

Quantification of Porosity Changes Due to Precipitation of Cement Materials with ICP

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Abstract

The Japanese Atomic Energy Agency (JAEA) is being carried out an URL (underground research laboratory) project in Mizunami city, central Japan. The project is a purpose-built generic URL project that is planned for a scientific study of the deep geological environment as a basis of research and development for geological disposal of nuclear wastes. In the project, groundwater flow model has been developed using the commercial software ConnectFlow, which is capable to generate hydrogeological models but it cannot account chemical processes. COMSOL is capable to include chemical processes in a hydrogeological model using iCP (interface COMSOL-PHREEQC) (Nardi et al., 2014).

The URL geometry and the spatial distribution of hydrogeological conditions (hydraulic conductivity and porosity fields) have been implemented in COMSOL from the ConnectFlow files (Figure 1 and Figure 2). iCP uses COMSOL to solve the flow and the transport. The chemical part is solved by PHREEQC (Parkust and Apelo, 2013). The groundwater flow model has been implemented using the Darcy's law physic and the transport process is implemented using a custom physic interface named as Molal Solute Transport (Nardi et al., 2014b).

The objective of this study was to develop a reactive transport model that reproduces the evolution of the hyperalkaline plume in the bedrock, which is supposed to be generated by the concrete degradation. Precipitated secondary minerals can reduce porosity up to the point to induce important changes on hydraulic conductivity.

The URL consists on two 500 m deep shafts and several galleries. In addition to concrete lining, grout has been injected to minimize groundwater inflow volume. There are up to three meters of rock filled by grout between the non-perturbed rock and the URL walls. The chemical composition of the grout is half of the portlandite and the other half of the CSH (1.67). This is a simplified composition taking into account the principal components involved in the alkalinity. The chemical composition of the groundwater has been obtained from Iwatsuki et al. (2005). Basically, this groundwater is in equilibrium with granites and has a pH of about 8.5.

Preliminary 1D model predicts that the interaction between the hyperalkaline concrete pore water and the natural rock water produces the oversaturation of different minerals phases (Figure 3).

Results of the model show that the high pH plume caused by dissolution of portlandite and CSH extends downstream. High values of pH (pH>11) extend less than 100 m downstream (Figure 4). The mixing between the grout and the natural groundwater produces the precipitation of three minerals with a specific order. The first phases are CSH(0.83) and the hydrotalcite. The precipitation of hydrotalcite consumes most of the OH⁻ of the water and buffers the pH to about 10.5. The last mineral that precipitates is calcite, covering a larger extension in the rock and buffering the pH up to natural values.

This project shows that iCP is a flexible and powerful tool for quantitative integration of hydrogeology and geochemistry. iCP allows to incorporate geochemical processes in 3D large scale COMSOL models and for long-time simulations.

Reference

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Nardi, A., Idiart, A., Trincherro, P., de Vries, L. M. and Molinero, J., 2014a. Interface Comsol-PHREEQC (iCP), an efficient numerical framework for the solution of coupled multiphysics and geochemistry, *Computers & Geosciences*, doi:10.1016/j.cageo. 2014.04.011.

Nardi, A., Idiart, A., Trincherro, P., de Vries, L. M., Sainz, A. and Molinero, J., 2014b. iCP – interface COMSOL PHREEQC. User's manual v1.0.

Parkhurst, D. L. and Appelo, C. A. J., 2013. Description of Input and Examples for PHREEQC Version 3—A Computer Program for Speciation, Batch-Reaction, One-Dimensional Transport, and Inverse Geochemical Calculations. Chapter 53 of Section A, *Groundwater book 6, Modeling Techniques U.S. Geological Survey Water-Resources*.

Figures used in the abstract

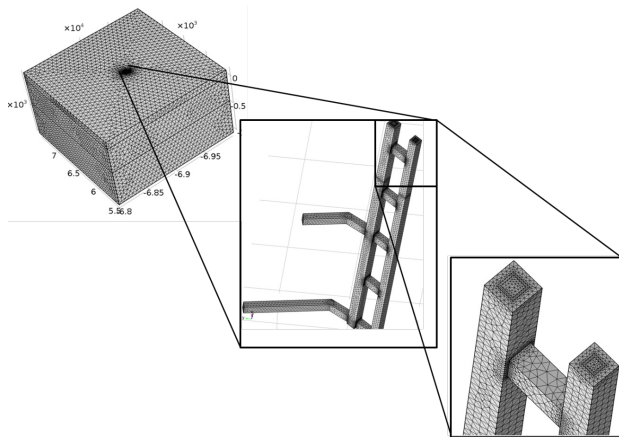


Figure 1: Detail of the URL geometry and the mesh employed in the groundwater flow model. The domain size is (2000x2000x1150 meters) and it is formed by 1,134,449 tetrahedral elements.

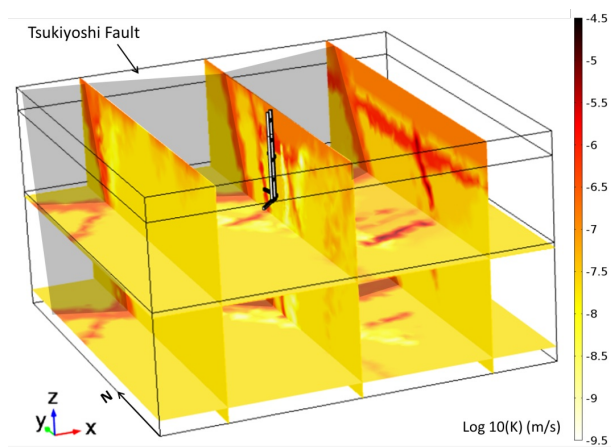
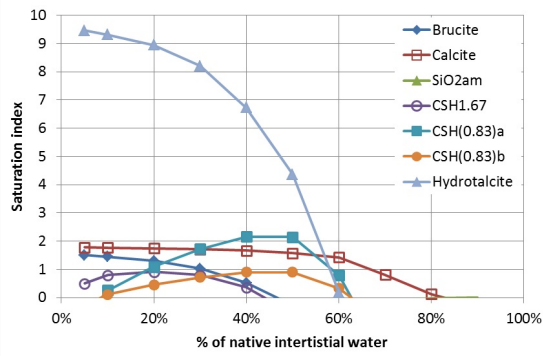


Figure 2: Hydraulic conductivity field of the rock domain implemented in COMSOL.



Mineral notation	Reaction	Log K*
Calcite	$\text{CaCO}_3 = +1.000\text{Ca} + 2.1.000\text{H} + +1.000\text{HCO}_3^-$	1.84897
Brucite	$\text{Mg}(\text{OH})_2 = +1.000\text{Mg} + 2.000\text{H}_2\text{O} - 2.000\text{H} +$	16.83953
Hydrotalcite_OH	$\text{Mg}_4\text{Al}_2(\text{OH})_{14} \cdot 3\text{H}_2\text{O} = 4\text{Mg}^{++} + 2\text{Al}(\text{OH})_4^- + 6\text{OH}^- + 3\text{H}_2\text{O}$	-56.0214
CSH _{en}	$(\text{CaO})_{1.666667}(\text{SiO}_2)(\text{H}_2\text{O})_{2.1} + 0.566667\text{H}_2\text{O} = 1.666667\text{Ca}^{++} + \text{SiO}(\text{OH})_3^- + 2.333334\text{OH}^-$	-13.1659
CSH(0.83a)	$\text{CaO}(\text{SiO}_2)_{2.4}(\text{H}_2\text{O})_{3.2} + 1.2\text{H}_2\text{O} = 2\text{Ca}^{++} + 2.4\text{SiO}(\text{OH})_3^- + 1.6\text{OH}^-$	-19.1991
CSH(0.83b)	$(\text{CaO})_{0.833333}(\text{SiO}_2)(\text{H}_2\text{O})_{1.3333} + 0.50003\text{H}_2\text{O} = 0.83333\text{Ca}^{++} + \text{SiO}(\text{OH})_3^- + 0.66666\text{OH}^-$	-8.0014
Portlandite	$\text{Ca}(\text{OH})_2 = +1.000\text{Ca} + 2.2.000\text{H}_2\text{O} - 2.000\text{H} +$	22.79937
SiO ₂	$\text{SiO}_2 + 10\text{H} + 1\text{H}_2\text{O} = \text{SiO}(\text{OH})_3^-$	1.475988

*Based on CEMDATA07 version 07.02 (14.08.2008)

Figure 3: Preliminary 1D model with mixing between grout material and natural groundwater. The image shows the saturation index of different mineral phases. The table shows the equilibrium constant for the different minerals phases involved in the model.

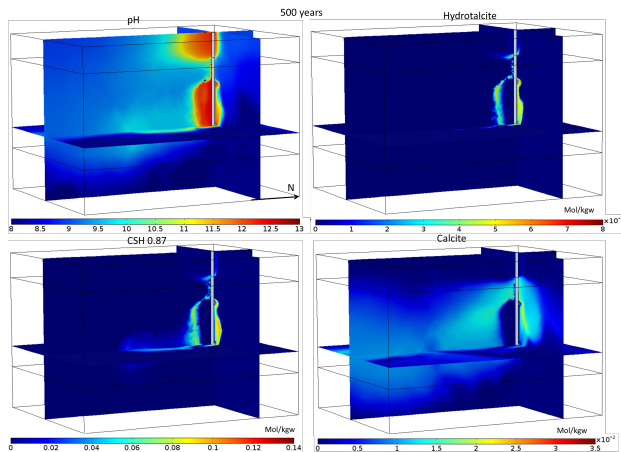


Figure 4: Groundwater pH and different mineral precipitation for a simulation time of 500 years. Groundwater flows southwards.