

# A Review of Some Reactive Aggregates from the United Kingdom with Reference to the Mechanism of Reaction and Deterioration

W.J. French

*Geomaterials Research Services Ltd.  
Billericay, UK*

## ABSTRACT

Some 40% of over 300 structures show evidence of alkali-aggregate reactivity. The reactions range from just detectable to serious destructive processes. The reactive aggregates are mostly highly polymictic sands and gravels but include crushed rocks. Reactive lithologies include chert, flint, siliceous ironstone, meta-argillite, greywacke to argillite, metaquartzite, siliceous volcanics, and calcitic dolomites. The expansive reactions mostly appear to take place within the aggregate and serious expansion and cracking is often related to the juxtaposition of two or more reactive particles. Cracks developed within the aggregate are thick in the central parts of the rock fragment and become thinner towards the aggregate surface. Gel occurs within the pores of the aggregate and issues from the rock fragments into cracks in the paste. These cracks are most damaging where a few reactive particles are close together and here the cracks tend to link the adjacent rock particles. In samples where aggregate particles have produced alkali-silicate gel it is sometimes found that very high levels of alkalis have been developed within other aggregate fragments without the particles themselves reacting. The reactions encountered do not appear to conform with the normal pessimum levels for particular lithologies and the uneven distribution of aggregate is thought to be of particular significance in producing damage. At a late stage in some reactions ettringite has been found to form in the gel and this appears to lead to further potential for deleterious expansion.

## 1. INTRODUCTION

This paper summarises observations made by the author on more than 300 concrete structures in the United Kingdom over a period of about a decade. Most of these structures showed cause for concern after an initial inspection and about 40% were found to show alkali aggregate reaction. Most of the concretes were placed in situ but some were precast and some prestressed.

Core samples and lumps have been studied with the petrological microscope in reflected and transmitted light. Many of the samples have been examined with the scanning electron microscope and analysed with the attached energy dispersive analytical system. Large thin sections, polished plates, and freshly broken surfaces of the concretes have been examined and modal compositions have been determined by point counting.

The volume proportions recorded include the amounts of aggregate, paste, voids, rock types, remnant clinker, portlandite, hydrated paste, and P.F.A and slag. Measurements and observations such as the number of microcracks per metre, the crystalline content of the cracks, the content of voids, and the degree of strain in quartz have also been made. Numerous samples have been studied in accelerated expansion tests, at 15<sup>o</sup>, 30<sup>o</sup>, and 38<sup>o</sup>C and some of these have been studied microscopically after testing.

Almost all the materials examined showed some defect though in some 30% of cases these were trivial. Microcracking is common with between 50 and 350 microcracks occurring per metre. In some 60% of the samples the microcracking could not be ascribed to an alkali-aggregate reaction and almost all of these cracks appeared to be due to shrinkage. The microcracks have led to serious deterioration of the paste of many samples through recrystallization with the formation of excessive amounts of portlandite, ettringite or thaumasite. Carbonation also occurs along some of the cracks, penetrating more deeply than can be detected by the phenolphthalein test. Penetration of de-icing salts along the microcracks is also of importance in producing accelerated deterioration of the paste and corrosion of reinforcement.

In the 40% or so of cases where an alkali-aggregate reaction was detected the expansion was not always of sufficient magnitude for deleterious expansion to have occurred. The techniques of study allow identification of alkali-silicate gel where the whole reaction system represents less than 0.01% by volume of the concrete. In a few such cases other more significant damage occurred as a result for example of salt attack or reinforcement corrosion. This might have been exacerbated by the reaction and the various processes are sometimes mutually enhancing.

The occurrence of reaction was found to vary greatly from sample to sample from a given structure and even within single samples. Three cores out of sixty from one structure for example showed a moderate degree of reaction while for the remainder no trace of reaction could be found. This reflected the distribution of rock types in the aggregate. Other variability in magnitude of reaction has also been found to be partly due to variation in the abundance of rock types but also reflects in part the location of the sample in the structure, accidental heterogeneity of composition of the mixture, and environmental factors. This variability is important because the diagnosis of incipient or low levels of reaction cannot always, or perhaps even commonly, be carried out using thin sections alone, however large and well made they may be. It has therefore been found necessary to prepare special polished plates from the concretes and to store these under standard conditions before examination in order to assess the magnitude and type of reaction taking place.

## 2. REACTIVE AGGREGATES

The reactive aggregates are mostly natural sands and gravels. Some of these are sea-dredged but most are from inland, fluvial, terrace and glacial gravel sources. They are highly polymictic and in a few instances have also been mixed with other gravels or crushed rock aggregate. Some of the gravels have been crushed but the particles showing reaction are generally the natural pebbles. Rarely, certain crushed rock aggregates were found to be reactive and one instance was found of slight reaction involving crushed flint.

The following five groups of reactive materials have been encountered: (i) chert (including extremely rare instances of slightly reactive flint) and siliceous ironstone; (ii) greywacke and related siltstones and argillites; (iii) low grade meta-argillites (e.g phyllite); (iv) fine grained acid volcanic rocks containing glass and devitrified glass; (v) metaquartzite. Two or more of these ingredients may occur together. The first three types have been found to take part in substantial expansive reactions with much cracking being generated. The glassy volcanic rocks occur in samples which also show reactions involving cherts. The evidence for reaction of metaquartzite suggests that this is at least an uncommon reaction and the almost ubiquitous

occurrence of associated chert always introduces an element of uncertainty in the diagnosis of the reactive rock type. Carbonate rocks have also been found to be reactive in a few structures. These are as usual fine grained calcitic dolomites with a small amount of clay grade silicate material.

### 3. MAGNITUDE OF REACTION

The magnitude of reaction encountered varies from a level which is scarcely detectable to complete degradation of the concrete. In the latter case all that remains in the worst places is the degraded paste and the loose degraded aggregate with abundant gel. This generally occurs locally within cores and in the structure and is associated with obvious random open cracking and abundant gel production throughout the concrete. The gel is readily seen on freshly broken surfaces and often fills voids. Such occurrences of total deterioration occur typically in foundations and in parts of structures which are against saturated substrates or frequently remain wet through poor drainage arrangements.

The magnitude of deterioration has been recorded on an arbitrary scale of from 0 to 10 where the totally decomposed material is 10 on the scale. About half the examples of alkali silica reaction encountered are on the first three steps of this scale which represent reaction which may not itself be structurally significant and where the amount of microcracking is not large and where the amount of gel produced is moderate to low and often of restricted occurrence. For these grades no field evidence occurs of the deterioration except in particularly adverse environments. Levels 4 to 6 represent a range of deterioration where there will typically be obvious field evidence of degradation but where the concrete remains coherent and relatively strong. For levels 7 to 10 there may well be major cracks present along with large amounts of gel and microcracking and it may be difficult to obtain coherent samples other than by careful selection of locality. Only in some seven or eight cases has this level of deterioration been observed. The most serious cases of reaction relate to the occurrence of cherts, argillites, and meta-argillites.

### 4. DIAGNOSIS OF REACTIVE AGGREGATE

In all cases of alkali-silica reaction encountered the occurrence of alkali-silicate gel is taken as the main diagnostic symptom. The amount of gel generated is not always related to the amount of micro and macrocracking observed and varies with the aggregate type. In the case of the greywacke-siltstone-argillite series, the amount of gel found is low where greywacke is involved but can be high where argillites are reactive. Gel in the case of greywacke tends to be viscous and immobile while that in the case of the reactive argillites is typically highly mobile and hygroscopic and issues onto broken surfaces. Reaction of meta-argillite has also been found to produce much gel. Moderate to large amounts of gel also occur where cherts, fine grained acidic rocks, or siliceous ironstones are involved in the reactions. The consistency of this gel varies from sample to sample and is sometimes free-flowing and in other cases highly viscous or dry.

Identification of reaction centres and the reactive aggregate types is generally difficult. Gel has been found to migrate along cracks, through pores in the paste, and may occur at some distance from the reaction centres. On polished plates it can often be seen to form a thin coating to a range of aggregate types but this is not regarded as evidence that the particles are themselves reactive. Microcracks along aggregate surfaces are commonplace, and it is to be expected that mobile gel will migrate along such cracks. It is usually possible to find gel issuing from within the reactive particle onto polished surfaces. This has been found to occur for cherts, siliceous ironstones, greywacke-type lithologies, argillites, and meta-argillites. These particles also frequently show internal decomposition and loss of mass and in some such occurrences radial microcracking occurs in the immediately surrounding paste. These indications of gel forming within the aggregate and the loss of mass of the aggregate are taken as positive evidence that the fragment has reacted, though even here ambiguities can arise in some instances.

In some concretes where members of the greywacke series appear to be reactive rocks, the amount of gel encountered is small and the amount of obvious microcracking is not large. The concretes often show evidence of significant deterioration out of proportion to the obvious evidence of reaction indicated by the abundance of gel. The electron microscope shows that extremely fine cracking occurs in these samples which cannot be seen in thin section. This fine microcracking may represent a general expansion and an increased permeability which could lead to general deterioration from various causes without major random cracking being apparent.

## 5. ALKALI DISTRIBUTION

The general level of alkalis has been found to be moderate to high with  $K_2O$  commonly exceeding  $Na_2O$ . Determinations of the alkalis have been made using the electron microprobe and are not therefore overall averages. Nevertheless they point to initial concentrations ranging from about 0.7 to 2.5 wt.% of the paste with common values being around 1.2% as total alkali equivalent. These results span the widely accepted boundary between alkali concentrations that are considered to be potentially deleterious and those that are considered safe. The concentration of the alkalis is usually extremely irregular with values exceeding 4% often occurring in the vicinity of particular aggregate fragments.

The alkalis can be exceptionally high inside some aggregate particles such as cherts and occasionally in greywackes and argillites. Spot analyses give over 70%  $K_2O$  in some chert particles where the  $K_2O$  occurs as minute well defined needles in pores in the heart of the chert particles. These needles may be precipitated from solutions which are evaporated during the sample preparation or be original solids. There is typically a minor amount of  $Na_2O$  in the analyses of these needles, together with some silica. The latter could represent contamination from the chert. In the outer part of the chert there is often a zone which contains small prismatic crystals of sodium silicate. These particular chert grains occur alongside other chert particles which have alkali silicate gel within their micropores. It is not clear why some of the chert particles have reacted to produce the gel and others have not. The alkali concentration gradients begin to fall towards the surfaces of the aggregate pieces and usually decline to background level in less than a millimetre from the aggregate surfaces unless microcracks containing gel are present.

## 6. MICROCRACKING IN AGGREGATE

Where reaction is clearly associated with particular aggregate types, it has been found that cracking occurs within the aggregate itself. The cracks are thickest in the centre of the aggregate particle and may be very thin indeed at the surface of the aggregate. There is sometimes an abrupt increase in the gape of the crack as it passes into the surrounding paste. Gel occurs in the pores and cracks within these aggregate pieces and is often seen to have built up in a series of layers in the cracks where they pass from the aggregate into the paste. This layered structure occurs for much of the crack length. In some samples cracks of two or more generations are present in the paste.

In some instances cracks occur within the aggregate but do not pass out into the paste. In other places where two or more reaction centres are close together, the cracks link the reactive particles. This pattern is considered to indicate that the expansive reaction takes place within the aggregate particles; the expansive forces generated by closely spaced aggregate particles causing cracks to propagate between the two expanding centres. Clusters of reactive particles appear to act as local expansion centres generating the larger macroscopic cracks and this non-uniformity in the distribution of aggregate is considered to be of importance in influencing the magnitude of deterioration produced. This variety of crack structure has been observed where the reactive particles are cherts, argillites and meta-argillites and occasionally greywackes. The cherts that develop the cracks are always natural pebbles with a structure in which an outer shell of very fine grained and apparently non-porous chert surrounds an obviously porous centre.

The electron microscope shows the chert to be microgranular throughout, with organic structures often present. The outer zone has extremely small pores, but its total porosity is actually very similar to that of the inner material. It is considered that this structure causes alkalis to be extracted from the paste through some process involving capillarity and concentrates them in the larger central pores of the pebbles. The high internal alkali concentration has not been found to occur where the chert particles are fractured or crushed before incorporation in the concrete. White flint has been found to extract alkalis when crushed but does so more strongly when present as zoned pebbles. Where cracks occur in argillites and meta-argillites, they tend to follow the bedding or cleavage planes. In both cases there is some evidence that these planes of weakness are acting as extremely small interconnected sheet-like capillaries which could draw material from their environment in the same manner as that postulated for the structured cherts.

It is unusual to find microcracking within siliceous ironstones that are reactive and these more often appear cavernous and can lose up to 50% of their mass in the reaction. Sometimes the surrounding paste contains dispersed gel and is coloured by the iron oxides. Metaquartzites often have microcracks along their surfaces which contain gel but in no case has it been possible to demonstrate unambiguously that the rock itself has reacted with loss of mass to produce the gel and the evidence for reaction is based on absence of other likely culprits. The degree and type of strain in the quartz in all the samples previously examined is being investigated to see if some general proposition can be found to indicate the potential for reactivity and to confirm whether reaction has occurred.

#### 7. SEQUENTIAL PROCESSES IN REACTIONS

The concretes examined vary from about 5 to 80 years old with most of those showing reaction being between 15 and 30 years. Early stages in the history of reaction have been identified by microscopic techniques before they become evident in the field. Small amounts of gel are produced and high localised alkali concentrations can be detected. Younger concretes, where the reaction is more obvious, show the presence of mobile clear gel which is demonstrably hygroscopic. This often issues with some slight pressure onto cut surfaces. In most older concretes (i.e. greater than 15 to 20 years), the gel is usually white, and has a dried or creamy consistency and does not obviously take in moisture. It is commonly carbonated and sometimes contains calcium.

In older concretes - especially those more than 25 years old - it is common to find that the gel is partly or largely replaced by needles of ettringite. This mostly occurs in the microcracks. This formation of ettringite has also been found to occur where reactive concretes from various structures have been subjected to accelerated expansion tests. The mineral has developed as a consequence of the test, and is associated with the use of slightly elevated temperatures and very high humidities. This form of recrystallization develops as a normal consequence of steam curing and it is suggested that continued expansion of concretes which have shown reaction could result from the growth of the sulpho-aluminate where the structures are maintained in a warm, humid environment.

#### 8. PESSIMUM PROPORTIONS

No evidence has been found that in any of the samples examined a normal pessimum proportion has occurred. For example, serious expansion has been found to occur where the amount of chert varies from 4 to 10% of the aggregate. Reactive chert has been found to make 2% of concretes containing 70% of reactive meta-argillite. Where the series greywacke to argillite is reactive, the rocks make from 5 to 30% of the aggregate and the amount of reaction tends to increase as the amount of the aggregate increases. Possibly reaction would be more severe if other proportions of the reactive constituents were to occur. Since the reaction often appears most damaging where the reactive constituents are clustered it is conceivable that pessimum proportions are established patchily.