Available online at www.sciencedirect.com

SCIENCE DIRECT®

Cement and Concrete Research 35 (2005) 177–183

CEMENT AND CONCRETE RESEARCH

ELSEVIER

Communication

Autogenous shrinkage of concrete: a balance between autogenous swelling and self-desiccation

Laurent Barcelo[a,*], Micheline Moranville[b], Bernard Clavaud[a]

[a]Lafarge, Laboratoire Central de Recherche, 95 rue du Montmurier, 38290 St. Quentin Fallavier, France
[b]Laboratoire Mécanique et Technologie, Ecole Normale Supérieure de Cachan, 61, Avenue du Président Wison, 94235 Cachan Cedex, France

Received 18 December 2002; accepted 17 May 2004

Abstract

According to physical analyses, the driving force of autogenous shrinkage of concrete is the change in the capillary pressure induced by self-desiccation in its cement matrix. Self-desiccation is caused by the balance between the absolute volume reduction (chemical shrinkage) and the building up of the capillary network. The aim of this study was to quantify the influence of the cement characteristics on the chain of mechanisms leading from hydration to autogenous deformations. Four parameters were selected: (i) for clinker, the amount of C_3A and free lime and the SO_3/K_2O ratio; (ii) for cement, the fineness. To master the experimental area, 16 cements were prepared at the laboratory from pure raw materials. An important number of characterizing techniques were used in the experimental study. Their choice was based on the important parameters drawn from the physical analysis: setting time, suspension–solid transition, hydration kinetics through isothermal calorimetry and nonevaporable water, chemical shrinkage, evolution of relative humidity, capillary porosity and autogenous shrinkage. Using different techniques allowed to determine the precise mechanism of action of each parameter. Results showed that these mechanisms are generally different, even if their macroscopic consequences may be identical. This point will probably be useful for modeling and determining the industrial keys reducing the autogenous shrinkage. The physical mechanisms involved in autogenous deformations were further understood. In particular, this study shows that initial autogenous shrinkage should be considered as a balance between the self-desiccation and an initial swelling phase. The influence of the four parameters considered on this last phenomenon were also characterized.

Keywords: Autogenous shrinkage; Self-desiccation; Chemical shrinkage; Autogenous swelling; Cement hydration

1. Introduction

Autogenous shrinkage of concrete is the deformation occurring at constant temperature with no moisture exchange with the environmental medium. Its basic driving force is chemical shrinkage: As hydration develops, the absolute volume decreases. The beginning of this deformation then starts with hydration, as soon as cement and water are in contact. From an engineering point of view, autogenous shrinkage can be split in two parts: autogenous plastic shrinkage that is generated during the induction period and self-desiccation shrinkage that occurs after setting. This paper focuses on the influence of cement parameters on the early self-desiccation shrinkage of cement pastes at a W/C of 0.3.

2. Driving forces of autogenous shrinkage

Previous works on cement pastes [1,2] showed that autogenous plastic shrinkage was of the same order of magnitude as chemical shrinkage and ended at the suspension–solid transition (Fig. 1) when all the grains are in contact and the apparent volume change is restrained. In other works, this suspension–solid transition has been related to the setting time [3,4].

Following the suspension–solid transition and due to self-restriction, the autogenous shrinkage becomes only a small part of chemical shrinkage. Within the matrix, some

---

* Corresponding author. Tel.: +1-514-738-1202; fax: +1-514-738-1124.
E-mail address: laurent.barcelo@lafarge-na.com (L. Barcelo).

0008-8846/$ – see front matter © 2004 Elsevier Ltd. All rights reserved.
doi:10.1016/j.cemconres.2004.05.050

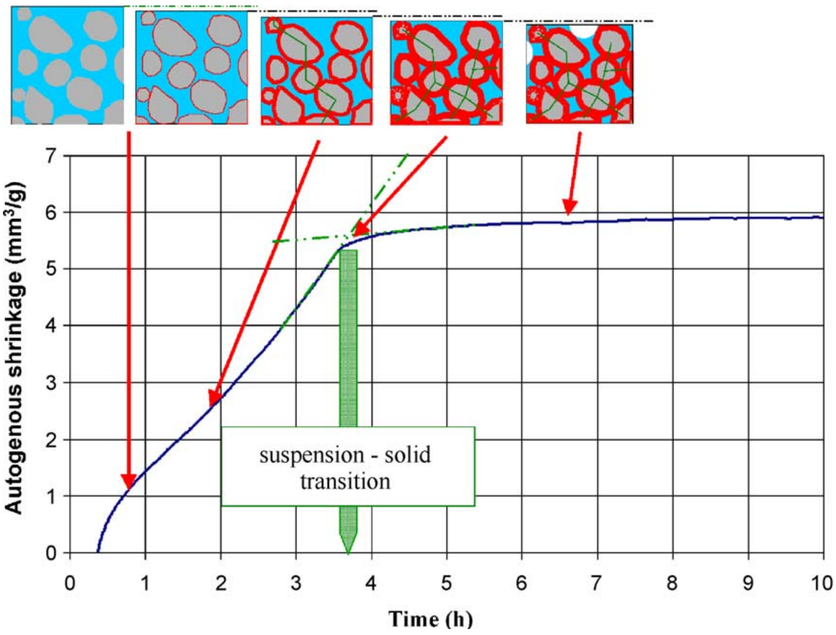

Fig. 1. Autogenous shrinkage of cement paste at W/C = 0.3 [1].

voids corresponding to the difference between the two kinds of shrinkage appear. The self-desiccation that is the balance between this amount of voids and the total porous network increases. Three mechanisms explain the relation between self-desiccation and autogenous deformation in the hardened state, i.e., self-desiccation shrinkage [5]: the variation of (i) capillary pressure, (ii) disjoining pressure and (iii) specific surface of hydrates.

If those three mechanisms can explain qualitatively the self-desiccation shrinkage, only the change in capillary pressure can do it quantitatively [3,5]. According to this mechanism, the self-desiccation results in the formation of 'water–air' meniscus, and Laplace's law indicates the difference in the pressure at each side of the meniscus Eq. (1):

$$P_{\mathrm{gaz}} - P_{\mathrm{liq}} = \frac{2\sigma}{r} = \frac{2\sigma\cos(\alpha)}{R} \quad \text{for spherical meniscus} \qquad (1)$$

where P_{gaz} and P_{liq} are, respectively, the pressures in the air and in the water, σ is the surface tension of water, r and R are, respectively, the radius of the meniscus and of the capillary and α is the wetting angle of the solid with water.

This equation shows that, when self-desiccation increases, i.e., when the meniscus appears in smaller pores, the pressure in water decreases. Thus the self-desiccation shrinkage is the viscoelastic response of the material.

According to Kelvin's law, Eq. (2), a given capillary pressure is equivalent to a specific relative humidity. The monitoring of the evolution of the relative humidity within the matrix is then similar to the monitoring of the capillary pressure.

$$P_{\mathrm{gaz}} - P_{\mathrm{liq}} = -\frac{\mathrm{RT}\rho}{M}\ln(H_{\mathrm{R}}) \qquad (2)$$

Where R is the ideal gas constant, T is the temperature, ρ is the specific gravity of water, M is the molar mass and H_{R} is the relative humidity.

The overall mechanism can be summed up as proposed in Fig. 2.

The aim of this paper is to characterize the influence of four cement characteristics on early self-desiccation shrinkage and to describe their effects on the last part of the above mechanisms. The chosen parameters are:

1. the fineness of the cement through the Blaine value;
2. the amount of free lime in the clinker;
3. the amount of C_3A,
4. the sulfate to alkali ratio in the clinker.

The first and third parameters where chosen because previous studies have shown their importance (e.g., Ref. [6]). The fourth parameter was chosen because it controls the C_3A crystalline form. When SO_3/K_2O ratio is higher

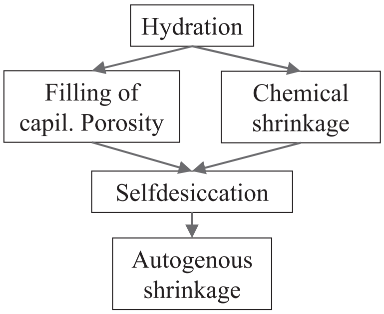

Fig. 2. Accepted mechanism for autogenous shrinkage in the hardened paste.

Table 1
Chemical composition of the 16 model cements

	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	SO_3	TiO_2	Mn_2O_3	P_2O_5	L.O.I.
Cement #1	22.78	4.22	3.61	67.07	0.24	1.41	0.02	0.4	0	0	0.06	0.39
Cement #2	22.47	4.3	3.56	67.34	0.25	1.41	0.02	0.39	0.03	0	0.05	0.38
Cement #3	22.77	5.64	1.02	68.36	0.24	1.43	0.02	0.43	0.01	0	0.05	0.25
Cement #4	22.45	5.5	0.98	68.44	0.28	1.59	0.02	0.53	0.03	0	0.04	0.34
Cement #5	22.78	4.2	3.62	67.26	0.31	0.9	0.02	0.79	0.03	0	0.04	0.19
Cement #6	22.34	4.22	3.59	67.26	0.32	0.91	0.03	0.81	0.07	0	0.03	0.26
Cement #7	22.75	5.56	1.01	68.63	0.33	0.86	0.02	0.71	0.03	0	0.04	0.26
Cement #8	22.53	5.56	0.99	69.03	0.33	0.81	0.02	0.69	0.04	0	0.03	0.26
Cement #9	22.78	4.22	3.61	67.07	0.24	1.41	0.02	0.4	0	0	0.06	0.39
Cement #10	22.47	4.3	3.56	67.34	0.25	1.41	0.02	0.39	0.03	0	0.05	0.38
Cement #11	22.77	5.64	1.02	68.36	0.24	1.43	0.02	0.43	0.01	0	0.05	0.25
Cement #12	22.45	5.5	0.98	68.44	0.28	1.59	0.02	0.53	0.03	0	0.04	0.34
Cement #13	22.78	4.2	3.62	67.26	0.31	0.9	0.02	0.79	0.03	0	0.04	0.19
Cement #14	22.34	4.22	3.59	67.26	0.32	0.91	0.03	0.81	0.07	0	0.03	0.26
Cement #15	22.75	5.56	1.01	68.63	0.33	0.86	0.02	0.71	0.03	0	0.04	0.26
Cement #16	22.53	5.56	0.99	69.03	0.33	0.81	0.02	0.69	0.04	0	0.03	0.26

than 1 in the clinker, K_2O is mainly combined with SO_3. Nevertheless, when this ratio is lower than 1, only a part of K_2O can be combined with SO_3. The remaining part is in solid solution in the crystal structure of clinker phases, mainly in C_3A, ferrite and belite. An important amount of K_2O in C_3A stabilizes the orthorhombic form. A low amount of K_2O stabilizes the cubic form.

The second parameter was chosen because it is well known that free lime is able to generate swelling [7].

3. Materials and experiments

3.1. Materials

To estimate the real influence of the four selected parameters, 16 model cements were prepared in the laboratory from pure raw materials to avoid the role of foreign elements present in the raw materials extracted from quarries. This avoids all the hidden parameters that can be created by the differences in the process of manufacturing of industrial cements. The conception, manufacturing and control of those model cements are described in Ref. [1].

The chemical composition of the 16 model cements is given in Table 1. The low level of impurities content is a consequence of the use of pure raw materials. Nevertheless, the presence of solid solutions has been validated by microprobe analysis during SEM experiments [1].

The value of the four parameters for each cement is given in Table 2. The quantitative analysis of C_3S and C_2S was done under an optical microscope. The total amount of aluminate phases (C_3A+C_4AF) was thus deduced. Because it was not easily distinguished under microscope, the C_3A-to-C_4AF ratio was estimated through Bogue equations.

Table 2
Mineralogical composition and value of the four parameters for the 16 model cements

	C_3S	C_2S	C_3A	C_4AF	free CaO	SO_3/K_2O in clinker	Blaine fineness (m^2/kg)
Cement #1	72	10	5 🟦	11	0.4 🟦	0.33 🟦	317 🟦
Cement #2	74	9	5	11	1.3 🟧	0.33	314
Cement #3	75	8	13 🟧	3	0.4 🟦	0.35	287
Cement #4	79	5	13	3	1.2 🟧	0.39	306
Cement #5	72	13	5 🟦	11	0.7 🟦	1.03 🟧	309
Cement #6	70	11	5	11	1.5 🟧	1.05	315
Cement #7	76	9	13 🟧	3	0.6 🟦	0.97	290
Cement #8	75	9	13	3	1.5 🟧	1.00	299
Cement #9	72	10	5 🟦	11	0.4 🟦	0.33 🟦	408 🟧
Cement #10	74	9	5	11	1.3 🟧	0.33	390
Cement #11	75	8	13 🟧	3	0.4 🟦	0.35	381
Cement #12	79	5	13	3	1.2 🟧	0.39	391
Cement #13	72	13	5 🟦	11	0.7 🟦	1.03 🟧	377
Cement #14	70	11	5	11	1.5 🟧	1.05	414
Cement #15	76	9	13 🟧	3	0.6 🟦	0.97	377
Cement #16	75	9	13	3	1.5 🟧	1.00	399

Table 3
High and low values taken by the four parameters

	SO_3/K_2O in clinker	C_3A	Free CaO	Blaine Fineness
High value	1.01	13.1	1.4	392
Low value	0.35	5.1	0.6	305

Fineness in m^2/kg.

Sixteen cements were prepared from the eight clinkers ground at different finenesses. The C_3A orthorhombic form for the low values of SO_3/K_2O in the clinker was characterized by XRD, SEM and optical microscopy [1].

The values taken by each of the four parameters can be qualified by high or low value (Table 3).

Only pure cement pastes at a water-to-cement ratio of 0.3 were studied.

3.2. Mixing procedure

Cement pastes were prepared in a kenwood mixer, with a 2.5-min mixing procedure [1]. Just after mixing, the cement pastes were deaired in a mixer at a pressure of 100 mbars.

3.3. Experiments

Three typical experiments monitored autogenous shrinkage and self-desiccation at early age: volumetric autogenous shrinkage, linear autogenous shrinkage and relative humidity measurements.

Volumetric autogenous measurement starts 30 min after casting. The cement paste is poured into a latex membrane and sealed, then put in a rotating device, avoiding the bleeding effects. Previous results showed that latex membranes did not completely prevent moisture exchange. A correction to this artifact has been proposed [1].

For linear autogenous shrinkage measurement [1,3], the cement paste is poured in PTFE cylinders of 2-cm diameter and 16-cm height. The samples are rotating after casting to avoid bleeding. Samples are then removed from the mold and wrapped in two layers of aluminum autoadhesive foils. Measurements of linear autogenous shrinkage start 9 h after the beginning of mixing.

Relative humidity measurements are done in an apparatus developed by Boivin [3]. The cement paste is poured in a specific device allowing the measure of the relative humidity 30 min after casting. During the measurement, a strict temperature control is necessary to obtain good-quality results. The small variations of temperature remaining can be corrected through calculation.

4. Results and discussion

The comparison between self-desiccation and autogenous shrinkage has been carried out on results from relative humidity measurements and linear autogenous shrinkage. Figs. 3 and 4 plot, respectively, linear autogenous shrinkage versus relative humidity at ages of 50 and 100 h after casting. Figs. 5 and 6 plot, respectively, the increase of linear autogenous shrinkage versus the loss of relative humidity in the ranges of ages 20–50 and 50–100 h.

It is obvious that a clear relation cannot be established between self-desiccation and autogenous shrinkage at very early age. Here, this relation starts to be detected only after 50 h. Therefore, self-desiccation does not appear to be the sole driving force of autogenous shrinkage during the first or the two first days.

The volumetric autogenous shrinkage measurement method was used to monitor the very early age shrinkage,

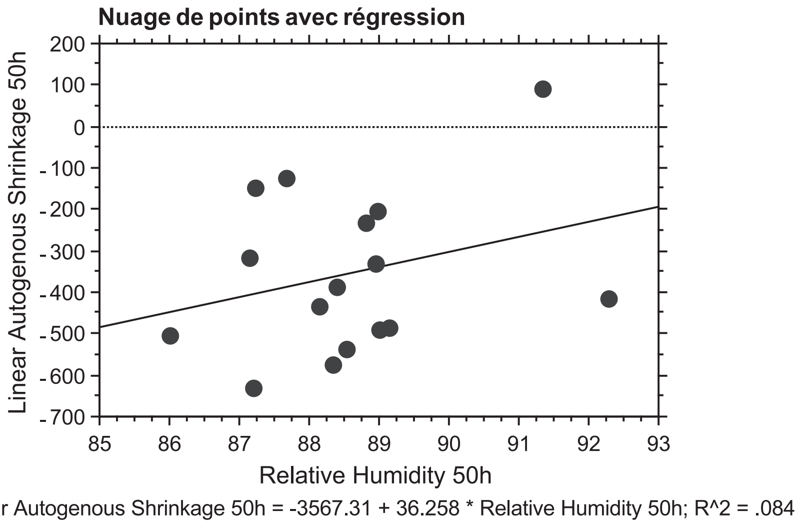

Fig. 3. Linear autogenous shrinkage (μm/m) vs. relative humidity for the 16 cement pastes at 50 h after mixing.

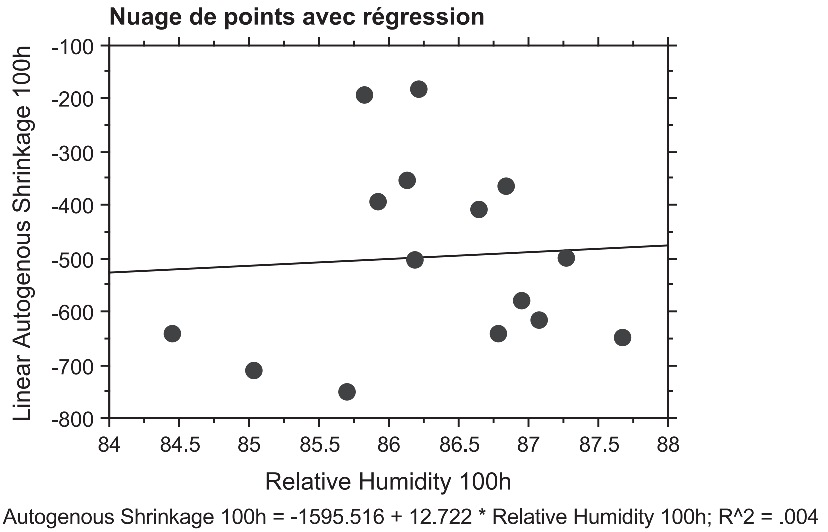

Fig. 4. Linear autogenous shrinkage (μm/m) vs. relative humidity for the 16 cement pastes at 100 h after mixing.

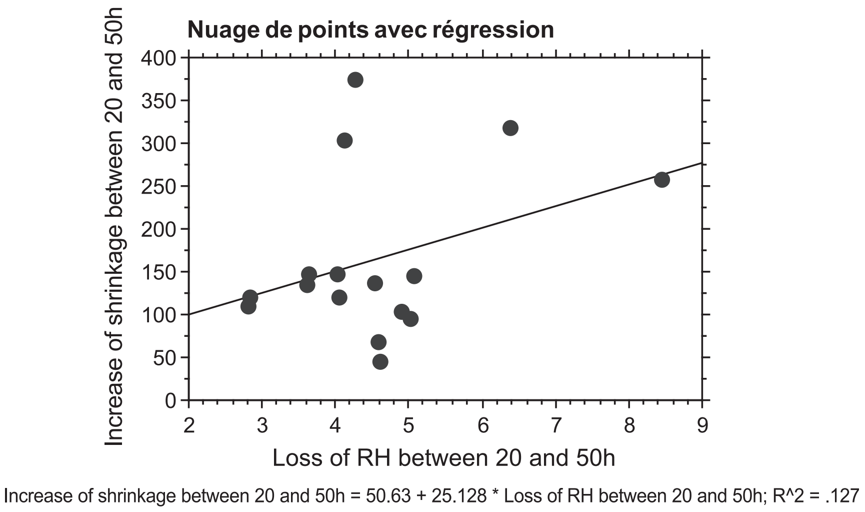

Fig. 5. Increase of linear autogenous shrinkage (μm/m) vs. loss of relative humidity for the 16 cement pastes between 20 and 50 h after mixing.

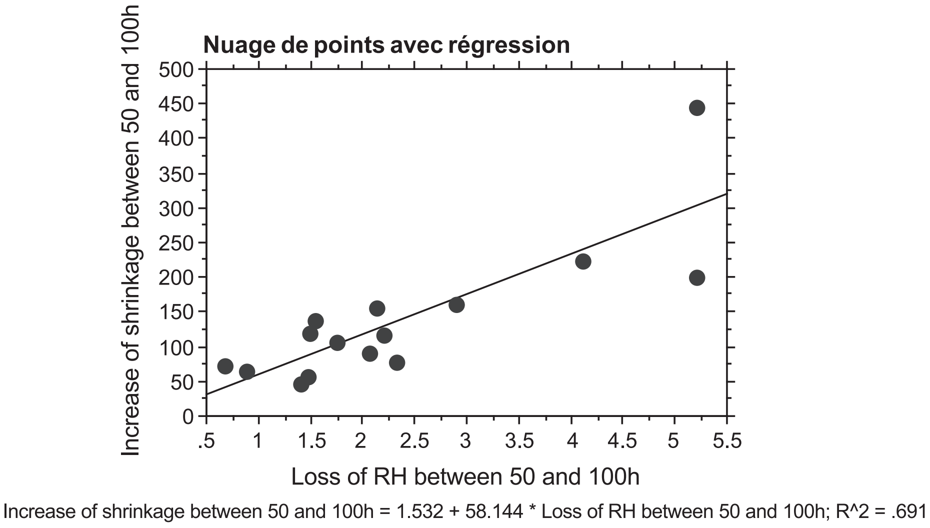

Fig. 6. Increase of linear autogenous shrinkage (μm/m) vs. loss of relative humidity for the 16 cement pastes between 50 and 100 h after mixing.

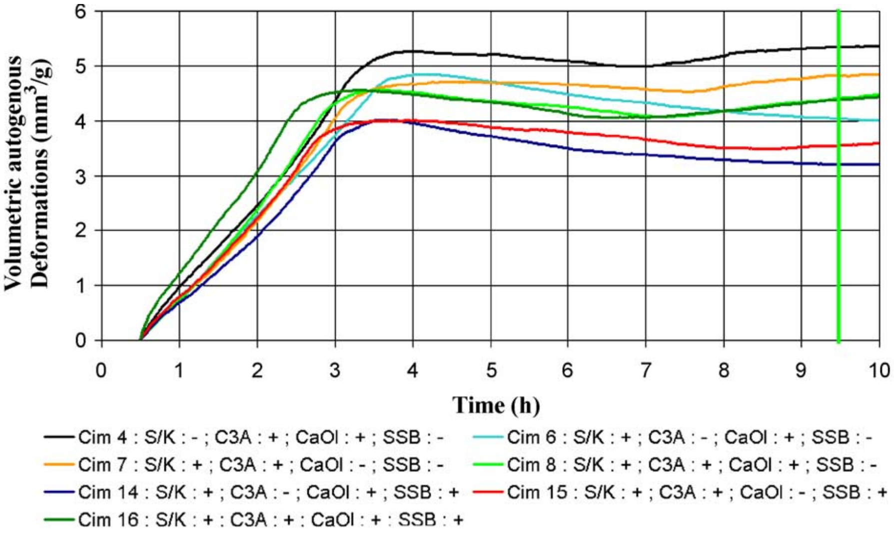

Fig. 7. Volumetric autogenous shrinkage from the casting, for the cement pastes undergoing swelling after setting.

just after setting. Results show that a lot of cement pastes have swollen for several hours (Fig. 7). Moreover, the variance analysis shows that this swelling is not distributed randomly but can clearly be related to the amount of free lime and to the sulfate to alkali ratio of the clinker (Fig. 8). The highest values of $SO_3/K_2O_{clinker}$ ratio and highest values of free lime both lead to a decrease of initial shrinkage or to an increase of initial swelling. Events clearly appear as if this initial autogenous deformation was a balance between an initial autogenous swelling and the self-desiccation, the relative importance of the swelling being stronger at the beginning and decreasing later on.

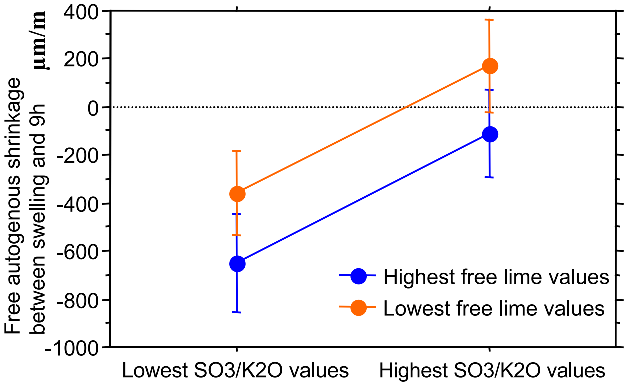

Fig. 8. Very early autogenous deformations for different values of SO_3/K_2O ratio in the clinker and free lime.

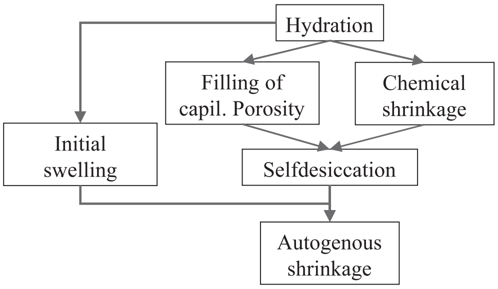

Fig. 9. Proposed mechanism of autogenous shrinkage at early age.

The mechanism of autogenous shrinkage at an early age can be summed up in Fig. 9. Initial autogenous swelling was seen on cement paste at higher W/C [8,9]. The fact that this was not measured at lower W/C can be explained by:

- The fact that, if autogenous shrinkage is a balance between self-desiccation and initial swelling, we can only measure the resulting deformation. As W/C decreases, the relative importance of self-desiccation becomes more important;
- As the initial swelling occurs just after the suspension–solid transition, only volumetric measurements can monitor them. Previously, those measurements were suffering from important artifacts that made the results difficult to analyse.

5. Conclusion

It is clear that autogenous shrinkage mechanism cannot be only explained by the physical mechanism of self-desiccation induced by the volume balance of hydration. It seems that the creation of the early hydrated products results in an autogenous swelling phenomenon that decreases with time. For modeling, the early volume changes has to be seen as a coupling between the chemistry of hydration and physics of water. The sulfate-to-alkali ratio of the clinker and the amount of free lime seems to change the intensity of this swelling phase.

References

[1] L. Barcelo, Influence des caractéristiques des ciments sur la structuration et le comportement dimensionnel des matériaux cimentaires au jeune âge, Ecole Normale Supérieure de Cachan, Cachan, France, 2001.

[2] L. Barcelo, S. Boivin, P. Acker, J. Touin, B. Clavaud, Early age shrinkage of concrete: Back to physical mechanisms, Concr. Sci. Eng. 3 (10) (2001) 85–91.

[3] S. Boivin, Retrait au jeune âge du béton: développement d'une méthode expérimentale et contribution à l'analyse physique du retrait endogène, Ecole Nationale des Ponts et Chaussées, 1999.

[4] H. Justness, P. Clemmens, P. Depuydt, D. Van Gemert, E.J. Sellevold, Correlating the deviation point between external and total chemical shrinkage with setting time and other characteristics of hydrating cement paste, Shrinkage of Concrete, RILEM, 2000.

[5] C. Hua, P. Acker, A. Ehrlacher, Analyses and models of the autogenous shrinkage of hardening cement paste: 1. Modelling at macroscopic scale, Cem. Concr. Res. 25 (7) (1995) 1457–1468.

[6] E. Tazawa, S. Miyazawa, Influence of cement composition on autogenous shrinkage of concrete, 10th International Conference on Cement Chemistry, Goteborg, 1997.

[7] H. Le Chatelier, Sur les changements de volume qui accompagnent le durcissement des ciments, Bull. Soc. Encourag. Ind. Natl. 5 (5) (1900) 54–57.

[8] A. Kheirbek, Influence des paramètres de formulation sur les retraits endogène et de dessiccation de la pâte de ciment, Université de Cergy-Pontoise, 1999.

[9] V. Baroghel-Bouny, A. Kheirbek, Effect of mix-parameters on autogenous deformations of cement pastes. Microstructural interpretations, Shrinkage of Concrete, RILEM, 2000.