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Autogenous deformation and RH-change in perspective

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Abstract

Autogenous deformation and change of the relative humidity (RH-change) have been described and registered for a century. However, it is only within the last decade that these phenomena have received appreciable attention. The reason for this is that autogenous deformation and autogenous RH-change are phenomena of special importance within high-strength (high-performance) concrete technology, and a significant utilization of these concretes did not take place until the early 1980s. In the present paper an historical overview of autogenous deformation and RH-change is given. In addition, due to the present status of this research field both terminology and measuring techniques are described in detail. Finally, some expectations for future research in this field are given.

Keywords: Shrinkage; Humidity; High-Performance Concrete; Self-Desiccation

1. Introduction

A characteristic feature of high-strength concrete is a low porosity and a discontinuous capillary pore structure of the cement paste. This is encompassed by keeping a low water-cement ratio with the aid of superplasticizers and by adding silica fume to the mixture. From a material point of view these modern concretes generally possess some highly advantageous properties compared to traditional concrete. Examples of these include good workability in the fresh state, higher strength and improved durability. However, these types of concrete have also proved to present some problematic properties, such as autogenous deformation and RH-change.

Potentially, these properties may be used beneficially. Autogenous RH-change may improve the frost resistance and may be used to shorten the time until an impervious, moisture sensitive covering can be applied to the concrete. Correspondingly, autogenous deformation may cause micro-cracking, which may increase the tensile strain capacity and have a stress equalizing effect. Furthermore, autogenous deformation may lead to strength increase due to clamping pressure on aggregates or incorporated fibres [1]. However, these benefits are, generally, considered secondary to the negative effect of

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cracking due to autogenous deformation. Micro-cracks due to restrained autogenous shrinkage may connect into a continuous crack pattern and form macro-cracks. Such cracks constitute a serious problem with regard to strength, durability and aesthetics.

No contemporary, international review paper exists on autogenous deformation and RH-change. A good overview of research on autogenous deformation carried out in Japan is given in a report from the Japan Concrete Institute [2]. A comprehensive state-of-the-art-report which was published in 1981 by a RILEM commission [3] is outdated.

2. Precursors through curiosity – milestones in the past

A very early description dates back to the year 1900. One of the pioneers of cement research, Le Chatelier, describes self-desiccation and commences a systematic registration of the material properties of cement paste, see Figure 1. He states that it is fundamental to distinguish between the *absolute volume* and the *apparent volume* of a hardening cement paste [4]. With the terminology used in this paper (cf. section 4.2) a change in the absolute volume means chemical shrinkage, and a change in apparent volume means autogenous deformation.

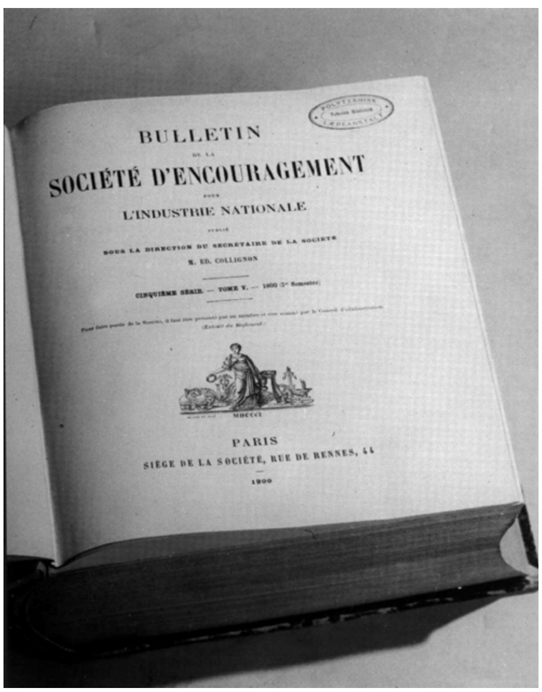

Figure 1. In the year 1900 Le Chatelier published a paper [4] which dealt with the volume change of a hardening cement paste: *Sur les changements de volume qui accompagnent le durcissement des ciments*. Autogenous deformation is covered by the descriptions given by Le Chatelier in this paper. Since then this phenomenon has been treated regularly in the literature.

Early reported measurements can be found in the literature from the beginning of the 1900s. In 1927, Jesser [5] reported measurements of autogenous RH-change for cement mortars with w/c-ratios of 0.24-0.36 and noted that the internal relative humidity may be 90% after 1 month of hardening. In 1928, Neville and Jones [6] described an apparatus for measurement of volumetric deformations of cement paste during sealed hardening at a constant temperature. In 1934, Lynam [7] was perhaps the first to define the term *autogenous shrinkage*; shrinkage which is not due to thermal causes or to loss of moisture to the air.

In 1940, Davis [8] published experimental results for autogenous deformation. He observed that the magnitude of autogenous shrinkage is in the range 50-100 µstrain after five years of hardening. Compared with thermal deformations and drying shrinkage this autogenous shrinkage is relatively small. For this reason little attention has been paid to autogenous deformation in both concrete practice and research for many years.

As a result of Powers' pioneering research [9] around 1940-1950, it became possible to describe and perform theoretical calculations of the phase composition of a hardening cement paste. This enabled Copeland and Bragg [10] and Powers [11] to perform a calculative analysis of self-desiccation measurements. It was realised that at sufficiently high w/c-ratios self-desiccation will not take place. For pure water-cement pastes undergoing sealed hydration appreciable self-desiccation will only occur if the w/c is below 0.40-0.45.

Further insight was given by L'Hermite et al. [12] and Buil [13]. Based on a comparison of normal drying shrinkage with autogenous deformation they showed that self-desiccation shrinkage, which is a result of chemical shrinkage, is the major cause of autogenous deformation.

In 1981 a RILEM commission presented an elaborate exposition of the properties of set concrete at early ages [3]. Measuring techniques and data analysis of autogenous deformation were also thoroughly covered by this state-of-the-art-report. With respect to cracking at early ages, the summary and suggestions for further research focus on plastic shrinkage and thermal deformation and do not include autogenous deformation.

3. Successors out of necessity – the HSC/HPC era

During the early 1980s a significant progress in concrete technology started: High-Strength / High-Performance concrete (HSC/HPC). This was partly enabled through the development of effective superplasticizers which made it possible to produce workable concretes with low w/c-ratios. However, the real breakthrough for the high-strength concrete technology was due to the arrival of silica fume. In less than 5 years, silica fume was accepted as a supplementary cementitious material, and compressive strengths of workable concretes of 100-150 MPa could be produced [14]. Today, silica fume appears as the de facto basis for high-strength/high-performance concrete technology.

With respect to self-desiccation these new types of concrete are considerably different from traditional concretes. This is due to both microstructural and chemical factors. The tendency of a concrete for self-desiccation is controlled by:
- the *chemical shrinkage* accompanying the cementitious reaction,
- the *sensitivity to the relative humidity* of the cementitious reaction,
- the *pore structure* of the concrete.

Low w/c-ratio and silica fume addition lead to a significant refinement of the pore structure. Compared with the hydration of Portland cement, the pozzolanic reaction of silica fume has a high chemical shrinkage and is relatively insensitive to a drop in the relative humidity [15]. All these factors promote self-desiccation and self-desiccation shrinkage. In addition, high-strength concretes also have a higher binder content per unit volume, which further increases shrinkage.

Despite effective control of both water-loss and temperature variations during hardening, cracking in these high-strength concretes was reported from concrete practice in the mid 1980s [16,17]. Subsequent laboratory investigations in the late 1980s clearly showed that the observed cracking phenomena were due to an increased autogenous deformation of high-strength concretes [18]. However, this did not become commonly recognized until the mid 1990s when a significant increase in the research on autogenous deformation and RH-change took place. Since 1996, international workshops entirely devoted to this subject have been held annually [19-23].

Figure 2 illustrates this rendering. Note the 10 years' delay from the introduction of high-strength concrete until the importance of research in autogenous deformation and RH-change was generally accepted.

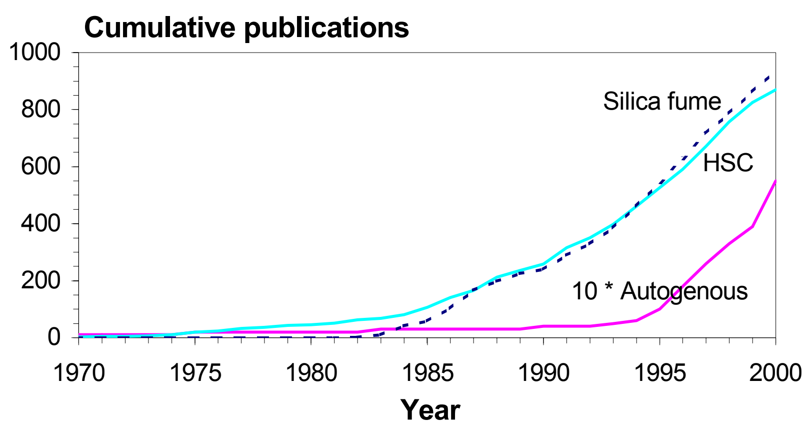

Figure 2. Number of publications registrered by Computerized Engineering Index [24] within different subfields of concrete technology: 1) Silica fume, 2) High-strength concrete (HSC) and 3) Autogenous deformation and RH-change. The number of publications within the latter subject has been multiplied by 10 for easy comparison. In the figure Compendex is used to illustrate the apperance and growth of different subfields of concrete technology - not to illustrate the complete amount of literature.

An important contribution to the understanding of self-desiccation and self-desiccation shrinkage was given by Hua et al. [25]. Based on measurements of autogenous deformation, degree of hydration and mercury intrusion porosimetry they showed that capillary stresses in the pore water are the dominating physico-chemical phenomenon which links self-desiccation and self-desiccation shrinkage.

4. Present status

Today the total amount of literature on autogenous deformation and RH-change is substantial. More than 100 papers deal entirely with these phenomena. Awareness of the importance of this subject has been established both in the scientific community and in concrete practice; research groups in more than 10 countries are presently investigating these phenomena. This was not the case 10 years ago.

According to measurements presented in the literature, autogenous deformation and RH-change are influenced by many factors, for example w/c-ratio, cement composition, silica fume content, fineness of cement and silica fume, volume of aggregate and exposure temperature. Unfortunately, there is a remarkable lack of agreement on the extent of influence of the different factors. Examples of apparently conflicting results include the applicability of the maturity concept to autogenous deformation [26], whether observed expansion after setting is an artefact [27] and the influence of silica fume addition [27-29].

Strictly speaking, consensus is limited to general characteristics such as:

- Autogenous RH-change (self-desiccation) is not able to proceed below approximately 75% RH.
- Dissolved salts in the pore solution may lower the relative humidity by at least 1-3%.
- A lowering of the w/c-ratio promotes autogenous shrinkage and RH-change.
- After setting, autogenous deformation developed during a couple of weeks hardening may amount to more than 1000 µstrain for a cement paste.
- Cracking may develop during restrained autogenous deformation.

4.1 Measuring techniques

Problems with the measuring techniques seem to be the cause of a large part of the disagreement found in the literature. Barcelo et al. [30] have demonstrated how difficult it is to interpret results based on different measuring techniques. It is fundamental to solve these problems with measuring techniques before a reasonable discussion of the influence of factors on autogenous deformation and RH-change can take place. When measuring results are presented and analysed in the literature, it should not be a matter of question to the reader whether the results were significantly influenced by for example bleeding, temperature variations, moisture loss or external forces. The measuring error may easily exceed the measured quantity if the measurements are not performed very carefully.

4.1.1 Techniques for measuring autogenous deformation

Measurements of autogenous deformation have been carried out in two fundamentally different ways, viz. measurement of volumetric deformation and measurement of linear deformation. Volumetric measurement of autogenous deformation is frequently performed by placing the fresh cement paste in a tight rubber balloon immersed in water.

The change in volume of the cement paste is measured by the amount of water displaced by the immersed sample, for example, by measuring the weight change of the immersed sample (buoyancy). Linear measurement of autogenous deformation is frequently performed by placing the cement paste in a rigid mould with low friction. The length change of the cement paste is recorded by a displacement transducer at the end of the specimen.

Both methods of measurement have advantages and disadvantages. These have been debated for a long time [13,30-33]. One advantage of the volumetric method is the possibility of commencing the measurements immediately after casting. Before setting, measurements have to be volume based since the plastic state prevents an unabiguous definition of length. In contrast, the lack of a steady contact between the rubber balloon and the cement paste is a considerable disadvantage of the volumetric method. A film of water, e.g. due to bleeding, or entrapped air at the surface of the cement paste may obstruct this contact significantly. During the hydration process the water or entrapped air will be sucked back into the cement paste as a consequence of chemical shrinkage. In this way the internal volume reduction may also be erroneously measured as an outer deformation; the volume of the rubber balloon is the combined volume of the cement paste and the volume of the surface water or entrapped air. Since the chemical shrinkage is considerably larger than the autogenous deformation, this may lead to a substantial error. In addition, Buil [13] mentions that the pressure caused by a tight rubber balloon could damage the weak structure during setting. Furthermore, volumetric measurement results of autogenous deformation seem to be associated with large scatter.

One advantage of the linear method is the firm anchorage of the measuring points to the set cement paste. This reduces the above-mentioned problems greatly. At the same time, this is a disadvantage of the linear method since the measurements cannot be carried out before the cement paste has set. The linear method has an additional problem: the risk of restraining the cement paste. In the very first hours after setting, the cement paste is too weak to overcome the friction against a rigid mould. However, this problem can be reduced by lubricating the mould.

A special corrugated mould system which combines the advantages of linear and volumetric measurement has been suggested by Jensen and Hansen [33], see Figure 3. Before set, the corrugated mould in fact transforms the volumetric deformation into a linear deformation, and after set a normal, well-defined linear deformation is measured. In this way, it is possible to commence linear measurements directly after casting.

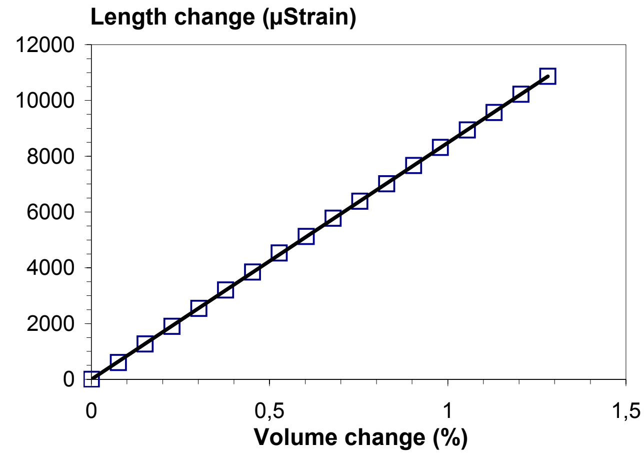

Figure 3. With a special, corrugated mould it is possible to perform linear measurements directly after casting. This is due to a greater stiffness in the radial than in the longitudinal direction of the mould. Before set the volumetric deformation of the cement paste is, therefore, transformed into a linear deformation, as shown in the graph.

Gagné et al. have suggested a volumetric technique where chemical shrinkage and autogenous deformation can be measured simultaneously on the same sample [34].

4.1.2 Techniques for measuring autogenous RH-change

From a point of view of measuring technique, autogenous RH-change has a significant advantage compared with autogenous deformation: The relative humidity is a physico-chemical state which can be measured on a given sample independently of the previous history of the sample. Contrary to this, a reference point has to be defined for autogenous deformation. But apart from that, major difficulties are also connected with measurement of autogenous RH-change.

Measurements of autogenous RH-change have been carried out in many different ways. A typical procedure is to place cement paste in a small, sealed and thermostated container. The internal relative humidity of the cement paste will equilibrate the relative humidity of the air inside the container, which in turn is measured by a humidity sensor. Normally, the cement paste is crushed in order to equilibrate the relative humidity faster. The measurements of relative humidity may be performed continuously or discretely.

Relative humidity can be measured in many different ways. To follow autogenous RH-change, however, only electronic sensors for continuous measurement are relevant. Unfortunately, they need frequent and extensive calibration in order to bring down the measuring accuracy to about ±1% relative humidity. Even for high-quality sensors drift may exceed 1% RH per month. A lack of thermal equilibrium between sensor and sample is also a source of errors. These gradients may be due to insufficient thermostatic control or heat of hydration. Near saturation at room temperature a temperature difference of 1°C will lead to a measuring error of 6% RH. Furthermore, condensation on the sensor may occur if the sample has a higher temperature than the sensor. As long as the condensation persists it will lead to measuring errors, and some sensors may in addition require a new calibration.

Moisture loss from the sample may lead to very significant measuring errors. Cementitious systems prone to self-desiccation will, typically, also be sensitive to moisture loss. This is because such cement pastes normally have a fine pore structure and a low amount of evaporable water. The moisture loss may arise from repeated measurements on the same sample, where the sealing is broken at each measurement, or from invisible cracks in the sealing and the like.

4.2 Terminology

The lack of agreement in the literature not only concerns the measuring techniques, but also the applied terminology. A confusion of nomenclature exists as different terms are used for the same phenomenon, and different phenomena are decribed by the same term [28]. A comparison of recent publications clearly illustrates this [2,27,35,36]. Below, a terminology is suggested based on definitions from physical chemistry, see Figure 4.

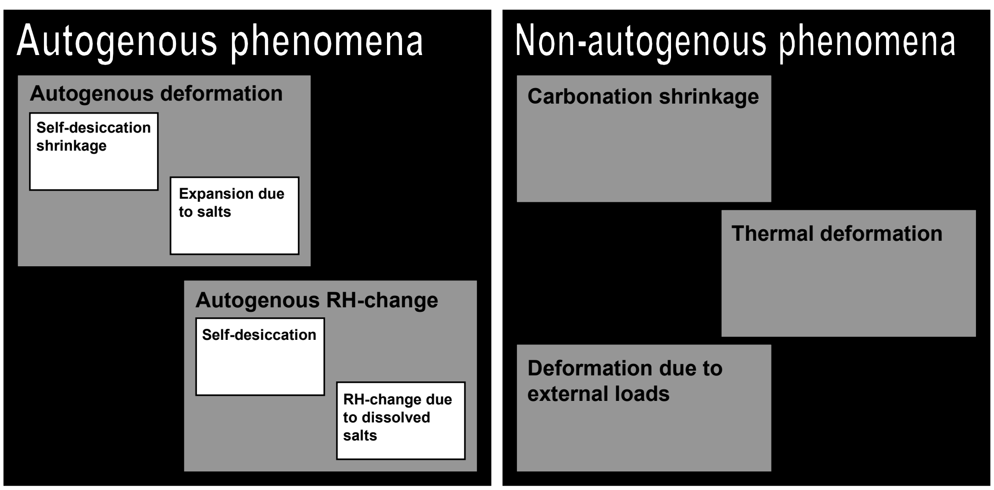

Figure 4. Graphic representation of applied terminology; self-desiccation and self-desiccation shrinkage are proper subsets of autogenous relative humidity change and autogenous deformation, respectively [28].

Chemical shrinkage: Internal volume reduction associated with the hydration reactions in a cementitious material.

Autogenous deformation: The bulk deformation of a closed, isothermal, cementitious material system not subjected to external forces.

Autogenous relative humidity change: The change of internal relative humidity in a closed, isothermal, cementitious material system not subjected to external forces.

Self-desiccation shrinkage: Autogenous deformation of a set cementitious material system caused by chemical shrinkage.

Self-desiccation: Autogenous relative humidity change of a set cementitious material system caused by chemical shrinkage.

4.2.1 Comments on the definitions

To define autogenous conditions, concepts from physical chemistry have been used here: a closed, isothermal system not subjected to external forces. The word *closed* signifies that no exchange of matter takes place between the cementituous material and the surroundings. Most important, exchange of water with the surroundings leads to non-autogenous deformation and non-autogenous RH-change, but other substances such as CO_2 and SO_4^{2-} may also be important in this connection. The word *isothermal* signifies that the temperature is kept constant. Autogenous deformation and RH-change also take place even though the temperature is not constant or when matter is exchanged with the surroundings. But in that case the autogenous phenomena are superimposed and modified due to the external influence. The autogenous phenomena may also be modified due to bleeding. However, the definitions here only concern a homogeneous, isotropic material system.

Chemical shrinkage is the consequence of reaction products occupying less space than the reacting materials in a hydrating cement paste. Before set this internal volume reduction may be completely converted into a bulk deformation of the system, so-called *setting shrinkage*. With the above definition setting shrinkage is a constituent part of autogenous deformation. After set chemical shrinkage creates inner, empty cavities if the cement paste is kept sealed. Due to the formation of menisci, chemical shrinkage leads to an RH-decrease and a shrinkage of the cement paste, i.e. self-desiccation and self-desiccation shrinkage. Parallel with the chemical shrinkage, a change of the pore structure of the cementitious material takes place. As mentioned previously, this has importance for the magnitude of the developed self-desiccation.

Principally, the pore structure may change without a matching chemical shrinkage, e.g. due to the so-called Ostwald ripening of the hydration products. The deformation and RH-change linked to this are included in autogenous deformation and RH-change, but not in self-desiccation and self-desiccation shrinkage. Figure 5 and 6 illustrate some of the phenomena discussed above.

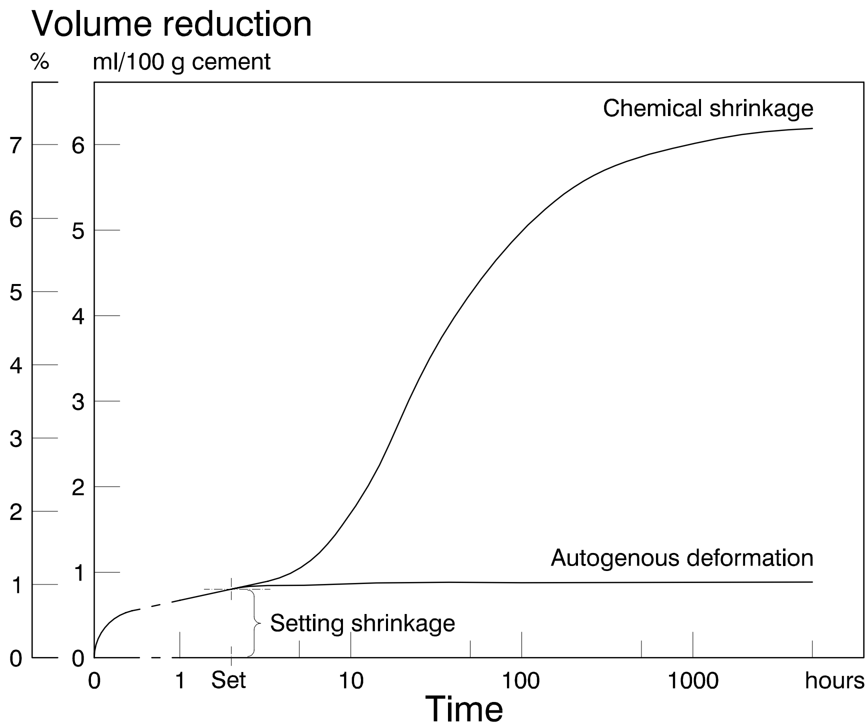

Figure 5. Schematic development of volume changes, which take place in a sealed cement paste system. Before set chemical shrinkage and autogenous deformation may be identical. This volume reduction is referred to as setting shrinkage, and may amount to about 1%. After set the chemical shrinkage may be 50 times larger than the autogenous deformation on a volume basis.

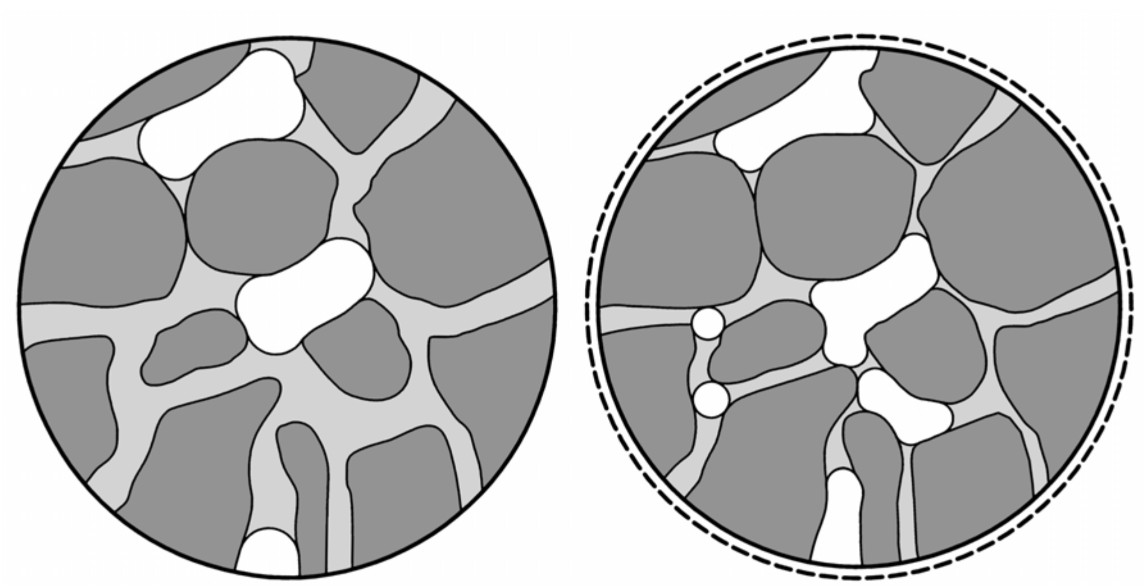

Figure 6. Schematic representation of a cross section of a hydrating cement paste. Left: low degree of hydration, right: high degree of hydration. Solid matter (hydration products, unhydrated cement, silica fume, etc.) is shown dark grey, pore water is light grey and empty pore volume is white. The Figure illustrates the following consequences of the hydration process: a) reduction of the amount of pore water due to binding in the hydration products, b) increase of the amount of solid material, c) refinement of the pore structure, d) formation of empty pore volume due to chemical shrinkage, e) decrease of the radius of curvature of the menisci, and f) bulk shrinkage due to increased tensile stresses in the pore water i.e. self-desiccation shrinkage.

5. Future

We think that the increased attention paid to autogenous deformation and autogenous RH-change will eventually lead to more consensus of nomenclature. Terms which lack logic or do not intuitively describe a specific phenomenon will inevitably disappear from the terminology.

The same fate will apply to the measuring techniques. In the literature there is an ongoing sound discussion, and awareness of the pitfalls and agreement on the usefulness of different measurement techniques must be the outcome of this. Perhaps, a round robin test could be useful in this process.

The major part of the reported research on autogenous deformation and RH-change is phenomenological. The lack of causal explanations limits the applicability of such research. Especially for autogenous deformation and RH-change it is difficult to apply phenomenological research, since these properties are strongly influenced by the experimental conditions and a large number of material parameters. Further research must be based on physical, chemical and thermodynamic examinations of underlying mechanisms for proper understanding of these phenomena and prediction of behaviour.

In some cases an analytical or experimental examination of these mechanisms may be very difficult to apply. However, during the last decade computer modelling has developed into a stong tool for simulation of the chemical and physical aspects of portland cement hydration [37]. Recent application of computer modelling to autogenous deformation and RH-change indicates that this approach may be useful for the understanding and prediction of these phenomena [38].

Limitation of autogenous deformation and RH-change, for example by so-called internal curing, is another topic which may receive increased attention in the coming years. It has been known for a decade that incorporation of saturated lightweight aggregate into the concrete mixture may provide internal curing and, thereby, minimize the autogenous shrinkage [39,40]. However, major problems are connected to this technique, both for the fresh, hardening and hardened concrete. Examples of these problems are: change of consistency, separation of the light porous phase and strength reduction. Moreover, it should be borne in mind that the water curing only limits the autogenous shrinkage. If the relative humidity of the hardened concrete is reduced later on due to equilibration with the surroundings, drying shrinkage will occur. Much additional research in this area is needed.

The importance of autogenous deformation and RH-change for concrete practice seems to be commonly acknowledged today. As a result of this, we expect that a worldwide explicit implementation in building codes will take place. The first indications of this can already be found [35,41].

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