Century-long durability of concrete structures: Expansiveness of hydration and chemo-mechanics of autogenous shrinkage and swelling

Zdeněk P. Bažant
Departments of Civil, Mechanical and Materials Science Engineering, Northwestern University, Evanston, USA

Saeed Rahimi-Aghdam
Department of Civil and Environmental Engineering, Northwestern University, Evanston, USA

ABSTRACT: Computations of long-time hygrothermal effects in concrete structures necessitate a physically based model for autogenous shrinkage and swelling of hardened portland cement paste. The present goal is to propose such a model. As known since 1887, the volume of cement hydration products is slightly smaller than the original volume of cement and water. However, this does not mean that the hydration reaction causes contraction of the cement paste and concrete. According to the authors' recently proposed paradigm, the opposite is true for porous cement paste as a whole. The growth of C-S-H shells around anhydrous cement grains pushes the neighbors apart and thus causes *volume expansion* of the porous cement paste as a whole, while the nanoscale volume contraction of hydration products contributes to porosity. The growth of ettringite and portlandite crystals may also cause additional expansion. On the material scale, the expansion always dominates over the contraction, i.e., the hydration per se is, in the bulk, *always expansive*, while the source of all of the observed shrinkage, whether autogenous or due to external drying, is the compressive elastic or viscoelastic strain in the solid caused by a decrease of chemical potential of pore water, with the corresponding decrease in pore humidity, increase of solid surface tension and, mainly, decrease of disjoining pressure. The low density C-S-H and high density C-S-H are distinguished in the proposed model. The selfdesiccation, shrinkage and swelling can all be predicted from one and the same unified model, as confirmed by comparisons with with the existing experimental evidence. The model is ready for use in finite element programs.

1 INTRODUCTION

The interaction of creep, shrinkage, pore humidity changes, water transport, and cement hydration is a complex problem, which has been debated for a long time. Although various useful models for particular phenomena have been developed, no comprehensive model capturing all these phenomena has been available until its general form was presented in Bažant et al. (2015).

One problem has been that most models assume the autogenous shrinkage, drying shrinkage and swelling to terminate with a horizontal asymptote. This assumption has been erroneous, caused by the widespread use of misleading plots in linear time scale and utter scarcity of long time tests. Nevertheless, there exist a few data (Persson 2002, Brooks & Wainwright 1983) whose logarithmic plots document that no asymptotic bound is in sight even after 30 years (Figs. 1a and 1b).

Here it is argued that they both the autogenous shrinkage and swelling will evolve logarithmically for decades, probably even centuries. What is the source of such behavior and how can it be modelled?

To answer this key question, let us begin with the autogenous shrinkage. Following Bažant et al. (2015), we consider its primary cause to be the decrease of pore relative humidity h due to selfdesiccation, no different to decrease of h due to external drying. According to RILEM TC 196-ICC (Kovler & Jensen 2007), the selfdesiccation is described as "the reduction of h in a sealed system when empty pores are generated". Obviously, the ultimate cause of selfdesiccation must be the hydration reaction.

However, the autogenous shrinkage, like drying shrinkage, must be getting prolonged by the *viscoelastic* behavior (i.e., creep plus elasticity, Bažant et al. 2015) of the solid skeleton of hardened cement paste loaded by pore pressure, reduced by a certain porosity factor. This is the second independent cause of long-term evolution of deformations.

The primary source of long-term autogenous shrinkage is the long-lived hydration process. Yet it has been widely believed that hydration terminates
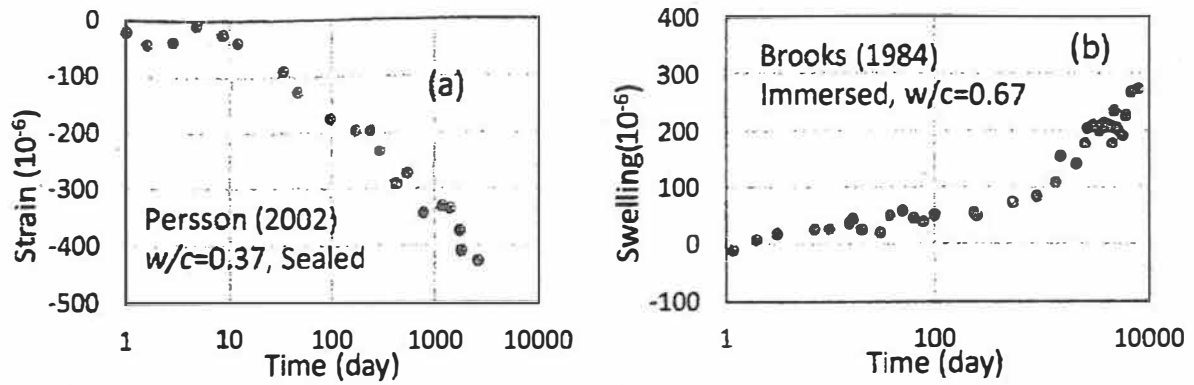
Figure 1. Long-term autogenous shrinkage and swelling experiments. a) autogenous shrinkage, b) swelling.

in about a year. But that is correct only in specimens thin enough to dry up uniformly within a year. In the cores of massive walls exposed to drying, high enough h may persist for decades, even centuries. What makes the hydration process long-lived and progressively slower is that, after the first day, the anhydrous cement grains become enveloped in contiguous shells of cement hydrate. The shells are highly impermeable and allow only a very slow transport of water toward the anhydrous grain interface. In addition, as these shell gradually thicken, the pore relative humidity h (and thus the chemical potential of water, μ) decrease, which slows down and further prolongs the hydration. This phenomenon was recently mathematically described by Rahimi-Aghdam et al. (2017).

As shown in this work, if h in the pores does not drop below about 0.7, the hydration reaction continues for several decades, probably even centuries. This must cause similar long-time evolution of autogenous shrinkage.

There is another phenomenon that doubtless prolongs greatly the autogenous shrinkage—the aging *viscoelasticity*, i.e., the creep plus elastic deformation. Maintaining local thermodynamic equilibrium requires equality of the chemical potential μ in all the phases of pore water. So a decrease of pore humidity h must produce tensile stress changes in all the phases of water (vapor, liquid, adsorbed and hindered adsorbed). These stress changes must be balanced by compressive stress changes, whichin turn must cause compressive elastic deformation and creep in the solid skeleton of cement paste.

In water immersion, most concretes are swelling. As several experiments show, the swelling is a also long-lived phenomenon. As documented by a few tests, concrete specimens immersed for a decade continue to expand logarithmically with no bound in sight. In immersed specimens, pores near the surface are saturated, $h = 1$, and those in the core undergo selfdesiccation until the saturation

front arrives. From this it is clear that the expansion cannot be driven by pore humidity. Hence, it must be driven by chemical expansion during hydration. Since it is inconceivable for the hydration to be both expansive in water immersion and contractive without immersion, we have only two candidate hypotheses—the hydration is *always either contractive or expansive.*

If it were the former, the swelling under water immersion would not be physically explicable. But if it is the latter, the autogenous and drying shrinkage is explicable. Indeed, the tensile stress changes in pore water due to drying or selfdesiccation must be balanced by compression in the solid skeleton of cement paste, which will inevitably cause viscoelastic compressive strain in the solid skeleton. Although contrary to traditional thinking, this leads to a new paradigm (Bažant et al. 2015):

The deformation of porous hardened cement paste caused by the hydration process is always expansive.

At the dawn of cement research, it was established by Le Chatelier, and later confirmed by Powers and others, that the cement hydration reaction is always contractive, i.e., the volume of the cement gel produced by hydration is always smaller than the sum of the original volumes of anhydrous cement and water. But now we claim that this is true only on the level of nanoscale microscopic powder. In the bulk, with a sufficient pore volume growth and constant relative humidity in the pores, a porous material *must always expand*, even if the volume of solid in the material decreases.

In hardened portland cement paste, the growth of two contacting C-S-H shells around anhydrous cement grains must push the neighbors apart and thus cause volume expansion of the porous cement paste during hydration (Bažant, Donmez, Masoero, & Aghdam 2015). The compression force within each pair of contacting neighbors, akin the crystal growth pressure, must be balanced by overall tension in the solid skeleton of cement

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paste. Like hydration, this tension evolves for decades, and so must the swelling. So the long-term hydration causes not only autogenous shrinkage, but also swelling. Furthermore, the long-time loading of the solid skeleton by pore pressure must, of course, produce not only elastic deformation but also creep.

It should be noted that several studies suggested that the source of swelling should be the growth of ettringite crystals (Taylor, Famy, & Scrivener 2001, Diamond 1996). But this could be only one minor contribution and can not be the basic cause of the long-term expansion, for two reasons. First, the ettringite crystals do not form with a delay of many years. Rather, they dissolve in early age and thus cannot affect long-term swelling. Second, the long-term swelling can be seen in any type of cement regardless the amount of C_3A phase.

Therefore, even if we would admit the ettringite crystal growth to be cause swelling in the initial days, it could not explain the swelling in the long term. The final possible source of long-term swelling is Alkali-Silica reaction (ASR). Although ASR can cause considerable swelling, but only if the alkali-content of the cement paste be high enough and aggregates be reactive (Bažant & Rahimi-Aghdam 2016, Rahimi-Aghdam, Bažant, & Caner 2016) that is usually not the case in normal laboratory experiments.

2 EQUATIONS GOVERNING BOTH SHRINKAGE AND SWELLING

For normal saturated elastic porous material, the volumetric strain ϵ is related to the volumetric mean stress σ and the fluid pore pressure p through the equation:

$$\epsilon = \frac{\sigma_{eff}}{K} = \frac{\sigma + bp}{K} \tag{1}$$

where K is the bulk modulus of the dry skeleton and b is the Biot coefficient which is calculated as (Biot & Willis 1957):

$$b = 1 - \frac{K}{K_s} \tag{2}$$

Here K_s is the bulk modulus of the solid skeleton; for cement paste in particular, K_s is the bulk modulus of C-S-H gel and can be considered as $K_s = 45$ MPa (Nielsen 1991). The foregoing equations are valid only for saturated pores. In the cement paste and other partly saturated media, the equations need to be modified so as to consider several pore fluids. For instance, unsaturated cement paste contains liquid water, water vapor, adsorbed water and air. Usually, water vapor and air (gaseous phases) are considered together as one gas phase.

To deal with unsaturated case, we need the average pore fluid stress p^*. In the case of cement paste:

$$p^* = S_w p_w + S_g p_g \tag{3}$$

where p_g and p_w are, respectively, the pressure in gas and liquid water phases. The adsorbed water can be, for our purpose, ignored; S_w and S_g are the saturation degrees of liquid water and gas:

$$S_w = \frac{\phi_w}{\phi}; \quad S_g = \frac{\phi_g}{\phi} = 1 - S_w \tag{4}$$

where ϕ = total porosity, ϕ_w = porosity occupied by water and ϕ_g = porosity occupied by gas.

Drying caused a decrease of pressure, p_w, in the liquid phase in cement paste pores, which has three sources:

1. increase in capillary tension ($p_c < 0$) according to the Kelvin-Laplace (KL) theory,
2. decrease Δp_d of disjoining pressure p_d in nanopores (Bažant 1972, Bažant & Bazant 2012, Bažant & Jirásek 2017), and
3. decrease δp_s of pressure p_s caused by increase $\Delta \gamma$ of solid-fluid surface tension that results from a decrease of spreading pressure in adsorbed water layer on the surface on nano scale globules of C-S-H. Probably Δp_d is most important.

Fortunately, all the three sources of pressure decrease depend on the relative humidity, h, in a similar way, as follows:

$$p_i = C \ln h \tag{5}$$

Therefore, it is logical to consider p_w to depend on the relative humidity similarly:

$$p_w = C_w \ln h \tag{6}$$

Coussy et al. (2004) derived the modified relations for unsaturated cement paste, omitting cement paste aging and creep. Here we adopt their formulation but extend it to include the aging and creep. Based on Coussy et al. (2004), the variation of solid matrix free energy $\psi(\epsilon, \phi)$ can be calculated as,

$$d\psi(\epsilon, \phi) = \sigma d\epsilon + \pi d\phi \tag{7}$$

where π is the equivalent pressure, which can be calculated as:

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$$\pi = p^{\bullet} - U; \quad U = \int_{S_w}^{1} (p_g - p_w) dS_w \quad (8)$$

For normal saturated porous materials, the change of the free energy of solid can be calculated as,

$$d\psi(\epsilon, \phi) = \sigma d\epsilon + p d\phi \quad (9)$$

Comparing Eqs. 9 and 7, we can conclude that the equivalent pressure π in an unsaturated medium acts the same way as the pressure p in a saturated medium. Using Eqs. 3, 4 and 8, the variation of equivalent pressure can be calculated as:

$$d\pi = S_w dp_w + S_g dp_g \quad (10)$$

It should be noted that Coussy's use of $p^{\bullet}$ is a simplification of the physical reality, since the water molecules cannot pass from one pore to the next while remaining in the vapor or liquid state. Rather they must first enter the adsorbed water phase and pass to the neighboring capillary pore through nanopore necks filled by hindered adsorbed water (Bažant 1972, Bažant & Jirásek 2017). Thus, what really matters is the gradient of the spreading pressure (or chemical potential μ) along the hindered adsorbed water layer. Effectively, we assume this gradient to be proportional to grad $p^{\bullet}$ (or grad h, or grad μ). Note that, the better and more detailed method to consider the effect of pores with significantly different sizes is considering three-phase medium (Bažant, Chau, & Rahimi-Aghdam 2017).

For cement based materials, the pressure p_g usually remains close to atmospheric pressure that is significantly smaller than liquid water pressure p_w. Therefore, we can neglect p_g and, using equation 6, write:

$$d\pi = S_w dp_w = C_w S_w \frac{dh}{h} \quad (11)$$

Now that we have the equation for equivalent pressure change, we can calculate the volumetric strain considering the creep:

$$\epsilon(t) = \int_0^t J(t - t') d\sigma_{eff}(t') \quad (12)$$

where t' is the time of applying load increment $d\sigma_{eff}(t')$ and J is the non-aging creep compliance function. Using Biot coefficient b and the equivalent pressure π, we can calculate $d\sigma_{eff}$ as:

$$d\sigma_{eff} = d\sigma + bd\pi = d\sigma + bC_w S_w \frac{dh}{h} \quad (13)$$

Until now we have not considered the effect of swelling due to the hydration process. To this end, we can consider an additional pressure (akin to crystal growth pressure) that is induced by the macro-expansive hydration process, σ_{hyd}. For simplicity, we can assume it to depend linearly on the hydration degree. Adding this pressure, we can rewrite equation 13 as:

$$d\sigma_{eff} = d\sigma + bd\pi = d\sigma + b\left(C_w S_w \frac{dh}{h} + C_{hyd} d\alpha + d\sigma_{cry}\right) \quad (14)$$

where C_{hyd} is a calibration parameter and $d\sigma_{cry}$ represents any possible additional pressure due to crystal growth. This study focuses on the swelling due to hydration, and so we assume $d\sigma_{cry} = 0$.

It should be mentioned that aging has a considerable effect on the stiffness of cement-based materials. For simplicity, we consider the stiffness of cement paste to be related to the hydration degree linearly:

$$K(\alpha) = K_u\left(\frac{\alpha - \alpha_{set}}{\alpha_u - \alpha_{set}}\right) \quad (15)$$

The Biot coefficient depends on the bulk modulus K and can be calculated as

$$b(\alpha) = 1 - \frac{K(\alpha)}{K_S} \quad (16)$$

Finally, we must introduce a realistic equation for the creep compliance of solid skeleton, J, loaded by pore water pressure. This compliance is doubtless different from the creep compliance of the cement paste as a whole (which includes the load-bearing adsorbed water in nanopores). For limited time periods, we can ignore the aging due to hydration and, for lack of deeper information, we assume a simple power function:

$$J(t - t') = c_0 + c_1(t - t')^n \quad (17)$$

where c_0 corresponds to the instantaneous response; constant c_1 and exponent n are two calibration parameters.

Now we try to predict the autogenous shrinkage for cement pastes with different water-cement ratios, w/c. Their value changes not only the amount of selfdesiccation but also the strength and stiffness. Therefore, to predict the autogenous shrinkage correctly, we need a model that can predict the stiffness, E, for different cement pastes correctly. In the literature, there are several empirical models for predicting the stiffness of concrete from
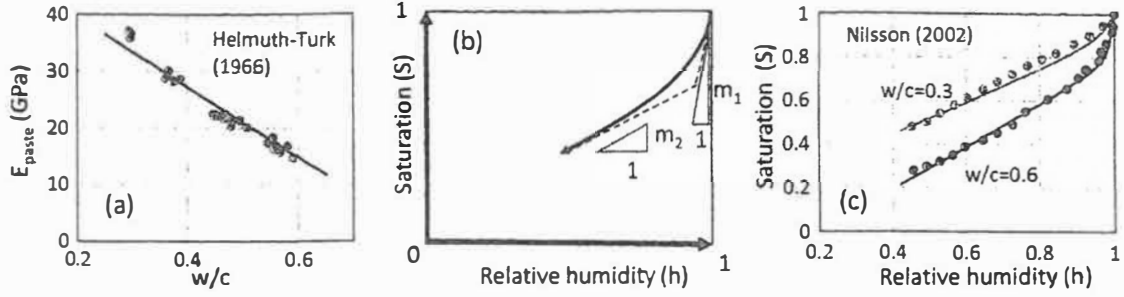
Figure 2. The stiffness of cement paste and desorption isotherm. a) Stiffness of cement paste with different water content, b) bi-linear desorption isotherm and c) experimental vs. simulated desorption isotherm for cement pastes with different water content.

its strength f'_c, but they are not suitable for the hardened cement paste. Therefore, we propose the following simple, empirically calibrated, equation that can predict the stiffness of cement paste for different w/c (Figure 2a):

$$E = A - Bw/c, \quad A = 52GPa, B = 62GPa \quad (18)$$

The Poisson ratio for cement pastes with different w/c is considered the same, $v = 0.2$.

3 DESORPTION AND ABSORPTION ISOTHERM

At constant temperature, the relation between evaporable water w_e and relative humidity h is known as the sorption isotherm. More specifically, the desorption isotherm describes w_e as a function of h at decreasing water content (or drying), while the absorption isotherm (sometimes called just the sorption isotherm) applies to increasing water content (wetting). The sorption isotherms exhibit pronounced hysteresis, which means that the absorption isotherm lies significantly below the desorption isotherm. Here we focus on drying and propose a simple equation for the desorption isotherm.

To have dimensionless parameters, the sorption isotherm is sometimes defined as a relation between the relative humidity, h, and the saturation degree, S (rather than the water content), which is what is adopted here. It makes it easier to compare the desorption isotherms for concretes with different water contents.

As shown by several studies (Nielsen 1991, Xi, Bažant, & Jennings 1994), the desorption isotherm is highly nonlinear and its shape depends significantly on the concrete properties. Based on experiments, the most important parameters are the water-cement ratio and age. In addition, some studies show that adding certain admixtures can change desorption isotherm significantly. Especially,

adding silica fume makes the pore sizes more uniform and makes steeper the decrease of pore relative humidity at states below full saturation (Baroghel-Bouny, Mounanga, Khelidj, Loukili, & Rafaï 2006, Jiang, Sun, & Wang 2006). Various methods to obtain the desorption isotherm have been proposed, but either they are too complicated to use or necessitate several calibration parameters hard to determine.

This study proposes some simple empirical equations for the desorption isotherm. Based on experimental results, for high relative humidities ($h > 50\%$) desorption isotherm can be fairly approximated by two straight lines. Accordingly, the isotherm is formulated as a smooth curve that has these two straight lines as asymptotes (see Fig. 2b), as follows:

$$dh = k(h,\alpha)dS \quad (19)$$

$$\frac{1}{k(h,\alpha)} = m_2 + (m_1 m_2)\frac{1}{1 + \left(\frac{1-h}{1-h^*}\right)^2} \quad (20)$$

where $k(h, \alpha)$ is the inverse slope of the desorption isotherm; m_1 and m_2 are the slopes of the two straight lines (which can be more simply used to approximate desorption isotherm), and h^* is the intersection point of the two straight lines. These parameters can be empirically estimated as follows:

$$m_1 = \frac{f_a}{c_{SF}}\left[2.4 + \left[(\frac{w/c - .13}{0.19})^{1.5}\right]\right] \quad (21)$$

$$m_2 = c_{SF}^{0.5} 1.18w/c^{0.4} \quad (22)$$

$$h_s = 1 - fc_{SF}\frac{0.03}{w/c^2} \quad (23)$$

$$f_a = (\alpha_u - \alpha_0)/(\alpha - \alpha_0) \quad (24)$$

Parameter c_{SF} is included to account for possible addition of silica fume. In its absence $c_{SF} = 1$, and

generally, for a finite specific content ξ_{SF} of silica fume (mass of silica fume/mass of cement):

$$c_{SF} = 1 + 2\xi_{SF} \tag{25}$$

Fig. 2c compares the calculation results (solid curves) with the measured data using aforementioned equations. As it can be seen, the simulated desorption isotherms are in a good agreement with the experimental results. It should be mentioned that some studies showed some minor effect of temperature on isotherm, but for simplicity this effect has been neglected.

4 SELFDESICCATION

According to RILEM TC 196-ICC, the selfdesiccation is defined as "the reduction in the internal relative humidity of a sealed system when empty pores are generated". It can occur in any concrete cured under sealed conditions, but its effects are quite dependent on the sizes of the generated empty pores. The size of empty pores depends mainly on the water-to-cement ratio (w/c), the degree of hydration, the particle size distribution of cement, and the type of admixtures. The pore size is smaller for lower w/c and finer cements. A smaller pore size leads to more pronounced selfdesiccation. Furthermore, adding some fine admixtures such as the silica fume decreases the pore size and thus increases the value of selfdesiccation.

To calculate the selfdesiccation correctly, we should start with the hydration reaction and the amount of water that is used in this reaction. The ordinary portland cement (OPC) consists of various phases such as alite (C_3S), belite (C_2S), calcium aluminate (C_3A), tetracalcium aluminoferrite (C_4AF) and gypsum, as well as minor other phases (we use cement chemistry notation in which C, S, A and F stand, respectively, for CaO, SiO_2, Al_2O_3 and Fe_2O_3) (Taylor, 1997). C_3S and C_2S are two major components of OPC and their ratio depends on cement type. To measure the amount of consumed water in the hydration reaction, for simplicity we consider just C_3S and C_2S and we assume the cement to be composed of 8% C_3S and 20% C_2S. The nano-level hydration reaction of C_2S and C_3S can be summarized as (Qomi, Krakowiak, Bauchy, Stewart, Shahsavari, Jagannathan, Brommer, Baronnet, Buehler, Yip, et al. 2014),

$$C_2S + 2.1H \rightarrow C_{1.7}SH_{1.8} + 0.3CH$$
$$C_3S + 3.1H \rightarrow C_{1.7}SH_{1.8} + 1.3CH$$

where $C_{1.7}SH_{1.8}$ and H are, respectively, the typical C-S-H type found in OPC pastes and water. Table 1 summarizes the molar volume and density of different components in hydration reactions. Note that, in the aforementioned hydration reactions, the C-S-H pores were assumed to be empty, and so it is necessary to include additional water that is trapped in C-S-H pores.

Recently it became well established (Constantinides, Ulm, & Van Vliet 2003, Jennings 2000, Tennis & Jennings 2000) that two types of C-S-H are produced during hydration reaction; low density C-S-H (LD), with porosity 36%, and high density C-S-H (HD) with porosity 26%. Jennings and Thomas (2000) showed that, for different w/c values, the ratio between these two C-S-H types varies. For instance their model (JT model) predicted that for w/c = 0.45, there are 50% HD and 50% LD while for w/c = 0.25 there are 80% HD and 20% LD. Therefore, the average porosity of C-S-H, ϕ_{gp}, mainly depends on w/c. Here, for simplicity, we assume a linear relation limited by lower and upper bounds, as follows:

$$0.27 < \phi_{gp} = 0.28 + .2(w/c - 0.3) < 0.35 \tag{26}$$

The lower and upper bounds on ϕ_{gp} are introduced because considering only the LD or HD phases may be insufficient.

Having quantified the hydration relation and calculated the porosity of C-S-H, we can calculate the total volume of water, ξ_{gc}, per unit volume of cement, that is used in hydration to produce a C-S-H gel, the volume of empty pores, ξ_{bw}, and the volume of water that fills gel pores, ξ_{fp}:

$$\xi_{wc} = \xi_{bw} + \xi_{fp} \tag{27}$$

$$\xi_{bw} = w_{C_2S}\xi_{bw}^{C_2S} + w_{C_3S}\xi_{bw}^{C_3S} \tag{28}$$

$$\xi_{fp} = \phi_{gp}\xi_{gc}S_{gp} \tag{29}$$

where S_{gp} = saturation degree of gel pores; w_{C_2S} and w_{C_3S} = volume ratio of C_2S and C_3S respectively; $\xi_{bw}^{C_2S}$ and $\xi_{bw}^{C_3S}$ = volume of water that is consumed to hydrate one unit volume of C_2S and

Table 1. The molar volume and density of calcium-silicate phases present in the cement paste.

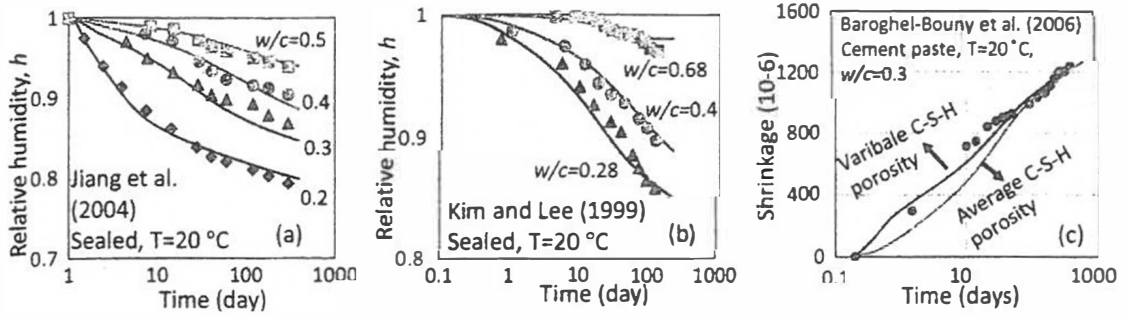| Phase	Molar volume (cm^3/mol)	Density (g/cm^3)
C_3S	72.9	3.15
C_2S	52.7	3.28
H_2O	18.0	1.0
C-S-H	110.1	2.05
CH	33.1	2.24

Figure 3. Self-desiccation and autogenous shrinkage. a) predicted vs. experimental relative humidity decrease in sealed specimens (self-desiccation) for experiment by Jiang et al. (2005), b) predicted vs. experimental relative humidity decrease in sealed specimens (self-desiccation) or experiment by Kim and Lee (2005), and c) predicted vs. experimental autogenous shrinkage using both average porosity and variable porosity.

C_3S, respectively, and produce C-S-H gel with empty pores. If we assume that 80% of cement is the C_3S and 20% C_2S, we have.

$$\xi_{bw} = 0.2(37.8/52.7) + 0.8(55.8/72.9) = 0.755$$

$$\xi_{gc} = 0.2(110/52.7) + 0.8(110/72.9) = 0.487$$

Since the gel pores are smaller than the capillary pores, in the literature the gel pores have always been considered as saturated. But Rahimi-Aghdam et al. (2017) show that this cannot be true and that some of the gel pores must be treated like unsaturated capillary pores. Otherwise the water in gel pores would always have a uniform chemical potential and we could not explain the stoppage of hydration at a certain low relative humidity. In this Study, we assume 33% of gel pores to have the same saturation degree as the capillary pores, and 67% the same as nanopores (which, at high relative humidities, are always saturated). Accordingly, for a high relative humidity, the saturation degree of capillary pores is:

$$S_{gp} = 0.67 + 0.33 S_{cp} \tag{30}$$

Fig. 3a,b show the predicted selfdesiccation using the aforementioned parameter. As it can be seen, the predictions agree with the experiments well. In spite of that, the predicted autogenous shrinkage curves show some delay (see Fig. 3c). This should be due to the fact that, initially, most of produced C-S-H gel is LD and, as hydration reaction goes on, the ratio of HD increases. This phenomenon may be formulated as,

$$\phi_{gn} = r_{LD}\phi_{LD} + r_{HD}\phi_{HD} = 0.36 r_{LD} + 0.26(1 - r_{LD}) \tag{31}$$

$$r_{LD} = \exp\left(-c_{w/c} \frac{\alpha}{\alpha_u}\right) \tag{32}$$

$$c_{w/c} = \frac{0.17}{(w/c - 0.05)^4} \tag{33}$$

where r_{LD} and r_{HD} are the volume fractions of the C-S-H gel produced. Using either the average porosity or a variable porosity C-S-H, one finds the predicted selfdesiccation curves to be almost the same but, as seen in Fig. 3c, the use of average porosity leads at early times to cause a delay in the prediction of autogenous shrinkage. Therefore, only the variable humidity method is employed here.

5 AUTOGENOUS SHRINKAGE PREDICTIONS

To check how good the predictions of the present model are, we begin with the experiments of Jiang et al. (2006), in which the autogenous shrinkage of cement pastes was measured for different water-cement ratios. Fig. 4a compares the calculated curves with the measured points. As seen, the agreement is good. Note that the model was calibrated only by matching of the measured bulk modulus of the cement paste with $w/c = 0.25$, and all the rest was predicted.

The second set of experiments considered here is a similar set by Baroghel-Bouny et al. (2005). As seen in Fig. 4b, the predicted results are in good agreement with the experimental ones except for the case of the highest water content, $w/c = 0.50$, in which the experimental results show a greater initial swelling than predicted. The reason for this error is probably the extra initial swelling due to growth of ettringite crystals, which is not considered in the present model.
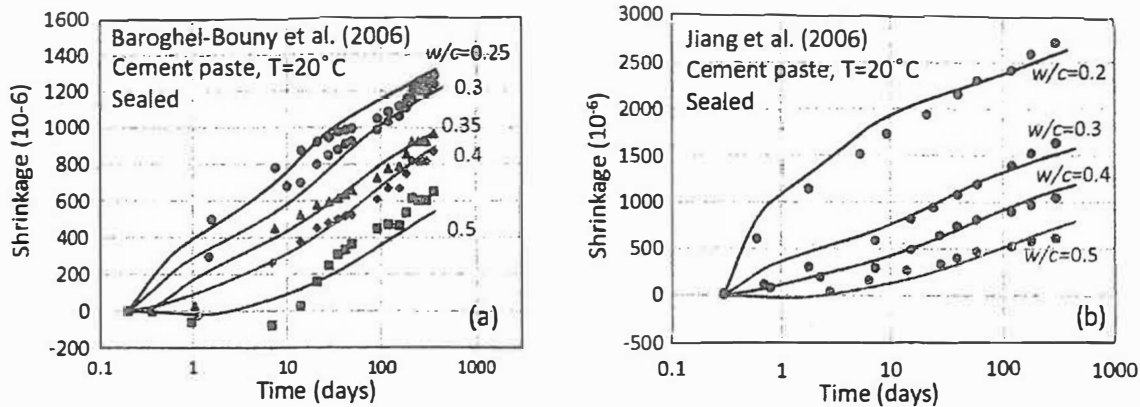
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Figure 4. Comparison of predicted curves with measured values of autogenous shrinkage for cement pastes with different watercement ratios.

6 SIZE EFFECT ON SWELLING UNDER WATER, TRANSITING FOR LARGE SIZES TO SHRINKAGE

Despite lack of experimental evidence, it is here predicted that there must be a large size effect on the diffusion of water that gradually fills the self-desiccated pores to 100% humidity. This diffusion is much slower than drying. As shown in Bažant (1975) (see also (Bažant & Jirásek 2017)), the amount of water that needs to be delivered to the propagating front of wetting is large because the humidity at the selfdesiccated front must be raised to 100% before the wetting front can advance farther.

Thus the interface condition at the advancing wetting front gives a large interface sink of water, which greatly slows down the advance of the diffusion front of wetting. Note that a similar (negative) sink is absent at the front of drying which, therefore, propagates much faster.

The result is that structural members thicker than about 0.2 m must be expected to always shrink (autogenously) under water for at least a century (Miyazawa & Monteiro 1996), due to selfdesiccation of most of their interior volume. The swelling of the (relatively thin) surface layer is cancelled by inducing a compressive stress parallel to the surface, which is beneficial as it prevents the formation of surface cracks.

7 CONCLUSION

The new hypothesis that, on the porous material scale, the hydration reaction is always expansive (Bažant et al., 2015) allows grand unification of models for drying shrinkage, autogenous shrinkage and swelling under water immersion, and especially their long-time (multi-decade and century

long) evolution. These diverse phenomena can all be predicted from one and the same material model, even for decade-long (and probably century -long) durations. When used in computer-aided design, this grand unification should be helpful for ensuring century-long durability, which is now seldom achieved for many large concrete structures.

ACKNOWLEDGMENT

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