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# Different manifestations of the alkali-silica reaction in concrete according to the reaction kinetics of the reactive aggregate

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## Abstract

The alkali-silica reaction (ASR) is caused by the presence of reactive aggregates in contact with sufficiently alkaline pore solution and a moisture level above 80%, which leads to the formation of expansive products that cause cracking and deterioration of the structures. Petrographic analysis of the ASR damaged microstructure provides information about the detection, analysis, and progress of the reaction. A stereobinocular and a polarizing optical microscope were used in order to observe and establish the relationship between ASR development in rapid or slow-reacting aggregates. The origin, mineralogical composition and fabrics of the constituent of the two types of aggregates were analyzed first. The progress of reaction was then studied on four concrete samples analyzing the different characteristics and textural patterns imprinted on the aggregates and the mortar. From the study it follows that the mineralogy and fabric of the rocks involved are responsible for different manifestations of the reaction in the aggregate and in the interfacial transition zone, causing damage that can diminish the strength and durability of concrete.

*Keywords:* Alkali-silica reaction; Aggregate; Petrography; Microstructure; Durability

## 1. Introduction

The alkali-aggregate reaction is an expansive reaction between the alkalis in the pore solution of concrete and minerals in the aggregates, under humidity conditions. According to the literature there are basically two types of deleterious reactions. The alkali-silica reaction (ASR), Stanton [1] and the alkali-carbonate reaction (ACR), Katayama [2], Tang Mingshu [3]. Their names are derived from the composition of the rock involved in the reaction, microstructural and external manifestations being somewhat different. It is also advisable to divide the ASR into two groups based on the evolution rate of the reaction as a consequence of the rock origin. In nature rocks are found that contain a variety of minerals of the silica group, where silica usually has different thermodynamic states. The first group of rocks is composed of vitreous or amorphous species; volcanic glass and opal are two typical examples, they react quickly with deleterious effects about a year after the structure was built, but this period is also dependent on environmental conditions.

The second group of rocks includes crystalline rocks. These rocks have undergone deformation and significant thermal changes due to a number of tectonic processes acting during the evolution of the Earth's crust. One of the constituent minerals, quartz, can be found in a strained and/or deformed state. This group shows a slow or delayed reaction with deleterious expansive effects that can be seen after many years, 6 to 10, or even at later ages. In both cases the reaction exhibits a typical behavior, the aggregate is attacked by the concrete pore solution that contains $OH^-$, $K^+$, $Na^+$, and $Ca^{+2}$ ions. Because of this reaction a sodium and potassium silicate gel that has the ability to absorb water is formed, McGowan and Vivian [4], which is

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the fundamental cause of expansion and subsequent concrete cracking, Diamond [5]. Therefore, it is important to study the internal microstructure of concrete, John et al. [6] since it contains information that will allow identifying the pathology that affects the structure, assessing the degree of progress and, together with other information, forecasting the future evolution of the reaction to take precautions, carry out structure maintenance, and predict service life.

In this paper alkali-reactive aggregates are identified, a brief introduction to the two main groups that cause the ASR is given, and then the results on the study of ASR-affected concrete samples are given. These concrete samples were taken from the structures in service and from laboratory test specimens, and contain aggregates of different origins that produce either rapid or slow reactions. Common techniques have been applied for carrying out the petrographic examination with a stereobinocular and a polarizing microscope.

Polished sections of each of the samples were observed to establish how the deleterious reaction develops according to the type of reactive aggregate contained in each concrete sample. The aggregate was first analyzed in relation to its origin and mineralogy, and then the attack characteristics were observed as well as the resulting textures imprinted on the aggregate and the paste.

## 2. Methodology

Samples were taken from the aggregates of different origins and hardened concrete. Polished sections were made to be subsequently analyzed by a 60X sterobinocular microscope, an OLYMPUS BHSM2 UMA polarizing microscope, and an image analyzer Image-Pro Plus, version 4.1. Other supplementary techniques such as staining with tetraamine copper sulfate and ultraviolet fluorescence with uranyl acetate, Natesaiyer [7,8], were used for identifying the reaction products.

## 3. Characteristics of the different types of ASR aggregates

In previous studies, Giovambattista et al. [9], Batic and coworkers [10,11], Ponce et al., [12], performed on hardened concrete samples it was observed that according to the type of reactive aggregate the ASR exhibits different manifestations. Two different types of aggregates can be grouped as per their reaction rate—those that exhibit a rapid reaction and those that have a slow or delayed reaction, Fournier and Berube [13]. The behavior and manifestations in concrete are directly related to the mineralogy of the aggregates, different compositions, and their own microstructures that are closely linked to their origin.

### *3.1. Rapid-reacting aggregates*

These aggregates are obtained from rocks composed of minerals that have significant initial alkali reactivity. They are:

#### *3.1.1. Volcanic igneous rocks*

This set of rocks is formed by the rapid cooling of the magma when it is exposed to the atmosphere or to environments with a marked thermal contrast. The quick heat loss of the silicate melt causes the formation of abundant crystalline nuclei that grow quickly forming a large number of small crystals. This is reflected in grain sizes that range from fine to very fine. When cooling is too quick, crystalline nuclei cannot be formed and thus no crystal formation occurs. There is, instead, a natural amorphous material called volcanic glass, which lacks an ordered atomic structure because the atoms did not have enough time to accommodate themselves and form crystalline networks, Best [14]. In this case, the internal free energy increases because of this disorder, which implies that there is metastability in the structure and therefore glass becomes a potentially reactive substance. The amount of glass in a volcanic rock is governed by its thermal history, severe overcoolings form larger amounts of glass that can either form entirely vitreous rocks (obsidians, pumices, perlites, vitrophyres) or be a component in a volcanic groundmass as in rhyolites and basalts. These latter rocks are frequently used as concrete aggregates and therefore glass becomes their main reactive component. Fig. 1 depicts a vitroclast in the mortar of a concrete showing its fluidal structure, marked with a dash line (L); this is an example of quick cooling.

#### *3.1.2. Sedimentary rocks*

The second type of rocks analyzed are sedimentary rocks. Since different types of mineral varieties of the silica group are usually involved in their origin, chalcedony being one of them, this variety occurs in micro- to cryptocrystals with a habit that ranges from fibrous to radiated fibrous and can be seen only under an optical and electronic microscope. It is softer than quartz and denser than opal. The latter is another component of the silica group, which has a characteristic semicrystalline structure since it is partly amorphous and partly consists of very small *lepispheres* (silica spheres) composed of successive layers of cristobalite and tridymite, of equal sizes, arranged as a packed structure. Moreover, water is usually present in its structure and normally varies between 4% and 9%, Deer et al. [15]. These two forms of silica are derived from the precipitation from low-temperature fluids or from the alteration of primary minerals.

Sedimentary rocks cover a range of varieties depending on the different formation processes including meteorization,

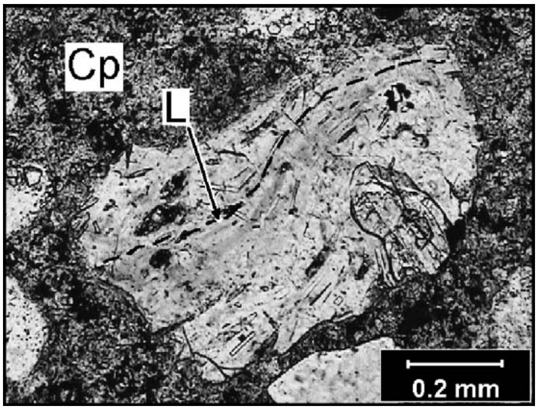

Fig. 1. Vitreous fluidal structures in a vitroclast, (Cp) cement paste. Concrete thin section viewed through parallel polarizers.

transport and deposition of sediments, chemical or biochemical precipitations, etc. A brief classification based on these factors is given below:

A. Clastic: they are derived from the weathering of pre-existing rocks. Their textural arrangement corresponds to clasts agglutinated by a finer phase called matrix, a third phase called cement is usually present, and both of them hold the larger grains together. Examples of these lithologies are lutites, limolites, sandstones, conglomerates, etc. The cements have two main compositions, siliceous and carbonate, and many times they are the only materials cementing the clasts. From the reactivity point of view siliceous cement is usually composed of a mixture of microcrystalline quartz, chalcedony, and opal, a mineralogy that causes a fast reaction rate.

B. Chemical and biochemical: their origin is related to the precipitates from inorganic reactions in water forming minerals such as microcrystalline quartz, chalcedony, opal, and calcite. The rocks consist of chert, limestone, and dolomites. Biochemical sedimentary rocks are derived from organic activity (animals and sea plants) forming rocks whose composition is similar to the above.

### *3.2. Slow-reacting aggregates*

The kinetics of these aggregates is mainly related to strained quartz grains, which are thermodynamically unstable and have high free energy in their crystalline networks as a result of their deformation. This type of quartz is mainly present in deformed granitic rocks and metamorphic rocks of varying grades.

#### *3.2.1. Granitic rocks*

They correspond to plutonic igneous rocks. Their origin is linked to the slow cooling of the magma forming completely crystalline textures that generally consist of crystals larger than 1 mm. One of their basic constituent minerals is quartz, which is the most important for ASR development, and in this type of rock it may range between 20% and 60%. These rocks have been usually subjected to thermotectonic events where quartz crystals have crystalline defects that render it unstable with respect to the alkalis in the concrete.

#### *3.2.2. Metamorphic rocks*

They are rocks that due to the changes in the prevailing physical and chemical environmental conditions undergo changes in their mineralogy, structure, and chemical composition, Bucher and Frey [16]. They are mainly characterized by the presence of a penetrative planarity in all the rock volume as the result of differential pressures. Their textural and mineralogical characteristics are directly related to the composition of the parent rock (protolith) and to the increase in the metamorphic grade imposed. They cover from slates, phyllites (lower metamorphic grade) to schists, gneisses, granulites, migmatites, the latter three with a greater metamorphic grade where quartz is abundant according to the original composition of the rock subjected to metamorphism.

Quartz is one of the constituent minerals in this type of rock. It always exhibits deformation signs, from low to high degrees (mylonites), it is elongate and lineated, which make these rocks potentially reactive, Blum et al. [17].

## 4. Characteristics and composition of the aggregates studied

A series of aggregates were analyzed for their reaction kinetics in concrete. The rapid-reacting aggregate is made up of a clastic sedimentary rock (orthoquartzite) whose main outcrops occur in the Province of Chaco, Argentina. They are heterogeneous deposits composed of quartzites (orthoquartzites) and sandstones, with minor intercalations of calcareous rocks and clays occurring as layers or pockets. More than 95% of the rock consist of rounded to subrounded quartz clasts. It does not have a matrix, so the material holding the grains together is the siliceous cement composed of microcrystalline quartz, chalcedony, and opal, which makes the rock highly reactive. The development of the clastic texture, quartz clasts (Qc) cemented by opal (Op) and chalcedony (Ca) is shown in Fig. 2. The optical properties of opal that, as it is amorphous, it appears darkened (isotropic), are also shown; chalcedony can be seen in a fine bright fibrous microcrystalline mosaic.

The slow-reacting aggregates studied cover three rock types. The first one is a granitoid of granitic–granodioritic composition showing deformation signs; the second is a granodioritic orthogneiss. They both come from the Paleoproterozoic metamorphic igneous basement of the Río de La Plata craton, Province of Buenos Aires, Dalla Salda et al. [18]. The third one is made up of quartz-mica schists from the Sierras Pampeanas in the Province of Tucumán, Argentina.

The granitic–granodioritic granitoid consists mainly of quartz, alkaline feldspar, and plagioclase with varying amounts of biotite, and minor hornblende and magnetite. Its grained texture shows clear signs of deformation, especially subgrain formation (Sg) and quartz recrystallization, tapered twins in plagioclases (Tt), myrmekites (My), and kinking in micas are further evidence of deformation, as shown in Fig. 3a.

The granodioritic orthogneiss shows a large amount of recrystallized quartz, alkaline feldspar and plagioclase, which are

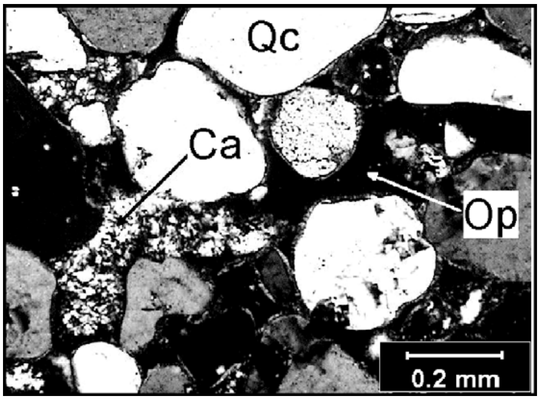

Fig. 2. Clastic texture and optical properties of opal and chalcedony in orthoquartzite. Thin section viewed through crossed polarizers.

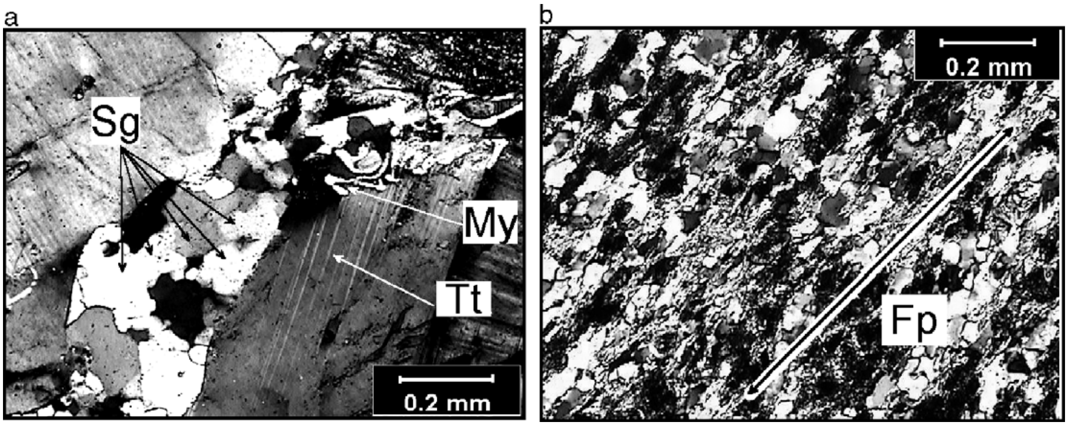

Fig. 3. (a) Granitoid showing its grained texture with deformation signs. Thin section viewed through crossed polarizers. (b) Microstructure of granitic orthogneiss. Thin section viewed through crossed polarizers.

partially altered to illite–sericite, and clinozoisite. A continuous foliation given by sericite orientation and quartz elongation can be distinguished. The direction of foliation planes (Fp) is shown by a black arrow in Fig. 3b.

Quartz-mica schists consist mainly of quartz, muscovite, and in lower proportions biotite, plagioclases, alkaline feldspar and opaque minerals. Recrystallized strained quartz, the slow-reacting mineral, is abundant, fine-grained (between 20 and 250 μm), anhedral, with undullose extinction and usually diffuse crystalline boundaries. Schists show marked penetrative planarity forming a continuous schistosity given by the elongation of felsic minerals (quartz–feldspar) and the parallel orientation of muscovite sheet.

## 5. Reaction pattern and textural characteristics in concrete according to the aggregate reaction kinetics

The intrinsic properties of the rocks used as concrete aggregates are exposed to alkali attack giving rise to deleterious signs that appear both in the aggregates and in their neighborhood with highly contrasting characteristics.

The behavior of a concrete whose coarse aggregate was Chaco orthoquarzite, concrete 1 (C1), was studied first. In this clastic sedimentary rock the mineral grains are cemented by siliceous cement that, as stated above, is highly alkali reactive, especially opal. The clasts are partially or totally surrounded by the cement (like islands), Fig. 2. Therefore, there is a large aggregate surface covered by this material that is exposed to pore solution attack.

Observations performed on the concrete containing this aggregate showed that the reaction dynamics in orthoquartzite starts when the highly alkaline pore solution produces, at the interfacial zone (Iz) between orthoquartzite (Oq) and the mortar (Mo), a homogeneous reactive front (Hrf) that attacks the siliceous cement of rock. This causes a strong dissolution of the cement due to the high pH and the formation of a reaction rim (Rr) with gel development (G) in that zone, Fig. 4a. The alkaline solution penetrates the reactive sites (siliceous cement) of the aggregate leaving dissolution zones, pushing the quartz grains apart and causing an increase in porosity. Fig. 4b depicts a thin section of (C1); at the interface (I) between the cement paste (Cp) and the orthoquartzitic aggregate it shows how opal cement (Op) has been attacked leaving dissolution zones (Dz), unbound quartz grains, and gel formation (G) at the interface.

Following the reaction dynamics, the gel formed from reaction sites exerts pressure, thereby causing cracks through which it moves to other places such as pores, the cement paste, interfaces, etc. Fig. 4c, shows a microcrack (Mck) filled with gel that runs through the interface of the orthoquartzitic aggregate (Oq) and migrates to the cement paste (Cp).

As a result of this attack, dissolution, gel formation and migration the siliceous cement that binds the clasts together is partially or totally removed from the aggregate zones adjacent to the interface. Therefore there is a cohesion loss of the mineral grains and clear unbinding with the mortar (Mo), thereby giving a "dissolution texture" evidenced by reaction rims (Rr) that in the aggregates analyzed at the age of 90 days in some cases reached a thickness greater than 1 mm, Fig. 4d. The attack proceeds to the interior of the aggregate, as shown in Fig. 4e, where gel formation (G) and its expansion cause cracks (Ck).

The slow-reacting aggregates show patterns and textures that are markedly different from the others analyzed before. These differences lie mainly in the mineralogical type and the structural condition of this mineralogy. In this type of aggregate strained quartz has a key role as the reactive mineral, Dolar-Mantuani [19], French [20]. Owing to its arrangement and present deformation degree in granitic and metamorphic rocks it seems to be responsible for the reaction textures developed.

Granitic rocks consist mainly of 3 minerals—alkaline feldspar, plagioclases, and quartz. During crystallization plagioclases are the first to be formed, contact between the crystals develops until a dense arrangement is formed leaving voids or pores. These places are then filled with alkaline feldspar and quartz, which explains why the latter has anhedral shapes (without well-developed crystal faces). This natural and intrinsic way of quartz crystallization in the aggregate, its potential strain level and recrystallization grade would be the main conditioning factors in the accessibility and degree of attack of the pore solution. This would then have a direct influence on the reaction dynamics and textures. When granitic rocks, like metamorphic rocks, undergo deformation and thermal changes, as in the case of those used in this study, quartz develops typical microstructures in response to the superimposed strain. Microstructures are

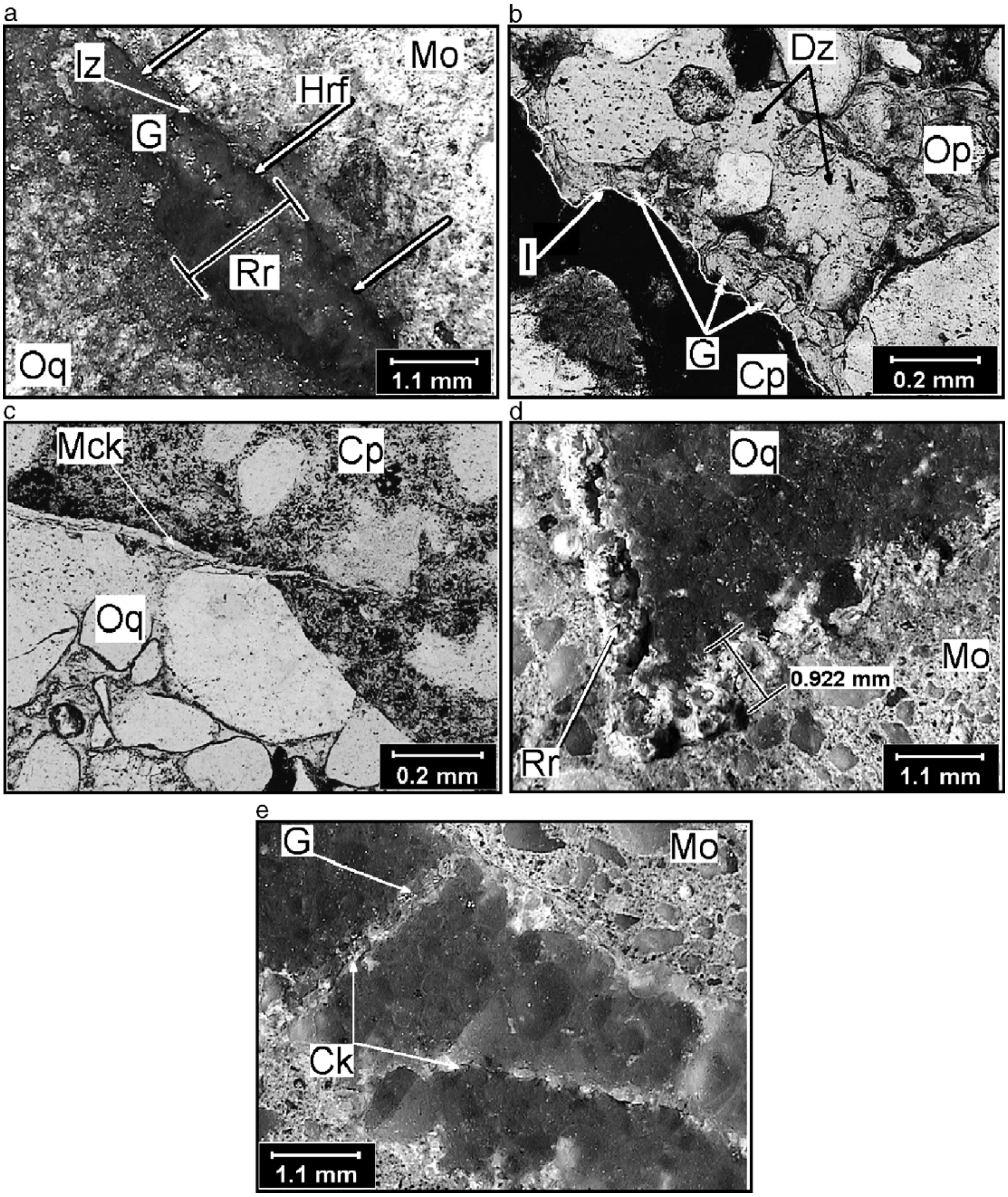

Fig. 4. (a) Reaction pattern. Polished section viewed with a stereobinocular microscope. (b) Dynamics and microstructural characteristics of the ASR. Thin section viewed through parallel polarizers. (c) Different pathways for gel migration. Thin section viewed through parallel polarizers. (d) "Dissolution texture" in orthoquartzite (Oq), thickness 0.922 mm. Polished section viewed with a stereobinocular microscope. (e) Attack toward the interior of the aggregate. Polished section viewed with a stereobinocular microscope.

the result of the triggering of certain mechanisms that tend to decrease the crystal free energy that increased in the deformation processes, Hobbs [21].

There are two basic mechanisms that tend to decrease the accumulated strain. The first one involves the "recovery" process that tends to shorten, group and cancel crystal defects, the resulting microstructures exhibiting deformation bands and subgrains. The second mechanism is recrystallization, which also tends to free crystals from deformation, grain boundary migration, subgrain rotation and nucleation being the mechanisms developed, Passchier and Trouw [22].

Each particular mechanism is more or less effective in displacing crystal dislocations and hence decreasing their free energy. All of them can act simultaneously, so quartz in a slow-reacting aggregate can usually be recovered at different levels depending on the prevailing fluids and temperature conditions. The alkaline solution could then attack strained quartz in different ways according to its susceptibility to react, which is directly related to its degree of deformation and/or recovery. Following this line of thought, in the same aggregate there would be sites more reactive than others.

## 6. Pore solution accessibility to reactive zones

The access of pore solution to reactive zones in slow reacting aggregate is restricted to two pathways. The first and the most immediate one is on the aggregate surface when the alkalis find crystals of quartz potentially metastable and susceptible to be attacked, Wigum [23]. Attack in localized areas rather than a homogeneous reactive front develops. The other likely pathway for pore solution access is a weak surface through which the pore solution can go into the rock mass and interact with reactive zones Jensen [24], Spry [25]. These surfaces involve microcracks and diaclases that in these aggregates could be of natural origin, for instance rapid tectonic decompression of rock massifs causing sets of cracks in the rock, or artificial when the aggregates are crushed. Foliation planes present in metamorphic and deformed granitic rocks also contribute to the access of pore solution, Fig. 3b. The development of natural cracks (Nck) in the granitic–granodioritc aggregate (Gr) of concrete 2 (C2) is shown in Fig. 5a.

## 7. Texture and reaction dynamics of slow-reacting aggregates

The textures observed and analyzed in these aggregates indicate differences in the reaction mechanism. The second concrete sample analyzed (C2) contains granitic–granodioritic

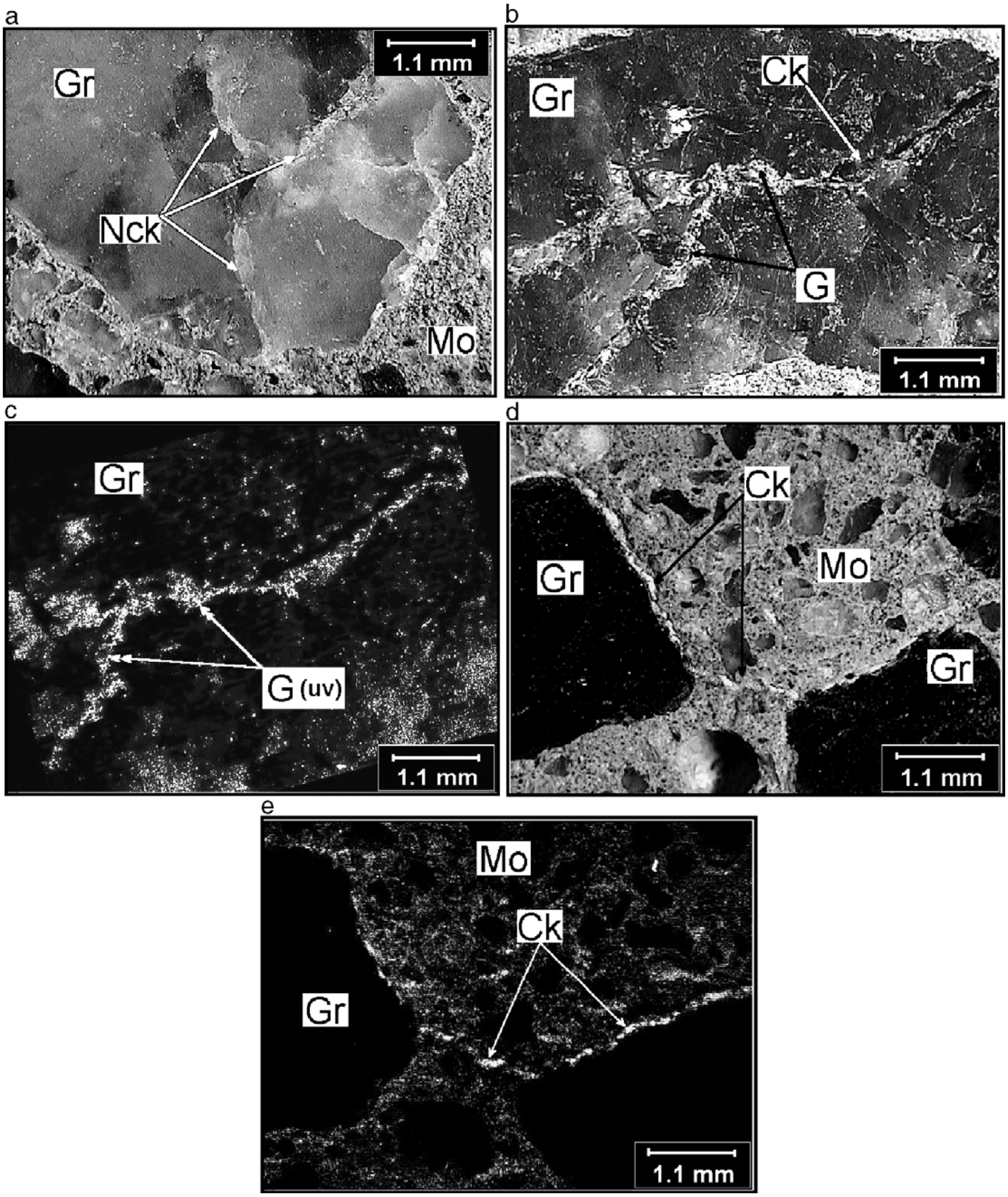

Fig. 5. (a) Granitoid cracking (Gr), (Mo) mortar. Polished section viewed with a stereobinocular microscope. (b) Alkali access and reaction through a crack. Polished section viewed with a stereobinocular microscope. (c) Gel fluorescence. Polished section viewed with a stereobinocular microscope, (uv). (d) Gel migration through the interface and mortar (Mo). Polished section viewed with a stereobinocular microscope. (e) Gel manifestations by ultraviolet fluorescence. Polished section viewed with a stereobinocular microscope, (uv).

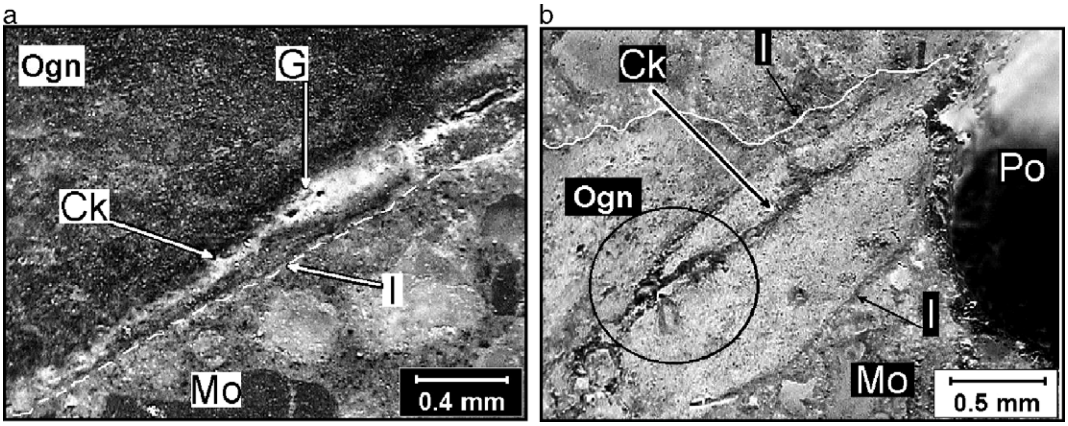

Fig. 6. (a) Aggregate cracking that runs parallel to the interface (I), Mortar (Mo). Polished section viewed with a stereobinocular microscope. (b) Effect of dissolution inside orthogneiss. Polished section viewed with a stereobinocular microscope and inverted light.

granitoid (Gr). Alkali attack occurs through a crack (Ck), which acts as a pathway. The access of the pore solution causes reaction at the reactive zones; the crack lips are attacked and eroded at the sites that are susceptible to react, as evidenced by the crack widening and the presence of gel (G) inside it, Fig. 5b. Gel fluorescence G(uv) inside the crack, when it is stained with uranyl acetate and then exposed to ultraviolet radiation, is shown in Fig. 5c.

On forming, the gels create pressure and migrate from reaction sites and penetrate the cracks (Ck), pores, and even the interface and the cement paste, as shown in Fig. 5d and e. In the latter, uranyl acetate has been applied to show the gels that under ultraviolet light exhibit a typical fluorescence.

The third concrete (C3) contains granitic–granodioritic orthogneiss (Ogn) as the coarse aggregate. This rock has a texture similar to that of the above sample; there are specific zones of attack and the reaction occurs mainly through internal cracks (Ck) in the aggregate. The cracks have been eroded, with dissolution zones on their sides and gel formation (G), as shown in Fig. 6a and b. The latter is viewed with inverted light (special photography generated by an image analyzer) to show cracking. The encircled zone shows the effects of dissolution on the crack lips, possibly with gel migration to the pore (Po).

The fourth and last concrete studied (C4) is a sample from a dam from the province of Tucumán, northwest of Argentina. One of the reactive aggregates contained in this concrete is a quartz–mica schist, whose petrologic description was given above. Here again strained quartz microstructure and foliation planes have a key role. Fig. 7a shows that schist (Qms) has developed incipient foliation that runs from the bottom left-hand corner to the upper right-hand corner. Consequently, a microcrack (Mck) filled with gel (G) runs almost parallel to the foliation planes. The gel protrudes like a plug toward the cement paste and continues through it. Pore solution access to the reactive zones (recrystallized strained quartz) probably occurred through the foliation planes of the metamorphic rock. The gel subsequently expanded and migrated through cracks developed through parallel weak planes. Fig. 7b depicts the reaction pattern of quartz-mica schists (Qms) showing internal microcracks (Mck) in the aggregate that prograde toward the cement paste (Cp). The interfaces, however, do not show any deleterious signs, their sides are clean and homogeneous, so the weak planes in the aggregate are of outmost importance for alkali access to the reaction zones. The deleterious reaction manifests itself mainly inside the aggregate with low or specific reaction at the interfaces.

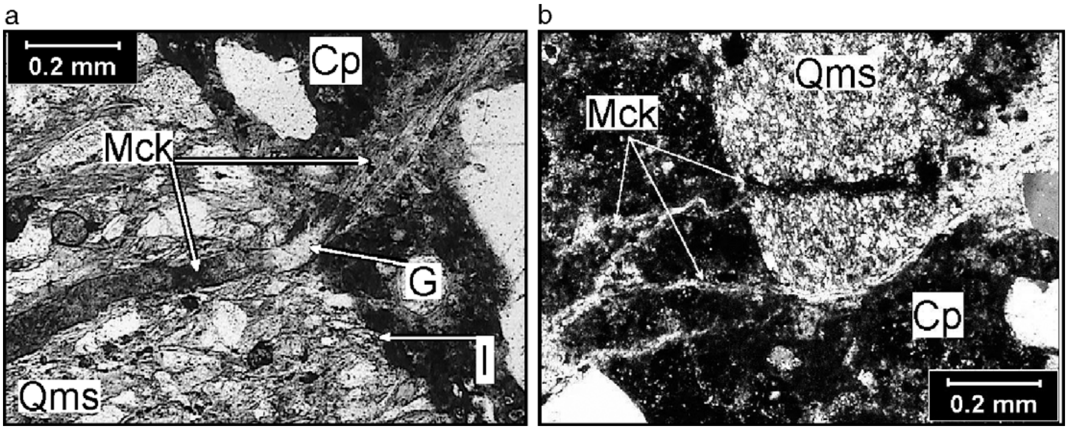

Fig. 7. (a) Quartz-mica schist (Qms).Thin section viewed through parallel polarizers. (b) Textural reaction pattern in quartz-mica schist. Thin section viewed through crossed polarizers.

## 8. Concluding remarks

### 8.1. Textural pattern of the concrete containing rapid-reacting aggregates

- The first concrete sample analyzed (C1) contains Chaco orthoquartzite. In this aggregate the reaction starts by the interaction of the pore solution and the siliceous cement, which is composed of highly reactive minerals (opal and chalcedony) and binds the rock quartz grains together. This homogeneous reactive front causes the dissolution, reaction, displacement and loss of the binding material leaving important voids or weak zones between the aggregate and the cement paste. This is what the authors have called "dissolution texture." Microcracking inside the aggregate as a result of gel expansion generally goes from reactive sites to the cement paste, or continues along the interfaces (strongly attacked by the reaction) and deviates to the interior of the mortar.
- The reactive rock aggregates of sedimentary origin, with clastic textures, where cement is composed of strongly reactive minerals, will form "dissolution textures" at the interfaces with the concrete cement paste. This causes a decrease in mechanical stability and subsequent decrease in concrete strength. Furthermore, there is a significant increase in permeability in that zone, which facilitates the circulation of aggressive fluids increasing the risk of the concrete being affected by other pathologies.

### 8.2. Textural pattern of the concretes containing slow-reacting aggregates

- The alkali reactivity of slow-reacting aggregates is directly related to the relative thermodynamic stability of strained quartz, which is one of the aggregate constituents. Defects on the crystalline structure lead to an increase in the free energy of crystalline networks, which turns silica metastable and susceptible to be attacked by concrete alkali pore solution.
- The slow-reacting aggregates contained in the other three concrete samples (C2, C3, C4) show a similar deleterious reaction pattern.
- The deleterious reaction of these aggregates in concrete is heterogeneous as the reactive strained quartz is only present at some sites, on the surface and/or inside the aggregate, which is linked to the intrinsic arrangement in the rock as a result of its origin. Other factors that contribute to the access of pore solution ions to the interior of the rock are cracks and foliation planes in the rock mass.
- Concrete samples (C2) and (C3) showed that their main reaction texture is related to internal cracks in the aggregate. These cracks have eroded and widened lips, and are filled with gel. The textural relationship of the aggregate and the paste is quite different from that of (C1). In this case, interfaces with gel occurrence, which comes mainly from inside the aggregate, no reaction rims or unbound areas, and discrete penetration of microcracks into the cement paste transporting the reaction products to the adjacent pores can be observed.
- The analysis of the thin sections of (C4) reveals the importance of the internal weak surfaces of aggregates. In this case there are foliation planes that not only act as pathways for the pore solution but also as weak planes through which the internal stress caused by gel expansion can dissipate. Hence the planes widen and form microcracks that will then take the reaction material out of the aggregate to penetrate even the cement paste.
- Based on the above statements it follows that to study and evaluate the reactivity of slow-reacting aggregates the following should be assessed:
  - *Thermodynamic stability of silica*: this involves an evaluation of the strained and/or deformation state of quartz in the aggregate.
  - *Pathways for access of the pore solution to the aggregate*: this is directly related to the rock fabric and structure, including cracks, diaclases, foliations, lineations, alteration state, friability, etc.

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