

Modeling of Lixiviation-Mechanical Coupling Behavior of Fiber Reinforced Concrete

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Abstract: A lixiviation-mechanical coupled model is proposed for fiber reinforced concrete; both the influence of chemical degradation on short/long term mechanical behavior and the effect of mechanical loading on the diffusion coefficient can be considered. The elastic mechanical properties are written as function of chemical damage. A Drucker–Prager typed criterion with associated flow rule is used with taking into account of mechanical damage and chemical damage. An empirical equation including the chemical damage is adopted to describe the creep strain. The diffusion coefficient is defined as a positive function of mechanical damage. The proposed model is implemented into COMSOL and validated by experimental data. The proposed model can well describe the lixiviation-mechanical coupling behavior of fiber reinforced concrete.

Keywords: lixiviation-mechanical coupling, chemical damage, fiber reinforced concrete

1. Introduction

Fiber reinforced concrete is widely used in many engineering constructions, such as nuclear waste containment structures, dams, tunnels and so on. The durability of these materials has to be assessed over long periods of time. Stability analysis of these structures requires the understanding and modeling of mechanical behaviors under complex multi-axial loading conditions. Extensive experimental investigations (Feldman and Beaudoin, 1974; Terrien, 1980; Yurtdas et al., 2006) and constitutive models (Mazars, 1986; Pietruszczak et al., 1988; Sellier 2006) have been performed on the mechanical behavior of cement based material.

In addition, Fiber reinforced concrete is subjected not only to mechanical loading but also to chemical degradation when they are in contact with aggressive environment such as rain, water, acid fluids and so on. More recently, in the framework of sustainable

development, it is of great concerns of storage and sequestration of nuclear waste, toxic residual fluids and gas. It is clear that the durability analysis of structures requires the identification and modeling of long term behavior undergoing chemical attacks. A lot of research efforts have been contributed to experimental investigations (Buil et al.1992; Adenot, 1992; Gérard et al, 1996; Carde et al, 1996;) and numerical modeling (Tognazzi, 1998; Ulm et al, 1999; Le Bellégo et al, 2003; Mainguy et al. 2000 etc.) of chemical degradation in cement based material, just to mention a few.

The objective of this study is to propose a lixiviation-mechanical coupling model in COMSOL for fiber reinforced concrete. For this purpose, this paper firstly presents the instantaneous mechanical model, creep model and lixiviation model, then the proposed models are combined into a coupling model and applied to simulate the experimental results of fiber reinforced concrete. In section 2, the hydration model, instantaneous mechanical model, creep model and lixiviation model are proposed for fiber reinforced concrete. In section 3, the proposed models are combined into a coupled model to simulate the creep test under lixiviation.

2. Model characterization

In this section, we will present a model to describe the lixiviation-mechanical coupling behaviour of fibre reinforced concrete, which is chose as the storage structures in French national agency for nuclear waste management (Andra).

2.1 Instantaneous mechanical model

In elastoplastic model, the plastic yield function is defined in a form of Drucker-Prager typed plastic criterion.

$$f_p(\sigma_{ij}) = q + \eta p - K = 0 \quad (1)$$

$$\text{with: } p = \frac{1}{3} \text{tr} \sigma_{ij}, \quad q = \sqrt{3J_2}, \quad J_2 = \frac{1}{2} S_{ij} : S_{ij},$$

$S_{ij} = \sigma_{ij} - \frac{1}{3} \sigma_{ij} \delta_{ij}$, p is the first stress invariant and q is the second stress invariant. The two material parameters are defined by:

$$\eta = \frac{1}{\sqrt{3}} \left(\frac{2Rc}{Rt + Rc} - 1 \right) \quad (2)$$

$$K = \frac{2}{\sqrt{3}} \left(\frac{Rt \cdot Rc}{Rt + Rc} \right) \quad (3)$$

where Rc and Rt compression strength and tension strength.

2.2 Creep model

For the fibre reinforced concrete which is properly cured, the creep tests (Camps 2008) at constant temperature show the creep strain rate is related with stress, time and chemical damage variable:

$$\dot{\epsilon}^{cpc} = A_c \left(A_1 + A_2 m t^{m-1} \right) f_3(d_c) \sigma \quad (4)$$

$$\dot{\epsilon}^{cpt} = A_t \left(A_1 + A_2 m t^{m-1} \right) f_3(d_c) \sigma \quad (5)$$

where $\dot{\epsilon}^{cpc}$ and $\dot{\epsilon}^{cpt}$ denote the creep strain rate under compressive and tensile stress, respectively. The parameters A_c and A_t are introduced to describe the difference between compression and tension. The model parameters A_1 , A_2 , A_c and A_t can be determined by fitting method based on the experimental data.

2.3 Lixiviation model

At a natural (reference) state, the calcium concentration in pore fluid Ca^{2+} is in equilibrium with that in solid skeleton Ca^{solid} . When the cement paste is in contact with aggressive fluid on its exterior boundary, the chemical equilibrium is broken. The calcium is dissolved from solid skeleton and is diffused inside pore fluid. The chemical dissolution and diffusion of calcium ions must verify the general mass balance equation. The mass balance equation reads:

$$\frac{\partial(\phi Ca^{2+})}{\partial t} = -\nabla \cdot \left[-D(Ca^{2+}) \nabla Ca^{2+} \right] \quad (6)$$

$$+ M_{Ca^{solid} \rightarrow Ca^{2+}}^0$$

Ca^{2+} is the calcium concentration of the pore solution, ϕ is the porosity. $D(Ca^{2+})$ is the effective diffusion coefficient which can depend on calcium concentration in pore fluid, temperature and mechanical deformation of

material (Gérard, 2000; Mainguy, 2000). The last term $M_{Ca^{solid} \rightarrow Ca^{2+}}^0$ describes the quantity of calcium dissolved in the liquid phase per unit of time which can be rewritten as:

$$M_{Ca^{solid} \rightarrow Ca^{2+}}^0 = -\frac{\partial Ca^{solid}}{\partial t} \quad (7)$$

$$= -\frac{\partial Ca^{solid}}{\partial Ca^{2+}} \cdot \frac{\partial Ca^{2+}}{\partial t}$$

Introducing Eq. (7) into Eq.(6), one gets the generalized diffusion equation of calcium concentration in pore fluid:

$$\left[\phi(Ca^{2+}) + Ca^{2+} \frac{\partial \phi(Ca^{2+})}{\partial Ca^{2+}} + \frac{\partial Ca^{solid}}{\partial Ca^{2+}} \right] \frac{\partial Ca^{2+}}{\partial t} \quad (8)$$

$$= \nabla \cdot \left[D(Ca^{2+}) \nabla Ca^{2+} \right]$$

In Eq. (8), the first two terms correspond to effect of porosity change on calcium diffusion process and the third one is related to phase change of calcium ions. Generally, the effect related to the phase change $\frac{\partial Ca^{solid}}{\partial Ca^{2+}}$ is

dominant compared to the other ones (Delagrave, 1997). Therefore, it is possible to simply the generalized diffusion equation by neglecting the first two terms in Eq. (8), Eq. (8) can be simplified as following:

$$C(Ca^{2+}) \frac{\partial Ca^{2+}}{\partial t} = D(Ca^{2+}) \nabla^2 Ca^{2+} \quad (9)$$

where, $C(Ca^{2+}) = \frac{\partial Ca^{solid}}{\partial Ca^{2+}}$ is the source

term calcium, ∇^2 is the Laplace operator. This equation is a partial differential equation that describes the calcium ions diffusion process in pore fluid. Note that this diffusion equation is strongly non-linear in nature due to the dependency of diffusion coefficient on various factors and also to nonlinear phase change equation.

In order to solve Eq.(9), the two terms $C(Ca^{2+}) = \frac{\partial Ca^{solid}}{\partial Ca^{2+}}$ and $D(Ca^{2+})$ still need to be determined. The first term represents the derivative of calcium ions in solid skeleton with respect to calcium concentration in pore fluid. Therefore, it should be determined from the chemical equilibrium curve between Ca^{solid} and Ca^{2+} . This equilibrium curve should be determined from laboratory data proposed by Buil (1992), also used by Gerard et al. (1998) according to the previous works

of Adenot (1992). In this work, we have adopted the data obtained by Adenot (1992) on cement paste.

According to Eq. (9), the diffusion coefficient is the main factor controlling the kinetics of diffusion of calcium concentration in pore fluid. A series of studies (Bentz and Garvoczi, 1992; Gérard, 1996) have shown that the diffusion coefficient depends on the volume fractions of various hydrate species, temperature and possibly mechanical deformation. Under isothermal conditions and taking into account the effect of mechanical deformation on diffusion process, we have adopted the following relations:

$$D = D(Ca^{2+}, d_p) = D_e(Ca^{2+})f(d_p) \quad (10)$$

where $D_e(Ca^{2+})$ denotes the diffusion coefficient of fibre reinforced concrete without mechanical damage. The term $f(d_p)$ is introduced to describe the effect of mechanical deformation. $D_e(Ca^{2+})$ can be determined by the existing laws (Tongnazzi 1998, Peycelon 2006, Camps 2008, Sellier 2010) according to the paste porosity.

Further, as the chemical dissolution kinetics is controlled by diffusion process, it is obvious that the diffusion coefficient should depend on the chemical nature of fluid. Consider the pure water as a reference fluid and the corresponding diffusion coefficient as the reference value. The actual diffusion coefficient associated with another aggressive fluid can be obtained by multiplying the reference value by an appropriate amplification factor. Let λ be such a multiplier, the actual diffusion coefficient corresponding to a specific aggressive fluid is given by $\lambda D_e(Ca^{2+})$. Therefore, Eq.(9) can be written

as following under aggressive fluid:

$$\frac{\partial Ca^{solid}}{\partial Ca^{2+}} \frac{\partial Ca^{2+}}{\partial t} = \lambda D_e(Ca^{2+}) \nabla^2 Ca^{2+} \quad (11)$$

The determination of the multiplier λ is based on relevant experimental data in different dissolution conditions; for instance, comparing the diffusion kinetics between pure water and nitrate ammonium.

From the mechanical point of view, the increase of porosity due to chemical leaching can be considered as a damage process, which is described by a scalar valued chemical damage variable d_c :

$$d_c = \phi - \phi_0 \quad (12)$$

where ϕ_0 is the reference value of connected porosity of sound material. Taking into account calcium mass balance equation and chemical equilibrium relation, the chemical damage can be expressed in the following form:

$$d_c = d_{c\max} \left[1 - e^{-b_{dc}(Ca^{2+} - Ca^{2+}|_0)} \right] \quad (13)$$

where $d_{c\max}$ is the asymptotic values of chemical damage. b_{dc} is a model parameter which is used to describe the evolution rate of chemical damage. $Ca^{2+}|_0$ and Ca^{2+} denote the initial (in sound condition) amount and actual concentration of calcium in pore fluid, respectively.

3. Simulation results

In order to simulate the creep compression and 4 point creep bedding tests subjected to ammonium-nitrate leaching, the lixiviation model, elastoplastic model and creep model are combined into a lixiviation-mechanical coupling model.

On one hand, it needs to consider the influence of mechanical deformation on the diffusion coefficient. According the study of Gerard (1998), the mechanical deformation (microcracks) will affect the diffusion of the ions in the material. In our study, it is assumed that only the plastic deformation affect the diffusion coefficient. Therefore, the variation of the diffusion coefficient caused by plastic deformation is defined by the following function:

$$D(d_p) = \lambda D_e (1 + \alpha_D d_p) \quad (14)$$

$$d_p = \begin{cases} \bar{d}_{pc} \left[1 - \exp(-b_{pc}\gamma^p) \right], & \text{compression} \\ \bar{d}_{pt} \left[1 - \exp(-b_{pt}\gamma^p) \right], & \text{tension} \end{cases} \quad (15)$$

where λD_e is the value of diffusion coefficient in the absence of mechanical damage. α_D is a model parameter which is introduced to describe the effect of mechanical damage. Eq. (14) proceeds from the assumption that the diffusion coefficient should be affected by mechanical damage, within the bounds of the coefficients for undamaged concrete and of totally cracked concrete where the diffusion in

free water is expected to be recovered on macrocrack faces. Hence, $D_e(1 + \alpha_D d_p)$ cannot exceed the diffusion coefficient in free water which is about $10^{-9} \text{ m}^2/\text{s}$. Therefore, the values of α_D must verify $0 \leq \alpha_D \leq 100$.

On the other hand, it needs to consider the influence of leaching on the mechanical parameters. The influence of leaching on the parameters in elastoplastic model has been defined in the previous part. In addition, the chemical degradation will change the creep behaviour of concrete which subjected to the coupling effect of stress and chemical degradation in the long term. Therefore, the term $f_3(d_c)$ in Eq. (4) and (5) is introduced to describe the effect of chemical degradation on creep strain rate, Eq. (4) and (5) can be expressed as following:

$$\dot{\epsilon}^{cpc} = A_c (A_1 + A_2 m t^{m-1}) (1 + \alpha_{dc} d_c) \sigma \quad (16)$$

$$\dot{\epsilon}^{cpt} = A_t (A_1 + A_2 m t^{m-1}) (1 + \alpha_{dc} d_c) \sigma \quad (17)$$

where α_{dc} is a model parameter which is used to describe the extent of effect of chemical damage on creep strain rate. Its value can be determined by fitting method based on experimental data. The values of model parameter are given in Table 1.

After the determination of these model parameters, the lixiviation-mechanical coupled model is applied to simulate 4 point bedding tests subjected to ammonium-nitrate leaching. Fig. 1 shows the distribution of Ca^{2+} in pore fluid and longitudinal strain of 4point bedding tests of CEM VF after 18 month leaching of ammonium-nitrate. We can find the significant influence of mechanical deformation on the dissolution of calcium in solid skeleton, the concentration near strain localisation is smaller than the one of other zone. The numerical results of flexility evolution of CEM IF and VF are given in Fig. 2. The evolution rate of creep strain of specimen with ammonium-nitrate leaching is greater than the one of sound specimen.

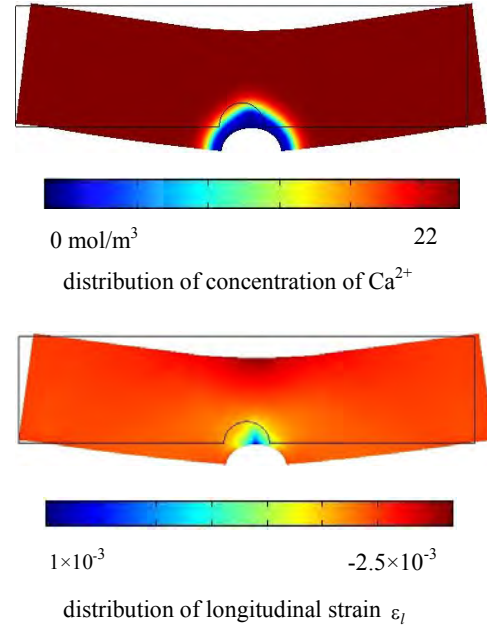


Figure 1 Distribution of Ca^{2+} in pore fluid and longitudinal creep strain ϵ_l^c of 4 point bedding creep tests of CEM VF subjected to ammonium-nitrate leaching

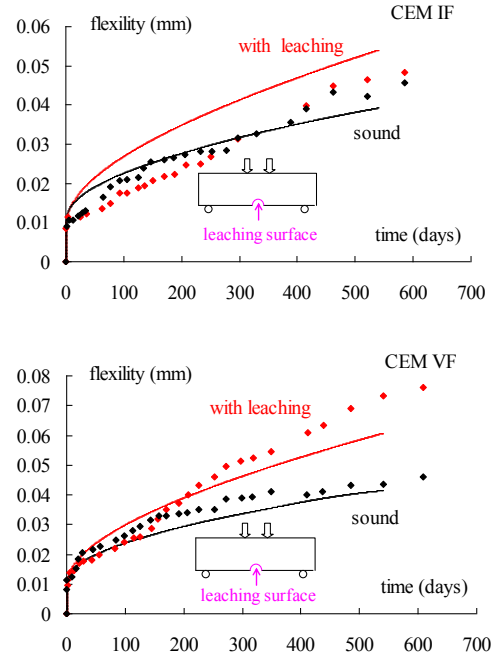


Figure 2 Numerical results of 4 point bedding creep tests subjected to ammonium-nitrate leaching of CEM IF and VF

4. Conclusions

A model was proposed to simulate the lixiviation-mechanical coupling behavior of fiber reinforced concrete. A series of numerical models, including instantaneous mechanical model, creep model and lixiviation model have been set up in COMSOL, and then these models were combined into a lixiviation-mechanical coupling model. The experimental data of 4 point creep bedding tests subjected to ammonium-nitrate leaching were simulated.

5. References

1. Adenot F., 1992. Durabilité du béton: caractérisation et modélisation des processus physiques et chimiques de dégradation du ciment. Thèse de Doctorat, Université d'Orléans, France.
2. Bentz D.P., Garboczi E.J., 1992. Modelling the leaching of calcium hydroxide from cement paste: effects on pore space percolation and diffusivity. *Materials and Structures* **25**, 523-533.
3. Bourgeois F., Burlion N. and Shao J.F. 2002, Modelling of elastoplastic damage in concrete due to desiccation shrinkage, *Int. J. Numer. Anal. Meth. Geomechanics*, **26**, 759-774.
4. Buil M., Revertégat E., Ollivier J.P., 1992. Modelling cement attack by pure water. In ASTM STP-1123, Gilliam T.M. and Wiles C.C.(Eds.), American Society for Testing and Materials, West Conshohocken, USA.
5. Camps, G., 2008. Etude des interactions chemo-mécaniques pour la simulation du cycle de vie d'un élément de stockage en béton. PhD Thesis, Université Paul Sabatier Toulouse.
6. Carde C., François R., Torrenti J.-M., 1996. Leaching of both calcium hydroxide and C-S-H from cement paste: modelling the mechanical behavior. *Cement and Concrete Research* **22**, 1257-1268.
7. Chen D., Yurtdas I., N. Burlion and J.F. Shao. 2007, Elastoplastic damage behavior of a mortar subjected to compression and desiccation, *Journal of engineering mechanics*, 464-472.
8. Delagrave A., Pigeon M., Marchand J. and Revertégat E., 1997. Influence of chloride ions and pH level on the durability of high performance cement paste. *Cement and Concrete Research* **26**, 749-760.
9. Feldman R.F. and Beaudoin J.J., 1974. Microstructure and strength of hydrated cement. Proc of the 6th International Symposium on the chemistry of cement, Moscou.
10. Gérard B., 1996. Contribution des couplages mécanique-chimie-transfert dans la tenue à long terme des ouvrages de stockage de déchet radioactifs. Thèse de Doctorat, E.N.S.de Cachan et Université de Laval.
11. Gérard B., Pijaudier-Cabot G. and Laborderie C., 1998. Coupled diffusion-damage modelling and the implications on failure due to strain localisation. *International Journal of Solids and Structures* **35**, 4107-4120.
12. Gérard B. and Marchand J., 2000. Influence of cracking on the diffusion properties of cement-based materials: Part I: Influence of continuous cracks on the steady-state regime. *Cement and Concrete Research* **30**, 37-43.
13. Kuhl D., Bangert F., Mescheke G., 2004. Coupled chemo-mechanical deterioration of cementitious materials, Part I: Modeling. *International Journal of Solids and Structures* **41**, 15-40.
14. Kuhl D., Bangert F., Mescheke G., 2004. Coupled chemo-mechanical deterioration of cementitious materials, Part II: Numerical methods and simulations. *International Journal of Solids and Structures* **41**, 41-67.
15. Le Bellégo C., Gérard B., Pijaudier-Cabot G., Dubé J.-F., Molez L. 2003. Coupled mechanical and chemical damage in calcium leached cementitious structures. *Journal of Engineering Mechanics*, **129**(3), 333-341.
16. Mainguy M. Tognazzi C. Torrenti J.M., Adenot F., 2000. Modelling of leaching in pure cement paste and mortar. *Cement and Concrete Research* **30**, 83-90.
17. Mazars, J., 1984. Application de la mécanique de l'endommagement au comportement non linéaire et à la rupture de béton de structure. Thèse de doctorat d'Etat, Université Paris VI, France.

18. Peccelon, H., Blan, C., Mazoin, C., 2006. Long term behaviour of concrete, influence of temperature and cement binders on the degradation (decalcification/hydrolysis) in saturated conditions. *Revue Européenne de Génie Civil-10/2006 Concrete behaviour assessment*, pp. 1107–1125.
19. Pietruszczak, S., Jiang, J., Mirza, F.A., 1988. An elastoplastic constitutive model for concrete, *Int. J. Solids & Structures* **24**(7), 705-722.
20. Ramtani, S., Berthaud, Y., Mazars, J., 1992. Orthotropic behavior of concrete with directional aspects: modelling and experiments. *Nucl. Eng. Des.* **133**, 97–111.
21. Sellier, A., 2006. Modélisations numérique pour la durabilité des ouvrages de génie civil. Dissertation for Habilitation à Diriger des Recherches, Université Paul Sabatier, Toulouse.
22. Terrien M., 1980. Emission acoustique et comportement mécanique post-critique d'un béton sollicité en traction. *Bulletin de liaison de Pont et Chaussées*, 105, pp. 65-72.
23. Tognazzi C., 1998. Couplages fissuration – dégradation chimique dans les matériaux cimentaires: caractérisation. Thèse de doctorat de l'INSA de Toulouse.
24. Ulm F.-J., Torrenti J.-M, Adenot F., 1999. Chemoporoplasticity of calcium leaching in concrete. *Journal of Engineering Mechanics* **125**, 1200-1211.
25. Yurtdas I., Xie S., Secq J., Burlion N., Shao J.-F., Sibai M., 2006. Etude expérimentale du couplage entre le comportement thermo-hydro-mécanique et la dégradation chimique d'une pâte de ciment pétrolière. Rapport (LML-MGC-03-03/A) pour TOTAL.
26. Waller V. Relations entre composition des bétons, exothermie en cours de prise et résistance en compression. PhD thesis, ENPC Paris, 1999; p. 297.

6. Appendix

Table 1: Values of model parameters

parameter	CEM IF and VF
α_D	40
α_{dc}	10
\bar{d}_{pt}	0.8
b_{pt}	10^5
\bar{d}_{pc}	0.5
b_{pt}	10^2