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CHEMO-PORO-MECHANICAL MODELING OF CEMENTITIOUS MATERIALS (DIFFUSION-PRECIPIATION-CRACKING)

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Abstract

The present work focuses on the impact of the chemical degradations of cementitious materials such as Delayed Ettringite Formation (DEF) on the overall material properties. DEF is an endogenous pathology due to the crystallization of ettringite within voids and cracks. The crystallization pressure in the porous cement paste induces swelling and cracking by differential expansion. The study aims to characterize the evolution of effective material properties (diffusion coefficient, apparent tenacity) with respect to DEF. A non-linear chemo-mechanical modeling is proposed where the entire diffusion-precipitation-pressurization-crack process is solved in a staggered approach. The diffusion-precipitation mechanism is translated by a rough chemical model. The resulting local volume fraction of ettringite is estimated by a finer micro-mechanical-based model using the effective elastic properties of the cement paste. The crack initiation and propagation is estimated with a dedicated cohesive zone model including pressure effect. Some applications of the model are presented.

Keywords: Delayed Ettringite Formation, Poromechanics Reactive Transport, Cracking, Cohesive Zone Model, Analytical Homogenization

1. INTRODUCTION

1.1 Context

In context of the lifetime extension of nuclear power plants, the French “Institut de Radioprotection et de Sûreté Nucléaire” (IRSN) leads research on the ageing of cementitious materials. Previous works dealt with chemo-thermo-mechanical at early age and hydro-mechanical modifications on the long term; these works induce delayed strain and degradation of concrete [1]. Since few years, IRSN has been interested in swelling reactions as alkali silica reaction and Delayed Ettringite Formation (DEF) [2]. The present work focuses on the impact of the DEF on the overall material properties at the aggregate scale (mesoscale), mainly on the diffusion coefficient and on the apparent tenacity of the concrete.

1.2 Delayed Ettringite Formation physics

DEF is an endogenous pathology due to early age chemical reactions. Some elevated thermal conditions induce dissolution of primal ettringite and later on due to moisture environment ettringite can precipitate again within voids and cracks. The ettringite crystallization pressure in the porous cement paste induces both swelling and cracking by differential expansion. These cracks are preferred location for ion diffusion and further ettringite precipitations [3]. It is commonly admitted that DEF and associated swelling depend on:

- Temperature:
 - At early age, depending on the kinetic and the maximum value of heat treatment, swelling varies [4]. It seems that these variations are induced by the quantity of aluminum available [5]
 - During the lifetime of the material, the temperature seems also to play an indirect role (via various chemo-diffusion processes) both on the swelling rate and on the maximum swelling [6]
- Humidity environment: DEF only develops in an environment with a high relative humidity [2]
- Cement composition: water-cement ratio [7], the cement paste fineness and the alkali proportions modify the chemical processes leading to DEF [8]
- Microstructure of aggregates: nature [3] [9], size effect and volume fraction [2] also play a role in the chemo-mechanical coupling that leads to ettringite-induced swelling.

The experimental swelling depends on chemical reactions (moisture environment leads to alkalis leaching that induces sulfate desorption) and on the material properties. To model this phenomenon, it is necessary to consider coupled chemical reaction, transport of ions, and mechanical response.

1.3 Delayed Ettringite Formation modeling

At least, two types of modeling exist to represent the impact of swelling in a cemented structure:

- Hydro-mechanical coupling: these models couple drying and swelling. The swelling depends on saturation threshold and maximum strain [4] [10]. The work of [10] depicts the difficulty to obtain these values due to a strong dependence on material properties.
- Chemo-mechanical coupling: some models are based on thermodynamic chemistry to evaluate the quantity of ettringite which precipitates and its mechanical impact [5] [6]. Recently a semi empirical chemo-mechanical modeling [11] was proposed to exhibit intelligible results for structure applications [12]

As previously reported by several authors [5] and [10], a strongly coupled modeling should improve predictions of swelling kinetics. To our knowledge, there exists no modeling with a strong coupling between transport properties and mechanical balance applied to DEF. The aim of the study is to characterize the degradation of material properties by crystallization pressure, with a coupled diffusion model within cracks and to estimate the overall mechanical response.

2. CHEMO-PORO-MECHANICAL MODEL

The strongly coupled phenomena suggest a chemo-mechanical modeling. In our model, the entire diffusion-precipitation-pressurization-crack process is solved in a staggered approach.

Before performing a simulation, the cement composition is estimated with a chemical model [13] and the mortar poro-mechanical and diffusion properties are evaluated by analytical homogenization. The chemical model does not take into account alkali and thermal impact. The goal is to estimate the influence of clinker composition and water-cement ratio on effective properties. The steps of the simulation are (Figure 1):

- First, compute the transport of the chemical species. This model follows the framework of [15] and the crack have a resistive impact on crack normal flow, decreases when the crack density increases,
- Second, knowing the ions in solution, the amount of volume of the precipitate is evaluated by a chemical system,
- Third, a local overpressure is calculated to feed the poro-mechanical model and the pressure in the cracks. The crack initiation and propagation are based on a dedicated cohesive zone model [14].

The associated numerical framework is Xper [14]. This software allows to simulate the fracture dynamics of heterogeneous materials, from crack initiation to non-smooth frictional post-fracture.

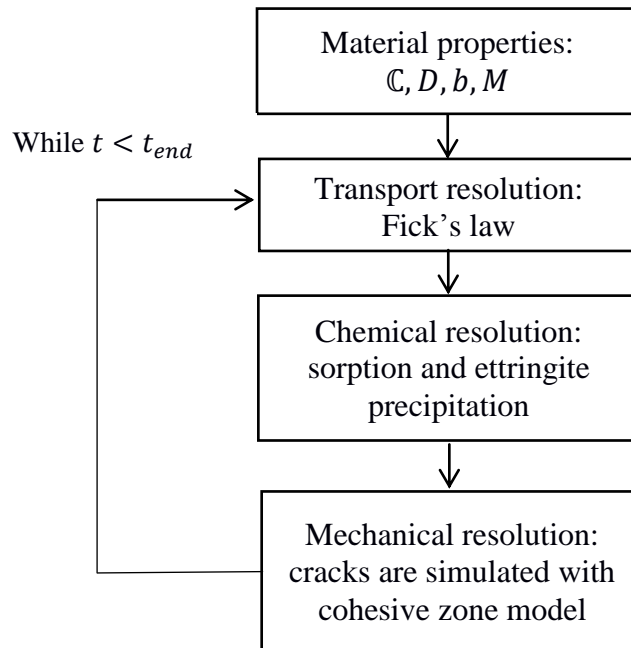


Figure 1: Diagram of the model

2.1 Analytical homogenization

The retained homogenization schemes follow recommendations of the literature [16] [17]:

- Cement paste: Mori-Tanaka for poro-mechanical properties, Maxwell for effective diffusion coefficient
- Mortar: generalized self-consistent scheme for poro-mechanical and effective diffusion properties

2.2 Transport resolution

The concrete is assumed to be immersed. Fick's law is used to solve the transport phenomena:

$$\frac{\delta \phi C_i}{\delta t} = \nabla \cdot (D \nabla C_i) + r_i + \varphi_t \quad (1)$$

where ϕ is the porosity, C_i is the ion concentration, D is the effective diffusion coefficient, r_i is the chemical source term and φ_t is the transversal flux through the crack [15]. The transversal flux depends on diffusion coefficients of the crack D_{crack} , and on the concentration difference between the crack lip:

$$\varphi_t = D_{crack} \frac{\Delta C}{lc} \quad (2)$$

where lc is a characteristic length for the gradient estimate. The transversal flux has a resistive impact on the concentration field. It is assumed that the diffusivity increases with the crack opening $[u_N]$, until the critical value $[u_{cri}]$ is reached [18]. Once this threshold is reached, the diffusivity is the same as in water, $D_0 \approx 10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$:

$$\text{if } u_N < u_{cri} = 100 \text{ } \mu\text{m} \text{ then } D_{crack} = D_0 \frac{[u_N]}{[u_{cri}]} \quad (3)$$

else $D_{crack} = D_0$

2.3 Chemical resolution

The chemical model takes into account sulfate desorption by alkali leaching and ettringite precipitation. We use the desorption model from [5]:

$$C_{(SO_4^{2-})ad} = k C_{SO_4^{2-}} (C_{Na^+})^{0.2} \quad (4)$$

where k depends on temperature and on C-S-H concentration in the solid. The model is restricted to sulfate and aluminate ions.

2.4 Mechanical resolution

The dynamic fracture of heterogeneous materials is studied through a micromechanical multibody modelling. The modelling consists in introducing Frictional Cohesive Zone Model [14] between each adjacent elements of a finite element mesh:

- The bulk behaviour is described with a visco-poro-mechanical model without any damage,
- A frictional cohesive zone model is inserted at element interfaces to take into account cracking and post pressure impact.

2.4.1 Bulk behaviour: visco-poro-mechanical model

The visco-poro-mechanical model describes the impact of the precipitation on the porous viscoelastic matrix. The viscoelasticity is described through a Zener rheological model (Figure 2). This viscoelastic constitutive law is equivalent to a Pointing Thompson model usually used to describe primary creep [1] [12]:

$$\underline{\underline{\sigma}} = \mathbb{C} : \underline{\underline{\varepsilon}} + \mathbb{V}_M : (\mathbb{C} + \mathbb{C}_M) \underline{\underline{\dot{\varepsilon}}} - b P \underline{\underline{I}} \quad (5)$$

where \mathbb{C} , \mathbb{C}_M , and \mathbb{V}_M are respectively elasticity fourth order tensor, Maxwell elasticity tensor and Maxwell viscous tensor. The internal stress is defined by the hydrostatic pressure P and the Biot coefficient b . The pressure law considers only positive value [5] [11] [12]:

$$P = M \langle V_{ett} - \langle V_0 + b \operatorname{tr}(\underline{\underline{\varepsilon}}) \rangle_+ \rangle_+ \quad (6)$$

where $\langle X \rangle_+ = 0$, if $X \leq 0$ and $\langle X \rangle_+ = X$, if $X > 0$, M is the bulk Biot modulus of ettringite, V_{ett} is the volume of ettringite, and V_0 is a connected pore volume threshold.

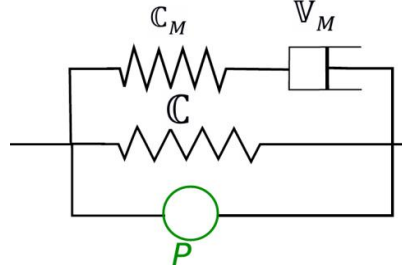


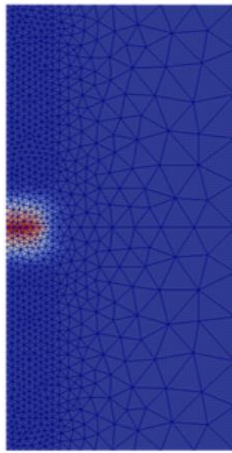
Figure 2: Rheological scheme of the model (Zener model with poro-mechanical)

2.4.2 Interface model: frictional cohesive model with post cracking pressure

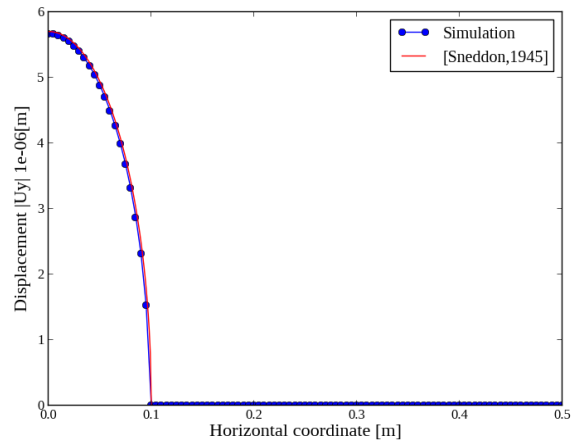
The cohesive model is based on [14] and [19], and considers post cracking pressure. The model takes into account normal and tangential components of the displacement jump through the crack lips $[\underline{u}]$ and gradually introduces a post-rupture pressure $P(t)$, see equation (5). For a crack of normal n , the cohesive stress vector $\underline{\sigma}_{adh}$ depends on a surface damage described by β ($\beta = 1$: healthy interface, $\beta = 0$: broken interface) and on a damageable surface elastic tensor $\underline{\underline{K}}(\beta)$:

$$\underline{\sigma}_{adh} = \underline{\underline{K}}(\beta)[\underline{u}] - (1 - \beta)^\alpha P(t)n \quad (5)$$

where $\alpha > 0$. The implementation of this new cohesive law with progressive transition resistance-damage-swelling is validated on a cubic elemental volume ellipsoidal pressure [20] (Figure 3). The pressure is only taken into account in the mechanical part of the model and evolves with respect to time.



a)



b)

Figure 3: Study of crack opening pressured in an impermeable matrix a) displacement field, only one quarter of cube simulate and b) comparison with analytical solutions [20]

3. APPLICATION

The stress-free swelling of a concrete sample (11 x 22 cm) is studied. The sample has an aggregate volume fraction of 0.3, with radius of 6 mm. The mortar is composed of 50 % of sand and the cement is a CEM I with water-cement-ratio of 0.3. Diffusion boundaries conditions take place only for alkali ions. The sample is immersed in a solution without alkali: an homogeneous Dirichlet boundary condition is applied on all boundaries. Initial concentrations of adsorbed sulfate by C-S-H and free aluminates are assumed. An axial displacement is prescribed along the longitudinal direction. The system is solved in 2D under plane strain assumption. The poro-mechanical and diffusion parameters are estimated by an analytical homogenization technic with a cement composition described in [17]. Viscoelastic properties are evaluated by means of value in the literature (ratio of Young modulus and Kelvin's modulus, and characteristic time). The cohesive parameters are described in [15]. The sample is initially without crack.

The first results show diffuse cracking due to DEF mostly at the interface between mortar and granulates, which seems to agree with experimental observations [4] (Figure 4 a). Ettringite precipitates in the entire sample (Figure 4 b). The localization of precipitation depends strongly on the microstructure: diffusion coefficient aggregates and cracks. The longitudinal displacement field depends on crack localization and boundary conditions (Figure 4 c). The swelling is thus not homogenous in the sample.

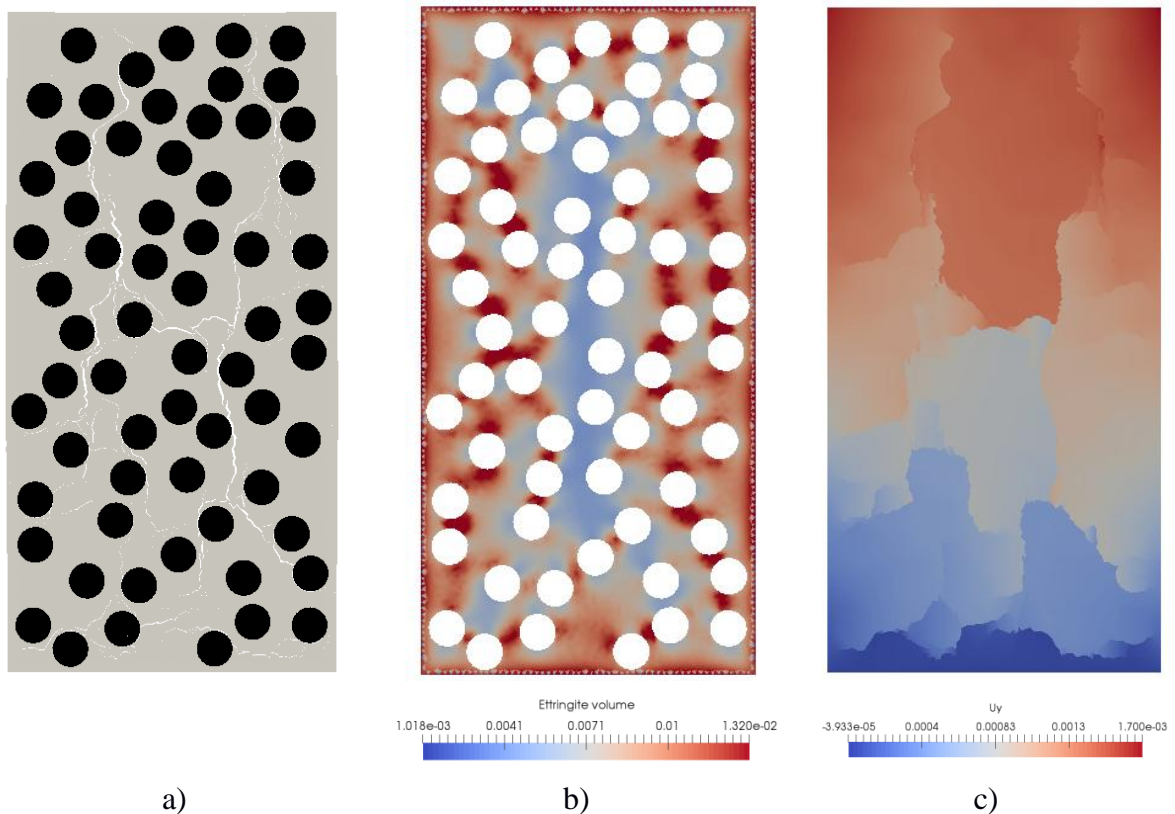


Figure 4: a) cracking profile (crack in white, mortar in gray and granulates in black), b) ettringite volume field [m^3] (granulates in white) and c) longitudinal displacement [m]

The numerical macroscopic expansion (Figure 5 a) is less than in the reported average experimental results of [4]. Initial aluminates concentration and adsorbed sulfate should not be present in enough quantities. Since, in this simulation the cohesive zone model was not coupled to the chemical reaction, the post cracking pressure law is not take into account. At last, the Figure 5 b shows the evolution of the crack density with the macroscopic volumetric strain. It is observed that the density increases abruptly for a value of 0.4%. These cracks could have a real impact on the diffusion properties which can not be neglected. This could be explained by the fact that we do not have a real sigmoid curve for the expansion curve.

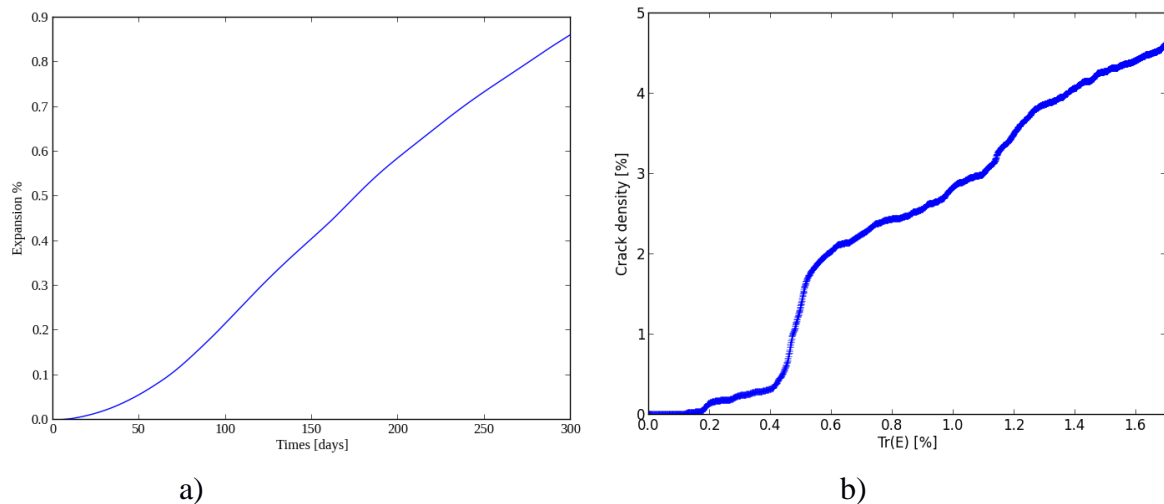


Figure 5: First coupling test, a) expansion measured on the left side of the numerical sample and b) evolution of crack density with volumetric strain

To model the impact of cracks on the macroscopic swelling kinetic, the reactive transport through the crack needs to be modeled as well as the link between chemistry and post cracking pressure law.

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