Bangombe (Gabon)

Description: The natural fission reactor of Bangombe (named BA 145 after the borehole number by which it was recognized) is one of the reactor zones found in the old Franceville sedimentary basin, at about 30 km south from the main Oklo uranium deposit. It is located at the foot of the manganese plateau, at the interface between the FA and FB formations (see review for Oklo). It is thus located in the discharge area of groundwaters having infiltrated the manganese plateau and the overlying FB pelites. Its specificity is to be located in a very shallow zone : the fossile reactor core lies only 12 m from the present ground surface and is thus also submitted to weathering. The core of the reactor is thin (< 15 cm for a lateral extension of about 10 m). Uraninite grains are embedded in a clay (illite, chlorite) rich matrix. Organic matter is abundant in the reactor. The FA formation, at the top of which the reactor zone is located, is an alternation of medium- and coarse-grained sandstones with intercalated fine-grained and black sandstones, vesicular sandstones and conglomerates. They show a great lateral and vertical variability and are very fractured. The pores and intergranular spaces are filled by a cement composed of chlorite, illite, quartz, opaque minerals, Fe-oxyhydroxides. Solid bitumen occurs in form of nodules, aggregates or fragments and also in fracture fillings. Two main types of bitumen are distinguished : oxidized bitumen and non mineralized bitumen. Solid bitumen is often associated with uraninite, galena, pyrite. Coffinite and LREE-phosphates were found in the secondary porosity of sandstones, beneath the reactor zone. U(VI)-SO_4 – PO_4 phases (uranopelite, zippeite, torbernite…) are also observed in microfractures around the reactor. An age of 76 500 +/- 6 800 y was assigned to a torbernite crystal.

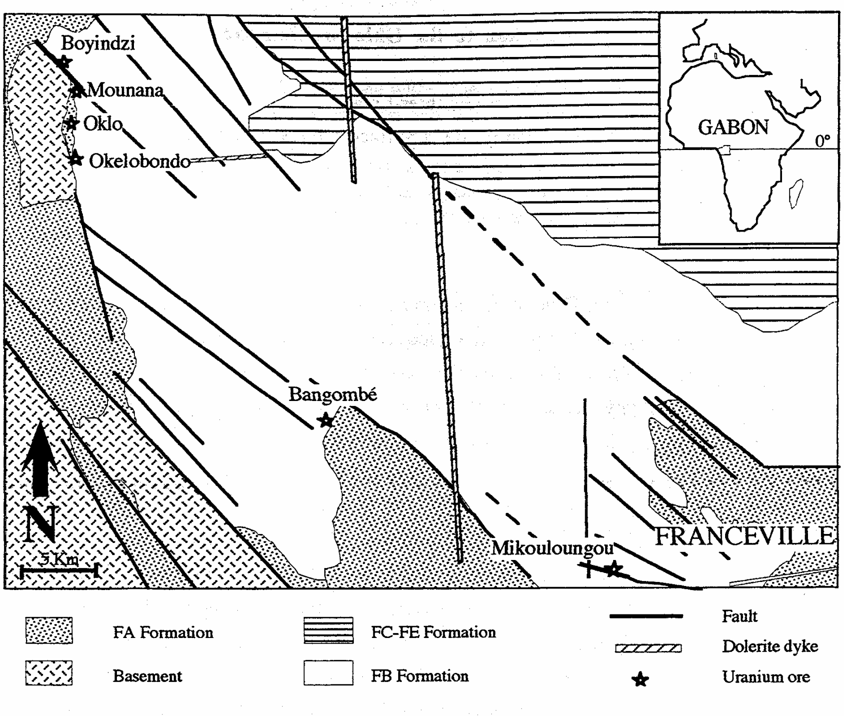

Figure 1: Map of the Franceville basin showing the location of the Oklo and Bangombé reactors (after Gauthier-Lafaye)

Several zones have been identified in the FB layers overlying the Bangombe reactor :

1) an “oxidised zone”, ~2.5 m thick, characterised by alternating red and indurated brown pelites, brecciated sandstones, black pelites. This transition zone between the FA and FB formations is not observed in all boreholes. The brown-black and red pelites represent the horizon overlying directly the reactor core. The goethite content is very high (5-40 %). Quartz and clays are present in equal portions. Kaolinite ranges from 15 to 20 wt %. Observations show that the clay matrix has been completely transformed by the precipitation of the iron oxyhydroxides. Horizontal and vertical fracture networks cross-cut the rock and are filled with ferric phosphates and Mn rich oxyhydroxides. In the uppermost part of the “oxidised zone”, two massive crusts (~ 3 cm thick

each) are developed, an iron crust (71 % goethite, 21 % illite, 8 % quartz) overlying a manganese crust (vernadite, lithiophorite, pyrolusite, cryptomelane).

2) a horizon of preserved black shales: grey-green pelites overlain by ochre-grey pelites. The content in chlorite decreases from the grey-green facies to the ochre-grey facies, showing a transition towards the weathered overburden. The major clay component is globally illite; a variable content in kaolinite (0-20 wt%) is observed. The dominant iron oxide is hematite; high goethite contents are observed in more altered zones. In the less altered black shales of this horizon, REE-Sr-Pb-crandallites were observed in association with illite.

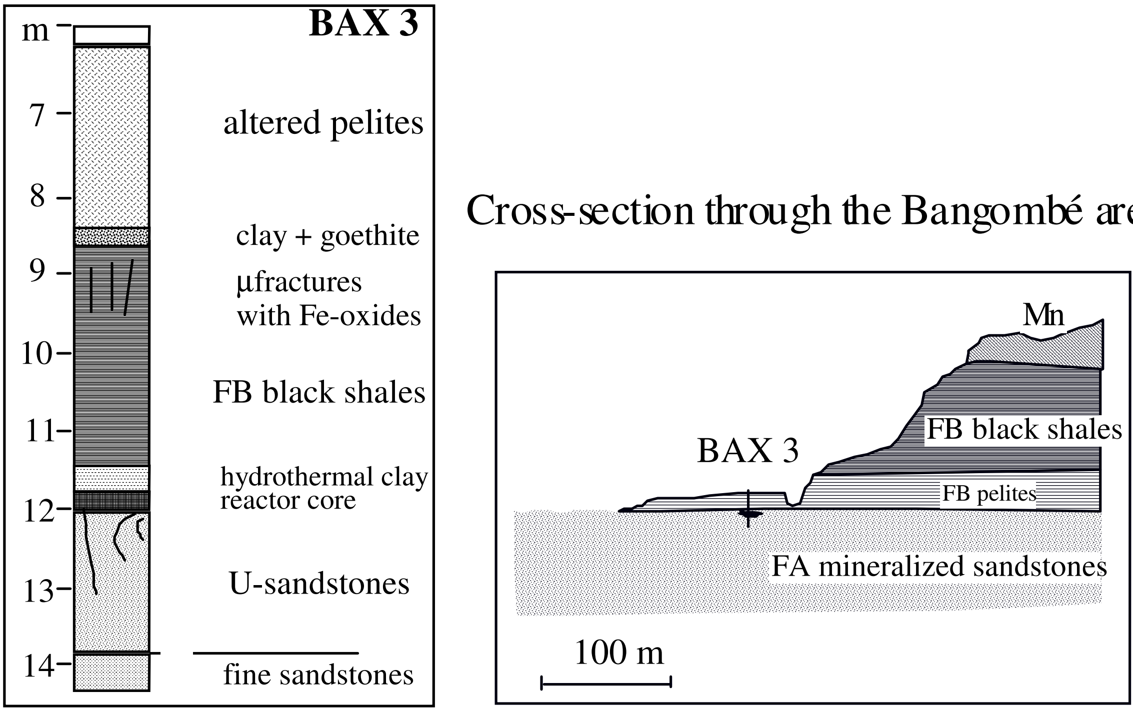

Figure 2: Stratigraphic section of the Bangombe reactor zone (bore hole BAX 3) from the soil to the FA sandstones (after Bros)

3) a "weathered zone", about 4 m thick, in which four facies are differenciated (white, pink, ochre and brown coloured clays). These materials result from the ferralitic alteration of black shales. The bulk composition of these facies is quite homogeneous and dominated by clay minerals, detrital quartz and Fe-oxyhydroxides (mainly goethite and to a lesser extent hematite). Illite is the major clay component, in association with neoformed kaolinite. Iron and manganese coatings are common. The differenciation of these four facies is mainly related to the quantity and nature of iron oxyhydroxides.

4) a soil layer, sometimes transformed to a mottled clay containing allochtonous fragments from the Bangombe plateau. The clay fraction mainly consists of illite, kaolinite and chlorite; mixed layer minerals (illite-smectite and chlorite-vermiculite) are also present. The chlorite content is higher in the less evolved soil layers (up to 30 %). Goethite, hematite and lepidocrocite reach contents up to 15 %.

The reactor zone of Bangombe was mainly studied in the EC Oklo-Phase II project. It was used as a natural laboratory to observe U and REE migrations and mineralogy, to test reactive transport codes and hydrogeological models and also conceptual models used to describe the migration of elements in the near field of a waste repository. In particular, depleted uranium in the groundwaters was used to trace water/rock interactions affecting the core of the reactor zone.

Figure 3: Aerial view of the Bangombé site; white spots indicate the location of boreholes (see Fig. 4) (Photograph by F. Gauthier-Lafaye, CGS, Strasbourg)

Major outputs of the studies on the Bangombe site are related to the following issues:

<u>Spent fuel and uraninite stability in a perturbed environment</u>

The Bangombe reactor is, among all reactor zones studied in the Franceville basin, the closest to the ground surface. Uraninite frequently shows corrosion features. However, observed secondary U(VI) containing phases, although well identified, appear to be negligible in mass compared to the progress of alteration in some places. This raises questions about the factors controlling the stability of U(VI) phases, e. g. sulfate availability. In several places in and around the RZ core, uraninite and U(IV) minerals (coffinites) have in contrast been well preserved, probably due to the redox buffering by bitumen.

One of the PA issue related to the long-term behaviour of spent fuel and raised during Oklo Phase II project is the evaluation of scenarios and conditions that would lead the spent fuel to progressively convert to a coffinite type product. Investigations are till in progress in this area.

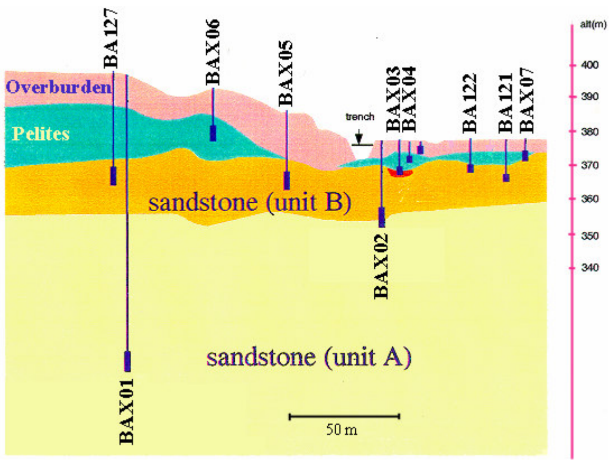

Figure 4: Vertical section of the Bangombe site showing the implantation of piezometers and water sampling devices. The reactor zone is depicted in red.

Radionuclide behaviour at the near-field/far-field interface ; radionuclide migration and retardation mechanism through sorption and precipitation with far-field components

REE concentration level in waters ranges from 3 to 20 ppt. A HREE enrichments is observed in waters, except for one borehole crossing the reactor (BAX03) in which enrichment in LREE is assigned to high fission yield of Nd. In Bangombé, several retention/complexation processes of REE and U are in competition. Fe-Phosphate coatings on Fe-oxides are highly enriched in HREE and show a strong retention of U(VI). The aqueous complexation by carbonates is active but seems to be outcompeted by phosphates.

LREE elements are enriched in phosphate minerals (apatites, monazites, florencite, Rhapdophane, crandallite, françoisite) and UO_2, coffinite LREE elements are also affected by sorption on clays. The qualitative and quantitative modeling of U soprtion in a complex clayey material is a major output of the studies performed on the Bangombe site.
The interest has been raised on specific mineral phases that act as radionuclide scavengers, in particular apatites.

Colloid studies performed in Bangombe indicate that, even in a superficial, dynamic groundwater system subjected to subtropical climate, the contribution of colloïds to U transport is very low. The nature of colloïdal particles nevertheless deserves further investigations (role of phosphates for REE ?).

Integration and modelling

Hydro-geochemical and reactive transport models were tested and used in order to reproduce the effects of water-rock interactions, to describe the geochemical evolution of the system and to assess migration time-scales. In particular a flow model around the reactor zone (from topography, rainfall, isotope data, lithography) was built and calculations of the transport of U around the reactor zone were performed. The prediction of the migration of present day produced ^{36}Cl, ^{129}I and ^{239}Pu was also attempted. Multivariable mixing and mass balance calculations were used to interpret groundwater data. The understanding and modelling of major redox buffer systems is an important outcome of this study and has applications in other sites, as the identified control mineral phases are encountered in many environments.

Several issues were raised :

- control of the redox (role of the organic matter, simulated by a dissolution rate of graphite).
- transformation of UO_2 to coffinite : no reliable logK for coffinite, low temperature conversion of UO_2 to coffinite is not understood (role of phosphate incorporation ?), 80% conversion predicted/ 8% observed
- variability with time in borehole Eh measurements

Relevance: The Bangombe site gives relevant analogies for several processes that are involved in the PA of spent fuel disposal and migration retardation:

- alteration and durability of uraninite in a zone of redox transition
- migration and retention of U, Th, REE, metals in a complex clayey material
- initial presence of actual fission products and transuranics give some qualitative clues on the behaviour of Tc, Pu, Np
- effectiveness of several natural redox buffers
- trapping of REE by phosphate minerals
- complexation and mobility of U and REE elements under the control of various ligands (silicate, phosphate, sulfate)

In addition, the geochemical observations conducted on this site offer several benchmarks for testing our understanding of speciation and migration of trace elements. The Bangombe datasets are thus important reference points for the estimation and calibration of integrated migration models.

Position(s) in the matrix tables: The Bangombé natural analogue study has direct applications to the long term behaviour of spent fuel waste forms (chemical integrity, radiolysis). Migration in clayey materials (EBS or near field rocks) is another important area of application. The potential role of colloids has also been addressed during the Bangombé project.

Limitations: Due to the long and complex history of the site, it is difficult to isolate simple subsystems that have not been influenced by the variations of boundary conditions. As for other NA studies, mass balances cannot be achieved easily except in rare cases for very small subsystems (eg UO_2 grains) for which immobility or strong mobility of specific elements can be established and quantified.

Presence of bitumen influenced redox conditions ; in an actual waste disposal, redox buffering by steel canister will probably also play an important role. It is however difficult to assess if these two buffering processes will have similar effects on the long term.

The accumulation of lead in the UO2 grains (due to U decay) could also bias the comparison with man made spent fuel.

Quantitative information: Quantitative information has been obtained in several areas that will give guidance to build realistic migration scenarios:

- Distribution of U and REE between different fractions of a complex clayey material
- Influence of colloïds on migration

Uncertainties: The main uncertainties are associated with the initial boundary conditions of the system which are not well defined.

Time-scale: Several time scales are involved in this system:
- the age of the U deposit (2 Gy)
- the duration of fission reactions (several to several tenth of ky) associated with a specific hydrothermal activity
- the duration and age of successive tectonic and hydrothermal events in the Franceville basin
- the flow regime and residence time of groundwaters in present day subsurface conditions (influence of oxygen)
- Two specific time windows seem to provide useable information : the period (about 1 My) at the time of reactor functioning and the recent period (since ~1 My before present) during which subsurface conditions have prevailed and led to U remobilizations.

PA/safety case applications: None known.

Communication applications: None known.

References:
There are more than 100 papers and reports published on the natural fission reactors at Bangome and Oklo. A few of the most recent and relevant are listed below.

Brookins D.G., Lee M.J., Mukhopadhyay B., and Bolivar S.F. 1975. Search of fission-produced Rb, Sr, Cs and Ba at Oklo. Le phénomène Oklo, IAEA, Vienne, p. 401-414.

Cowan G.A. 1976. A natural fission reactor. Scientific American. v.235, p. 36-47.

De Laeter J.R., Rosman K.J.R. and Smith C.L. 1980. The Oklo natural reactor: cumulative fission yields and retentivity of the symetric mass region fission-products. Earth and Planetary Science. Le

Hidaka H., Masuda A., Fujii I. and Shimizu H. 1988. Abundance of fissiogenic and pre-reactor natural rare-earth elements in a uranium ore sample from Oklo. Geochemical Journal, 22, 47-54.

Gauthier-Lafaye F., Holliger P., Ipavec S., Ledee V., Menager M.T. et Menet C. 1989. Compte rendu de mission - Oklo- Avril 1989. Rapport CEA.

Gauthier-Lafaye F., Weber F., and Ohmoto H. 1989 Natural fission reactors of Oklo. Economic Geology, vol. 84, p. 2286-2295.

Brookins D.G. 1990. Radionuclide behavior at the Oklo nuclear reactor, Gabon. Waste Management, 10, 285-296.

Nagy B., Gauthier-Lafaye F., Holliger Ph., Davis D.W., Mossman D.J., Leventhal J.S., Rigali M., and Parnell J. 1991 Role of organic matter in containment of uranium and fissiogenic isotopes at the Oklo natural reactors. Nature, Vol. 354, 12 Dec. 1992, p. 472-475.

Raimbault L., et Peycelon H. 1992. Oklo, natural analogue for transfer processes in a geological repository. Rapport d'avancement 1992. Contrat C.C.E. FI 2W CT 0071.

von Maravic H. 1993. Oklo working Group meeting. Proceeding of the second joint CEC-CEA progress meeting held in Brussels, April 1992. Ed. H. von Maravic, Report EUR 14877 EN.

Gauthier-Lafaye F. 1994. Oklo, Natural analogue for radionuclide transfer processes in a geological repository. Migration of Radionuclides in the Geosphere. MIRAGE Project- 3rd phase. Final Meeting, 1994. Brussels, 15-17 Nov. 1994.

Trotignon L; Michaud V., Louvat D 2000 Application of the Oklo natural analogue study to nuclear waste disposal model, test and development. n: Louvat D. et al., 2000, European Commission, Nuclear Science and Technology, OKLO Working Group Proceedings of the final meeting OKLO-natural analogue Phase II project held in Cadarache, France, from 20 to 21 May 1999, EUR 19137EN, 401-408, (2000)

Zetterström L. (2000) Oklo – A review and critical evaluation of literature. SKB TR-00-17, 37 p., SKB, Stockholm, Sweden.

Del Nero M., Salah S., Clement A. and Gauthier-Laaye F. 2002 Retention processes of uranium and REE in the Bangombé natural reactor zopne, Gabon. Eighth EC Natural Analogue Working Group Meeting. Proceeding of an international workshop held in Strasbourg, France from 23 to 25 March 1999. EUR 19118. H. Von Maravic, W. R Alexander (Edtors). p. 261-270.

Gauthier-Lafaye F. 2002 2 billions years old natural analogs for nuclear waste disposals: the natural nuclear fission reactors in Gabon (Africa). C. R. Physique. Acad. Sc., Paris. 3, 839-849.

Gauthier-Lafaye F., Stille P. and Del Nero M. 2002. 2.0 old natural nuclear wastes: the natural nuclear reactors in Gabon. The 2002 Goldschmidt Conference, August 18-23, 2002. Davos, Switzerland.

Added value comments: None to add.

Potential follow-up work: The Oklo uranium mine closed in year 2000. The reactor zone of Bangombé remains accessible for scientific purposes (the CEA is owner of this reactor zone). Several academic and applied institutions have identified a potential follow-up work on the site of Bangombe. Several key-issues have been identified on which progress in knowledge and understanding is needed:

Spent fuel behaviour:
He trapping in UO_2 grains : measurement of He concentration, derivation of diffusion coefficients
Athermal diffusion in UO_2 grains : study of the distribution of FP daughters
Coffinitisation : combine lab and field observations, link with other NA, study the role of PO_4, derive thermodynamic and kinetic constants

Migration/retention processes understanding:
REE retention mechanisms : role of PO_4, Fe-Mn oxides,
Fracture system in the sandstone
REE partitioning
U retention mechanisms

Controls of trace elements

Reactive transport modelling at different scales (UO2 grain scale, RZ scale, fractured systems)

Keywords : uraninite, dissolution, clay, pelites, sandstones, Oklo, Bangombe, U, Th, REE, fission products, coffinite, migration, modelling, phosphate, radiolysis, colloids, redox buffers, bitumen

Reviewers and dates : L. Trotignon (October 2004)