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Mineral distribution of carbonated Portland cement, an indicator of the coupled effects of transport, chemistry and mechanics

J. Corvisier (1), A. Fabbri (1), S. Bernard (1), F. Brunet (1), A. Schubnel (1), B. Goffé (1), G. Rimmelé (2), V. Barlet-Gouédard (2)

(1) Laboratoire de Géologie, Ecole Normale Supérieure, CNRS UMR8538, Paris, France, (2) Schlumberger, Riboud Product Centre (SRPC), Clamart, France (corvisier@geologie.ens.fr)

A major concern of underground CO_2 storage in depleted oil and gas reservoirs or in deep saline aquifers is to ensure the long-term wellbore integrity. CO_2 injection causes cement pore water acidification and leads to different dissolution/precipitation mechanisms. Portland cement strongly reacts with wet supercritical CO_2 or CO_2-satured water, especially in the context of deep geological storage. The corresponding alteration process is characterized by a complex series of dissolution/carbonation fronts (Barlet-Gouédart *et al.*, 2007) related to the diffusion of aqueous species and also has an impact on the hydromechanical cement properties.

Scanning Portland cement samples which were experimentally carbonated (at 90˚C and 28 MPa) with a Raman Spectrometer using near infrared laser to minimize fluorescence effects often encountered when dealing with cement phases, allowed us to distinguish three $CaCO_3$ polymorphs: calcite, aragonite and vaterite. Raman maps show that vaterite which is the least stable polymorph is mainly located at the vicinity of the carbonation front, consistently with the Ostwald rule which predicts that metastable phases first crystallize in such systems far from equilibrium.

A computer program purely diffusive 1D transport with simple boundary conditions (e.g. infinite source of pure CO_2-H_2O fluid) has been build; it is able to model the coupled effects of chemical reactions and fluid transport and also to handle the competition between their respective kinetics. The obtained simulations reproduce well

the dissolution and carbonation fronts observed in laboratory experiments with the same boundary and P-T conditions. A porosity profile composed of three main zones is predicted from sample centre to rim: a dissolution zone where portlandite partly dissolved; a carbonation zone where the competition between the different polymorphs can be tested and a dissolution backfront where carbonates are dissolved due to acidic boundary conditions.

In order to link this work with the induced mechanical effects, tri-axial press experiments combined with P and S elastic wave velocities have been performed. They allowed us to characterize the density and the distribution of cracks within the samples. These micro-cracks are likely to concentrate at the carbonation front and may result from carbonation due to pore pressure increase. Even if experimental decompression side-effects cannot be ruled out, pore pressure calculation indicates that the first option is the more likely.

Reference:

Barlet-Gouédard, V., Rimmelé, G., Goffé, B. and Porcherie, O. (2007) Well technologies for CO_2 geological storage: CO_2-resistant cement. *Oil & Gas Science and Technology - Rev.IFP*, **62**, 325-334.