

# Use of Reactive Aggregate Powders to Mitigate ASR Expansion

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## Abstract

The purpose of this research was to test a method for reducing ASR expansion that could be more economical and compliant than the use of supplementary cementing materials. This method consists in using the reactive aggregate itself as mineral admixture in order to decrease the deleterious expansion due to ASR. The results show that, in most cases, reactive aggregate powders (RAP) reduce the expansion of concrete. It is assumed that RAP react with Portlandite to produce low Ca/Si C-S-H which can adsorb a significant quantity of alkalis and thus lead to a decrease of the alkalinity of the pore solution, to a level sufficiently low to limit the attack of coarser reactive aggregates. This hypothesis is supported by a quantitative evaluation of the alkalinity of the pore solution of concrete containing RAP, performed using a modified version of the hydration model CEMHYD3D.

## 1. Introduction

In the decade following the discovery of the alkali silica reaction (ASR) by Stanton, recommendations, such as the use of non-reactive aggregate or the use of low alkali cement, were made to avoid or to mitigate the disorders due to this reaction. At the same time, it was established that blended cements made of OPC mixed with pozzolanic admixtures were efficient in reducing ASR expansion. The use of such admixtures (fly ash, silica fume, ground granulated slag) is now current practice to prevent disorders due to ASR.

An analysis of the literature concerning other admixtures obtained by grinding rocks shows that the papers can be classified in two categories:

- (i) Papers related to ground admixtures added to mixtures containing a reactive aggregate coming from a different source. Many references concern this category, particularly Hanna [1], Stanton [2], Bennett and Vivian [3], Pepper and Mather [4], Abdun-Nur [5], Ming-Shu et al. [6,7], Asgeirsson [8] and Kodama and Nishino [9].
- (ii) Papers devoted to fine admixtures associated with the reactive aggregate, from which they were obtained by grinding. Only a few references are found in this category. For example, Bian et al. [10] worked on mortars made of andesite, and they replaced between 30 and 70% of cement by this ground aggregate (fineness ranging between 300 and 1100 m<sup>2</sup>/kg). They showed that ground andesite

reduced the expansion of mortars. However, high replacement rates and high finenesses were needed for powders to be efficient, thus limiting the interest of the solution in practice. Guédon-Dubied et al. [11] found that, when filler from reactive siliceous limestone was added to mixtures containing the same reactive aggregate, expansions were reduced to about the half of those occurring in concrete with no filler. The aim of this paper is to verify whether reactive aggregate powder (RAP) is an efficient and general solution to counteract ASR expansion due to the coarse aggregate. So tests were performed on concrete using four reactive aggregates with their ground fillers.

## 2. Materials

The binder was an ordinary Portland cement CEM I 52.5R, as specified by European Standard EN 197-1, with a specific weight of 3.15 and a fineness of 350 m<sup>2</sup>/kg (Blaine). Its chemical composition (Table 1) shows a moderately high content of Na<sub>2</sub>O<sub>eq</sub> (0.8%).

Four coarse aggregates were chosen:

- A siliceous aggregate B (quarry located at Brix, in Brittany, France) which is a metaquartzite containing very small quantities of muscovite and amorphous silica in the intergranular cement. This rock, chosen in France as a standard reactive aggregate, presents low kinetics of silica dissolution.
- A siliceous limestone T (quarry located in Tournaisis, North of France), mainly composed of calcite with a few percent of dolomite. When observed at microscopic level, diagenetic silica is seen to consist mainly of quartz and traces of feldspars, muscovite and kaolinite. This aggregate is also a French standard reactive rock, with a slow rate of silica dissolution.
- An opaline aggregate O, which is very deleterious and commonly reported in laboratory experiments as a reference mineral. The aggregate used in this study was an “opal CT”, according to the classification of Jones [12]. The glassy phase was predominant, the crystallised phase being composed essentially of tridymite and a few percent of quartz.
- A crushed waste glass G, resulting from the crushing of window glasses. It was chosen because the reuse of waste glasses has been envisaged by several workers in the last decade [13-17]. It presented an amorphous structure, as determined by X-ray diffraction.

Reactive aggregate powders b, t, o and g were obtained by the crushing and sieving at 80 µm of aggregates B, T, O and G, respectively. The chemical compositions of these RAP appear in Table 1. In order to study the influence of fineness in counteracting expansion, the powders were ground to a specific surface area (Blaine) ranging from 400 to 650 m<sup>2</sup>/kg, depending on the aggregate.

Table 1 - Chemical compositions of cement, aggregates and RAP.

	Cement	B	b	T	t	O	o	G	g
SiO <sub>2</sub>	20.1	94.1	87.7	20.0	15.4	93.0	92.7	72.1	66.7
Al <sub>2</sub> O <sub>3</sub>	5.6	1.6	4.0	1.3	1.7	0.0	0.0	1.0	0.7
Fe <sub>2</sub> O <sub>3</sub>	2.0	0.3	1.0	0.6	1.1	0.2	0.3	0.1	0.1
CaO	62.5	0.2	0.4	40.6	43.6	0.2	0.2	6.3	7.2
MgO	3.1	0.1	0.2	1.2	1.5	0.1	0.1	3.7	4.1
K <sub>2</sub> O	0.9	0.7	0.9	0.4	0.5	0.1	0.1	0.1	0.2
Na <sub>2</sub> O	0.2	0.2	0.1	0.4	0.5	0.2	0.2	12.7	13.4
SO <sub>3</sub>	3.2	0.1	0.1	0.3	0.2	0.9	1.1	0.1	0.2
L.O.I	1.7	0.7	1.1	34.7	34.9	5.5	6.0	0.4	0.7

### 3. Experimental methods

The concrete samples were made, stored and tested according to French Standard P 18 454 [18]. This test consisted of evaluating the performance of a concrete with respect to AAR by measuring the expansion of 7x7x28 cm prisms stored at 60°C and 100% R.H. After mixing, the samples were stored in a moist room at 20°C for 24 hours. Then, the concrete prisms were placed vertically on grids in watertight containers containing 35 mm of water (the concrete was not in contact with the water). Each concrete was tested on three prisms. The containers were placed in a reactor generating an atmosphere at 60°C and 100% R.H.. Expansion measurements were taken after the containers and their prisms had been cooled for 24 hours at 20°C. Immediately after each measurement, the prisms were put back into their containers, which were kept in the reactor at 60°C until the next measurement. The last measurement was taken at 83 weeks.

Details of the concrete mixtures, including the slump (modified Abram's cone of 200mm) and density, are given in Table 2. Each mixture is designated by a capital letter, representing the reactive aggregate used, followed by a small letter for mixtures containing RAP. The water content of the concrete was set between 190 and 210 l/m<sup>3</sup>. A significant amount of cement was used (between 429 and 467 kg/m<sup>3</sup>) in order to obtain high alkali content, and it was decided to enhance alkali content to 5 kg/m<sup>3</sup> by adding NaOH to the water.

For concretes B and T, the coarse aggregate (4-16mm) was composed of reactive aggregate and the sand (0-4mm) was non-reactive quartz. For concrete O, the amount of opal was limited to 36% of the coarse aggregate (8-16mm), in order to take into account its pessimum content; the rest of the aggregate was non-reactive quartz sand (0-8mm). In concrete G, the reactive aggregate resulted from the crushing of waste window glass, so its maximum size was limited by the thickness of the window glass, i.e. 4 mm. Hence, the coarse aggregate > 4mm was non-reactive quartz.

RAP used as replacement of 20% of the sand (by weight) had finenesses of 400, 600, 650 and 400 m<sup>2</sup>/kg for powders b, t, o and g, respectively.

Table 2 - Composition, density and slump of concrete mixtures.

	B	T	O	G	B-b	T-t	O-o	G-g
Cement (kg/m <sup>3</sup> )	477	477	469	452	462	467	438	442
Water (l)	212	212	208	201	205	208	195	197
W/C	0.44	0.44	0.44	0.44	0.44	0.44	0.44	0.44
Superplasticizer (l)	0	0	0	0	2.46	2.49	4.87	4.91
RAP (kg/m <sup>3</sup> )	0	0	0	0	122.4	113.5	119.4	136.8
Sand 0.08-4 (kg/m <sup>3</sup> )	631	617	639	699	490	454	478	546
Agg 4-8 (kg/m <sup>3</sup> )	(NR)	(NR)	(NR)	(NR)	(NR)	(NR)	(NR)	(R)
Agg 8-16 NR (kg/m <sup>3</sup> )	235	248	138	200	228	242	129	195
Agg 8-16 R (kg/m <sup>3</sup> )	(R)	(R)	(NR)	(NR)	(R)	(R)	(NR)	(NR)
Density (kg/m <sup>3</sup> )	888	929	345	0	861	910	323	0
Slump (cm)	2443	2445	2403	2319	2371	2397	2252	2272
	4	4	4	4	3	7	11	5

(R) = reactive aggregate; (NR) = non-reactive aggregate

#### 4. Experimental results

The length variations of the concrete prisms cured 83 weeks at 60°C and 100% R.H. are shown in Figure 1. In a first analysis, it is noteworthy that all the aggregate powders reduced the expansion of concrete. Freshly broken surfaces were observed by scanning electron microscopy (SEM) coupled with energy dispersive spectrometry (EDS). All concretes without RAP reacted and produced large amounts of ASR gel. These gels had various morphologies, from smooth cracked surfaces to lamellar and acicular crystals. Concretes containing RAP did not present any ASR gel on the broken surfaces and only very small amounts of gel were detected in some voids.

##### Concretes B and B-b

The reduction in the final expansion due to RAP b reached a significant value of 89%, due to a delayed start of the reaction and a lowering of the expansion speed (Table 3). The concrete B-b meets the recommendations of the standard NF P18-454 [18], its expansion at the age of 3 months being lower than the limit of 0.02%.

##### Concretes T and T-t

The activity of the RAP t was moderate, resulting in an expansion reduction of 36%. Expansion at the age of 3 months remained far above the limit of 0.02%. Thus the use of concretes T and T-t should be prohibited. In a work carried out to determine the alkali-reactivity of another Tournai limestone, Guédon-Dubied et al. [11] showed that

incorporation of reactive limestone filler (15 at 30% by weight) reduced expansion at 1 year. The observed reduction of 50% was greater than in the present work, but the expansion of their concrete without filler was 0.065% at 1 year, a much lower value than in our case (0.17%).

### Concretes O and O-o

RAP o was very efficient to reduce expansion (96%) Practically the concrete O-o meets recommendations of standard NF P18-454 [18], its expansion being only the half the authorized level.

### Concretes G and G-g

The analysis of the experimental results was not easy at all, because there were three possible points of view.

- (i) On a phenomenological level, the expansion of concretes G was very high (1.07%). The addition of aggregate powder was very interesting since it led to a great reduction of expansion (90%).
- (ii) A strict examination of the performance of concrete G-g in relation to standard NF P18-454 [18] led to approval of this mixture since its expansion was lower than 0 at 5 months.
- (iii) From a practical point of view, it would be negligent to implement such a concrete. The reason lies in the fact that, even though expansion started after a very long delay (1 year) due to RAP, it still reached a high level: the expansion at 83 weeks was fivefold the limit suggested by the standard at 5 months.

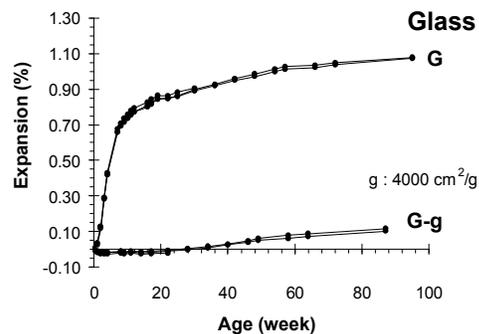
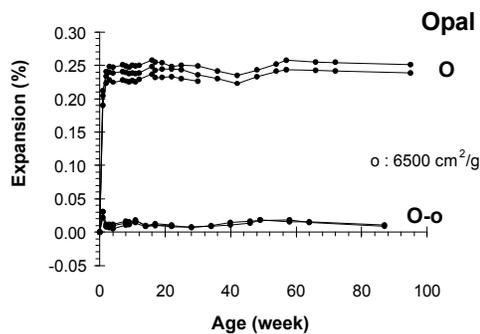
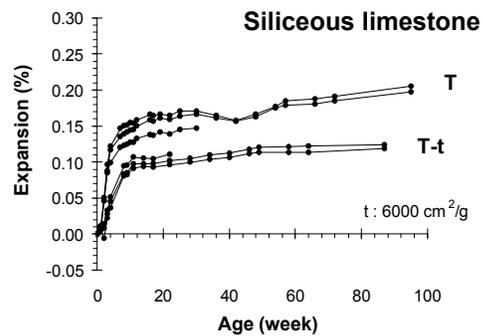
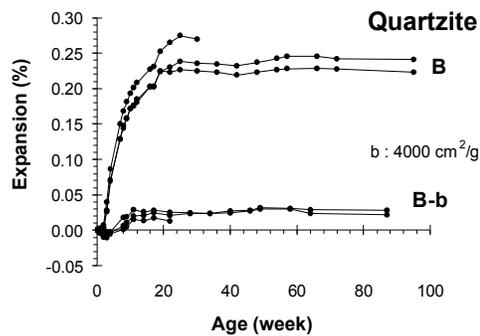


Figure 1 – Expansion of concretes versus time (60°C and 100% RH).

Table 3 – Influence of aggregate powders on the properties of concrete (relative to ASR).

			B	T	O	G
Expansion (83 weeks)	Without powder	Final expansion (%)	0.234	0.200	0.245	1.07
		Max expansion rate ( $\mu\text{m}/\text{m}/\text{week}$ )	212	395	2070	1025
	With powder	Reduction of expansion (%)	89	36	96	90
		Reduction of expansion rate (%)	75	63	98	97
		Acceptance criteria P18-454*	yes	no	yes	yes
	Microstructure	Without powder	ASR products visible by SEM	yes	yes	yes
With powder		ASR products visible by SEM	disappeared	disappeared	disappeared	disappeared

\* expansion at 3 months lower than 0.02%

### 5. Hypothesis concerning the mechanism by which aggregate powders counteract the deleterious effects of reactive aggregates.

The activity of reactive aggregate powders (RAP) as ASR-reducers may be explained using the same mechanisms as proposed for pozzolans. When RAP are dispersed in a cement paste, they release silica, resulting in a lowering of the Ca/Si ratio in C-S-H. Many authors [19-21] have 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.

This hypothesis was evaluated in two steps. Firstly, experiments were carried out in order to confirm that reactive aggregate powders acted as pozzolans. The results [22] showed that:

- all RAP consumed CH, proportionally to their reactive silica content;
- the use of these fines in pastes made with  $\text{C}_3\text{S}$  led to a decrease of the Ca/Si ratio in C-S-H;
- higher amounts of alkalis were fixed by C-S-H in cement pastes containing RAP.

The second step consisted of a numerical simulation to quantify the alkalinity of the pore solution of concrete containing reactive aggregate powders.

#### 5.1 Models

The simulation was based on two models. The first one was a model for the hydration of cement, which simulates the dissolution, diffusion and

precipitation phases of ionic species in cement paste. The program CEMHYD3D was chosen among the different models available in the literature. CEMHYD3D is proposed by the NIST (National Institute of Standards and Technology, USA) and was developed by Bentz [23-26]. It has been used for more than 10 years by many researchers of the scientific community and the hydration module seems to give pertinent results according to the numerous papers published in recent years [27-31]. Its principle of operation can be found elsewhere [23-26]. One item of information given by the model is the volume of C-S-H having a Ca/Si ratio of 1.8 and 1.1, produced by the hydration of calcium silicates and pozzolans, respectively. Abundant literature highlights the different consumptions of alkalis depending on the types of C-S-H produced. So these data could then be used for the calculation of the alkalinity of the pore solution.

A second model had to be used to evaluate the consumption of alkalis, depending on the nature of C-S-H produced by the hydration of cement and RAP particles present in the elementary volume.

The calculation of the alkalinity of the pore solution was based on the approach of Hong and Glasser [21]. They defined an experimental coefficient of distribution of alkalis,  $K_d$ , which characterizes the distribution of alkalis between the two types of C-S-H and the pore solution (Equation 1). A value of  $K_d = 1$  means that the quantities of alkalis in C-S-H and the pore solution are the same. For our tests, the values proposed by Hong and Glasser [21] were 0.4 for ordinary C-S-H (Ca/Si  $\approx$  1.7) and 3 for "pozzolanic C-S-H" (Ca/Si  $\approx$  1.1). It was assumed that  $K^+$  and  $Na^+$  had the same value of  $K_d$ .

$$K_d = \frac{\text{alkalis in C-S-H (mM / g)}}{\text{alkali concentration in pore solution (mM / mL)}} \quad (\text{Equation 1})$$

According to this approach, it can be supposed that alkalis are mostly present in three phases: anhydrous phase, C-S-H and pore solution. The application of the mass conservation law leads to equation 2: the quantity of alkalis dissolved at time t ( $Alk_{dissolved}$  in mol) is equal to the sum of alkalis in the pore solution ( $Alk_{solution}$ ), in the C-S-H ( $Alk_{CSH}$ ) and in the pozzolanic C-S-H ( $Alk_{PCSH}$ ).

$$Alk_{dissolved} = Alk_{solution} + Alk_{CSH} + Alk_{PCSH} \quad (\text{Equation 2})$$

The distribution of alkalis between the pore solution and the two types of C-S-H depends on the volume of each constituent (Equation 3), and it is evaluated from coefficients  $K_d$  (Equations 4 and 5).

$$Alk_{dissolved} = [Alk_{solution}] \cdot V_{solution} + [Alk_{CSH}] \cdot V_{CSH} + [Alk_{PCSH}] \cdot V_{PCSH} \quad (\text{Equation 3})$$

where  $[Alk_{solution}]$ ,  $[Alk_{CSH}]$  and  $[Alk_{PCSH}]$  are the alkali concentrations (mol/L) in the pore solution, CSH and pozzolanic CSH present in the hydrated paste according to volumes (L)  $V_{solution}$ ,  $V_{CSH}$  et  $V_{PCSH}$ .

$$K_d^{CSH} = \frac{[Alk_{CSH}]/d_{CSH}}{[Alk_{solution}]} \quad (\text{Equation 4})$$

$$K_d^{PCSH} = \frac{[Alk_{PCSH}]/d_{PCSH}}{[Alk_{solution}]} \quad (\text{Equation 5})$$

where  $d_{CSH}$  and  $d_{PCSH}$  are the densities (g/cm<sup>3</sup>) of the two types of C-S-H.

By combining equations 3, 4 and 5, the alkali concentration (mol/L) of the pore solution is obtained (Equation 6).

$$[Alk_{solution}] = \frac{Alk_{dissolved}}{V_{solution} + K_d^{CSH} \cdot d_{CSH} \cdot V_{CSH} + K_d^{PCSH} \cdot d_{PCSH} \cdot V_{PCSH}} \quad (\text{Equation 6})$$

$K_d^{CSH}$  and  $K_d^{PCSH}$  were found in the literature [21] and the densities  $d_{CSH}$  and  $d_{PCSH}$  were taken to be 2.12 and 1.69 g/cm<sup>3</sup> [32]. Finally, parameters  $Alk_{dissolved}$ ,  $V_{solution}$ ,  $V_{CSH}$  and  $V_{PCSH}$  were calculated using CEMHYD3D at each step of the hydration reaction. This kind of approach has already been proposed [33].

## 5.2 Results

Numerical simulations were carried out on the cement pastes of our concretes by neglecting all particles larger than 80 μm. So, only cement, fines particles and water were used in the model. The simulations of hydration were made for RAP having reactive fractions of 10% (fine t), 25% (fine b), 50%, 75% and 100% (fine o). In the case of fine g (glass), the high alkali content was taken into account for the calculation of the alkalinity of the pore solution.

Figure 2 gives the alkalinity of the pore solution (mol/L) after 28 days of hydration as a function of the reactive fraction of RAP. It can be seen that about 65% of reactive silica is necessary to stay under a threshold of 0.5 mol/L. This threshold is a mean value of those found in the literature and for which only small expansions are observed on mortars and concretes: 0.4 to 0.5M [34], 0.65M [35], 0.25M [36], 0.3M [37], 0.45 to 0.67M [38].

Concrete containing RAP of opal presented the lowest alkalinity. This concrete also gave the lowest expansion (Figure 1). The use of fines from siliceous limestone only led to a small reduction of the alkalinity, and so a small reduction of the expansion.

Figure 3 gives the relation between reductions of the expansions of the concretes (Table 3) and the calculated alkalinity of the pore solution. It can be seen that the reductions of expansion and alkalinity are in the same sense. The alkali concentration alone does not explain the expansions of the concretes, since other parameters are to be considered (e.g. the

amount of reactive silica in the aggregates). However, a good correlation between relative expansions and alkalinity of pore solution is obtained. A threshold value of about 0.7 mol/L is observed, below which the expansions are reduced by more than 80%.

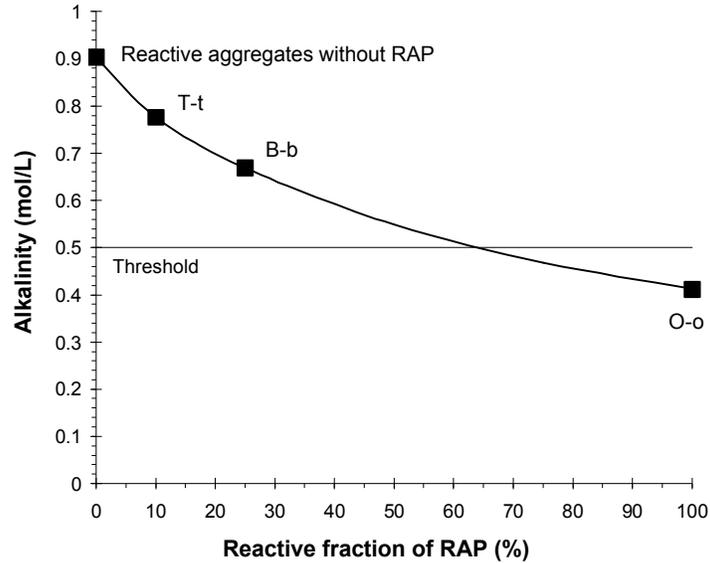


Figure 2 – Numerical simulations of the alkalinity of the pore solution of concretes with RAP, cured 28 days at 60°C.

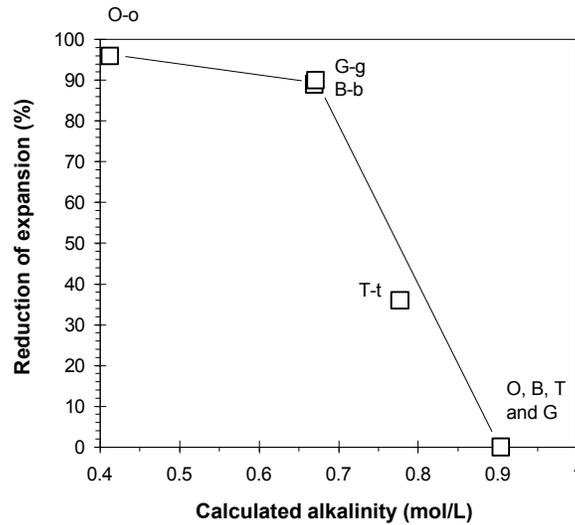


Figure 3 – Relation between the reduction of expansion measured on concretes and the alkalinity at 28 days calculated from numerical simulations.

## 6. Conclusion

The experiments reported in this paper were intended to verify the following hypothesis: the harmful effects of ASR-expansions induced by an alkali-reactive aggregate can be counteracted by the beneficial activity resulting from the addition of a fraction of this aggregate ground into a fine state (RAP).

The work was carried out on concretes incorporating four alkali-reactive aggregates, with or without RAP added. The concretes were cured at 60°C and 100% RH and their expansions were measured up to 83 weeks. In these conditions, which are different from actual conditions in the field, the following conclusions can be drawn:

- (i) The addition of RAP to concrete containing the same coarse reactive aggregate can reduce or suppress ASR expansion.
- (ii) The efficiency of such a RAP depends on its petrographic composition, content and fineness.
- (iii) Another consequence of the addition of RAP is the invisibility of ASR products on the broken surfaces of concretes examined by scanning electron microscopy. However, it cannot be asserted that the production of gel is effectively prevented; perhaps it forms in particles too small and diffuse to be detected at this level of magnification.
- (iv) It is assumed that RAP react with Portlandite to produce low Ca/Si C-S-H which can adsorb a significant quantity of alkalis and thus lead to a decrease of the alkalinity of the pore solution, to a level sufficiently low to limit the attack of coarser reactive aggregates. This hypothesis is supported by a quantitative evaluation of the alkalinity of the pore solution of concrete containing RAP, performed using a modified version of the hydration model CEMHYD3D.

Before any attempt to extrapolate from laboratory to field concrete, our findings have to be verified on other aggregates and more attention must be paid to the influence of the amount of RAP on their activity. In the event of successful verifications, the method of counteracting the ASR expansion due to a reactive aggregate by addition of a finely ground fraction of this aggregate would be attractive from a practical point of view, since this addition could be done at the quarry or at the concreting site.

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