

# ALKALI AGGREGATE REACTION AND DELAYED ETTRINGITE FORMATION: COMMON FEATURES AND DIFFERENCES

Renaud-Pierre Martin <sup>1\*</sup>, Céline Bazin<sup>1</sup>, François Toutlemonde<sup>1</sup>

<sup>1</sup>Ifsttar, Université Paris Est, Bridges and Structures Department, Paris, France

## Abstract

Alkali-Aggregate Reaction (AAR) and Delayed Ettringite Formation (DEF) are both internal swelling processes that can affect concrete. In the field, the two reactions are often observed acting simultaneously. To manage the affected structures, it is necessary to provide numerical tools able to predict the mechanical state of the construction in a reliable way. This implies to have a precise understanding of the two phenomena acting alone or combined. This paper is aimed to point out the common features and the differences between AAR and DEF. The final objective is to determine which concepts can be used for the two processes and which aspects need a separate approach. After a brief review of the literature, results of tests performed at Ifsttar for the two reactions and relative to expansive and mechanical properties are analyzed.

**Keywords:** internal swelling reactions, couplings, expansion, mechanical effects

## 1 INTRODUCTION

Alkali Aggregate Reaction (AAR) and Delayed Ettringite Formation (DEF) are internal swelling processes that can affect concrete. They lead to an expansion of the material and induce generally cracking and degradation of the mechanical properties. This implies problems in terms of serviceability, structural integrity [1,2,3] and durability since cracking favors the ingress of external species prone to initiate other degradations [4,5]. Because of their different chemical mechanisms, these two reactions are mostly studied separately in the literature. A large amount of research has been performed in the last decades for AAR [6,7] and DEF [8,9,10]. However AAR and DEF often act simultaneously in the field. Due to similar macroscopic effects, some controversies remain in the identification of the deleterious causes [11,12]. To deal with the affected structures, it is thus necessary to precisely understand the chemo-mechanical effects of each reaction.

This paper proposes a comparison of AAR and DEF in order to identify their common features and differences, based on the literature and the feedback of studies performed at the French Institute of Science and Technology for Transport and Civil Engineering (Ifsttar, formerly LCPC). The corresponding results can be used to further investigate the macroscopic effects of these processes or to develop numerical tools to re-assess the affected structures [1,2]. Firstly, a literature review is proposed to illustrate some of the available results regarding the swelling mechanisms and their influencing parameters. AAR and DEF effects at the scale of the material are then studied, including their impact on the evolution of the mechanical properties.

## 2 MECHANISMS AND INFLUENCING PARAMETERS

### 2.1 Mechanisms

AAR has been firstly identified many decades ago [13]. Because of the great amount of research done in this field, the basic mechanisms of the reaction are nowadays well known. AAR mainly implies alkali,

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\* Correspondence to: renaud-pierre.martin@ifsttar.fr

hydroxyle and calcium ions as well as the silica contained in the aggregates to form the alkali-silica gel [6]. Due to water adsorption of the gel, expansive forces appear due to osmotic [14] or hydrostatic [15] pressure. The beginning of the expansion takes place within the aggregate containing the reactive silica [4].

DEF has been more recently identified on structures [8]. Its definition used to be very controversial. In this paper, the term of DEF describes any heat-induced internal sulfate attack [16]. The temperature increase generally takes place at early age due to precast processes [8] or to the heat of hydration developed in mass concrete [5]. However, heating in hardened concrete can occur and induce damage [17]. The theories concerning the mechanism of expansion remain quite controversial but always implies the effect of crystallization pressure of ettringite in hardened concrete. Two main theories are proposed: either cement paste expansion [9,16] or expansion at the past-aggregate interface [8,18]. It is also suggested that expansion could be initiated according to the first mechanism and then result of a combination of the two processes [9].

Thus, although the two reactions have similar macroscopic effects, the microscopic causes are very different. If AAR can occur in a whole structure due to the use of a reactive aggregate, it is much less likely for DEF where the necessary temperature elevation is generally restricted to the core of massive elements. It follows that the structural effects are quite different. Due to its mechanism of expansion, DEF is in some research considered to induce isotropic expansion [9], at least at the beginning. On the contrary, AAR is anisotropic [7,14]. Such differences have obviously to be taken into account in the models.

## 2.2 Main influencing parameters

### *Alkali content*

In a concrete, the majority of the alkalis comes from the cement although some can be released from aggregates [4]. Alkalis have a great influence on the development of AAR: not only they are reactants for the gel formation but also they ensure a high internal pH propitious for AAR. The expansion increases with the alkali content [19]. Concerning DEF, the presence of alkalis strongly decreases the stability of ettringite [20], especially when temperature increases [16]. Thus, during the heat treatment necessary for DEF, the presence of alkalis will tend to lower the formation of (non deleterious) primary ettringite and thus release sulfates either in the pore solution [21] or adsorbed by the CSH [20]. These sulfates provide reactants to form ettringite in the hardened material after cooling. The presence of alkalis at early age is thus prone to develop DEF. On the contrary, in hardened concrete, a high amount of alkalis will tend to hinder [20] or even avoid [22] DEF to occur since the desorption of the sulfates bound with the CSH will be slower in this case [22].

### *Temperature*

The influence of temperature is very complex since it has various effects on the composition of the pore solution: this parameter should thus not be considered separately but as a part of coupled processes [23]. The storage temperature has a strong influence on the expansive behavior of the affected materials. Generally, an increase of temperature is considered to trigger AAR, at least in terms of kinetics [4,14,24] and explains why performance tests are generally performed at high temperatures (38 to 60°C). The influence on the ultimate expansion does not seem to be as clear as for the kinetic. For some authors, the storage temperature has no real effect on the final expansion [14] whereas for others, the ultimate expansion is inversely proportional to the temperature [24].

Since the precipitation of ettringite is strongly related to the temperature, DEF effects are linked to the thermal history of the material: on the one hand, an “atypical” heating determines if DEF will occur while on the other hand, the storage temperature influences the characteristics of expansion. Above a threshold of 65-70°C [8], it is mostly believed that an increase of temperature or duration of heating increases the kinetic and the ultimate value of expansion [25,26,27]. Concerning the storage temperature, a heating implies an

increase of ettringite solubility decreasing the expansion [28]. However, a temperature increase implies an increase of the kinetics which can be explained by enhanced ionic exchanges at higher temperatures [25].

#### *Aggregate properties*

The content of reactants of the aggregates determines the amount of gel that can be created and thus influences the ultimate expansion and its kinetics [4]. Some kind of aggregates exhibit a so-called “pessimum” effect: for a given alkali content, it exists a reactive aggregate mix maximizing the expansion. The size of the particles has an influence on the rate of expansion, its final value and the induced damage on the material [4]. The results in the literature are various and seem difficult to be compared since the experimental techniques used are very different [29]. It seems that the size of aggregate corresponding to the greatest expansion is strongly correlated to its nature and its composition [23,29]. To take these complex couplings into account, some attempts of modeling have been performed [29]. This issue is nowadays of great interest because it determines the size of the aggregates used in the performance tests [23].

The physical nature of the aggregates seems to have an influence on the expansions related to DEF. Thus, limestone aggregates are mostly reported to delay the swellings as compared to siliceous ones [30]. This effect is supposed to be the consequence of a better paste-aggregate interface either due to a more angular shape or to lower thermal differential dilations between paste and aggregates [9]. As for AAR, the size of the aggregates has an influence on the expansion properties. The use of fine materials has been shown to decrease [31] or to increase the expansion [32] and no clear conclusion has been established so far.

#### *Concrete porosity*

For both reactions, the porosity has a determining influence on the expansive behavior. On the one hand, it drives the mobility of the reactants and thus influences the rate of expansion while on the other hand, an increase of porosity allows expansive products to form without exerting pressure [7,14,33]. Besides, a decrease of the w/c ratio induces a decrease of porosity and corresponds to better mechanical properties allowing the material to better withstand the pressure [7]. Porosity is even sometimes considered to explain the anisotropy of the AAR swellings [14]. As an expansion tank, the porosity partly defines the crystallization pressure of ettringite: the smaller and the less connected the porosity, the greater the expansion [9].

#### *Humidity*

Humidity is known to be one of the most influencing parameter for AAR and DEF: the expansion increases with humidity [7,14,22]. Water is necessary as a transportation medium but also as a part of the expansive process by sorption in the silica gel [23] or as constituent of ettringite [9]. The expansion is driven by the internal humidity of concrete [2,34] and appears above a humidity threshold reported to be about 80-85 % of Relative Humidity (RH) for AAR [35] and 90-92% for DEF [36]. However, RH is a function of the pore structure, the temperature, the chemical conditions and the moisture history and thus, this threshold values depend on the concrete studied [23]. In terms of saturation, Martin established a threshold of 0.93-0.95 for DEF depending on the nature of the concrete [5]. Concerning AAR, for ordinary concrete, the humidity provided by mixing water is generally high enough to generate expansion [7,14] contrarily to DEF [5].

#### *AAR and DEF possible competition*

AAR and DEF act frequently combined in the field. Due to their similar macroscopic effects, it is quite difficult to quantify precisely the deleterious contribution of each reaction [10]. Moreover, it exists mechanical couplings (the degradation due to a reaction weakens the material and promotes the other one) as well as chemical couplings [20] between the reactions leading to a possible AAR initiator effect on DEF [37].

## *Conclusion*

Numerous studies have been performed in the past to better understand AAR and DEF basic mechanisms. Except from the alkali content, all other parameters studied in this review seem to have similar qualitative effects on the expansions. A high alkali content will enhance AAR long term expansions while any alkali consumption (leaching, formation of gel) will favor DEF. In this context, researches were led at Ifsttar to quantify the effects of expansions on the material properties and are described thereafter.

## **3 EFFECTS ON MATERIAL PROPERTIES**

### **3.1 Expansions**

#### *Concrete mixes*

The tests presented in this paper were mostly performed during two extensive experimental programs relative to AAR [7] and DEF [5]. The different concrete mixes used are given in Table 1. Three different types of reactivity were obtained by mixing different cements and aggregates: an AAR mix (A), a DEF mix (D) and a combined AAR and DEF mix (AD). Two different cements were used: the cement C1 is a CEM I 52.5 and has been chosen because of its high content of alkalis ( $N_{a_2O_{eq}}=0.92\%$ ) to ensure AAR-prone chemical conditions; the cement C2 (CEM I 52.5 R) has a high aluminates, sulfates and alkalis content which is propitious for DEF to occur (respectively 3.46, 4.30 and 0.83 wt. %). For all mixes, the total alkali content was increased by adding  $K_2O$  in the mixing water up to a  $N_{a_2O_{eq}}$  value of 1.25% for A and 1% for D and AD. For A, a non-reactive limestone sand and reactive limestone aggregates were used. The D mix contains non-reactive siliceous (NRS) sand and aggregates. The AD mix contains the NRS sand and reactive limestone aggregates. All these mixes were designed to get similar mechanical performances and transfer properties.

After casting of the specimens, a 28-days curing time was applied before starting any test. For the A specimens, the elements were unmolded 24 hours after casting and then stored under an aluminum-foil sealing at ambient temperature to prevent materials from drying. For D and AD specimens, a heat treatment in water characterized by a plateau at 81°C during 72 hours was applied just after casting to simulate a mass concrete thermal history and thus induce DEF [38]. The specimens were unmolded at the end of the process and then stored during a 28-days period under aluminum-strips sealing at ambient temperature.

#### *Free expansion test*

The tests were performed on cylinders (0.11m in diameter, 0.22m in height) drilled out either from cylinders of 0.16m in diameter and 0.32m in height for the A specimens or from blocks ( $0.28 \times 0.28 \times 0.23m^3$ ) for D and AD specimens to provide an homogenous repartition of the aggregates in the samples. The cylinders were stored either immersed in tap water or sealed under an aluminum-foil cover to prevent the materials from drying. The storage temperature was constant and equal to 38°C. For each storage type, three identical specimens are cast for the mix A and four cylinders are used for the D and AD mixes. The vertical strains (i.e. parallel to the direction of casting) were monitored with a digital extensometer: stainless steel studs were glued on locations placed at 120 degrees from each other, allowing to take three measurements of the axial expansion of each specimen. The water content was monitored by weighing the specimens.

#### *Results and discussions*

Figure 1 represents the evolution of vertical expansion and the corresponding mass variations for the immersed and sealed specimens. The dots represent the mean value of the parameters and the error bar the standard deviation. The magnitude and the kinetics of the swellings can be quantified using Equation 1 [14,27,37]. Table 2 gives the estimation of the corresponding parameters for the different specimens.

$$\varepsilon(t) = \varepsilon_{\infty} \cdot \frac{1 - e^{-\frac{t}{\tau_c}}}{1 + e^{-\frac{t - \tau_L}{\tau_c}}} \cdot F(t) \text{ with } F(t) = \begin{cases} 1 & \text{if AAR} \\ \left(1 - \frac{\phi}{\delta + t}\right) & \text{with } \phi > \delta \text{ else} \end{cases} \quad (1)$$

DEF is generally believed to have stronger effects than AAR in terms of swellings. This observation is consistent with the field investigations. A comparison of the A and D immersed specimens emphasizes a higher magnitude of expansion for the DEF-affected materials. After the onset of expansion, DEF develops faster than AAR ( $\tau_c$ , which is inversely proportional to the slope of the curve, is smaller). However, it must be noted that these conclusions may not be relevant if other cement, aggregates or thermal history are used.

The combination of AAR and DEF for these specific materials leads to an earlier onset of the expansion for the AD specimens compared to the D ones stored in water (the latency time  $\tau_L$  is reduced). This phenomenon has been attributed to an initiator effect of AAR on DEF in this particular case [37]. AAR is believed to be triggered by the initial heat treatment and thus induces damages (microcracks) and decrease of the alkali content of the pore solution. Both of these effects are prone to enhance DEF.

Although the water content of the A and D sealed specimens remained equivalent during the test, it can be noted that significant expansions appeared in the case of AAR while drying shrinkage was measured for DEF (see Figure 2.c and 2.d). This phenomenon is consistent with a lower humidity threshold for AAR (see section 2). Moreover, no expansion is observed for the AD specimens; contrarily to the immersed case, the combination of DEF and AAR does not help to initiate DEF, at least during the monitoring period.

#### *Water-uptake mechanism*

The expansions in a moist atmosphere are associated with a water uptake (see Figure 1.b). Assuming that, for specimens kept under water, the expansion leads exclusively to the formation of cracks immediately filled with water (i.e. the volume variation of the specimen  $\Delta V$  is equal to the volume of water entering the specimen  $\Delta V_{\text{water}}$ ), it is possible to link the volume expansion  $\varepsilon_{\text{Vol}}$  to the mass variation of the specimen (see Equation 2 where  $V_{\text{ini}}$  is the initial volume of the specimen,  $\Delta m$  its mass variation,  $m$  its initial mass,  $\rho_{\text{concrete}}$  and  $\rho_{\text{water}}$  the specific gravity of concrete and water respectively and  $d_{\text{concrete}}$  the concrete density).

Another possibility to assess the volume expansion is to use the strain monitoring performed on the cylinders as given in Equation 3. In this relation,  $\varepsilon_{\text{vert}}$  and  $\varepsilon_{\text{hor}}$  correspond respectively to the vertical and horizontal expansion, i.e. the swelling measured in parallel and perpendicularly to the casting direction. The anisotropy coefficient  $k$  corresponds to the ratio between  $\varepsilon_{\text{vert}}$  and  $\varepsilon_{\text{hor}}$ . For DEF, in the absence of data and based on the literature review [9], we consider an isotropic process corresponding to a value of  $k$  equal to 1 ( $\alpha=3$ ). Based on the investigations of Multon [7], a ratio of 1.58 for the concrete A ( $\alpha=2.27$ ) was established.

$$\varepsilon_{\text{Vol}} = \frac{\Delta V}{V_{\text{ini}}} = \frac{\Delta V_{\text{water}}}{V_{\text{ini}}} = \frac{\Delta m}{\rho_{\text{water}}} \cdot \frac{\rho_{\text{concrete}}}{m} = \frac{\Delta m}{m} \cdot \frac{\rho_{\text{concrete}}}{\rho_{\text{water}}} = \frac{\Delta m}{m} \cdot d_{\text{concrete}} \quad (2)$$

$$\varepsilon_{\text{Vol}} = \varepsilon_{\text{vert}} + 2 \cdot \varepsilon_{\text{hor}} = \varepsilon_{\text{vert}} + 2 \cdot \frac{\varepsilon_{\text{vert}}}{k} = \left(1 + \frac{2}{k}\right) \cdot \varepsilon_{\text{vert}} = \alpha \cdot \varepsilon_{\text{vert}} \quad (3)$$

Figure 2 compares the volume expansion estimated thanks to Equations 2 and 3. Two main phases can be defined. Firstly, the estimation derived from the mass monitoring is higher than the one calculated

with the expansion. It indicates that during this phase, our assumption stating that water uptake is only the result of filling the crack opened by expansion is false. At this time, water ingress is probably mainly driven by capillary sorption as it can be seen for non-reactive concretes. Upon a vertical expansion of about 0.03% probably corresponding to cracking, a linear relation exist between the two estimations. The slopes are 0.939 and 1.074 for the A and D specimens respectively (close to 1) and indicate that our assumption may be valid.

### 3.2 Degradation of the mechanical properties

Figure 3 illustrates the effect of expansion on the Young's modulus (E) and the compressive strength (Rc) complementing results described in the case of AAR [7,14,39,40,41] and DEF [21,26,42]. AAR and DEF seem to have a common effect inducing a decrease of the Young's modulus, at least during the initiation of expansion. An increase of the modulus is sometime observed after the initial degradation induced by AAR. This is attributed to the filling of the cracks by silica gel [7,43]. Similarly, an increase of the dynamic modulus of elasticity is sometimes reported for DEF [42] and is attributed to the formation of ettringite in the cracks.

AAR and DEF have different effects on the compressive strength. It is frequently reported that AAR-affected materials only suffer from small decrease of Rc or even exhibit higher performances. This is usually attributed to a "healing effect" thanks to a continuous hydration of the cement due to the water ingress through AAR cracks. On the contrary, DEF-affected specimens suffer from a severe decrease of Rc. This effect could be explained by a competition between the degradation process and the continuous hydration: for AAR, if the material expands slowly without extensive cracking, the healing effect could be efficient enough to stabilize or increase the compressive strength; on the contrary, due to DEF expansions higher and growing faster, the healing process should not be able to fill the cracks, thus leading to a decrease of Rc.

### 3.3 Influence of expansions on the shrinkage behavior

#### *Experiments*

In this section, the concretes A and D are considered. The same monitoring techniques as in section 3.1 were used. Tests with the D mix were performed on cylinders of 0.11m in diameter and 0.22m in height [5]. For the A mix, cylinders of 0.16m in diameter and 0.32m in height were used [7]. To assess the effect of the expansions on the shrinkage behavior of the materials, specimens initially immersed as described in section 3.1 were dried in a 30% RH atmosphere at 38°C after the stabilization of the expansion (i.e. after 676 days in water for the A specimens and 538 days for the D and AD specimens). In parallel, other specimens of same composition and geometry were dried identically before the expansions occur, 28 days after casting.

#### *Results and discussion*

For both reactions, the expansions are strongly irreversible: a late drying leads to a limited shrinkage compared to the magnitude of swelling (see Figure 4). Free shrinkage is believed to be proportional to the water content of a material [44]. In a "real structure", the relation between the strain and the mass variation is not linear anymore because of restrained deformations. However, it exists a linear domain considered to be representative of the hydrous contraction: a linear part typical of the drying shrinkage of the concrete can be identified. To characterize the shrinkage process in beams tested in laboratory, Multon [7] and Martin [5] compare the shrinkage measured on cylinders to the ratio between the mass variation of the outer surface (2cm thick) and its ultimate value: thus, the shrinkage of the outer surface is linked to its drying progress.

Results of this computation are given in Figure 4. For the AAR-prone specimens, the expansion has no clear effect on the behavior since the slopes of the linear parts are similar and respectively equal to 0.0015 and 0.0016 before and after expansion. On the contrary, the slope is significantly increased after DEF (from 0.0013 to 0.0032) and illustrates its improving effect on the drying shrinkage.

## 4 CONCLUSIONS

Common features and differences between AAR and DEF have been listed in the context of the management of affected structures. In particular, the knowledge of the AAR field that can be transcribed to DEF (and conversely) could be pointed out while other fields need further investigations.

Although the reactions have similar macroscopic effects, the microscopic phenomena are very different. In particular, the expansion mechanisms might imply the anisotropy of AAR contrarily to DEF. Moreover, the presence of alkalis has an antagonist effect: high alkali content tends to delay DEF while it is prone to generate extensive AAR. Coupling the two reactions could trigger the expansions, the consumption of alkalis to form gel leading to the precipitation of ettringite. Finally, moisture, storage temperature and concrete porosity have a similar effect on AAR and DEF expansions.

The tests performed confirm the generally observed higher magnitude of DEF expansion. The combination of AAR and DEF led to an earlier onset of the expansion which could be explained by an initiator role of AAR. Besides, the formation of cracks during expansion has been linked to the water uptake.

Both AAR and DEF lead to a decrease of the Young's modulus that can be explained by cracking of the matrix. The effect on the compressive strength is not so clear: while it is believed to be limited with AAR, DEF strongly affects this characteristic. Higher kinetic and magnitude of DEF could explain this result.

At least for limited magnitude of expansion, AAR does not seem to have an influence on the shrinkage behavior of the materials. On the contrary, the drying shrinkage is increased for DEF affected specimens, probably due to the great amount of macro-cracks created during expansion.

All these results can be used as a basis to develop numerical models of AAR and/or DEF affected structures. If their general outline can be similar for both reactions, some aspects have to be treated carefully depending on the reaction considered. In particular, the effects on mechanical characteristics and the evolution of the moisture transfer properties have to be taken into account differently for the two reactions.

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TABLE 1: Concrete mixes (kg/m<sup>3</sup>).  
\* NRL=Non Reactive Limestone; NRS=Non Reactive Siliceous; ARL=Alkali Reactive Limestone;  
 $d_{min}/d_{max}$ =minimal and maximal particle size (mm)

| Mix | Reactivity | Cement type | Cement | Water | NRL* |     | NRS* |      | ARL* |      |       |       | Na <sub>2</sub> O <sub>eq</sub> |
|-----|------------|-------------|--------|-------|------|-----|------|------|------|------|-------|-------|---------------------------------|
|     |            |             |        |       | 0/5  | 0/2 | 4/8  | 8/12 | 4/7  | 7/10 | 10/14 | 14/20 |                                 |
| A   | AAR        | C1          | 410    | 205   | 621  | -   | -    | -    | 175  | 92   | 319   | 536   | 1.25%                           |
| D   | DEF        | C2          | 410    | 188   | -    | 854 | 100  | 829  | -    | -    | -     | -     | 1%                              |
| AD  | AAR+DEF    | C2          | 410    | 188   | -    | 797 | -    | -    | 114  | 29   | 843   | -     | 1%                              |

TABLE 2: Parameters estimation for the specimens (\* in days).

| Mix | Storage | $\epsilon_{\infty}$ | $\tau_C^*$ | $\tau_L^*$ | $\phi^*$ | $\delta^*$ |
|-----|---------|---------------------|------------|------------|----------|------------|
| A   | Water   | 0.23%               | 26         | 63         | -        | -          |
| A   | Scaled  | 0.11%               | 60         | 127        | -        | -          |
| D   | Water   | 1.58%               | 11         | 83         | 18       | 120        |
| AD  | Water   | 1.55%               | 9          | 62         | 17       | 120        |

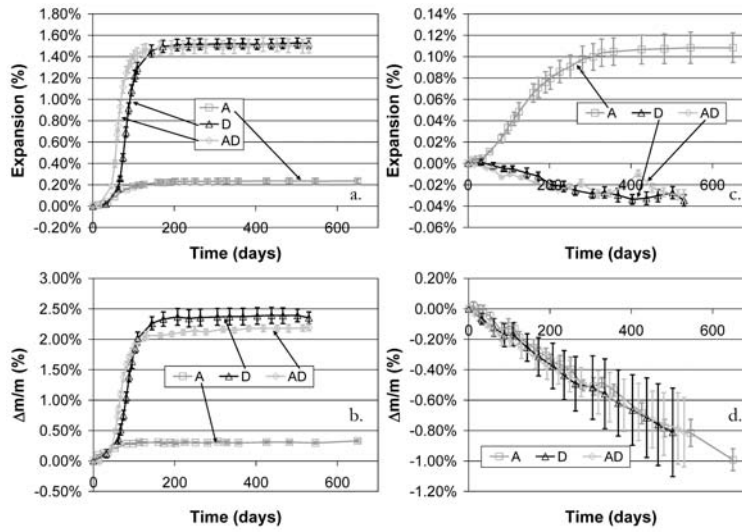


FIGURE 1: Vertical expansion and mass monitoring of the specimens stored in water (a and b respectively) and sealed (c and d respectively).

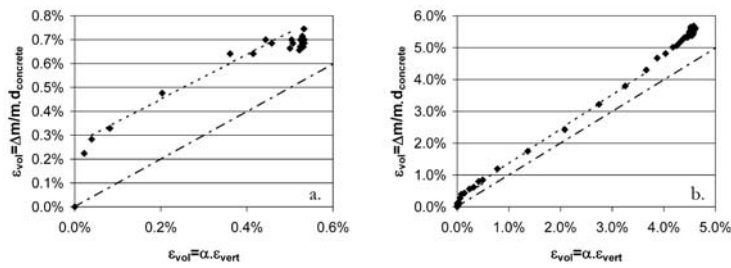


FIGURE 2: Volume expansion for AAR (a) and DEF (b) affected specimens (mixes A and D).

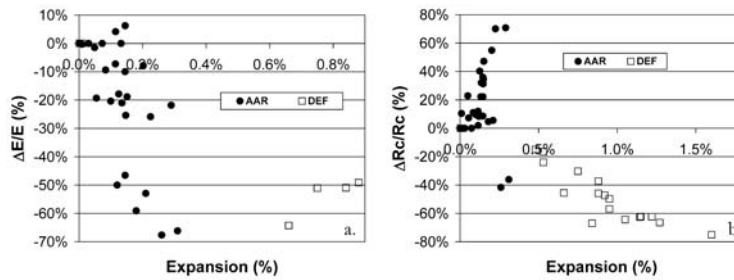


FIGURE 3: Relative variations of the Young's modulus E (a) and of the compressive strength  $R_c$  (b) as a function of expansion.

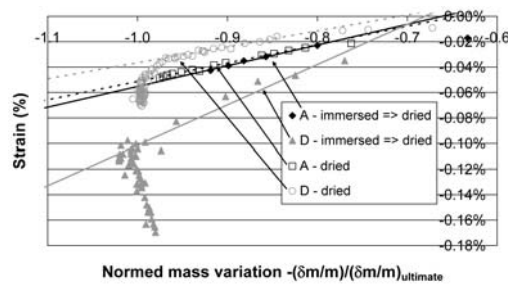


FIGURE 4: Evolution of the shrinkage as a function of the relative mass variation.