Carbonation of a Portland cement under geological conditions

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Industrial context: Geological storage of CO_2

Key issue: to guaranty the containment of CO_2: well integrity

- Objectives:
  - To study the long-term evolution of portland cement exposed to CO_2 thanks to reactive transport modeling (COORES™-Arxim©).
  - To produce quantitative data on cement carbonation mechanisms
  - To validate reactive transport simulations
- Experimental program:
  - Mineralogy changes in cement samples due to CO_2-rich water exposure
  - Carbonation kinetics under geological conditions
  - Chemical composition of the pore water

Carbonation tests - experimental conditions

- Ageing of cement in CO_2-rich water
  - $P(CO_2)$ = 50bar (725PSI)
  - T = 80°C (176°F)
  - Ageing durations: 14, 28, 87 days
- Periodical samplings of the ageing solution during the tests
  - Determination of aqueous species concentrations
  - Speciation calculations

Pressurized vessel

Characterization after carbonation

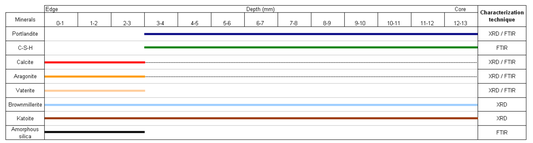
Cement sample aged during 14 days. (L=50mm, Ø=25mm)

Slice cut in the cylinder (w=5mm)

Prismatic specimen cut in the slice (w=5mm) and used for analysis

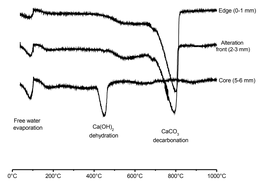
- Cutting of a prismatic specimen in pieces from the edge to the core: for analysis of pieces by XRD, FTIR and TGA
- SEM and EPMA analysis on a prismatic specimen

Mineralogy changes: XRD and FTIR

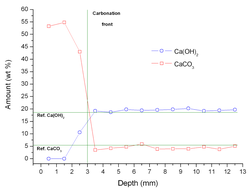


Evolution of the mineralogy from the edge (left) to the core (right) of the sample aged during 14 days in CO_2-rich water. (Dotted lines are equivalent to traces)

Quantification of portlandite and carbonates: TGA

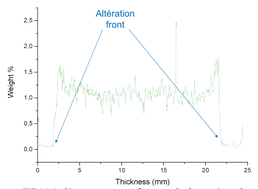


TGA results from sample aged during 14 days under CO_2-rich water

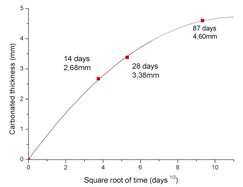


Evolution of $Ca(OH)_2$ and $CaCO_3$ concentrations from the core to the edge of the sample aged during 14 days

Carbonation kinetics



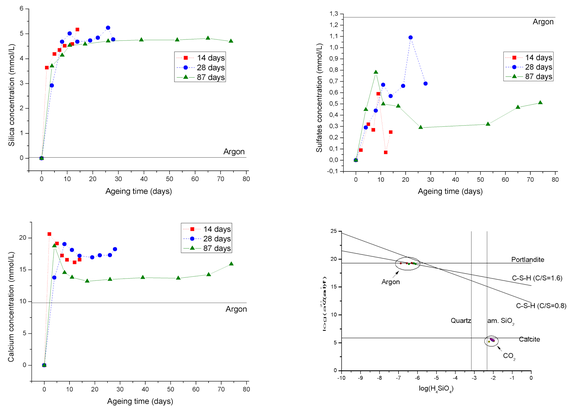
EPMA line scans for sulphur in the sample aged during 14 days under CO_2-rich water



Evolution of the alteration thickness with squared root of time determined from results of EPMA for sulphur.

- Literature review:
  - Carbonation thickness does not evolve linearly with $t^{1/2}$
    - Carbonation of a cement paste under geological conditions is not a diffusion controlled mechanism
    - Hypothesis: Carbonation reactions are the rate-controlling processes (Thiery et al. 2007)

Composition of the pore water



Conclusions and Perspectives

- Conclusions:
  - Accurate determination of the evolution of the mineralogy from the core to the edge of the cylindrical samples and of the carbonation depth thanks to EPMA measurement
  - Carbonation in CO_2-rich water is not a diffusion-controlled process (in agreement with Kutchko et al. 2008)
- Undergoing work:
  - Accurate quantification of the 3 calcium carbonates : calcite, aragonite, vaterite (FTIR)
  - A 6-month carbonation test to clarify the alteration kinetics
  - Development of a carbonation model taking into account the kinetics of chemical reactions
  - Geochemical modeling of carbonation experiments is in progress with COORES™-Arxim©

References

- Rimmelé et al., Cement and Concrete Research, 2008
- Kutchko et al., Environmental Science and Technology, 2008
- Carey et al., International Journal of Greenhouse Gas Control, 2007
- Thiery et al., Cement and Concrete Research, 2007
- Arxim: http://www.emse.fr/~moutte