

Scale effect of alkali-silica reaction (ASR) expansion

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Abstract

The effect of particle size of reactive aggregates on ASR expansions has been largely studied. In spite of all these experimentations, conflicting results exist concerning the aggregate size which leads to the highest ASR expansion. Therefore, it is still difficult to generalize about the effect of the aggregate size. Most of research works clearly showed a pessimum effect for aggregate size larger than 100 μm , which can be explained by the time necessary for hydroxide ions to diffuse into the aggregate. However, all the results available in the literature were obtained using different experimental conditions and the coupled effects with other important parameters, such as specimen size used in the expansion tests, have been often neglected. This paper aims to investigate this coupled effect of specimen size and aggregate size on ASR expansion. Two parts are mainly presented and discussed:

1) Experimental results of mortar tests which were conducted with four aggregate sizes of siliceous limestone (0-315, 315-630, 630-1250 and 1250-2500 μm) and three specimen sizes (20x20x160, 40x40x160 and 70x70x280 mm) at 60°C in 1 mol/l NaOH. These results highlight a combined effect of the aggregate and specimen sizes on ASR expansions. It appears that, even with a 'specimen size / aggregate size' ratio higher than 100, this scale effect still exists.

2) A microscopic model which is based on the ions diffusion into the aggregate and the mechanical damage theory to predict the expansion of a Representative Elementary Volume (REV) of concrete. The experimental expansions are analysed with this model and an empirical function is proposed to take into account the scale effect.

Keywords: alkali-silica reaction (ASR), particle size, specimen size, expansion, scale effect

1 INTRODUCTION

Alkali-Silica Reaction occurs in all types of structures which contain alkali-reactive aggregates. The range of the aggregate size causing the highest ASR expansion varies along with the nature and composition of the aggregate, and significant differences were observed between rapid and slow alkali-reactive aggregates. Numerous papers discuss the effect of particle sizes of reactive aggregates on ASR expansion [1-7]. Research has shown that the expansion induced by ASR increases as the reactive particle size is reduced [6]. Other authors obtained insignificant expansion when the sizes of the reactive particles were under 50 to 150 μm [5,7]. In some cases involving opal aggregates, very small particles led to significant ASR expansions [2]. Other research works showed a pessimum effect for particle size, but for particles much larger than 100 μm [2,4,6,8]. Besides, the factor of "specimen size" was largely less studied [9,10]. Experiments performed with different ratios 'aggregate size to specimen size' showed the effect of the size of the specimens on ASR expansions measured. Lower expansions were measured on

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the smallest specimens [9,10]. The difficulty to generalize results about the effect of the particle size of reactive aggregates on expansion could be explained by the differences of experimental conditions, and particularly the combined effects of aggregate size and specimen size on ASR expansion. This paper aims to investigate and quantify this scale effect and the influence of the reactive silica content of aggregate on this effect in order to have a better understanding of expansion tests.

2 EXPERIMENTAL CONDITIONS AND RESULTS

2.1 Experimental conditions

Expansion was measured on mortar prisms with a water/cement of 0.5 and a sand / cement of 3. Mixtures were adjusted by adding NaOH in the mixing water. For the reference specimens, the sand was only composed of non reactive marble. For all the other specimens, the sand contained 30% of reactive aggregate and 70% of non-reactive marble. The prisms were stored in NaOH solution at 60°C. The experimentation presented in this paper aimed to study three particular points:

Effect of alkali concentration

The aim of this part was to study three NaOH concentrations of immersion close to the standardized conditions of 1 mol/l (0.77, 1 and 1.25 mol/l). Reactive siliceous limestone was used to study the effect of alkali concentration on ASR expansion. The specimen and aggregate sizes were 20 x 20 x 160 mm and 315-1250 μm , respectively.

Effect of reactive silica content

The second point concerned the effect of the reactive aggregate nature on ASR expansion. Four reactive aggregates with different reactive silica contents were chosen and tested (Table 1). Opal (O) is known to be very reactive and to present large expansion if the amount of available alkali is sufficient. Quartzite (Q) and siliceous limestone (SL) are usually less reactive but can exhibit significant expansion in concrete. Quartz aggregate (QA), which contains mostly quartz, is considered as non-reactive. The aggregates were used to cast 20x20x160 mm specimens with reactive aggregate 315-1250 μm (15% of 315-630 and 15% of 630-1250 μm). The specimens were kept in 1 mol/l NaOH solution.

Combined effect of aggregate and specimen sizes

Four reactive aggregate size classes: C1 (0-315 μm), C2 (315-630 μm), C3 (630-1250 μm) and C4 (1250-2500 μm) and three specimen sizes 20x20x160 mm, 40x40x160 mm and 70x70x280 mm were used to research the combined effect of aggregate and specimen sizes on ASR expansion. Specimens were stored in 1 mol/l NaOH solution. Siliceous limestone was used as reactive aggregate.

2.2 Experimental results

Effect of the alkali concentration

The final expansions (i.e. at 360 days) for the three concentrations 0.77, 1.0 and 1.25 mol/l were 0.67%, 0.64% and 0.60% respectively (Figure 1). The difference between two consecutive concentrations appeared to be small considering that the standard deviation for the specimens was in the range of 0~0.02%. Between 0.75 and 1.25 mol/l, the alkali concentrations could be considered to have little influence on long term ASR expansions in conditions of abundant alkali.

Effect of the reactive silica content

The ASR expansions obtained for the four aggregates are plotted in Figure 1. The specimens containing opal is the most reactive with fast expansion and an asymptotic value of about 1.35%. The specimens with siliceous limestone (SL) exhibit rapid expansion, but the final expansions were lower (about 0.6%). The specimens with quartzite aggregate (Q) presented a slow expansion rate but reached a

final expansion of about 0.55%. ASR expansion of the specimens containing quartz aggregate (QA) was about 0.14%. At the end of the experiment, the specimens containing opal were greatly damaged and cracked (crack width of about 425 μm), while the other specimens showed cracks width lower than 10 μm .

Combined effects of aggregate and specimen sizes

The ASR expansions obtained for mortars containing the reactive siliceous limestone with various aggregate sizes and cast in specimens of different sizes are given in Figures 2 to 4. First, the specimens containing small reactive particles (0-315 μm) had the smallest expansion (lower than 0.15%). The ASR expansions were significant and higher than 0.5% for the three other aggregate sizes. About the amplitude of the final ASR expansion, the largest specimens showed the highest ASR expansion. This was particularly significant on the large aggregate class 1250-2500 μm (ASR expansions were twice as large for the 70 mm size specimens as for the others). ASR expansions present a pessimum effect with the reactive aggregate size (with a final expansion larger for the aggregate size 315-630 μm) for measurements performed on the smallest specimens (20x20x160 mm, 40x40x160 mm) but not for the largest ones. These observations show the significant combined effect between specimen size and aggregate size.

Scale effect

In order to understand the scale effect (combination of the aggregate and specimen sizes effects on expansion), the ASR development can be described in a three-phase model:

1. Ions (hydroxyls and alkalis) present in the cement paste reach and diffuse into the aggregates (Figure 5a).
2. Alkalis react with reactive silica and create ASR gel in and/or around the aggregate particle (according to the repartition of the reactive silica in the aggregate). A part of the gel can fill the connected porous volume surrounding the aggregate in a thickness t_c without leading to damage [8,11-13] (Figure 5b). It can explain why the smallest aggregates lead to small expansion. Indeed, for a same content of reactive silica, more of the gel can migrate in this connected porosity for the smallest aggregates than for larger aggregates. Thus, little ASR gel is available to cause expansion [8,12]. ASR gel exerts pressures to the surrounding aggregate and cement paste when the connected porosity is filled (Figure 5b). The pressure causes cracking of the aggregate and cement paste. The propagation of cracks in small specimens containing large aggregate can be rapid and can occur for low pressures, while it is more difficult for larger specimens contained smaller aggregates and needs higher pressure (Figure 5b). In the framework of the fracture mechanics, the maximal normal stress in the vicinity of the aggregate which induces a pressure on the cement paste is given by:

$$\sigma(r) = \frac{K_I}{\sqrt{2\pi r}} \quad (1)$$

With σ the maximal normal stress at the point M located at r, the distance from the edge of the aggregate (Figure 5b) and K_I , the stress intensity factor obtained from the relation:

$$K_I = p_g \cdot f\left(\frac{R_a}{L}\right) \quad (2)$$

where f is a function increasing with the ratio R_a/L , with R_a , the radius of the aggregate and with L , the dimension of the specimen. Assuming that the aggregates have spherical shapes, and p_g , the gel pressure at the time-step t can be assessed from [8]:

$$p_g(t) = M_g \left\langle \frac{4}{3} \pi R_a^3 V_{gel}^{mol} s \zeta(t) - \varphi \frac{4}{3} \pi [(R_a + t_c)^3 - R_a^3] \right\rangle \quad (3)$$

with M_g the gel elastic modulus, s the reactive silica content, ζ the chemical advancement of the alkali-silica reaction (which depends on temperature, moisture and alkali conditions), V_{gel}^{mol} the molar volume of the gel, φ the concrete porosity, and t_c the thickness along which the gel can migrate from the aggregate to the cement paste.

The normal stress at a given distance from the aggregate is larger if the size of the aggregate is large compared to the size of the specimen. Thus, the tensile strength at the boundary of the specimen can be reached for a smaller pressure (Figure 5b) and cracks can be initiated faster.

3. Once concrete cracks, ASR-gels can be accommodated by cracking without creating supplementary pressure [14,15]. Then, the pressure in the gels falls, which stops the expansion.

If the pressure necessary to cause cracks is lower for small specimens containing large aggregate, the pressure of the gel will fall sooner in the case of the specimens 20x20x160 mm containing the aggregate 1250-2500 μm than for the specimens 70x70x280 mm containing the same aggregate. It explains the smallest expansion measured at the end of the experiment (Figures 2 and 4). It can explain the pessimum effect of ASR expansion with aggregate size. ASR expansion was lower for the largest aggregates in the smallest specimens. As explained just above, the specimens 20x20x160 mm containing the aggregate 1250-2500 μm were more affected by the scale effect than the specimens containing the aggregate 315-630 μm : a larger aggregate induced a greater stress intensity factor (Equation 2). Therefore, the part of reactive silica consumed when cracking appeared is lower in the specimens containing the largest aggregate. Once cracks occur, a large part of the gel is accommodated by the cracks, and the gel produced after cracking leads to little supplementary pressure and expansions stops before the chemical reaction is stopped for a final expansion lower for the largest aggregate. This is consistent with experimental determination of degree of reaction performed with chemical attack [15] and SEM image analysis [16]: expansion stopped while the degree of reaction keeps on increasing. The pessimum effect is not an intrinsic phenomenon but is due to the scale effect which depends on the 'specimen size / aggregate size' ratio.

3 MODELLING ASR EXPANSION

The modelling used in this paper is an improvement of the microscopic model developed by Multon et al. [12]. It was based on previous models [11-13] and attempted to predict the damage and the expansion of a Representative Elementary Volume (REV) of concrete containing a mix of reactive aggregates of different sizes. The reaction between the reactive silica and the alkalis was determined through the mass balance equation, which controls the diffusion mechanism in the aggregate and the fixation of the alkali in the ASR gels. The mechanical part of the model is based on the damage theory in order to assess the decrease of stiffness of the mortar due to cracking caused by ASR, and to calculate the expansion of the REV [12]. Some modifications were made in this model: the threshold of alkalis has been re-evaluated in accordance to the results presented just above and the combined effects of specimen and aggregate sizes and reactive silica content on expansions were taken into account.

3.1 Presentation of the model

The reactive aggregates and the REV containing the surrounding concrete paste were assumed to be spherical [12]. The diffusion of ions in the aggregate controls mainly the kinetics of the chemical attacks and thus of the expansion. At the beginning of ASR, hydroxyl ions attack the reactive silica of the aggregate. In the previous model, a threshold of alkali concentration of 0.625 mol/l of Na⁺ under which ASR did not occur was considered [12]. It was based on experimental data which showed that with alkali content lower than 3 kg per m³ of concrete, no ASR-expansion was observed [17,18] in tests performed in usual conditions (in air with RH upper than 95%). In this work, all the specimens were kept immersed in alkali solution and thus, the alkali was supplied in abundant quantities. The experiments performed on specimens immersed in three alkali concentrations (0.77, 1 and 1.25 mol/l) showed very small differences. Considering a threshold of 0.625 mol/L, the expansion of the mortars kept in 0.77 mol/l solution should be significantly slower than the expansion of the mortars kept in 1.25 mol/l solution. The results of this assumption will be studied in the next part.

ASR-expansion calculated by the model is imposed by the effective volume of ASR-gel. The effective volume of gel is deduced from the total volume of gel which is proportional to the number of moles of ASR gel formed after the attack of the reactive silica:

$$V_g = \sum_i n_g^i \times V_{gel}^{mol} \quad (4)$$

with n_g^i (mol): the number of moles of ASR gel produced by the aggregate size i , V_{gel}^{mol} (m³/mol): the molar volume of the gel.

Whatever the distribution of the silica in the aggregate, a part of the reactive silica is in contact with the cement paste: the ASR-gel formed from this reactive silica can permeate through the connected porous volume and partly fill it. It has been assumed that the proportion of gels filling the connected porosity without creating pressure was the total volume of porosity filled by the gel with an equivalent thickness t_c , which was supposed to be constant with the size of the aggregate [13]:

$$V_{por} = \frac{4}{3} \pi \left((R_a + t_c)^3 - (R_a)^3 \right) \varphi \quad (5)$$

With R_a : the aggregate size, t_c : the thickness of gels (around aggregates) filling the porosity and φ : the porosity.

Once cracking appears in the specimens, a part of the gel which is not accommodated by the porosity $\langle V_g - V_{por} \rangle$ is assumed to be accommodated by the cracks without producing significant pressure. This part depends on the scale effect: the larger the aggregate compared to the specimens size, the higher the volume of ASR-gel accommodated by the cracks. This phenomenon is also affected by the reactive silica content of the aggregate. The higher the reactive silica content is, the stronger is the non-linearity due to cracking. A reduction of the volume of effective gel (gel not accommodated by porosity and cracks) is assumed that applies equally to all the aggregates of a given size:

$$V = \langle V_g - V_{por} \rangle^+ \times \exp\left(-c \frac{\phi}{1} s^x\right) \quad (6)$$

With V : the effective volume of gel, c : the scale effect coefficient, Φ : the aggregate diameter, l : the specimen size, s : the reactive silica content (mol/m^3 of aggregate) and χ : silica content exponent.

The pressure imposed to the aggregate and thus to the cement paste depends on the effective volume of gel V . The effective volume of gel is then used in the mechanical modelling presented in [12] to deduce the resulting expansion.

3.2 Comparison with experiments

Table 2 sums up the various parameters used in the modelling process. Three parameters of the physicochemical modelling (thickness of the connected porous interface zone t_c , molar volume of ASR gel V_{gel}^{mol} , scale effect coefficient c – equations 5 and 6) were obtained by curve fitting to obtain the final expansions measured on the specimen of size 70 x 70 x 280 mm with the four different size classes (0-315, 315-630, 630-1250 and 1250-2500 μm) of the siliceous limestone. The value of χ was determined by fitting the final expansion of mortars containing opal as aggregate. Coefficient of alkali diffusion in aggregate D_a depends on the nature of the aggregate. It was obtained through curve fitting with the results obtained on specimen of size 20 x 20 x 160 mm (Figure 1). The coefficients of diffusion in aggregate (D_a) obtained were equal to $5.0 \times 10^{-14} \text{ m}^2/\text{s}$ for opal, $2.0 \times 10^{-14} \text{ m}^2/\text{s}$ for the siliceous limestone, $4.0 \times 10^{-16} \text{ m}^2/\text{s}$ for quartzite and $2.0 \times 10^{-16} \text{ m}^2/\text{s}$ for quartz aggregate.

Expansions obtained with modelling are compared with the measurements in Figure 1. Except for the expansion kinetics of specimens containing opal, both kinetics and final expansions obtained by the calculations are in accordance with the measurements for the four types of aggregates. The description of the expansion kinetics by the model is globally quite good: presence of a latent time before the initiation of the expansion, followed by a high rate of expansion and ending by a low rate to reach the final expansion. Concerning the effect of the alkali concentration of the immersion solution, the difference between the final expansion predicted by the model (0.70%) and the mean measured expansion (0.65%) is lower than 10%. The calculation does not show any differences in final expansions between the three alkali concentrations. Indeed, the model assumes that all the reactive silica is consumed in the three conditions and thus, the volume of ASR gel created is the same. The aim of this part was to verify the ability of the model to evaluate the effect of the alkali concentration surrounding the specimens. It was obtained by the assumption exposed above: in the case of specimens immersed in alkali solutions, no alkali threshold has to be considered. This assumption was well supported by the experimental results. The curve obtained by the model for opal did not fit well with experimental results. It could be likely attributed to crack opening for opal specimens. Indeed, the specimens containing opal aggregate present much larger cracks than specimens of the same size cast with the other aggregates. These cracks could cause a great increase in the diffusion phenomenon, which could accelerate the reaction while the model considers the diffusion as constant in spite of cracks opening. The comparison of the calculated final expansions obtained for the three specimen sizes (20 x 20 x 160 mm, 40 x 40 x 160 mm, 70 x 70 x 280 mm) and the four aggregate sizes of the siliceous limestone with the measurements is shown in Figure 6. Concerning these values, only the measurements performed on the largest specimens were used in the parameters identification. The calculated results seem to be in good agreement with the measurements (Figure 6). The differences between calculated and measured expansions are lower than 15% except for the expansions obtained on the smallest specimens (20x20x160mm) containing the largest aggregate (1250-2500 μm) with a difference of about 30% (Figure 6).

4 CONCLUSIONS

This paper aims to present the results of an investigation to quantify the combined effects of aggregate and specimen sizes on ASR expansion and the influence of the reactive silica content of aggregate on this effect. In order to quantify these effects on ASR expansion, a microscopic model was improved. A scale effect, combination of the effects of aggregate size and specimen size on ASR expansion, has been highlighted by experimentation. Thus, the well-known pessimum effect of aggregate size on ASR-expansion appears not to be an intrinsic phenomenon of ASR-expansion but to depend on the size of the specimen used to perform the expansion test. Considering that the scale effect is inevitable for specimen (ratio between specimen and aggregate sizes should be larger than 100 to decrease significantly the effect), experimental results of expansion tests should be analysed in respect to this effect. The modelling proposed to take into account the scale effect with an empirical relationship. Calculations are in good agreement with experimentations for expansion tests performed with ratio between the specimen and the aggregate sizes lower than 10 for the four studied aggregates.

5 REFERENCES

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TABLE 1: Reactive silica contents of aggregates

Reactive silica (SiO_2)	SL	O	Q	QA
Percentage in mass (%)	6.9	50.4	7.6	2.7
Content (mol/m^3 of aggregate)	3000	21900	3300	1170

TABLE 2: Parameters of modelling

Parameter	Symbol	Identification	Value	Units
<i>Aggregate</i>				
Reactive silica content	s	measurement	D.N.A.*	mol/m^3
Coefficient of diffusion	D_a	curve fitting	D.N.A.*	m^2/s
Porosity	p	usual value	0.01	%
<i>Paste</i>				
Porosity of mortar	φ	measurement	18.0	%
Thickness of the connected porous interface zone	t_c	curve fitting	14.0×10^{-6}	m
<i>Gel</i>				
Molar volume of ASR gel	$V_{\text{gel}}^{\text{mol}}$	curve fitting	4.2×10^{-4}	m^3/mol
Scale effect coefficient	c	curve fitting	0.037	$(\text{mol/m}^3)^{-\chi}$
Silica content exponent	χ	curve fitting	0.75	-

* depends on the nature of the aggregate

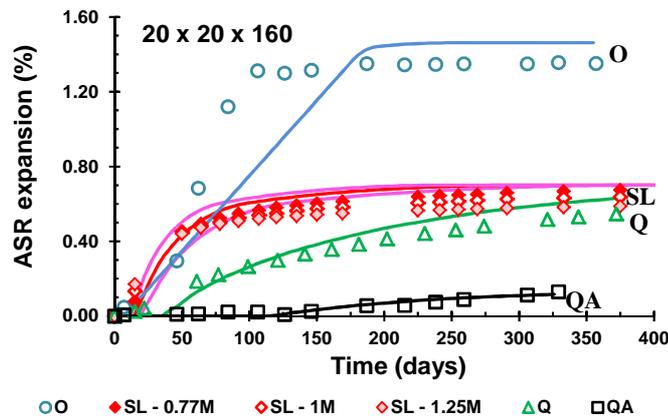


FIGURE 1. Comparison of calculated and measured expansions according to the alkali concentration of the immersion solution (0.77, 1.0 and 1.25 mol/l) and to the nature of the aggregate (O: opal, SL: siliceous limestone, Q: quartzite and QA: quartz aggregate)

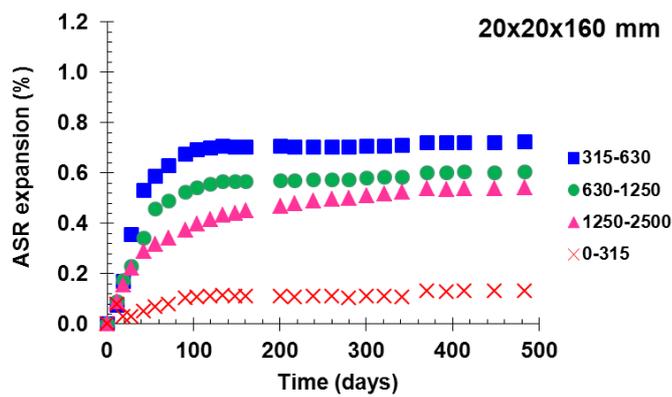


FIGURE 2. ASR expansions in prismatic specimens 20x20x160 mm (SL: siliceous limestone)

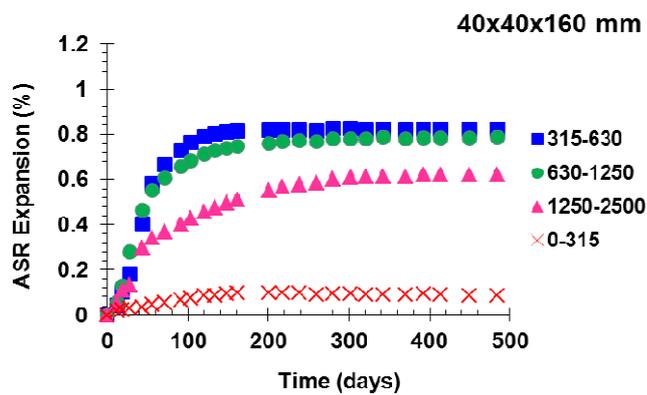


FIGURE 3. ASR expansions in prismatic specimens 40x40x160 mm (SL: siliceous limestone)

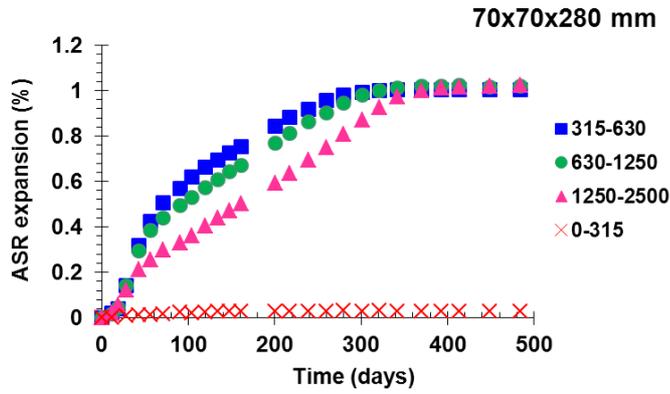


FIGURE 4. ASR expansions in prismatic specimens 70x70x280 mm (SL: siliceous limestone)

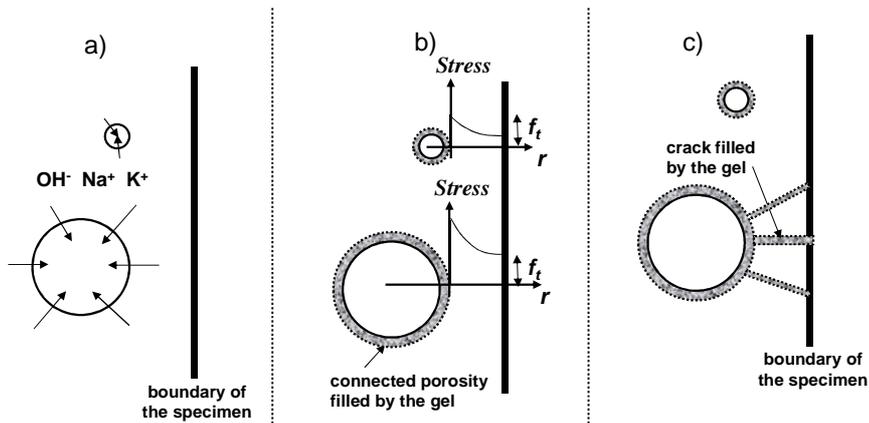


FIGURE 5. Description of the ASR development in three phases

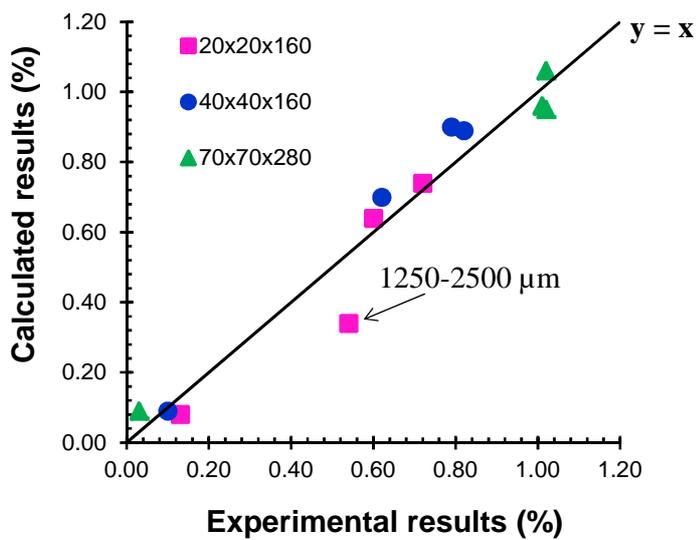


FIGURE 6. Comparison of calculated and measured expansion of specimens with siliceous limestone