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Cement/clay interactions: feedback on the increasing complexity of modeling assumptions

Nicolas C.M. Marty¹, Isabelle Munier², Eric C. Gaucher¹, Christophe Tournassat¹, Stéphane Gaboreau¹, Chan Quang Vong¹, Benoit Cochepein², F. Claret¹

1. BRGM, 45060 Orleans Cedex, France (n.marty@brgm.fr)

2. ANDRA, 1/7 Rue Jean Monnet, F-92298 Châtenay-Malabry Cedex, France

Cementitious materials will be widely used in French concept of radioactive waste repositories. During their degradation over time, in contact with geological pore water, they will release hyper-alkaline fluids rich in calcium and alkaline cations. This chemical gradient likely to develop at the cement/clay interfaces will induce geochemical transformations. The first simplified calculations based mainly on simple mass balance calculation led to a very pessimistic understanding of the real expansion mechanism of the alkaline plume (see examples in Gaucher & Blanc, 2006). However, geochemical and migration processes are much more complex because of the dissolution of the barrier's accessory phases and the precipitation of secondary minerals. To describe and to understand this complexity, coupled geochemistry and transport calculations are a useful and a mandatory tool. Furthermore, such sets of modeling when properly calibrated on experimental results are able to give insights on larger time scale unreachable with experiments. Since approximately 20 years, numerous papers have described the results of reactive transport modeling of cement/clay interactions with various numerical assumptions. For example, some authors selected a purely thermodynamic approach (Adler *et al.*, 1999; Burnol *et al.*, 2006; Gaucher *et al.*, 2004; Trotignon *et al.*, 2006) while others preferred a coupled thermodynamic/kinetic approach (De Windt *et al.*, 2008; 2004; Fernandez *et al.*, 2010; Marty *et al.*, 2009; Savage *et al.*, 2010; 2002; Soler, 2003; Soler *et al.*, 2011; Steefel & Lichtner, 1994; 1998; Trotignon *et al.*, 2007; Vieillard *et al.*, 2004; Watson *et al.*, 2009). Unfortunately, most of these studies used different and not comparable parameters as space discretization, initial and boundary conditions, thermodynamic databases, clayey and cementitious materials, etc... This study revisits the types of simulations proposed in the past to represent the effect of an alkaline perturbation with regard to the degree of complexity that was considered. The main goal of the study is to perform simulations with a consistent set of data and an increasing complexity. In doing so, the analysis of numerical results will give a clear vision of key parameters driving the expansion of alteration fronts and mineralogical transformations. In addition, the validation of some simplifying hypotheses in previous approaches will be tackled. In the present paper, the following modeling cases are considered:

- alkaline fluid (Dirichlet boundary condition) in contact with clay stone;
- consideration of a cementitious medium (CEM I, pH 12.5);
- introduction of alkaline cations (CEM I, pH 13.2);
- consideration of zeolites as secondary phases;
- transition from 1D Cartesian geometry to 1D radial;
- incorporation of reaction kinetics;
- investigation of a damaged zone (EDZ) around the drift;
- feedback effect of mineralogical transformations on the porosity and then on diffusive transport.

In these calculations the clay will be the Callovo-Oxfordian clayrock. The constituent phases selected here are established according to the "Chlorite(CCa2)/Illite(IMt2)" model proposed by Gaucher *et al.* (2009). The concrete system is modelled as an ordinary Portland cement (CEM I) consisting of portlandite, CSH with Ca/Si=1.6, ettringite, minor quantities of hydrotalcite and monocarboaluminate as proposed by Blanc *et al.* (2010a; b).

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