



HAL
open science

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

Nicolas C.M. Marty, I. Munier, Eric C. Gaucher, Christophe Tournassat, Stéphane Gaboreau, Chan Quang Vong, B. Cochevin, Francis Claret

► **To cite this version:**

Nicolas C.M. Marty, I. Munier, Eric C. Gaucher, Christophe Tournassat, Stéphane Gaboreau, et al.. Cement/clay interactions: feedback on the increasing complexity of modeling assumptions. International meeting "Clays in Natural and Engineered Barriers for Radioactive Waste Confinement", Oct 2012, Montpellier, France. hal-00701808

HAL Id: hal-00701808

<https://brgm.hal.science/hal-00701808v1>

Submitted on 26 May 2012

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

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).

References:

- Adler, M., Mäder, U. K., and Waber, H. N., 1999. High-pH alteration of argillaceous rocks: an experimental study. *Schweiz. Mineral. Petrogr. Mitt.* 79.
- Blanc, P., Bourbon, X., Lassin, A., and Gaucher, E. C., 2010a. Chemical model for cement-based materials: Temperature dependence of thermodynamic functions for nanocrystalline and crystalline C-S-H phases. *Cement and Concrete Research* 40, 851-866.
- Blanc, P., Bourbon, X., Lassin, A., and Gaucher, E. C., 2010b. Chemical model for cement-based materials: Thermodynamic data assessment for phases other than C-S-H. *Cement and Concrete Research* 40, 1360-1374.
- Burnol, A., Blanc, P., Xu, T., Spycher, N., and Gaucher, E., 2006. Uncertainty in predictions of transfer model response to a thermal and alkaline perturbation in clay. *Tough symposium 2006 - Lawrence Berkeley National Laboratory - Berkeley - California - USA.*
- De Windt, L., Marsal, F., Tinseau, E., and Pellegrini, D., 2008. Reactive transport modeling of geochemical interactions at a concrete/argillite interface, Tournemire site (France). *Physics and Chemistry of the Earth, Parts A/B/C* 33, Supplement 1, S295-S305.
- De Windt, L., Pellegrini, D., and van der Lee, J., 2004. Coupled modeling of cement/claystone interactions and radionuclide migration. *J. Contam. Hydrol.* 68, 165-182.
- Fernandez, R., Cuevas, J., and Mader, U. K., 2010. Modeling experimental results of diffusion of alkaline solutions through a compacted bentonite barrier. *Cement and Concrete Research* 40, 1255-1264.
- Gaucher, E. C., Tournassat, C., Pearson, F. J., Blanc, P., Crouzet, C., Lerouge, C., and Altmann, S., 2009. A robust model for pore-water chemistry of clayrock. *Geochim. Cosmochim. Acta* 73, 6470-6487.
- Gaucher, E.C., and Blanc, P., 2006. Cement/clay interactions - A review: Experiments, natural analogues, and modeling. *Waste Management* 26, 776-788.
- Gaucher, E., Blanc, E., Matray, J. M., and Michau, N., 2004. Modelling diffusion of an alkaline plume in a clay barrier. *Appl. Geochem.* 19, 1505-1515.
- Marty N. C. M., Tournassat C., Burnol A., Giffaut, Eric C. Gaucher, 2009. Influence of reaction kinetics and mesh refinement on the numerical modelling of concrete/clay interactions. *Journal of Hydrology* 364, 58-72.
- Trotignon, L., Devallois, V., Peycelon, H., Tiffreau, C., and Bourbon, X., 2007. Predicting the long term durability of concrete engineered barriers in a geological repository for radioactive waste. *Physics and Chemistry of the Earth, Parts A/B/C* 32, 259-274.
- Trotignon, L., Peycelon, H., and Bourbon, X., 2006. Comparison of performance of concrete barriers in a clayey geological medium. *Physics and Chemistry of the Earth, Parts A/B/C* 31, 610-617.
- Savage, D., Benbow, S., Watson, C., Takase, H., Ono, K., Oda, C., and Honda, A., 2010. Natural systems evidence for the alteration of clay under alkaline conditions: An example from Searles Lake, California. *Applied Clay Science* 47, 72-81.
- Savage, D., Noy, D., and Mihara, M., 2002. Modelling the interaction of bentonite with hyperalkaline fluids. *Appl. Geochem.* 17, 207-223.
- Soler, J. M., 2003. Reactive transport modeling of the interaction between a high-pH plume and a fractured marl: the case of Wellenberg. *Appl. Geochem.* 18, 1555-1571.
- Soler, J. M., Vuorio, M., and Hautajärvi, A., 2011. Reactive transport modeling of the interaction between water and a cementitious grout in a fractured rock. Application to ONKALO (Finland). *Appl. Geochem.* 26, 1115-1129.
- Steefel, C. I. and Lichtner, P. C., 1994. Diffusion and reaction in rock matrix bordering a hyperalkaline fluid-filled fracture. *Geochim. Cosmochim. Acta* 58, 3595-3612.
- Steefel, C. I. and Lichtner, P. C., 1998. Multicomponent reactive transport in discrete fractures II: infiltration of hyperalkaline groundwater at Maqarin, Jordan, a natural analogue site. *Journal of Hydrology* 209, 200-224.
- Vieillard, P., Ramirez, S., Bouchet, A., Cassagnabere, A., Meunier, A., and Jacquot, E., 2004. Alteration of the Callovo-Oxfordian clay from Meuse-Haute Marne Underground Laboratory (France) by alkaline solution: II. Modelling of mineral reactions. *Appl. Geochem.* 19, 1699-1709.
- Watson, C., Hane, K., Savage, D., Benbow, S., Cuevas, J., and Fernandez, R., 2009. Reaction and diffusion of cementitious water in bentonite: Results of 'blind' modelling. *Applied Clay Science* 45, 54-69.