Chemo-Hydro-Mechanical analysis of Bituminized Waste swelling due to water up-taking: Experimental and model comparisons

G. Melot[1,2], P. Dangla[2], S. Granet[1], S. M'Jahad[1], JB. Champenois[3], A. Poulesquen[3]

[1] IMSIA, UMR9219, EDF-CNRS-CEA-ENSTA Paris Tech, France
2 Univ Paris-Est Laboratoire Navier (UMR 8205) CNRS, Ecole des Ponts ParisTech, IFSTTAR, France
3 CEA, DEN, DTCD, SEAD, LCBC, F-30207, Bagnols sur Cèze, France

Introduction

Bituminized Waste materials (BW) are composed of precipitation sludge from the chemical reprocessing of spent nuclear fuel, immobilised in bitumen matrix. According to the radiological activity, geological storage could be the reference solution for this kind of wastes. Under geological disposal conditions, and after up to hundred thousand years, BW will undergo water re-saturation from host rock. Water up-taking by BW will first induce free swelling until all different types of void existing in the storage environment are filled. Secondly swelling could occur in contact with host rock and generates potential stresses. For those reasons, hydro-mechanical behaviour of BW has to be investigated.

Various models have already been developed to describe water up-take by bituminized waste.

The COLONBO model has been developed by the French Alternative Energies and Atomic Energy Commission (CEA). This operational model describes the evolution as a function of time of the pore structure, the water up-take by the leached bituminized waste and the corresponding salts and radionuclides release in leachates. (Sercombe 2006, Gwinner 2006). This model is based on diffusive mechanisms.

A Chemo-Hydro-Mechanical (CHM) model has been developed by the UPC (University Polytechnic of Catalonia) (Mokni *Applied Clay Science* 2010, Mokni *Journal of Nuclear Materials* 2010, Mokni 2011, Mokni *Transport in porous med*ia 2011) for Belgian BW. This model is based on a classical poromechanical approach considering the balance equations for water, dissolved salts, crystals salt and solid phase. These equations include osmosis and coupled transport phenomena: diffusion and permeation. The dissolution/precipitation of salts is managed by an additional equation. Moreover, the mechanical behaviour takes creep deformations into account.

In a previous paper (Mélot 2017) a simplified model has been proposed, considering one single soluble salt embedded in the bitumen matrix. Constitutive equations of this model have been derived from a classical CHM model, as described by the UPC. For the present work, this model has been implemented in three dimensions with a finite element scheme.

This work aims at comparing numerical and experimental results. Calibration of the model and interpretation have been done for free swelling tests of a BW incorporating 40 wt. % of NaNO3.

Theoretical model

Main part of the constitutive equations of the current CHM model has been presented in a previous paper (Mélot 2017). New equations have been included in order to take into account creep deformations. The main points of the model are reported in the following part.

Bituminized waste can be considered as a porous bitumen matrix with salt crystal inclusions. The medium is composed of four species: water (w), salt as solute in liquid phase (s), salt as crystal (c) and the bitumen porous matrix. In the Chemo-Hydro-Mechanical (CHM) model, the following assumptions have been made: one soluble salt is considered (for ex. NaNO3), the medium is water saturated and the gravity is neglected. This model includes salts dissolution, elastic and creep deformation, osmosis, diffusion of salt and water, advection and compressibility of water. The porosity and the properties of the bitumen matrix evolution during the leaching are also taken into account.

Experimental data and calibration

In the present work, a set of simplified BW product has been elaborated by incorporating an increasing content of NaNO3. Leaching experiments were performed at 25°C by casting 20 g of BW product in 50 mm-diameter polyethylene flasks. Each flask was then filled up with 40 mL of demineralized water and sealed from the atmosphere. Resulting leachates were renewed after fixed period of time (1, 7, 14, 28, 59, 126, 237, 384, 475 and 680

days). For each renewal, BW samples were weighted once leachates were pulled out in order to determine the amount of water uptake. Leachates were collected and analyzed by using ionic chromatography (Metrohm 940 Professional IC Vario) to measure the amount of nitrate and sodium ions that have been leached.

The model has been calibrated by using previously reported experimental data. We also use porosity profiles measured during leaching test, which have been previously published (Gwinner 2006, Gwinner *Thesis* 2004, Gwinner 2004).

The permeability, the osmotic efficiency coefficient and the viscosity of the bitumen have been calibrated. Evolution of the parameters has been investigated. An example of comparison is shown for one typical case in Figure 1.

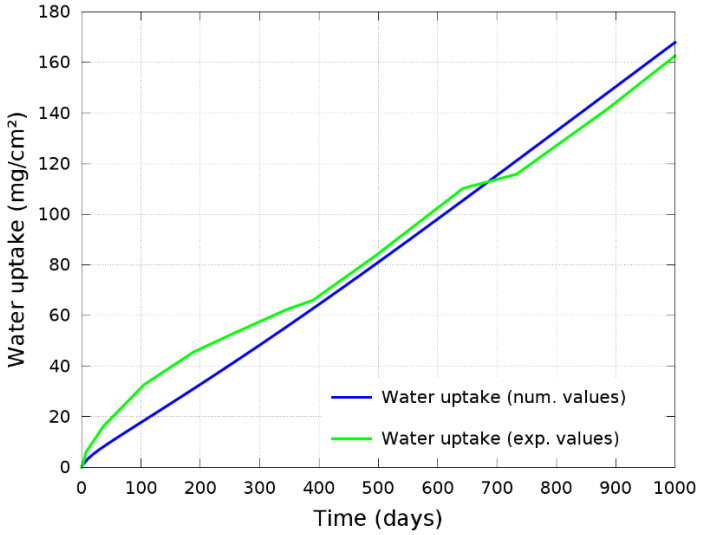

Figure 1: Evolution as a function of time of the uptake water during the leaching by demineralized water of a BW composed of 40 % wt. of $NaNO_3$ - 25°C
Comparison between experimental and numerical values

Conclusions and further works

The CHM model is able to reproduce main leaching results (mass of water absorbed, mass of leached salt and porosity profiles) for several specific cases. Further experimental comparisons are needed to completely validate the theoretical model and to improve the calibration. Moreover, the model could be enhanced in further works in order to reproduce bi-salt behaviour.

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