m thick, the pressure gradient changes suddenly until it reaches 0.15 to 0.20 bars/m. The pressure gradient between two undercompacted zones depends upon the porosity and permeability of the rocks. Therefore, depending on the local environment and the diagenetic evolution, the pressure gradient may change from one area to another, and with time.

In the center of the Franceville basin, if it is assumed that the stratigraphic column (after basin uplift) consists of 1,000 m of FA, 1,000 m of FB, and 1,000 m of the upper FC-FD-FE formations, then the vertical fluid pressure gradient can be estimated, as shown in Figure 20. The presence of an undercompacted zone at the bottom of the FB formation (1,500-2,000 m deep) is assumed. Figure 20 shows that if the overpressured fluid at point A (2,000 m deep) reaches the upper levels following the hydrostatic gradient pathway, the hydrofracture pressure gradient is intersected at point B, 750 m above point A. This suggests that if an undercompacted zone is connected by a fault or an aquifer to a zone located 750 m stratigraphically higher and capped by impermeable layers such as the FB black shales, then hydrofracturing may occur. Hence, this process is possible in the Franceville basin, where normal faults may have throws up to 1,000 m, putting the FA reservoir in tectonic contact with the FB impermeable level.

The FA sandstones consist of numerous layers with variable lithology acting as fluid drains with variable permeabilities. Since the tensile strength of the sandstones is lower perpendicular to the bedding than



FIG. 20. Pressure gradient in the Franceville basin: hydro-fracturing conditions.

parallel to the bedding (Gratier, 1984), hydrofracturing mostly occurs parallel to the bedding. However, in flat-lying sediments, where hydrofracturing is parallel to the bedding, the fluid pressure in a fracture must be greater than the lithostatic pressure. This is possible when the fluid pressure increases very quickly and/or during a compressive tectonic phase. For instance, during shearing the maximal stress tends to be horizontal (Hubbert and Willis, 1957).

This model explains the systematic association of hydraulic fracturing and the ore deposits, which are always in the highest levels in the basin and associated with tectonic structures where shear deformations are important. The contact zone between the FB pelites and the FA sandstones is also highly hydrofractured and mineralized, because the FB pelites form an impermeable cap which traps the overpressured fluids coming from deeper, undercompacted zones. A similar process has been described by Phillips (1972) for mineralization occurring in normal faults and breccia zones affecting Ordovician and Silurian sediments in central Wales.

Carbon isotope data

It has been seen that uranium mineralization is related to organic matter which may correspond to the reduction potential permitting U^{+6} to be reduced to U^{+4} , resulting in uranium precipitation. However, uranium is also related to calcite and we know that carbonate cements are important in the lower FA formation which contains the assumed source rocks for uranium (radioactive conglomerates) and in which sandstone is mostly oxidized. Therefore it is assumed that uranium mineralization occurred where two types of fluids mixed: a reduced fluid related to the oil migration from the FB black shales to the FA reservoirs and petroleum traps, and an oxidized uraniumbearing fluid coming from the FA formation. Carbon isotope analyses were performed in order to test this

hypothesis. Analyses were made on organic matter coming from all stratigraphic levels of the Francevillian column and corresponding to the various types: barren and mineralized organic matter and migrated or sedimentary organic matter. Carbon isotope analvses were also made on mineralized and barren carbonates of the FA formation and on dolomite layers of the FB-FC formations. All the results are given in Table 4 using the δ notation in units per mil relative to the PDB standard and with a precision of 0.5 per mil. Data synthesized on the histogram of Figure 21 show clearly that values of $\delta^{13}C_{PDB}$ for calcites in mineralized sandstone range between those of the organic matter, whatever its type and barren carbonates. Such a result agrees with the assumption that carbon of the calcite in mineralized sandstone came from organic matter (with depleted δ^{13} C) and from the uraniumbearing fluids migrating into the FA sandstone reservoir (with enriched δ^{13} C).

Conclusions

During the history of the Franceville basin, three main periods may be distinguished: sedimentation, burial, and uplift. Each of these periods has important effects for uranium mineralization in the Franceville basin.

Sedimentation period: Uranium and oil source-rock occurrences

The sedimentation of the FA and FB Series took place about 2,100 Ma. The Francevillian basin was first filled by FA fluviatile clastic sediments such as

Sample no.	Formation	$\delta^{13}C_{\text{organic}}$	Sample no.	Formation	$\delta^{13}C_{organic}$
A Organia mattar			SE0 Al	Upper FA	
A. Organic matter			BA9-1	Upper FA	-22.00
Dolomite layers			BA2-2	Upper FA	-24.90
9	FB	-25.80	Oklov = -290	Upper FA	-21.80
29	FB-C	-37.70	$KA27^1$	Upper FA	-26.45
30	FB-C	-39.50		opper 1 M	20.40
Black shales with high car	bonate content		Reactors		
10	Lower FB	-33.10	GL 1009 ²	Upper FA	-26.90
19	Upper FB	-24.50	Oklo 22812	Upper FA	-20.80
13	Upper FB	-29.90	GL 1022 ²	Upper FA	-24.00
20	Upper FB	-34.30	GL 222 ²	Upper FA	-25.40
17	Upper FB	-36.40	GL 2556 ²	Upper FA	-24.30
15	Upper FB	-34.00	GL 522 ²	Upper FA	-22.70
16	Upper FB	-34.70			
14	Upper FB	-25.70			
21	Upper FB	-35.80	Sample no.	Formation	$\delta^{13}C_{mineralized}$
22	Upper FB	-38.20	r		mineralized
18	Upper FB	-34.30	B . Carbonates		
Black shales			Dolomite lavers		
11	Middle FB	-27.10	9	FB	3.50
23	Upper FB	-36.00	28	FB-FC	4.70
24	Upper FB	-37.50	29	FB-FC	6.30
OK110 ¹	FBla	-23.20	30	FB-FC	2.60
BA30bis1	FB1b	-22.60			
Al ¹	FB2	-30.30	Calcite in barren sandstor	ne	
31	FB-C	-36.90	1	FA	-2.80
32	FC	-46.20	_		
Al ¹	FC	-44.10	Dolomite cement		
$1V^1$	FC	-44.10	2	FA	2.10
$1V^1$	FD	-45.30	3	FA	2.40
Bitumen				_	
26	FB-c	-42.20	Calcite in mineralized sandstone		
27	FB-C	-45.40	OK 75, 373.4 m ³	Upper FA	-15.04
			OK 72, 371.8 m ³	Upper FA	-7.73
Mineralized FA sandstone	s		OK 72, 363.4 m ³	Upper FA	-15.94
BA30bis ¹	Upper FA	-25.30	OK 99, 408.1 m ³	Upper FA	-13.75
BA11'	Upper FA	-27.20	KN 289 m ^o	Upper FA	-12.22

TABLE 4. δ^{13} C (‰) of organic matter (A) and carbonates (B)

Analyses without any mark were made at the Max-Planck Institute, Mainz

¹ Analyses made at the Société Nationale Elf Aquitaine at Pau

² Analyses made at the Federal Institute for Geosciences, Hannover

³ Analyses made at the Pennsylvania State University



FIG. 21. Carbon isotope compositions of the various organic matters and carbonates in the Franceville basin.

conglomerates and sandstones. We suggest that these conglomerates are the source rocks for uranium. We have seen that these rocks are radioactive and red colored. Table 5 shows that these conglomerates have the highest uranium content relative to other sedimentary units of the Francevillian Series which are known only in reduced state. Furthermore, uranium in these conglomerates is mainly contained in uraniferous thorites appearing at the present time as altered minerals which have been thus able to release uranium. Assuming that all the economic uranium ore deposits of the Franceville basin contain 40,000 tons of uranium—an area of 4×10^3 km² for the Franceville basin surface—and that oxidized fluids leached 4 ppm of uranium migrating through radioactive conglomerates, one may calculate that only 1 m of these conglomerates is needed to produce the uranium quantity of the Franceville basin deposits. It therefore seems possible that such rocks are sources of the uranium.

These fluviatile sediments were overlain by deltaic and marine deposits. Marine transgression came from the southeastern part of the basin and progressed toward the northwest, creating areas within the basin with various thicknesses of fluviatile and deltaic sediments. After the FA sedimentation, a collapse event occurred in the Franceville basin allowing marine transgression and deposition of thick FB black shale.

The uranium ore deposits occur in FA sandstones deposited in a deltaic environment. This sedimentary environment favors the accumulation of oil because the rock reservoirs are stratigraphically adjacent to the oil source rocks. In the Franceville basin, the FB black shales appear to be the main source rocks for petroleum trapped in the FA sandstone reservoirs (plain deltaic sandstones). Thus the main uranium ore deposits are located in areas where the deltaic plain sediments (reservoir rocks) are close to the marine source rocks, e.g., in the northwestern edge of the basin where the deltaic plain sediments are very thin and overlain by thick FB black shales. In the central part of the basin the deltaic sediments are thicker and prodeltaic sediments are well developed. Therefore, the FB oil source rocks are far away from the good reservoir rocks and the uranium deposits are less important. In this central part of the Franceville basin, oil can occur in the deltaic plain sediments only when tectonic faulting brings the sediments into contact with the FB black shales. The thickness of the fluviatile sediments can also control the uranium deposits sedimentologically. At the edge of the basin, where the uranium deposits are the most important, the fluviatile deposits and the radioactive conglomerates are thicker (500-1,000 m) than in the central part of the basin (100-400 m).

Burial of the Franceville Series: Oil generation and uranium mobility

During the early phase of burial, the FA sandstones with no organic matter (fluviatile sandstone and sandstone deposited in the deltaic plain) contained oxidizing fluids, which probably consisted of connate water. These fluids produced the red-colored sediments and probably bore uranium. As the oxidized fluids began to migrate, the complete Franceville Series and especially the FB black shales reached P-T conditions of the oil window. The resulting petroleum migrated into the FA sandstones when silicification began, as is illustrated by inclusions of organic matter in quartz overgrowths. Petroleum migration also produced the reduction fronts with green-colored sediments. By this time the upper FA formation was buried to a depth of about 4,000 m as recorded by fluid inclusions in diagenetic minerals such as quartz overgrowths and dolomite cements of sandstones. The temperature reached at this depth was about 180°C; maturation temperature beyond the oil window was not attained and oil was still mobile.

 TABLE 5.
 Total Uranium Content in the Formations of the Franceville Basin

		Total U (ppm)			
Formation	Facies	Avg	Range	n	
FD	Black shales	6.90	2-10	15	
FC	Black shales	5.20	2 - 9	5	
FC	Chert and dolomite	2.60	1 - 9	11	
FB 2	Black shales	1.90	1 - 2.5	7	
FB 1	Black shales	3.50	1 - 15	48	
FB 1	Pelites	1.9	1 - 10	10	
FA	Sandstones	4.98	1 - 22	48	
FA	Conglomerates	9.60	2 - 25	11	
Basement	Gneiss and schists	2.64	0.3-9	27	

Uplift of the Franceville basin and uranium mineralization event

The major thermic event recorded by the diagenetic clay minerals of the Franceville basin is temporally associated with the third tectono-metamorphic phase affecting the mobile zone of Ogooué. It is thus possible that this tectonic event resulted also in the uplift of the Franceville basin, ending its burial stage. Uplift of the basin is recorded in fluid inclusions which are posterior to the siliceous diagenesis. Microthermometric data of fluid inclusions in calcite and barite minerals related to the uranium mineralization suggest that temperature decreased to about 140°C, corresponding to an uplift of about 1,000 m. The resulting decrease of pressure and the occurrence of undercompacted zones in the FB black shale played a major role in the hydrofracturing process which took place where sandstone reservoirs were capped by the impermeable FB sediments. This hydrofracturing created the secondary porosity and permeability in previously well-silicified sandstones, allowing migration of hydrocarbons and uranium-bearing fluids. We suggest that uranium mineralization took place during this uplift stage, in structural high levels of the basin which are also good traps for hydrocarbons (Fig. 22). Uranium mineralization occurred in these areas when reduced fluids related to hydrocarbon migrations met the oxidized uranium-bearing fluids migrating through the FA sandstone reservoir. Major tectonic structures may represent good pathways for these fluids, whose origin is still uncertain.

Assuming 10 percent porosity for the sandstones, 10 ppb of dissolved uranium in water, and FA sandstones 1 km thick, one can calculate that 40,000 tons of uranium corresponds to 10 times the water volume located in the FA sandstones of the Franceville basin. This very simple computation shows that significant water circulation in sandstone is needed in order to explain such uranium deposits, and we assume that this circulation was easier during the uplift stage of



FIG. 22. Main pathways for the fluids flowing in the Franceville basin during the uranium mineralization stage.

the basin when fracturing was important. On the other hand, water should stay oxidized in order to mobilize uranium. This condition allows us to assume that uranium-bearing fluids should stay oxygenated and thus the more probable type for these fluids seems to be meteoric fluids. Such an origin for the uranium-bearing fluid seems most possible during an uplift stage of the basin. This fluid should have migrated through radioactive conglomerates and FA sandstones with sulfate and dolomite cements resulting in its uranium enrichment and high salinity. Such a uranium-bearing fluid with high salinity is also described for fluids associated with uranium mineralization of unconformity-type deposits in Canada and Australia (Ypma and Fuzikawa, 1980; Pagel, 1983) where ores with high uranium contents are found.

Acknowledgments

This study would not have been possible without the logistic and financial support given by the Compagnie des Mines d'Uranium de Franceville. We wish to thank particularly J. P. Pfiffelmann, J. F. Chauvet, and J. L. Dronne and all geologists in Gabon for their support. The Commissariat à l'Energie Atomique (France) also supported this study. Thanks are also due to H. Ohmoto, K. Geer, S. Poulson, and N. Clauer for the helpful comments and suggestions and to Schidlowsky for carbon isotope analyses.

June 26, 1987; March 13, 1989

REFERENCES

- Bohomme, M., Gauthier-Lafaye, F., and Weber, F., 1982, An exemple of lower Proterozoic sediments: The Francevillian in Gabon: Precambrian Research, v. 18, p. 87–102.
- Cortial, F., 1985, Les bitumes du Francevillien (Protérozoique inférieur du Gabon 2000 m.a) et leurs kérogènes. Relations avec les minéralisations uranifères: Unpub. 3rd cycle thesis, Strasbourg, Univ. Louis Pasteur, 183 p.
- Cortial, F., Gauthier-Lafaye, F., Weber, F., and Oberlin, O., 1988, Caractérisation des matières carbonées associées aux gites d'uranium du Francevillien au Gabon (Proterozoique inferieur, 2000 M.a). Implications métallogéniques: France, Bur. Recherches Géol. Min. Doc. 158, p. 415-444.
- Devillers, C., and Menes, J., 1978, Contribution du plomb et du thorium à l'histoire des réacteurs d'Oklo, *in* The natural fission reactors: Vienna, Internat. Atomic Energy Agency, p. 495–509.
- Dunoyer de Segonzac, G., 1969, Les minéraux argileux dans la diagenèse. Passage au métamorphisme: Strasbourg, Service Carte Géol. Alsace—Lorraine, Mem. 29, 320 p.
- Durand, B., and Oudin, J. L., 1979, Exemple de migration des hydrocarbures dans une série deltaique: Le delta de la Mahakam, Indonésie, *in* World Petroleum Cong. 10th Bucarest, 1979, Proc. PD.1, p. 1–90.
- Ellis, A. J., 1970, Quantitative interpretation of characteristics of hydrothermal systems: Geothermics Spec. Issue 2, p. 516-528.
- Evans, C. R., McIvor, D. C., and Magara, K., 1975, Organic matter, compaction history and hydrocarbur occurrence, Mackensie delta, *in* World Petroleum Cong., 9th, Tokyo, 1975, Proc., v. 2, p. 149-157.
- Gancarz, A. J., 1978, U-Pb age $(2,05 \times 109 \text{ years})$ of the Oklo uranium deposit, *in* The natural fission reactors: Vienna, Internat. Atomic Energy Agency, p. 513–520.

- Gauthier-Lafaye, F., 1986, Les gisements d'uranium du Gabon et les réacteurs d'Oklo. Modèle métallogénique de gites à fortes teneurs du Protérozoique inférieur: Sci. Géol. Mém., v. 78, 206 p.
- Gauthier-Lafaye, F., and Weber, F., 1981, Les concentrations uraniféres du Gabon: Leur association avec des gites à hydrocarbures fossiles du Protérozoique inférieur: Acad. Sci. [Paris], Comptes Rendus v. 292, p. 69–74.
- 1984, Effets des déformations postérieures à une diagenèse siliceuse d'enfouissement sur le comportement microthermométrique des inclusions fluides: Cas du réservoir gréseux uranifère du Francevillien (Gabon): Acad. Sci. [Paris] Comptes Rendus, v. 299, p. 555–560.
- 1988, Le mécanisme de la fracturation hydraulique dans la métallogenèse de l'uranium en série sédimentaire. L'exemple du gisement d'Oklo, Gabon, *in* Les gisements métallifères dans leur contexte géologique: France, Bur. Recherche Géol. Min. Doc. 158, p. 445–465.
- Gratier, J. P., 1984, La déformation des roches par dissolutioncristallisation: Unpub. Sci. thesis Univ. Grenoble, 315 p.
- Hagemann, R., Lucas, M., Nief, G., and Roth, E., 1974, Mesures isotopiques du rubidium et du strontium et essais de mesure de l'age de la mineralisation de l'uranium du réacteur naturel d'Oklo: Earth Planet. Sci. Letters, v. 23, p. 170–176.
- Haubensack, C., 1981, Environnement des grés Protérozoiques et des indices uranifères du secteur Kiene dans le basin de Franceville (République Gabonaise). Aspect sédimentologiques et géochimiques: Unpub. 3rd cycle thesis, Strasbourg, Univ. Louis Pasteur, 105 p.
- Hilbert, R., 1979, PVT-Daten von Wasser und Wassrigen Natriumchlorid-Losungen bis 873 k, 4000 Bar und 25 Gewichtsprozent NaCl: Freiburg, Hochschul Verlag-Freiburg, 213 p.
- Holliger, P., Gauthier-Lafaye, F., Mougel, X., and Weber, F., 1989, Uranium deposition and Pb loss ages at Oklo as determined by ion microprobe [abs.]: European Union of Geosciences, Terra, Abstracts, v. 1, no. 1, p. 344. March 1989.
- Hubbert, M. K., and Willis, D. G., 1957, Mechanics of hydraulic fracturing: Am. Inst. Mech. Engineers Trans., v. 210, p. 153– 168.
- Jones, P. H., 1969, Hydrodynamics of geopressure in the northern Gulf of Mexico basin: Jour. Petroleum Technology, v. 21, p. 803-810.
- Lagache, M., and Weisbrod, A., 1977, The system: Two alkali feldspars-KCl-NaCl-H₂O at moderate to high temperatures and low pressures: Contr. Mineralogy Petrology, v. 62, p. 77–101.
- Lancelot, J. R., Vitrac, A., and Allegre, C. J., 1975, The Oklo natural reactor: Age and evolution studies by U-Pb and Rb-Sr systematics: Earth Planet. Sci. Letters, v. 25, p. 189–196.
- Magara, K., 1978, Compaction and fluid migration: Devel. Petroleum Sci., v. 9, 319 p.
- 1981, Mechanisms of natural fracturing in a sedimentary basin: Am. Assoc. Petroleum Geologist Bull., v. 65, p. 124– 132.
- Oberlin, A., 1977, La microscopie electronique moderne comme moyen d'analyse: Analusis, v. 5, Part 2, p. 85-101.
- Oberlin, A., Boulmier, J. L., and Villey, M., 1980, Electron microscopic study of kerogen microtexture, *in* Durand, B., ed., Kerogen: Paris, Technip, p. 191-241.

- Openshaw, R., Pagel, M., and Poty, B., 1978, Phases fluides contemporaines de la diagenèse des grés, des mouvements tectoniques et du fonctionnement des réacteurs nucléaires d'Oklo, *in* The natural fission reactors: Vienna, Internat. Atomic Energy Agency, p. 267–293.
- Pagel, M., 1983, Les modèles proposés pour la genèse des gisements d'uranium liés spatialement aux discordances protérozoiques: Univ. Nancy, Geologie Géochimie Uranium Mém. 1, p. 355–380.
- Perrodon, A., 1980, Géodynamic pétroliere: Centre Recherche Explor. Prod. Elf-Aquitaine, Mém. v. 2, 381 p.
- Phillips, W. J., 1972, Hydraulic fracturing and mineralization: Geol. Soc. London. Jour., v. 128, p. 337-359.
- Poty, B., Stadler, H. A., and Weisbrod, A. M., 1974, Fluid inclusions studies in quartz from fissures of western and central Alps: Suisse Minéralogie Pétrographie Bull., v. 54, p. 717–752.
- Riedel, W., 1929, Zur Mechanik geologischen Brucherscheinungen: Zentralblatt fur Mineralogie, Geologie, und Palaontologie, p. 354–368.
- Steiger, R. H., and Jäger, E., 1977, Subcommission on geochronology: Convention on the use of decay constants in geo- and cosmochronology: Earth Planet. Sci. Letters, v. 36, p. 359– 362.
- Tuttle, O. F., 1949, Structural petrology of planes of liquid inclusions: Jour. Geology, v. 57, p. 331–356.
- Vachette, M., 1964, Ages radiométriques des formations cristallines d'Afrique Equatoriale (Gabon, Republique Centrafricaine, Tchad, Moyen Congo): Univ. Clermont-Ferrand, Ann. Faculté Sci., v. 25, Géol. Min. no. 8, p. 31–38.
- Vanderbroucke, M., Rouzaud, J. N., and Oberlin, A., 1978, Etude géochimique de la matière organique insoluble (kerogene) du minerai uranifère d'Oklo et des schistes apparentès du Francevillien, *in* The natural fission reactors: Vienna, Internat. Atomic Energy Agency, p. 307–332.
- Weber, F., 1968, Une série precambrienne du Gabon: Le Francevillien. Sédimentologie, géochimie, relations avec les gites minéraux associés: Strasbourg, Service Cartes Géol. Alsace-Lorraine Mém. 28, 328 p.
- Weber, F., Schidlowski, M., Arneth, J. D., and Gauthier-Lafaye, F., 1983, Carbon isotope geochemistry of the lower Proterozoic Francevillian series of Gabon (Africa) [abs.]: Terra Cognita, v. 3, p. 220.
- White, D. E., 1965, Saline waters of sedimentary rocks: Am. Assoc. Petroleum Geologists Mem. 4, p. 342-366.
- 1970, Geochemistry applied to the discovery, evaluation and exploitation of geothermal energy resources: Geothermic Spec. Issue 2, v. 1, p. 58–80.
- Wilcox, R. E., Hardin, T. D., and Seley, D. R., 1973, Basic wrench tectonics: Am. Assoc. Petroleum Geologists Bull., v. 57, p. 74– 96.
- Winkler, H. G. F., 1976, Petrogenesis of metamorphic rocks: New York, Heidelberg, Berlin, Springer-Verlag, 334 p.
- Ypma, P. J., and Fuzikawa, K., 1980, Fluid inclusion and oxygen isotope studies of the Nabarlek and Jabiluka uranium deposits, Northern Territory, Australia, *in* Ferguson, J., and Goleby, A. B., eds., Proceedings of International Uranium Symposium on Pine Creek Geosyncline, Sydney, June 4–8, 1979: Vienna, Internat. Atomic Energy Agency, p. 375–395.