EFFECTS OF AGGREGATE SIZE AND ALKALI CONTENT ON ASR EXPANSION

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Abstract

Attempts at modeling ASR expansion are usually limited by the difficulty of taking into account the heterogeneous nature and size range of reactive aggregates. This work is a part of an overall project aimed at developing models to predict the potential expansion of concrete containing alkali-reactive aggregates. The paper gives measurements in order to provide experimental data concerning the effect of particle size of an alkali-reactive siliceous limestone on mortar expansion. Results shows that no expansion has been measured on the mortars using small particles (under $80 \,\mu\text{m}$) while the coarse particles (0.63-1.25 mm) gave the largest expansions (0.32%). When two sizes of aggregate were used, ASR-expansions decreased with the amount of small particles. A model is proposed to study correlations between the measured expansions and parameters such as the size of aggregates and the alkali and reactive silice contents.

Keywords: particle size, alkali content, model, reactive silica.

1 INTRODUCTION

Reassessment of Alkali-Silica-Reaction-damaged structures (bridges and dams) is of first importance for engineering structures owners. The gel volume formed by the chemical reaction can be used as input data of structural models [1]. One of the main difficulties is to assess this gel volume [2]. Microscopic models [3-6] could be one method of assessing it. Such models should be able to predict the differences of expansions with the evolution of all influencing parameters (size of aggregate, silica content, alkali content...) and have to be compared with experimental results.

Numerous papers deal with the effect of particle size of reactive aggregates on the expansion due to ASR. Experimentations have been performed on several types of aggregates. It seems that the aggregate size causing the highest ASR expansion depends on the nature and composition of the aggregate. Significant differences have been observed between rapid and slow alkali-reactive aggregates. Opal was one of the earliest and most widely used aggregates in laboratory studies of the size effect [7-11]. Investigations on the size effect have also been performed with different kinds of silica glass, fused silica, waste silica glass, andesite, siliceous limestone, quartzite, greywacke, chert, mylonite, flint and sandstone [12-17]. In spite of all these studies, it is difficult to generalize about the effect of the particle size of reactive aggregates, since conflicting results exist concerning the most damaging size which leads to the highest ASR expansion. All the results available in the literature were obtained using different experimental conditions and the coupled effects with other important parameters, such as Na/Si ratio, have been often neglected.

A few papers deal with the effect of size for reactive siliceous limestone [6-8, 18]. This type of aggregates has been used in many damaged structures in France. Therefore, in order to test models to predict the potential expansion of concrete containing such alkali-reactive aggregates, tests have been performed to provide experimental data concerning the effect of particle size of an alkali-reactive siliceous limestone on mortar expansion. The paper presents the experimental results (expansion measurements performed during more than 500 days) and gives data necessary for model development.

16 mix-designs have been studied and a special attention has been paid to the proportions of alkalis (Na_2O_{eq}) in the mixtures and reactive silica in the aggregate. First, the paper presents the experimental conditions of the tests. Then, the measurements of ASR-expansions are presented in two parts: experiment on mortars containing one reactive size (0-80 µm, 80-160 µm, 160-315 µm, 315-

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 $630 \,\mu\text{m}$; $630-1250 \,\mu\text{m}$, $1250-2500 \,\mu\text{m}$) and experiment on mortars containing mixes of two size of aggregate (0-80 μm and $1250-3150 \,\mu\text{m}$) with increasing 0-80 μm reactive aggregate content. At last, a model is presented to analyze the experimental results.

2 EXPERIMENTAL CONDITIONS

2.1 Materials

The cement used was a standard CEM I 52,5R with a specific gravity of 3.1 and a specific surface area (Blaine) of 400 m²/kg. Its chemical composition is given in Table 1. The aggregates were crushed sands (jaws crusher): a non-reactive marble (NR) and a reactive siliceous limestone (R). The chemical composition of aggregates NR and R are given in Table 1. The non-reactive marble was mainly composed of calcite. The reactive siliceous limestone contained mostly calcite and quartz, with traces of dolomite, feldspars and phyllosilicates. In order to control the particle size distribution of aggregates in the mortars, the aggregate samples were divided into several particle size fractions: 0-80, 80-160, 160-315, 315-630, 630-1250, 1250-2500 and 1250-3150 μ m). Details of the aggregate combinations are given in 2.3.

2.2 Methods

Expansion was measured on mortar prisms (2x2x16cm) with a sand (1613 kg/m^3) to cement (538 kg/m³) ratio of 3. The mortar prisms were kept in sealed bags at 20°C for 27 days after demoulding (24 hours after casting). The prisms were stored at 60°C, placed on grids in watertight containers containing 20 mm of water (mortar bars were not in contact with water) 28 days after mixing. Salt (K₂SO₄) was added to the water (above saturation) in order to maintain a relative humidity above 95% in the containers and to try to avoid condensation on specimens. Expansion was measured using the scale micrometer method (specimens had shrinkage bolts in the two extremities). Each measurement was the mean of three values from three replicate specimens. Expansion measurements were performed after the containers and the prisms had been cooled for 24 hours at 20°C.

In order to reach distinct Na/Si ratios, mixtures were adjusted to alkali contents (Na₂O_{eq}) of 6.2, 8.1 and 9.9 kg of alkali per m^3 of mortar by adding NaOH in the mixing water. An alkali-free superplasticizer was used (0.5% dry matter of cement mass) in mortars to achieve a proper set in the molds.

For the first part, the water-cement ratio was 0.5. For the second part, the largest content (40%) of fine reactive particles (0-80 μ m) absorbed too much water during the casting, the mortar was too dry to be cast with a ratio of 0.5 (even with a superplasticizer). Therefore, the water-cement ratio has been increased to 0.6.

2.3 Experimental program

Size effect

The first experimentation presented in this paper concerns the study of size effects of the reactive siliceous limestone on ASR expansions. In this part, six particle size fractions were studied: 0-80, 80-160, 160-315, 315-630, 630-1250 and 1250-2500 μ m in mortars M1, M2, M3, M4, M5, M6, respectively. These mortars contained 8.1 kg of alkali per m³ of mortar. In order to obtain significant expansions with the aggregate studied in this paper, the content of reactive aggregate had to be at least 30%. The particle size distributions were obtained by adding 30% of reactive aggregate of the six different fractions to 70% of a continuous size distribution (0-2500 mm) of non reactive aggregate. Therefore, the particle size distributions are different for the six mortars. The effect of such differences in particle size distribution on the porosity of mortars has been studied on mortars containing only non reactive particles. Measured porosities, using AFPC-FREM method [19], lied between 17.3 and 18.2%. The differences did not appear to be significant compared to difference of 1% which can be obtained during measurements made on three specimens of a same mortar. Porosity will be measured on the reactive mortars at the end of the measurements of expansions and can be considered as input data for model analysis.

Effect of fine reactive aggregate

For the second part of the experimentation, reactive particles of two sizes were used: 0-80 μ m and 1250-3150 μ m, on mortars containing 6.2 and 9.9 kg of alkalis per m³ of mortar. Aggregates in all mortars had an equivalent particle size distribution and were composed of 40% of 0-80 μ m particles, 30% of 315-630 μ m particles and 30% of 1250-3150 μ m particles. For the five reactive mortar mixtures studied, all the 315-630 μ m particles were non reactive and all the 1250-3150 μ m particles

were reactive. Only the nature of 0-80 μ m particles changed: mortars M7, M8, M9, M10 and M11 contained 0%, 10%, 20%, 30% and 40% of 0-80 μ m reactive aggregate and 40%, 30%, 20%, 10% and 0% of 0-80 μ m non reactive aggregate, respectively. Therefore, mortars M7, M8, M9, M10 and M11 contained a total of 30%, 40%, 50%, 60% and 70% of reactive particles, respectively. Moreover, measurements on two reference mortars with only non reactive aggregates were performed (one for each alkali contents). It should be noted that the difference of sizes used in the two parts is due to the fact that the results come from two studies carried out simultaneously.

3 EXPERIMENTAL RESULTS

3.1 Size effect

The experimental results are presented in Figures 1 and 2. The ASR-expansions presented in these figures were obtained by subtracting the expansion of the reference mortar (without reactive aggregate) from the total expansion, as already proposed by some authors [20-22]. The long-term expansion of the reference mortars was 0.03%. Kinetics of the ASR-expansions measured during 500 days are given in Figure 1. After 500 days of exposure in 60°C and 95% R.H., mortars containing small reactive particles (0-80 and 80-160 μ m) show ASR-expansions lower than about 0.01% (Figure 2). ASR-expansions have been measured for all the other mortar using particles larger than 160 μ m: 0.057% for 160-315 μ m reactive particles, 0.315% for 315-630 μ m particles, 0.328% for 630-1250 μ m particles and 0.267% for 1250-2500 μ m particles.

3.2 Effect of fine reactive aggregate

Measurements of the ASR-expansions have been performed for more than 500 days for the two alkali contents (Figures 3 and 4). The ASR-expansions presented were obtained by subtracting the expansion of the reference mortar from the total expansion. The long-term expansions of the reference mortars were 0.02% and 0.03% for the low and the high alkali contents, respectively. For the two alkali contents, the mortars containing only the large reactive particles (M7) show the largest ASR-expansions. Figure 5 shows the last ASR-expansion measured, related to the amount of fine reactive particles in mortar mixtures. ASR-expansion decreased with the amount of fine reactive particles in the mortars: the more the mortar contains fine reactive particles, the smaller the ASR-expansions.

4 MODEL AND DISCUSSION

4.1 Size effect

For this reactive limestone, no ASR expansion was measured for small reactive particles (under $160 \,\mu\text{m}$). ASR-expansion appeared for particles whose diameter is higher than $160 \,\mu\text{m}$ (Figure 2). However, for the same content of reactive particles, the expansion is smaller for the $160-315 \,\mu\text{m}$ reactive particles than for the $315-630 \,\mu\text{m}$ reactive ones. The critical particle size which causes ASR-expansions is around 200 and 300 μm . ASR-expansions increased with the size of reactive particles between 0 and $630 \,\mu\text{m}$. The ASR-expansion is quite the same for the $315-630 \,\mu\text{m}$ particles and for the $630-1250 \,\mu\text{m}$ ones. At last, the larger reactive particles show lower expansions (Figure 2). This pessimum effect has already been observed for other reactive aggregates [9, 12]. During the experiment, the authors observed that cracks were wider for the specimens containing the larger reactive particles. If the cracks are wider, the ASR gel could more easily migrate in them, thus explaining the lower expansions. This pessimum effect could be then a scale effect between the size of the reactive particles and the size of the specimens. More experiments with larger particles of reactive aggregate, over $2500 \,\mu\text{m}$, are necessary to verify this assumption.

The increase of ASR-expansion with the size of the reactive particles can be explained by the effect of the porosity connected to the reactive aggregate. The expansion of the mortar is caused by the expansions of the ASR-gels. It can be assumed that this expansion is caused by the volume variation of the aggregate (in presence of gel). By sake of simplicity, the authors assume in this paper that the mortar expansion is proportional to the aggregate expansion:

$\varepsilon_{mor} = k\varepsilon_{agg}$

If k=1, the mortar expansion is equal to the aggregate expansion. In the reality, k has to be lower than 1, since the mortar expands less than the aggregate due to mechanical effect of cement paste. The decrease of the value of the parameter k corresponds to the increase of the restraint of the cement paste on the aggregate. During ASR, when the aggregate swells, the cement paste is submitted to tensile stresses. Therefore, the effect of restraint is limited and thus the parameter k should not be too small. During the formation of the ASR-gel, a part of the gel can migrate through the porosity close to the aggregate without causing expansions. Therefore, the aggregate expansion can be written for one aggregate 'i':

$$\varepsilon_{agg} = \varepsilon_i = \frac{\left\langle V_{gel_i} - V_{poro_i} \right\rangle^+}{V_{agg_i}}$$

With:

 $-\langle X \rangle^+$: the positive part of X: if X<0, $\langle X \rangle^+ = 0$ else $\langle X \rangle^+ = X$

- $V_{gel,i}$ the volume of ASR gel formed in only one reactive particle, thus:

$$V_{gel_i} = v_{gel} V_{agg_i}$$

With: v_{gel} : the volume of gel per m³ of reactive aggregate

- V_{agg_i} : the volume of one reactive particle: $V_{agg_i} = \frac{4}{3}\partial_i R_i^3$ with R_i : the radius of the reactive

particle.

- $V_{pow_{\perp}}$: the volume of the porosity close to the reactive aggregate in which the ASR gel can migrate without causing expansion. Assuming that the gel can migrate along a same distance of the aggregate l_i whatever the size of the aggregate, the volume of the porosity is:

$$V_{poro_i} = \frac{4}{3}\pi \cdot \left(\left(R_i + l_c \right)^3 - R_i^3 \right) p_{mor} \text{ with } p_{mor} \text{ it porosity of the mortar.}$$

In order to carry out this calculation, three parameters must be determined: $l_c - v_{gd} - k$. In this paper, the authors considered several values for k between 0.1 and 1 (for k = 0.1, the aggregate swells 10 times more than the mortars), and l_c and v_{gd} have been calculated to minimize the deviation between the calculated value e_{mor} and the measured one. The results of the calculations of the parameters are given in Table 2 and the calculated expansions are plotted in Figure 6. Whatever the values of k, all the curves are quite similar and only one curve has been plotted (named 'model1'). For the lower values of k, the values of l_c and v_g are maximal (Table 2). Indeed, the larger is the cement paste restraint, the larger must be the volume of ASR-gel to obtain significant expansion; and if there is a lot of gel, l_c has to be large to prevent expansion for the small reactive particles. Whatever the value of the parameter k (higher than 0.1), the value of the parameter l_c is quite small (less than 10 µm) and can be reached by the ASR-gel. Even if the cement paste largely restraints the aggregate, this calculation can explain the increase of expansion with the size of the reactive particles.

4.2 Effect of fine reactive aggregate

For mortars containing two sizes of reactive particles, the larger was the content of reactive aggregate, the smaller were the ASR-expansions (Figure 5). In fact, the increase of the content of reactive aggregate was due to the increase of the smallest reactive particles ($0-80 \mu m$), while the content of the largest reactive particles ($1250-3150 \mu m$) was the same. The previous part showed that the smallest reactive particles did not cause expansions, however the largest ones largely expanded. As the content of large particles was the same for all the five mortars (for one alkali content), expansions could be similar. However the experimentation showed that the expansions decreased with the increase of the content of the small reactive particles. This effect had already been shown on concrete [23] and can be explained by the consumption of the alkalis by the small reactive particles. When aggregate powders are dispersed in a cement paste they release silica, resulting in a lowering of the Ca/Si ratio in C-S-H. It has been established that the ability of these low Ca/Si C-S-H to fix alkalis is enhanced. The depletion of free alkalis lowers the pH of the pore solution and, consequently, reduces the attack of reactive aggregates. Finally the expansion is reduced or suppressed.

High alkali content implied deeper aggregate attack, and thus a larger volume of gel per m³ of reactive aggregate. However, a lot of papers showed that for low alkali contents (under a threshold of 3 or 4 kg/m³), no expansion occurred. The volume of gel per m³ of reactive aggregate can be considered as proportional to alkali content of the mortar minus by a threshold taken equal to a mean value of $AC_{tb} = 3.5$ kg/m³:

$$v_{gel} = v_{gel_0} \frac{\left\langle AC - AC_{th} \right\rangle^+}{\left\langle AC_0 - AC_{th} \right\rangle^+}$$

with: v_{gd_0} : the volume of gel per m³ of reactive aggregate determined in the previous part, AC: the alkali content of the mortar,

 AC_{tb} : the value of the threshold in alkali (taken equal to 3.5 kg/m³),

 AC_0 : the alkali content of the mortar of the previous part (8.1 kg/m³),

The volume of gel created by the reactive particle of one size depends on the content of the reactive aggregates of this size compared to the content of all the reactive aggregates of the mortar. Indeed, if there are only large particles, all alkalis can react with these particles; but when small particles are added, the alkali content which reacts with the large particles has to decrease. It can be assessed by:

$$v_{gel_i} = v_{gel_0} \frac{\left\langle AC - AC_{th} \right\rangle^+}{\left\langle AC_0 - AC_{th} \right\rangle^+} \frac{CRA_i}{CRA_{tot}}$$

with: CRA_i : the content of the reactive particles of size 'i' (kg/m³) CRA_{bac} : the content of reactive aggregate in the mortar (kg/m³).

For several reactive particles, the aggregate expansion is the sum of the expansions of all the reactive aggregates:

$$\varepsilon_{agg} = \sum_{i} \varepsilon_{i}$$

Calculations were performed for the two alkali contents: Figure 7 gives the measured expansions versus the expansions assessed with the model with the parameters determined for k=1 in the previous part (black points). As shown in Figure 7, the model gives values not too far from the measured expansions. The mean deviation between calculated and measured expansions is about 30%. It can be partly explained by the deviation between the model and the measurement for the largest particle for the parameter determined in the previous part. The value calculated for the largest particle was about 30% higher than the measurement (Figure 6). The aggregate can be better represented by the parameters of the curve 'model 2' in Figure 6 (k=1, l_c = 0.65 µm and v_g= $3.03.10^{-3}$ m³). Figure 7 shows that the assessment is better with these parameters (gray points in Figure 7) with a mean deviation of about 15%.

The model uses only proportionality between the parameters of volume of gel, alkali content and content of reactive aggregate. It appears to be efficient to assess ASR-expansions in this case for which mix designs and environmental conditions are perfectly known.

5 CONCLUSION

This paper presented the experimental measurements performed on 16 different mix-designs containing reactive siliceous limestone with a special attention to the proportions of alkalis (Na₂O_{eq}) and reactive silica in the mixtures. Small reactive particles (under about 160 μ m) do not cause expansions while coarse particles (0.63-1.25 mm) show the largest expansions (0.32%). The model presented shows that it can be explained by the migration of ASR-gel in the porosity very close the reactive aggregate (less than 10 μ m). The experimental results on mortars containing two sizes of aggregates with increasing silica content show that ASR-expansions decreased with the amount of small particles. The volume of gel per m³ of reactive aggregate used in the calculations have been considered as proportional to alkali content of the mortar minus by a threshold in alkali content and to the content of the reactive aggregates of each size compared to the content of all the reactive aggregates of the mortar. With these assumptions, the model predicts the expansions of the mortars containing two sizes of reactive aggregates. This approach will be used as a basis for the development of future models to assess the potential expansion of concrete containing alkali-reactive aggregates.

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TABLE 1: Chemical composition of cement and aggregates (% by mass).

	Fraction*	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	Na ₂ O _{eq}	SO3	LOI
Cement	-	20.1	5.6	2.0	62.5	3.1	0.2	0.9	0.8	3.2	1.7
Non-reactive marble	-	-	-	-	54.4	0.49	0.001	-	-	0.01	43.0
Reactive siliceous limestone	1250-3150 μm	20.0	1.3	0.6	40.6	1.2	0.4	0.4	0.7	0.3	34.7

TABLE 2: Model parameters.

k	0.1	0.25	0.5	0.75	1.
l _c (μm)	6.1	2.5	1.3	0.85	0.65
v_{gel} (m ³ /m ³)	37.10-3	14.8.10-3	7.4.10-3	4.95.10-3	3.74.10-3



Figure 1: ASR-expansions of mortars containing 30% of reactive particles of size 0-80, 80-160, 160-315, 315-630, 630-1250, 1250-2500 μ m and 70% of continuous 0-2500 μ m non reactive sand.



Figure 2: ASR-expansions of mortars containing 30% of reactive particles of size 0-80, 80-160, 160-315, 315-630, 630-1250, 1250-2500 μ m and 70% of continuous 0-2500 μ m non reactive sand after 500 days at 60°C and 95% RH.



Figure 3: ASR-expansions of mortars containing variable reactive aggregates content (mix of reactive particles of two sizes: $0-80 \mu m$ and $1250-3150 \mu m$) for 6.2 kg of alkalis per kg of mortar.



Figure 4: ASR-expansions of mortars containing variable reactive aggregates content (mix of reactive particles of two sizes: 0-80 µm and 1250-3150 µm) for 9.9 kg of alkalis per kg of mortar.



Figure 5: Last ASR-expansions measured versus the percent of 0-80 µm reactive particles.



Figure 6: ASR-expansion versus mean reactive particle size: comparison between model and experiment.



Figure 7: Comparison between model and experiment for mortars containing two sizes of reactive particles for two alkali contents.