

# Coupled Chemo-Hydro-Mechanical analysis of Bituminized Waste swelling due to water up-taking

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## Abstract

This paper presents results and discussions about mechanisms and their couplings during bituminized waste water up-taking. Bituminized Waste Products(BWP) are produced by an industrial reprocessing of radioactive waste (low or medium activity and long life). Geological storage is the reference solution for this kind of wastes. Under geological disposal conditions, and after a period of hundred thousand years, BWP will undergo water re-saturation from the host rock. A one dimensional chemo-hydro-mechanical numerical model has been implemented with a finite volume scheme. Using this model, the role of each mechanism and coupling has been evaluated by simulating water up-taking tests under constant volume. Indeed, the impact of dissolution permeation, diffusion and osmosis has been investigated. Moreover, the dependency of material parameters with porosity variations has been studied. This numerical simulation has shown that osmosis is the principal mechanism of water up-taking. Nevertheless, other mechanisms might not be negligible.

**Key words:** Bituminized waste Products (BWP) / Chemo-Hydro-Mechanical analysis / Swelling / Diffusion / Osmosis

## 1 Introduction

Bituminized Waste materials (BWP) were produced by an industrial reprocessing of radioactive waste qualified low or medium activity and long life (LA-LL and MA-LL). BWP is composed of salt crystals immobilised in a bitumen matrix. These salts are present in precipitation sludge produced during the chemical reprocessing of spent nuclear fuel. They could be soluble (e.g.  $\text{NaNO}_3$ ) or weakly soluble (e.g.  $\text{BaSO}_4$ ). BWP have been produced, by the past, in different places in different countries including France, Belgium and Japan. The mechanical properties of the bitumen differ from each other. In the same way, there are various natures and proportions of crystal salts for these different BWP: Belgium BWP are largely occupied by  $\text{NaNO}_3$  while French contains weakly soluble salts. All of chemistry characteristics had to be considered especially under leaching conditions.

Geological storage is the reference solution for this kind of wastes (MA-LL). Under geological disposal conditions, and after a period of hundred

thousand years, storage will undergo water re-saturation from the host rock. Water up-taking by BWP will first induce free swelling in order to fill all different types of void existing in the storage disposal (void in primary canister, void in concrete container and void in rock vault). Then swelling could continue in contact with the host rock and under special stress conditions (confined conditions).

Various models have already been developed for 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 of the pore structure, the water up-take and the salt and radionuclides release of leached bituminized waste [1][2]. This model is based on diffusive mechanisms.

A Chemo-Hydro-Mechanical (CHM) model has been developed by the UPC (University Polytechnic of Catalonia)[3][4][5][6] 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 this paper a simplified model is proposed, considering one single soluble salt. Constitutive equations of this model have been derived from the CHM model of UPC. This simplified model has been developed to evaluate the part of each mechanism and coupling. It has been implemented in one dimension with a finite volume scheme.

The constitutive model is presented in the first part of this paper. Then a validation is proposed on an experiment on a BelgiumBWP. Therefore, previously published measurements of amount of salt leached during BWP water up-tacking are used. These results were obtained by the experimental program on Eurobitume still in progress at the Belgian Research Center SCK. CEN[7]. The diffusion coefficient is also calibrated by this set of tests and confrontations are detailed.

The major aim of this paper is the evaluation of diffusion and osmosis impact on mechanical and chemical behaviour. Therefore, numerical simulations of water up-taking under constant volume have been provided. Moreover, an investigation has been lead on the impact of material parameter variations due to porosity increasing. Finally, this work enhanced the importance of experimental calibration of some of these parameters.

## 2 Constitutive model

Bituminized waste can be considered as a verylow-permeable material containing one or several different salt crystals. In the present model it was chosen to focus only on one kind ofsoluble salt. This approximation is reasonable when one salt is predominant.A coupled model is required in order to describe consequences of water uptake on such a material. Swelling is one of these consequences and is governed by two principal mechanisms. The first mechanism is the solvent transport leading to the dissolution of salt crystals. During dissolution,volume increases, leading to global swelling of the bitumen matrix. The second mechanism is osmotic flows which lead directly to a water overpressure in the pore due to the chemical gradient (salt concentration gradient). The bitumen matrix plays the role of a semi-permeable membrane allowing an increase of the pore water pressure in the bituminized waste (osmotic overpressure). The chemical part of this model deals with the precipitation/dissolution of salt crystals present in the bitumen matrix. It is the principal

driving force of water up-taking, leading to salt saturation in pore water and increasing the porosity. The porosity created by the salt crystals' dissolution allows advective and diffusive transport of water and salt through the bitumen matrix. Permeability is linked to this porosity increase. The mechanical behaviour is strongly dominated by creep-deformation. But in the present paper water up-taking simulations are only done under constant volume: It seems reasonable to assume that an elastic mechanical model will be sufficient.

The following assumptions have been made:

- One soluble salt is considered ( $\text{NaNO}_3$ )
- The medium is water saturated
- Gravity is neglected

The medium is composed of four species:

- Water (e)
- Salt as solute in liquid phase (s)
- Salt as crystal (c)
- Bitumen porous matrix

The model is based on the mass balance equations for water (1), dissolved salt(2) and crystals salt (3):

$$\frac{\partial}{\partial t}(\omega_l \rho_l \phi_l) + \nabla j_e = 0 \quad (1)$$

$$\frac{\partial}{\partial t}(\omega_s \rho_l \phi_l) + \nabla j_s - \frac{dm}{dt} = 0 \quad (2)$$

$$\frac{\partial}{\partial t}(\rho_c \phi_c) + \frac{dm}{dt} = 0 \quad (3)$$

Where  $\omega_l(-)$  denotes the mass fraction of water in the liquid phase,  $\rho_l(kg/m^3)$  the liquid density,  $\phi_l(-)$  the volume fraction of the liquid phase,  $j_e$  the total flux of water,  $\omega_s(-)$  the mass fraction of solute in the liquid phase,  $j_s$  the total flux of solute,  $\rho_c(kg/m^3)$  the crystal density and  $\phi_c(-)$  is the volume fraction of crystals. The total fluxes of water and solute are detailed later in this paper. The time derivative term  $\frac{dm}{dt}$  is the dissolution/precipitation term. The function for this term is derived from the one used in the UPC model [4]:

$$\frac{dm}{dt} = \phi_c \sigma_c \beta \xi \left[ \left( \frac{\omega_s}{(\omega_s)_{sat}} \right)^\theta - 1 \right]^\eta \quad (4)$$

Where  $\sigma_c (m^2/m^3)$  is the specific surface,  $\beta (kg.s^{-1}.m^{-3})$  the rate constant,  $\xi (-)$  the sign of the reaction (positive for dissolution and negative for precipitation),  $(\omega_s)_{sat} (-)$  the mass fraction of solute in the liquid phase at saturation, and  $\theta (-)$  and  $\eta (-)$  are

experimental calibration parameters (the value 1 was chosen in the present work).

Water and solute fluxes are built from permeation, osmosis and diffusion mechanisms. We will describe in the following each of them.

The Darcy law is used to describe liquid flux through the porous bitumen matrix due to pressure gradient.

$$J_{Darcy} = -\frac{k}{\mu} \nabla p \quad (5)$$

Where  $J_{Darcy}$  is the Darcean flux,  $k$  ( $m^2$ ) the permeability,  $\mu$  ( $Pa \cdot s$ ) the dynamic viscosity of the water and  $p$  ( $MPa$ ) the pore pressure.

The bitumen matrix play the role of a semi-permeable membrane. That means that solute transport through the bitumen matrix is restrained when water transport is free. To describe the quality of the membrane a parameter  $\tau$  (-), named the efficiency coefficient, is used. When  $\tau = 1$  the semi-permeable membrane is perfect, solute cannot transport through it. When  $\tau = 0$  the membrane has no effect. A chemical gradient in this kind of material induces osmotic pressure. The liquid flux due to this osmotic pressure gradient is derived from the Darcy law previously introduced. It can be written as:

$$J_{Osm} = \frac{k}{\mu} \frac{R \cdot T \cdot \rho_l}{M_s} \tau \nabla \omega_s \quad (6)$$

Where  $J_{Osm}$  denotes the osmotic flux,  $R$  ( $J \cdot mol^{-1} \cdot K^{-1}$ ) the ideal gas constant,  $T$  ( $K$ ) the temperature and  $M_s$  ( $g/mol$ ) the molar mass of the solute.

The diffusive flux is defined by the Fick's law. Diffusion fluxes are limited by the semi permeable membrane. The diffusion coefficient is corrected to take into account this mechanism:

$$J_{diff} = -D_0(1 - \tau) \rho_l \nabla \omega_s \quad (7)$$

Where  $J_{diff}$  is the diffusive flux and  $D_0$  ( $m^2 \cdot s^{-1}$ ) the diffusion coefficient. The term  $D_0(1 - \tau)$  is named effective diffusion coefficient.

The liquid total flux  $J_l$  is the addition of the Darcean and osmotic fluxes:

$$J_l = J_{Darcy} + J_{Osm} \quad (8)$$

The diffusion contribution is taken into account by a non-advective flux of salt ( $J_s$ ):

$$J_s = J_{diff} - \tau \cdot (\rho_l \omega_s) J_l \quad (9)$$

Total fluxes of solute ( $j_s$ ) and water ( $j_e$ ) are a combination of the non-advective and advective fluxes such as:

$$j_s = J_s + (\rho_l \omega_s) J_l \quad (10)$$

$$j_e = -J_s + \rho_l(1 - \omega_s) J_l \quad (11)$$

The mechanical responses of the BW during water up-taking has to be managed in combination with transport and chemical response. From a mechanical point of view the BW is considered as a homogenized mixture. For a material with no volume forces (gravity is neglected) the total stress equilibrium can be written as:

$$\nabla \sigma = 0 \quad (12)$$

Where  $\sigma$  ( $MPa$ ) denotes the total stress. The total stress can be decomposed referring to the Terzaghi principle:

$$\sigma = \sigma' - p \quad (13)$$

Where  $\sigma'$  ( $MPa$ ) is the effective stress. For the present analysis the mechanical model is based on the perfect elasticity equation. The Hooke's law can be written for a monodimensional problem as:

$$\sigma' = E \varepsilon = E \nabla u \quad (14)$$

Where  $\varepsilon$  (-) is the deformation and  $u$  (m) the displacement.

### 3 Constant volume test case description

In order to study the capacity of the model to reproduce the behaviour of BW, water up-taking tests under constant volume are modelled. This test is described in Mariën et al. (2013) [7].

The sample has a length of 10 mm. Due to symmetry condition, half of the BW sample is represented (Fig. 1). At left of the domain, BW is in contact with water at low  $NaNO_3$  concentration. This water injected in the sample (called free water) is at the atmospheric pressure. At the other side, classic symmetry boundary conditions are used. In both sides of the domain, displacements are blocked.

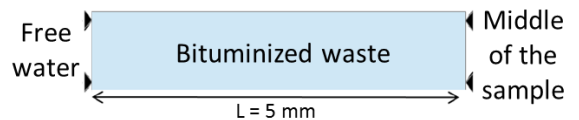


Fig. 1 Schematic representation of the model domain

The bituminized waste initially contains sodium nitrate crystals occupying 16% of the total volume i.e.  $\phi_c = 0.16$  (corresponding to 28% in the weight). The sample is water saturated. The porosity is defined as the volume fraction accessible by the liquid phase. We assume a small initial value: 1% of the total volume ( $\phi_l = 0.01$ ). This initial value is purely numerical. Initially, we consider pore water pressure equal to 1 atm. The rest is occupied by the bitumen matrix (83% of the volume). The water is initially considered  $NaNO_3$  saturated (initial mass fraction of solute in water:  $(\omega_s)_{t=0} = (\omega_s)_{sat} = 0.47$ ).

#### 4 An experimental validation

Measurements of amount of leached salt during water up-taking tests under constant volume are published in Mariën et al. (2013) [7]. Fig. 2 shows the evolution with the square root of time. These experimental results are used in the present paper to explore the model's ability to reproduce the leaching kinetic.

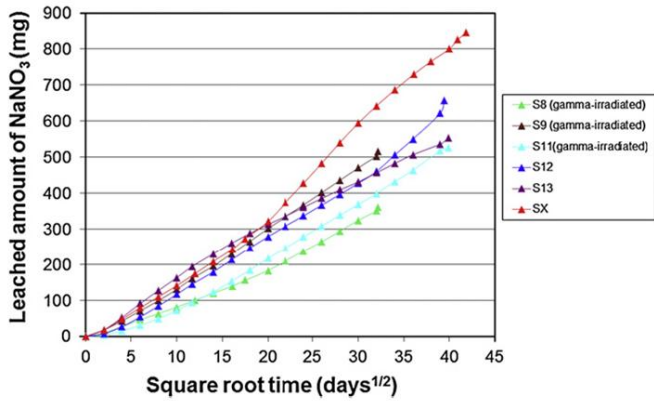


Fig. 2 Cumulative amount of  $NaNO_3$  that has leached out of Eurobitume sample (diameter 38 mm, height 10 mm) with 28.5 wt%  $NaNO_3$  being hydrated in nearly constant volume conditions with 0.1 M KOH [7]

In the present study, material parameters values are the classical ones used to model Eurobitume behaviour [6]:

Table 1 - Material parameters

Parameter	Notation	Value
Intrinsic permeability	$k$	$3.10^{-26} m^2$
Efficiency coefficient	$\tau$	0,95
Liquid density	$\rho_l$	$1000 kg.m^{-3}$
Crystal salt density	$\rho_c$	$2260 kg.m^{-3}$
Dissolution rate	$\beta$	$10^{-5} kg.s^{-1}.m^{-3}$
Crystal specific surface	$\sigma_c$	$148 m^2.m^{-3}$
Solute molar mass	$M_s$	$85 g.mol^{-1}$
Dynamic viscosity of water	$\mu$	$10^{-3} Pa.s$

The diffusion coefficient is used to calibrate the numerical model.

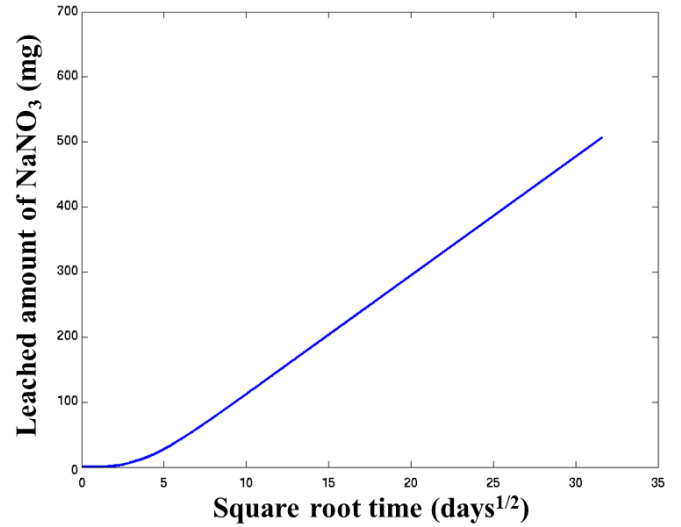


Fig. 3 Numerical results of cumulative amount of  $NaNO_3$  that has leached out of the modelled domain.

The numerical results are presented in the Fig. 3. The present model shows a good ability to reproduce the qualitative kinetic of the BW leaching. The calibration of the model give the value of the diffusion coefficient:  $D = 5.10^{-16} m^2.s^{-1}$ . This quantitative value should be considered cautiously and the present work is about qualitative analysis.

As a conclusion, this experimental validation point out the ability of the numerical model to reproduce the leaching kinetic. This numerical model seems appropriate to make a qualitative analysis of the behaviour of BWP water up-taking. Quantitative results will be discussed in further publications.

#### 5 Numerical results analysis

In this paper, the contribution of diffusion and osmosis are first investigated. Secondly, the impact of the coupling, i.e. the variation of material parameters with other quantities, is analysed. Precisely, we focus on the influence of porosity on permeability, diffusion and osmosis.

In the following, we present the evolution of the solute mass fraction as an indicator of the transport kinetic and the pore water pressure as an indicator for the hydraulic and mechanical behaviour. Indeed total stresses are directly dependent on this value.

##### 5.1 Diffusion impact

The goal of this part is to study the impact of the diffusion on the kinetic and mechanical behaviour. Two water up-taking tests are simulated with two diffusion coefficients  $D_0 = 1,3.10^{-15} m^2.s^{-1}$  and  $D_0 = 2,6.10^{-14} m^2.s^{-1}$ . All others parameters are constants.

Fig. 4 to Fig. 7 show respectively for coefficients  $D_0 = 1,3 \cdot 10^{-15} \text{ m}^2 \cdot \text{s}^{-1}$  (cf. Fig. 4 and Fig. 6) and  $D_0 = 2,6 \cdot 10^{-14} \text{ m}^2 \cdot \text{s}^{-1}$  (cf. Fig. 5 and Fig. 7), the solute mass fraction and the pore pressure profiles along the sample at several time.

We observe that, with the increasing of the diffusion coefficient:

- i. The transport kinetic increases
- ii. The pore water pressure increases

The diffusion coefficient increase implies an acceleration of the velocity of the solute transport through the bitumen matrix. Consequently, the solute mass fraction in the pore water decreases faster causing the enhancement of crystals salt dissolution kinetic. As volumetric mass of crystallized salt is superior to liquid density, the salt volume increases with dissolution. This generates pore pressure increases about 90 percent.

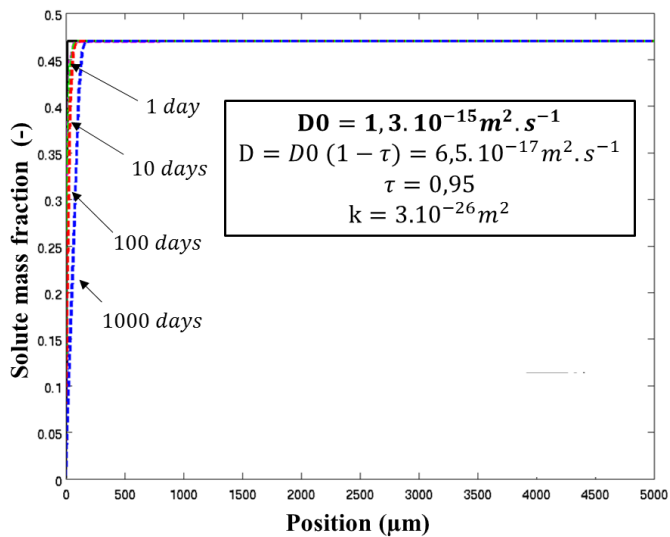


Fig. 4 Solute mass fraction profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with the diffusion coefficient  $D_0 = 2,6 \cdot 10^{-14} \text{ m}^2 \cdot \text{s}^{-1}$ , the effective diffusive coefficient  $D = D_0(1 - \tau) = 1,3 \cdot 10^{-15} \text{ m}^2 \cdot \text{s}^{-1}$ , the efficiency coefficient  $\tau = 0,95$  and the permeability  $k = 3 \cdot 10^{-26} \text{ m}^2$ .

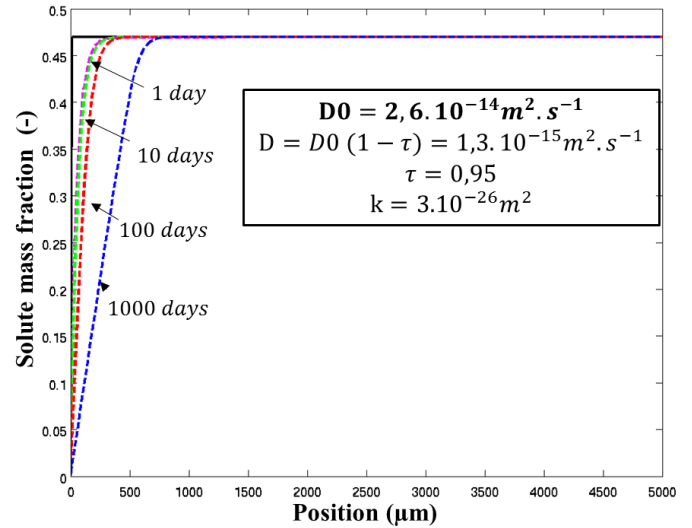


Fig. 5 Solute mass fraction profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with the diffusion coefficient  $D_0 = 1,3 \cdot 10^{-15} \text{ m}^2 \cdot \text{s}^{-1}$ , the effective diffusive coefficient  $D = D_0(1 - \tau) = 6,5 \cdot 10^{-17} \text{ m}^2 \cdot \text{s}^{-1}$ , the efficiency coefficient  $\tau = 0,95$  and the permeability  $k = 3 \cdot 10^{-26} \text{ m}^2$ .

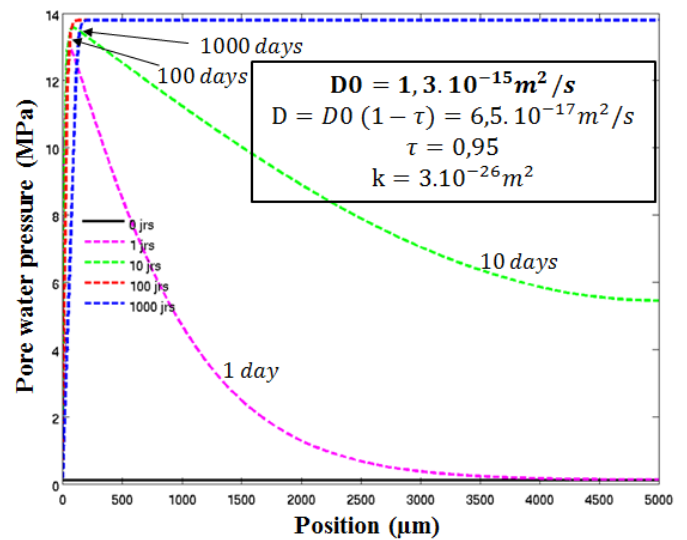


Fig. 6 Pore water pressure profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with the diffusion coefficient  $D_0 = 2,6 \cdot 10^{-14} \text{ m}^2 \cdot \text{s}^{-1}$ , the effective diffusive coefficient  $D = D_0(1 - \tau) = 1,3 \cdot 10^{-15} \text{ m}^2 \cdot \text{s}^{-1}$ , the efficiency coefficient  $\tau = 0,95$  and the permeability  $k = 3 \cdot 10^{-26} \text{ m}^2$ .

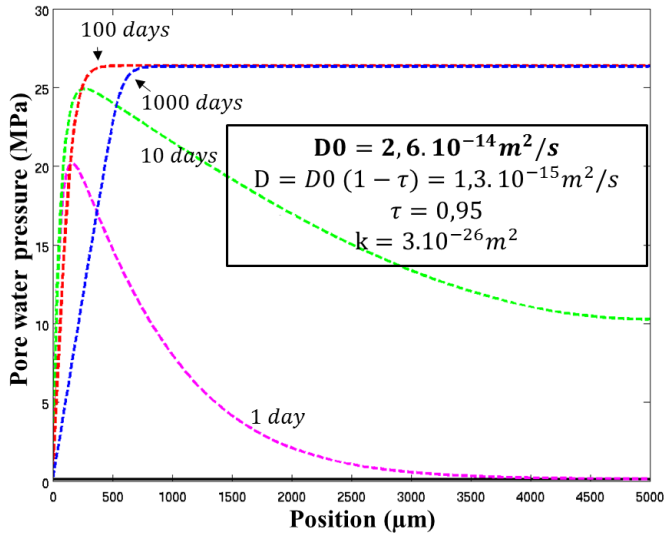


Fig. 7 Pore water pressure profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with the diffusion coefficient  $D_0 = 1,3 \cdot 10^{-15} \text{ m}^2 \cdot \text{s}^{-1}$ , the effective diffusive coefficient  $D = D_0(1 - \tau) = 6,5 \cdot 10^{-17} \text{ m}^2 \cdot \text{s}^{-1}$ , the efficiency coefficient  $\tau = 0,95$  and the permeability  $k = 3 \cdot 10^{-26} \text{ m}^2$ .

### 5.2 Osmosis impact

The goal of this part is to study the impact of the osmosis on the kinetic and mechanical behaviour. Two water up-taking tests are simulated with two efficiency coefficients  $\tau = 0$  and  $\tau = 0,95$ . All other parameters are constants. However the effective diffusive coefficient  $D$  is impacted by the osmosis efficiency coefficient  $\tau$ . Indeed it can be written as  $D = D_0(1 - \tau)$ . The value of the diffusion coefficient is adapted in order to obtain the same value of effective diffusive coefficient in both cases.

The Fig. 8 and Fig. 9 show the evolution of the solute mass fraction profile in the sample during the water up-taking respectively for  $\tau = 0$  and  $\tau = 0,95$ . Both results are similar and the kinetic does not seem to be influenced by the variations of the efficiency coefficient. In both cases, the effective diffusive coefficient is the same, so the diffusive flux is unchanged. Solute advection with the water flux is slowed with the increasing of the efficiency coefficient but in this case the impact is negligible. That can be explained by the low level of liquid fluxes due to the low permeability of the bitumen matrix ( $k = 3 \cdot 10^{-26} \text{ m}^2$ ).

The Fig. 10 and Fig. 11 show the evolution of the pore pressure profile in the sample with the two coefficients ( $\tau = 0$  and  $\tau = 0,95$ ). The pore water pressure strongly increase, about 60 percent, due to osmosis mechanism.

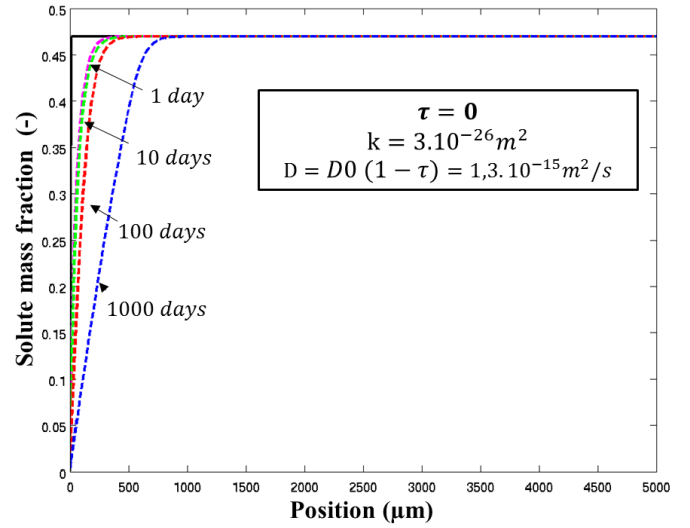


Fig. 8 Solute mass fraction profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with the efficiency coefficient equal to 0.

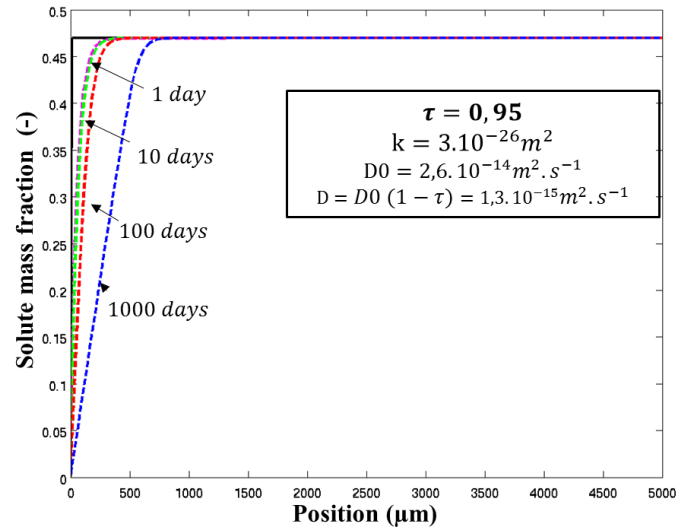


Fig. 9 Solute mass fraction profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with efficiency coefficient equal to 0,95, the permeability  $3 \cdot 10^{-26} \text{ m}^2$  and the effective diffusive coefficient  $D = D_0(1 - \tau) = 1,3 \cdot 10^{-15} \text{ m}^2 \cdot \text{s}^{-1}$ .

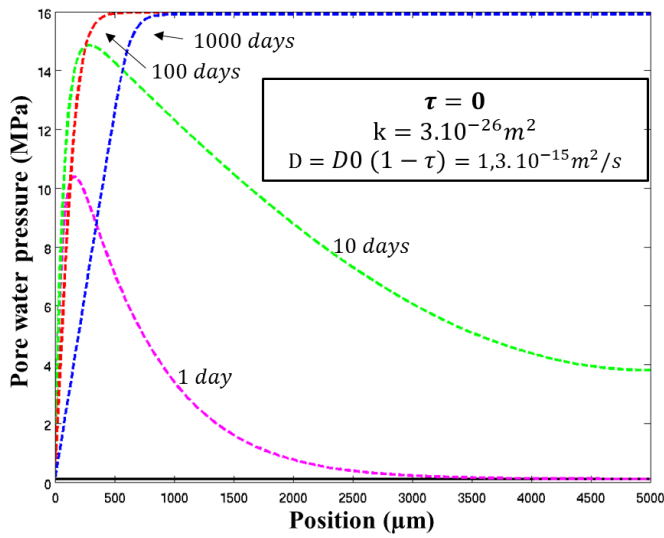


Fig. 10 Pore water pressure profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with efficiency coefficient equal to 0, the permeability  $3.10^{-26}m^2$  and the effective diffusive coefficient  $D = D0(1 - \tau) = 1,3.10^{-15}m^2 \cdot s^{-1}$ .

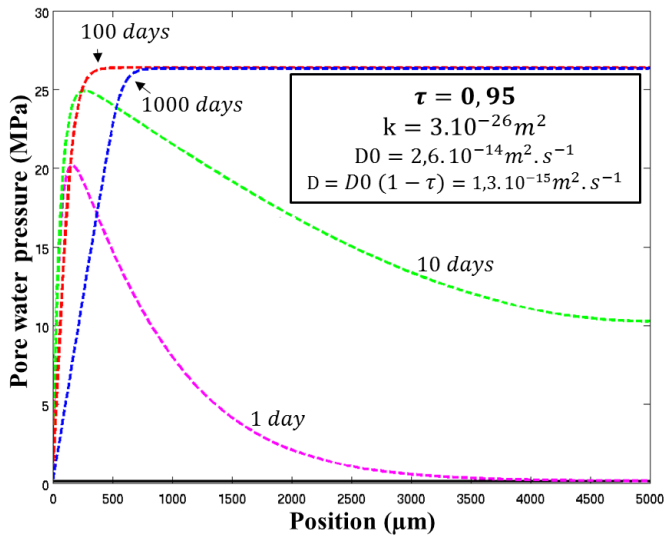


Fig. 11 Pore water pressure profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with efficiency coefficient equal to 0,95, the permeability  $3.10^{-26}m^2$  and the effective diffusive coefficient  $D = D0(1 - \tau) = 1,3.10^{-15}m^2 \cdot s^{-1}$ .

### 5.3 Permeability, diffusion and osmosis evolution impact

During the BW leaching, salt crystals dissolution occurs. This implies an increase of the porosity. Initially salt crystal occupying 16% of the volume and porosity 1%. That means that a complete dissolution of crystal could multiply the void volume (porosity) by 17 (e.g. Fig. 12). This mechanism strongly modifies the material properties. That's why the permeability, the diffusion coefficient and the efficiency coefficient need to be considered as function of the porosity. The following evolution equations are proposed in order to study the qualitative impact of porosity[6]:

$$k = f(\phi_l) = k_i \left( \frac{\phi_l}{\phi_{li}} \right)^3 \left( \frac{1 - \phi_{li}}{1 - \phi_l} \right)^2 \quad (15)$$

$$D0 = f(\phi_l) = D0_i \frac{\phi_l}{\phi_{li}} \quad (16)$$

$$\tau = f(\phi_l) = \tau_i \frac{\phi_{li}}{\phi_l} \quad (17)$$

Where  $\phi_l$  denotes the porosity,  $\phi_{li}$  the initial porosity,  $k_i$  the initial permeability,  $D0_i$  the initial diffusion coefficient and  $\tau_i$  the initial efficiency coefficient. These functions are postulated, experimental calibration can be expected in further works, in order to apply this model to French BWP

In order to understand the whole problem, the impact of each parameter evolution has to be studied. That's why variation laws have been simulated separately to analyse their impact on the transport kinetic and the mechanical behaviour.

#### 5.3.1 Permeability variation impact

The impact of the variation of the permeability is studied without osmosis mechanism ( $\tau = 0$ ). The Fig. 12 shows the evolution of the porosity profiles in the sample and Fig. 13 shows permeability profiles in the sample at different times.

- i. The kinetic does not seem to be influenced by the variations of the permeability (cf. Fig. 8 and Fig. 14) whereas liquid velocity should increase with the permeability. That means that the solute transport by the advection is negligible compared to the solute transport by the diffusion.
- ii. The pore water pressure strongly decreases when the variation of the permeability is taken into account (cf. Fig. 10 and Fig. 15). The value of the pressure is divided about 16. The increases of the permeability allow important liquid fluxes as said previously. That implies a quick release of the pore water pressure in the sample (cf. Fig. 15).

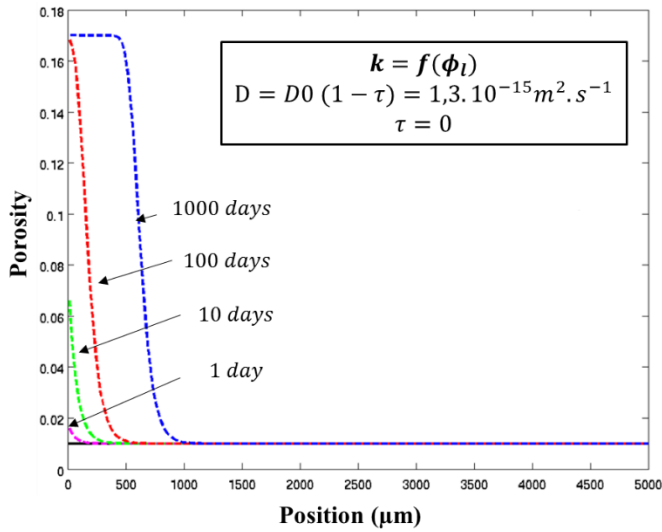


Fig. 12 Porosity profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration

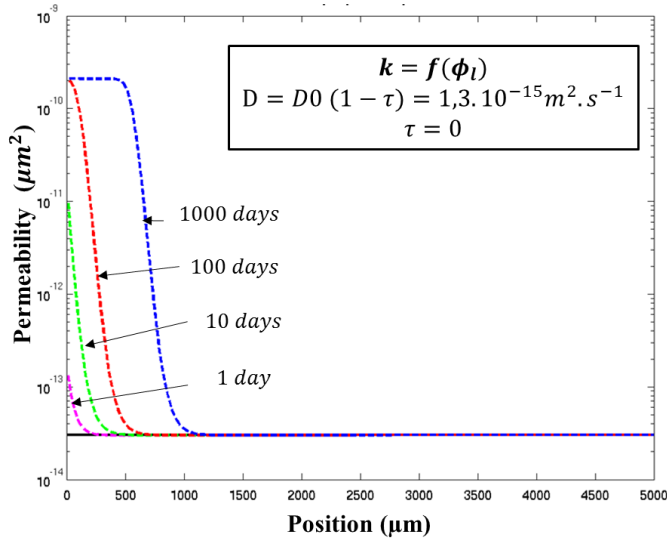


Fig. 13 Permeability profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration.

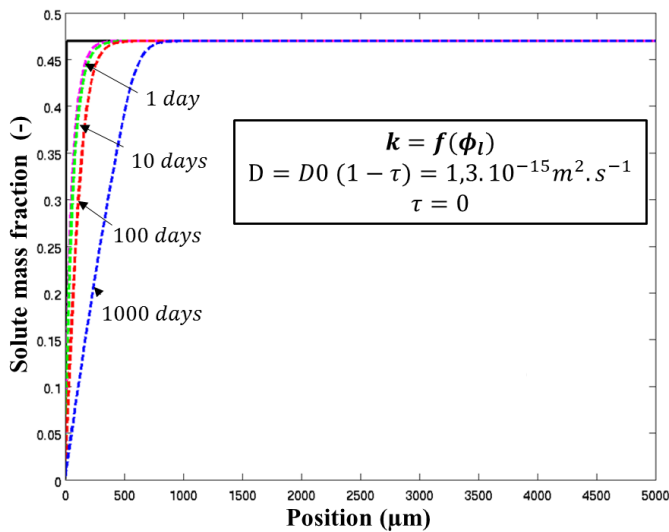


Fig. 14 Solute mass fraction profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with the permeability

function of the porosity  $k = f(\phi_l)$ , the diffusive coefficient  $D = 1,3 \cdot 10^{-15} m^2 \cdot s^{-1}$  and the efficiency coefficient  $\tau = 0$ .

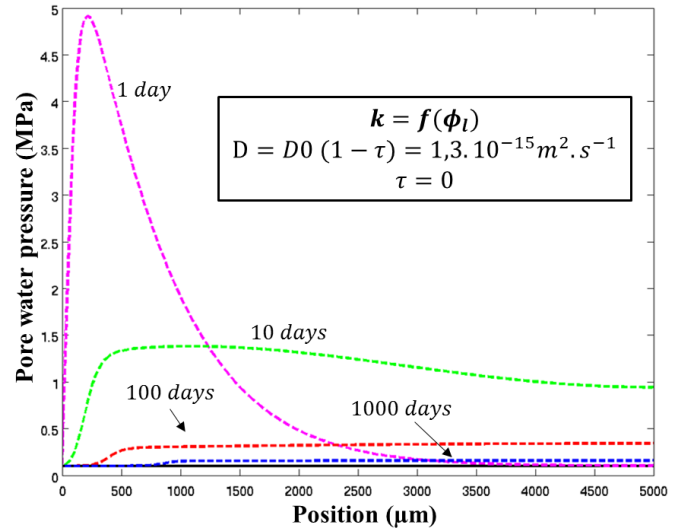


Fig. 15 Pore water pressure profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with the permeability function of the porosity  $k = f(\phi_l)$ , the diffusive coefficient  $D = 1,3 \cdot 10^{-15} m^2 \cdot s^{-1}$  and the efficiency coefficient  $\tau = 0$ .

### 5.3.2 Diffusion coefficient variation impact

Impacts of the variation of the diffusion coefficient (cf. Fig. 16) are analysed without permeability nor osmosis variation.

- i. As diffusion coefficient increases with the porosity during the leaching, the kinetic increases (cf. Fig. 4 and Fig. 17). This is in accordance with observation made in 5.1.
- ii. In the same way, and as observed in 5.1, the pore water pressure strongly increases with diffusion (cf. Fig. 6 and Fig. 18).

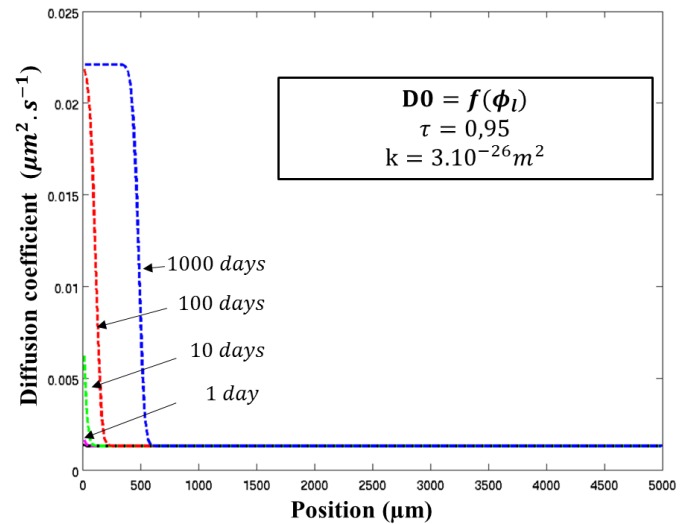


Fig. 16 Diffusion coefficient profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration.



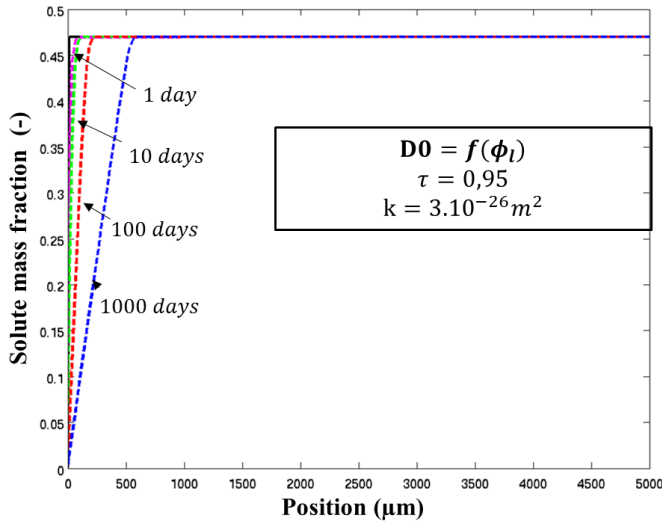


Fig. 17 Solute mass fraction profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with the diffusive coefficient  $D = f(\phi_I)$ , the efficiency coefficient  $\tau = 0,95$  and the permeability  $k = 3.10^{-26} m^2$ .

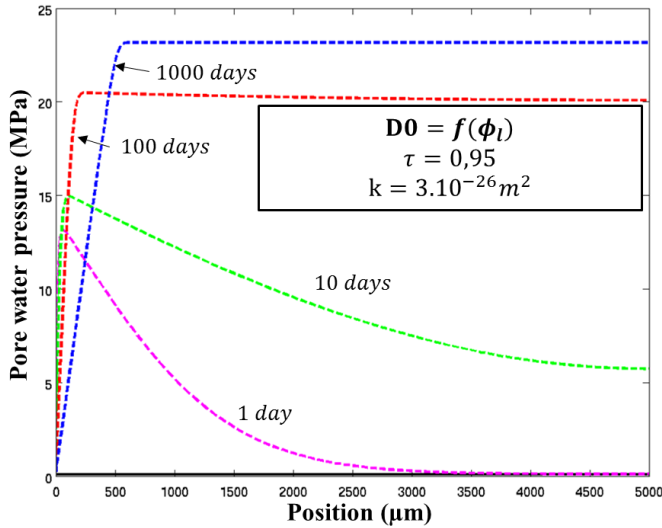


Fig. 18 Pore water pressure profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with the diffusive coefficient  $D = f(\phi_I)$ , the efficiency coefficient  $\tau = 0,95$  and the permeability  $k = 3.10^{-26} m^2$ .

### 5.3.3 Efficiency coefficient variation impact

Impacts of the variation of the efficiency coefficient are analysed. During the BW leaching the bitumen matrix become more and more porous. That is why the efficiency coefficient  $\tau$  decreases (cf. Fig. 19). Consequences are:

- i. The kinetic increases (cf. Fig. 4 and Fig. 20) with the efficiency coefficient drop. This is due to the logical increasing of the effective diffusive coefficient:  $D = D0(1 - \tau)$ . As seen in 5.1, the rise of diffusive mechanism is linked to the kinetic increasing.
- ii. The pore water pressure moderately increases with the efficiency coefficient drop (cf. Fig. 6 and Fig. 21). This could be

explained by the opposite effects of the diffusion rise (as seen in 5.1) and the efficiency coefficient decrease (as seen in 5.2). The combined mechanisms finally induce a small pore water pressure rise. In this case, the diffusion has a more significant impact than the osmosis.

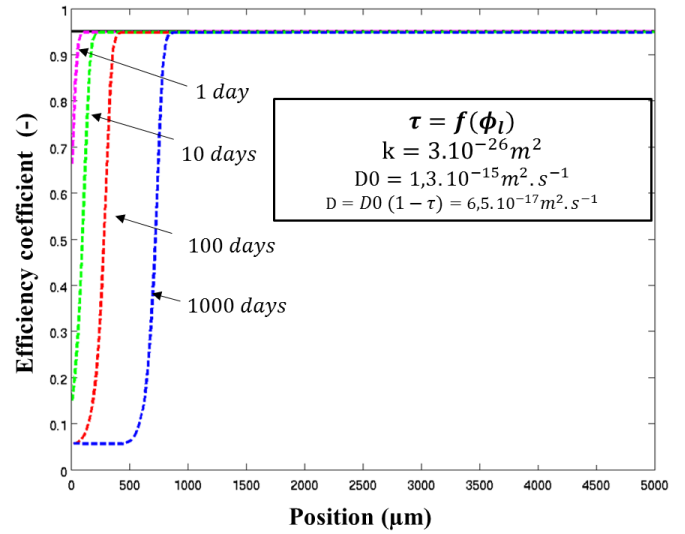


Fig. 19 Efficiency coefficient profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration.

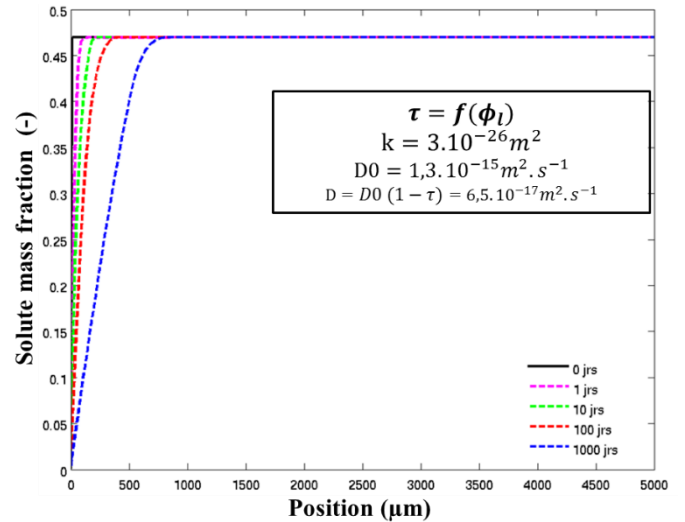


Fig. 20 Solute mass fraction profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with the efficiency coefficient  $\tau = f(\phi_I)$ , the permeability  $k = 3.10^{-26} m^2$  and the diffusive coefficient  $D = 1,3.10^{-15} m^2.s^{-1}$ .

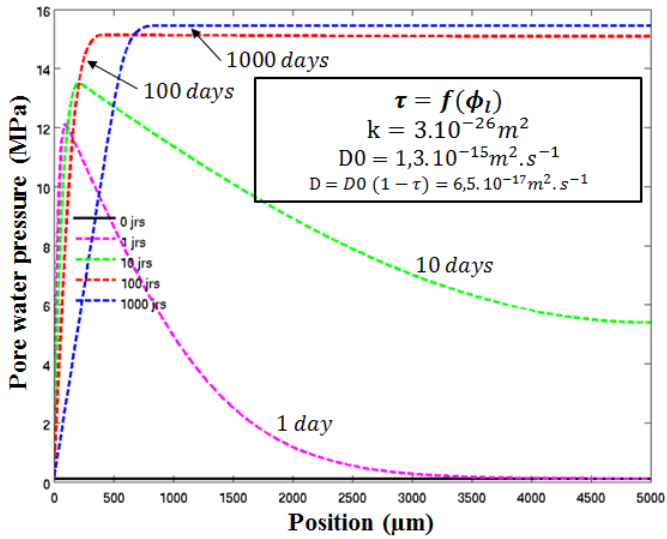


Fig. 21 Pore water pressure profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration with the efficiency coefficient  $\tau = f(\phi_l)$ , the permeability  $k = 3.10^{-26} m^2$  and the diffusive coefficient  $D = 1,3.10^{-15} m^2.s^{-1}$ .

### 5.3.4 Global simulation

When the permeability, the diffusion coefficient and the efficiency coefficient vary with the porosity, the results of leaching Bituminized waste are shown on Fig. 22 to Fig. 24. Fig. 22 shows a faster transport kinetic than the other cases. That is well explained by the previous studies (cf. 5.3.1, 5.3.2 and 5.3.3). Each parameter's dependence of porosity induces a kinetic increase.

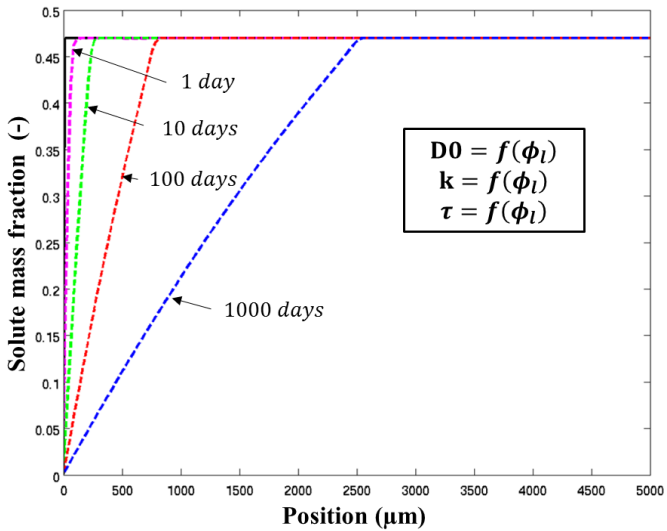


Fig. 22 Solute mass fraction profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration

The Fig. 23 shows an important pressure release rapidly after the hydration beginning. The total stress behaviour, presented in the Fig. 24, is directly related to the pore water pressure evolution. With our convention, compression is negative. First, it can be noticed that the compression increases at the beginning of the hydration. That is mainly due to the osmosis and

the dissolution. Second, after approximately ten days, a decrease of the compression is observed. That is due to osmosis decreasing and enhancing of Darcy fluxes.

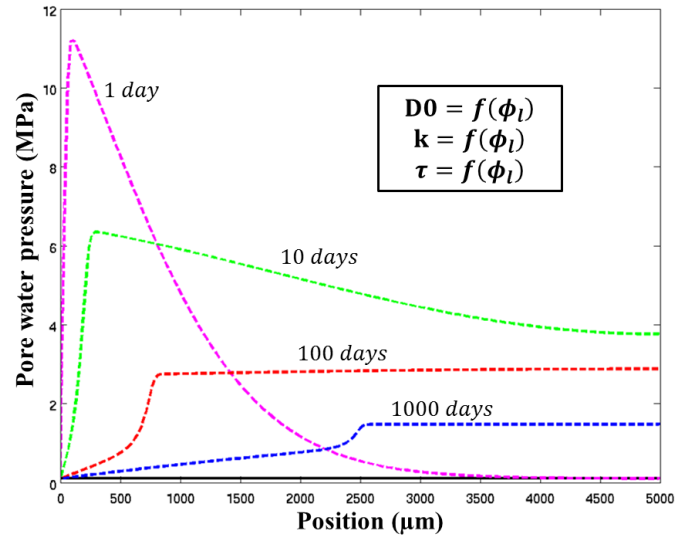


Fig. 23 Pore water pressure profiles in an Eurobitume sample after 1, 10, 100 and 1000 days of hydration

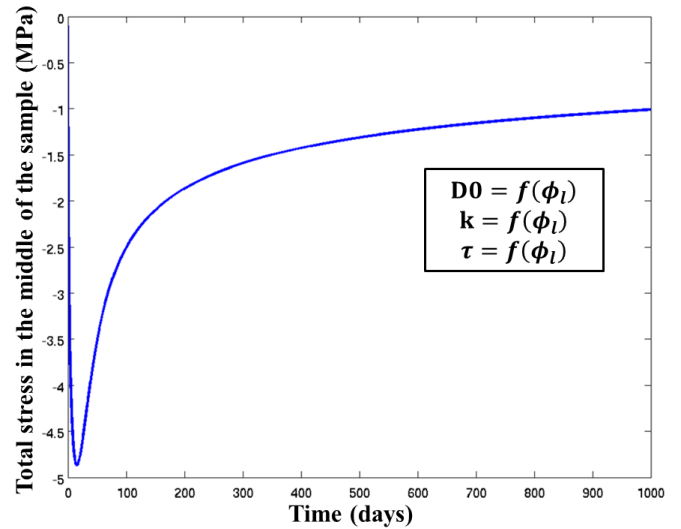


Fig. 24 Evolution of the total stress in the middle of the sample during the hydration

## 6 Conclusions and further works

In this paper, the behaviour of Bituminized Waste Products (BWP), containing essentially some  $NaNO_3$  salts, in contact with free water and in confined conditions (constant volume) has been studied. In this condition, leaching could generate overpressure and mechanical consequences.

A numerical poromechanical model has been proposed including Darcy's Flow, diffusion and osmotic mechanisms. The dissolution/precipitation of one salt is taken into account and perfect elasticity is assumed. The formulation has been implemented in one dimension finite volume with MATLAB.

The contribution of permeability, diffusion and osmosis have been investigated including their evolution with porosity.

Numerical results have shown that the transport kinetic is mainly ruled by the diffusion. From a mechanical point of view, the osmosis is the main factor of pore water pressure (and stress) increasing. The diffusion plays also a significant role. The simulation analyses have pointed out strong couplings between mechanisms, notably because of the impact of porosity on material parameters. At the end, with this model, it seems impossible to separate those mechanisms and their coupling unless to miss reality.

Consequently, more experimental calibrations are needed to verify material parameters and evolution laws. Further investigations are needed on the mechanical behaviour, especially on creep deformation, in order to simulate water up-taking tests in free swelling conditions.

In future works, we will focus on French BWP and will analyse the comportment of several weakly soluble salts and consequences on porous media.

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