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CARBONATION OF A PORTLAND CEMENT UNDER GEOLOGICAL CONDITIONS
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Geological storage of CO2 could be a viable way of limiting the effect of anthropogenic carbon dioxide emissions on the global warming. However, the containment of the gas has to be ensured and the understanding of how CO2 could leak out of the sequestration formation is of great importance. The loss of the integrity of one or several wells located on the storage site represents the greatest risk of CO2 leakage. For example, cement carbonation is one of the mechanism which can impair sealing capacity of a well. The knowledge of the long-term evolution of a hardened portland cement exposed to CO2-rich fluids is therefore a key issue to ensure confidence CO2 geological storage. Reactive transport modelling appears as the most reliable way to forecast the cement annular at very long term. However, reactive transport codes require reliable input data, calibration and validation with respect to experimental data in order to accurately predict the evolution of cement mineralogy and porosity at long term. In the recent years, carbonation of portland cement has been extensively investigated leading to the identification of alteration mechanisms. However, the published results dealing with the carbonation mechanisms are essentially qualitative ones. Yet, the calibration of geochemical modeling requires quantitative data on both the cement matrix mineralogy changes and the chemical composition of the water. Besides, the evolution versus time of the altered depth for a cement paste exposed to CO2-ladden fluids has not been clarified yet. Some published results showed a linear evolution of the carbonated depth profile as a function of the square root of time although others works proposed different equations. The purpose of this study is to investigate the mineralogy changes in cement samples due to CO2-rich water exposure in order to get quantifying data. The second goal is to clarify the carbonation kinetics under geological conditions, and to propose an analytical model allowing to describe the evolution of the carbonation depth with the ageing time. The conditions of experiments were 80°C under 5 MPa of CO2. The ageing experiments lasted 14, 28 and 87 days. The evolution of the mineralogy and chemical changes were studied by combining several analytical techniques such as X-ray diffraction (XRD), infrared spectrometry (FTIR), thermogravimetric analysis (TGA) and electron probe micro-analysis (EPMA). TGA allowed to quantify both portlandite and calcium carbonates amounts at different depths from the core to the edge of the samples. These methods showed that, after a 87-day exposure, the core of the samples remained unaltered whereas, at the edge of the specimens, portlandite is fully dissolved and calcium carbonates precipitated. FTIR measurements showed the formation of amorphous silica and the Calcium depletion of calcium silicate hydrates in the carbonated zones.
The alteration thickness determined by EPMA increased from about 2-2.5 mm for the 14-day aged sample to about 4-4.5 mm for the 87-day aged sample. The carbonation depth profile does not evolve linearly with the square root of time. Our findings suggest that the carbonation is not necessarily a diffusion-controlled process.
Moreover, the regular samplings of the ageing solution during the experiments allowed us to determine the composition of the pore water. The quantification of the different species in the solution (Iron, Aluminum, Calcium, Silica, Sodium, Potassium and Sulfates ions) are used for speciation calculations which help us to determine the saturation state of minerals present in the cement. The speciation calculations are performed with Arxim, a geochemical code developed by Ecole des Mines-Saint Etienne and IFP Energies nouvelles.
Modelling of the carbonation experiments with Arxim are under progress.