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COUPLED DIFFUSION-DAMAGE MODELLING AND THE IMPLICATIONS ON FAILURE DUE TO STRAIN LOCALISATION

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Abstract—For underground cement-based structures, the leaching of pore solution ions by pure water is one of the reference scenario of degradation. The penetration of this chemical attack leads to a loss of mechanical properties of the material. Under mechanical loading, progressive damage can develop too. In order to evaluate the coupling between these chemical and mechanical mechanisms of degradation (increase of porosity and cracking) a model is developed. The chemical attack kinetics follows a non-linear diffusion equation based on the mass conservation of the calcium ions in concrete. The mechanical behaviour is modelled according to a scalar damage constitutive relation. After a brief presentation of the model, the influence of damage development on the chemical attack kinetics is analysed. It is shown that due to the chemical degradation (activated by diffusion mechanisms) localisation due to strain softening can be at the origin of the failure. In spite of the chemical coupling, the problem of equilibrium is ill posed at the inception of strain localisation. A non local damage model is implemented in order to alleviate this problem. A preliminary example of finite element computations illustrates the different regimes of damage on a bending beam subjected to a chemical attack and to a variable deflection.

INTRODUCTION

Concrete has been widely used in the construction of nuclear waste containment structures (concrete packages, surface vaults). The durability of these concrete barriers has to be assessed over long periods of time, typically 300 years for surface repositories. In that context, the reliable prediction of their long-term behaviour requires a sound knowledge of the various deterioration mechanisms that affect the structures over their life time, and the extensive use of simulation tools. This paper presents the main features of a modelling approach which has been developed in order to predict the long-term behaviour of low-level nuclear waste concrete barriers. The main originality of the approach resides in the fact that the model considers chemical and mechanical actions simultaneously and their mutual actions.

Of concern are short lived waste where irradiation and residual heat effects are negligible. The waste containers are generally kept in surface repositories, and it is reasonable to believe that these barriers are not subjected to any significant hydraulic gradient over their lifetime. It is generally accepted that a constant flow of pure water along the external surfaces of the concrete barriers is most critical. It initiates a slow leaching of calcium from the cement paste which is followed by micro structural changes such as an increase of porosity. These micro structural alterations are likely to increase both the diffusion and the

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permeation, and detrimentally affect the material properties. Even if the containers are not expected to be subjected to severe mechanical loads, several thermo-mechanical studies have clearly demonstrated that the cement hydration can initiate cracks in these structures (see e.g. Didry, 1993). Obviously, the development of these cracks should be influenced by the pore structure alterations induced by the calcium leaching process. Simultaneously, the presence of these cracks should affect the mass transfer properties of the material, and have a certain influence on the evolution of the calcium leaching process. Therefore, the development of the model requires the description of the evolution of the mechanical behaviour of concrete and its transfer properties as the micro structural alteration of the material develops, and at the same time the description of the evolution of the kinetic of alteration as mechanical damage develops.

For this purpose, we will start from a mechanical model based on damage mechanics. We will use a scalar damage model proposed by Mazars (1984) in order to model the response of concrete to a mechanical load. In section two of the paper, the mechanical model is enriched with an additional ageing variable which captures the influence of the chemical degradation. At the same time, the parameters in the diffusion equation which governs the chemical action are set to depend on damage in order to capture the influence of micro-cracking on the chemical process. The resulting model exhibits strain softening. In Section 3 the conditions for strain localisation are examined and it is found that it is necessary to incorporate an internal length in the model in order to avoid ill-posedness of the governing equations at the inception of localisation. Hence, a non local damage model is substituted to the mechanical model. Finally, a finite element computation is briefly presented and documents the capabilities of the model which remain to be validated against experimental data on structures.

MODEL DESCRIPTION AND FUNDAMENTAL BASES

Several possibilities exist for modelling progressive damage and fracture in quasi-brittle materials. One possibility is micromechanics and a second one is phenomenological modelling. Micromechanics refers directly to the micro structure of the damaged material, e.g. the shape and amount of defects (e.g. dissolved hydrate solids), and thus provides a direct connection with the type of damage which can be observed under the chemical action. Phenomenological models, however, are better suited for solving large scale problems because micromechanics of interacting cracks or voids in the presence of sharp gradients due to damage localisation is difficult to handle in practical situations. Considering the industrial applications (design of nuclear waste barriers), and the need to couple few complex physico-chemical mechanisms with the mechanical behaviour concrete, it has been preferred to work with a phenomenological approach.

Mechanical damage

In the present analysis, we use an isotropic scalar damage model due to Mazars (1984) which is briefly reviewed below. Detailed presentations of this constitutive relation and the determination of the model parameters can be also found in (Saouridis, 1988). In this model, the mechanical effect of progressive micro-cracking due to external loads (without load sign changes) is described by a single internal variable which degrades the Young's modulus of the material. The constitutive relations are:

$$\sigma_{ij} = (1-d)\Lambda_{ijkl}\varepsilon_{kl} \tag{1}$$

where σ_{ij} and ε_{ij} are the components of the stress and strain tensors, respectively ($i, j, k, l \in [1,3]$), Λ_{ijkl} are the initial stiffness moduli, and d is the damage variable. The material is initially isotropic, with E and ν the initial Young's modulus and Poisson's ratio, respectively. Damage is a function of the positive strains which means that it is mainly due to micro cracks opening in mode I. The following scalar called equivalent strain is defined:

$$\tilde{\varepsilon} = \sqrt{\sum_{i=1}^{3} (\langle \varepsilon_i \rangle_+)^2} \tag{2}$$

where $\langle \varepsilon \rangle_+ = 0$ if $\varepsilon_i < 0$ and $\langle \varepsilon_i \rangle_+ = \varepsilon_i$ if $\varepsilon_i \geqslant 0$; and ε_i are the principal strains. $\tilde{\varepsilon}$ is the variable that controls the growth of damage according to the following conditions:

$$F(\tilde{\varepsilon}) = \tilde{\varepsilon} - \kappa \tag{3a}$$

$$\text{if } F(\tilde{\varepsilon}) = 0 \quad \text{and} \quad \dot{F}(\tilde{\varepsilon}) = 0 \quad \text{then} \begin{cases} d = h(\kappa) \\ \kappa = \tilde{\varepsilon} \end{cases} \text{with the condition } \dot{d} \geqslant 0 \quad \text{else} \begin{cases} \dot{d} = 0 \\ \dot{\kappa} = 0 \end{cases} \tag{3b}$$

The two conditions $F(\tilde{\varepsilon}) = 0$ and $\dot{d} \geqslant 0$ define the damage surface, that is the reversible domain. In this domain, damage must not decrease because the rate of dissipated energy due to damage must remain positive or zero according to the second principle of thermodynamics (Lemaitre, 1996). Initially $\kappa = \kappa_0$, where κ_0 is the threshold of damage. κ_0 is the tensile strain at which damage is initiated. Typically it corresponds to the point where the maximum tensile stress f_t is reached in a uniaxial test:

$$\kappa_0 = \frac{f_t}{E} \tag{4}$$

In the evolution law, generically denoted as $h(\kappa)$ in eqns (3), damage is decomposed into two parts, d_t for tension and d_c for compression. d is computed from the relation (5) which combines these two types of damage:

$$d = \alpha_t d_t + \alpha_c d_c \tag{5}$$

The factors α_t and α_c are expressed as non dimensional functions of the principal strains (for detailed information see Mazars, 1984). They are such that in uniaxial tension $\alpha_t = 1$, $\alpha_c = 0$, and $d = d_t$, and $\alpha_t = 0$, $\alpha_c = 1$, and $d = d_c$ in uniaxial compression. The damage variables are functions of the softening parameter κ:

$$\begin{cases} d_t = 1 - \dfrac{\kappa_0(1-A_t)}{\kappa} - \dfrac{A_t}{\exp(B_t(\kappa-\kappa_0))} \\ d_c = 1 - \dfrac{\kappa_0(1-A_c)}{\kappa} - \dfrac{A_c}{\exp(B_c(\kappa-\kappa_0))} \end{cases} \tag{6}$$

Constants A_c, B_c, A_t, B_t are model parameters.

Description of leaching and chemical damage

In the case of leaching of pore solution ions in concrete by pure water, one of the most important phenomena (from the mechanical point of view) is that leaching produces an increase of the porosity of the material as certain constituents are being dissolved. As the porosity increases, the mechanical properties of the material decrease (Saito *et al.*, 1994; Gérard *et al.*, 1995; Carde, 1996). Since the calcium is the dominant element of the hydrated phases, the calcium concentration has often been chosen as a state variable (Greenberg, 1960; Berner, 1992). The equilibrium between the solid calcium phases (CaO/SiO_2) and the

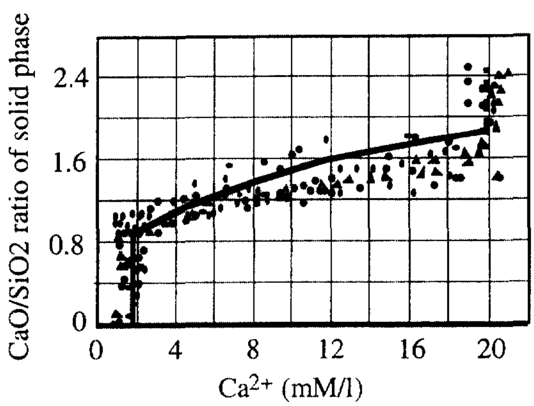

Fig. 1. CaO/SiO$_2$ ratio of the hydrated cement paste as a function of calcium concentration C in solution.

calcium concentration of the pore solution denoted as C is shown on Fig. 1. The equilibrium is characterised by a curve with two steps: one step is associated to the dissolution of Portlandite (at about 20 mmol/l of calcium in pure solution); the second step is associated to the total dissolution of C-S-H (about 2 mmol/l of calcium in pore solution). From a physical point of view, it is commonly accepted that this phenomenon can be described by a diffusion equation because the limiting factor in the development of the chemical attack is the diffusion of the calcium ions from the reaction sites. Hence, the kinetics of leaching is described with the local calcium mass conservation assuming a local equilibrium (the kinetics of dissolution reactions is very fast compared to diffusion process):

$$f(C)\frac{\partial C}{\partial t} = \mathrm{div}(D(C,d)\,\mathrm{grad}(C)) \tag{7}$$

where C is the concentration of calcium in the pore solution, $D(C,d)$ is the global diffusion coefficient of calcium and $f(C)$ is a function whose aim is to account for phase changes and dilution of pore solution due to the increase of porosity. Equation (7) is a non linear diffusion equation because of phase changes and because the diffusion coefficient is a function of the volume fraction of hydrates in the cement paste and of its chemically and mechanically induced variations. The parameters of the model are calculated from the chemistry of the cement used. Figure 2 shows the evolution of $f(C)$ and $D(C, d = 0)$ with

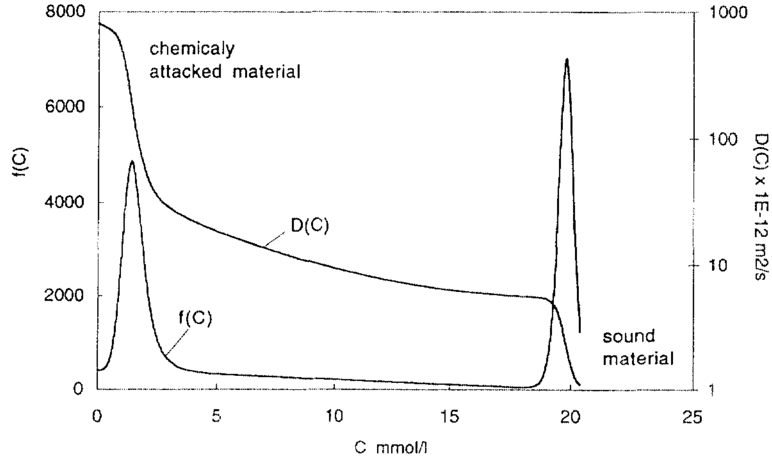

Fig. 2. Phase change function $f(C)$ and diffusion coefficient of calcium $D(C)$ as a function of calcium concentration C in pore solution.

the concentration of calcium ions C, as predicted by the chemical model (for details see Gérard, 1996).

The influence of the chemical reaction and the dissolution of material constituents in concrete is introduced in the constitutive relation by adding a new ageing state variable V (Gérard *et al.*, 1995). Since the dissolution of the material constituents increases the porosity of the material, we expect that its mechanical influence should be similar to void growth and thus produces a degradation of the material properties in an isotropic fashion. Therefore, it may seem logical to introduce this variable in the stress–strain relation in a way similar to mechanical damage. It takes a value between zero (for unattacked or chemically sound material) and one for totally dissolved material. The question is now to determine how chemical damage interacts with mechanical damage in the stress–strain relation and thus in the state potential defining the free energy in the material. If we assume that chemical damage and mechanical damage are two independent state variables in the free (recoverable) mechanical energy, a multiplicative combination seems more appropriate. This combination means that mechanical damage represents essentially the influence of microcracks while chemical damage describes void growth and that the two are not interacting. Therefore, the stress–strain relations in the presence of both mechanical and chemical damages are rewritten from eqn (1):

$$\sigma_{ij} = (1-d)(1-V)\Lambda_{ijkl}\varepsilon_{kl} \tag{8}$$

Note that an additive combination of the two types of damage could have been considered as well with a different meaning of the overall value of damage since it would have been the sum of the mechanical and chemical damage without any distinction on the type of damage involved, except in the conditions of growth of damage. We have preferred to use eqn (8) which clearly separates the two contributions referring to two different physical mechanisms. V is a function of the concentration C of calcium in pore solution:

$$V = g(C) \tag{9}$$

where g is an experimentally determined function. The evolution of the chemical damage variable with the calcium concentration was determined from micro-hardness experiments on leached cement paste samples (Gérard, 1996). Figure 3 shows the comparison of the expression of g, which is a generalisation of the proposal by Fagerlund (1973), with

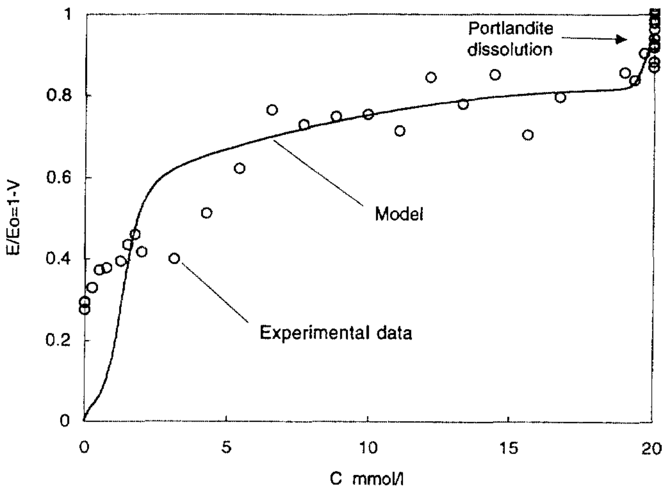

Fig. 3. Ageing variable evolution $V(C)$ as a function of calcium concentration C.

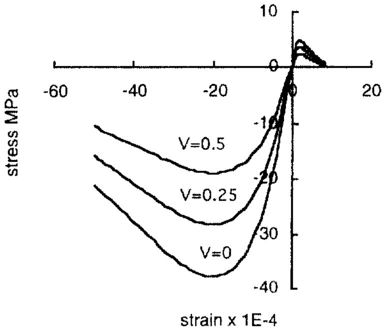

Fig. 4. Mechanical constitutive law of leached concretes. When $V = 0$ the material is chemically sound.

experimental data. Figure 4 presents the compression–tension behaviour of a concrete sample at different levels of homogeneous chemical damage.

As we have seen in eqn (7), the mechanical evolution of damage will affect the diffusion of the ions in the material too. The variation of the diffusion coefficient due to damage is, in the following, arbitrarily fixed as a S function (Gérard, 1996):

$$D(C,d) = \max(D(d), D(C)) \quad \text{and} \quad D(d) = D_0 + D_{\max}\left[1 - \frac{1}{1+\left(\dfrac{d}{d_{cr}}\right)^n}\right] \qquad (10)$$

where D_0 is the value of the diffusion coefficient in the absence of chemical attack, the tritiated water diffusion coefficient of the material is used (about 10^{-12} m^2/s). $D_{\max}$ is its value for a completely cracked material, n and d_{cr} are model parameters ($n = 5$, $d_{cr} = 0.4$). Equation (10) proceeds from the assumption that the diffusion coefficient should be affected by 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, $D_{\max}$ cannot exceed the diffusion coefficient in free water which is about 10^{-9} m^2/s (we took here $D_{\max} = 0.8 \times 10^{-9}$ m^2/s). It is important to note that eqn (10) does not derive from experimental observations because experimental data are hardly available. Since we took the largest possible range of variation of the diffusion coefficient and since an increase of the diffusivity inevitably induces an increase of the chemical attack and damage rates, the proposed model should yield pessimistic predictions and results which remain on the safe side as far as the lifetime prediction of structures is concerned.

ANALYSIS OF STRAIN LOCALISATION

In mechanical problems, the unrealistic features of damage localisation due to strain softening are usually circumvented by the introduction of an internal length in the continuum description which scales the localisation process and controls the size of the damage/strain localisation zone (Pijaudier-Cabot and Benallal, 1993; de Borst *et al.*, 1993). Motivations for introducing such an internal length in the continuum description are based, for instance, on micromechanics of interacting crack systems (Bazant, 1987).

In order to determine whether the chemical/mechanical model needs the same approach, we are going to investigate the occurrence of bifurcation in an infinite body from a homogeneous state of deformation, mechanical and chemical damages and calcium concentration denoted as $(\varepsilon_{ij}^0, d^0, C^0, V^0)$. In the foregoing analyses, the stress–strain relations will be linearised, assuming that the loading condition is satisfied everywhere (i.e. mechanical damage grows everywhere).

Infinitesimal perturbations about equilibrium in an infinite body where strain and damage are initially homogeneous have to satisfy the two governing equations:

$$\mathrm{div}(\dot{\sigma}) = 0 \tag{11a}$$

$$f(C^0)\frac{\partial \dot{C}}{\partial t} = \mathrm{div}(D(C^0)\,\mathrm{grad}(\dot{C})) \tag{11b}$$

It is important to note that eqns (11) are written in a rate format which means that we are looking for perturbations of the initial state of equilibrium within a small time interval δt which can be eliminated from the equations. Substitution of the constitutive relations [eqns (2)–(6), (8)] in the equation of equilibrium eqn (11a) yields:

$$[(1-V^0)\Lambda^{\mathrm{t}}_{ijkl}\dot{\varepsilon}_{kl}]_{,j} = (1-V^0)\Lambda^{\mathrm{t}}_{ijkl}[\dot{\varepsilon}_{kl}]_{,j} = \left[\frac{\partial g(C)}{\partial C}\Lambda^{\mathrm{u}}_{ijkl}\varepsilon^0_{kl}\frac{\partial C}{\partial t}\right]_{,j} \tag{12}$$

where $\Lambda^{\mathrm{t}}_{ijkl}$ and $\Lambda^{\mathrm{u}}_{ijkl}$ are the coefficients of the tangent and unloading operators, respectively, in the mechanical model where the chemical action is omitted. Note that the right hand-side term in this equation can be expressed as a function of the initial state and the evolution of $\dot{C}$. This term does not affect the nature of the partial differential equations of equilibrium and the condition of equilibrium in the chemo-mechanical model is of the same nature as in the mechanical model alone. More precisely, the nature of eqn (12) is governed by the nature of eqn (13):

$$\Lambda^{\mathrm{t}}_{ijkl}[\dot{\varepsilon}_{kl}]_{,j} = 0 \tag{13}$$

which is the governing equation in the mechanical problem in the absence of chemical action. Therefore, the equations of equilibrium (11a) become ill-posed at the inception of strain localisation. In the following, we will deal with the simple one-dimensional case and document this assertion more thoroughly.

One-dimensional problem

We will restrict here the analysis to a one-dimensional continuum. The assumptions are the same as in the previous analysis. We look for possible bifurcation points depending on the initially homogeneous state. In a one-dimensional setting, the equations governing the mechanical and chemical infinitesimal perturbations are:

$$\frac{\partial \dot{\sigma}}{\partial x} = 0, \quad f(C^0)\frac{\partial \dot{C}}{\partial t} = D(C^0, d^0)\frac{\partial^2 \dot{C}}{\partial x^2} \Leftrightarrow \frac{\partial \dot{C}}{\partial t} = D^*\frac{\partial^2 \dot{C}}{\partial x^2} \tag{14}$$

Bifurcation exists if there are several possible solutions to this system of partial differential equations. The stress–strain relation in the rate form is:

$$\dot{\sigma} = (1-V^0)E_{\mathrm{t}}\dot{\varepsilon} - \frac{\partial g}{\partial C}E_{\mathrm{u}}\varepsilon^0\dot{C} \tag{15}$$

with

$$E_{\mathrm{t}} = (1-d^0)E - \frac{\partial h}{\partial \varepsilon}E\varepsilon^0, \quad E_{\mathrm{u}} = (1-d^0)E$$

With eqn (15) and with the compatibility conditions, the two governing equations are rewritten in term of the perturbation sought, i.e. in term of the rate of displacement and the rate of concentration of calcium:

$$(1-V^0)E_t\frac{\partial^2 \dot{u}}{\partial x^2}-\frac{\partial g}{\partial C}E_u\varepsilon^0\frac{\partial \dot{C}}{\partial x}=0 \tag{16a}$$

$$\frac{\partial \dot{C}}{\partial t}-D^*\frac{\partial^2 \dot{C}}{\partial x^2}=0 \tag{16b}$$

Equation (16b) is a parabolic equation, which means that infinitesimal perturbations ought to be time dependent. Hence, chemical coupling is going to introduce a time scale related to diffusion. This equation can be solved separately:

$$\dot{C}=M\int_0^z \exp(-z^2)\,\mathrm{d}z \quad \text{with } z=\frac{x}{\sqrt{D^*t}} \tag{17}$$

where M is a constant which depends on the boundary conditions. Substitution of this solution in eqn (16a) yields the displacement and strain rates under the condition that the tangent modulus E_t is not zero and that the ageing variable is not one:

$$\dot{u}=K\int_0^x\left[\int_0^z \exp(-z^2)\,\mathrm{d}z\right]\mathrm{d}x, \quad \text{with } K=\frac{\frac{\partial g}{\partial C}E_u\varepsilon^0 M}{(1-V^0)E_t}$$

$$\dot{\varepsilon}=\frac{K}{M}\dot{C} \tag{18}$$

According to these equations, the strain rate distribution is entirely controlled by the concentration of calcium ions in the bar. It follows from these solutions that bifurcation cannot exist and that the solution exists, is unique, and depends linearly on the boundary conditions before the inception of strain softening, i.e. before the tangent modulus vanishes.

At this particular point, the first term in eqn (16a) vanishes and the equilibrium condition has no solutions because the concentration rate in calcium ions cannot satisfy at the same time the diffusion and equilibrium equations (except when it is zero identically). It is still possible, however, to find a solution in the dynamic regime. In this situation, the second-order time derivative of the velocity would be proportional to the gradient of the concentration rate.

In the softening regime, the nature of the equations of equilibrium is that of the mechanical problem alone. Accordingly, bifurcation to a discontinuous strain rate distribution may occur. Therefore, solving the governing equations in static's is an ill posed problem and there is a need for regularising the equation of equilibrium. This situation is similar to the case of porous saturated soils where strain localisation may occur (see e.g. Rudnicki, 1983) and where localisation limiters need to be implemented (Larsson *et al.*, 1996).

Non local damage and localisation analysis

In order to avoid ill-posedness due to strain softening, the mechanical model can be modified and enriched with an internal length (Pijaudier-Cabot and Bazant, 1987). We have chosen here the non local version of the damage model described in the first section due to Saouridis (1988). The non local variable $\bar{\varepsilon}$ which represents the average of the equivalent strain eqn (2) over the representative volume surrounding each point in the material is defined:

$$\bar{\varepsilon}(x)=\frac{1}{V_r(x)}\int_\Omega \psi(x-s)\tilde{\varepsilon}(s)\,\mathrm{d}s \quad \text{with } V_r(x)=\int_\Omega \psi(x-s)\,\mathrm{d}s \tag{19}$$

where Ω is the volume of the structure, $V_r(x)$ is the representative volume at point x, and $\psi(x-s)$ is the weight function:

$$\psi(x-s) = \exp\left(-\frac{4\|x-s\|^2}{l_c^2}\right) \tag{20}$$

l_c is the internal length of the non local continuum. This length is usually taken in the calculations as $l_c \approx 3d_a$ in which d_a is the maximum size of the aggregate in normal strength concrete (Bazant and Pijaudier-Cabot, 1989).

The averaged effective strain $\bar{\varepsilon}$ is the variable that controls the growth of damage. Therefore $\bar{\varepsilon}$ is substituted to $\tilde{\varepsilon}$ in the loading function and evolution equations [eqns (3) and (6)] of the mechanical model. The role of the internal length in this type of non local model has been elucidated by Pijaudier-Cabot and Benallal (1993). It controls the wave length of the bifurcation mode at the inception of softening and prevents perturbations with an arbitrary wave length from being solutions of the equilibrium conditions. Furthermore, the fracture energy of an infinite specimen is proportional to this internal length (Mazars and Pijaudier-Cabot, 1996). One assumption of the coupled chemo-mechanical non local model is that the chemical degradation affects also the fracture energy. The fracture energy of a homogeneously attacked material depends on ageing, on the internal length and on the softening parameters in the evolution of damage A_t and B_t [eqns (6)]. It can be demonstrated from eqn (8) that this energy decreases linearly as the ageing variable increases.

We have seen that the equations governing equilibrium in the coupled chemomechanical problem are of the same nature as the equations of equilibrium in the absence of chemical effects eqns (11) and (12). Hence, it is expected that the non local chemomechanical model yields the same regularisation properties as the non local damage model without chemical effects. As an example, let us consider again the one-dimensional model in the case of an infinite bar. In the non local model, the stress–strain relation becomes (monotonic loading):

$$\dot{\sigma} = (1-V^0)\left[(1-d^0)E\dot{\varepsilon} - \frac{\partial h}{\partial \varepsilon}E\varepsilon^0\int_\Omega \psi^*(u)\dot{\varepsilon}(x+u)\,\mathrm{d}u\right] - \frac{\partial g}{\partial C}\dot{C}E_u\varepsilon^0 \tag{21}$$

with

$$E_u = (1-d^0)E$$

where ψ^* is the normalised weight function (i.e. divided by the representative volume). The equation governing equilibrium of small perturbations is an integro-differential equation whose closed-form solution can hardly be found. As an approximation, one can expand the term in the integral of eqn (21) in Taylor series up to the second order and transform eqn (11a) into a differential equation:

$$\int_\Omega \psi^*(u)\dot{\varepsilon}(x+u)\,\mathrm{d}u \approx \int_\Omega \psi^*(u)\left(\dot{\varepsilon}(x) + \frac{\partial\dot{\varepsilon}}{\partial x}u + \frac{\partial^2\dot{\varepsilon}}{\partial x^2}\frac{u^2}{2}\right)\mathrm{d}u \approx \dot{\varepsilon}(x) + \alpha\frac{\partial^2\dot{\varepsilon}}{\partial x^2} \tag{22}$$

with

$$\alpha = \int_\Omega \psi^*(u)\frac{u^2}{2}\,\mathrm{d}u$$

The first-order derivative of the rate of strain vanishes because of the symmetry of the weight function. Note that the dimension of α is m^{-2}, the inverse of the square of an internal length. Upon substitution of eqns (21) and (22) in the governing eqns (11), the following system of equations is obtained:

$$(1-V^{0})\left\{E_{t}\frac{\partial\dot{\varepsilon}}{\partial x}-\alpha E^{*}\frac{\partial^{3}\dot{\varepsilon}}{\partial x^{3}}\right\}=\frac{\partial g}{\partial C}E_{u}\varepsilon^{0}\frac{\partial\dot{C}}{\partial x} \tag{23a}$$

$$\frac{\partial\dot{C}}{\partial t}-D^{*}\frac{\partial^{2}\dot{C}}{\partial x^{2}}=0 \tag{23b}$$

with

$$E_{t}=(1-d^{0})E-\frac{\partial h}{\partial\varepsilon}E\varepsilon^{0},\quad E^{*}=\frac{\partial h}{\partial\varepsilon}E\varepsilon^{0},\quad E_{u}=(1-d^{0})E$$

The solution of eqn (23a) is the sum of the solution of the homogeneous differential equation and a particular solution. Again, the homogeneous differential equation corresponds exactly to the situation where chemical effects do not appear. The solution of the homogeneous equation is:

$$\dot{\varepsilon}(x)=A\cos(kx)+B\sin(kx)+C\quad\text{with}\,k=\sqrt{\frac{-E_{t}}{\alpha E^{*}}} \tag{24}$$

where A, B, C are unknown constants which depend on the boundary conditions. A particular solution of eqn (23a) is:

$$\dot{\varepsilon}(x)=\int\frac{\partial g}{\partial C}E_{u}\frac{\varepsilon^{0}}{k(1-V^{0})}v(x)\,\mathrm{d}x$$

$$v(x)=\int-\exp\left(-\frac{x^{2}}{4D^{*}_{t}}\right)\cos(kx)\,\mathrm{d}x\sin(kx)+\int\exp\left(-\frac{x^{2}}{4D^{*}_{t}}\right)\sin(kx)\,\mathrm{d}x\cos(kx) \tag{25}$$

The rate of strain is the sum of the two solutions in eqns (24) and (25). The characteristic dimension (wave length) of the solution in eqn (24) is governed by the mechanical internal length through the quantity α. The characteristic length/time involved in eqn (25) depends on the diffusion process. Hence the two relevant length scales of the problem interact in the solution.

The above analysis indicates that finite element solutions should properly converge towards a mesh independent, unique, solution upon element size refinement with a gradient model or a non local one for which the approximation in eqn (22) holds. The same conclusion is expected (although not demonstrated) without the approximation of eqn (22) where the non local model is degenerated to a gradient model because well-posedness of the equations in the coupled problem is controlled by well-posedness of the equilibrium equations in the mechanical problem. The bifurcation analysis provides also some useful information concerning the element size which is to be used in optimal meshes. In the present case of leaching of pore solution by pure water in concrete, the diffusion coefficient D^{*} is extremely small, in the range of 10^{-10} m^2/s. One can calculate for a small perturbation/time interval the order of magnitude of the penetration depth (characteristic length) of the perturbation of the chemical attack as:

$$l_{d}\approx\sqrt{D^{*}t} \tag{26}$$

The order of magnitude of this length at the inception of localisation where the time interval is small is in the range of 0.1 mm. As a consequence, there is an obvious need for very small finite elements if converged solutions (with respect to the discretisation size) are sought for small time intervals near failure. On the other side, the internal length in the mechanical problem is classically in the range of a few centimetres for concrete. It follows that there is a mismatch between the two relevant length scales of the chemical and mechanical problems, respectively. This mismatch should be looked at as a computational problem since the governing equations are well posed. In an optimised computational scheme, different discretisations according to the respective length scales, or a multiple scale analysis should be preferred for solving the chemical and mechanical problems.

NUMERICAL SIMULATIONS

The model has been implemented in the finite element code Castem 2000 developed by the French atomic agency (Jeanvoine and de Gayffier, 1995). A staggered scheme has been used for solving the coupled equilibrium-diffusion equations. This scheme is outlined in Fig. 5. The coupled chemo-mechanical problem is solved in a semi-coupled fashion. Given a time increment, the diffusion equation is integrated first assuming that damage and diffusivity are constant. The time integration method implemented for this purpose is a two-step time scheme which is explicit with a second order accuracy (Dupont *et al.*, 1974). Then, the ageing variable is updated and the mechanical problem is solved for the considered time/load increment with a constant ageing variable. Afterwards, damage and diffusivity are updated and the computation of the next time step can start. The mechanical equation is solved with a modified Newton–Raphson algorithm with constant stiffness matrix, equal to that of the elastic problem. This algorithm is not very effective compared to a standard Newton–Raphson technique which requires the derivation of a (consistent) tangent operator. The implementation of a non local integral damage model would, however, severely complicate the derivation of a tangent operator which is non symmetric and has a large bandwidth due to non local interactions (Pijaudier-Cabot and Huerta, 1991). This staggered solution scheme may not be extremely robust. In particular, it is quite sensitive to the size of the time/load increments. Small time steps have been employed in the prototype computations that are presented below in order to improve the accuracy of the computation. Six nodded finite elements were used for the spatial discretisation. The element size was refined at mid-span, in the expected localisation zone, where the size of the (square) elements was approximately 0.5 cm.

In order to illustrate the influence of the chemical process on failure, a three-point bending beam subjected to an external deflection and a chemical attack by pure water on the tensile face (bottom) is presented (Fig. 6). This is a plane stress calculation performed on a beam of unit thickness. Figure 7 shows the applied deflection vs time. Initially, the tensile stress at the bottom is about 3.5 MPa (about 60% of the tensile strength). The internal characteristic length of the concrete is 15 mm and the model parameters in the mechanical model are $E = 3500$ MPa, $\nu = 0.2$, $\kappa_0 = 1.5 \times 10^{-4}$, $A_t = 1.0$, $B_t = 6000$ (damage does not occur in compression in this example). Chemical damage propagates from the bottom to the upper faces of the beam. Due to leaching, the strain increases in the central part of the beam and mechanical damage is triggered. Figure 8 shows the maximum penetration depth of the chemical attack and of damage for $d = 0.99$ along the cross section

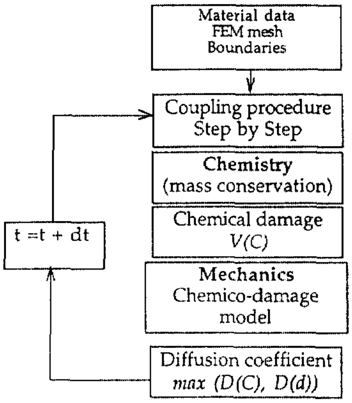

Fig. 5. Organisation of the numerical simulations.

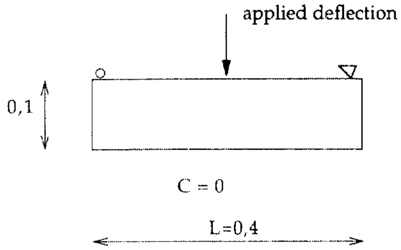

Fig. 6. Three points bending beam subjected to leaching on the tensile face and to an applied deflection at mid-span.

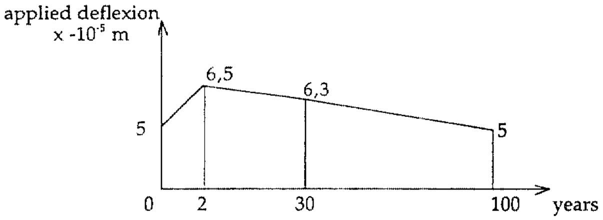

Fig. 7. Applied deflection vs time.

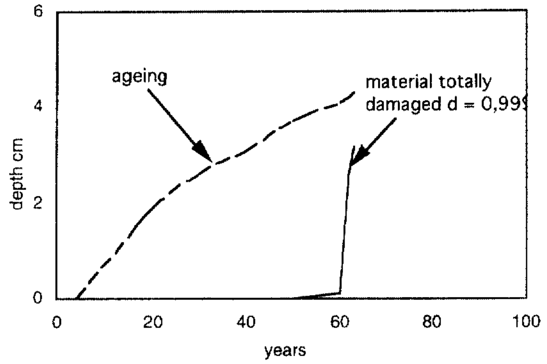

Fig. 8. Leaching and damage penetration depth vs time along the central axis.

of the beam at mid span. During the first 50 years, the chemical attack propagates as a square root function of time, which is typical of diffusion. Mechanical damage is not initiated. After 60 years a sharp evolution of damage is noticed with a failure of the beam at about 64 years. In this problem, the degradation of the elastic stiffness is first controlled by the diffusion process with a very small length scale. Since the elastic strains are increased when the degradation proceeds, mechanical damage starts to develop until strain localisation occurs. At this point, the mechanical length scale of the problem becomes relevant and it is this quantity which governs the failure process.

Several numerical simulations have been run with the same mesh and different internal lengths in the non local model. In these computations, the applied deflection was that of Fig. 7. The predicted times to failure are collected in Table 1. We found that the higher the internal length is, the longer the service life of the beam is predicted. The internal length appears to have a great influence on the quantification of damage propagation, and more

Table 1. Evolution of the time to failure of bending beams with the internal length

Internal length	0 cm	1 cm	1.5 cm	2 cm
Time to failure	18 years	30 years	64 years	> 300 years

particularly on the inception of failure. Therefore it is clear that this model parameter deserves a special attention as far as its experimental determination is concerned. For practical purposes, it would be useful to assess the accuracy of the experimental determination of the internal length in order to evaluate the degree of accuracy of computational predictions. Future studies should definitely concentrate more thoroughly at understanding better the sensitivity of this internal length and also the influence of the calcium and ageing distributions on the inception of strain localisation.

CONCLUSIONS

The leaching of pore solution ions by pure water is one of the reference scenario of degradation for underground cement-based structures. The penetration of this chemical attack leads to a loss of mechanical properties of the material and external loads may create damage at the same time. In order to describe the coupling between these chemical and mechanical mechanisms a model has been developed and implemented in a finite element code.

Chemically induced damage introduces, via a diffusion equation, an additional intrinsic time scale which can be regarded also as a scaling of the localisation of damage process leading to failure. Chemical coupling cannot, however, act as a localisation limiter and the difficulties encountered when dealing with strain softening in a mechanical problem are still to be solved. One possibility explored in this paper is the implementation of a non local model which incorporates an internal length scale and preserves the uniqueness of the solutions to boundary/initial value problems. Finite element computations of beams subjected to an external deflection show that the growth of damage at low load levels is first governed by the diffusion equation controlling the chemical process. At the inception of strain localisation, the internal length in the mechanical model becomes the leading scale as damage propagates in a region of the structure which is not affected by leaching.

Same as in the mechanical problem, the mechanical internal length scale controls the energy dissipation at failure and the instability of the structure. This underlines the necessity of determining with a good confidence the internal length in order to assess the service life period of concrete confinement barriers.

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