ALKALI-SILICA REACTIVITY OF SOME COMMON ROCK TYPES A GLOBAL PETROGRAPHIC ATLAS

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Abstract

The correct identification of potentially alkali-silica reactive aggregates is important for the prevention of ASR in concrete. Though a number of standards for assessment of concrete aggregate by petrography are available, distinction of potentially deleterious from innocuous rock types can be problematic. The application of geological nomenclature alone is insufficient as the geological history and hence mineralogical texture of a given rock type may strongly influence its performance in concrete. As one of the goals of RILEM TC 219-ACS, a worldwide photo atlas is being developed in order to be a guide for petrographers in the identification of the mineral composition and textures which are characteristic of alkali-reactive rocks. The atlas is based on micrographs of rock types recognized as potentially deleterious by field performance and/or laboratory expansion testing. It is aimed to serve as an independent reference work and intends to unify rock terminology in order to improve the petrographic characterization of aggregates.

Keywords: alkali-silica reaction, deleterious forms of silica, petrographic methods

1 INTRODUCTION

Since alkali-silica reaction (ASR) in concrete was identified for the first time in 1940s, a huge number of papers and books have been published in order to try to understand the phenomena involved. However, the discussion still standing around this theme reflects the fact that the identification of potential reactivity of an aggregate seems to be a great challenge far from being reached. The correct identification of potentially alkali-silica reactive rock types is important for the prevention of ASR in new concrete. The deficient knowledge of aggregates is responsible for part of the large amount of money spent every year on rehabilitation of damaged structures [1]. The methods that have been developed over the years for the identification of aggregate constituents and characterization of the alkali-reactivity potential are commonly cascaded in two subsequent stages:

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- Firstly, visual/optical identification and quantification by petrographic assessment of both raw unprepared material and thin-sections;
- Secondly, assessing the performance of the aggregates under accelerated laboratory conditions by testing expansion potential.

Although a number of standards for assessment of concrete aggregate by petrography are available (e.g. [2-7]), the distinction of potentially deleterious from innocuous rock types can be problematic and it is strongly dependent on the personal experience of the petrographer in the region in question.

The application of geological nomenclature alone has shown to be insufficient as the geological history and development of a given rock type, or a particular mineralogical, textural/microstructural aspect may strongly influence its performance in concrete [5]. Figure 1 shows an example of a metamorphic rock that can be named in very different ways. The rock contains epidote/clino-zoisite, titanite and chlorite, next to quartz and plagioclase, and could be called a calc-silicate rock based on mineral content alone. Texturewise, the rock contains both ductile and fragile deformed parts and could hence be designated as a cataclasite. However, field observations reveal that the rock should be correctly named as a deformed metavolcanic rock with chlorite, epidote and titanite from late-stage alteration and hydration. This illustrates the importance of field information for correct application of rock nomenclature, however, field information is not always available, eg. for unconsolidated sedimentary materials with unknown provenance. Neither 'calcsilicate' nor 'metavolcanic' are listed in [4], defining the limitations of this standard for rocks with uncommon names. The more elaborate AAR-1 [5], regarding potential reactivity of aggregates, circumnavigates the issue by avoiding rock nomenclature as a criterion determining alkali-reactivity, and instead focuses on the nature and qualities of the silica (Σ =quartz+chalcedony+opal+moganite) present, including but not limited to sutured or interlobate grain boundaries, grain size and subgraining, undulatory extinction, deformation lamellae and (micro-) porosity.

To answer the many doubts raised by the characterization of aggregates by petrographic methods, one of the goals of RILEM TC 219-ACS is revision of the original 2003-edition of AAR-1, to further improve its reliability and efficacy upon implementation of the procedure. Eventually, the guideline text will be supplemented with a photo atlas with worldwide coverage, showing thin-section micrographs of selected rock types with confirmed alkali-reactivity observed in field structures or expansion testing. Whereas a plethora of atlases (see listing in [8]) is available as *aide-de-camp* for the identification of specific groups of minerals or rocks, such is lacking for identification of alkali-reactive rock types in concrete aggregate, at least with worldwide coverage.

The atlas aims to serve as an independent reference work for anyone interested, and intends to help to unify rock nomenclature and terminology in order to improve the petrographic identification of alkalireactive rock types in aggregate for concrete. The atlas builds on the work carried out for European aggregates under the PARTNER project [9; http://vefur.honnun.is/farin/petroatlas/webindex.htm] and the previous RILEM committee dealing with ASR. A preliminary version of the atlas presenting selected rock types is scheduled for printed publication in 2013. It is hoped that later versions will also comprise electronic access.

2 MATERIALS AND METHODS

2.1 General

The study is based on petrographic assessment of thin-sections from aggregates for concrete, from various countries around the world. Delegates attending the 13th ICAAR in Trondheim, 2008, or the 12th EMABM in Dortmund, 2009 [8], or the 13th EMABM in Ljubljana, 2011, have been invited to contribute to the atlas by supplying sample materials. At present, sample materials for the atlas have been contributed by

specialists in Argentina, Brazil, Canada, Germany, Iceland, Japan, Norway, Russia, Singapore, South Africa, Spain, Sweden, Turkey and USA. In addition, seven thin-sections from concrete prisms from the EU Partner Project were studied, namely those containing aggregates from Belgium, Denmark, Germany, Italy, Norway and UK.

2.2 Materials

Petrographic assessment was conducted on different types of sample materials, kindly shared by RILEM colleagues and/or other atlas contributors:

- hand specimen/rock fragments from quarries,
- grab samples from natural gravel deposits,
- concrete samples from field structures, or concrete prisms after expansion testing, and/or
- thin-sections of such materials.

To prepare thin-sections where not yet available, billets were cut and mounted with araldite on a standard sized 26×48 mm carrier glass, following procedures outlined in [10]. Excess material was cut off with a diamond blade, the specimen impregnated with epoxy resin to reinforce the sample material, seal off porosity and reduce preparation artefacts, then cured up to $\sim 70^{\circ}$ C until set. Thin-sections of 30μ m thickness were lapped by hand using silicon carbide powder, and finally polished with 0.25 μ m diamond grit. Some of the ready-made thin-sections were already impregnated with yellow-fluorescent dye.

2.3 Assessment and analysis

Mineralogical and textural characteristics of the samples were obtained by the analysis of thin-sections, which were studied using a Nikon Eclipse E400POL petrographic microscope in plane polarized light (PPL) and crossed polarized light (XPL), as applicable also in fluorescence (FL). Photomicrographs were acquired using a Zeiss MRc5 camera and Axiovision software.

3 RESULTS

3.1 General

The past three years have been spent on the development of the atlas project, starting with specification of what type of data should be included ideally, and what is practically feasible. After that, the main objective has been the acquisition of suitable sample materials with confirmed and documented deleterious alkali-reactivity. The most reliable way to identify alkali-reactive aggregate constituents is through concrete petrography of an ASR-damaged field structure. Exposure to real life conditions precludes any artefacts from laboratory expansion testing, while thin-section petrography enables assessment of coarse and fine aggregate fractions at the same time. However, concrete samples represent only a minority of the total number contributed. Alkali-reactive sample materials from different provenances that have been assessed until now (see Table 1) appear rather consistent regarding the type of rock and the character of the reactive silica. In the following, rock types are grouped in the traditional manner (ie. igneous, sedimentary, metamorphic), and those features thought to govern alkali-reactivity are interpreted in terms of rock nomenclature (eg. [11-13]; also see on nomenclature discussion in [8] and references therein). Some of the samples studied here have already been described in detail elsewhere in the published literature.

3.2 Volcanic and plutonic igneous rocks

The most common igneous rocks used as aggregates for concrete are volcanic basalts and andesites *s.l.* and rhyolites, as well as plutonic granites. Figure 2 presents typical examples of different textures. Deleterious alkali-reactivity has been observed in basalt, dacite, andesite and rhyolite in Japan, New Zealand and Iceland

[14-16] attributed to primary cristobalite and rhyolitic glass, and possibly secondary opal or chalcedony of secondary origin (alteration). Batic et al [17] reported that some structures in Argentina made with basalt showed signs of AAR and [18] performed expansion tests on basaltic rocks from Turkey and concluded that the glassy matrix was responsible for the observed reactivity, possibly by 'virtue' of partial devitrification. Due to the fine grained nature of the glassy matrix in volcanic rocks, the identification of reactive forms of silica may be challenging. After initial thin-section petrography, bulk whole-rock chemistry (XRF), electron microscopy (SEM, TEM) and X-ray diffraction (XRD) are often required to characterize free silica present. In summary, there seems to be consensus that the alkali-reactivity of volcanic igneous rocks is primarily attributable to undesired dissolution of the glassy matrix. Such consensus appears lacking for coarse-grained crystalline plutonic igneous rocks, which show great variation in performance, as much in field structures as in expansion testing. Some of the disagreement seems to be related to the gradual (structural, textural) transition of a granite without preferential orientation via a granite with flow texture into a gneiss with layering and banding, and upon increasing deformation into a mylonite. Even in the field, such a transition may be very inconspicuous especially if it happens over large distances, and where granite ends and gneiss begins is in most cases rather arbitrary. Additional factors complicating correct identification include lack of experience of the petrographer with local materials, undisclosed identity of the deposit and/or unknown geological position, the component being one constituent of a polymict particulate material, among others.

The key factor determining alkali-reactivity of igneous rocks seems to be the presence of microcrystalline quartz, say $\leq 60 \mu m$ (eg. [19]) and/or the presence of microcracks (eg. [20]). The designation of the rock depends much on local geological context and experience, as elaborated above. Finally, laboratory testing reveals substantial differences in expansion [21-25].

3.3 Sedimentary rocks

The most common consolidated clastic sedimentary rocks used as aggregate for concrete are greywacke, various types of sandstone/siltstone, argillite, and others. A number of papers have been published on sedimentary rocks in concrete causing damage (eg. [26,27]). Figure 3 shows selected examples of common sedimentary aggregate rocks in thin-section. Sandstone/siltstone, greywacke, argillite, claystone and related rocks are composed of predominantly siliceous detritus ('clasts') from pre-existing igneous, metamorphic, or sedimentary rocks, liberated by weathering and erosion, transported and deposited by sedimentary processes, and consolidated by subsequent compaction and cementation. Consequently, mineral content, particle size distribution, texture and structure are subject to tremendous variation, reflecting provenance as well as sedimentary processing and history. The mineral content of the neogenic cement or matrix may differ greatly from the sedimentary clasts [28] and may be composed of carbonate, clay minerals, zeolites or other minerals, or of neogenic quartz cement syntactically deposited on detrital quartz particles (eg. [29]). In fluorescence thin-section petrography, interstitial/intergranular pore space between the detrital grains is often observed open and connected, facilitating access for (concrete pore) fluid to the rock particles' interior. In endeavours correctly to reflect the vast possible variation in both detrital and neogenic mineral content, texture and structure, nomenclature of sedimentary rocks is very varied, and several systems have gradually found wide acceptance (eg. [12]).

The true nature of alkali-reactivity of carbonate rock material is still a matter of dispute (eg. [30]). Recently, 'cryptic quartz' (ie. undetectable in thin-section petrography) present in the carbonate matrix has been identified as responsible for deleterious reaction of certain limestones [31,32] (eg. Kingston limestone). Identification required liberation through selective dissolution in excess acid and XRD analysis of the insoluble residue. Some other limestone types appear to show reaction of dolomite with concrete pore fluid, though the expansive mechanism remains uncertain.

Flint and/or chert occur worldwide in certain natural deposits exploited for concrete aggregate, notably in north-western Europe. Flint and chert are composed of micro- to cryptocrystalline quartz [33], and porous varieties are considered particularly alkali-reactive, even at low modal contents. Their reactivity can be attributed to the fine-grained nature of the quartz and areas of defective lattice, as well as to the presence of silica varieties chalcedony, opal and quite possibly moganite [29,34,35], a little known silica polymorph apparently common in flint/chert worldwide [36].

3.4 Metamorphic rocks

Metamorphic rocks are formed from igneous, sedimentary, or metamorphic precursor rocks by submission to pressure, temperature, deformation, and fluids under geological conditions, a process that may be reiterated several times. Mineral content, bulk whole-rock composition, structure, texture and fabric may all undergo very substantial changes reflecting the sum of metamorphic conditions, but modern analytical techniques allow identification of both precursor and metamorphic path in most cases (see eg. [37]). Consequently, systematic nomenclature of metamorphic rocks reflecting precursor, mineral content and structure/texture is complicated (see e.g. [38]). Metamorphic rocks as aggregates with pronounced foliation/cleavage and fine grain size are mostly used in locations where access to alternative aggregate resources is impracticable or non-feasible. The alkali-reactivity potential of metamorphic rock types, such as phyllite, schist, mylonite and cataclasite have, however, been extensively investigated [34,35,39]). Figure 4 presents typical examples of phyllite and cataclasite rock textures.

The fine grain size of the quartz (ie. <60µm) in e.g. meta-greywacke, phyllite, hornfels presents a large surface area for alkali attack, whereas intense deformation of quartz in mylonite (under ductile conditions) or cataclasite (brittle conditions) introduces strain in the crystal lattice [28,40]. Recrystallization of deformed quartz reduces grain size by formation of subgrains, as well as it reduces lattice strain in the new grains. Thinsection petrography of metamorphic rocks thus requires detailed assessment not only of mineral content, but also (micro-) structure, texture and fabric with particular emphasis on quartz, notably grain size, presence of undulatory extinction, (low, high) angle grain boundaries and/or subgraining, deformation lamellae, as well as contacts with adjacent grains (curvate, serrate, interlobate) and the overall mineral fabric, among others.

4 DISCUSSION

The alkali-reactivity of volcanic rocks has been investigated widely in eg. Japan, China, Iceland, Australia, New Zealand, Turkey, Argentina and Brazil (eg. [14-18,41-46]. The presence of rhyolitic (=rich in Si, poor in Fe,Mg) glass in the matrix is generally accepted as an indicator for the alkali-reactivity potential of the rock. Katayama et al [43] imply that fresh basaltic rocks are essentially innocuous, but may become alkali-reactive upon alteration with possible deposition of opaline silica and devitrification of matrix glass. In contrast, the reactivity of coarse grained plutonic rocks like granite, diorite and related rock types is primarily attributed to (modal) content of (microcrystalline) quartz, grain size, and/or lattice strain (eg. [19,21,22,47,48]). In addition, microcracks in the aggregate particles provide easy access to quartz grains in their interior, as suggested in [20]. Thus, alkali-reactivity seems to correlate with the total available surface area of quartz, also including subgrains. Lattice strain and subgrain development (eg. from thermal stress upon cooling) in quartz are very common in metamorphic rocks too (from thermal stress as well, or from tectonic deformation *sensu stricto*), and a strict distinction between plutonic and metamorphic rocks is in this respect hardly useful. Partial recrystallization of quartz by thermal effects [37] may to some extent explain the decreased reactivity of recystallized (blasto-) mylonite, but additional factors prevent a simple 1:1 correlation as certain non-recrystallized mylonites exhibit negligible expansion (eg. Table 1 in [35]). Wenk et al [48]

demonstrated a positive correlation between ASR expansion and dislocation density in the quartz lattice, which seems to support field experience.

The reactivity of sedimentary rocks, whether siliceous or calcareous, is often attributed to the presence of opaline silica, or more commonly various forms of microcrystalline or cryptocrystalline quartz in chert/flint and siliceous limestones, or as chalcedony and possibly moganite. Internal porosity seems a crucial factor, and can be identified using fluorescence-impregnation petrography and scanning electron microcopy.

Aggregate particles that are identified as alkali-reactive in mortar bars or concrete prisms from laboratory expansion testing have to be regarded with certain skepticism. They have been exposed to elevated temperature, moisture, and boosted alkali content, to provoke expansion in a mere 14 days instead of several decades in a typical field structure. The aggressive elevated conditions may lead to false positives, the time-limited exposure to a false negative [49].

5 CONCLUSIONS

The present work summarizes the attempt that is being developed aiming to contribute to an easier identification of reactivity of aggregates by petrographic methods. Although based on the most well-known classifications of rock types and on the international nomenclature, the petrographic atlas under development is focused on representative views obtained under the petrographic microscope in order to highlight and explain the potentially alkali-reactive of aggregates based on the analysis and description of the different forms of silica. The application of geological nomenclature alone has shown to be insufficient as the geological history and development of a given rock type, or a particular mineralogical, textural/microstructural aspect may strongly influence its performance in concrete

Different types of rocks, implying different mineral compositions, microstructural textures and provenance, exhibit reactive components which are characteristic of each type/group of aggregates. Therefore, the features that are more common for each rock type are explained: strain features for metamorphic rocks such as mylonites and schists; microcrystalline quartz in igneous rocks such as granite and granodiorite; volcanic glass, tridymite and cristobalite in volcanic rocks; chalcedony, opal and cryptocrystalline quartz in sedimentary rocks, including siliceous limestones.

Various methods are presented in the literature related to the classification of aggregates regarding reactivity. The objective of the petrographic atlas is to help in the identification of aggregates, assuming that petrography should be the first method to be applied and that it is the one with better ratio benefit/time. However, much work remains to be done in order to minimize the dependence on the experience of each petrographer. The regional/local experience is of upmost importance and should be taken into account when giving a designation according to traditional nomenclature.

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TABLE 1: List of the main types of aggregates received so far.		
Igneous	Sedimentary	Metamorphic
basalt	flint/chert	phyllite
volcanic glass	quartz sandstone	schists
andesite	greywacke	cataclasite
rhyolite	siliceous limestone	mylonite
granodiorite	siltstones and argillites	gneiss
granite		meta-arenite
trachyte	sand and gravel (with diverse rock	pseudotachylite
microdiorite	fragments)	



FIGURE 1: Example of a rock for which different designations can be attributed: calc-silicate rock, regarding the mineralogical composition; cataclasite, regarding the texture (PPL and XPL).



FIGURE 2: Igneous rocks: a) rhyolite showing vitreous texture (PPL); b) andesite with volcanic glass in the matrix (PPL); c) microcrystalline quartz in granite (XPL).



FIGURE 3: Sedimentary aggregates: a) greywacke with microcrystalline quartz; b) sandstone with silicate minerals and microcrystalline quartz in the cement; c) siliceous limestone (XPL).



FIGURE 4: Metamorphic aggregates: a) cataclasite showing strained quartz; b) phyllite with microcrystalline quartz; c) quartzite showing dynamic re-crystallization and incipient polygonization of quartz crystals (XPL).