

## Oxidation Processes of the Natural Nuclear Reactor of Okélobondo (Gabon): Reactive Transport Modelling

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Natural fission reactions took place in Oklo (Gabon) during the Proterozoic Age (210<sup>9</sup> years), in the basal sediments of the Franceville Basin. The fission products have been preserved in massive bodies of uraninite and, at present day, these deposits are close to the topographic surface (outcropping in some cases) and have been mined. The Okélobondo uranium deposit is part of this uraniferous system. It is located at a depth of about 300 m and is affected by oxidation and dissolution processes. The presence of high pH-Eh water at depths similar to those expected for radioactive waste confinement is unusual. It is important to determine whether the processes leading to this situation are site specific or whether they might represent a common phenomenon.

Two types of groundwaters are distinguished in a pH-Eh diagram (Gurban et al., 1996): Type I, characterised by pH-Eh distributions parallel to the Fe<sup>+2</sup>-Fe(OH)<sub>3</sub> equilibrium and, Type II, oxidising, slightly basic and subsaturated with respect to uraninite. These data suggest that the Fe<sup>+2</sup>-Fe(OH)<sub>3</sub> equilibrium plays an important role in controlling the chemistry of the Type I water. On the other hand, the relative abundance of Mn with respect to Fe, and the distribution of the Type II water samples close to equilibrium with Mn-minerals, suggest that the pair Mn<sup>+2</sup>/Mn<sup>+3</sup> could play a significant role in controlling the pH-Eh of this type of water.

In order to identify the main geochemical processes which control the water composition, reactive transport modelling has been applied on three conceptual 1D models (along characteristic flow paths) and a 2D model at 25 °C. The reactive transport

calculations were performed with the computer code RETRASO, developed by the UPC-CSIC group (Saaltink et al., 1998).

The chemical composition and the pH-Eh values of the water sampled in the upper part and in the western margin of the Okélobondo system (Type I water) can be explained in terms of the interaction of a meteoric recharge with the pelites, dolomitic complexes and sandstones. The dissolution of Fe-silicates and the precipitation of Fe(OH)<sub>3</sub> maintained the Eh along the Fe<sup>+2</sup>-Fe(OH)<sub>3</sub> equilibrium, reaching values close to 0.0 volts. Uraninite does not dissolve at the low Eh values obtained. This could account for the lower U content in the water samples from the pelites and dolomitic complexes above the Okélobondo deposit. The high Mn/Fe ratio and the high pH-Eh values of the water sampled in the lower part of the Okélobondo system (Type II water), are attributed to the interaction of the recharge with the Mn-deposits. Three key factors are required to obtain the chemistry of type II water: 1) a fast dissolution of a Mn-phase, 2) a slower dissolution of a Fe-phase and, 3) a continuous source of alkalinity. As a result, uraninite dissolves at the high Eh values obtained in deep, and radiogenic isotopes are released in groundwater.

The variability of the analytical data could be explained by mixing water and environmental changes in the infiltration water (pH and alkalinity could modify rhodochrosite dissolution and Fe(OH)<sub>3</sub> precipitation), and by changes in the parameters which control mineral dissolution and precipitation, which are mineral reactive surfaces and kinetic laws.

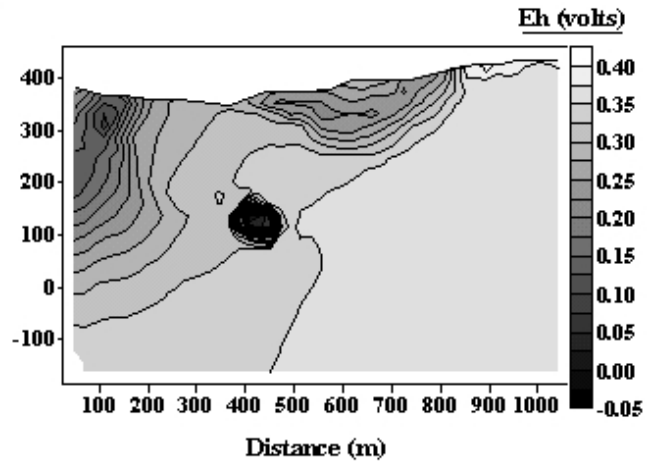
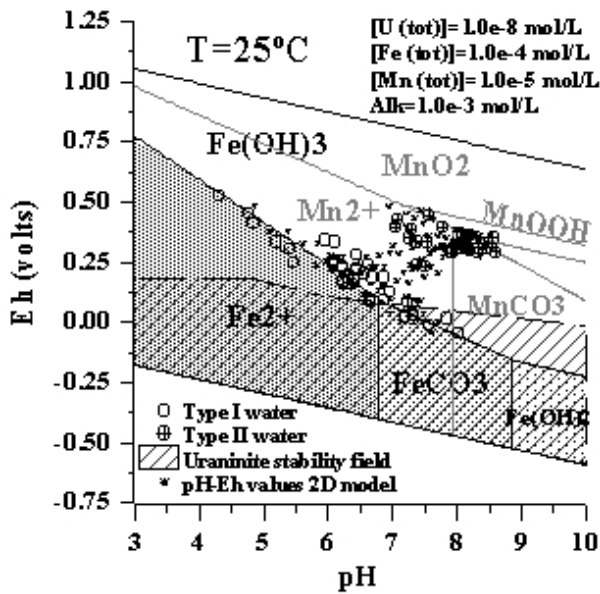


Figure 1: pH-Eh diagram and the Eh field obtained in the Okélobondo system.

Saaltink, MW, Ayora, C & Carrera, J, *Water Resources Research*, **34**, 1649-1656, (1998).

Gurban I, Ledoux E, Madé B, Salignac AL, Winberg A, Smellie J, Louvat D & Toulhoat P, *Oklo, analogue naturel de stockage de déchets radioactifs (Phase I). CE Sciences et Techniques Nucléaires*, *EUR 16857/3*, **3**, 117, (1996).