Oklo (Gabon)

Description: The natural fission reactors of Oklo were discovered in 1972. Routine analyses performed in the Commissariat à l'Energie Atomique (CEA) during the processing of uranium ores coming from the Oklo mine revealed isotopic anomalies (depletion in ^{235}U). Further investigations (Bodu et al. 1972, Neuilly et al.,, 1972 ; Naudet, 1991) showed that these ^{235}U depleted ores were coming from specific zones of the Oklo deposit in which sustained fission reactions occurred about 2 Gy ago. Several of these natural reactor zone (called RZ) which have usually the shape of metric to decametric lenses, where discovered in the Oklo mine and in the nearby Okelobondo site.

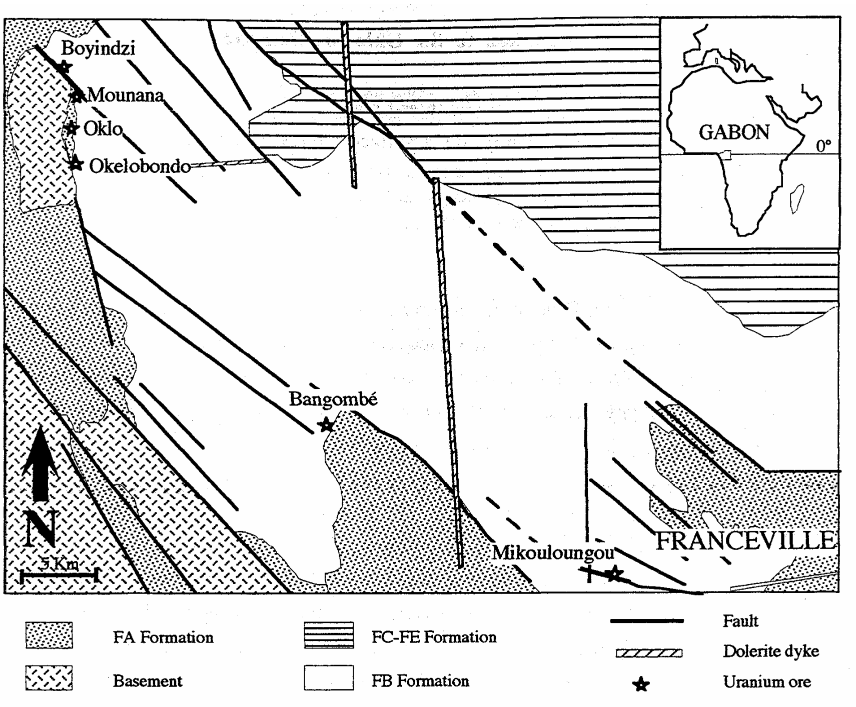

Figure 1: Simplified geological map of the Franceville basin (after Gauthier-Lafaye)

Most scientific results on the Oklo reactors were obtained during three major projects:

The Franceville Project (1972-1978). The aim was to acquire samples and data from several RZ, to observe local modifications induced by the reactors in the rocks, to understand and explain the reactor divergence and functioning, to constrain neutronic parameters of reactor cores (Naudet, 1991).

The EC funded Oklo Phase I project (1991-1995). This project was a intended to study the Oklo reactors and their environment as analogues for the long term stability of irradiated fuel in a waste disposal and for the migration of uranium and fission products. In this respect, the Oklo Phase I project was focused on samples from RZ 7-9, 10 and 13 and developed hydrogeological characterisation, hydrogeochemical studies and migration modelling in the Oklo, Okelobondo and Bangombé sites (Blanc, 1996 ; Holliger and Gauthier-Lafaye, 1996 ; Gurban et al., 1996)

The EC funded Oklo Phase II project (1996-1999). This project was a further step in the use of the Oklo site as an analogue for the migration of radionuclides in a natural environments. Boundary conditions were better constrained by a reassessment of the geological history of the site, leaching experiments of Oklo uraninites were conducted and compared to the leaching of PWR fuel, migration mechanisms were described and modelled, the confinement properties and the stability of uraninites and other mineral phases were evaluated, potential effects of colloïds and microbes were explored and an important effort was paid to the modelling of migrations at a large space scale.

The Oklo uranium mine definitely closed in year 2000. The reactor zone of Bangombé (see review file for this natural analogue) remains accessible for scientific purposes (the CEA is owner of this RZ).

Geological setting and history:
The Oklo uranium deposit is located on the SW margin of the Francevillian precambrian sedimentary basin (Haut-Ogouoé, Gabon). The Francevillian series (~2.3 to 2.1 Gy age, 1000 to 4000 m thick) lies unconformably on an archean crystalline basement (~2.7 Gy age). The Francevillian series is divided into five formations labelled FA at the base to FE at the top. The FA (100-1000 m thick, 400 m in Oklo) is a sandstone and conglomerate horizon (fluviatile deposit). The FB formation (deltaic environment) consists mainly of pelites (black shales) in its lower part and evolves towards sandstones at the top. FC, FD and FE formations are volcano-sedimentary rocks with chert and dolomite intervals.

Identified U mineralisations are all located in the FA formation. The Oklo U deposit is located in the ~10 m thick uppermost sandstone level (layer C1) of FA formation, just under the FB pelites. Upon progressive burial of the sedimentary series, organic matter migrated and was trapped in the layer C1, while early diagenetic evolutions affected the sediments (~2.1 Gy, chloritization, illitisation, silicification, max. burial depth of 4 000 m).

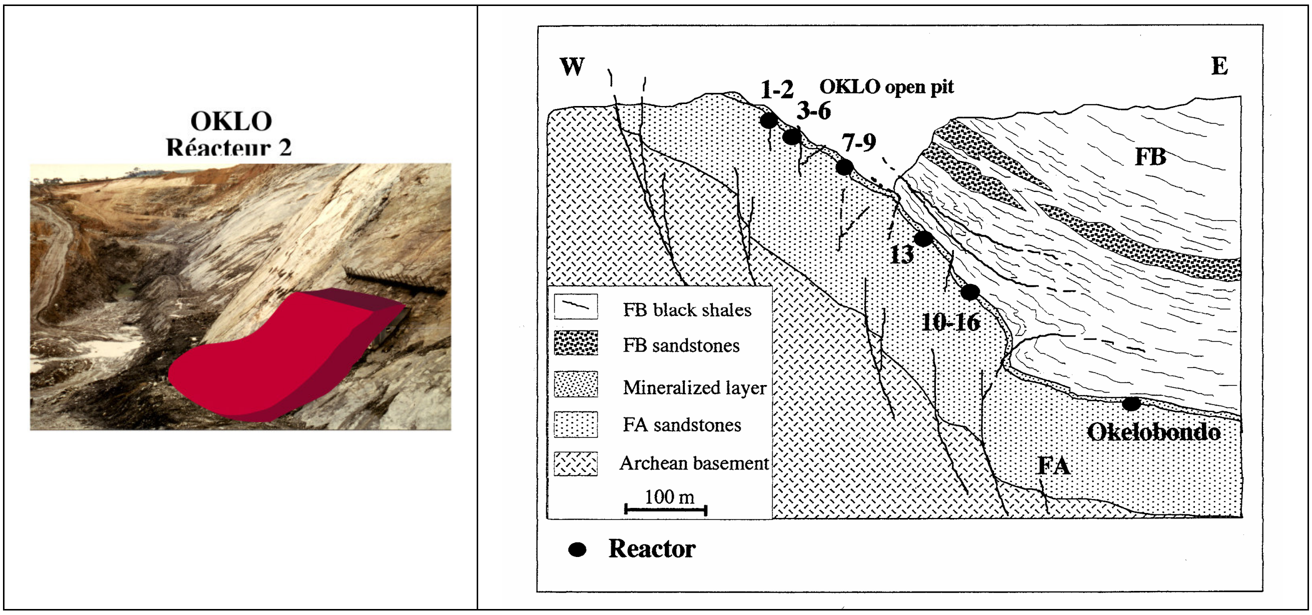

Figure 2: Left : visual reconstruction of Reactor Zone 2 in the Oklo pit. Right : location of reactor zones along the FA/FB interface. (after Gauthier-Lafaye, CGS, CNRS, Strasbourg)

The basin uplift is thought to have started soon after (~2.05 Gy). The U concentrations occurred when U(VI) bearing fluids met reducing conditions in the oil traps where organic matter accumulated. Soon after (1.97 Gy), conditions for the fission reactions were met in several zones of C1 layer, in which the U concentration ranged from 20 to 60 wt%. This local event is associated with local and intense hydrothermal modifications around reactor zones (desilification of the sandstones, formation of “Argile de pile” clays, hydrofracturation, collapse of the roofs and walls of the reactor due to mass loss). The local p-T conditions around the reactor zone are in the range of 30 to 40 Mpa and 300 to 400°C (compared to regional temperatures of ~140°C at the depth of interest).

Local redox conditions were also altered by radiolysis. Important but local elemental migrations, affecting actinides and fission products, are associated with the reactor zone function. The duration of reactor operation, based on the recalculated ^{239}Pu production and other isotopes, ranges from 20 000 to 800 000 y, depending on the reaction zones. Several important events affecting the

Franceville basin were then recorded during the continuous uplift of the sedimentary series. In particular, regional episodes of distension at 980 My (dolerite intrusion in Mikouloungou) and 780 My (dolerite intrusion in Oklo) are linked with marked hydrothermal events. The 780 My dolerite dyke in Oklo affected strongly RZ 13, situated at only 15 m of the intrusion and is associated with important elemental migrations. Several other events are recorded in the Franceville basin, in particular the opening of the South Atlantique Ocean. In mine measurements of U depletion in groundwaters and stable isotope data, also evidence that recent water circulations continue to slowly alter the reactor zones.

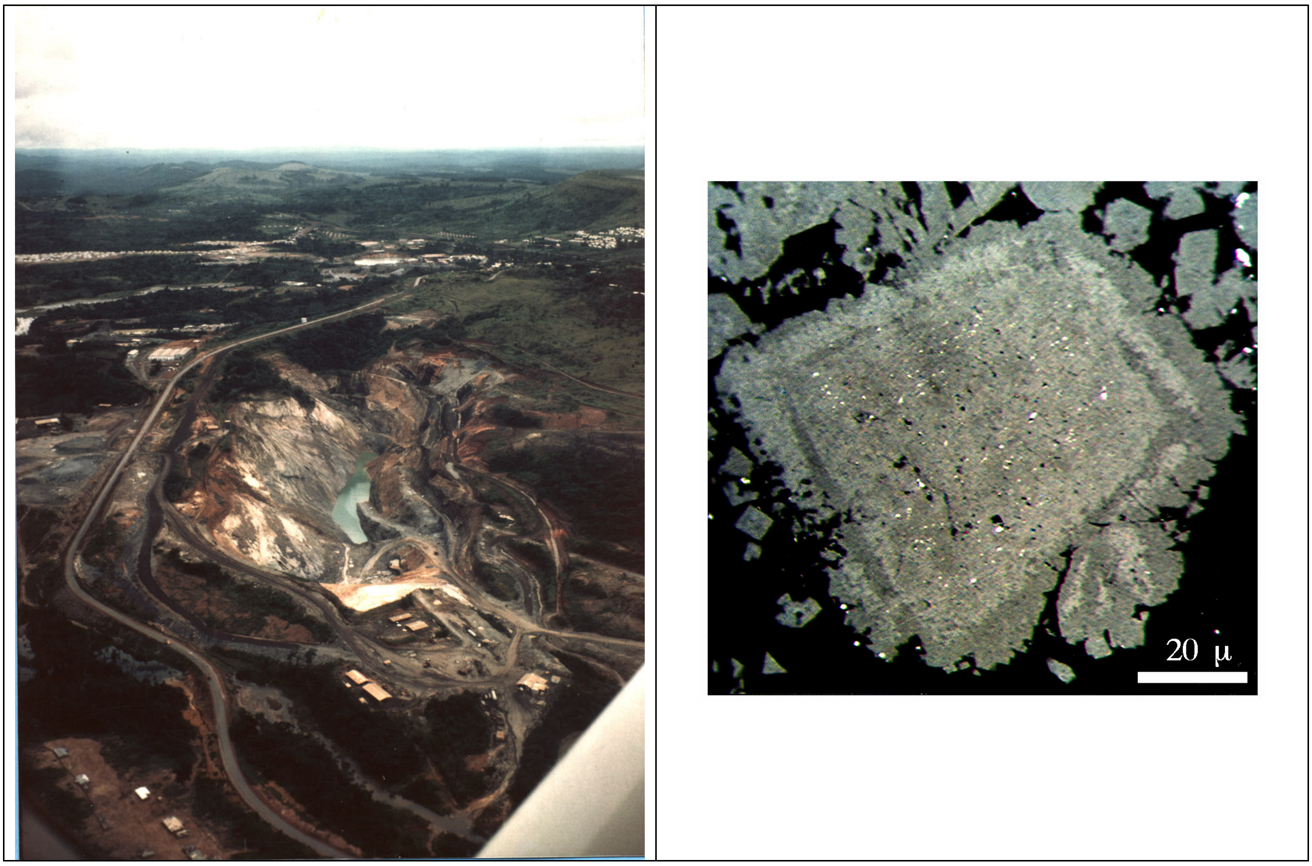

Figure 3: Left : Aerial view of the Oklo pit at the end of the 1990 years. Right : Uraninite crystal (Reactor Zone 10) (after Gauthier-Lafaye, CGS-CNRS, Strasbourg)

<u>Summary of the different reactor zones:</u>
Reactor zones numbered 1 to 9 were studied within the framework of the Franceville project and were then mined out, after collection of samples. Reactor 15 was mined out before the start of the EC projects. Organic matter was abundant in reactor zones 7 to 9.

Reactor zones 11, 12 and 14 have not been confirmed after reconnaissance during the mining explorations.

Reactor 10 was one of the most studied reactors. It has the shape of a 15 m width and 30 m length lens. Organic matter is abundant in and around the reactor. The core and the clays are well developed and the top of the reactor collapsed. Apatite crystals were found in the clays.

Reactor 13 is located very close to the dolerite dyke (15 m). The dolerite intrusion perturbed a lot the reactor core and led to important mobilisations of major and minor elements (e. g. sulfur, lead, arsenic,). It is a small reactor zone (6 m width x 15 m length).

Reactor 16 was the last discovered reactor in Oklo ; it has been sampled both on wall-face and by coring.

Reactor OK84 was found in the Okelobondo site, at the south end of the Oklo mine, in the deeper part of the mine. It was recognised by drill-holes. Organic matter has been found in abundance in and around the reactor. Because of his deep location, it has been far less affected by meteoritic alteration than for exemple the RZ9 or the Bangombé reactor.

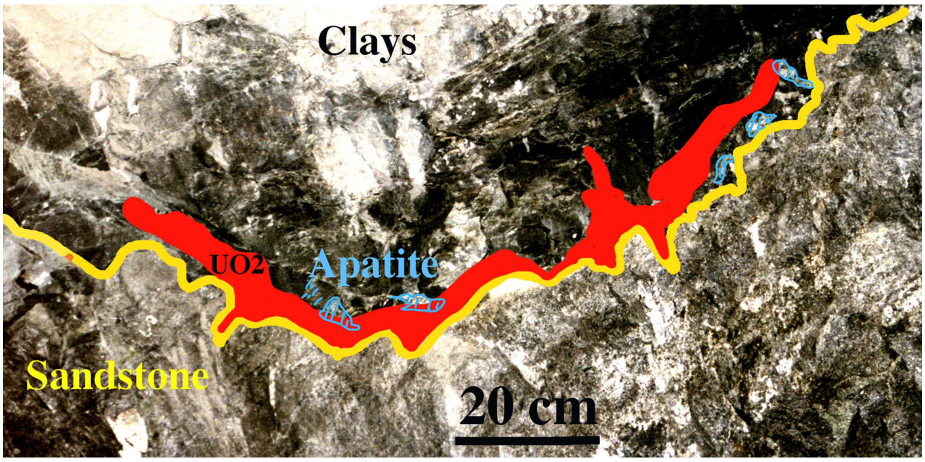

Figure 4: Reactor zone 10 (Gauthier-Lafaye, CGS, CNRS, Strasbourg)

The source term and the analogy with PWR spent fuel:
One of the aim of the initial investigations on the Oklo reactors was to understand how the nuclear fission reaction could start, last sufficiently to affect large zones of the deposit and then eventually stopped. In addition, these investigations were of primary importance to assess, at least qualitatively, mass balances on actinides and fission products. This analysis showed indeed several interesting analogies and differences between "Oklo fuel" and manmade spent fuel:

- comparable initial $^{235}U/^{238}U$ ratio but less complete nuclear reactions in Oklo than in a PWR (by a factor of 2 to 3); in addition the "burn rate" was far less in Oklo uraninites than in a PWR fuel, leading thus at lower
- association of platinum metals (Pd, Ru, Rh, …) in metallic aggregates that are similar to insoluble inclusions in modern spent fuel. ^{99}Tc is shown to have been associated with these aggregates. Mo, Te and Bi are also found in aggregates.
- build-up of large concentrations of radiogenic lead in the Oklo uraninite

An important feature of the Oklo reactor zones, is that intense hydrothermal alteration occurred in an around the affected layers, at temperatures up to 400°C. In addition, due to the age of the Oklo event, radioactive fission products and transuranics have totally decayed and the reconstruction of their past migration relies on the interpretation of mass spectra of their daughters (baryum for cesium, xenon for iodine, etc). A recent study of noble gas isotopes trapped in Al phosphates of RZ 13 (Meshik et al., 2004) showed that the Oklo natural reactors worked in a pulsed way, like geysers.

Studies of uraninite have shown that this mineral retained significant amounts of the low solubility and non volatile fission products even under the hydrothermal conditions prevailing while fision reactions were taking place. The preservation of the uraninite themselves was probably favoured by the buffering of redox conditions by organic matter. It occurs as densely packed, up to 1 mm sized idiomorphic and brecciated grains, dispersed in an illitic and/or organic matter matrix. Uraninite has however often been subjected to a partial corrosion and coffinitisation. The lead-content varies from about 25 % (in pristine uraninites) to nearly 0 % in partially altered uraninite grain-boundaries. Th, lanthanides and Zr are well retained in the uraninite.

Short term laboratory dissolution studies were undertaken on Oklo uraninites providing datasets that are consistent with other similar studies on uraninite or spent fuel.

Criticality:

The understanding obtained on the Oklo reactors shows that criticality in a natural setting is difficult to achieve and, if it occurs at all, it is a self limiting process. This point is important for the consideration of waste forms that may contain significant amounts of fissile isotopes (e. g. spent fuel).

Radiolysis effects:

Investigations of fluid inclusions have shown the importance of radiolysis effects on water : H2 and O2 were detected in fluid inclusions at the edge of reactor 10. Hematite caps found above or around several reactor zones are interpreted as a consequence of locally oxidising conditions. Dissolution/reprecipitation of uraninite upon locally highly oxidative and reducive environments across reactor zones is also strongly suggested.

Trapping by specific host phases:

Several fission products were lost by the uraninites (among which Cs, Rb, Ba, I, Sr). Indications exist of their trapping in different phases of the clay rich surrounding (e. g. apatites, organic matter) but it is impossible to make a complete mass balance. Plutonium trapping is also evidenced in the surrounding chlorites and apatites, most probably at the time of reactor function. Investigations have shown that the apatites, that are stable in deep conditions, may evolve to other secondary phosphates in surface or perturbated conditions ; in this case, a redistribution of trapped REE is evidenced ; this should also affect actinides. Apatites found in Oklo also trapped U, Cs, Sr, Rb, Mo, I, Zr and Ba.

As mentionned above, Tc has probably been trapped in platinum group aggregates. Investigations of Ru isotopes in reactor OK84 (Okelobondo) showed that the loss in 99Tc ranged from 1-3 % in the less porous lower part of the reactor to up to 20 % in the more porous upper region of the ore. The trapping of mobilized Tc in chlorites and Mn-oxides is probable.

Migration studies:

All elements present in the reaction zones were, at a moment or another, subject to some remobilisation. In addition, several elements, like REE may have both a fissiogenic and a non fissiogenic origin. Finally, different reactor zones have experienced various initial and boundary conditions which sometimes moderates the conclusions of observations at a specific point. The presence of clay rich materials (chlorites, illite, kaolinite) inside and around the reactor zones is a major feature in Oklo and has led to numerous observations of mm to m scale elemental migrations.

As shown by the anomalous $^{235}U/^{238}U$ ratio, U migrated around reaction zones at distances from 10 cm to about 1 m. It was trapped in clays (see also the review on Bangombé) and also precipitated in secondary U(VI) bearing phases. Several migration episodes occurred, including during recent alterations by supergene waters. Th seems to have been fairly immobile ; anomalies observed in the walls of reactor 10 evidence a mobility of ^{236}U before decay to ^{232}Th.

REE were studied both to elucidate the nuclear fission process and to evaluate their migration in clay rich environments. The differential behaviour of LREE and HREE is linked to the increasing strength of the carbonate complexes. Their trapping occurs in clays (illite, chlorite, smectite) in Mn and Fe-oxyhydroxides and in phosphates.

Evaluation of present day low-level production and migration of ^{36}Cl, ^{129}I and ^{239}Pu has also been done.

Geochemical, transport and reactive transport modelling:

Several types of PA support models have been tested at Oklo ; the Oklo project supplies a reference dataset. In particular, the combination of blind prediction modelling tests and geochemical investigations gives a consistent picture of speciation and solubility controls in oxic to anoxic media. Test cases developed during Oklo Phase II project in order to describe coupled flow and chemistry are an important milestone for the involved teams and should help PA modellers to chose relevant hypotheses.

Colloïds, microbes:
see Bangombé review.

Relevance: The Oklo studies are relevant in several respects to studies on waste disposal and elemental migration :

- understanding the UO_2 stability and degradation from anoxic to oxic conditions
- evidencing the effects of radiolysis in the near-field
- indirect/direct evidence of the trapping of actual fission products in generic phases (clays, phosphates, oxy-hydroxides, …)
- benchmark for the simulation of reactive-transport at redox interfaces

Position(s) in the matrix tables: Spent-fuel, near-field, clay, migration, radiolysis

Limitations:.Long and complex geologic history making difficult the reconstruction of all alteration events. Mass balances are difficult to assess for mobile elements except for some systems at the UO_2 grain scale. Accumulation of lead inside UO_2 grains due to decay may bias the comparison with man made spent fuel.

Quantitative information: Characteristic migration distances for several elements on a geologic time scale

Uncertainties: Main uncertainties linked with the reconstruction of the geological history
Some perturbations due to the mining activities

Time-scale: Several time scales to be considered :
– duration of criticality inside reactor zones (less tha 1 My), accompagnied with a specific hydrothermal activity
– long term and slow uprising of the Franceville basin (more than 1.9 Gy) accompagnied by punctual thermal, tectonic and/or hydrothermal events
– recent perturbations due to the influence of surficial waters (less than 100 ky to 10 y)

PA/safety case applications: The Oklo study has been used for the development of conceptual models of criticality and for understanding redox buffering.

Communication applications: The Oklo natural analogue plays a unique role among natural analogues because it has focused the interest of a large scientific community and also of the public: how could it possibly happen for a fission reactor to be formed and work spontaneously in the environment ?

A large part of the public however doesn't know the existence of Oklo and presenting this natural analogue frequently exerts a specific fascination. Relating this specific analogue to waste disposal issues has been done in films (e.g. IPSN) or in advertisements (e.g. SKB).

Many aspects of the natural reactors of Oklo are nevertheless till areas of scientific investigations.

References:

Blanc P-L. 1996. Oklo-Natural analogue for a radioactive wate repository (phase 1). Volume 1. Acquirement of the project. in European Commission, Nuclear science and technology series, VI, 123 pp. ISBN 92-827-7448-1

Bodu R., Bouzigues H., Morin N., and Pfiffelmann J.P. (1972) Sur l'existence d'anomalies isotopiques rencontrées dans l'uranium du Gabon. C.R. Acad. Sci. Paris 275, 1731-1734.

Bracke G. Salah S., Gauthier-Lafaye F. 2001 Weathering process at the natural fission reactor of Bangombé. Environmental Geology, 40, 403-408.

Bros R. 1993. Géochimie isotopique (Sm-Nd, Rb-Sr, K-Ar, U) des argiles du bassin Protérozoïque de Franceville et des réacteurs d'Oklo (Gabon). Thèse Université Strasbourg, ULP. 1993

Bros, R., Carpena, J., Sere, V., Beltritti, A. 1996. Occurence of Pu and fissiogenic rare earth elements in hydrothermal apatites from the fossil naturel nuclear reactor 16 of Oklo (Gabon) Radiochim. Acta, 74, 277-282

Bros R, Hidaka H., Kamei G. and Ohnuki T. 2003 Mobilization and mechanisms of retardation in the Oklo natural reactor zone 2 (Gabon)—inferences from U, REE, Zr, Mo and Se isotopes, Applied Geochemistry, 18, 1807-1824

Bruno J., Duro L. and Grive M. (2002) The applicability and limitations of thermodynamic geochemical models to simulate trace element behaviour in natural waters. Lessons learned from natural analogue studies. Chemical Geology, 190, 371-393.

Casas I., De Pablo J., Giménez J., Torrero M.E., Bruno J., Cera E., Finch R.J. and Ewing R.C. 1998. The role pf Pe, pH, and carbonate on the solubility of UO2 under nominally reducing conditions. Geochim. et Cosmochim. acta, 62, p. 2223-2231.

Casas I., De Pablo J., Perez I., Gimenez J., Dura L. and Bruno J. (2004) Evidence of uranium and associated trace element mobilization and retention processes at Oklo (Gabon), a naturally radioactive site. Environ. Sci. Technol., 38, 3310-3315.

Curtis D., Benjamin T., Gancarz A., Loss R., Rosman K., DeLaeter J., Demore J.E., and Maek W.J. 1989 Fission product retention in the Oklo natural fission reactors. Applied Geochemistry, 4, p. 49-62

Dubessy J., Pagel M., Beny J.M., Christensen H., Bernard H., Kosztolanyi C. and Poty B. 1988. Radiolysis evidence by H2-O2 and H2-bearing fluid inclusions in three uranium deposits. Geochim. Cosmochim. Acta, 52, 1155-1167.

Eberly, P., Ewing, R.C., Janeczek, J. and Furlano, A. 1996 Clays at the natural nuclear reactor at Bangombé, Gabon: Migration of actinides. Radiochim. Acta 74, 271-275

Gauthier-Lafaye F. 1997. The last natural nuclear fission reactor. Nature, 387, p. 337.

Gauthier-Lafaye F. 1977 Oklo et les gisements d'uranium du Francevillien : aspects tectoniques et métallogéniques. Thèse 3° cycle, ULP- Strasbourg, 82 p.

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

Gauthier-Lafaye F., Holliger P. and Blanc P-L. 1996. Natural fission reactors in the Franceville basin, Gabon: a review of the conditions and results of a critical event in a geologic system. Geochimica et Cosmochimica Acta. 60, 4831-4852.

Hagemann and Roth, 1978. Relevance of the studies of the Oklo nuclear reactors to the storage of radioactive wastes. Radiochimica Acta, 25, 241-247.

Hidaka H. and Gauthier-Lafaye F. 2000. Redistribution of fissiogenic and non-fissiogenic REE, Th and U in and around natural fission reactors at Oklo and Bangombé, Gabon. Geochim. et Cosmochim. Acta. 64, 2093-2108.

Hidaka, H.; Konishi, T.; Masuda, A. 1992 Reconstruction of Cumulative Fission Yield Curve and Geochemical Behaviors of Fissiogenic Nuclides in the Oklo Natural Reactors Geochem. J. 26, p.227-239.

Hidaka H., Shinotsuka K. and Holliger P. 1993. Geochemical behaviour of 99Tc in the Oklo natural fission reactors. Radiochimica Acta, 63, 19-22.

Hidaka H., Sugiyama T., Ebihara M. and Holliger P. 1994. Isotopic evidence for the retention of 90Sr inferred from excess 90Zr in the Oklo natural fission reactors: implication for geochemical behaviour of fissiogenic Rb, Sr, Cs and Ba. Earth and Planetary Science Letters, 122, 173-182.

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.

Horie K., Hidaka H. and Gauthier-Lafaye F. 2004 Isotopic evidence for trapped fissiogenic REE and nucleaogenic Pu in apatite and Pb evolution at the Oklo natural reactors. Geochem. Chimica Acta. 68, 115-125.

Janeczek J. and Ewing R. 1992. Dissolution and alteration of uraninite under reducing conditions. Journal of Nuclear Materials, 190, 157-173.

Janeczek J. and Ewing R. C. (1996). Florencite-(La) with fissiogenic REE from a natural reactor at Bagombé, Gabon. Amer. Mineral., 81, 1263-1276.

Langmuir D. (1978) Uranium solution-mineral equilibria at low temperatures with applications to sedimentary ore deposits. Geochim. Cosmochim. Acta, 42, 547-569.

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. Letters, 50, 238-246.

Mathieu. R. 1999. Reconstitution des paléocirculations fluids et des migrations élémentaires dans l'environnement des réacteurs nucléaires naturels d'Oko (Gabon) et des argilites de Tournemire France). Thèse de l'Institut National de Polytechnique de Lorraine. Nancy. Sept. 1999.

Mathieu R., Zetterström L., Cuney M., Gauthier-Lafaye F. and Hidaka H. 2001. Alteration of monazite and zircon and lead migration as geochemical tracers of fluid paleocirculations around the Okl-Okélobondo and Bangombé natural nuclear reaction zones (Franceville basin, Gabon). Chemical Geology. 171, 147-171.

Meshik A. P., Hohenberg C. M. and Pravdivtseva O. V. (2004) Record of cycling operation of the natural nuclear reactor in the Oklo/Okelobondo area in Gabon. Physical Review Letter, 93(18), 182302-1-4.

Menet C., Ménager M.T., et Petit J.C. 1992 Migration of radioelements around the new nuclear reactors at Oklo: Analogies with a high-level waste repository. Radiochimica Acta, 58/59, 395-400

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.

Naudet R. 1991 Oklo : des réacteurs nucléaires fossiles. Paris : Collection du Commissariat à l'Energie Atomique. Eyrolles, Paris (France), 685 p.

Neuilly M., Bussac J., Frejacques C., Nief G., Vendryes G. and Yvon J. 1972 Sur l'existence dans un passé reculé d'une réaction en chaîne naturelle de fission, dans le gisement d'uranium d'Oklo (Gabon). C.R. Acad. Sci. Paris, 275, p. 1847-1849

Oversby V.M. 1996. Criticality in a high level waste repository, A review of some important factors and an assessment of the lessons that can be learned from the Oklo reactors. SKB Technical Report 96-07 (June 1996).

Pourcelot L. 1997 Les réacteurs de fission naturels du Gabon. Contribution à l'étude des conditions de stabilité d'un site naturel de stockage de déchets radioactifs (2 Ga). Thèse de l'Université Louis Pasteur, Strasbourg.209p.

Salas J. and Ayora C. (2004) Groundwater chemistry of the Okélobondo uraninite deposit area (Oklo, Gabon) : two-dimensional reactive transport modelling. J. Cont. Hydrology, 69,115-137.

Salah S. 2000. Weathering processes at the natural nuclear reactor of Bangombé (Gabon). Identification and geochemical modeling of the retention and migration mechanisms of uranium and rare earth elements. Thèse ULP, Dec. 2000.

Savary V. and Pagel M. 1997. The effects of water radiolysisi on local redox conditions in the Oklo, Gabon, natural fission reactors 10 and 16. Geochim. et Cosmochim. Acta, 61, p. 4479-4494.

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

Added value comments: Recent work published by different authors shows the potential of Oklo to challenge the imagination and skill of researchers, with benefit for a large community, from the academic to the industrial application.

Potential follow-up work: On stored samples there is potential for studies on UO_2 long term behaviour (He diffusion, coffinitisation), on the retention of FP (technetium, iodine). At the Bangombé site there remains the interest of several institutes for a further investigation on the migration of FP and U (see Bangombé review for more detail).

Keywords: near-field, UO_2, spent fuel, clay, redox front, nuclide containment, chemical alteration, radiolysis, nuclide release, dissolution, precipitation, leaching, THC, nuclide retardation, far-field, sandstone, oxidation, reduction, technetium, REE, plutonium, uranium, iodine, reactive-transport modelling

Reviewers and dates: Laurent Trotignon (April 2004)