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Early age shrinkage of concrete: back to physical mechanisms

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ABSTRACT

Solving a problem such as early age cracking of concrete calls for a physical understanding of the driving mechanisms behind early age shrinkage and creep. Among those mechanisms, the physical and mechanical consequences of cement hydration volume balance and the percolation of the mineral network will be reviewed. The capillary pressure induced by the water leaving the porosity of the material and the visco-plastic behavior of C-S-H products will also be discussed in this article. The different phases observed during the development of autogenous shrinkage of concrete will be identified and explained through those mechanisms.

1. INTRODUCTION

Autogenous shrinkage of concrete has been studied for several years. Up to the middle of the nineties, the research work has been often related to the determination of new testing methods [1-5]. Since then, this topic has received less attention, even though experimental artefacts could still be suspected in some measurements [6-8]. The determination of those artefacts is essential to insure the validity of parametric studies. In order to find, understand and solve the artefacts and direct the choice of relevant parameters for parametric studies, it is necessary to better understand the physical mechanisms involved in early age behavior of concrete. This will be the focus of this article.

2. CHEMICAL SHRINKAGE

It is now well known that hydration of hydraulic binders leads to a decrease in the absolute volume of the reactants [9].

Table 1 – Volumetric calculation for C_3S hydration

Product	Nb. of moles	Molar Weight (g/mol)	Density	Volume (ml)
C_3S	1	228	3.13	72.8
H	5.3	18	1	95.4
$C_{1.7}SH_4$	1	227.2	2.01	113.0
CH	1.3	74	2.24	42.9

Let us consider a C_3S hydration reaction [10] at early age:

$$C_3S + 5.3H \longrightarrow C_{1.7}\,SH_4 + 1.3CH$$

It is obvious that the mass balance is respected. However, as it has been done for instance by Justnes [10], it is possible to write the reaction in a volumetric way [Cf. Table 1].

From this calculation, for the hydration of 1 mole (72.8 ml) of C_3S, the consequences of the hydration can be considered in two ways:

1. **Total volume decreases** from 168.2 ml (72.8 + 95.4) to 156 ml (113 + 42.9). This decrease (7.3% of the initial volume) is the amount of chemical shrinkage for complete hydration of C_3S. We can notice that the value of 7.3% is very dependent on the value taken for the density of C-S-H.
2. During the same time, the **solid volume increases** drastically: from 72.8 ml to 156 ml (214% of the initial solid volume). The consequence of this solid volume increase is the building up of the microstructure and the refinement of the porous network during hydration.

Chemical shrinkage versus hydration degree

Chemical shrinkage can be measured continuously during several days [11-13] (weighing method). Fig. 1 gives an example of such a measurement.

Except for very low degrees of hydration (1 or 2% of hydration), chemical shrinkage can be linearly related to the degree of hydration measured by isothermal calorime-

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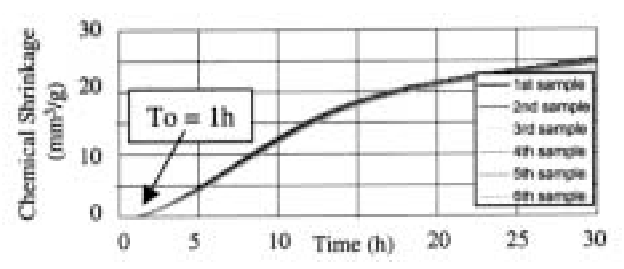

Fig. 1 – Chemical Shrinkage. Cement paste, W/C = 0.3.

try [7] [Cf. Fig. 2]. Nevertheless, it is important to notice that chemical shrinkage measurement is made under continuous water supply. The kinetics of development of degree of hydration is certainly overestimated in comparison to cement paste undergoing self-desiccation under sealed conditions. Fig. 2 also shows that the ultimate chemical shrinkage is very weakly dependent on the water / cement ratio (w/c). Consequently, as it has also been reported by Geiker [12], for a given cement, the chemical shrinkage curves can be used to quickly determine an acceptable degree of hydration vs time.

From the curves obtained in Fig. 2, it is possible to extrapolate the ultimate chemical shrinkage at about 80 mm³ per gram of cement for total hydration. If 100 g of cement are mixed at a w/c of 0.5, and if we consider that it is possible to reach total hydration, the contraction obtained by extrapolation is 8 cm³ (80 mm³ * 100) for 82 cm³ of cement paste. This value of ultimate chemical shrinkage is quite consistent with the generally taken value of 8 to 10% of the volume of cement paste [1, 14].

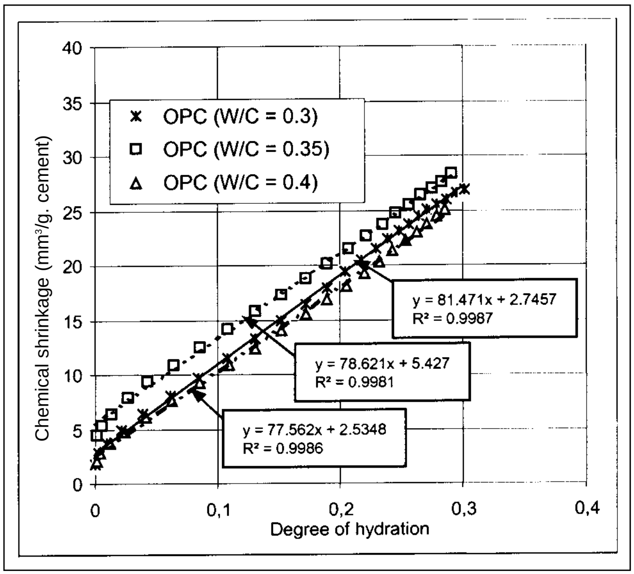

Fig. 2 – Relation between chemical shrinkage and degree of hydration from [7].

3. AUTOGENOUS SHRINKAGE

Autogenous shrinkage is the shrinkage occurring at constant temperature, without any moisture exchange with the surrounding medium. It is the external volume variation resulting from chemical shrinkage.

3.1 Autogenous shrinkage versus chemical shrinkage

Fig. 3 represents the variations of absolute volume caused by hydration reactions (chemical shrinkage), along with the resulting variations of apparent volume [17, 19].

During the suspension period (phases 1 and 2 in Fig. 3), the solid grains are isolated in a connected liquid phase. The material cannot resist the volume variations imposed by the hydration reactions. It is well understood [3, 7, 15], and experimentally assessed [6, 7, 16] that, during this period, chemical shrinkage and autogenous shrinkage coincide.

During phase 3 in Fig. 3, as hydration proceeds, the rigidity of the material gradually increases, due to an increase in connectivity of the solid particles. There are some parts of the material where the volume variations imposed by the hydration reactions are locally hindered by solid contacts. As it will be described further in this article, the macroscopic consequence for the material is a decrease in the slope in the curve representing autogenous shrinkage versus chemical shrinkage. Initially, this decrease remains small (about 5-10%). It is during this period that the first solid path within the material is created: it is the mineral percolation threshold. The movements of the particles belonging to this first solid network (which statistically appears throughout the whole volume of the material) are hindered in only one direction. This percolation does not confer yet a solid type of behavior to the material.

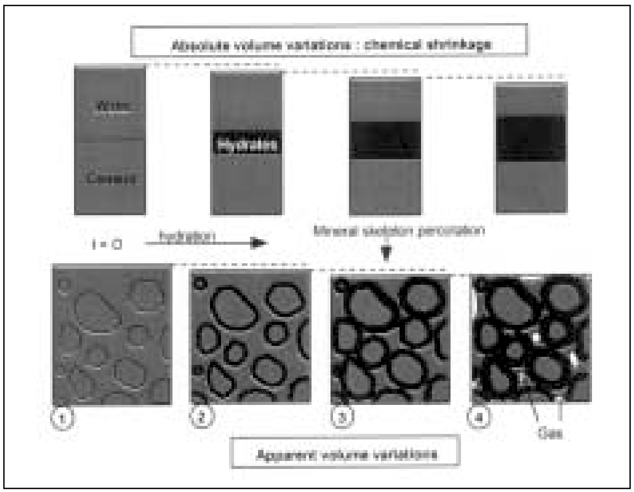

Fig. 3 – Self-desiccation as a consequence of chemical shrinkage (from [19] and [17]).

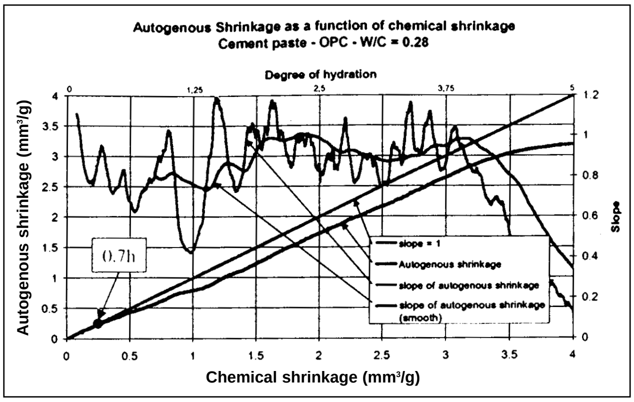

Fig. 4 – Comparison between chemical and autogenous shrinkage of cement paste. W/C = 0.28 (Approximate degree of hydration calculated on the basis of chemical shrinkage value).

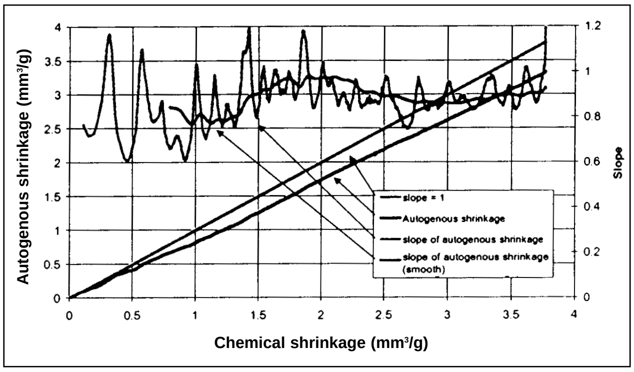

Fig. 5 – Comparison between chemical and autogenous shrinkage of cement paste. W/C = 0.3.

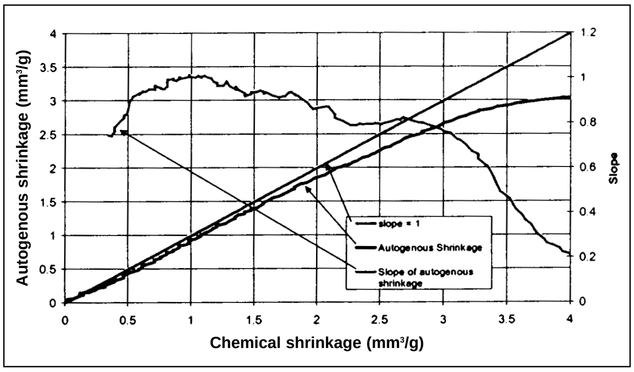

Fig. 6 – Comparison between chemical and autogenous shrinkage of cement paste. W/C = 0.35.

Between phase 3 and phase 4, a transition occurs for the cement paste from a liquid to a solid type of behavior. During this suspension-solid transition, the movements of the particles belonging to the solid network become hindered in all three directions. After that point, the volume variations caused by hydration are not compatible with the acceptable mechanical deformations of the mineral skeleton, and a significant volume of gas develops in the capillary porosity of the material. This phenomenon is called self-desiccation and has been extensively reported in the literature [3, 15, 17].

The previously described evolution of autogenous shrinkage versus chemical shrinkage has been experimentally assessed, with a very good level of accuracy. For that purpose, two types of measurements were used: (1) the "weighing method" for chemical shrinkage measurement, (2) a continuous, rotating method for volumetric autogenous shrinkage measurement (developed at Lafarge Research Center) [Cf. Fig. 4]. The rate of signal acquisition was set at 10 seconds to allow precise drawing of slopes[1]. In this case, it is obvious that autogenous shrinkage coincide very well with chemical shrinkage for the first minutes of hydration. After that moment, while remaining in the same order of magnitude, the rate of autogenous shrinkage slows down to about 95% of the rate of chemical shrinkage. This result was reproduced for the three following w/c, 0.28, 0.3 and 0.35 [Cf. Figs. 4, 5 and 6][2]. A better understanding of this behavior would be very beneficial. If these results reflect the actual behavior of the material (*i.e.* the rate of autogenous shrinkage becomes lower than the rate of chemical shrinkage quite early), it implies that self-desiccation may start very early, just a few minutes after casting, far before setting.

3.2 Mineral percolation threshold

The mineral percolation threshold, previously described as the first solid path within the material, may be determined using ultrasonic technique [18]. According to Boumiz [18], the threshold corresponds to the minimum value in the compressional velocity. At the beginning, compressional waves go through the liquid phase and the decrease in the compressional velocity traduces the increase in the tortuosity of the liquid phase in the capillary porosity as hydration goes on. After the mineral percolation threshold, the waves go progres-

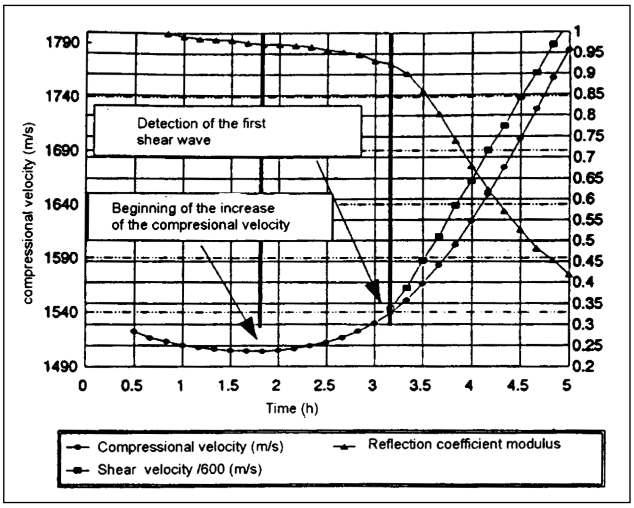

Fig. 7 – Ultrasonic technique (from [18]).

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[1] The use of moving average is nevertheless necessary.

[2] Other experiments have shown variability in the initial slope value (from 0.75 up to 1) depending on the tested mix compositions [7]. The cause of these variations is still not fully understood at that point.

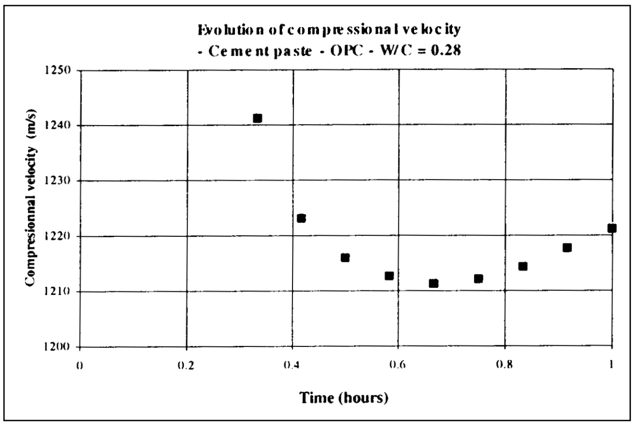

Fig. 8 – Ultrasonic measurement.

sively through the mineral skeleton; the compressional velocity then increases [Cf. Fig. 7][3].

Fig. 8 shows the evolution with time of the compressional velocity of a cement paste (that of Fig. 4). This measurement shows that the mineral percolation threshold appears around 40 minutes after casting. This point corresponds quite well with the time when autogenous shrinkage and chemical shrinkage start do diverge [Cf. Fig. 4].

3.3 Suspension-solid transition

The suspension-solid transition can be determined on curves such as those presented in Figs. 4 and 6, at the time when the slope of the curve drastically decreases. Fig. 9, which represents results of Fig. 6 on a larger scale, shows the change in slope associated with the suspension-solid transition (about 80% decrease). It appears from Fig. 9 that after the suspension-solid transition, the amplitude of the apparent volume variations – autogenous shrinkage – is very small in comparison with the intensity of the absolute volume variations – chemical shrinkage – (about 15%). The mechanism responsible for these apparent volume variations is the capillary pressures induced by the apparition of the gaseous phase within the material. This mechanism is thoroughly described by Hua [17].

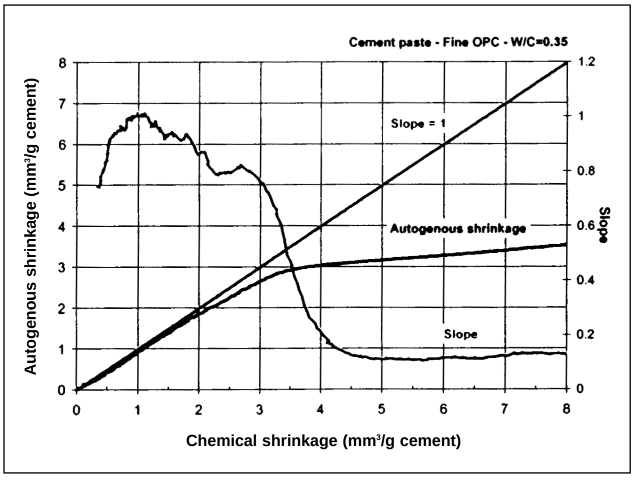

Fig. 9 – Autogenous versus chemical shrinkage – the suspension-solid transition occurs when the slope of the curve drastically decreases.

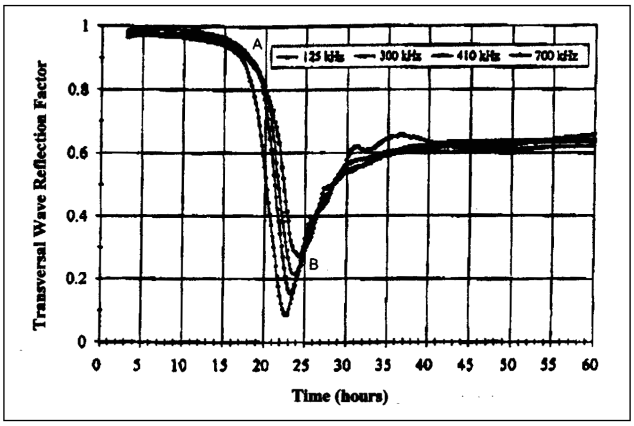

Fig. 10 – Transversal Wave Reflection Factor at different frequencies as a function of time – Reactive Powder Concrete type of mix design [20].

Assessment of suspension – solid transition by acoustic technique

According to recent work done with an ultrasonic technique [20], it is possible to monitor the solid percolations in 1D (mineral percolation threshold) and 3D (suspension-solid transition) through the evolution of the Transversal Wave Reflection (TWR) factor at different frequencies. For Morin, 1D percolation should correspond to the moment when the TWR factor starts to be dependent on the frequency used (point A in Fig. 10). On the contrary, 3D percolation should correspond to the moment when the TWR factor is not dependent anymore on the frequency (point B in Fig. 10).

Suspension – solid transition versus Vicat needle setting time

Initial and final set are measured in a standardized way through the penetration of a needle within the cement paste, until it reaches specified values. Such measurements have been performed on the pastes tested for chemical and autogenous shrinkage, according to the NF EN 196-3 standard (but using the w/c of interest, instead of the w/c corresponding to a standardized consistency). This was done in order to define what is refered to as the suspension-solid transition in comparison with a more standardized definition of setting [7]. Fig. 11 presents the result of such a comparison for the example of Figs. 6 and 9. It appears on Fig. 11 that the values

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(3) It would be theoretically possible to associate the mineral percolation threshold with the detection of the first shear wave crossing the material. However, due to dissipation of the wave within the matrix, the first shear wave is detected only after the percolation threshold (the sensor is not sensitive enough to detect such an attenuated signal).

Table 2 – Suspension-solid transition versus initial and final set values by Vicat needle [7].

	Fine OPC – SSB = 5355 cm²/g			SSB = 3385 cm²/g	
	W/C = 0.3	W/C = 0.35	W/C = 0.4	W/C = 0.3	W/C = 0.4
Suspension-solid transition	2.9h	3.2h	3.8h	3.8h	7h
Initial set (Vicat needle)	2h	2.7h	3h	3.2h	4.7h
Final set (Vicat needle)	3.3h	4h	5.6h	4.5h	8.7h

obtained for initial and final set correspond to a lower and upper bound of the suspension solid-transition taken at the knee of the curve. In our case, this value corresponds to 3.2 hours after the water-cement contact, and to about 4% in terms of degree of hydration. Similar results have been obtained on different mixes, as illustrated in Table 2.

It appears from this table that the suspension-solid transition occurs earlier for low w/c or for finer cements. These trends are easily conceivable, since both a decreased w/c or an increased fineness should lead to earlier and more uniformly distributed solid-solid contacts.

3.4 Capillary pressure and viscoplasticity of cement paste

As shown by micromechanical analysis [21] and recently confirmed by microindentation testing [22], creep is a specific characteristic of cement pastes, mostly dependent on the C-S-H phase. Aggregates (including non-pozzolanic fillers and residues of non-hydrated clinker grains) shall be considered as elastic inclusions, excepted some of them, like sandstone and some granites, which exhibit a few creep. This small creep may be important if these inclusions represent an important solid volume fraction. Creep tests on pure cement paste exhibit delayed strains which are 5 to 10 times higher than concrete ones [23]. These deformations are rather not recoverable (as observed after unloading) and cement paste has to be considered as highly viscoplastic. Thus, resulting creep values of concrete as well as the shape of its curves can be predicted by using micromechanics modelling [24, 25].

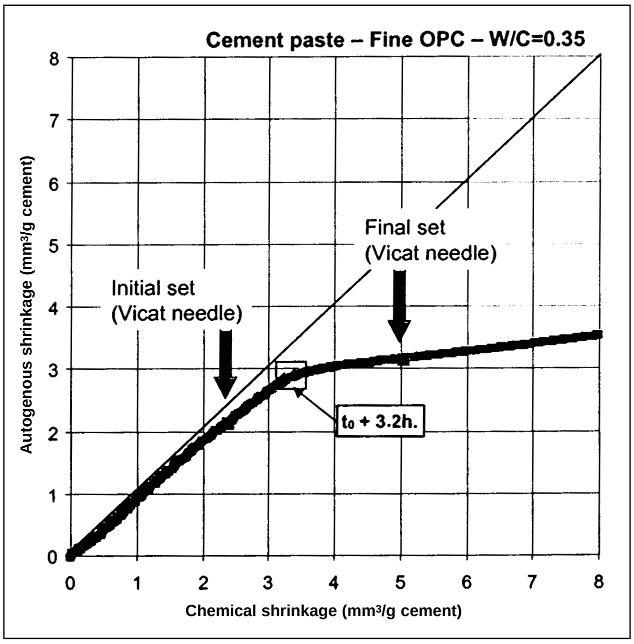

Fig. 11 – Suspension-solid transition versus Vicat needle penetration [7].

The direct consequence is that, during hardening of concrete, hydration consumes a higher volume of water than the additional volume of solid created, and all aggregates (including non-pozzolanic fillers and residues of non-hydrated clinker grains) become progressively compressed, in a quasi-hydrostatic compressive state. The C-S-H phase is mainly in tension, more precisely under a 3-D *deviatoric* stress tensor (2 directions in tension, 1 in compression) and creep occurs as a viscous *shear* strain in the C-S-H layers (*leaf sliding,* which plays the same role as dislocation migration at grain interfaces in metals).

This complex pattern of *eigenstresses* was considered for explaining long-term shrinkage, which rate clearly cannot be explained by the long-term hydration rate, but appears as a form of creep due to capillary pressure [26]. This 3-D *(tension-tension-compression)* stress field leads to shear strain in C-S-H binder which reduces quite uniformly the distance between grain surfaces and thickness of the binder layer which separate two adjacent grains. As this thickness is not uniform and highly scattered, stress concentration or even punching can in some

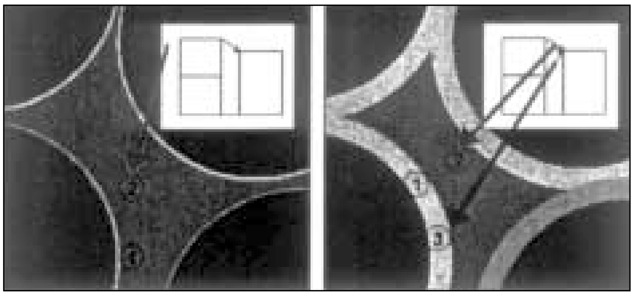

Fig. 12 – Chemical shrinkage as the driving force of a potential swelling. On the left : 1) Anhydrous cement particles dissolve from the surface and 2) C-S-H and CH precipitates in the capillary porosity. The absolute volume decreases.
On the right : 1) Anhydrous cement particles dissolve from the surface, 2) the calcium ions diffuse through the C-S-H layer and portlandite precipitates in the capillary porosity but 3) the C-S-H precipitates where anhydrous dissolve. Even if the overall absolute volume decreases, locally, the solid volume increase may induce swelling stresses between cement particles.

points drastically increase with time, progressively concentrating the stress distribution to a diffuse network of granular pathways.

Creep curves obtained on pure cement paste can provide the elastoplastic constitutive law of C-S-H phase for modelling this micromechanical process, as well as the effect of a constant (external) loading, *i.e.* creep. But the major effect of such an external loading (deviatoric, as the applied solicitation is never hydrostatic) is probably increasing the scatter (disorder and amplitude of stress range of values) of the stress values in C-S-H layers.

3.5 Swelling phenomenon

Swelling phenomenon in autogenous shrinkage experiments is quite often caused by re-absorption of bleeding water [2, 3]. Other mechanisms such as crystallization pressure of growing hydration products such as ettringite or portlandite have also been described [27, 28]. Such mechanisms have been suggested because it was thought that chemical shrinkage that leads to an absolute volume reduction could not explain swelling. Actually it is not the case. At the beginning of hydration, anhydrous cement dissolves from the surface of the grain and combines with water to precipitate [Cf. Fig. 12, left side]. When the layer of C-S-H becomes sufficiently thick, the hydration becomes diffusion controlled [29] and a more or less porous layer of hydrates covers the surface of the anhydrous. It has been reported [30] that during this period, several C-S-H formation mechanisms are possible and, among them, topochemical reaction of C_3S [31] [Cf. Fig. 12, right side]. Water has to go through the C-S-H porous layer to allow crystallization of C-S-H in the given volume of dissolved C_3S. It is also reported that portlandite certainly crystallizes outside the C-S-H layer [31]. Table 1 shows that the volume of the formed C-S-H is larger than the initial volume of C_3S (according to these data, the volume of C-S-H represents 155% of the volume of the anhydrous). The fact that topochemical reaction of anhydrous product may lead to swelling has been already proposed [30]. This "crystallization" of inner C-S-H is likely to impose stresses in the overlapping C-S-H layer [Cf. Fig. 12] and may finally lead to swelling.

4. CONCLUSION

This article gives an overview of the physical mechanisms involved in autogenous shrinkage. Experimental measurements of absolute and apparent volume variations allowed us to accurately and continuously follow the structuring of the material from a suspension to a porous solid. Two events in this structuring can be distinguished: the mineral percolation threshold and the suspension-solid transition. Though self-desiccation begins at the mineral percolation threshold, its consequences on the apparent volume are clearly visible only after the suspension-solid transition. Both events can also be detected by acoustic techniques. Regarding the long-term component of autogenous shrinkage, the importance of creep in the C-S-H phases under the effect of capillary pressure was also emphasized in this article. In the future, other driving mechanisms (such as percolation of the gaseous phase, mechanisms responsible for autogenous swelling) still need to be more thoroughly investigated.

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