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# The carbonation of recycled concrete aggregate affected by alkali-silica reaction

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## Abstract

*Recycled concrete aggregate (RCA) produced by crushing concrete from structures affected by alkali-silica reaction (ASR) can induce expansion and damage in new concrete produced using the RCA even when preventive measures are implemented to control ASR. The damage can be prevented by carbonating the RCA prior to its use in new concrete. Laboratory data are presented from a study using RCA produced from crushing mortar containing a highly-reactive (Jobe) sand and a high-alkali cement. The mortar was crushed to produce coarse aggregate in the size range from 5 to 20 mm. A portion of the RCA was carbonated prior to use. Concrete samples were then produced with the carbonated RCA (RCA-CO2) and non-carbonated RCA (RCA-ASR). The binder used in the concrete was a blend of 80% low-alkali cement plus 20% fly ash. This blended cement has been shown to be effective in preventing ASR expansion when used with the Jobe aggregate. Concrete prisms containing RCA-ASR expanded significantly (0.162% in 2 years) when stored over water in sealed containers at 38°C whereas concrete produced with RCA-CO2 did not (< 0.040% at 2 years). The mechanism is discussed. In the case of the uncarbonated RCA there is a sufficient concentration of alkali hydroxides within the mortar portion of the RCA to fuel ASR with the reactive silica in the sand particles. However, in the carbonated RCA the concentration of alkali hydroxides is significantly reduced by the carbonation process and this prevents ASR expansion despite the abundance of reactive silica present in the RCA.*

*Keywords*: carbonation; preventive measures; recycled concrete aggregates

## 1.  INTRODUCTION

The consideration of concrete as a sustainable building material is related to its relatively low embodied emissions per unit mass [1] and low operating emissions associated with the use stage of a building in service [2]. The sustainability of concrete is further demonstrated by considering it to be a recyclable construction material; returned or reclaimed concrete can be crushed for use as aggregates in new construction [3]. The practice not only reduces waste disposal but helps to conserve natural resources and can realize economic benefits [4].

Recycled concrete processing is often aligned with the demolition of a structure at the end of its service life. In many cases, end of service life is associated with a reduced performance particularly with respect to durability. Insofar as adhered paste is a part of a recycled concrete aggregate (RCA) it can contain ionic species that reflect the nature and exposure history of the source concrete. Such species, like alkalis and chlorides, can have a chemical impact in the new concrete that is unlike that of natural aggregates that might otherwise be used [5].

One common deleterious durability concern in concrete, particularly one that can be relevant to the end of its service life, is alkali-silica reaction (ASR). Siliceous aggregates in the concrete undergo an expansive reaction with alkali hydroxides from the binder phase. Recycled concrete aggregates (RCA) produced from ASR-affected concrete can contain both reactive silica and additional alkalis either in the adhered mortar or from ASR gel already present in the RCA [6]. The problem of alkali-silica reactions in recycled concrete is a challenge to manage since a shift in the gradation of reactive particles and concentration of alkalis (possibly through crushing of the concrete and/or dilution by changing the concrete to a component of a new concrete mix) can become closer to the pessimum content (a proportion that is an ideal balance between reactive silica and available alkali that will produce the highest expansion) in the recycled aggregate concrete (RCA) than in the original concrete.

The study of RCA produced from recycled concrete that had been affected by ASR has shown that it can be as deleteriously reactive as the siliceous limestone aggregate originally contained within the

concrete [7]. As with reactive aggregate, the expansion could be mitigated by replacing part of the cement in the binder with SCMs albeit at greater levels than required for the original virgin reactive aggregate. Fine RCA was observed to be less reactive than coarse RCA likely due to a reduced proportion of reactive constituents in the small size fraction. Alkali silica reaction in concrete has also been observed to be induced by mortar adhered to recycled aggregate [8] and the amount of adhered mortar can affect the level of reactivity in an aggregate [9]. In limiting the impacts it has been observed that reducing the available alkalis appears to have a greater impact than reducing the calcium availability [10]. There is a clear and obvious challenge to recycling ASR-affected or ASR-susceptible concrete for use as aggregates.

It has been established that deliberately pre-carbonating RCAs can improve both the quality of the aggregates and the mechanical properties of concrete produced with such aggregates [11–14]. It is theorized that CaO from the binder combines with carbon dioxide to form $CaCO_3$ that precipitates in the pore space of the mortar component present in RCAs and improves the microstructure [15]. The $CO_2$ treatment of recycled concrete aggregates has been observed to reduce the water absorption of the aggregates and reduce the transport properties of concrete (as measured by bulk electrical conductivity, chloride ion permeability and gas permeability) made with the aggregates as compared to untreated RCA [15].

The $CO_2$ treatment of a recycled concrete aggregate and its impact on chemical fluxes into and out of the treated mass may has some similarity to the $CO_2$ treatment of cement solidified wastes [16]. It has been observed that $CO_2$ solidification of paste cylinders comprised of dried wastes (mainly heavy metal hydroxides) and cement greatly reduced the leachability of metals contained within the forms. It has been suggested that some metals can be preferentially incorporated in the silica-rich rims of decalcified cement grains, as in the calcite infilling porosity [17].

The present work considers the impacts of carbonating a recycled concrete aggregate that is produced from mortar comprised of a reactive sand and a high-alkali cement. The alkali–silica reaction (ASR) in concrete occurs by reaction of reactive silica phases in the aggregate with alkali and hydroxide ions in the pore solution of the hydrating cement to produce a hydrous alkali silicate gel [18,19]. The reaction depends not only on sufficient chemical driving forces (alkali concentration and the aggregate's reactivity) but also on the transport of alkali ions. If mobility of the alkali ions in the RCA adhered paste can be reduced, then expansive gel formation may be inhibited.

## 2. MATERIALS AND METHODS

### 2.1 Materials

A high-alkali (HA) and low-alkali (LA) Portland cement, and a single source of low-calcium fly ash (FA) were used in the study; the chemical composition of the cementing materials is given in Table 2.1.

Table 2.1: Composition of Cementing Materials

|        | $SiO_2$ | $Al_2O_3$ | $Fe_2O_3$ | CaO  | MgO  | $Na_2O$ | $K_2O$ | $SO_3$ | $Na_2Oe$ |
|--------|---------|-----------|-----------|------|------|---------|--------|--------|----------|
| HA PC  | 19.6    | 5.33      | 2.12      | 61.8 | 2.36 | 0.30    | 1.24   | 4.17   | 1.12     |
| LA PC  | 21.6    | 4.81      | 2.10      | 62.1 | 2.43 | 0.22    | 0.36   | 3.41   | 0.46     |
| FA     | 48.5    | 19.0      | 13.6      | 5.24 | 2.26 | 2.52    | 1.87   | 2.45   | 3.75     |

A single source of highly-reactive mixed quartz/chert/feldspar sand (JB) was used in the study. Concrete mixtures incorporated either a non-reactive siliceous gravel (NC) or a non-reactive natural river sand (NF). A solution of 30% lithium nitrate (LN) was used.

### 2.2 Production and Treatment of Recycled Concrete Aggregate (RCA)

Prisms (75 x 75 x 300 mm) were cast using a mortar produced with high-alkali Portland cement (HA) and Jobe sand (JB) using sand:cement:water = 3:1:0.5. The prisms were sealed in plastic bags and stored at 23°C for 3 months. The mortar prisms were then fractured into chunks using a hammer and the chunks passed through a jaw crusher to reduce the particle size to pass a 20-mm sieve. The sub-20 mm material was screened on a 5-mm sieve to remove the fine fraction. The 20-5 mm material was used for all testing and was identified as RJC.

The 20-5 mm material was air-dried in the laboratory prior to one of the following treatments:

- RJC: untreated recycled Jobe-concrete aggregate
- RJC-LN: RJC immersed in 30%-LiNO$_3$ solution for 28 days
- RJC-CO2: RJC aggregate stored at 55 to 65% RH in a CO$_2$-enriched atmosphere (1% CO$_2$) for 91 days. A schematic of the carbonation chamber is shown in Figure 2.1. During carbonation, larger particles of RCA were periodically removed from the chamber, crushed with a hammer and sprayed with phenolphthalein solution to determine whether uncarbonated material remained. Beyond 56 days of carbonation the phenolphthalein remained colourless for all particles tested. The absence of Ca(OH)$_2$ was also confirmed after 91 days by testing crushed particles using thermogravimetric analysis (TGA).
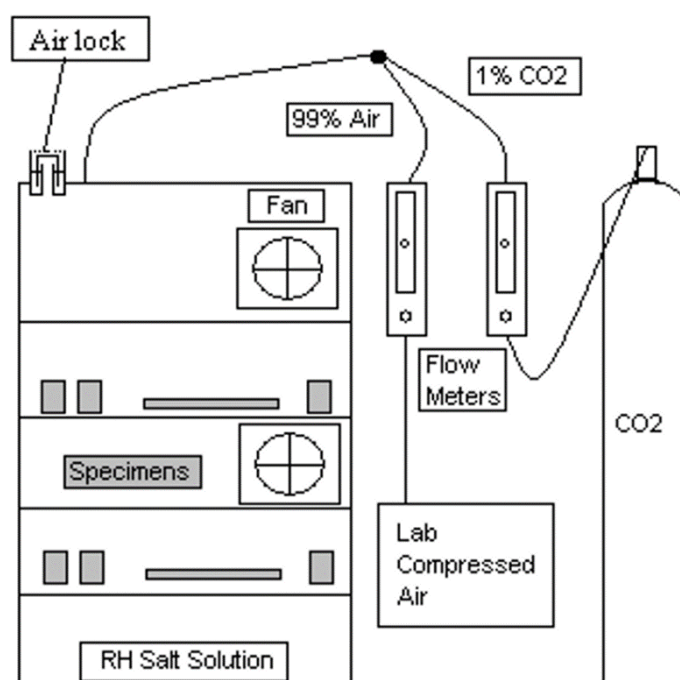


Figure 2.1: Schematic of Carbonation Chamber

## 2.3 Testing of RCA for Alkali-Silica Reaction (ASR)

The Jobe sand (JB), the untreated RCA (RJC) and the treated RCA (RJC-LN and RJC-CO2) were tested using the concrete-prism test (ASTM C1293). Table 2.2 presents the concrete mixtures that were tested in this study. It was originally intended to also test the lithium-treated RCA in a mix with high-alkali cement but there was insufficient RCA available to permit this.

Table 2.2: Composition of Concrete Prism Tests

| Mix ID | Cementing Material | Reactive Aggregate | Non-Reactive Aggregate |
|---|---|---|---|
| HA-JB | HA Cement | JB Sand | Non-Reactive Coarse |
| LAFA-JB | LA Cement + Fly Ash | | |
| HA-RJC | HA Cement | RJC untreated | Non-Reactive Fine |
| LAFA-RJC | LA Cement + Fly Ash | | |
| HA-RJC-CO2 | HA Cement | RJC Carbonated | |
| LAFA-RJC-CO2 | LA Cement + Fly Ash | | |
| LAFA-RJC-LN | LA Cement + Fly Ash | RJC Lithium-Treated | |

Briefly, the concrete-prism test involves producing concrete prisms (75 x 75 mm x 250-mm gauge length) that are fitted with stainless-steel inserts at the ends to allow length-change measurements to be made. The prisms are stored over water in sealed containers stored at 38°C and are periodically removed to determine changes in length and mass. The concrete mix design incorporates 420 kg/m$^3$ of cementing material and w/cm in the range of 0.42 to 0.45. The cementing material was comprised of either 100% high-alkali cement, designated HA, or a combination of 80% low-alkali cement plus 20% fly ash, designated LAFA. Note that none of the concrete mixtures used in this study were boosted with NaOH during mixing. The alkali loading (from the Portland cement) was 4.70 and 1.55 kg/m$^3$ Na$_2$Oe, respectively, in mixes produced with HA Cement and LA Cement + Fly Ash. All concrete prism expansion tests were monitored for 2 years after which time petrographic examination was performed on selected concrete mixes.

## 3. RESULTS

### 3.1 Concrete with Jobe Sand

Figure 3.1 shows the expansion of the concrete prisms containing alkali-silica reactive Jobe sand and non-reactive coarse aggregate. As expected the concrete containing high-alkali (HA) cement expanded very rapidly showing deleterious levels of expansion (> 0.04%) and cracking after just 3 months; the total expansion at 2 years was 0.550%. The concrete produced with Jobe sand in combination with low-alkali (LA) cement and fly ash (FA) did not exhibit deleterious expansion (> 0.04%) or any visible cracking throughout the two-year period (0.039% at 2 years).
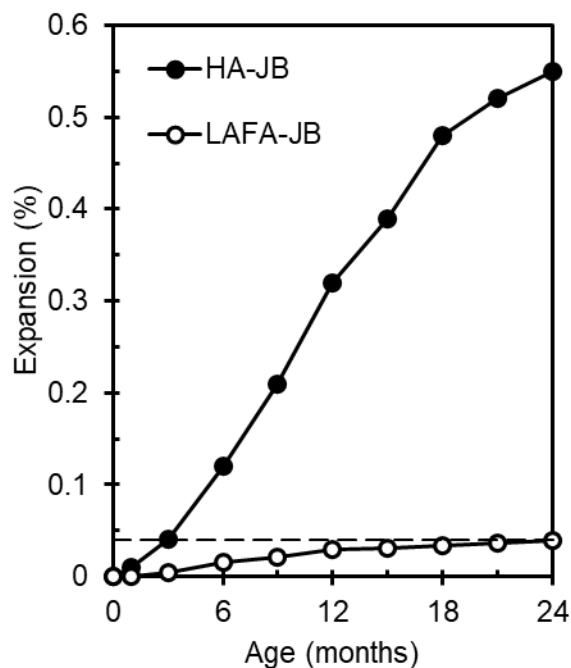


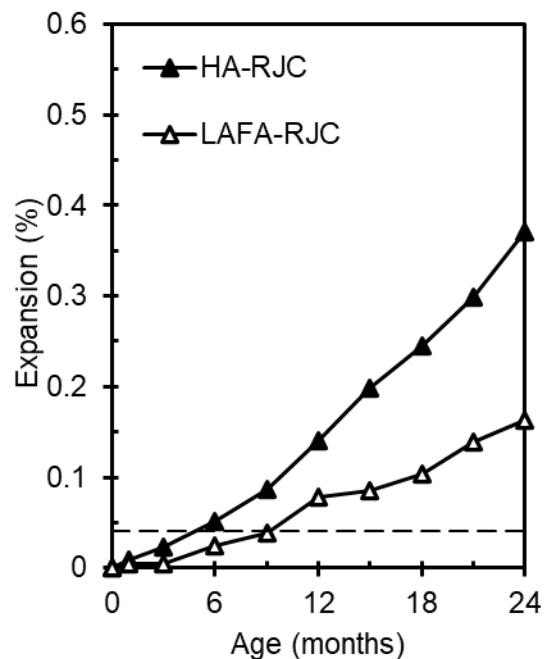Figure 3.1: Expansion with Jobe Sand

Figure 3.2: Expansion with Untreated RJC

### 3.2 Concrete with Untreated Recycled Concrete Aggregate (RJC)

Figure 3.2 shows the expansion of concrete prisms produced with the untreated RJC material as the coarse aggregate and a non-reactive fine aggregate. Concrete containing the RJC and HA cement expanded rapidly but compared with the mix with HA cement and Jobe sand, the rate of expansion was slower and the ultimate expansion was less. The concrete with the RJC material and the low-alkali cement/fly ash combination (LAFA) also expanded but to a lesser degree than with the HA cement. Petrographic examination of concrete prisms with HA and LAFA cement at 2 years revealed evidence of ASR within the particles of the untreated RJC and cracking emanating from some of these particles.

### 3.3 Concrete with Carbonated Recycled Concrete Aggregate (RJC-CO2)

Figure 3.3 shows the results for concrete produced with the carbonated RJC material (RJC-CO2) as the coarse aggregate and a non-reactive fine aggregate. Concrete with HA cement did show some expansion which was significantly lower than the other concretes with HA cement. The concrete produced with the LAFA combination did not exhibit deleterious expansion throughout the 2-year period; the expansion at 2 years was just 0.024% and there was no evidence of surface cracking at that time. Petrographic examination of prisms at 2 years revealed the presence of ASR gel and cracking within the $CO_2$-treated RJC particles in the concrete produced with HA cement but no evidence of ASR was found in the concrete produced with LAFA cement.

### 3.4 Concrete with Lithium-Treated Recycled Concrete Aggregate (RJC-LN)

Figure 3.4 shows expansion data for the lithium-treated RJC aggregate (RJC-LN) and the 80/20 combination of LA cement/fly ash (LAFA). The concrete showed a small amount of deleterious expansion, 0.042%, at 18 months and this was accompanied by very faint surface cracks measuring approximately 0.1 mm in width. The expansion did not increase beyond this amount with further exposure; indeed, the 2-year expansion was slightly lower at 0.033%. There was no associated mass loss of the prisms between 18 months and 2 years, so the small amount of shrinkage is not attributed to moisture loss (desiccation) of the concrete. Petrographic examination of these prisms revealed some evidence of alkali-silica gel formation in the lithium-treated RAC particles but no evidence of cracking emanating from the particles into the surrounding paste. Unfortunately, there are no results for the combination of HA cement and RJC-LN as there was insufficient RAC to produce this concrete.
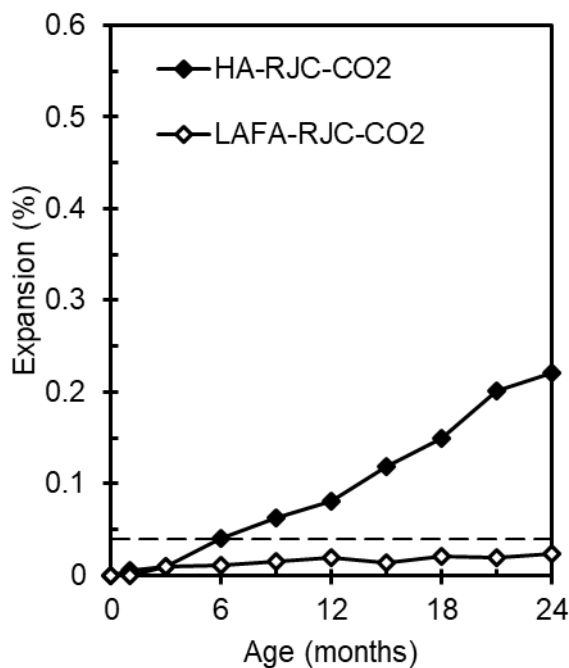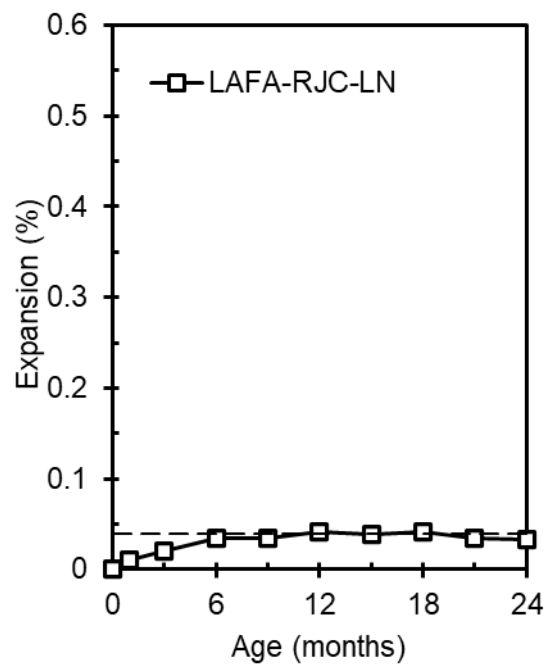


Figure 3.3: Expansion with Carbonated RJC    Figure 3.4: Expansion with Li-Treated RJC

## 4. DISCUSSION

The data show that the use of recycled concrete aggregate produced from ASR-affected concrete can lead to expansion and cracking of new concrete unless the aggregate is treated. Previous workers [7] have shown that such expansion can be prevented by using suitable amounts of pozzolans, but that the amounts required are more than that needed for virgin reactive aggregate. In the case studied here, expansion was observed even when the binder was comprised of a low-alkali cement (0.46% $Na_2Oe$) in combination with 20% of a low-calcium fly ash. This same cementitious material combination (LAFA) did not lead to expansion and cracking of the concrete with the virgin Jobe aggregate.

In the case of the expansion and cracking observed with concrete comprising the untreated RJC aggregate and the LAFA cementing system, it is proposed that the source of alkalis required to promote ASR in the new concrete is within the RJC aggregate itself. Figure 4.1 shows a schematic of the RJC (left) and the fresh concrete produced with the RJC (right). The RJC was produced with HA cement and Jobe sand, and was just 3 months old when the fresh concrete was mixed. At this age there is still considerable potential for further reaction within the RJC particles as both alkali hydroxides and reactive silica are still present. The potential for further ASR within the particles is demonstrated by the amount to expansion observed beyond 3 months in the concrete containing HA cement and virgin Jobe sand (HA-JB in Figure 3.1).

Carbonating the RJC prior to use in fresh concrete will reduce the pH from somewhere in excess of 13.1 to approximately 8 and there will be insufficient alkali hydroxides within the particles to sustain ASR. Consequently, producing fresh concrete with the carbonated RJC-CO2 aggregate and a "low-alkali binder system" such as LAFA results in no expansion because there is an insufficient supply of alkali hydroxides in the system to fuel ASR despite the abundance of reactive silica (e.g. unreacted Jobe) that remains. If, however, this same aggregate (RJC-CO2) is combined with a "high-alkali binder system" such as HA cement, expansion can result as the alkali hydroxides that are present in the fresh paste can diffuse into the carbonated recycled aggregate particles and react with any remaining unreacted silica (Jobe) in these same particles.

An alternative approach to "neutralizing", by carbonation, the alkali hydroxides that remain in the original cement paste phase of the RCA particles is to "balance" the concentration of $Na^+$ and $K^+$ ions with a sufficient concentration of $Li^+$ ions. It is well-established [20] that the expansion of concrete containing certain alkali-silica reactive aggregates can be prevented by adding a sufficient quantity of lithium. Generally, the amount of lithium required increases as the availability of sodium and potassium increase and it has been shown that establishing a lithium-to-sodium-plus-potassium-molar ratio of [Li]/[Na+K] ≥ 0.74 is usually sufficient [20]. The lithium-treatment of the RJC was borderline effective when the RJC-LN material was combined with the "low-alkali binder system" (LAFA). It is suspected that this treatment would be less effective if RJC-LN was combined with a "high-alkali binder system" as the value of [Li]/[Na+K] will likely be diminished. It should be noted that lithium-based admixtures are not effective in preventing ASR expansion with all types of alkali-silica reactive aggregate [20].



**Recycled Jobe Concrete Aggregate (RJC)**          **New Concrete Containing RJC**
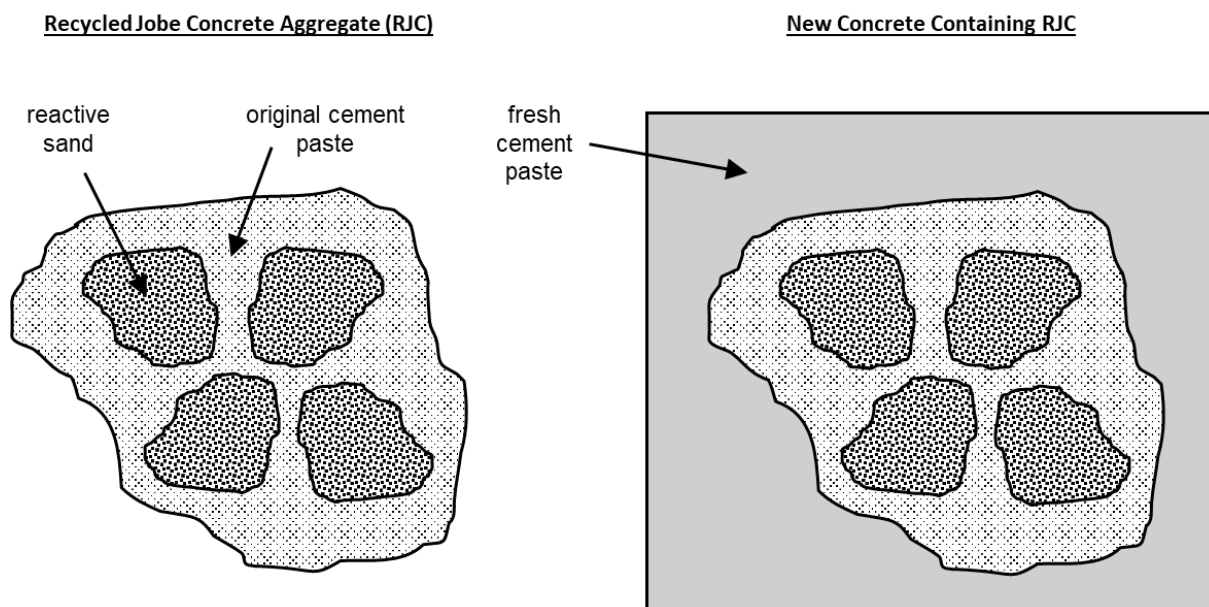
Figure 4.1: Schematic showing different phases within fresh concrete produced with RCA

The benefits of pre-carbonating RCA to improve certain properties of concrete have been reported in the literature [11–14] and have been mainly attributed to improvements in the microstructure of the mortar phase [15]. This study indicates that changes in the pH of the pore solution due to a reduction in the availability of alkali hydroxides after carbonation may be a further benefit if the RCA has been produced from concrete affected by ASR provided the availability of alkalis is also controlled when the

RCA is reused in new concrete. Although there have been numerous laboratory studies on carbonating RCA the commercial and technical viability of conducting this technology on an industrial scale has yet to be established but demonstration projects are underway in Europe (www.fastcarb.fr).

## 5. CONCLUSIONS

Recycled concrete aggregate (RCA) was produced by crushing and grading (20-5mm) three-month-old mortars containing high-alkali cement and highly-reactive sand (Jobe). This RCA was used either without treatment (RJC) or following carbonation (RJC-CO2) or lithium-soaking (RJC-LN) to produce fresh concrete using either a high-alkali (HA) cement or a combination of low-alkali cement plus fly ash (LAFA). From the results of concrete-prism expansions tests, the following conclusions can be drawn:

- ASR expansion can occur when the untreated RJC is used with a "low-alkali binder system" (LAFA).
- ASR expansion can be prevented by carbonating the RJC (RJC-CO2) and combining it with a "low-alkali binder system" (LAFA).
- ASR expansion can occur with the carbonated aggregate (RJC-CO2) is combined with a "high-alkali binder system" (HA).
- Limited ASR expansion (0.042%) accompanied by very fine microcracks (~ 0.1mm) was observed when the RJC was treated with lithium (RJC-LN) and combined with LAFA. Unfortunately, no data were obtained for RJC-LN combined with HA cement

It should be noted that this study used just one type of reactive aggregate which is considered to "very highly reactivity" by the criteria in ASTM C1778. Further testing is required to determine if the results of this study are applicable to aggregates of lower reactivity with different mineralogy.

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