

Multiscale poro-creep model for cement-based materials

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SUMMARY

Experimental observations clearly show that the relative humidity (h_r) conditions influence significantly the creep behavior of cement-based materials, indicating that the water present within these materials plays a crucial role. This work presents a creep model for hardened cement pastes (HCP), based on a multiscale homogenization approach. It takes into account both free and adsorbed water contained in the porosity and investigates their effects on the HCP macroscopic creep behavior. The calcium silicate hydrate phase is assumed to be linear viscoelastic, and the Mori–Tanaka scheme is applied in the Laplace–Carson space to the composite formed of porosity, calcium silicate hydrate, and the other main hydrated compounds (which behavior is linearly elastic) by making use of the correspondence principle. With this model, estimations of the evolution of the macroscopic creep behavior of HCP submitted to constant external loading are examined under different h_r and compared with available experimental data. Finally, a method for implementing the model in a finite element code is proposed, and simulations of standard creep tests are performed to assess its validity. Copyright © 2011 John Wiley & Sons, Ltd.

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1. INTRODUCTION

Cement-based materials are massively employed in the domain of construction, also being seen as attractive options in the context of nuclear waste storage. For these applications, the long-term durability (i.e. the ability to resist weathering action, chemical attacks, mechanical loadings, and other environmental actions) and performance of these materials are of particular importance. A good knowledge of the environment impact on their macroscopic properties is then a prerequisite for a precise calculation and dimensioning of structures. In particular, their time-dependent behavior is known to considerably affect the overall response of structures and may lessen their service life because of excessive deformations resulting in additional damage and cracking. To calculate the long-term deformations of a structure made of a cement-based material during a given design life, the macroscopic time-dependent stress–strain relation of this material and the effects of relative humidity (h_r), temperature, and other factors on that relation are required. It is well known that the relationships between microstructure and macroscopic properties are at the heart of modern materials science. It is therefore essential to have a good knowledge of the microstructure of cement-based materials as well as a precise understanding of the physical mechanisms at the origin of creep phenomenon in order to accurately predict their time-dependent behavior. One of the main characteristics of cement-based materials

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is that they exhibit a heterogeneous and hierarchical microstructure (e.g., [1]). Moreover, the main solid phases composing them behave differently regarding creep.

The time-dependent and rate-dependent behaviors of calcium silicate hydrate (C-S-H) phase when submitted to mechanical or hydric loadings is now well recognized (e.g., [2]), whereas crystallized hydrated compounds (for example portlandite) and aggregates (which represent about 70% of concrete in volume) have an elastic behavior. Therefore, it is quite difficult to develop realistic micromechanics-based models to reproduce the creep behavior of cement-based materials. Over the last 50 years, numerous models have been proposed for describing creep of concrete. The reader can refer to the papers of Bazant and co-workers [3–6], where a rather comprehensive list of publications on the subject is provided. Most of the proposed models are established at the macroscopic scale because of the complexity of the underlying physical mechanisms and microstructures. In some sophisticated models, dependence of the behavior upon the h_r and temperature has been introduced in a phenomenological way leading to nonlinear viscoelastic behavior (e.g. [7]). Consequently, the parameters involved in these models may lack a clear physical meaning and need being identified with specific experiments at various external conditions (h_r , stress level, temperature). These experiments have to be repeated further for each considered material, which makes the identification procedure long, fastidious, and expensive. Such considerations render micromechanics approaches attractive because they incorporate microstructure information and take into account phenomena at the corresponding scale, thus facilitating the physical interpretation of the involved parameters. It is then expected to considerably reduce the experimental needs owing to the increased predictive capacities of the models, which integrate morphology characteristics (volume fractions, geometrical forms, and arrangement. . .) and intrinsic physical properties (elastic moduli, creep functions. . .) of the different phases composing the materials. However, the main physical mechanisms generating creep are not totally understood and explained at the microscopic and nanoscopic scales, although the crucial role of water is widely recognized. Indeed, there are ample experimental evidence showing a dramatic influence of the external h_r (and then of the water content) on the concrete creep (e.g., [8]). The quantity of water contained in the material and its location either in large pores or confined in very narrow pores, where the influence of interfaces are significant, are of particular importance when addressing the problem of creep of cement-based materials (e.g. [7]). Another aspect, which considerably affects the creep properties, is the aging of the material, that is, the evolution of its microstructure caused by the hydration process (e.g., [3–7], [9,10]). In particular, it is well known that the time at which the external mechanical loading is applied, especially at an early age when the extent of the hydration reactions is low, influences greatly the material response in terms of short-term and long-term deformations.

The time-and-rate dependence exhibited by some phases of the cementitious materials can be modeled by a viscoelasticity or viscoplasticity approach. Whereas micromechanical schemes have been well-developed in the case of linear elasticity for estimating the effective moduli of multiphase materials, they are no more valid when one phase has a time-dependent behavior. In the particular case of linear viscoelasticity, this problem can be formally solved by using the correspondence principle (see e.g. [11]). This principle is based on the Laplace–Carson transform, which allows to convert a linearly viscoelastic problem in the time space into a linearly elastic one in the transform space. Consequently, the classical micromechanical models, such as the self-consistent scheme and the MT scheme, can be applied to estimate the effective properties of composite materials with linearly viscoelastic phases in the transform space [12–14]. Nevertheless, the effective viscoelastic properties in the time space cannot easily be derived from the effective elastic ones in the transform space. Except for some special cases [14], no analytical solutions are available, and numerical methods are necessary for achieving this inversion in the time space (e.g., [15]).

In the present paper, a comprehensive creep model is developed for hardened cement pastes (HCP) based on a multiscale homogenization approach. Particular attention is paid to the influence of the h_r on the creep properties. The effects of the water present in HCP under different conditions and states are explicitly taken into account. We consider that the hydration processes of the cement are such that no significant microstructure evolutions are expected when the external loading is applied (typically several months after the material confection), and consequently, the effects of aging are not taken into account. The paper is organized as follows. In section 2, the different types of pores are classified according to their sizes, and the respective forms of water, which may be encountered in those pores

are detailed. In section 3, a micromechanical creep model for HCP is elaborated. First, the static formulation of the C-S-H caused by the effects of mesopores is studied. Then, in the Laplace–Carson space, a multiscale homogenization method is applied with the help of the correspondence principle to obtain the macroscopic properties of HCP. The behavior of cement-based materials is modeled as the one of heterogeneous poroelastic materials, in which both mechanical and hydromechanical properties depend upon the intrinsic properties and volume fractions of the phases, porosity, and an equivalent pore pressure. The numerical method owing to Donolato [16] is adopted to invert the Laplace–Carson transform so as to obtain the effective viscoelastic properties in the time space. In section 4, the obtained numerical results are confronted to available experimental ones in order to identify the parameters involved. The proposed multiscale poro-creep model is further implemented in the FE code Cast3M in section 5; simulations of a few standard creep tests are performed and the corresponding numerical results are analyzed. Finally, in section 6, a few concluding remarks are provided.

2. MICROSTRUCTURE OF CEMENT-BASED MATERIALS

A detailed investigation of the microstructure of cement-based materials is primarily required to understand their creep mechanisms. As the effects of water are of particular importance in this regard, the role of the different types of water present in the multiscale porous microstructure should be studied carefully in addition to the specification of the phases involved and their mechanical properties, distribution, sizes, and shapes. The pore systems of cement-based materials are multiple. In particular, the individual pores may vary greatly both in size and in shape. The mechanical and physical behaviors of the fluid phases in a pore system depend strongly on the size ranges of the pores involved [17]. According to the degree of difficulty or ease with which water can be removed from the pores, they can be classified into nanopores, mesopores, and macropores. In mesopores whose size ranges from about 2–2.5 to 50 nm, capillary phenomena take place. For capillary phenomena, we refer to [18–20]. In nanopores of size inferior to 2–2.5 nm, water can be held by hydrogen bonding, and the interaction potential is significantly higher than that in wider pores owing to the proximity of the walls. Therefore, water in nanopores is significantly different from bulk water. In general, it can be considered as a thin film (see [21]).

The crucial effect of the C-S-H phase on the long-term deformations of cementitious materials, and in particular on creep, is now well recognized (see e.g., [22,23]). In the following, we overview the multi-scale microstructure, types, amounts, sizes of phases in HCP, and then we specify the role of water in the different types of pores in relation with creep behavior.

2.1. Multi-scale microstructure of hardened cement pastes

HCP has a fair complex microstructure (e.g., [24–26]). The characteristic sizes of the phases constituting HCP range from nanoscale to microscale. Referring to [2,27,28], HCP can be considered as having a three-level hierarchical microstructure (see Figure 1), which can be described as follows:

- At level I ranging from 1 nm to 10 nm, the C-S-H gel exhibits a layered microstructure. The inter-layer space is very small and varies from 5 to 25 Å. It has been suggested that a monomolecular water layer, called the interlayered water, fills in the space and is strongly held by hydrogen bonding. At this scale, the physical phenomena involved in the solid–liquid interactions are extremely complex, especially when the material is submitted to mechanical loads and weathering actions. Attraction and repulsion forces, as well as disjoining pressure, play an essential role. No simple and reasonable models are now available to elucidate the microstructure–property relationships.
- At level II ranging from 10 nm to 1 μm , the C-S-H matrix behaves, in general, as a heterogeneous material, which can be divided into the inner high-density C-S-Hⁱⁿⁿ and the outer low-density C-S-H^{out} [29]. They differ essentially by the respective volume fraction of porosity. For simplicity, no distinction between C-S-Hⁱⁿⁿ and C-S-H^{out} will be made in this paper. We assume that the mesopores with equivalent radius smaller than the capillary radius of the meniscus of the gas–liquid interface (see next section) are totally filled with water whereas those with larger size are recovered by a thin water film, called adsorbed water layer. The latter results from

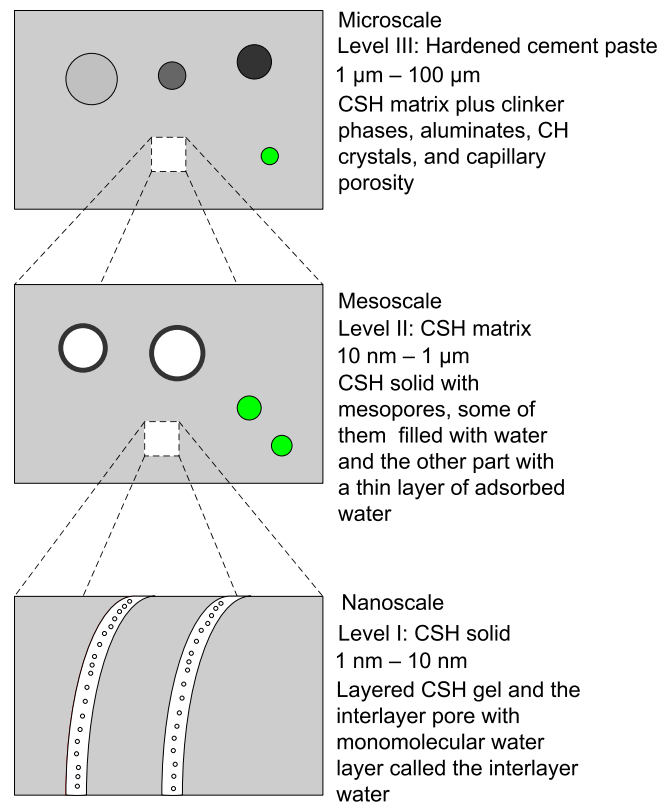


Figure 1. Multiscale microstructure of HCP.

chemisorption and physisorption phenomena. The respective proportions of pores filled with water and only covered by adsorbed water evolve as a function of the h_r . In this paper, these two types of water are taken into account, and their effects on the macroscopic creep are investigated. Referring to [30], the typical thickness t of an adsorbed water layer can be estimated by the following empirical formula:

$$t = 3.85 - 1.89[\ln(-\ln h_r)] \quad (\text{in } \text{\AA}) \quad (1)$$

- At level III ranging from 1 to 100 μm , the cement paste is considered as a composite in which the main hydrated crystals, that is, portlandite (CH) and calcium sulfoaluminate hydrates (CASH), as well as unhydrated clinker (UC) and capillary pores are embedded as inclusions in the homogenized C-S-H matrix. The capillary pores at this level are either totally filled with water or totally empty according to the degree of saturation, and the adsorbed water layers are neglected.

The previous description specifies the microstructure of HCP and classifies the different pores. It should, however, be noticed that other representations of HCP exist. In particular, as an alternative to the matrix-inclusion representation, some models describe the inner C-S-Hⁱⁿⁿ as a spherical layer surrounding the remaining unhydrated part of cement grains and the outer C-S-H^{out} phase either as a matrix (e.g., [31]) or a second spherical layer (e.g., [32,33]).

In the following, we examine the effects of the water present in the mesopores of HCP.

2.2. Physical properties of the different types of water in the mesopores of C-S-H

As mentioned earlier, there exist two types of water in the mesopores of C-S-H (see Figure 1 at level II), namely the capillary water and the adsorbed water. For simplicity, only spherical pores are considered in the sequel. Let us first write the Kelvin equation, which expresses the liquid–vapor equilibrium

under atmospheric conditions for the capillary water:

$$P_c = -\frac{\rho_l}{M_v} RT \ln h_r \quad (2)$$

where M_v is the molar mass of water, R the universal gas constant, T the absolute temperature, ρ_l the water density, and P_c the capillary pressure given by

$$P_c = p^g - p^l \quad (3)$$

with p^g and p^l denoting the gas and liquid pressures, respectively. Usually, the gas pressure p^g is small enough to be negligible with respect to the liquid pressure p^l for typical h_r conditions, so that

$$P_c = p^g - p^l \simeq -p^l. \quad (4)$$

Thus, Equation (2) reduces to

$$p^l \simeq \frac{RT}{V_m} \ln h_r. \quad (5)$$

In the C-S-H phase, the adsorbed water in the desaturated pores forms a thin layer on the pore solid wall surfaces, separating the solid and gas phases. The gas phase is therefore never in contact with the solid phase in such pores. This type of water can be physically considered as thin films, whose behavior is significantly different from that of the free capillary water. In the following, we focus our attention on the mechanical effects of the thin films of adsorbed water on the solid phase. From the standpoint of modelling, we adopt here the approach proposed by Rusanov [34,35], who extended the method of Derjaguin *et al.* [36], for modelling the thin water films. This approach consists in representing a thin liquid film as a layer with a finite thickness t and in introducing the disjoining pressure Π as an equilibrium characteristic. Let us consider the general case of an arbitrarily curved thin liquid film α of constant thickness t lying between a solid phase γ and a gas phase β , as shown in Figure 2. The inner and outer surfaces of the thin liquid film are denoted by Γ_1 and Γ_2 , respectively. The pressures p^α and p^β in the thin liquid film and in the gas β are assumed to be uniform. The normal pressure exerted by the thin film on the solid is denoted by p_N^s . Referring to the work of Rusanov [35], the mechanical equilibrium condition for the thin liquid film is expressed by the following equation:

$$p^\alpha - p^\beta = -\frac{2\gamma^f}{R_0} - (p_N^s - p^\alpha) \left(\frac{R}{R_0} \right)^2 \quad (6)$$

where R_0 and γ^f are the curvature radius and the surface tension of the gas–liquid interface, respectively, $R = R_0 + t$ designates the curvature radius of the pore surface (see Figure 2). It is noteworthy that the difference $\Pi = p_N^s - p^\alpha$ is generally defined as the (local) disjoining pressure. This equation will be

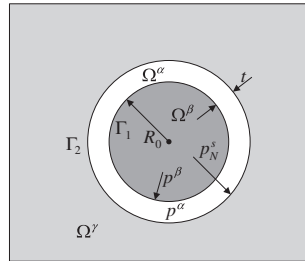


Figure 2. Thin spherical liquid film α with a finite constant thickness t between the solid phase γ and the liquid phase β .

used in the sequel to calculate the mechanical action p_N^s of the thin films adsorbed on the desaturated pore walls, which separate the solid phase from the gas one. It is important to recognize that this pressure integrates both the effects of the liquid–gas interface through γ^f and of the gas phase pressure p^β .

In this section, we have discussed the different types of water present in C-S-H. In what follows, we shall incorporate the effects of the adsorbed water and the capillary water in the mesopore spaces in describing the dependency of the creep behavior of HCP on the h_r .

3. MULTISCALE MODELLING

In this section, a multiscale homogenization method is now elaborated to account for the effects of the h_r on the macroscopic creep behavior of HCP whose microstructure is described in the previous section. We recall that the hydration chemical reactions are assumed complete so that aging phenomena (i.e. microstructure evolutions) are disregarded. For simplicity, only one type of C-S-H solid phase is considered, and its mechanical behavior is assumed to be linearly viscoelastic so as to be characterized by the following equation (see e.g. [37]):

$$\varepsilon\sigma(t) = \mathbb{S}_s(0) : \sigma(t) + \int_{-\infty}^t \frac{\partial \mathbb{S}_s(t-\tau)}{\partial t} : d\sigma(\tau) = \mathbb{S}_s(t) \circledast \sigma(t) \quad (7)$$

where $\mathbb{S}_s(t)$ is the fourth-order tensor function defining the creep behavior of the C-S-H solid phase and $\circledast$ is the Stieltjes convolution. In addition, HCP is modeled as having a two-scale microstructure: mesoscale and microscale structures (see Figure 1 at levels II and III). In the microscale microstructure, the solid inclusion phases are taken to be linearly elastic.

Via the Laplace–Carson (LC) transform, a linear viscoelastic problem can be transformed into a linear elastic problem in the LC space. The LC transform $\tilde{f}(s)$ of a function $f(t)$ is, in general, defined by (see e.g. [11,37]).

$$\tilde{f}(s) = s \int_0^\infty \exp(-st) f(t) dt. \quad (8)$$

Its inverse is formally given by the formula

$$f(t) = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} \frac{1}{s} \tilde{f}(s) \exp(st) ds. \quad (9)$$

Application of the LC transform to Equation (7) results in

$$\varepsilon(s) = \tilde{\mathbb{S}}_s(s) : \sigma(s) \quad (10)$$

which is identical to Hooke's law in the LC space. It is well known that the solution to a linearly elastic problem can be directly used as the solution to the associated linearly viscoelastic problem in the LC space. To obtain the solution to the latter in the time space, the inversion of the LC transform must be made. Except for some very simple cases, this inversion can be done only numerically.

3.1. Effects of the mesopore spaces in calcium silicate hydrate

As described in the previous section, at the mesoscale the C-S-H matrix consists of a solid phase in which mesopores are embedded. We consider that a part of the pores is filled with capillary water and the remaining part covered with a thin layer of adsorbed water (see Figure 1: Level II). A volume element Ω (see, Figure 4 (a)) is chosen so as to be representative of the C-S-H matrix. The subdomains occupied by the C-S-H solid phase, capillary mesopores and adsorbed water mesopores are designated by Ω^s , Ω^{cp} , and Ω^{aw} , respectively. Therefore the subdomain of the mesopores is given by

$$\Omega^{meso} = \Omega^{aw} \cup \Omega^{cp}, \quad (11)$$

and its volume fraction is defined by

$$\phi = \frac{|\Omega^{meso}|}{|\Omega|}. \quad (12)$$

The volume fractions of the capillary mesopores in mesopore space and in C-S-H are given by

$$\varphi_{cp} = \frac{|\Omega^{cp}|}{|\Omega^{meso}|}, \quad f_{cp} = \frac{|\Omega^{cp}|}{|\Omega|} = \varphi_{cp}\phi. \quad (13)$$

Similarly, for pore i containing an adsorbed water layer, the volume fractions φ_{aw_i} and f_{aw_i} are defined by

$$\varphi_{aw_i} = \frac{|\Omega^{aw_i}|}{|\Omega^{meso}|}, \quad f_{aw_i} = \frac{|\Omega^{aw_i}|}{|\Omega|} = \varphi_{aw_i}\phi. \quad (14)$$

As the adsorbed water layer exerts the pressure p_N^s on the pore surface, the latter is, from the mechanical point of view, equivalent to a pore saturated with a fluid of pressure p_N^s (see Figure 3, [38]). From Equation (6), we obtain p_N^{is} in pore i with radius R_{i0} :

$$p_N^{is} = \frac{1}{(1 - \delta_i)^2} \left[-\frac{2\gamma}{R_{i0}} + \delta_i(\delta_i - 2)p^l \right] \quad (15)$$

where $\delta_i = t/R_{i0}$. In the LC space, the average stress in Ω is mathematically expressed by

$$\tilde{\Sigma} = (1 - \phi) \langle \tilde{\sigma} \rangle_s - \phi \left(\varphi_{cp} \tilde{p}^l + \sum_i^n \varphi_{aw_i} \tilde{p}_N^{is} \right) I \quad (16)$$

where n is the total number of the spherical mesopores, $\langle \cdot \rangle_s$ designates the volume average over the solid phase Ω_s :

$$\langle g \rangle_s = \frac{1}{|\Omega_s|} \int_{\Omega_s} g d\Omega_s \quad (17)$$

and I is the second-order identity tensor. Introducing the size distribution function $\psi(r)$ of the adsorbed water mesopores, we have

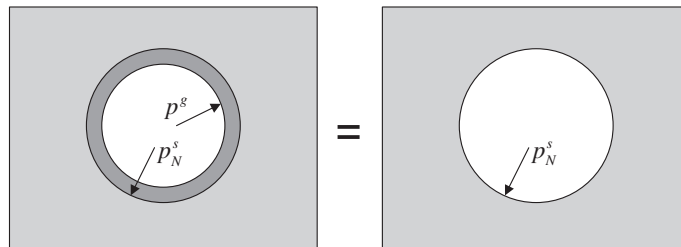


Figure 3. A replacement procedure based on the action of p_N^s because of the thin film on the pore wall.

$$\sum_i^n \varphi_{aw_i} \tilde{p}_N^{is} = \int_{R_{min}}^{R_{max}} \psi(r) \varphi_{aw}(r) \tilde{p}_N^s(r) dr, \quad (18)$$

where R_{min} and R_{max} are the minimal and maximal radii of the spherical mesopores with adsorbed water. We can obtain R_{min} via the Laplace equation $p_c = 2\gamma/(r_{min} - t)$, while assuming all pores with radius $r \leq r_{min}$ to be saturated. For the spherical pore shapes, we have

$$\varphi_{aw}(r) = \rho^t \frac{4\pi}{3} r^3 \quad (19)$$

where ρ^t is the pores density calculated by

$$\rho^t = \frac{\phi}{\int_0^{R_{max}} \frac{4\pi}{3} r^3 \psi(r) dr}. \quad (20)$$

Therefore, referring to the works [39,40] and using the equivalency for p_N^s , the problem in question can be decomposed into two sub-problems (see Figures 4b and 4c). The macroscopic constitutive law of C-S-H matrix in the LC space can be expressed by the following relationship involving h_r through the value of p^{CSH} :

$$\tilde{\Sigma} = \tilde{\mathbb{C}}_{CSH} : \tilde{E} - p^{CSH} \tilde{B}_{CSH}. \quad (21)$$

In the preceding formula,

$$p^{CSH} = \varphi_{cp} \tilde{p}^l + \sum_i^n \varphi_{aw_i} \tilde{p}_N^{is}, \quad (22)$$

$\tilde{B}_{CSH}$ is the effective Biot tensor of the C-S-H matrix in the LC space determined by (e.g., [40,41])

$$\tilde{B}_{CSH} = I : \left(\mathbb{I} - \tilde{\mathbb{C}}_s^{-1} : \tilde{\mathbb{C}}_{CSH} \right), \quad (23)$$

and $\tilde{\mathbb{C}}_{CSH}$ and $\tilde{\mathbb{C}}_s$ are the effective stiffness tensors of the C-S-H matrix containing the mesopore spaces as inclusions and of the C-S-H phase, respectively.

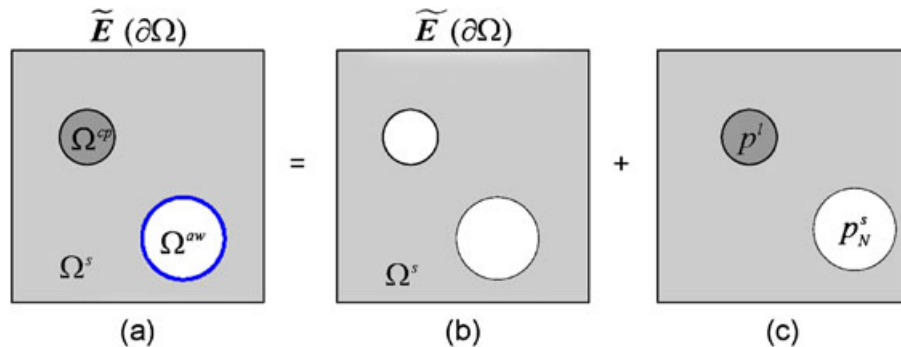


Figure 4. Simplified decomposition of the C-S-H problem involving the pore pressure p_l and the action of p_N^s because of the thin films.

Assuming that both $\tilde{\mathbb{C}}_s$ and $\tilde{\mathbb{C}}_{CSH}$ are isotropic, we have

$$\tilde{\mathbb{C}}_s = 3\tilde{k}_s\mathbb{J} + 2\tilde{\mu}_s\mathbb{K}, \quad (24)$$

$$\tilde{\mathbb{C}}_{CSH} = 3\tilde{k}_{CSH}\mathbb{J} + 2\tilde{\mu}_{CSH}\mathbb{K}. \quad (25)$$

where $\mathbb{J}$ and $\mathbb{K}$ designate the projection operators defined by

$$\mathbb{J} = \frac{1}{3}I \otimes I, \quad \mathbb{K} = \mathbb{I} - \mathbb{J}. \quad (26)$$

Using the well-known MT scheme (e.g., [42,43]), $\tilde{k}_{CSH}$ and $\tilde{\mu}_{CSH}$ are provided by

$$\tilde{k}_{CSH} = \tilde{k}_s \left[1 - \phi \frac{1}{1 - (1 - \phi)\tilde{\alpha}_s} \right], \quad (27a)$$

$$\tilde{\mu}_{CSH} = \tilde{\mu}_s \left[1 - \phi \frac{1}{1 - (1 - \phi)\tilde{\beta}_s} \right] \quad (27b)$$

with

$$\tilde{\alpha}_s = \frac{3\tilde{k}_s}{3\tilde{k}_s + 4\tilde{\mu}_s} \quad \text{and} \quad \tilde{\beta}_s = \frac{6}{5} \frac{\tilde{k}_s + 2\tilde{\mu}_s}{3\tilde{k}_s + 4\tilde{\mu}_s}. \quad (28)$$

The substitution of Equation (25) into Equation (23) yields

$$\tilde{B}_{CSH} = \tilde{b}_{CSH}I, \quad \tilde{b}_{CSH} = \left(1 - \frac{\tilde{k}_{CSH}}{\tilde{k}_s} \right). \quad (29)$$

3.2. Macroscopic creep properties of hardened cement pastes

In this section, we focus on the homogenization of HCP. For this purpose, HCP is considered as a multiphase poroelastic material consisting of a C-S-H matrix in which the four types of inclusions are distributed (see Figure 5(a)). The volume fractions of the CH crystals $\mathcal{Q}^{CH}$, CA $\bar{S}$ H $\mathcal{Q}^{CA\bar{S}H}$, UC $\mathcal{Q}^{UC}$, capillary pores $\mathcal{Q}^{CP}$, and C-S-H matrix are denoted by f^{CH} , $f^{CA\bar{S}H}$, f^{UC} , f^{CP} , and f^{CSH} , respectively. At this scale, the capillary pores are divided into those $\mathcal{Q}^l$ filled with water and those $\mathcal{Q}^v$ being empty. Because of the size of the capillary pores, we neglect the adsorbed water layers. The volume fraction of the capillary pores filled with water is denoted by f^l and that of the empty pores is noted by f^v . Therefore,

$$f^{CP} = f^l + f^v. \quad (30)$$

In the LC space, linear viscoelasticity acts as linear elasticity. Consequently, the principle of superposition permits to classically decompose the problem with the loading $(\tilde{E}, (-p^lI, -p^{CSH}\tilde{B}_{CSH}))$ applied on the poroelastic material into two sub-problems with the loadings $(\tilde{E}, (0, 0))$ and $(0, (-p^lI, -p^{CSH}\tilde{B}_{CSH}))$ (see Figure 5). The corresponding local strain and stress fields

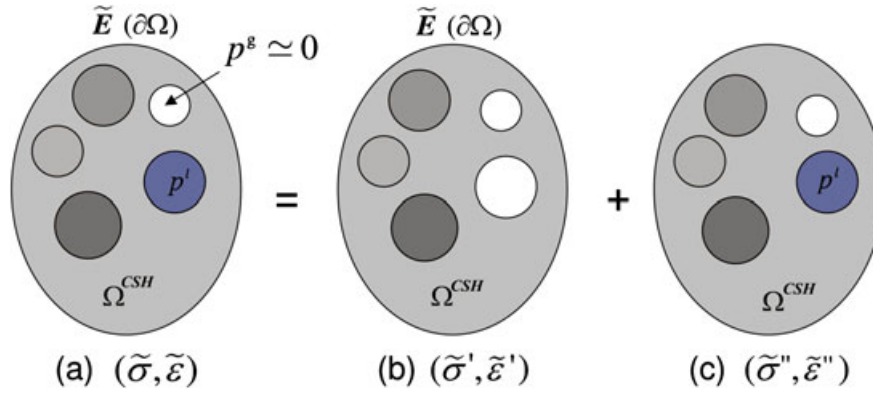


Figure 5. Decomposition of the problem of HCP: (a) overall loading $(\tilde{E}, (-p^l I, -p^{CSH} \tilde{B}_{CSH}))$ on the poroelastic material; (b) loading $(\tilde{E}, (0, 0))$; (c) loading $(0, (-p^l I, -p^{CSH} \tilde{B}_{CSH}))$.

are denoted by $(\tilde{\sigma}', \tilde{\varepsilon}')$ and $(\tilde{\sigma}'', \tilde{\varepsilon}'')$. Thus, we can write the following relations for the strain and stress averages over the representative volume element Ω_{HCP} of HCP:

$$\begin{aligned} \langle \tilde{\varepsilon} \rangle_{\Omega_{HCP}} &= \tilde{E}, & \langle \tilde{\varepsilon}'' \rangle_{\Omega_{HCP}} &= 0, \\ \langle \tilde{\sigma}'' \rangle_{\Omega_{HCP}} &= -p^{mix} \tilde{B}_{HCP} \end{aligned} \quad (31)$$

where $p^{mix} \tilde{B}_{HCP}$ is determined in the following. Using the principle of superposition, the macroscopic constitutive law in the LC space can be expressed by (see, e.g., [40,41])

$$\tilde{\Sigma} = \tilde{\mathbb{C}}_{HCP} : \tilde{E} - p^{mix} \tilde{B}_{HCP} \quad (32)$$

where $\tilde{\mathbb{C}}_{HCP}$ is the elastic stiffness tensor of HCP containing the solid phase and capillary pores. Note that the term $p^{mix} \tilde{B}_{HCP}$ may be expressed equivalently in the present study as:

$$p^{mix} \tilde{B}_{HCP} = p^{eff} I \quad (33)$$

because the material is isotropic, with p^{eff} the macroscopic effective pressure. In order to determine this term, Hill's lemma is applied to the equilibrated stress field $\tilde{\sigma}''$ and to the strain field $\tilde{\varepsilon}$, so that

$$\langle \tilde{\sigma}'' : \tilde{\varepsilon} \rangle_{\Omega_{HCP}} = \langle \tilde{\sigma}'' \rangle_{\Omega_{HCP}} : \langle \tilde{\varepsilon} \rangle_{\Omega_{HCP}} = -p^{mix} \tilde{B}_{HCP} : \tilde{E}. \quad (34)$$

By introducing the following definition of the pre-stress

$$\tilde{\sigma}^l = \begin{cases} 0 & \text{in } \Omega - \Omega_l \\ -p^l I & \text{in } \Omega_l \end{cases}, \quad \tilde{\sigma}^{CSH} = \begin{cases} 0 & \text{in } \Omega - \Omega_{CSH} \\ -p^{CSH} \tilde{B}_{CSH} & \text{in } \Omega_{CSH} \end{cases}, \quad (35)$$

we obtain the local stress tensor in the form:

$$\tilde{\sigma}'' = \tilde{\mathbb{C}}_s : \tilde{\varepsilon}'' + \tilde{\sigma}^l + \tilde{\sigma}^{CSH}. \quad (36)$$

where $\tilde{\mathbb{C}}_s$ designates the local stiffness tensor of the solid phase, which is either the C-S-H matrix, UC, CH, or CASH according as the material point is considered. Therefore,

$$\begin{aligned} \langle \tilde{\sigma}'' : \tilde{\varepsilon}' \rangle_{\Omega_{HCP}} &= \langle \tilde{\varepsilon}'' : \tilde{\mathbb{C}}_s : \tilde{\varepsilon}' \rangle_{\Omega_{HCP}} + f^l \langle \tilde{\sigma}^l : \tilde{\varepsilon}' \rangle_{\Omega_l} \\ &\quad + f^{CSH} \langle \tilde{\sigma}^{CSH} : \tilde{\varepsilon}' \rangle_{\Omega_{CSH}}. \end{aligned} \quad (37)$$

Recalling Equation (31) and introducing the concentration tensor $\tilde{\mathbb{A}}$ defined by

$$\tilde{\varepsilon} = \tilde{\mathbb{A}} : \tilde{\mathbb{E}}, \quad (38)$$

we can write

$$\langle \tilde{\sigma}'' : \tilde{\varepsilon}' \rangle_{\Omega_{HCP}} = \left(-f^l \mathbb{I} : \tilde{\mathbb{A}}_{\Omega_l} - f^{CSH} p^{CSH} \tilde{\mathbb{B}}_{CSH} : \tilde{\mathbb{A}}_{\Omega_{CSH}} \right) : \tilde{\mathbb{E}} \quad (39)$$

where $\tilde{\mathbb{A}}_{\Omega_l}$ and $\tilde{\mathbb{A}}_{\Omega_{CSH}}$ denote the relations between the macroscopic strain and the local strains in Ω_l and Ω_{CSH} , respectively. Then, considering the fact that the two Equations (34) and (39) hold for any $\tilde{\mathbb{E}}$, we have

$$p^{mix} \tilde{\mathbb{B}}_{HCP} = f^l p^l \mathbb{I} : \tilde{\mathbb{A}}_{\Omega_l} + f^{CSH} p^{CSH} \tilde{\mathbb{B}}_{CSH} : \tilde{\mathbb{A}}_{\Omega_{CSH}}. \quad (40)$$

Note that, for $p^{CSH} = p^l$, we recover the results for classical poroelastic materials. However, in the present paper, taking into account the effects of the adsorbed water leads to $p^{CSH} \neq p^l$, and we then have a more general formulation for the poroelastic problem.

Following the Mori–Tanaka method [42] and [44], the concentration tensor $\tilde{\mathbb{A}}_r^{MT}$ corresponding to phase r reads

$$\begin{aligned} \tilde{\mathbb{A}}_r^{MT} &= \left[\mathbb{I} + \tilde{\mathbb{P}}_r^{CSH} : \left(\tilde{\mathbb{C}}_r - \tilde{\mathbb{C}}_{CSH} \right) \right]^{-1} \\ &: \left\langle \left[\mathbb{I} + \tilde{\mathbb{P}}_i^{CSH} : \left(\tilde{\mathbb{C}}_i - \tilde{\mathbb{C}}_{CSH} \right) \right]^{-1} \right\rangle_{\Omega_{HCP}}^{-1} \end{aligned} \quad (41)$$

and the effective (homogenized) elastic tensor $\tilde{\mathbb{C}}_{HCP}^{MT}$:

$$\begin{aligned} \tilde{\mathbb{C}}_{HCP}^{MT} &= \left\langle \tilde{\mathbb{C}}_i : \left[\mathbb{I} + \tilde{\mathbb{P}}_i^{CSH} : \left(\tilde{\mathbb{C}}_i - \tilde{\mathbb{C}}_{CSH} \right) \right]^{-1} \right\rangle_{\Omega_{HCP}} \\ &: \left\langle \left[\mathbb{I} + \tilde{\mathbb{P}}_i^{CSH} : \left(\tilde{\mathbb{C}}_i - \tilde{\mathbb{C}}_{CSH} \right) \right]^{-1} \right\rangle_{\Omega_{HCP}}^{-1} \end{aligned} \quad (42)$$

where $\tilde{\mathbb{P}}_r^{CSH}$ is the Hill tensor of inclusion phase r defined by $\tilde{\mathbb{P}}_r^{CSH} = \mathbb{S}_r^{CSH} : \mathbb{C}_{CSH}^{-1}$ with $\mathbb{S}_r^{CSH}$ being the Eshelby tensor for an inclusion of phase r embedded in the C-S-H matrix, and $\langle \cdot \rangle_{\Omega_{HCP}}$ is the volume average over the domain Ω_{HCP} , given, for example for a field A , by

$$\langle A \rangle_{\Omega_{HCP}} = \sum_i^n f_i \langle A \rangle_i = \sum_i^n f_i A_i. \quad (43)$$

Above, f_i are the phase volume fractions and A_i the phase volume averages of A .

From the Equation (41), we have

$$\begin{aligned} \tilde{\mathbb{A}}_{\Omega_i}^{MT} &= \left[\mathbb{I} - \tilde{\mathbb{P}}_{\Omega_i}^{CSH} : \tilde{\mathbb{C}}_{CSH} \right]^{-1} \\ &: \left\langle \left[\mathbb{I} + \tilde{\mathbb{P}}_i^{CSH} : \left(\tilde{\mathbb{C}}_i - \tilde{\mathbb{C}}_{CSH} \right) \right]^{-1} \right\rangle_{\Omega_{HCP}}^{-1} \end{aligned} \quad (44)$$

and

$$\tilde{\mathbb{A}}_{\Omega_{CSH}}^{MT} = \left\langle \left[\mathbb{I} + \tilde{\mathbb{P}}_i^{CSH} : \left(\tilde{\mathbb{C}}_i - \tilde{\mathbb{C}}_{CSH} \right) \right]^{-1} \right\rangle_{\Omega_{HCP}}^{-1}. \quad (45)$$

The substitution of Equations (42), (44), and (45) into Equation (32) gives the effective properties of HCP in the LC space.

In the isotropic case and for spherical inclusions, $\tilde{\mathbb{P}}_r^{CSH}$ has the expression

$$\tilde{\mathbb{P}}_r^{CSH} = \frac{\tilde{\alpha}_{CSH}}{3\tilde{k}_{CSH}} \mathbb{J} + \frac{\tilde{\beta}_{CSH}}{2\tilde{\mu}_{CSH}} \mathbb{K} \quad (46)$$

with

$$\begin{aligned} \tilde{\alpha}_{CSH} &= \frac{3\tilde{k}_{CSH}}{3\tilde{k}_{CSH} + 4\tilde{\mu}_{CSH}}, \\ \tilde{\beta}_{CSH} &= \frac{6}{5} \frac{\tilde{k}_{CSH} + 2\tilde{\mu}_{CSH}}{3\tilde{k}_{CSH} + 4\tilde{\mu}_{CSH}}. \end{aligned} \quad (47)$$

Therefore, the concentration tensors $\tilde{\mathbb{A}}_{\Omega_{CSH}}^{MT}$ and $\tilde{\mathbb{A}}_{\Omega_i}^{MT}$ are given by

$$\begin{aligned} \tilde{\mathbb{A}}_{\Omega_{CSH}}^{MT} &= \left\langle 1 - \frac{(\tilde{k}_i - \tilde{k}_{CSH})}{\tilde{k}_i + \frac{4}{3}\tilde{\mu}_{CSH}} \right\rangle_{\Omega_{CSH}}^{-1} \mathbb{J} \\ &+ \left\langle 1 - \frac{\tilde{\mu}_i - \tilde{\mu}_{CSH}}{\tilde{\mu}_i + \tilde{H}_{CSH}} \right\rangle_{\Omega_{CSH}}^{-1} \mathbb{K} \end{aligned} \quad (48)$$

and

$$\begin{aligned} \tilde{\mathbb{A}}_{\Omega_i}^{MT} &= \left(1 + \frac{3\tilde{k}_{CSH}}{4\tilde{\mu}_{CSH}} \right) \left\langle 1 - \frac{(\tilde{k}_i - \tilde{k}_{CSH})}{\tilde{k}_i + \frac{4}{3}\tilde{\mu}_{CSH}} \right\rangle_{\Omega_{HCP}}^{-1} \mathbb{J} \\ &+ \left(1 + \frac{(\tilde{k}_{CSH} + 2\tilde{\mu}_{CSH})}{\frac{3}{2}\tilde{k}_{CSH} + \frac{4}{3}\tilde{\mu}_{CSH}} \right) \left\langle 1 - \frac{\tilde{\mu}_i - \tilde{\mu}_{CSH}}{\tilde{\mu}_i + \tilde{H}_{CSH}} \right\rangle_{\Omega_{HCP}}^{-1} \mathbb{K}. \end{aligned} \quad (49)$$

with $\tilde{H}_{CSH} = \tilde{\mu}_{CSH} \frac{3\tilde{k}_{CSH} + 4\tilde{\mu}_{CSH}}{\tilde{k}_{CSH} + 2\tilde{\mu}_{CSH}}$. The effective stiffness tensor $\tilde{\mathbb{C}}_{HCP}^{MT}$ is given by

$$\tilde{\mathbb{C}}_{HCP}^{MT} = 3\tilde{k}_{HCP}^{MT} \mathbb{J} + 2\tilde{\mu}_{HCP}^{MT} \mathbb{K} \quad (50)$$

where

$$\tilde{k}_{HCP}^{MT} = \left\langle \tilde{k}_{CSH} + \frac{4}{3} \tilde{\mu}_{CSH} \frac{\tilde{k}_i - \tilde{k}_{CSH}}{\tilde{k}_i + \frac{4}{3} \tilde{\mu}_{CSH}} \right\rangle_{\Omega_{HCP}} \left\langle 1 - \frac{(\tilde{k}_i - \tilde{k}_{CSH})}{\tilde{k}_i + \frac{4}{3} \tilde{\mu}_{CSH}} \right\rangle_{\Omega_{HCP}}^{-1}, \quad (51)$$

$$\tilde{\mu}_{HCP}^{MT} = \left\langle \tilde{\mu}_{CSH} + \tilde{H}_{CSH} \frac{\tilde{\mu}_i - \tilde{\mu}_{CSH}}{\tilde{\mu}_i + \tilde{H}_{CSH}} \right\rangle_{\Omega_{HCP}} \left\langle 1 - \frac{\tilde{\mu}_i - \tilde{\mu}_{CSH}}{\tilde{\mu}_i + \tilde{H}_{CSH}} \right\rangle_{\Omega_{HCP}}^{-1}. \quad (52)$$

Therefore, we obtain

$$\begin{aligned} p^{mix} \tilde{B}_{HCP} &= f^l p^l \left(1 + \frac{3 \tilde{k}_{CSH}}{4 \tilde{\mu}_{CSH}} \right) \left\langle 1 - \frac{(\tilde{k}_i - \tilde{k}_{CSH})}{\tilde{k}_i + \frac{4}{3} \tilde{\mu}_{CSH}} \right\rangle_{\Omega_{HCP}}^{-1} I \\ &+ f^{CSH} p^{CSH} \left(1 - \frac{\tilde{k}_{CSH}}{\tilde{k}_s} \right) \left\langle 1 - \frac{(\tilde{k}_i - \tilde{k}_{CSH})}{\tilde{k}_i + \frac{4}{3} \tilde{\mu}_{CSH}} \right\rangle_{\Omega_{HCP}}^{-1} I. \end{aligned} \quad (53)$$

This relation directly provides the expression of the equivalent effective pressure p^{eff} introduced in Equation (33) and acting at the macroscopic scale in the presently considered case of isotropic material.

So far, we have derived the expressions for estimating the effective properties of HCP in the LC space. To obtain the associated properties in the time domain, we need to invert the LC transform. In this work, we adopt the method proposed by Donolato [16], which has the main advantage of providing an analytical, although approximate, expression of the inverse of any function defined in the LC space. Introducing the variable $u = 1/\xi$, the LC transform (8) becomes

$$g(u) = \tilde{f}(1/u) = 1/u \int_0^\infty \exp\left(-\frac{t}{u}\right) f(t) dt. \quad (54)$$

Following [16] (see also [45]), an approximate function $f_n(t)$ of the inverse of the LC transform $\tilde{f}(s)$ is obtained by

$$f(t) \approx f_n(t) = \frac{1}{n!} u^{n+1} \frac{du^n [u^n g(u)]}{du^n} \Big|_{u=\frac{t}{n+1}}. \quad (55)$$

In this equation, the integer n characterizes the order of derivation of the function $g(u)$ and also defines the level of precision of the approximation: $f_n(t)$ will be closer to the exact solution $f(t)$ for greater values of n . Practical examples will be provided and analyzed in section 5.

4. CREEPING OF HARDENED CEMENT PASTES AT DIFFERENT RELATIVE HUMIDITY VALUES

In this section, we apply the model developed earlier to estimate the time-dependent behavior of an HCP at different constant values of the h_r . A cylindrical specimen of diameter equal to 8 mm and height equal to 20 mm is first subjected to drying at a given constant external h_r until the mass water

balance is achieved. It then undergoes a constant uniaxial compressive stress of -20 MPa. The corresponding experimental results are reported in [8].

We propose to model the C-S-H, for simplicity, as a linearly viscoelastic material with a time-independent Poisson's ratio $\nu_0 = 0.25$ as in [46] and with a time-dependent shear modulus represented by a generalized Maxwell model comprising five elements:

$$\mu(t) = \mu_0 + \sum_{i=1}^5 \mu_i \exp\left(-\frac{t}{\tau_{i0}}\right) \quad (56)$$

where μ_i and τ_{i0} are the relaxation constants and the retardation times, respectively. The porosity of the C-S-H is taken to be $\phi = 25\%$. The hypothesis that the Poisson's ratio of the C-S-H is time-independent implies that the following relation holds:

$$\tilde{k}_s = \psi_0 \tilde{\mu}_s \quad \text{with} \quad \psi_0 = \frac{2(1 + \nu_0)}{3(1 - 2\nu_0)}. \quad (57)$$

The substitution of this equation into Equation (28) leads to

$$\tilde{\alpha}_s = \frac{1 + \nu_0}{3(1 - \nu_0)} \quad \text{and} \quad \tilde{\beta}_s = \frac{2(4 - 5\nu_0)}{15(1 - \nu_0)}. \quad (58)$$

Then, we have

$$\tilde{k}_{CSH} = \psi_k \psi_0 \tilde{\mu}_s, \quad \tilde{\mu}_{CSH} = \psi_\mu \tilde{\mu}_s \quad (59)$$

with

$$\psi_k = \left[1 - \frac{\phi}{1 - (1 - \phi) \left(\frac{1 + \nu_0}{3(1 - \nu_0)} \right)} \right], \quad \psi_\mu = \left[1 - \frac{\phi}{1 - (1 - \phi) \left(\frac{2(4 - 5\nu_0)}{15(1 - \nu_0)} \right)} \right]. \quad (60)$$

Introducing Equation (59) into Equation (29) yields

$$\tilde{b}_{CSH} = 1 - \psi_k \quad (61)$$

Equations (59) and (61) describe the properties of the C-S-H solid phase in the LC space. The composition of HCP is indicated in Table I whereas the mechanical properties of the elastic inclusions are listed in Table II.

In general, cement-based materials exhibit several pore size classes (e.g. [47,48]). The mesopores size distribution in C-S-H is not precisely known in general, principally because it is extremely difficult to characterize experimentally. In a first attempt and without loss of generality, we then assume that the

Table I. Estimated volume fractions of the different phases in HCP [46].

Phase	Volume fraction
CSH	$f^{CSH} = 0.51$
CASH	$f^{CASH} = 0$
CH	$f^{CH} = 0.17$
CP	$f^{CP} = 0.11$
UC	$f^{UC} = 0.21$

Table II. Mechanical properties of the elastic inclusions in HCP [46].

Phase	k (GPa)	μ (GPa)
CH	40	16
UC	105.2	44.8

radii of the mesopores are uniform and equal to 1.6 nm, corresponding to a typical averaged value of C-S-H porosity [47]. This assumption also allows to simplifying the analytical expressions of the model. For $r_{min} = 1.6$ nm, Equation (2) gives $h_r = 0.51$. Then, for $h_r < 0.51$, the mesopores in C-S-H are assumed to be covered with a thin layer of adsorbed water whose thickness can be estimated by Equation (1), whereas for $h_r \geq 0.51$, the C-S-H matrix is completely saturated.

In the case of the uniaxial constant loading under consideration, the behavior of HCP is described by:

$$\tilde{E}_{yy} = \left(\frac{1}{9\tilde{k}_{HCP}^{MT}} + \frac{1}{3\tilde{\mu}_{HCP}^{MT}} \right) \tilde{\Sigma}_{yy} + \frac{\eta_{\Omega_{CSH}} \left[f^I p^I \left(1 + \frac{3}{4} \frac{\tilde{k}_{CSH}}{\tilde{\mu}_{CSH}} \right) + f^{CSH} p^{CSH} \tilde{b}_{CSH} \right]}{3\tilde{k}_{HCP}^{MT}}. \quad (62)$$

Using the Donolato method to perform the inversion of the LC transform (with $n = 3$) and the experimental results of Roelfstra [8] corresponding to $h_r = 100\%$, the constants μ_i and τ_{i0} in Equation (56) can be numerically identified, and their values are given in Table III.

With the parameters identified earlier, uniaxial compression tests are simulated for different values of h_r . Comparisons with experimental data are shown in Figure 6 for $h_r = 100\%$, 81%, and 44% in terms of the time evolution of the strain ϵ_{yy} corresponding to the loading direction. We observe that the model does not predict a satisfying response for $h_r \neq 100\%$ as the magnitude of the numerical strain ϵ_{yy} increases whereas it decreases according to the experimental results. This is probably because of the significant role of the interlayer water on the viscoelastic properties of C-S-H, which is not explicitly taken into account in the model. Indeed, as already mentioned, the physical phenomena appear much too complex to be completely captured through some simple and identifiable models. On the other hand, it also implies that the capillary pressures acting in the mesopores and macropores are not sufficient, according to the model, to explain the observed h_r dependency of the macroscopic behavior. One important consequence seems to be that the viscoelastic behavior of the C-S-H should evolve as a function of h_r . More precisely, it appears that this phase should progressively rigidify for decreasing h_r , which agrees with other approaches in the literature (see e.g. [7]). This also means, at first sight, that the viscoelastic behavior retained for the C-S-H phase is too simplistic.

In the sequel, we propose to modify the preceding model so as to more correctly reproduce the experimental data, by introducing a dependence of the C-S-H phase behavior upon h_r at the mesoscale. To conserve a global coherent approach, we decide to keep both the linear viscoelastic behavior for the C-S-H matrix (needed for applying the LC transform) and the framework of micromechanics. The basic idea is as follows: the desaturated mesopores in the C-S-H at level II (see Figure 1) are assumed to directly contribute to its apparent strengthening. To this aim, they are simply replaced by elastic inclusions, which are equivalent to considering that the adsorbed water layer rigidifies the surrounding C-S-H matrix, in addition to the action of the pressure p_N^s defined in Equation (15). It should be noticed that this approach would probably also be extended to level I involving the C-S-H matrix at the nanoscale. However, in the present study, this phase has been considered to simplify as homogeneous at this scale; as

Table III. Values of the coefficients for the generalized Maxwell model with 5 elements.

Element	0	1	2	3	4	5
μ (GPa)	1.398	2.520	2.084	1.999	1.104	0.973
τ_{i0} (Days)		0.05	0.5	5.0	50.0	500.0

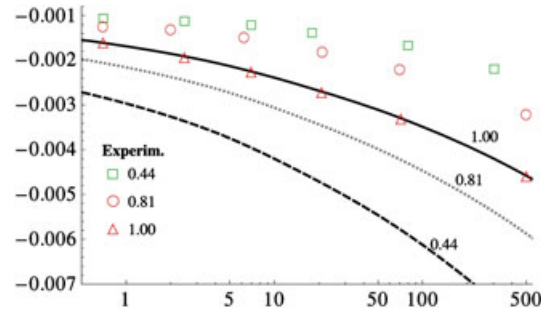


Figure 6. Numerical time evolution of the strain ϵ_{yy} without correction of the retardation times and experimental data of [8] for a uniaxial compression creep test with $\Sigma_{yy} = -20$ MPa.

a consequence, the model should not be used for h_r lower than 0.3–0.4, corresponding to the upper limit where C-S-H desaturation starts. Again, we leave these complex aspects related to the nanoscopic behavior of C-S-H for subsequent investigations. The proposed improvements necessitate some modifications in the model, in particular, in the Equations (27a), (27b) and Equation (23) together with (29), which should include the effects of the equivalent elastic inclusions. Rewriting the term $p^{CSH} \tilde{B}_{CSH}$ appearing in Equation (21) in the same way as in Equation (40), that is, involving the strain localization tensors $\tilde{\mathbb{A}}_{\Omega_i}$ of the inclusions i subjected to the microscopic homogeneous stresses p_N^s , we have:

$$p^{CSH} \tilde{B}_{CSH} = \varphi_{cp} \tilde{p}^l I : \tilde{\mathbb{A}}_{\Omega_{cp}} + \sum_i^n \varphi_{awi} \tilde{p}_N^{is} I : \tilde{\mathbb{A}}_{\Omega_i}, \quad (63)$$

where the expressions of $\tilde{\mathbb{A}}_{\Omega_{cp}}$ and $\tilde{\mathbb{A}}_{\Omega_i}$ are given in Equation (41) provided the subscript *CSH* is replaced by *s*. The mechanical properties k_{ei} and μ_{ei} of the equivalent elastic inclusions appear in both tensors in the denominator and also in the numerator for $\tilde{\mathbb{A}}_{\Omega_i}$. Likewise, the bulk and shear moduli defined in Equations (59)–(60) are modified following the Mori–Tanaka scheme as:

$$\begin{aligned} \tilde{k}_{CSH} &= \tilde{\mu}_s [\psi_0 (1 - \phi_l) + 4\phi_{ei} \alpha_{ei}] [1 + 3/4 \phi_l \psi_0 - 3\phi_{ei} \alpha_{ei}]^{-1}, \\ \tilde{\mu}_{CSH} &= \tilde{\mu}_s [1 - \phi_l + (8 + 9\psi_0) \phi_{ei} \beta_{ei}] [1 + \phi_l + 6(2 + \psi_0) \phi_{ei} \beta_{ei}]^{-1}. \end{aligned} \quad (64)$$

with

$$\alpha_{ei} = \frac{k_{ei} - \psi_0 \tilde{\mu}_s}{3k_{ei} + 4\tilde{\mu}_s}, \quad \beta_{ei} = \frac{\mu_{ei} - \tilde{\mu}_s}{6(2 + \psi_0) \mu_{ei} + (8 + 9\psi_0) \tilde{\mu}_s} \quad (65)$$

where ϕ_{ei} is the volume fraction of the emptied mesopores replaced by elastic inclusions. The results of

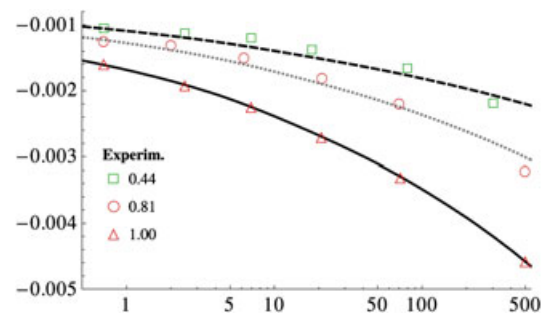


Figure 7. Numerical time evolution of the strain ϵ_{yy} with correction of the retardation times and experimental data of [8] for a uniaxial compression creep test with $\Sigma_{yy} = -20$ MPa.

simulations obtained with the new approach are presented for the different values of h_r on Figure 7, with the following adjusted values of properties for the equivalent elastic inclusions: $k_{ei}=50GPa$ and $\mu_{ei}=30GPa$. Clearly enough, the agreement between simulations and experimental results is now quite good, indicating that the modifications introduced for strengthening the C-S-H phase when h_r decreases are relevant.

5. NUMERICAL STUDY

5.1. Numerical implementation of the model in a FE code

The model elaborated and identified foregoingly has been implemented in the FE code Cast3M developed at CEA. As already mentioned, one of its main characteristics and interests is that the macroscopic time-dependent mechanical parameters are explicitly determined in the time space with the help of the Donolato method (see Equation (55)). The implementation of a new material behavior in an FE code generally requires specifying the stress tensor and, more precisely, its increment at the current time step. In this section, we consider only mechanical loadings (i.e. $p^{mix}=0$) for simplicity, although more general cases with $h_r \neq 100\%$ pose no problem. Under this condition, the stress–strain relation in the LC space can be expressed by

$$\tilde{\sigma}(s) = \tilde{R}(s)\tilde{\epsilon}(s). \quad (66)$$

The inverse transform of the preceding equation yields the time-dependent stress tensor:

$$\sigma(t) = L^{-1}[\tilde{R}(s)\tilde{\epsilon}(s)] \quad (67)$$

which is delivered by using Equation (54) with

$$g(u) = \tilde{R}(1/u)\tilde{\epsilon}(1/u) = r(u)\epsilon(u). \quad (68)$$

Note that the preceding relations involve different orders of derivation of the functions $r(u)$ and $\epsilon(u)$ through Equation (54). Moreover, whereas $\tilde{R}(s)$ is known explicitly, the tensor $\tilde{\epsilon}(s)$ has to be calculated as it integrates the history of the local loading.

Classically, the FE numerical solution of a time-dependent problem entails the discretization of both space and time domains. Then, $\tilde{\epsilon}(s)$ must be expressed for all time steps, provided that its values are known at the beginning and the end of the current time step i spanning from t_i to t_{i+1} . In fact, the strain at the beginning of the current time step is the converged FE solution of the previous one, and at the end, it results from the estimation of the nonlinear solution procedure of the FE code, based on the modified Newton–Raphson method. To express $\tilde{\epsilon}(s)$, we propose to consider two linearization methods in $[t_i, t_{i+1}]$: the first one consists in constant stepwise strains (i.e., constant for each time step), which is the most simple approximation of the real loading strains (see the left picture of Figure 8). Applying the LC transform, the general formulation of the stepwise strains $\tilde{\epsilon}(s)$ at the current time step i is:

$$\begin{aligned} \tilde{\epsilon}_i(s) &= \epsilon_0 s \int_0^{t_1} \exp(-st)dt + \cdots + \epsilon_{i-1} s \int_{t_{i-1}}^{t_i} \exp(-st)dt + \epsilon_i s \int_{t_i}^{\infty} \exp(-st)dt \\ &= \epsilon_0 + \Delta \epsilon_1 \exp(-st_1) + \cdots + \epsilon_{i-1} \exp(-st_{i-1}) + \Delta \epsilon_i \exp(-st_i) \end{aligned} \quad (69)$$

where ϵ_0 is the initial strain at t_0 and $\Delta \epsilon_i$ is the strain increment at the time step i . This yields a particularly simple expression of $\epsilon_i(u)$ as follows:

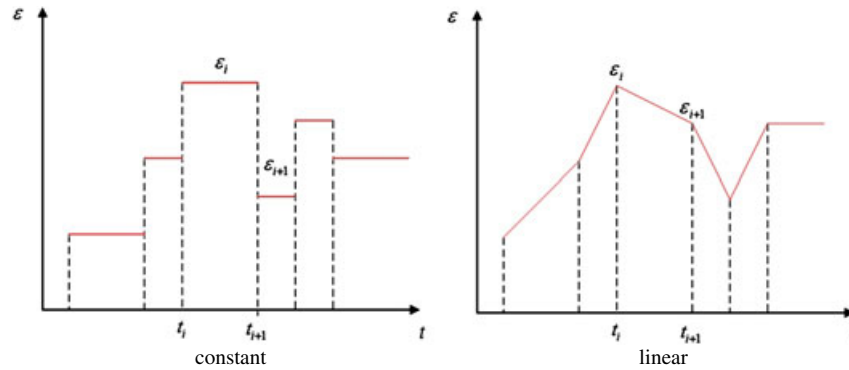


Figure 8. Illustration of the strain discretization methods: constant stepwise (left) and linear stepwise (right).

$$\epsilon_i(u) = \epsilon_0 + \Delta \epsilon_1 \exp\left(-\frac{t_1}{u}\right) + \cdots + \Delta \epsilon_{i-1} \exp\left(-\frac{t_{i-1}}{u}\right) + \Delta \epsilon_i \exp\left(-\frac{t_i}{u}\right). \quad (70)$$

The second linearization method considers a linear stepwise strain approximation, as depicted in the right picture of Figure 8. In this case, at the time step $[t_i, t_{i+1}]$, we have:

$$\epsilon(t) = \frac{\epsilon_{i+1} - \epsilon_i}{t_{i+1} - t_i} (t - t_i) + \epsilon_i. \quad (71)$$

Thus, the LC transform can be expressed as:

$$\tilde{\epsilon}_i(s) = \epsilon_0 + \frac{1}{s} \left[\frac{\Delta \epsilon_1}{\Delta t_1} (1 - \exp(-st_1)) + \frac{\Delta \epsilon_2}{\Delta t_2} (\exp(-st_1) - \exp(-st_2)) + \cdots + \frac{\Delta \epsilon_i}{\Delta t_i} (\exp(-st_{i-1}) - \exp(-st_i)) \right] \quad (72)$$

and the corresponding expression of $\epsilon_i(u)$ takes the form

$$\epsilon_i(u) = \epsilon_0 + u \left[\frac{\Delta \epsilon_1}{\Delta t_1} \left(1 - \exp\left(-\frac{t_1}{u}\right)\right) + \frac{\Delta \epsilon_2}{\Delta t_2} \left(\exp\left(-\frac{t_1}{u}\right) - \exp\left(-\frac{t_2}{u}\right)\right) + \cdots + \frac{\Delta \epsilon_i}{\Delta t_i} \left(\exp\left(-\frac{t_{i-1}}{u}\right) - \exp\left(-\frac{t_i}{u}\right)\right) \right]. \quad (73)$$

Again, it is important to remark that with these expressions, the (approximate) time-dependent stress formulation is provided explicitly by Equation (55) and can be implemented directly in the FE code Cast3M. This is clearly an advantage of the method adopted here: from a rather complex model developed in the LC space, the Donolato method allows us to obtain an explicit, although approximate expression of the stress tensor.

5.2. Numerical analysis of the model response

In this subsection, we analyze, although very simple examples, the influences of the strains discretization as presented earlier and of the time steps on the model response. The effects of the approximation caused by the finite value of the order of derivation n of the Donolato method (see Equation (55)) are also discussed. We then consider the 2D plane deformation problem of a long cylinder of square cross section. The boundary conditions are such that the problem can be considered as

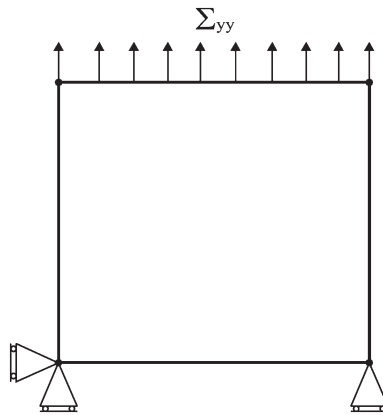


Figure 9. Schematic of the 2D structure subjected to a uniaxial creep loading.

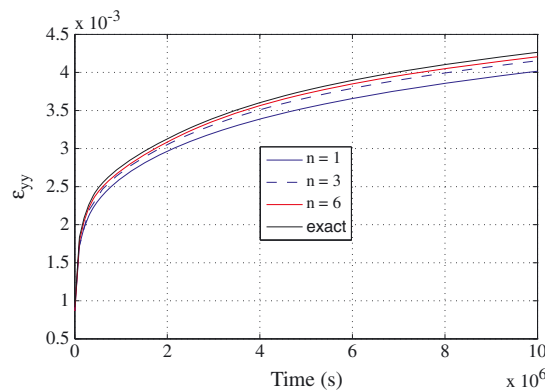


Figure 10. Time evolution of the strain ϵ_{yy} obtained analytically with various values of the order of derivation n in the Donolato method for a creep test with $\Sigma_{yy}=20$ MPa and comparison with the exact solution.

one-dimensional (i.e. the displacements of the edge opposite to the loaded one is prescribed to be 0 in the loading direction; see Figure 9). The properties of the materials are defined in section 4.

Figure 10 presents the curves depicting the time evolutions of the macroscopic strain ϵ_{yy} obtained analytically for different values of the derivation order n of the Donolato method, in the case of creep loading with $\Sigma_{yy}=20$ MPa and fully saturated material (i.e. $p^{mix}=0$). The exact solution is also plotted on this figure. We observe some differences between the curves, in particular when n is low. Increasing n leads to a more precise response. The value $n=3$ retained for this study appears to be a reasonable compromise between the approximate solution accuracy and the formulation complexity.

Figure 11 shows the FE time evolutions of the strain ϵ_{yy} obtained for the two cases of constant stepwise and linear stepwise strains as described earlier and with the creep loading $\Sigma_{yy}=20$ MPa and $p^{mix}=0$. We remark that these two FE curves are very close to the analytical solution, as shown in the same figure. This indicates that the two strain discretization methods provide very accurate approximations of the exact solution. In the following, we retain the constant stepwise strain method for the computations, because it is simpler and gives sufficiently precise results.

Finally, Figure 12 illustrates the influence of the time step on the numerical response in the case of the same creep loading as in Figures 10 and 11. We observe that the differences between the three curves obtained with $\Delta t=10^3$, $\Delta t=10^4$, and $\Delta t=10^5$ s are very small (Figure 12 top). They appear more clearly in the second picture of Figure 12, which represents a narrow zoomed region of the curves. From these results, it is seen that a time step around $\Delta t=10^4$ s is sufficient to getting a good accuracy in the present simulation case. Obviously, this value of time step cannot be viewed as a universal one. The maximal time step to be used for the calculations depends more generally on the whole

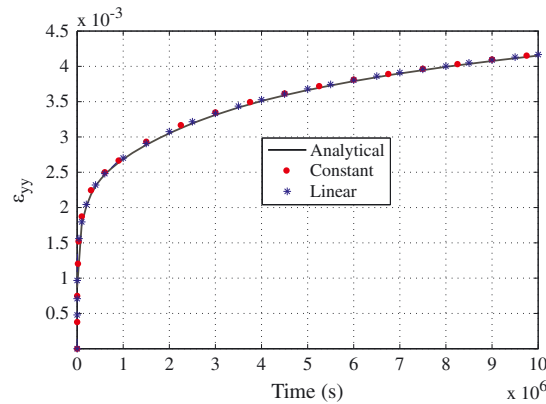


Figure 11. FE numerical evolutions of the strain ϵ_{yy} with constant and linear stepwise strain discretization and analytical curve ($n=3$) for a creep test with $\Sigma_{yy}=20$ MPa.

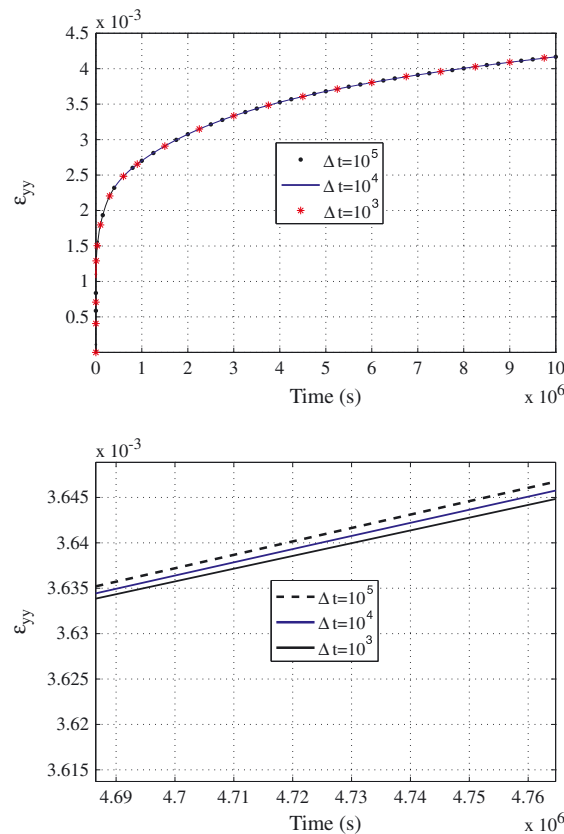


Figure 12. Influence of different time steps on the FE results (up); zoom of a portion of the curves (down).

considered problem and not only on the discretization method adopted. Indeed, numerous factors, in particular the external loading and the initial conditions as well as the viscoelastic properties, significantly affect the response and its temporal evolutions. The time step should be adapted accordingly.

This section has presented simple examples of FE calculations performed with the model in the 2D situation. The aim was mainly to evaluate the two strain discretization methods proposed and their influence on the numerical results. As both methods provide accurate results, and the time steps turn out to impact the response very moderately, we consider that the implementation of the model is effective. The next stage of the study will consist in integrating the proposed creep model into a thermo–hydro–mechanical formulation so as to carry out simulations of concrete structures

subjected to long-term thermal and hydric loadings. Because of the length of the present paper, this aspect will be investigated and developed in a subsequent study.

6. CONCLUSIONS

In the present paper, a simple micromechanical creep model accounting for the effects of h_r on the creep behavior has been developed for HCP. In this model, the main hydrated phases simplified as elastic spherical inclusions and the pores considered as spherical voids subjected to water pressures are randomly distributed in the C-S-H taken as a viscoelastic matrix. The Mori–Tanaka method is then applied to estimate the effective mechanical properties of HCP in the LC space. An analytical approximate inversion of the expressions of these properties is obtained by applying the Donolato method. Comparisons between numerical simulations and experimental data are presented for various values of the h_r in the case of uniaxial compression. These comparisons indicate that the creep properties of the C-S-H phase, which are assumed here for simplicity to be linear viscoelastic, should vary with h_r to obtain a correct agreement between numerical and experimental results. This dependence is ensured in this study by assuming that the mesopores, which progressively empty when h_r decreases, are replaced by elastic inclusions, which then strengthen the surrounding C-S-H matrix. Although this approach gives satisfying results, a better understanding of the role played by the interlayer water in C-S-H would be necessary to more accurately characterizing the creep behavior of cement-based materials.

The resulting explicit formulation in the time space of the stress–strain relation makes it possible to quite easily implement the model in a FE code, provided a relevant discretization of the strain tensor at each time step is carried out. FE simulations of simple creep tests show a very good agreement between numerical and analytical results, which mean that the implementation of the micromechanical model is effective.

The next stage of this study will be to integrate the proposed creep model in a coupled thermo–hydro–mechanical formulation so as to perform simulations of concrete structures subjected to thermomechanical loadings with induced hydric transfers. This will entail extending the model to concrete material by considering that it is composed of an HCP matrix in which are distributed the aggregates viewed as a particulate phase. Moreover, the effects of the temperature on the creep properties of the C-S-H will be investigated and taken into account. This work is currently in progress. In addition, further experiments are still needed for an advanced validation of the model, in particular, in the case of triaxial loadings.

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REFERENCES

1. Taylor HFW. *Cement Chemistry*, Telford, 1997.
2. Jennings H-M. A model for the microstructure of calcium silicate hydrate in cement paste. *Cement and Concrete Research* 2000; **30**:101–116.
3. Bazant Z-P. Prediction of concrete creep and shrinkage: past, present and future. *Nuclear Engineering and Design* 2001; **203**:27–38.
4. Bazant Z-P, Hauggaard A-B, Baweja S. Microprestress-solidification theory for concrete creep. I: aging and drying effects. *ASCE Journal of Engineering Mechanics* 1997; **123**:1188–1194.
5. Bazant Z-P, Hauggaard A-B, Baweja S. Microprestress-solidification theory for concrete creep. II: algorithm and verification. *ASCE Journal of Engineering Mechanics* 1997; **123**:1195–1201.
6. Granger L-P, Bazant Z-P. Effect of composition on basic creep of concrete and cement paste. *ASCE Journal of Engineering Mechanics* 1995; **121**:1261–1270.
7. Bazant Z-P, Cusatis G, Cedolin L. Temperature effect on concrete creep modeled by microprestress-solidification theory. *ASCE Journal of Engineering Mechanics* 2004; **130**:691–699.
8. Roelfstra P-E. *A Numerical Approach to Investigate the Properties of Concrete*. Ecole Polytechnique Fédérale de Lausanne: Numerical concrete, 1989.
9. Sercombe J, Hellmich C, Ulm FJ, Mang H. Modeling of early-age creep of shotcrete. I: model and model parameters. *ASCE Journal of Engineering Mechanics* 2000; **126**:284–291.

10. Scheiner S, Hellmich C. Continuum microviscoelasticity model for aging basic creep of early-age concrete. *ASCE Journal of Engineering Mechanics* 2009; **135**:307–323.
11. Mandel J. *Mécanique des Milieux Continus*. Gauthier-Villars: Paris, 1966.
12. Laws N, McLaughlin R. Self-consistent estimates for viscoelastic creep compliances of composite-materials. *Proceedings of the Royal Society of London. Series A. Mathematical, Physical and Engineering Sciences* 1978; **359**:251–273.
13. Brinson L-C, Lin W-C. Comparison of micromechanics methods for effective properties of multiphase viscoelastic composites. *Composite Structures* 1998; **41**:353–367.
14. Le Q-V, Meftah F, He Q-C, Le Pape Y. Creep and relaxation functions of a heterogeneous viscoelastic porous medium using the Mori–Tanaka homogenization scheme and a discrete microscopic retardation spectrum. *Mechanics of Time-Dependent Materials* 2007; **11**:309–331.
15. Lahellec N, Suquet P. Effective behavior of linear viscoelastic composites: a time-integration approach. *International Journal of Solids and Structures* 2007; **44**:507–529.
16. Donolato C. Analytical and numerical inversion of the Laplace–Carson transform by a differential method. *Computer Physics Communications* 2002; **145**:298–309.
17. Gregg S. *Adsorption, Surface Area and Porosity*. Academic Press, 1982.
18. Adamson A-W. *Physical Chemistry of Surfaces*. Wiley-Interscience, 1997.
19. Butt H-J, Graf K, Kappl M. *Physics and Chemistry of Interfaces*. Wiley-VCH, 2003.
20. Harald I. *Physics of Surfaces and Interfaces*. Springer, 2006.
21. Luth H. *Solid Surfaces, Interfaces and Thin Films*. Springer-Verlag Berlin and Heidelberg GmbH & Co. K, 2001.
22. Jennings H-M, Thomas J-J, Gevrenov J-S, Constantinides G, Ulm F-J. A multi-technique investigation of the nanoporosity of cement pastes. *Cement and Concrete Research* 2007; **37**:329–336.
23. Vandamme M, Ulm F-J. Nanogranular origin of concrete creep. *Proceedings of the National Academy of Sciences* 2009; **106**(26):10552–10557.
24. Richardson I-G. The nature of the hydration products in hardened cement pastes. *Cement and Concrete Composites* 2000; **22**:97–113.
25. Scrivener KL. Backscattered electron imaging of cementitious microstructures: understanding and quantification. *Cement and Concrete Composites* 2004; **26**(8):935–945.
26. Richardson I-G. The calcium silicate hydrates. *Cement and Concrete Research* 2008; **38**:137–158.
27. Bernard O, Ulm F-J, Lemarchand E. A multiscale micromechanics-hydration model for early-age elastic properties of cement-based materials. *Cement and Concrete Research* 2003; **33**:1293–1309.
28. Constantinides G, Ulm F-J. The effect of two types of C-S-H on the elasticity of cement-based materials: results from nanoindentation and micromechanical modeling. *Cement and Concrete Research* 2004; **34**:67–80.
29. Tennis P-D, Jennings H-M. A model for two types of calcium silicate hydrate in the microstructure of Portland cement pastes. *Cement and Concrete Research* 2000; **30**:855–863.
30. Quenard D, Sallee H. Water vapor adsorption and transfer in cement-based materials: a network simulation. *Materials and Structures* 1992; **25**:515–552.
31. Sanahuja J, Dormieux L, Chanvillard G. Modelling elasticity of a hydrating cement paste. *Cement and Concrete Research* 2007; **37**:1427–1439.
32. Bary B, Béjaoui S. Assessment of diffusive and mechanical properties of hardened cement pastes using a multi-coated sphere assemblage model. *Cement and Concrete Research* 2006; **36**:245–258.
33. Stora E, Bary B, He Q-C. Modelling and simulations of the chemo-mechanical behavior of leached cement-based materials. Leaching process and induced loss of stiffness. *Cement and Concrete Research* 2009; **39**:763–772.
34. Rusanov A-I. Surface thermodynamics revisited. *Surface Science Reports* 2005; **58**:111–239.
35. Rusanov A-I. Equilibrium thin liquid films. *Colloid Journal* 2007; **69**:39–49.
36. Derjaguin B-V, Churaev N-V, Muller V-M. *Surface Forces*. Consultants Bureau: New York, 1987.
37. Christensen R-M. *Theory of Viscoelasticity*. Dover Publications, 1982.
38. Bary B. Estimation of poromechanical and thermal conductivity properties of unsaturated isotropically microcracked cement pastes. to appear in *International Journal for Numerical and Analytical Methods in Geomechanics*, DOI: 10.1002/nag.969, 2011.
39. Chateau X, Dormieux L. Micromechanics of saturated and unsaturated porous media. *International Journal for Numerical and Analytical Methods in Geomechanics* 2002; **26**:831–844.
40. Dormieux L, Molinari A, Kondo D. Micromechanical approach to the behavior of poroelastic materials. *Journal of the Mechanics and Physics of Solids* 2002; **50**:2203–2231.
41. Dormieux L, Kondo D, Ulm F-J. *Microporomechanics*. Wiley, 2006.
42. Mura T. *Micromechanics of Defects in Solids*. Martinus Nijhoff: Dordrecht, The Netherlands, 1982.
43. Milton G-W. *The Theory of Composites*. Cambridge University Press, 2001.
44. Nemat-Nasser S, Hori M. *Micromechanics: Overall Properties of Heterogeneous Materials*. North Holland, 1998.
45. Lévesque M, Gilchrist M-D, Bouleau N, Derrien K, Baptiste D. Numerical inversion of the Laplace–Carson transform applied to homogenization of randomly reinforced linear viscoelastic media. *Computational Mechanics* 2007; **40**:771–789.
46. Bary B. Modélisation du fluage des bétons en température. CEA internal Report NT DPC/SCCME 08-437-A (in French), 2008.
47. Baroghel-Bouny V. Caractérisation des pâtes de ciment et des bétons. PhD thesis (in French), LCPC, 1994.
48. Baroghel-Bouny V. Water vapour sorption experiments on hardened cementitious materials: part I: essential tool for analysis of hygral behaviour and its relation to pore structure. *Cement and Concrete Research* 2007; **37**(3):414–437.