UNITED STATES DEPARTMENT OF THE INTERIOR

GEOLOGICAL SURVEY

PRELIMINARY RESULTS FROM

MICROANALYSES OF ORGANIC MATTER IN THE

LOWER PROTEROZOIC URANIUM ORES AT OKLO IN GABON

By

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INTRODUCTION

The uranium ore deposits of the approximately 2×10^9 years old Lower Proterozoic Francevillian Series at Oklo in the Republic of Gabon generally show an intimate association with organic matter. Minimally metamorphosed non-uraniferous rocks surrounding the ore deposits also are rich in organic matter. The organic matter in the Francevillian Series has been noted by various investigators. The first comprehensive investigation of kerogen in some of the Oklo uranium ores was reported by Cassou and others (1975). The association of the main uranium ore zones with bitumen, which has migrated during more than one geochronological episode and later resolidified, was described by Gauthier-Lafaye (1986). In addition to bitumen, syngenetic kerogen is also present in the Oklo uranium ores and in their environs. Intimate association of uranium mineralization with organic matter is known in various deposits, for example in the Archean Witwatersrand carbon seams in South Africa (Zumberge and others, 1978); in the Proterozoic Claude uranium ores at Cluff Lake in Canada (Leventhal and others, 1987) and in the Proterozoic uraniferous paleoplacers at Elliot Lake, Canada (Willingham and others, 1985; Nagy and Mossman, 1988, 1989). Uranium organic associations are also common in the Phanerozoic. However, the kerogenous uranium ores at Oklo are of particular interest to basic science, because part of them went critical approximately 2×10^9 years ago. Fission reaction originated in the high grade ores where water acted as the moderator; the natural fission reactors shut down when the contained water was expelled (Naudet, 1978). The temperature of the reactors during the fission reaction was estimated to be greater than approximately 400°C, (Holliger and others, 1978; Openshaw and others, 1978; Vidale, 1978). It has been estimated that nuclear criticality was sustained at Oklo for more than a hundred thousand years (Loss and others, 1988). The Oklo uranium ores contain the only non-anthropogenic fission reactors, based on presently available compelling analytical evidence.

Parts of the uranium ores at Oklo went critical because of fortuitous circumstances, which, however, need not be unique. The causes of criticality include the concentration of 25-60 percent uranium in the form of uraninite (Weber and others, 1975; Geffroy, 1975) in small, compact masses along with a suitable neutron moderator, in this case water and organic matter. Reactor No. 9 is a disk-shaped mass, approximately 6 m in the direction of its dip, 11 m along the strike and about 0.5 m thick. Such a mass was critical approximately 2×10^9 years ago, when the abundance of ^{235}U isotope was about 3.8 percent of all uranium. Water was present to act as the moderator. Also, there is no evidence that significant quantities of neutron absorbers, such elements as lithium, boron and a number of the rare earths, were present in the ore prior to criticality. These elements, called neutron poisons, have large neutron-capture cross sections and can prevent criticality. Fission in the natural reactors involved several processes. The major process was the thermal neutron induced fission of ^{235}U, and others were the thermal neutron induced fission of ^{239}Pu and the high energy neutron fission of ^{238}U. These processes produced fissiogenic nuclides; the proportions of each nuclide is different for each of the fission reactions. A number of the fission products have not been retained in reactor zone No. 9. This has been noted by Curtis and others (1989) who also pointed out that some fission products have been lost and others gained in samples taken along a cross section of this natural reactor zone.

The uranium ores at Oklo occur in the 2.15×10^9 year old (Bonhomme and others, 1982) Francevillian Series, which contains sandstones, shales,

conglomerates, cherts, dolomites and pyroclastic sediments. Virtually unmetamorphosed sediments are located at the eastern part of the Francevillian basin, where the Series is less than 1,000 meters thick (Gauthier-Lafaye and Weber, 1989). The Francevillian Series has been subdivided by Weber (1968) into five formations, FA at the base and FD and FE at the upper portions of the column. Uranium mineralization occurs in a thin layer designated as C1 in the FA formation. The complex structural and stratigraphic relationships defining the spatial relationships of the uranium ore bodies at Oklo have been described by Gauthier-Lafaye and others (1975) and Gauthier-Lafaye (1978). Figures 1 and 2 show the stratigraphic relationships of the ore bodies in the C1 layer and point out the deltaic depositional environment. The accretion of the uranium ore bodies at Oklo appears to be related to a deltaic depositional environment. Cowan (1976) suggested that syngenetic uranium dispersed in extensive pre-Francevillian igneous rocks, was released by weathering and subsequently accumulated in streambeds under reducing conditions. With the onset of an oxidizing environment, uranium became soluble and was carried downstream into the delta of the river system. Uranium became concentrated in the delta, through incorporation in decayed organic matter, which was perhaps, similar in degree of decomposition to organic matter found in the delta plain marshes of the Holocene, prograded Mississippi delta. In such organic-rich environments oxygen deficiency may develop, reducing the oxidized uranium species, causing their precipitation from solution and therefore their immobilization. After burial, and subsequent diagenesis, uranium deposited in such environments may lead to the formation of organic-rich uranium ore deposits, if reducing conditions prevail in the host rocks.

The validity of this model hinges on a number of factors, foremost the oxidizing environment necessary to solubilize uranium for long distance fluvial transport. This concept gained support recently from the work of Holland and Beukes (1989), who reported evidence for a substantial rise in the level of atmospheric oxygen between 2.2 and 1.9 x 10^9 yrs ago. Based on mineralogical and geochemical studies of samples collected in South Africa, P_{O_2} was 25 percent of PAL (present atmospheric level) if P_{CO_2} was >1 PAL during this time interval. Because such atmospheric composition could not be restricted to South Africa, the Oklo locality in Central Africa must have also been affected by it at the time of the accumulation of uranium, which later formed the Oklo deposits. Openshaw and others (1978) observed organic matter in fluid inclusions in quartz veins and noted that CO_2 does not seem to be a significant constituent and that only Cl^{-1} and SO_4^{-2} ions are the major components of the aqueous solutions. They suggested that uranium was probably transported in the form of uranyl sulfate. Because the fluid inclusion studies revealed an elevated temperature history of the host minerals, the question arises whether certain post-depositional and therefore later events, possibly prior to or occurring during criticality, or even subsequent to it, may have been oxidizing. Thin section studies of Francevillian Series rocks reveal a series of different generations of veins, some filled with oxidized and others with reduced mineral constituents deposited by percolating solutions. Gauthier-Lafaye and Weber (1989) noted that oxidized solutions developed during early diagenesis, followed by fluids which were reducing. The Oklo uranium ores and surrounding non-uraniferous strata contain an abundance of veins, generally sub-millimeter size in their width.

The organic carbon content (TOC) of the Francevillian sedimentary rocks varies between 0.5 and 15 percent (Cortial and others, 1989). Simpson and Bowles (1978) noted that the organic matter occurs in the form of both

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syngenetic kerogen and mobilized organic matter. Electron microscopic studies coupled with the dark field diffraction method indicate that the majority of organic matter in black shales contain moieties of varying sizes of molecular organization (Oberlin, 1977). Organic matter in the uranium ores and in the reactor have similar matrices, as will be shown below. The spatial distribution of organic matter varies among finely dispersed, minute particles, larger (up to 2-3 mm) globules and pore and vein fillings, some showing effects of oxidation (Gauthier-Lafaye and Weber, 1989). Some of the organic matter in veins was not oxidized and attests to the effects of reducing environments during diagenesis. Gauthier-Lafaye and Weber (1987) proposed that in the FA formation there are solidified bitumens generated by two different diagenetic episodes. The remobilized and globular organic matter is of interest because of its effects on uranium minerals and conversly by the effects of the uranium ores, such as radiation damage, on the organic matter. Remobilization of syngenetic kerogen by hydrothermal solutions to form globular and vein filling bitumens, caused by processes analogous to hydrous pyrolysis, has been proposed by Nagy and Mossman (1988, 1989) at the Elliot Lake uranium deposits that are localy rich in organic matter. Kerogen mobilization also occurred at Cluff Lake (Leventhal and others, 1987) and, to a smaller extent, at the Witwatersrand uraniferous deposits (Willingham and others, 1985) and elsewhere. The veins into which the viscous bitumen was injected, and in which it resolidified, evolved in fractures created by fluids under high pressures (Gauthier-Lafaye, 1986). Because the tensile strength of fine grained clastic rocks is lower perpendicular to the bedding plane than parallel to it, fracturing occurs mainly parallel to the bedding. Tectonic processes causing shearing set up maximum stresses which tend to be horizontal (Hubbert and Willis, 1957). Many, but not all, of the veins filled with organic matter occur in such spatial orientation at Oklo. The expulsion of water under high pressure and temperature contributed to extensive hydraulic fracturing in the reactor zones, surrounding deposits and adjacent rocks.

The properties of fluids expelled, or mobilized through the reactor zone, including their inorganic and organic components, the role of minerals and the pressure, temperature and compositions affected by the nuclear reactions, can be used elucidate the transport of fissiogenic elements and decay products, as well as the geochemistry of the Francevillian Series. The present study is focused on the geochemical effects of mobilized fluids on the organic matter, Oklo uranium ores and fission products.

SAMPLES

The organic matter in six samples from the Francevillian basin in Gabon were analyzed in relation to uranium content and the natural fission reactor at Oklo. The samples are designated:
KP 3, 4.60 m, reactor No. 9;
Crochon. 336. Oklo;
Boy Niv 235, RO 33.FC;
Boy Niv 235, RO 33.5;
LN 12, 113 m 50;
FB black shale;
These sample designations will be referred to in an abbreviated form as:
KP 3; Cro 336; Boy 33.FC; Boy 33.5; LN 12, and FB black shale. Sample KP 3 is from a drill core through the natural reactor No. 9. It is a shaley sandstone with high uranium and organic matter contents. Cro 336, from the northern part of the Oklo deposit, is a sandstone with high uranium and organic matter
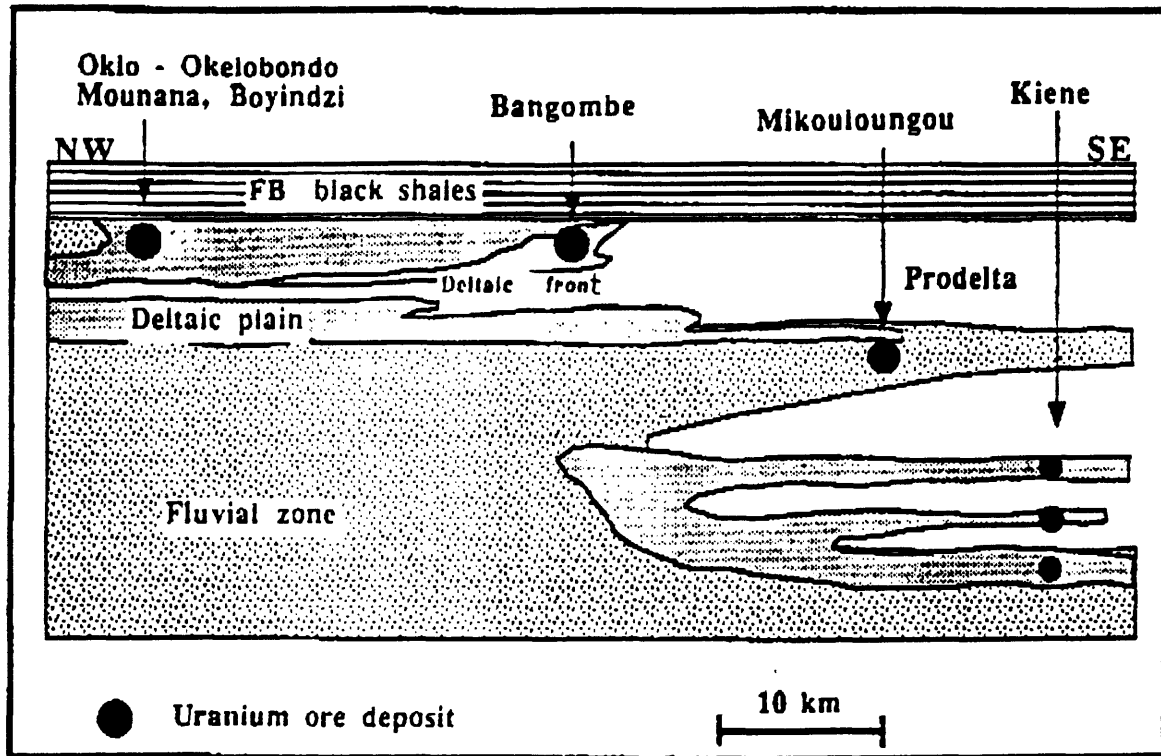
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Figure 1.   Paleogeographic reconstruction of the FA Formation in the
Franceville basin and location of the uranium ore deposits.
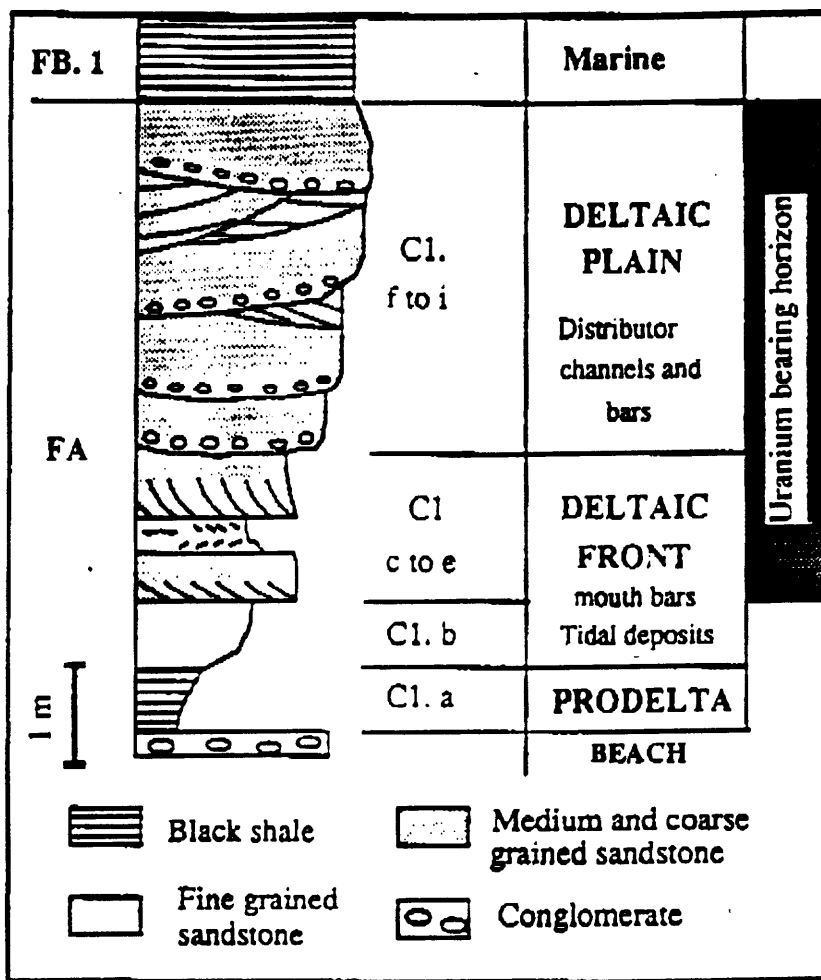
from Gauthier-Lafaye and Weber, 1989.

Figure 2.   Stratigraphic column and interpretation of the C1 layer
at the Oklo deposit.

From Gauthier-Lafaye and Weber, 1989.

contents. Boy 33.FC is a sandstone with high uranium and organic content; most of the organic matter occurs as flattened globules. This sample is from the Boyindzi deposit, located about 5 km from the Oklo reactor (fig. 1). Boy 33.5, from the same location and distance from the reactor as the previous sample, is a sandstone with globules of uraniferous organic matter. Its uranium content is lower than that of Boy 33.FC. The LN 12 sample consists of virtually pure, black and shiney organic matter. It is from the FB formation (fig. 2), district of Mounana, 25 km southeast from Oklo. It is a drill core sample from a layer or vein of organic matter probably lying parallel to the bedding planes. This sample is not mineralized with uranium. The FB black shale is an outcrop sample from near Franceville. This sample is 50 km southeast from the Oklo reactor and it is also not mineralized with uranium.

EXPERIMENTAL PROCEDURES

Scanning electron microscopy (SEM) was performed with an International Scientific Instruments SS-40* scanning electron microscope equipped with a Kevex system, which permitted semiquantitative energy dispersive analysis (EDS) of the elemental components of samples. Two samples were examined by SEM and analyzed by EDS. These were the un-mineralized and distant samples from the reactor, LN 12 and FB black shale. These samples were selected to examine possible regional metamorphic effects on Francevillian rocks and the morphological effects of diagenesis or metamorphism on their contained organic matter which was unaffected by the thermal and compositional overprint of processes originating from the natural reactors. The samples were polished with different grades of grinding powder; the final step consisted of polishing with 0.5 micrometer (micron) Al_2O_3 powder. The polished surfaces of the samples were coated with a 100 Å (10 nm) thick layer of carbon. Back-scattered scanning electron microscopic (BSEM) images were obtained and recorded on photomicrographs. This BSEM technique has been described by, for example, Pye and Krinsley (1983) and White and others (1984).

The primary analytical tool used in the molecular organic geochemical characterization for this study was pyrolysis-gas chromatography-mass spectrometry (py-gc-ms). The stepwise-pyrolysis-gas chromatography method is described in Leventhal (1981) and Leventhal and others, (1986, 1987) and is briefly summarized here. A small portion of sample (approximately 1-10 mg) is placed in a 2 mm x 2.3 cm quartz tube that is placed in the heating coil of the Chemical Data Systems Pyroprobe*. The sample is heated, in steps, for 10 seconds each at 450, 600, 750 and 900°C and pyrolysis products from each step are separated on a 15 m x 0.32 mm inside diameter (id) fused silica column with a 1.0 micron thick DB1 bonded liquid phase. The column was heated from 30°C to 260°C at 5°C/minute. The separated compounds were detected by a flame ionization detector.

For identification of non-n-alkane peaks a HP 5710A* gas chromatograph with a 25 m x 0.31 mm id fused silica capillary column with a 0.17 micron bonded of 5% phenyl methyl silicone and a helium flow of 1 cc/min was coupled to a Finnegan MAT model 700 ion trap detector operated in a dynamic mode. The column was heated from 90°C to 260°C at 4 or 8°C/ minute. Scanning rate for the detector was at 2 second intervals from m/z = 50 to 350. The ion trap detector was controlled by an IBM type XT* personal computer software and the data were stored and manipulated with the same system.

*Mention of brand name does not necessarily imply endorsement by U.S. Geological Survey.

Rock-Eval* pyrolysis was performed on samples and is described by Tissot and Welte (1978) using a Delsi model II with an organic carbon module. Briefly, Rock-Eval is a method for analysis of sedimentary organic matter that consists of heating of an aliquot (100 mg) of sample first isothermally at 250°C for 2 minutes and then from 250 to 600°C at 25°C/min and detecting the evolved materials. Volatile hydrocarbons (hc) released at temperature below 250°C are designated S_1 (mg hc/g). Pyrolysis products released above 250°C are designated S_2 (mg hc/g). The quantity S_2/%organic C is the hydrogen index (HI) that is correlative to the atomic H/C ratio. Thus a higher HI corresponds to a more organic hydrogen-rich or higher H/C containing material. Carbon dioxide released up to 390°C is called S_3 (mg CO_2/g); S_3/%org C is the oxygen index (OI) and is analogous to the atomic O/C ratio. A higher OI value corresponds to a more oxygen-rich organic material. The temperature at which the release of S_2 products is a maximum is called T_{max} and this value (not related to geologic temperatures) increases with thermal maturity. Kerogens with T_{max} below 420°C are thermally immature with respect to petroleum generation, whereas temperatures from 430 to about 455°C correspond to the petroleum generation zone. HI values of 800, 200, and 20 correspond to H/C (atomic) values of 1.6, 0.75 and 0.5 respectively. An OI value of 35 corresponds to an O/C (atomic) value of 0.18. Rock-Eval results can be displayed on a plot of HI vs OI, which is similar to a van Krevelen plot of H/C vs O/C (Tissot and Welte, 1978). However, Rock-Eval results must be interpreted with care and understanding of the samples and instrumental complexities (Leventhal, 1982).

Micro-focussed laser Raman spectroscopy was performed on a sample that was prepared by mounting a piece of the sample (1 mm^3 to 1 cm^3 in size) in epoxy and then polishing with 250, 450 and 600 grit silicon carbide paper with water as a lubricant. Other samples examined were flat surfaces of unpolished pieces of rock and some samples were organic-rich residues. The samples were viewed on an Olympus BH2* petrographic microscope with a 10x eyepiece and a 20x, 50x or 100x objective using a video monitor. The microscope optics were also used for the transmitting the incident laser beam to the sample and the Raman signal from the sample to the spectrometer. The laser was a Spectra Physics* model 2020, 25 watt argon laser using 544 nm light. The laser output was usually in the range of 60-90 milliwatts. The Raman spectroscopy was done on a Jobin Yvon* model U 1000 spectrometer manufactured by Instruments, S A (France). The slit of the monochrometer was 200 micrometers and the scan rate was either 1 or 2 sec in increments of 1 or 2 cm^{-1} from 1740 to 1060 cm^{-1}.

EXPERIMENTAL RESULTS

Backscattered scanning electron microscopic (BSEM) studies of the FB black shale revealed rounded or sub-rounded quartz grains, linear or occasionally twisted aggregates of layer lattice silicates and generally rounded and sub-rounded pyrite. The minerals were surrounded by a matrix of organic matter. The grain sizes of the minerals are small, approximately 0.5 to 10 microns, with a few pyrite and quartz grains as large as 50 to 100 microns, in certain areas only. The quartz occurred occasionally as <1 micron angular grains. The quartz often showed vacuoles or vesicles. The layer lattice silicates appear to be clay minerals; some of them as mixed layer aggregates, based on their Z contrast on the BSEM. Gauthier-Lafaye and Weber (1989) described illites in the Francevillian Series and noted that the structural parameters of these clay minerals are characteristic of diagenesis to minimal metamorphism. The rounded and sub-rounded pyrite often showed

hollow interiors, in which angular crystals were visible. These grains were probably authigenic pyrite of a younger paragenetic sequence. The FB black shale appears to be an organic rich, quartzose siltstone with clay and pyrite as prominent components. Its sedimentary texture is remarkably well preserved, attesting to minimal metamorphism characteristic of well preserved Proterozoic clastic sediments. BSEM examination of the LN 12 sample revealed an organic solid virtually free of mineral matter. However, other portions of this sample must contain a greater abundance of minerals, as shown by the TOC value (less than 80 percent) of the bulk sample. This organic matter is a brittle solid, showing conchoidal fractures induced during the grinding of its polished surface. This organic substance appears to be a resolidified and polymerized solid, which may have been emplaced in veins.

The results of the Rock-Eval analysis are given on table 1. An example of the pyrolysis-gc-ms for sample KP 3 is shown on figure 3.

Laser Raman spectroscopy is capable of distinguishing the different types of chemical bonds of carbon atoms and thus shows different spectra for organic carbon (carbon bonded to H), for amorphous carbon (non-crystallilne carbon), cryptocrystalline graphite (crystal domain of smaller than optical microscopic size) and well ordered (>1 micron) graphite. Figure 4a shows laser Raman spectra of 1 micron crystalline graphite that shows a single sharp absorption peak near 1600 cm^{-1} (Tunistra and Koenig, 1970). Figure 4b shows Raman spectra of graphite of smaller crystal dimensions that is less-well ordered (cryptocrystalline) than micron sized graphite but it is more mature than anthracite rank coal (Lespade and others, 1982). Figure 4c shows the Raman spectra of the coal series from sub-bituminous to anthracite rank (Beny-Bassez and others, 1981). Figure 4d shows a sample that is typical of the sediment-hosted disseminated gold deposit at Jerritt Canyon, Nevada (Leventhal and Hofstra, 1989) that has a crystal domain size of around 0.01 microns. Figure 5 shows the spectra from the unmineralized remote LN 12 sample and from the KP 3 mineralized sample from Oklo. We have found no effect on the Raman spectra by polishing or other aspects of sample preparation (see Pasteris, 1989, for discussion). The results are very similar, whether they are taken on a highly polished jasperoid, rock slabs polished with 600 grit silicon carbide paper under water, samples polished with 1 micron alumina paste, or a natural fracture surface, or samples within a jasperoid vein.

DISCUSSION AND CONCLUSIONS

All samples contain abundant organic matter, and the Rock-Eval pyrolysis results show that the organic matter is a highly thermally altered (over-mature relative to petroleum generation) kerogen. There are, however, differences between the nature of the organic constituents at various distances away from reactor No. 9. Surprisingly, the four uranium ore samples, including KP 3, have high oxygen index (OI) values, three of them have the higher hydrogen index (HI) values and these samples also show the lowest T_{max} values. Combined pyrolysis-gas chromatographic-mass spectrometric analysis of the organic matter in the six samples also show differences in molecular abundances (table 2). Only the remote samples (FB Black shale and LN 12) show traces of short chain n-alkanes. The sample from the reactor core No. 9 and both samples from Boyindzi (Boy 33.FC and Boy 33.5) gave more diverse aromatic pyrolysis products than the distant samples including naphthalene and mono- and dimethyl substituted naphthalenes. They also yield benzene, toluene, xylenes (C2-benzenes) and CH_4 and CS_2; these five moieties were also present in all samples.

SAMPLES FROM GABON

Samples	TOC	HI	OI	Tmax
Remote				
FB bl sh	10.3	4	4	541
LN 12	32.7	2	4	604
Boyindzi deposit				
BOY 33.5 lean	0.05	--	--	--
BOY 33.5 rich	57.01	14	17	489
BOY 33.FC	17.51	80	40	472
Oklo deposit				
Cro 366	44.1	2	49	541
KP 3	54.9	29	21	475

Table 1. Rock-Eval results. See text for explanation.

Chromatogram   D:\DATA\OKKP37   Acquired: May-10-1989   09:52:55
Comment: 8/MIN, 80-300
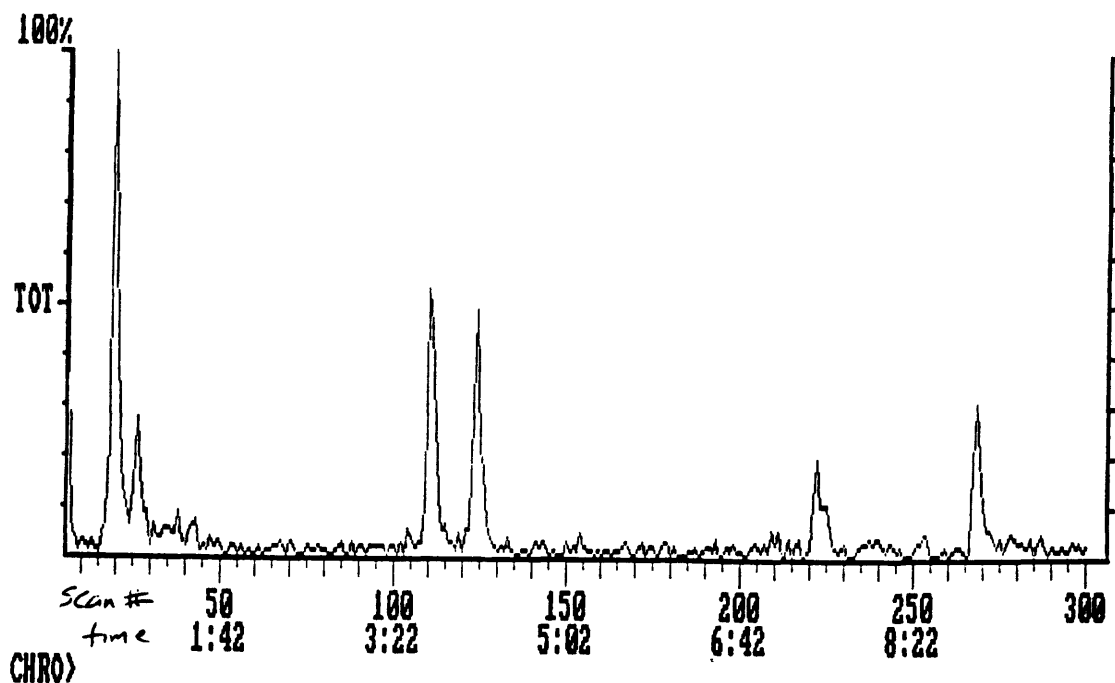Scan Range: 6 - 300   Scan: 6   Int = 594   @ 0:15 RIC: 100% =1684

Figure 3.

Results of the pyrolysis-gc-ms of the 750°C step of sample KP
Total ion current chromatogram shows peaks for non-n-alkane
pyrolysis products: S, toluene, C2-benzenes, methyl
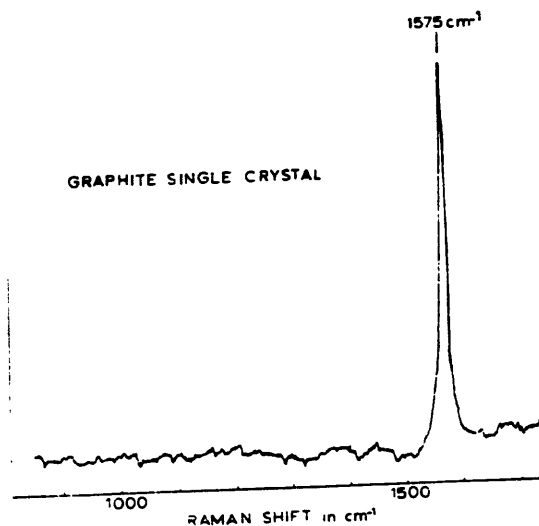naphthalenes, C2-naphthalenes and S

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Figure 4a. Raman spectra of well crystalline graphite (from Tunistra and Koenig, 1970).
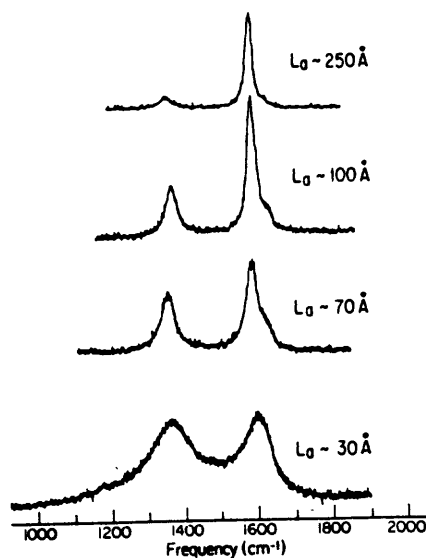

Figure 4b. Raman spectra of smaller crystal sizes of graphite/carbon (250 Å = 0.025 microns) (from Lespade and others, 1982).
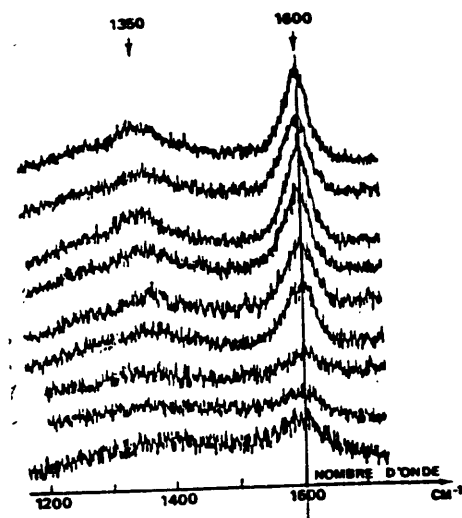
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Figure 4c. Raman spectra of the coal series, sub-bituminous on the bottom to anthracite rank on the top (from Beny-Bassez and others, 1981).
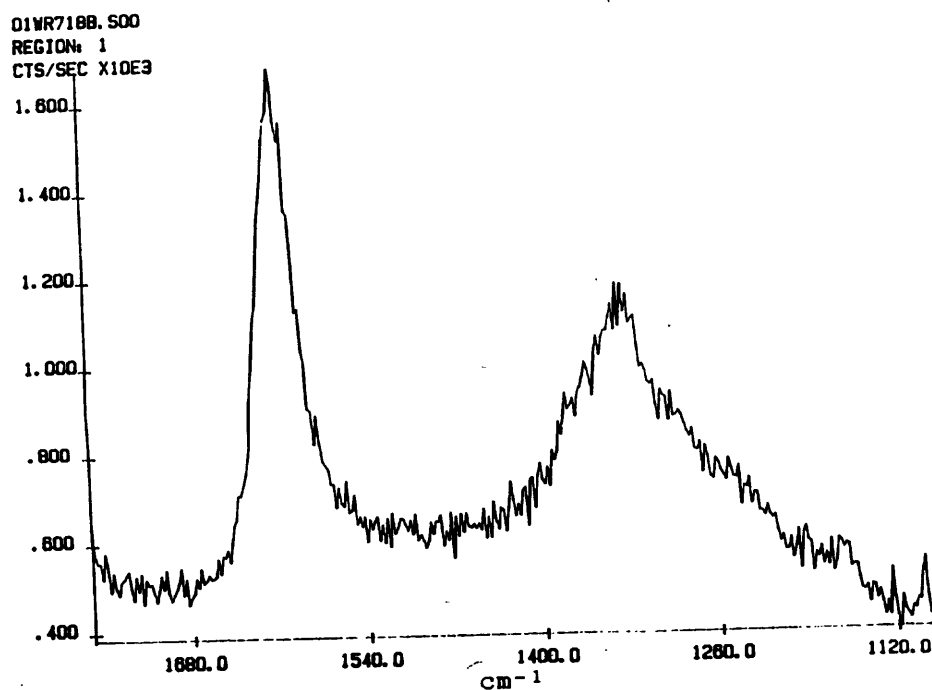

Figure 4d. Raman spectra of carbon from Nevada gold deposit. Note that the cm^{-1} scale is inverted, scanning was from higher to lower wavenumber.
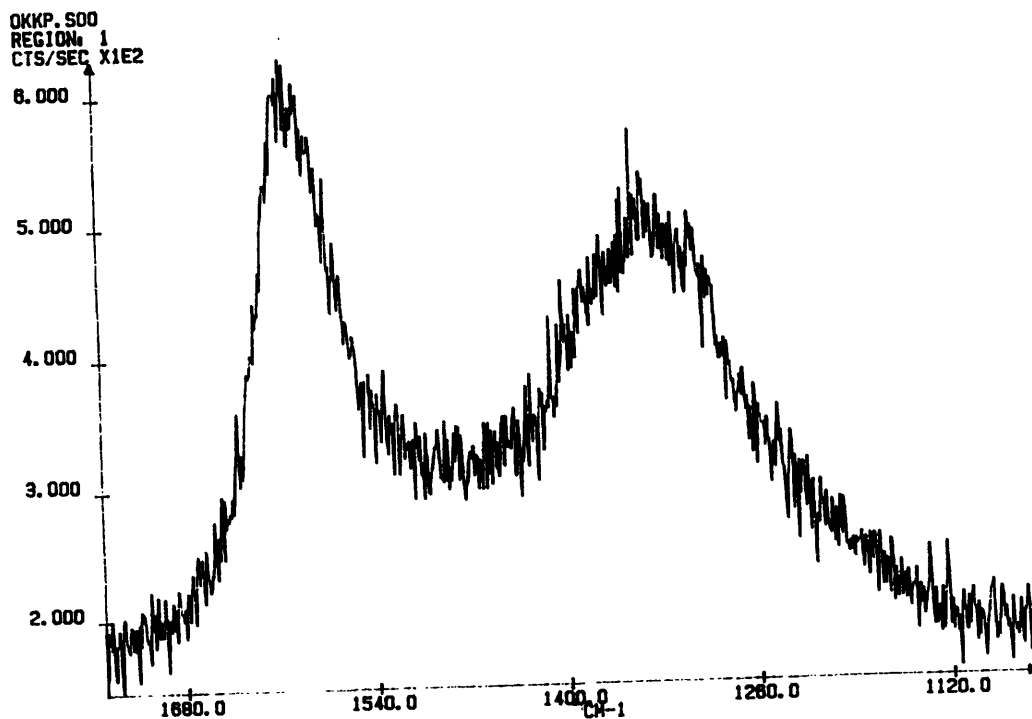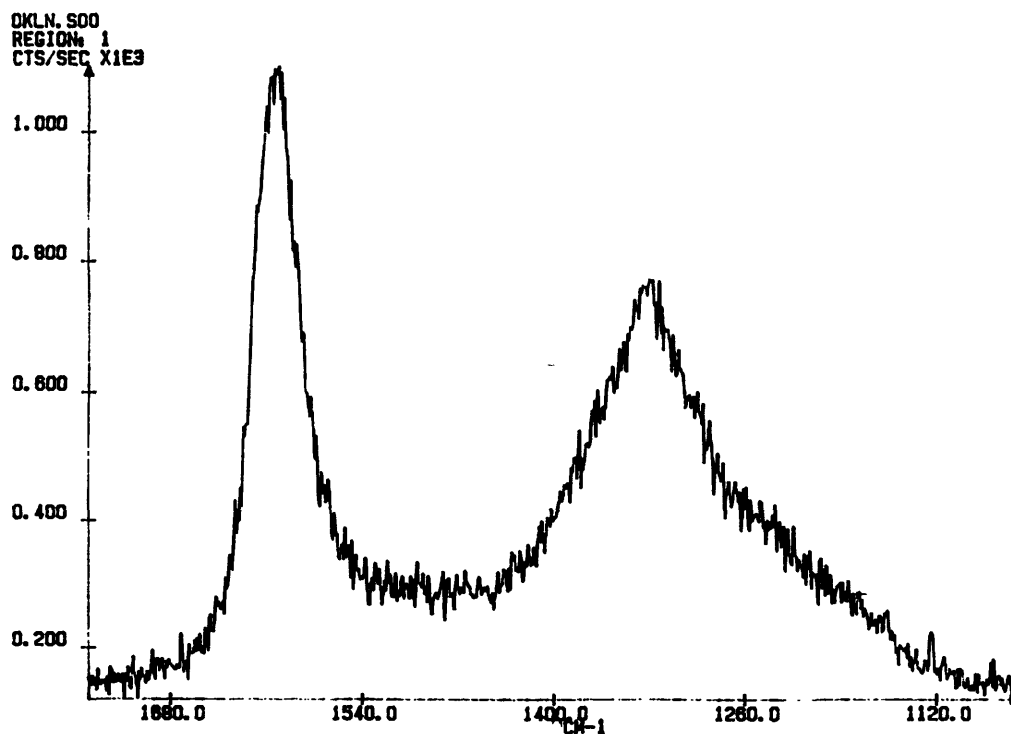
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Figure 5a. Laser Raman spectra from Oklo sample KP 3.

Figure 5b. Laser Raman spectra from sample LN 12 that is remote from the Oklo deposit.

SAMPLES FROM GABON

	Remote		Boyindzi		Oklo	
	FB Black Shale	LN 12	Boy 33.5	Boy 33.FC	Cro 336	Reactor 9: KP3
CH_4	p	p	p	p	p	p
CS_2	p	--	--	--	p	--
Benzene	M	p	p	p	p	p
C1-Benzene	M	M	p	M	M	M
n-alkanes	M	M	p	--	--	--
C2-Benzene	M	p	M	p	P	M
C3-Benzene	M	p	M	M	--	p
Naphthalene	p	--	M	M	p	M
Phenol	--	p	--	--	P	p
C1-Naphthalene	--	--	p	p	--	M
C2-Naphthalene	--	--	--	--	--	p
Elemental S	M	M	p	--	p	p

p = present   M = major moiety   -- not present

Table 2. Summary of pyrolysis-gc-ms results of samples

Laser Raman spectroscopy of all samples gives two broad peaks at around 1590 to 1600 and 1330 to 1360 cm^{-1}. An example of the results for a remote and a reactor sample are on figure 5. The two broad peaks rather than a sharp one at 1582 cm^{-1}, indicate that part of the organic matter is crypto-crystalline graphite and has not yet been matured to graphite with crystal domain as large as 1 micron. The samples that are remote from the reactor, FB Black shale and LN 12, showed the best peak near 1600 cm^{-1} (smallest width at half height), probably due to their very low uranium content having little effect on disrupting the crystallinity

Therefore, radiation in the uranium-rich zones may have decreased the apparent maturity of the kerogen by disordering the graphite lattice. The importance of these measurements are: 1) they confirm the maturity indicated by the Rock-Eval measurements and 2) they further elucidate the crypto-crystalline nature of the carbon.

The Rock-Eval, pyrolysis gc-ms and laser Raman results show a rather highly thermally altered to weakly metamorphosed organic mater that is now, at least in part, cryptocrystalline graphite. Relative to the remote samples, the U-rich samples seem to be less mature either due to radiation damage or to later addition of migrated organic matter. Either of these would explain the diversity of pyrolyzate moieties in the uranium-rich samples. In addition, water at elevated temperature in the reactor must also have affected the kerogen by the process of hydrous pyrolysis and that this would also affect clay-organic complexes. Hydrous pyrolysis would have produced liquid organic matter (bitumen), and mobilized and expelled it in aqueous solution as bitumen into neighboring rocks in the form of globules and vein fillings. Such a process appears to have occurred at Elliot Lake in Canada (Nagy and Mossman, 1988). If temperatures were high enough, organic matter may have been extracted from clay-organic complexes or clay-asphaltene type associations. Because such complexes favor the uptake and retention of polar compounds, including those with functional groups such as oxygen-bearing moieties, their extracts may account for the high oxygen index (OI) values as well as for the associated, somewhat lower T_{max}, organic matter shown by the uranium-rich samples. When the uranium and lead isotopic and fission product information (unpublished results) are taken into account, it is possible that both fluid migration and organic matter affected the redistribution of uranium and other isotopes during syngenetic and epigenetic events as has been suggested by Nagy and others (1989). In addition to their syngenetic kerogen, the uranium-rich samples may contain two types of bitumen that were generated and migrated during diagenesis and the operation of the natural reactor, and that both have subsequently been polymerized to a solid, in part by radiation damage.

ACKNOWLEDGMENTS

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