

DATING FLUID FLOW EVENTS IN A SHALLOW SEDIMENTARY BASIN: THE KEY CONTRIBUTION OF K-Ar GEOCHRONOLOGY OF AUTHIGENIC ILLITE

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In shallow parts of sedimentary basins, low-temperature diagenetic processes (<90-100°C) produce discrete mineralogical changes on both the carbonates and clays. These processes are somehow challenging to investigate as most of the geothermometers are at their application limits. Furthermore, the low concentrations of trace elements in the newly-formed minerals (e.g., REE in fluorite, U in calcite) complicate the dating attempts. In this context, this study presents petrographic and geochemical data acquired on secondary minerals filling the fractures, vugs and primary porosity of three aquifers of the Mesozoic sedimentary sequence from Paris Basin, France.

The Oxfordian and Middle Jurassic limestone aquifers located respectively above and below Callovian-Oxfordian claystones are essentially cemented by successive stages of blocky calcite. Isotopic tracing ($\delta^{18}\text{O}$, $\delta^{13}\text{C}$ and $^{87}\text{Sr}/^{86}\text{Sr}$) and data acquired on hosted fluid inclusions (salinity, δD of paleo-fluids) reveal that the physical-chemical properties of calcite-forming waters were rather different for each of the aquifers and from present-day groundwaters. The aquifer located in Lower Triassic siliciclastic sedimentary rocks is buried at a 2000m depth and evidences a diagenetic alteration, among which quartz overgrowth, adularia precipitation and widespread illitization. Illite nanometric particles (<0.02 and 0.02-0.05 μm fractions) were separated, analyzed (DRX, EDS microprobe, $\delta^{18}\text{O}$, $^{87}\text{Sr}/^{86}\text{Sr}$) and K-Ar dated. Two ranges of K-Ar ages were obtained: 183 ± 2 Ma (2 samples), 150 ± 2 Ma (7 samples) and an additional age at 116 ± 2 Ma. All ages are consistently younger than the Triassic sedimentation. They further strongly suggest that at least two mineralizing fluid flows occurred before the maximal burial of the sediments (Upper Cretaceous). These ages fit with the identified episodes of successive opening stages of the central Atlantic Ocean where increasing heat flows and associated fluid circulations have induced: (i) several Pb-Zn-F-Ba mineralizations along the margin of the basin, and (ii) extensive illitization in Triassic and Permian sandstones in many locations of Western Europe.

The study shows also that each studied aquifer underwent an individual and specific diagenetic history that highlights the role of the Callovian-Oxfordian claystones as an efficient hydrological barrier between the limestone units. This effective isolation is of major engineering importance as these claystones represent the currently studied target for a long-term geological disposal of radioactive waste.

Complementarity of computational molecular modelling and experimental techniques to study trace elements geochemistry

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Chemical reactions at the interfaces between soils components and water play an important role in numerous natural processes, like in particular the fate and behaviour of potentially toxic trace metals. One efficient approach has recently emerged to address this question. It consists in the combination of X-ray absorption spectroscopy (XAS) and isotopic techniques. Besides this experimental approach, computational molecular modelling can also contribute significantly to improve our understanding of the atomic-scale processes that control the biogeochemical cycles of trace elements. This presentation will expose the complementarity of both experimental and theoretical approaches. Within the same theoretical framework, it is now possible (i) to calculate the local structural and electronic properties of any atom in the studied system and compare these properties with XAS data, (ii) to model XANES spectra, which allows a better interpretation of spectral features, and (iii) to determine the equilibrium fractionation factors of traditional as well as non-traditional isotopes, associated with any structural site of the system. Several applications will be presented, including the arsenic adsorption complexes at the hematite surface [1], the calculated XANES spectra of 3d transition metal compounds [2], and the isotopic properties of Al- and OH-bearing hematite [3].

[1] Blanchard *et al.* (submitted) *GCA*. [2] Cabaret *et al.* (2010) *PCCP* **12**, 5619-5633. [3] Blanchard *et al.* (2010) *GCA* **74**, 3948-3962.