PETROGRAPHIC ASPECTS

ON SILICEOUS ALKALI REACTIVE AGGREGATES

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ABSTRACT

Problems in identifying siliceous alkali reactive
aggregates were discussed. Two methods, the general method
for Determination of Quartz Instability and the method of
the Undulatory Extinction Angle (in stressed quartz) were
applied on four aggregates. Some agreement has been shown
between the results by the Undulatory Extinction Angle me-
thod and expansion of two rocks in concrete resp. in sodium
hydroxide solution (Rock Cylinder method).

Petrographic identification of reactive aggregates is based on detailed knowledge of both the composition and texture of the rocks. Despite the fact that alkali reactivity is a chemical process physical characteristics based on texture are a significant factor. An understanding of the reaction phenomena is the basis for his work.

Alkali reactive aggregates are usually varieties of certain groups or classes of rocks which have similar but not identical properties. Not all of the rocks in a given group or class are alkali reactive and certainly not to the same degree. Acceptance of general descriptions of alkali reactive rocks, found in the literature, may lead to unnecessary and expensive testing of many rocks which otherwise could be identified petrographically as innocuous. Precise detailed descriptions of reactive varieties are needed to permit the exclusion of nonreactive aggregates which all too often are described as "potentially reactive". This term should be reserved for those varieties which show strong petrographic indications of reactivity and are distinctly similar to those varieties which have been shown to be reactive.

This paper deals with alkali reactive siliceous rocks and problems in identifying reactive aggregates which face the petrographer dealing with a large number of aggregates for which long term test data are not yet available. The second half deals with two methods of identifying unstable quartz which, it is assumed, can cause quartzbearing rocks to be reactive.

Alkali Reactivity of Metastable Silica Minerals

The classical subgroup, containing metastable silica minerals and rocks with opal, tridymite and cristobalite as well as the acid volcanics, is relatively easy to identify by petrographic methods. The acid volcanic glasses consist predominantly of silica, e.g. rhyolitic obsidians contain 72.4 or 73.6 percent of silica (Rittmann 1962 p.221). In the intermediate volcanic rocks such as andesites, the silica is concentrated in the glassy groundmass after the formation of intermediate feldspar and mafic phenocrysts. It is possible to look at the volcanic glasses as silica containing smaller amounts of other elements as "impurities". The alkali hydroxide probably attacks the silica selectively, in the same way it is assumed to attack cryptocrystalline (<0.04 mm) and aphanocrystalline (<0.004 mm) devitrified glasses. This can be determined using the scanning microscope as shown by

Gillott on rocks from Alert, N.W.T. (Gillott and Swenson 1973). It would be useful to know whether the crystallites in the volcanic glasses influence the degree of reactivity of such rocks and consequently the expansion of concrete. The presence of crystallites can be established with a polarizing microscope.

It is well known that small percentages of reactive rocks can cause severe deterioration of concrete. E.g., only two percent rhyolites and andesites included in the concrete of the Parker dam caused deterioration after $2\frac{1}{2}$ years (Meissner 1941). It is not easy to detect such small amounts of reactive rocks when making a relatively quick preliminary petrographic analysis of an aggregate. Opal-bearing rocks such as opaline shales and sandstones may be identified petrographically using other characteristic features of these varieties. This is usually done when previous information is available which indicates that special opal-bearing rocks might be present in the aggregate deposit.

Rocks Containing Alkali Reactive Quartz

Particular attention must be given to rocks which contain alkali reactive quartz which is the most stable modification of silica.

Crystallized cryptocrystalline volcanics and chert may be considered as gradational rocks between the silica mineral reactive subgroup and the reactive rock subgroup containing coarser grained reactive quartz. Varieties of chert may consist of opal, chalcedony or cryptocrystalline quartz, or combinations of any of these three (ASTM Designation C294). From the physico-chemical point of view it is understandable that chalcedony and even cryptocrystalline quartz are reactive, their grains having a large surface area. The alkali sensitivity of chalcendony promoting expansion is attributed by Diamond (1974) to its microporosity in the form of interconnected pore systems similar to that in opal. This explanation based for chalcedony on works of Kneller (1968) and others introduces a strong textural factor which is generally applicable for minerals and rocks. On the other hand, according to Gillott the dense interlocking microcrystalline texture of chert from Alert in the N.W. Territories, is no doubt in a large measure responsible for the extreme slowness of the alkali aggregate

reaction (Gillott and Swenson 1973).

Cherts are silicate rocks which frequently contian impurities
such as carbonates, clay minerals, iron oxihydrates (limonite) and pyrite.
An intricate relationship must exist between the types, amounts and
distribution of these impurities and the various silica minerals which in-
fluences the degree of reactivity of chert. Chert consisting of fine,
medium and coarse grained quartz is generally considered not to be reactive.
But alkali reaction occurs in such rocks to a minor degree. Paleozoic
weathered chert consisting of cryptocrystalline (< 0.04mm) and microcrystalline
(<0.2mm) quartz and some chalcedony, conisdered not to be reactive, has
been shown to produce gel in mortar bars after more than 2 years storage in
the laboratory under 35% RH (Dolar-Mantuani 1972a).

Alkali Reactive Silicate Rocks

A series of sedimentary and metamorphic silicate rocks have been
described as alkali reactive. "Some" argillites, siltstones, greywackes,
subgreywackes, arkoses, quartzites, slates, phyllites and quartzmica schists
have been found to be alkali reactive (Swenson 1957, 1972; Buck and K. Mather
1969; Dolar-Mantuani 1969, 1972; Duncan and Swenson 1969; Duncan, Gillott
and Swenson 1973; Gillott, Duncan and Swenson 1973; Idorn 1967; and others).
The series has increased recently and now includes granites, and charnokites
(hypersthene granites) (Gogte 1973) and granite gneisses (K. Mather 1973).

A common feature of all these rocks is that each contains a
relatively large percentage of quartz, but they vary greatly in grain size.
The reactivity of argillites, siltstones, slates and phyllites, rocks which
contain cryto- to microcrystalline or fine grained quartz or any variation
in the range of such grain sizes is explainable mineralogically and texturally
but it becomes more and more difficult to explain the reactivity of the
sandstone group which may contain medium to coarse grained quartz. In the
last group of quartz-rich rocks, some granites and granite gneisses were
proven to be alkali reactive when they developed strong cracking accompanied
by silica gel (K. Mather 1973).

With the exception of some quartzites which may be almost pure

silica, all of the silicate rocks listed contain various other minerals in addition to quartz. These may be considered as dilutants which diminish or delay the reactivity of the rocks but in addition to quartz the presence of clay minerals may be essential for the disintegration of concrete (Gillott 1970, and Gillott et al 1973). Great emphasis is given by Gillott to the presence of micaceous minerals, particularly vermiculite, in greywackes, argillites, phyllites and schists from Nova Scotia. Petrographic examinations of concrete containing similar varieties of the same rock types without the vermiculite porphyroblasts, from the Canadian Shield showed expansion and cracking accompanied by the production of distinct amounts of silica gel (Dolar-Mantuani 1971). The rocks were selected for examination of alkali reactivity becuase gel was found in cracks and along aggregate particles in concrete made with these rocks. Some quartzites were almost lacking in micas. It is not clear whether these rocks from the Canadian Shield exhibit a different type of alkali reactivity than those in Nova Scotia or if some phyllosilicate minerals are alkali reactive in addition to silica minerals. Cursory examinations of distressed concrete from a few structures in the Maritimes showed some gel at aggregate particles in which the characteristic vermiculite porphyroblasts found in some Nova Scotia beach deposits were not evident. Gillott also describes highly expansive greywackes from Alert which are petrographically similar to the alkali-expansive greywackes from the Appalachians in Nova Scotia but do not contain expansive vermiculite porphyroblasts.

While it is generally accepted that minerals occurring in minute grains are vulnerable to chemical attack by alkalies, the reactivity with portland cement of rocks containing medium or coarse grained quartz requires an explanation. It is postulated that this reactivity is due to defects in the quartz lattice sporadically suggested by several authors. The presence of strained quartz was used by Brown (1955) and Mielenz (1958) in the interpretation of alkali reactions with metamorphic quartzites.

Causes for Defective Crystal Lattice in Quartz

A defective crystal lattice of quartz may be caused by surficial indentations or chipping and pitting due to leaching (Carozzi 1960) or it occurs during replacement of other minerals, especially those with abundant secondary inclusions, such as feldspars. The impurities may be concentrated

at the surface of the quartz grains or may occur in rims close to the surface during secondary growth of this mineral (Carozzi). Such imperfections at the grain surfaces may decrease the resistivity of quartz to pressure effects and to chemical attack by alkalies.

The main physical factor which causes the formation of imperfections in the crystal lattice is the pressure developed especially during the exposure of the rock to metamorphic phenomena. The response of quartz to pressure has been repeatedly emphasized by metamorphic petrologists (e. g. Fairbairn 1953). Although Smaller (1974) states that crack patterns in granitic quartz are probably developed to homogenize strain in the original quartz crystals due to stress field changes, especially those due to the high-low transformation of quartz, Moss (1966, quoted in Moss et al 1974) considered that the major cause of stressing was rock deformation under temperature and pressure conditions.

The effects of pressure on quartz are evident in thin sections. S. Young (1975) who used differences of quartz crystal responses to stress to determine the source rock of quartz sands, lists a series of increased pressure effects on quartz. Undulatory, wavy extinction to various degrees, from slightly recognizable to distinctly wavy; the presence of deformation lamellae (0.002 mm in thickness) and deformation bands; polygonization of grains in which smaller sectors of a grain are differently oriented optically but recognizable with the analyser only; formation of elongated grains usually showing wavy extinction; suturing of quartz to quartz boundaries due to migration of dislocation; and finally cracking and fragmentation of larger grains. The wavy extinction was measured by de Hills and Corvalán (1964) using the exfinction angles in the same grain. The method of undulatory extinction angles, fracturing and granulation of quartz was used by Gogte (1973) to correlate the number of highly distressed quartz grains in granite, granodiorites, gneisses, charnockites, quartzites and schists with the expansion of mortar bars containing these rocks as aggregates. He reports that rocks containing 40 percent or more of such unstable quartz varieties were highly reactive, those with 30 to 35 percent of strained quartz were moderately reactive and rocks containing predominantly unstrained or recrystallized quartz showed negligible expansion. Stable varieties of quartz are characterized by non-undulose extinction, polgonal forms under 120 degrees and smooth crystal boundaries.

Examinations of Quartz Instability

In an attempt to assess the degree of instability of quartz as a means for recognizing quartz-bearing alkali reactive rocks, two methods were used:

(1) Determination of several properties characterizing quartz.

(2) Determination of the undulatory extinction angle in quartz.

General Method for Determination of Quartz Instability

In the general assessment of quartz instability the following properties were used:

(1) Grain size and shape

(2) Extinction (uniform to strongly undulose, estimated).

(3) Segmented undulosity, (formation of optical subgrains with somewhat different optical orientation).

(4) Type of quartz to quartz boundaries in polycrystalline quartz (straight or sutured to various degrees).

(5) Special features such as inclusions and cracks.

Specimens - The examinations of quartz were made on concrete specimens prepared from a crushed granodiorite rock and a crushed gravel, both from James Bay. The granodiorite contained about 20 percent quartz and the gravel consisted of 50 percent igneous, mostly acid, rocks and 38 percent gneisses. Each aggregate was tested in combination with both low (0.5%) and high (1.0% Na_2O equivalent) alkali cements and stored at 100^OF (38^OC) and approximately 100 percent RH. Quartz grains were examined using the point count method in thin sections of:

(a) crushed granodiorite in concretes with both high and low alkali cements

(b) polylithic aggregate made of crushed gravel in mortars with both high and low alkali cements.

Results and Discussion - 128 quartz grains were examined in the four thin sections. The results showed that differences among quartz grains were small in any of the properties in the four thin sections.

The analysis of the extinction types seemed to provide the most useful results. The following three categories were distinguished: uniform, wavy, and distinctly wavy extinction. Combining the results of all grains from the four thin sections, the percentages of quartz grains in each category were:

TABLE 1

(a) Uniform extinction	73%
(b) Wavy extinction	24%
(c) Distinctly wavy extinction	3%

Considering extinction types, polygonization and suturing of quartz to quartz outlines in particles consisting of either one or more quartz grains or part of a rock fragment in the concrete, the following results were obtained:

(1) The quartz grains were mostly of the stable type and only single grains had a distinctly wavy extinction

(2) The wavy extinction of monocrystal particles were more frequent in the aggregate crushed from the granodiorite but polycrystalline quartz particles were more frequent in the crushed gravel.

(3) The distinctly wavy quartz was of the polycrystalline variety.

(4) Quartz with sutured outlines was more frequent in crushed gravel than in granodiorite aggregate. This was in accordance with the fact that more than one third of the particles were gneisses.

Preliminary Evaluation of the Results - Whether the very low percentage of quartz grains with a distinctly wavy extinction or those with less distinct wavy extinction characterizing one quarter of the quartz grains examined are indicative of the degree of reactivity of both types of aggregates can not be decided for the time being. The four types of combinations of aggregates in mortars and concrete respectively, showed insignificant expansion but

the test specimens indicated some tendency of expansion after more than one year of testing as visible in Table 2.

TABLE 2

Days Tested	Expansion* in percent with	
	High Alkali Cement	Low Alkali Cement
Crushed granodiorite, James Bay, in concrete (CSA A23.2.24)		
520	0.017	0.021
600**	0.02	0.023
Crushed gravel, James Bay, in mortar bar (ASTM C227)		
300	0.022	0.028
600**	0.023	0.030

*Tests performed in the Laboratory of the Building Research Division of the Canadian National Research Council in Ottawa by Dr. P. E. Grattan-Bellow.

**Extrapolated extended line.

Evaluation of the Method (as performed) - When using this method for the identification of unstable quartz in concrete aggregate, it has to be realized that the results are partially influenced by the small aggregate particle sizes present in the thin sections of concrete. This affects the shape of the quartz grains. Small particle sizes may also conceal other features such as wavy extinction and polygonization and the formation of subgrains, all of which are observable in the quartz grains of larger particles.

Method of Undulatory Extinction Angle

Description of the Method and its Evaluation - According to De Hills and Corvalán the undulatory extinction angle is determined by selecting quartz grains with the highest birefringence colour and recording the angle of the first appearance of the extinction in the grain. The grain is then rotated to the position in which any extinction in the grain disappeared. This position is not easily determined and therefore it was repeated four times averaging the values for calculation of the undulatory extinction angle, which is the difference between these two readings. I found that a more

constant reading for the same grain is received by taking the position of the first and last extinction of a grain obtaining in this way the extinction range angle.

Specimens - The method was tried on two thin sections, one a feldspar-bearing quartzite and the other an orthoquartzite both from Sudbury, Ontario. The quartzite consists of poorly sorted fine grained quartz with slightly sutured outlines, and smaller sized feldspar grains composed of plagioclase and microcline. Fragments of a cryptocrystalline rock were very subordinate. Anhydral epidote grains occur in some interstices.

      The orthoquartzite consists of poorly sorted mostly rounded medium grained quartz, and some 20 percent of a matrix, two thirds of which is cryptocrystalline quartz, one fifth is microcrystalline quartz and the remainder is mica scales and flakes with some chlorite. The quartz sand grains are usually separated by mica scales.

Results and Discussion - The undulatory extinction angles and the extinction range angles were determined in 25 quartz grains of each rock. The results are shown in Table 3. The table contains also expansion data performed on concrete made with the crushed orthoquartzite and data for the rock cylinder of the feldspatic quartzite.

TABLE 3

Sudbury:	Orthoquartzite		Feldspatic Quartzite	
Angles in Degrees	Avg.	Range	Avg.	Range
Extinction Range Angle	7.9	2.5-18	6.3	2 - 14
Undulatory Extinction Angle	27.7	13.5-42.5	21.5	11.5 - 30

Expansion* Determined	In Concrete		As Rock Cylinder (ASTM C586)	
	Percent	Day	Percent	Day
	0.165	200	0.025	200
	0.190	400	0.037	450
	0.200	600	0.130	500

*Tests performed in the Laboratory of the Building Research Division of the N.R.C. in Ottawa by Dr. P. E. Grattan-Bellow.

The results show that an agreement is indicated between the extinction angles and the expansion values in that the rock with a lower value of the undulatory extinction angle (and of an extinction range angle) is also less expansive than the rock with higher values of both angles. Using the results by de Hills and Corvalán, the average value of the undulatory extinction angle of the quartz grains in the orthoquartzite indicates a rock which is moderately expansive and that in the feldspatic quartzite a slightly reactive rock.

When the general method for determination of quartz instability was applied to 246 grains in the orthoquartzite, the following results were obtained considering the degree of extinction and polygonization:

TABLE 4

Uniform extinction, no polygonization	65%
Wavy extinction and simple polygonization	30%
Distinct wavy extinction and polygonization	5%

The results indicated somewhat greater number of unstable quartz grains in the orthoquartzite in comparison with those in the first group, the granodiorite and gravel which were examined in thin sections of concrete and mortar respectively.

Summary and Conclusion

The results obtained using both the general method for Determination of Quartz Instability and that of the Undulatory Extinction Angle are only preliminary, and far more data must be obtained before any evaluation of precision and exactness of the methods can be attempted. There is no doubt that the method of the undulatory extinction angle which offers numerical results is more promising. Some agreement has been indicated between the average values of these angles in the quartz grains and the expansion of the rocks exposed to high alkali cement and to the 1 N sodium hydroxide solution, respectively. Whether one should use the angle of the first and last extinction in a grain, can only be decided when more data are available.

Also the usefulness of any of these methods is still not clear in spite of the promising results obtained by the method of the undulatory extinction angle. Although there is no doubt that these properties indicate an unstable quartz variety, they may not be sufficient or decisive for determining the chemical alkali reactivity of quartz and rocks containing it, an information which is badly needed not only in petrographic evaluation of alkali reactive aggregates but in the concrete technology in general. Further examination and studies on lattice imperfections of quartz is recommended.

The principle of determining the number and degree of defects in quartz grains might be satisfactory for determination of reactive quartz-bearing silicate rocks if a more precise instrument with automatic recording were used, for example the Leitz instrument used for textural determinations which permits determinations of defects in crystal lattices. Such methods would however not eliminate technological test methods but might decrease the amount of aggregate classified unnecessarily as "potentially reactive".

A final consideration still remaining unresolved is whether the rocks containing fine to coarse grained aggregate also contain crypto- or even aphanocrystalline quartz on surfaces of larger grains or in the interstices between them. If these smaller sized grains are present, it would be useful to know the amounts contained in alkali reactive rocks.

In conclusion, in spite of the different conditions to which alkali sensitive particles are exposed in concrete on a microscopic or submicroscopic scale, there must be specific properties which distinguish the alkali reactive from the non-reactive particles which seem for the time being identical. There could well be trace elements which trigger an alkali siliceous reaction.

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