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1 **Alkali-silica reaction (ASR) expansion: pessimum effect**
2 **versus scale effect**

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10
11 **Abstract**

12 The effect of aggregate size on ASR expansions has been largely studied and conflicting
13 results exist concerning the aggregate size that leads to the highest ASR expansion. Most of
14 the research works clearly show a pessimum effect of aggregate size on ASR expansion.
15 However, all the results available in the literature were obtained using different experimental
16 conditions and the combined effects of other important parameters, such as specimen size
17 used in the expansion tests, have often been neglected. This paper aims to investigate the
18 combined effect of specimen size and aggregate size on ASR expansion. Experimental results
19 highlight a scale effect, a combination of the effects of aggregate size and specimen size on
20 ASR expansion. This scale effect appears to be influenced by the reactive silica content of the
21 aggregate. Modelling at microscopic level is used to propose a quantification of this effect.

22
23 **Keywords:** alkali-silica reaction (ASR), particle size, specimen size, expansion, scale effect
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27 **1. Introduction**

28 Alkali-Silica Reaction (ASR) is a deleterious chemical reaction occurring in all types of
29 structures that contain alkali-reactive aggregates: dams, bridges, roads, breakwaters, etc. The
30 mechanism of expansion can be described in three main steps [1-4]: the diffusion of ionic
31 species (Na^+ , K^+ , OH^- , Ca^{2+}) into the aggregates, the disruption of the silanol and siloxane
32 bonds contained in the reactive silicate, and the reaction of alkali silicate with ionic species
33 (Na^+ , K^+ , Ca^{2+}) to form ASR gels. ASR gel induces pressure in the aggregate and in the
34 cement paste, leading to stresses and thus cracking. ASR expansion depends on numerous
35 parameters (amounts of alkali, reactive silica and water present, aggregate and specimen sizes,
36 etc.).

37 It seems that the range of aggregate size causing the highest ASR expansion varies with the
38 nature and composition of the aggregate. Numerous papers [5-23] discuss the effect on ASR
39 expansion of particle sizes of reactive aggregates like opal [5-12], various kinds of silica
40 glass, fused silica and waste silica glass, and so on [13-24]. Some research has shown that the
41 expansion induced by ASR increases as the reactive particle size is reduced [9,14,17,19].
42 Some authors obtained insignificant expansion when the sizes of the reactive particles were
43 under 50 to 150 μm [11,13,15,22]. Furthermore, the use of powder from reactive aggregates
44 like pozzolans, with particle sizes up to about 100 μm , has been developed to counteract the
45 effect of ASR [15,18,21]. Only a few exceptions with very small particles led to significant
46 ASR expansions, all involving opal aggregates [5-7, 12]. Other research works have shown a
47 pessimum effect for various types of aggregates, with pessimum values occurring in a wide
48 interval of particle sizes [4,7,8,10-13,16,20,23,24]. It is observed that, in some cases, the most
49 damaging size, leading to the highest ASR expansion, reaches more than 1 mm. The effect of
50 “specimen size” has been much less studied [25-28]. Experiments performed with different

51 'aggregate size to specimen size' ratios show the effect of the size of the specimens on the
52 ASR expansions measured. Lower expansions were measured on the smaller specimens
53 [25,26].

54 First, a literature review showed that conflicting results exist concerning the most damaging
55 reactive aggregate size, which leads to the highest ASR expansion. Secondly, experimentation
56 performed by Zhang et al. [26] suggested that the two effects of aggregate and specimen sizes
57 are combined. Therefore, the difficulty in generalizing results on the effect of the particle size
58 of reactive aggregates could be explained by the differences in experimental conditions, and
59 particularly the combined effects of aggregate size and specimen size on ASR expansion. This
60 paper aims to investigate and quantify this scale effect and the influence the reactive silica
61 content of aggregate has on the effect, in order to gain a better understanding of expansion
62 tests.

63 The development of an ASR model at the microscopic level is a major concern of researchers
64 trying to understand the mechanisms involved in the ASR and to predict future expansion.
65 Previous modelling investigated the different aspects of the reaction: the mechanical
66 consequences of ASR [29-32], the chemical mechanisms driving the attack of the aggregate
67 by hydroxyl ions [33] and their coupling [34-38]. These works do not take into account the
68 permeation of ASR gels in cracks which could be the main cause of the decrease of expansion
69 by pessimum or scale effects. The concepts of fracture mechanics can explain the cracking
70 phenomenon in the cases of aggregate in infinite matrix [39, 40] or of central penny-shaped
71 crack in sphere [41]. However, it is difficult to apply such models in the cases of specimens
72 containing aggregates with numerous interactions (interactions with other neighbour
73 aggregates and with specimen boundaries). The experiments presented in this paper were used
74 to point out the dependence of expansion with aggregate and specimen sizes. These results are
75 explained by fracture mechanics concepts. The teachings pointed out by fracture mechanics

76 are then used to propose a simplified relationship to take into account the scale effect in an
77 analytical modelling. Finally, the experimental results are used here to calibrate and discuss
78 the validity and the capability of the model.

79 **2. Experimental conditions and results**

80 ***2.1 Experimental conditions***

81 Expansion was measured on mortar prisms with a water/cement ratio of 0.5 and a sand (1512
82 kg/m³) / cement (504 kg/m³) ratio of 3. Mixtures were adjusted by adding NaOH to the
83 mixing water in order to have the same alkali concentration in the pore solution and in the
84 storage solution. For the reference specimens, the sand was composed of the non-reactive
85 marble only. For all the other specimens, the sand contained 30% of reactive aggregate and 70%
86 of non-reactive marble. The prisms were stored in NaOH solution at 60°C. The expansion
87 measurements were performed after the prisms had cooled to 20°C (~12 h) in the NaOH
88 solution. Each variation of length value was calculated as the mean of three values from three
89 replicate specimens measured using the scale micrometer method (specimens had shrinkage
90 bolts in the two extremities) [42,43]. The experimentation presented in this paper aimed to
91 study three particular points:

92 ***Effect of alkali concentration***

93 As explained above, the specimens were kept immersed in NaOH solution. The aim of this
94 part was to study three NaOH concentrations for immersion close to the standard conditions
95 of 1 mol/l (0.77, 1 and 1.25 mol/l). Reactive siliceous limestone was used to study the effect
96 of alkali concentration on ASR expansion. The specimen and aggregate sizes were
97 respectively 20 x 20 x 160 mm and 315-1250 µm (15% of 315-630 and 15% of 630-
98 1250 µm). Eighteen specimens (nine reactive and nine reference specimens) were used for
99 this part of the experimentation.

100 *Effect of reactive silica content*

101 The second point concerned the effect of the reactive aggregate nature on ASR expansion.
102 Four reactive aggregates with different reactive silica contents were chosen and tested. Opal
103 (O) is known to be very reactive and to give large expansion if the amount of available alkali
104 is sufficient. Quartzite (Q) and siliceous limestone (SL) are usually less reactive but can
105 exhibit significant expansion in concrete. Quartz aggregate (QA), which contains mostly
106 quartz, is considered as non-reactive. The silica contents of the aggregate [43] are given in
107 Table 1. The aggregates were used to cast 20x20x160 mm specimens with 315-1250 μm
108 reactive aggregate (15% of 315-630 and 15% of 630-1250 μm). The fifteen specimens were
109 kept in 1 mol/l NaOH solution.

110 *Combined effect of aggregate and specimen sizes*

111 Four reactive aggregate size classes: C1 (0-315 μm), C2 (315-630 μm), C3 (630-1250 μm)
112 and C4 (1250-2500 μm) and three specimen sizes: 20x20x160 mm, 40x40x160 mm and
113 70x70x280 mm were used to research the combined effect of aggregate and specimen sizes on
114 ASR expansion. The forty-five specimens (thirty-six reactive and nine non-reactive
115 specimens) were stored in 1 mol/l NaOH solution. Siliceous limestone was used as the
116 reactive aggregate.

117 **2.2 Experimental results**

118 **2.2.1 Effect of the alkali concentration**

119 The final expansions for the three NaOH concentrations of 0.77, 1.0 and 1.25 mol/l were
120 0.67%, 0.64% and 0.60% respectively (Figure 1). The difference between two consecutive
121 concentrations appeared to be small considering that the standard deviation for the specimens
122 was in the range of 0~0.02%. Between 0.77 and 1.25 mol/l, the alkali concentrations could be
123 considered to have little influence on ASR expansions in conditions of abundant alkali.

124

125 **2.2.2 Effect of the reactive silica content**

126 The ASR expansions obtained for the four aggregates are plotted in Figure 1. The specimens
127 containing opal were the most reactive, with fast expansion and an asymptotic value of about
128 1.35%. The specimens with siliceous limestone exhibited rapid expansion, but the final
129 expansions were lower (about 0.6%). The specimens with quartzite aggregate presented a
130 slow expansion rate but reached a final expansion of about 0.55%. ASR expansion of the
131 specimens containing quartz aggregate was about 0.14%. By the end of the experiment, the
132 specimens containing opal were seriously damaged and cracked (crack width of about 425 μm)
133 while the other specimens showed cracks with widths smaller than 10 μm (Figure 2). The
134 reliability of measured expansions could have been affected by such cracking. However, the
135 coefficient of variations obtained for three specimens was lower than 5%. It shows that the
136 expansion scattering stayed quite small in spite of the large cracks.

137 **2.2.3 Combined effects of aggregate and specimen sizes**

138 The ASR expansions obtained for mortars containing the reactive siliceous limestone with
139 various aggregate sizes and cast in specimens of different sizes are given in Figure 3. First,
140 the specimens containing small reactive particles (0-315 μm) had the smallest expansion
141 (lower than 0.15%), confirming results found in the literature. The ASR expansions were
142 significant and higher than 0.5% for the other three aggregate sizes. Concerning final ASR
143 expansion, the largest specimens showed the highest ASR expansion (Figure 4). This was
144 particularly significant on the large aggregate class 1250-2500 μm (ASR expansions were
145 twice as large for the 70x70x280-mm specimens as for the others). Moreover, ASR
146 expansions presented a pessimum effect with the reactive aggregate size (with a final
147 expansion larger for the aggregate size 315-630 μm than for 1200-2500 μm) for
148 measurements performed on the smallest specimens (20x20x160 mm, 40x40x160 mm) but

149 not for the largest ones (Figure 4). These observations show the significant combined effect of
150 specimen size and aggregate size.

151 **2.2.4 Scale effect**

152 *Mechanisms*

153 In order to understand the scale effect (combination of the aggregate- and specimen-size
154 effects on expansion), the ASR development can be described in four phases:

155 1. Ions (hydroxyls and alkalis) from the pore solution diffuse into the aggregates whatever the
156 reactive silica distribution in the aggregate (uniform or in veins – Figure 5-a).

157 2. Ions react with reactive silica and ASR gel is created in and/or around the aggregate
158 (according to the distribution of the reactive silica in the aggregate, a part of the reactive silica
159 can even be in direct contact with cement paste – Figure 5-b). A part of the gel can fill the
160 connected porous volume surrounding the aggregate without leading to damage [33-35,39]
161 (Figure 5-b) and also a part of the porosity of the aggregate close to the reactive site which
162 can lead to the cracking of veined aggregates [41]. This can explain why the smallest
163 aggregates lead to small expansion. For the same reactive silica content, more of the gel can
164 migrate in the connected porosity for the smallest aggregates than for larger aggregates. Thus,
165 a small amount of ASR gel is able to cause expansion [23,34,39]. It is easier for the cement
166 paste to accommodate ASR gel when the gel is created by a large number of small sites rather
167 than by a small number of scattered, larger sites.

168 3. ASR gel exerts pressures on the surrounding aggregate and cement paste when a part of the
169 connected porosity is filled (Figure 5-c) and the gel can no longer move in the porosity. In
170 order to assess the gel pressure p_g^a for the aggregate of size a , an analogy with previous
171 modelling [23, 34] can be made. In these works, the ASR mechanical effect was assumed to
172 be an imposed strain of the aggregate on the surrounding cement paste equal to:

$$\mathcal{E}_{imp}^a = \frac{\langle V_a^{gel} - V_a^{poro} \rangle^+}{V_a} \quad (1)$$

173 $\langle X \rangle^+$ is the positive part of X : if $X < 0$, $\langle X \rangle^+ = 0$ otherwise $\langle X \rangle^+ = X$

174 V_a^{gel} is the volume of ASR gel formed in reactive aggregates and V_a , the volume of the
 175 reactive aggregate, $V_a = \frac{4}{3}\pi \cdot R_a^3$ (if spherical shape is assumed for aggregate).

176 V_a^{poro} is the volume of the porosity in which the ASR gel can migrate without causing
 177 expansion. It was assumed that the proportion of gels filling the porosity connected to the
 178 reactive sites (cement paste and aggregate) without creating pressure was the total volume of
 179 porosity filled by the gel with an equivalent thickness t_c , which was assumed to be
 180 independent of the aggregate size:

$$V_a^{poro} = \varphi \frac{4}{3}\pi \cdot \left((R_a + t_c)^3 - R_a^3 \right) \quad (2)$$

181 where φ is the porosity of the mortar. In reality, it includes a part of aggregate porosity but, by
 182 sake of simplicity, the model considers the thickness t_c as the average distance of gel
 183 penetration which should depend on the gel pressure: penetration is more or less difficult due
 184 to the ASR gel viscosity, its surface tension and the size of the pores of the aggregate and of
 185 the cement paste surrounding the reactive sites [39]. Once the gel reaches t_c , it cannot
 186 penetrate in the paste anymore due to the combination of these three main parameters. They
 187 were assumed to be the same for all the aggregates and that's why the equivalent thickness t_c
 188 was assumed to be independent with the aggregate size.

189 In order to assess the effect of ASR-gel on the cement paste in term of pressure, similar
 190 relationship can be used. p_g^a , the gel pressure can be assumed to be proportional to M_g the gel
 191 bulk modulus and to the increase of volume due to ASR-gel production:

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

192 V_a^{gel} , the volume of ASR gel formed in reactive aggregates, is proportional to the number of
 193 moles of ASR gel formed after the attack of the reactive silica:

$$V_a^{gel} = n_a^{gel} \cdot V_{gel}^{mol} \quad (4)$$

194 with n_a^{gel} (mol): the number of moles of ASR gel produced by the aggregate a and V_{gel}^{mol}
 195 (m^3/mol): the molar volume of the gel.

196 The number of moles of ASR gels produced by the reaction is defined by the number of
 197 moles of silica attacked by alkalis and can be taken proportional to s the reactive silica content
 198 (in mol/ m^3):

$$n_a^{gel} = \frac{4}{3} \pi R_a^3 \cdot s \cdot \zeta(t) \quad (5)$$

199 $\zeta(t)$ is the chemical advancement of the alkali-silica reaction (which depends on temperature,
 200 moisture and alkali conditions and is assessed by the diffusion of ionic species in the
 201 aggregate and by the time necessary for the attack of the reactive silica by hydroxyl ions and
 202 for the formation of ASR-gels [34]).

203 Finally, p_g , the gel pressure at the time-step t , can be assessed from:

$$p_g^a(t) = M_g \left\langle \frac{4}{3} \pi R_a^3 \cdot V_{gel}^{mol} \cdot s \cdot \zeta(t) - \varphi \frac{4}{3} \pi [(R_a + t_c)^3 - R_a^3] \right\rangle \quad (6)$$

204 4. The pressure causes cracking of the aggregate and cement paste. Whatever the reactive
 205 silica distribution in the aggregate, the propagation of cracks in small specimens containing
 206 large aggregate can be rapid and can occur for low pressures, while the propagation is more
 207 difficult for larger specimens containing smaller aggregates and needs higher pressure (Figure
 208 5-d). In the framework of fracture mechanics [44-45], the maximal normal stress in the
 209 vicinity of an inclusion (e.g. the aggregate) that induces pressure on a matrix (e.g. the cement
 210 paste) is given by:

$$\sigma(r) = \frac{K_I}{\sqrt{2\pi r}} \quad (7)$$

211 With σ , the maximal normal stress at the point M located at distance r from the edge of the
212 inclusion (Figure 5-d) and K_I , the stress intensity factor obtained for a specimen in stress-free
213 conditions from the relation:

$$K_I = p_g^a \cdot f\left(\frac{R_a}{L}\right) \quad (8)$$

214 where f is a function increasing with the ratio R_a/L (which can be obtained in [41]), with R_a
215 the aggregate radius and L the dimension of the specimen (a : superscript relative to the size of
216 the reactive aggregate).

217 ***Smaller expansion in smaller specimen***

218 The larger the aggregate size compared to the specimen size, the larger the normal stress at a
219 given distance from the aggregate (Equations 7 and 8). Thus, the tensile strength at the
220 boundary of the specimen can be reached for a smaller pressure (Figure 5-d) and cracks can
221 be initiated earlier. Once concrete cracks, ASR-gels can be accommodated by cracking
222 without creating supplementary pressure [46,47] and can even leach off through the porosity
223 induced by cracking. Thus, the pressure in the gels falls, which stops the expansion.

224 If the pressure necessary to cause cracks is lower for small specimens containing large-sized
225 aggregate, the pressure of the gel will fall sooner in the case of the 20x20x160-mm specimens
226 containing the 1250-2500- μm aggregate than for the 70x70x280-mm specimens containing
227 the same aggregate, and thus cause smaller expansion.

228 ***Pessimum effect or scale effect***

229 The scale effect can also explain the pessimum effect of ASR expansion with aggregate size.
230 For the largest aggregates, a part of the difference of expansion can be explained by a delay in
231 the attack of the reactive silica by the hydroxyl ions due to the diffusion of the ions into the
232 aggregate [23]. Measurements confirmed that the expansion rate was slower in the largest
233 particles whatever the size of the specimens (Figure 3). However, even when the final value
234 was reached, ASR expansion remained lower for the largest aggregates in the smallest

235 specimens. As explained above, the 20x20x160-mm specimens containing the 1250-2500- μ m
236 aggregate were more affected by the scale effect than the specimens containing the 315-630-
237 μ m aggregate: a larger aggregate induced a greater stress intensity factor (Equation 8).
238 Therefore, the part of reactive silica consumed when cracking appeared is lower when
239 cracking appears in the specimens containing the largest aggregate. Once cracking occurs, the
240 gel is accommodated by the cracks and the gel produced after cracking leads to little
241 supplementary pressure. Expansion ceases before the chemical reaction stops, giving a final
242 expansion lower than that for the largest aggregate. This is consistent with the experimental
243 determination of the degree of reaction performed with chemical attack [47] and SEM image
244 analysis [48]: expansion stopped while the degree of reaction was still increasing. The
245 pessimum effect versus specimen size is not an intrinsic phenomenon; it is due to the scale
246 effect, which depends on the 'specimen size to aggregate size' ratio. In this experimentation,
247 only mortars were tested and the pessimum effect disappeared for aggregate of 1.25-2.5 mm
248 when tests were performed in specimens of 70 mm. For concrete, the pessimum still exists for
249 aggregate of 4-8 mm in specimens of 70 mm [24]. If the scale effect (combination of the
250 aggregate size effect and the specimen size effect) is identical for concrete, specimens of more
251 than 250 mm could be necessary to remove the pessimum effect for such aggregates.
252 Supplementary investigations are necessary to analyse this effect on concrete.

253 *Effect of the reactive silica content on the scale effect*

254 At last, it can also be used to analyse the effect of the reactive silica content on ASR
255 expansion (Figure 1). As shown in Figure 6, the final expansions measured on specimens
256 containing the different aggregates were not proportional to the reactive silica content.
257 Moreover, the specimens containing opal were much more damaged and cracked than those
258 with the other aggregates. This could have been due to the large concentration of reactive
259 silica contained in opal. A larger reactive silica content would induce a higher gel pressure

260 (Equation 6) and thus a greater stress intensity factor (Equation 8) for opal than for the
261 siliceous limestone. Therefore, the ratio between the total reactive silica and the reactive silica
262 consumed when cracking appeared was not the same. The part of reactive silica consumed
263 would be lower when cracking appeared in the specimens containing opal than in the
264 specimens containing the siliceous limestone. Once cracking had occurred, cracks
265 accommodated the gel, and expansion stopped before the chemical reaction finished, without
266 proportionality with the total reactive silica. This effect was probably very important in the
267 case of specimens containing opal in which cracks were very large at the end of the
268 experiment (Figure 2).

269 The concept of stress intensity factor appears to be important for an understanding of the
270 development of ASR expansion in concrete. The larger the stress intensity factor K_I is, the
271 faster cracks appear and the cracking could lead to reduction of the expansion through gel
272 accommodation and exudation. The ratios between specimen and aggregate sizes were very
273 high in these experiments. This shows that it could be difficult to use specimens large enough
274 to avoid the scale effect. One possibility would be to try to control the aggregate and the
275 specimen sizes so as to perform the test with similar stress intensity factors and thus to have
276 similar cracking conditions as proposed in [42,43] but this would be difficult to do,
277 particularly as far as controlling the aggregate size is concerned. Another possibility is to
278 understand the scale effect through a model that can be used to analyse expansion tests. This
279 is the aim of the modelling presented in the next part.

280 **3. Modelling ASR expansion**

281 The model used in this paper is an improvement on the microscopic model [34]. It was based
282 on previous models [33, 35-37,39] and attempted to predict the damage and the expansion of
283 a Representative Elementary Volume (REV) of concrete containing a mix of reactive

284 aggregates of different sizes. The reaction between the reactive silica and the alkali was
285 determined through the mass balance equation, which describes the diffusion mechanism in
286 the aggregate and the fixation of the alkali in the ASR gels. The mechanical part of the model
287 is based on damage theory in order to assess the decrease of stiffness of the mortar due to
288 cracking caused by ASR and to calculate the expansion of the REV [34]. Some modifications
289 were made in this model: the threshold of alkalis was re-evaluated in accordance with the
290 results present above and the combined effects of specimen and aggregate sizes and reactive
291 silica content on expansions were taken into account.

292 **3.1 Presentation of the model**

293 **3.1.1 Diffusion of alkali**

294 The diffusion of ions in the aggregate partly controls the kinetics of the chemical attacks and
295 of the expansion. At the beginning of ASR, hydroxyl ions attack the reactive silica of the
296 aggregate. Hydroxyl ions come from the pore solution and thus the external boundary of the
297 reactive aggregates is attacked first. If the reactive silica is uniformly distributed in the
298 aggregate, the reactive silica closest to the pore solution is attacked first (Figure 5-a top). If
299 the reactive silica occurs in veins, the penetration of hydroxyl ions is not uniform but veins in
300 contact with the pore solution are attacked first (Figure 5-a bottom). Therefore, in both cases,
301 the transport of ionic species in the aggregate can be modelled by an equivalent diffusion of
302 ions from the pore solution to the aggregate core. The diffusion is not the only phenomenon
303 which impacts the kinetics of ASR-expansion: the time necessary for the attack of the reactive
304 silica by hydroxyl ions and for the formation of ASR-gels is taken into account through a
305 depletion term in the mass balance equation [34]. This term models the alkali consumed by
306 the ASR-gel formation. This alkali consumption is assumed to be linear with the alkali
307 concentration in the aggregate. The temperature of storage for the experiment was 60°C. It
308 accelerates ASR considerably compared to the environmental conditions of real damaged

309 structures and can affect the nature of the gel. It impacts the values of the kinetic parameters
310 but should not affect the principle of the modelling.

311 As in the previous model [34], only the diffusion of ions in the aggregate was considered
312 since the kinetics of diffusion in aggregates was much slower than in cement paste [35].
313 Therefore, the alkali concentration at the surface of aggregates was assumed to be equal to the
314 concentration of the NaOH solution. The kinetics of the expansion was thus partly controlled
315 by the diffusion in the aggregate, which is one of the kinetic parameters of the model. This
316 assumption is relevant for small specimens or when there is no chemical exchange with the
317 environment but it can be discussed for larger specimens immersed in NaOH solution. In the
318 experiments presented here, the largest specimens showed the slowest rate of ASR-expansion
319 (Figure 3). This can be explained by the diffusion of the alkali into the specimens. The initial
320 alkali concentration in the pore solution was sufficient to initiate the reaction. Alkalis were
321 quickly consumed by the ASR gel and supplementary alkalis were necessary to maintain gel
322 production. The alkali came from the solution and diffused into the mortar. Thus, more time
323 was necessary for alkali to reach the centres of the largest specimens than the centres of
324 smaller ones. Thus an alkali gradient appeared in the specimens. In consequence, at a given
325 time, a gradient of expansion existed between aggregates located close to the external
326 boundary and aggregates located in the core of the specimen. Only complete discretization of
327 the whole specimen [24,31,32] would allow this difference of expansion to be considered.
328 The aim of the modelling used in this paper was to evaluate the material behaviour that could
329 be represented by measurements performed on small specimens. Therefore, the phenomenon
330 of diffusion in the specimen was not taken into account. This modelling can give a good
331 representation of the expansion kinetics of concrete damaged by ASR measured on small
332 specimens (with width smaller than 40 mm, for which the effect of the alkali gradient is
333 negligible).

334 **3.1.2 Threshold of alkali concentration**

335 In the previous model, a threshold of alkali concentration of 0.625 mol/l of Na⁺ was
336 considered, under which ASR did not occur [34]. It was based on experimental data showing
337 that, with an alkali content lower than 3 kg per m³ of concrete, no ASR-expansion was
338 observed [49-52]. In tests performed in non-saturated conditions (in air with RH above 95%),
339 the concentration of alkali played an important role in the attack of the silica. In the present
340 work, all the specimens were kept immersed in alkali solution and, thus, the alkali was
341 supplied in abundant quantities. The experiments performed on specimens immersed in three
342 alkali concentrations (0.77, 1 and 1.25 mol/l) showed negligible differences. Considering a
343 threshold of 0.625 mol/l, the expansion of the mortars kept in 0.77 mol/l solution should be
344 significantly slower than the expansion of the mortars kept in 1.25 mol/l solution. With this
345 threshold, the alkali concentration in the aggregate has to be higher than 0.625 mol/l before
346 the reactive silica is attacked. The gel production kinetics is then proportional to the
347 difference between the alkali concentration in the paste and the threshold. The alkali
348 concentration gradient between the paste and the aggregate remains too small to induce the
349 same reaction speed as for the other concentrations. This is not in accordance with the
350 expansion kinetics measured on specimens (Figure 1). Therefore, the gradient must be close
351 for the three concentrations and consequently the alkali consumption by the silica must begin
352 as soon as alkalis are present in the aggregate, without a threshold. The result of this
353 assumption is shown in Figure 7: if no threshold is taken into account, the expansions
354 determined by the modelling at 50 days are of the same order as the expansions obtained for
355 the measurements. The apparent threshold effect observed in experimentations [49-52] can be
356 explained by the alkali fixation in C-S-H. This fixation consumed a part of alkali which are no
357 more available in the pore solution and reduced the attack kinetics and consequently the ASR-

358 expansion kinetics. All these phenomena can be explained without considering a threshold of
359 alkali concentration for the attack of the silica.

360 **3.1.3 Effective ASR-gel**

361 The ASR expansion calculated by the model is imposed by the effective volume of ASR-gel.
362 The effective volume of gel is deduced from the total volume of gel, which is proportional to
363 the number of moles of ASR gel formed after the attack of the reactive silica (Equation 7).
364 Once cracking appears in the specimens, a part of the rest of the gel $\langle V_a^{gel} - V_a^{poro} \rangle$ is
365 assumed to be accommodated by cracks created in the aggregate and in the cement paste by
366 the ASR-gel pressure. This gel accommodation by cracks stops the increase of the ASR-gel
367 pressure. This part depends on the scale effect: the larger the aggregate in comparison to the
368 specimen, the greater the volume of ASR gel accommodated. This phenomenon is also
369 affected by the reactive silica content of the aggregate. As explained in the analysis of the
370 experiments, the greater the reactive silica content is, the stronger is the non-linearity due to
371 cracking. Fracture mechanics concepts show that the aggregates closest to the external
372 boundary are the first to produce cracks. In order to quantify this effect, it can be assumed that
373 these aggregates lead to less pressure than the aggregates located in the core of the specimens.
374 As for the diffusion in cement paste, this will lead to a gradient of deformation between the
375 external boundary and the core and thus to internal stresses. The scale effect appears to be a
376 highly non-linear phenomenon. Therefore, an exponential function is proposed to model the
377 consequences on ASR expansions. An empirical relationship to quantify the reduction of the
378 gel amount effectively used to assess the pressure is thus assumed and applies equally to all
379 the aggregates of a given size i without consideration for their location compared to the
380 specimen boundary:

$$V_a^{eff} = \langle V_a^{gel} - V_a^{poro} \rangle^+ \times \max \left[\exp \left(-c \frac{2R_a}{L} s^x \right) \right] \quad (9)$$

381 With V_a^{eff} : the effective volume of gel, c : the scale effect fitting coefficient and χ : the silica
382 content exponent.

383 The pressure imposed on the aggregate and thus on the cement paste depends on the effective
384 volume of gel. Moreover, if the distribution of the reactive silica is uniform in the aggregate,
385 the pressure will be isotropic (Figure 5-c). If the reactive silica is contained in veins in the
386 aggregate, the large number of aggregate particles in the specimens means that the orientation
387 of the veins in the specimens is randomly distributed and the mean resulting pressure can be
388 considered as isotropic too (Figure 5-c). The effective volume of gel is then used in the
389 mechanical model presented in [34] to deduce the resulting expansion.

390 **3.2 Comparison with experiments**

391 **3.2.1 Parameters**

392 Table 2 sums up the parameters used in the modelling, stating the symbols, the methods used
393 for the identification, the values and the units. Three parameters (unique, independent of the
394 other variables, and usable for all experiments and all aggregate types studied in this work) of
395 the physicochemical modelling (thickness of the connected porous interface zone t_c , molar
396 volume of ASR gel V_{gel}^{mol} , and scale effect fitting coefficient c – equations 4, 6 and 9) were
397 obtained by curve fitting on the final expansions measured on the specimen of size 70 x 70 x
398 280 mm with the four different size classes (0-315, 315-630, 630-1250 and 1250-2500 μm) of
399 siliceous limestone (SL – Figure 8). The parameters were first determined without
400 considering the effect of the silica content exponent ($\chi = 0$) since the specimens involving the
401 curve fitting contained the same aggregate. Once these three parameters had been assessed,
402 the value of χ was determined by fitting the final expansion of mortars containing opal as
403 aggregate (O – Figure 8). As explained above, the coefficient of alkali diffusion in aggregate,
404 D_a , was used as a kinetic parameter. This coefficient can depend on the nature of the
405 aggregate. It was obtained by curve fitting the results obtained on specimens of size 20 x 20 x

406 160 mm (Figure 9). The coefficients of diffusion in aggregate (D_a) thus obtained by curve
407 fitting were respectively equal to 2.0×10^{-15} , 5.0×10^{-14} , 4.0×10^{-16} and 2.0×10^{-16} m²/s for
408 the siliceous limestone, opal, quartzite and quartz aggregate. It can be noted that the values
409 were considerably lower than the usual value of diffusion coefficient determined for cement
410 paste (about 10^{-12} m²/s); the assumption of fast alkali supply at the surface of aggregate is thus
411 verified. However, these values appear to be very small compared to coefficient of diffusion
412 measured for quartzite [53]. It can be explained by the assumption used to determine the
413 kinetics of the formation of ASR-gels. In this modelling, the kinetics was driven by two main
414 phenomena: the diffusion of ions in the aggregates and the kinetics of the attack of reactive
415 silica by alkali and hydroxyl [34]. The attack was assumed to be linear with the alkali
416 concentration in the aggregate and only one parameter was used for all the aggregates. This
417 parameter was fitted for siliceous limestone [34]. In the reality, the mechanisms leading to the
418 gel formation are more complex and this simplified assumption can be responsible of the
419 overestimation of the speed of the silica attack. It can lead to underestimation of the speed of
420 diffusion. Improvements of the assumed kinetics of silica attack could lead to more realistic
421 coefficients of diffusion for aggregates. However, this kinetics of silica attack could depend
422 on the nature of the reactive silica and on the pH of the pore solution and several parameters
423 could be necessary to obtain relevant results which could be very difficult to measure.

424 **3.2.2 Calculations**

425 Expansions obtained by modelling are compared with the measurements in Figures 9 and 10.
426 Figure 9 shows the expansions obtained with 20 x 20 x 160-mm specimens containing two
427 aggregate sizes (15% of 315-630 and 15% of 630-1250 μ m) for the four natures of aggregate.
428 Only the coefficient of diffusion and the silica content exponent were fitted on these results.
429 The fitting concerning the final expansion of the siliceous limestone was only performed on
430 the largest specimens (70x70x280 mm) and for specimens containing only aggregate of the

431 same size. Except for the kinetics of expansion obtained for specimens containing opal, both
432 kinetics and final expansions obtained by the calculations were in accordance with the
433 measurements for the four types of aggregate. The description of the expansion kinetics by
434 the model is globally possible: presence of a latent time before initiation of the expansion,
435 followed by a high rate of expansion and ending by a low rate to reach the final expansion.

436 Concerning the effect of the alkali concentration of the immersion solution, the difference
437 between the final expansion predicted by the model (0.70%) and the mean measured
438 expansion (0.65%) is lower than 10%. The calculation does not show any difference in final
439 expansions between the three alkali concentrations: the model assumes that all the reactive
440 silica is consumed in the three conditions and, thus, the volume of ASR gel created is the
441 same, leading to the same ASR-expansion. The expansion rates obtained by the model at the
442 beginning of expansion are in good agreement with experiments. This confirms that, in the
443 case of specimens immersed in alkali solutions, no alkali threshold has to be considered
444 (unlike for specimens exposed to a saturated environment [34]).

445 The curve obtained by the model for opal did not fit the experimental results well. At the
446 beginning of the experiment, the slopes of the two curves were the same but, after about 50
447 days, the measurements showed a speeding-up of the expansions which was not obtained by
448 the calculation. This can probably be attributed to crack opening for opal specimens as the
449 specimens containing opal aggregate presented much larger cracks than specimens of the
450 same size cast with the other aggregates. First, cracks opening increased the apparent
451 expansion. These cracks could also cause a great increase in diffusion in the specimens, which
452 could accelerate the reaction.

453 The comparison of the calculated final expansions obtained for the three specimen sizes (20 x
454 20 x 160 mm, 40 x 40 x 160 mm, 70 x 70 x 280 mm) and the four aggregate sizes of the
455 siliceous limestone with the measurements is shown in Figures 10 and 11. Concerning these

456 values, only the measurements performed on the largest specimens were used for parameter
457 identification. The calculated results for all the combinations are in good accordance with the
458 measurements (Figure 10). The scale effect of ASR expansion pointed out in
459 experimentations is rather well-predicted (Figure 11). The differences between calculated and
460 measured expansions are lower than 15% except for the expansions obtained on the smallest
461 specimens (20x20x160 mm) containing the largest aggregate (1250-2500 μm), where the
462 difference is about 30% (Figure 10).

463 **3.2.3 Discussion**

464 The molar volume of ASR gel obtained by curve fitting in this work was about 170 cm^3/mol .
465 It was higher than the molar volume obtained in the previous modelling performed for
466 specimens kept in air at 95% relative humidity [34] and higher than the molar volume of C-S-
467 H (about 100 cm^3/mol [54]). The differences can be explained by the conditions of
468 conservation (in saturated air in [34] and immersed in NaOH solution in this work) which
469 could have had a marked effect on the gel morphology and thus on the gels' capability to
470 absorb water. The ASR gels were produced in pore solution with different water contents and
471 alkali concentrations. The compositions of the gel were therefore different and thus the molar
472 volume was different. Concerning the water, ASR gels formed in specimens kept in solution
473 can absorb much more water than those in humid air (R.H. >95%).

474 The thickness of the connected porous interface zone (t_c equal to 14.0 μm) was also higher
475 than that found in [34], which was about 1 μm . This value was determined to obtain the final
476 expansions of four aggregate sizes while only two aggregate sizes were studied in the
477 previous work. The molar volume of the gel was higher and therefore more connected porous
478 volume was necessary to accommodate the gel. The ASR gels could permeate over a long
479 distance because the molar volume was high and because the viscosity was small due to high
480 water and alkali contents. This value is in accordance with the results found in the literature

481 [55-56] showing that the interfacial transition zone between aggregate and cement paste can
482 reach 20 μm .

483 **4. Conclusion**

484 This paper has aimed to investigate and quantify the combined effects of aggregate and
485 specimen sizes on ASR expansion and the influence of the reactive silica content of aggregate
486 on this effect. In order to quantify these effects on ASR expansion, a microscopic model was
487 improved. The main conclusions can be summarized as follows:

488 1. A scale effect, combining the effects of aggregate size, reactive silica content and
489 specimen size on ASR expansion, has been highlighted by experimentation. A
490 temporary pessimum size effect had previously been shown to exist due to the speed
491 of the attack of the reactive silica on the aggregate: the larger the aggregate, the slower
492 the penetration of ions into the aggregate and thus the slower the expansion [23]. In
493 this paper, only stabilized expansions were compared, which pointed out another
494 pessimum effect. The pessimum size effect of stabilized expansion appears not to be
495 an intrinsic phenomenon of ASR expansion but to depend on the size of the specimen
496 used to perform the expansion test. For the largest specimens, no pessimum effect was
497 detected for the aggregate size used in this study. The scale effect has been explained
498 by fracture mechanics concepts: the larger the ratio between the aggregate and
499 specimen sizes, the larger the stress intensity factor around the aggregate and the faster
500 the cracking around the aggregate. After cracking, a part of the ASR gels can be
501 accommodated by cracks.

502 2. Considering that the scale effect is unavoidable for experimental specimens (the ratio
503 between specimen and aggregate sizes should be larger than 100 to significantly
504 decrease the effect), the results of expansion tests should be analysed with respect to

505 this effect. The modelling proposed took the scale effect into account through an
506 empirical relationship considering underlying linear fracture mechanics concepts.
507 Calculations are in good accordance with experiments for expansion tests performed
508 with ratios higher than 10 between specimen and aggregate sizes for the four
509 aggregates studied.

510 Finally, this paper points out the complexity of ASR expansion and states the numerous
511 parameters that have to be taken into account to obtain relevant calculations. Another
512 important conclusion is that stress-free expansions obtained on specimens cannot be directly
513 used as input parameters in structural models because they are strongly dependent on the
514 specimen size and on the conservation conditions during the expansion test. To be used,
515 stress-free expansion test results should be interpreted by considering the phenomena pointed
516 out in this work. Such an approach is possible: free expansion tests can be carried out to
517 assess the AAR advancement in aggregates, while the expansion in the conditions of the
518 damaged structures is assessed through a finite element inverse analysis able to combine the
519 fitting of the expansion measured on structures with the chemical advancement kinetics
520 deduced from laboratory tests [57].

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678 **TABLES**

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Table 1: Reactive silica contents of different aggregates [43]

<i>Reactive silica (SiO₂)</i>	<i>SL</i>	<i>O</i>	<i>Q</i>	<i>QA</i>
<i>Percentage by mass (%)</i>	6.9	50.4	7.6	2.7
<i>Content (mol/m³ of aggregate)</i>	3000	21900	3300	1170

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Table 2: Parameters of model

Parameter	Symbol	Identification	Value	Units
<i>Aggregate</i>				
Reactive silica content	<i>s</i>	measurement	D.N.A.*	mol/m ³
Coefficient of diffusion	<i>D_a</i>	curve fitting	D.N.A.*	m ² /s
Porosity	<i>p</i>	usual value	0.01	%
<i>Paste</i>				
Porosity of mortar	<i>p_{mort}</i>	measurement	18.0	%
Thickness of the connected porous interface zone	<i>t_c</i>	curve fitting	14.0 x 10 ⁻⁶	m
<i>Gel</i>				
Molar volume of ASR gel	<i>V_{gel}^{mol}</i>	curve fitting	1.7 x 10 ⁻⁴	m ³ /mol
Scale effect coefficient	<i>c</i>	curve fitting	0.037	(mol/m ³) ^{-χ}
Silica content exponent	<i>χ</i>	curve fitting	0.75	-

684 * depends on the nature of the aggregate

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686

687 **FIGURES**

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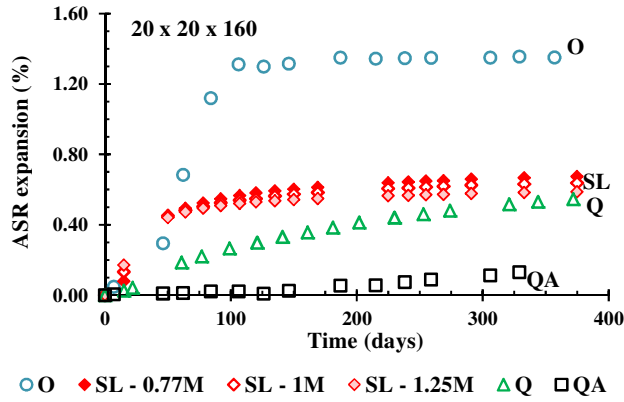


Figure 1: ASR expansions according to the alkali concentration of the immersion solution for SL (0.77, 1.0 and 1.25 mol/l) and to the nature of the aggregate (O: opal, SL: siliceous limestone, Q: quartzite and QA: quartz aggregate) stored in the 1 mol/l NaOH immersion solution

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(a)



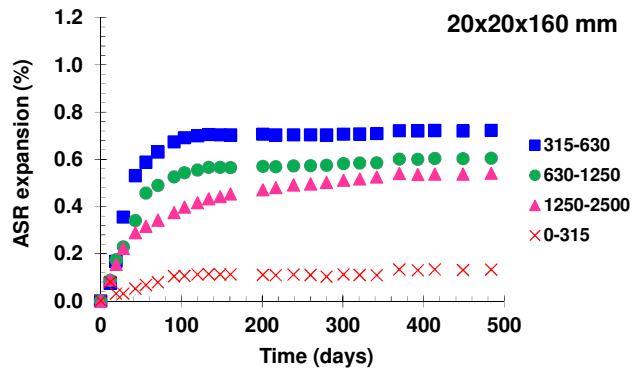
(b)

Figure 2: Cracking patterns of specimens cast with the siliceous limestone (a) and with opal (b)

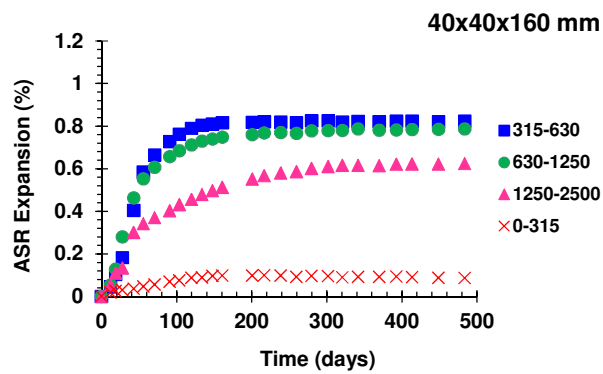
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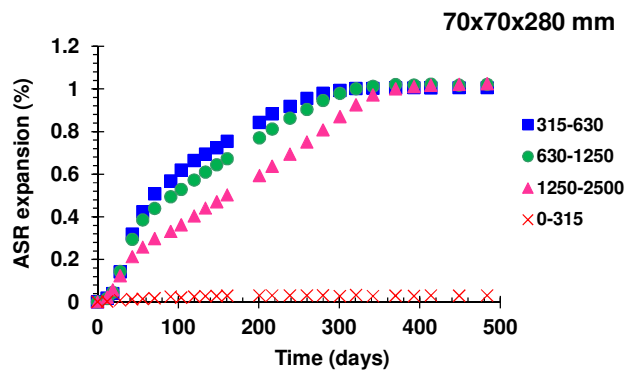
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(a)



(b)



(c)

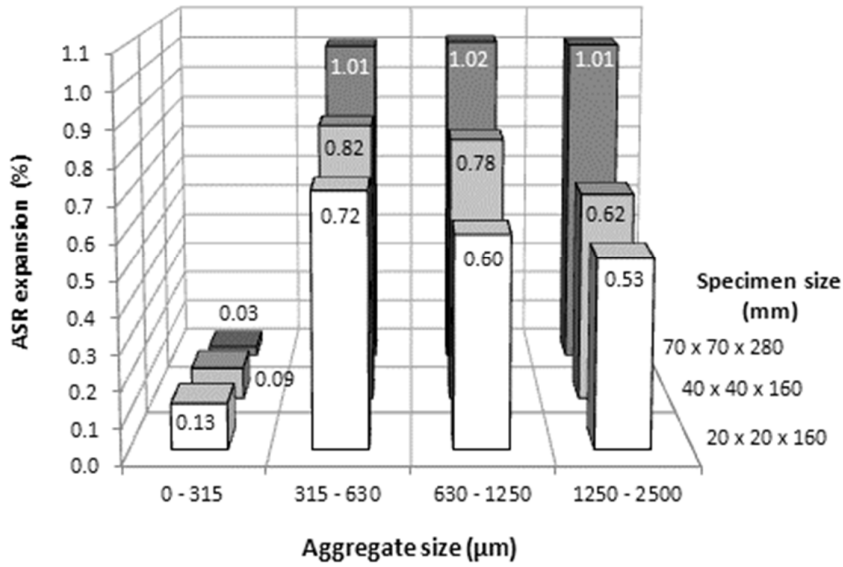
Figure 3: ASR expansions on prismatic specimens 20x20x160 mm (a), 40x40x160 mm (b) and 70x70x280 mm (c)

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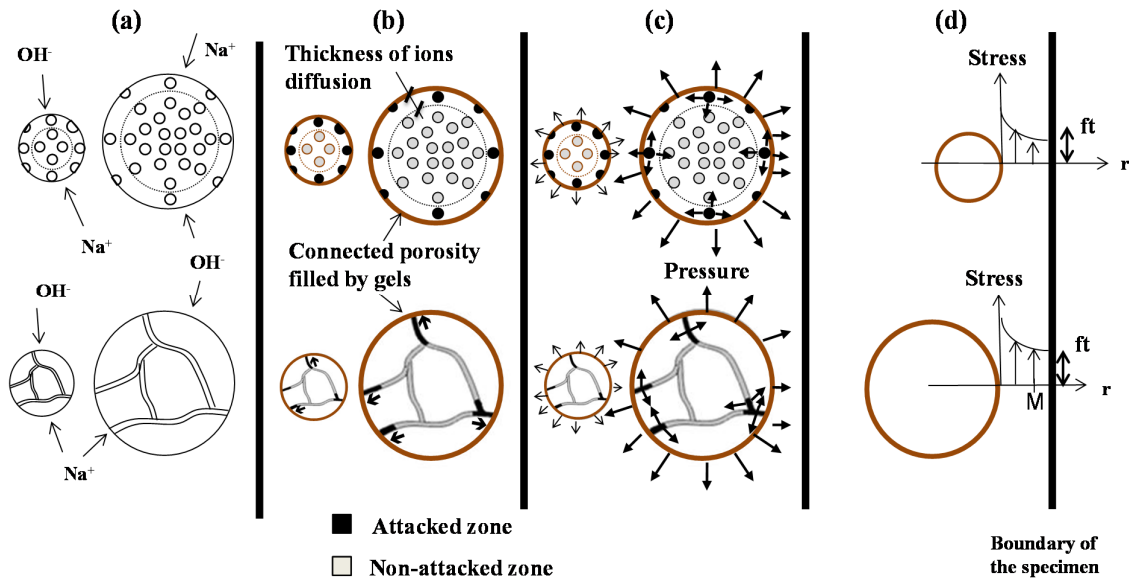
Figure 4: Final ASR expansions according to specimens size and aggregates size

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Figure 5: Four phases of ASR development: ion diffusion (a), reaction with the reactive silica (b), gel pressure (c), and stress development (d). Top line: diffuse reactive silica distribution, bottom line: veins

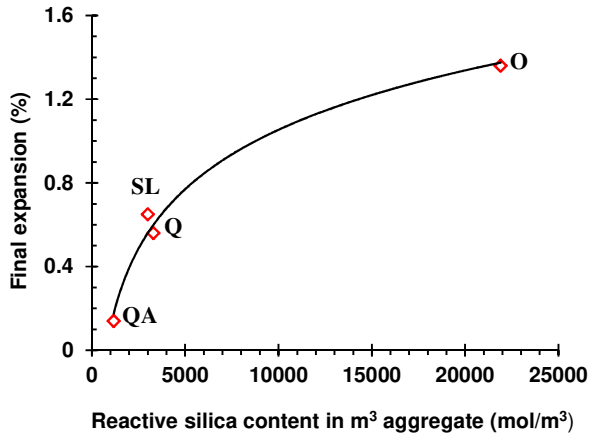


Figure 6: Final expansions according to reactive silica content

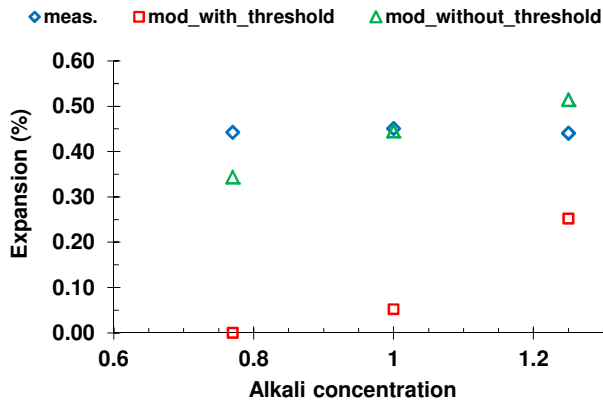


Figure 7: Measured and calculated expansions at 50 days according to the alkali concentration of the solution (calculations performed with or without the threshold of 0.625 mol/l)

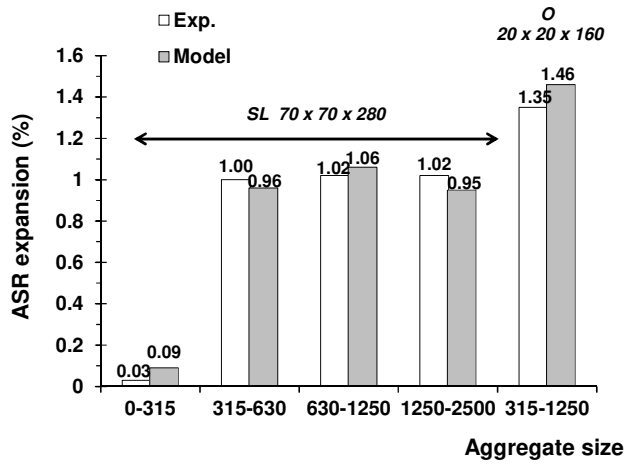


Figure 8: Identification of the modelling parameters

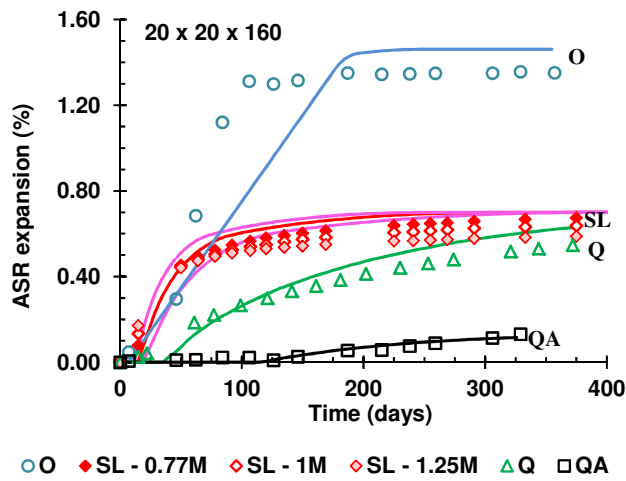
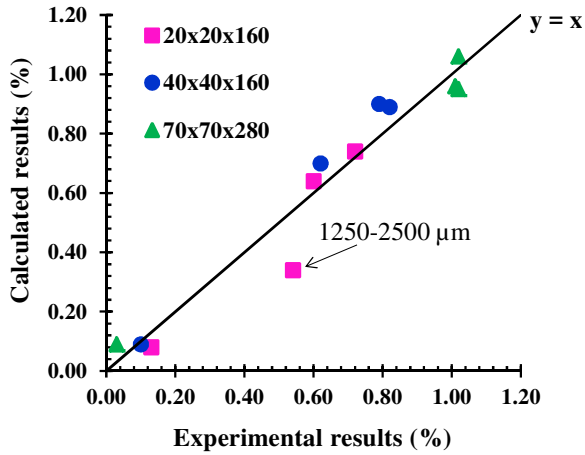


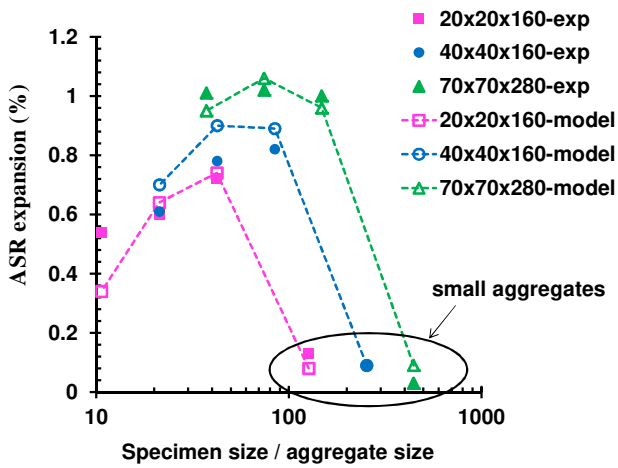
Figure 9: Comparison of calculated and measured expansions according to the alkali concentration of the immersion solution (0.77, 1.0 and 1.25 mol/l) and to the nature of the aggregate (O: opal, SL: siliceous limestone, Q: quartzite and QA: quartz aggregate)

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Figure 10: Comparison of calculated and measured final expansions for all the combinations between the sizes of aggregates and specimens with siliceous limestone



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Figure 11: Scale effect of ASR expansion: experimental and modelling results