

The mineralogical and petrological factors in Alkali Silica Reactions in concrete

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Abstract: Reactive silicas which can cause Alkali Silica Reactions (ASR) are opal, SiO₂ rich volcanic glass, chalcedony (or fibrous quartz) and tridymite. Opal is a hydrous cryptocrystalline or colloidal form of silica (SiO₂ · nH₂O) with a water content of around 6% to 10%. Silica-rich volcanic glass is a component of many young volcanic rocks. Chalcedony is a compact variety of silica composed of minute (crypto- or micro-crystalline) crystals of quartz (SiO₂) enclosing submicroscopic pores. Tridymite is a rare hexagonal high temperature polymorph of quartz with a S.G. of 2.28.

Opaline silica and silica-rich volcanic glass are regarded as the most reactive. Chalcedony, tridymite and some forms of crypto- and microcrystalline quartz occurring in acidic volcanics, chert and flint are regarded as of intermediate reactivity. Strained macrocrystalline quartz can cause ASR but only under certain conditions.

A factor which may influence the reactivity of particular aggregate is the proportion of reaction materials that is present in the aggregates. For a very reactive opaline silica the worst expansion may occur when it is found in the order of 2% to 5% (in the coarse aggregate), that is the pessimum. The pessimum for silica-rich volcanic glass is unknown. In the case of the strained macro-crystalline quartz, the pessimum being 100% that is quartzite containing wholly of strained quartz. The pessimum for the intermediate reactive silica is not known, but could well be higher than that of the opaline silica and definitely lower than that of strained quartz.

A multitude of rocks can cause ASR and there are many others which will not. Common rock types often used for aggregates can be grouped into: (i) Inherently reactive (ii) Potentially reactive if pessimum amounts of reactive silicas are present and (iii) Innocuous unless impregnated by secondary reactive silica minerals. Fine aggregates can also cause ASR to produce distress in concrete.

A sweeping ban on all volcanic aggregates for use in concrete may give a false sense of confidence for the control of ASR in Singapore. Banning of alkali rich cement/clinker is a better way of controlling ASR as well as other alkali-aggregate reactions.

INTRODUCTION

Alkali silica reactions (ASR) in concrete were first recognised in 1940 (Stanton, 1940a and 1940b; Rhoades and Mielenz, 1948) and because of its adverse effects on concretes have been discussed and studied by quite a number of persons (see Neville, 1976; Diamond, 1978; BRED, 1982). This reaction and its worse can cause cracks in concrete and disruption or failure of structures with disastrous results. Factors which control ASR are:

- i. Sufficient alkali solution in the pore structure of the concrete. The alkali that is available can come from external sources such as sea water and

introduced salts or from the cement or from the aggregates. Soluble alkalis from aggregates are generally insignificant while effects of sea water and introduced salts are secondary to alkalis in cement for causing ASR. Several authorities on concrete quote 0.6% Na₂O equivalent of alkalis in cement as the limit below which concrete are unlikely to suffer ASR even in combination with reactive aggregate. As the amount of cement used in concrete varies, therefore, the 0.6% Na₂O equivalent of alkalis cannot be used as an accurate limiting factor. Experiments performed in the United Kingdom (B.R.E.D., 1982) suggest a limit of 3 kg of Na₂O equivalent per cubic meter of concrete. On this basis if the amount of cement used in one cubic meter of concrete is 300 kg the tolerance should be 1.0% equivalent of Na₂O and if 500 kg, then it should be 0.6%.

- ii. Moisture is required for the concentration of the alkalis to cause ASR. Moisture is present in the concrete itself or often introduced as the result of immersion in water or due to condensation. Most of the authorities are of the opinion that ASR had only developed in structures which have been exposed to external sources of water. Alternate wetting and drying conditions allow the concentration of alkalis which favours the causing of ASR.
- iii. Presence of reactive silica minerals in the aggregates used in the concrete. This shall be the main topic to be discussed here.

SILICA MINERALS

Strong alkali solutions in concrete, which are introduced with the cement, can react with aggregates to form expansive gel. Two groups of minerals found in rock aggregates can cause such reactions. These two groups are carbonates (consisting of fine-grained dolomite and calcite with clay), and silicates (see Mindess *et al.*, 1981; Diamond, 1978; Neville, 1977; BRED, 1982; Rhoades and Mielenz, 1948). All silicate minerals react with strong alkalis to a higher or lesser degrees, though generally three sub-groups of these minerals can react strongly to produce distress in concretes. These are:

- (i) Silica minerals
- (ii) Zeolites
- (iii) Phyllosilicates (micas and clay)

Of interest in this paper are the silica minerals.

Silica minerals have a common formula of SiO₂ but they possess different crystal system that is they show polymorphism. Opal, always regarded as a silica mineral is an exception because it is an amorphous hydrate of silica. Besides opal, other known rock forming silica minerals are quartz, tridymite, cristobalite and coesite. Coesite, an extremely rare mineral is known to occur in fused rocks formed by meteorite impact. Cristobalite is another relatively rare mineral and has been known to occur in some volcanics only. Quartz, tridymite and cristobalite

show an α (or low temperature) and a β (high temperature) form each. Whereas, the high temperature polymorph of quartz has been known to occur in nature, the high temperature forms of tridymite and cristobalite are known only under experimental conditions (Deer *et al.*, 1967).

Some properties of the naturally occurring silica minerals are summarized in Table 1.

REACTIVE SILICAS

Not all known silica minerals are reactive. Silica minerals which give rise to ASR are opal, chalcedonic quartz and tridymite. Cristobalite and coesite which are quite rare minerals are tested to be reactive but however, in practice, they are rarely encountered in aggregates. Silica rich glass found in young volcanics are reactive (Rhoades and Mielenz, 1948; Allen, 1948; Ochard, 1973). BRED (1982) regards "cryptocrystalline" and "microcrystalline" quartz as reactive though Diamond (1978) is of the opinion that the strained varieties of these "two minerals" are likely to cause ASR only. Apparently this reference of "cryptocrystalline quartz" can probably be traced back to the investigations of Rhoades and Mielenz (1948); and Stanton (1940(a), 1940(b)) who state that "cryptocrystalline" rhyolite and dacite

Table 1. Reactive silica minerals.

Mineral	Formula	Crystal System	Variety	Abundance
α Quartz	SiO ₂	Trigonal	(i) <i>Normal Crystalline</i> (ii) <i>Fibrous</i> (or Chalcedonic) Flint Chert Jasper Agate Chalcedony	Very abundant Abundant in some sedimentary formations Not abundant but widely distributed in many rock types
β Quartz	SiO ₂	Hexagonal	Normal crystalline	Rare
Tridymite	SiO ₂	Orthorhombic	Granular crystalline and infilling	Quite rare
Cristobalite	SiO ₂	Tetragonal (Pseudo-cubic)	Infilling	Very rare
Coesite	SiO ₂	Tetragonal	Crystalline granular	Extremely rare
Opal	SiO ₂ nH ₂ O	Amorphous	Massive, infilling, encrusting	Rare

(Data in part based on Deer *et al.*, 1965; Dana, 1960; Kerr, 1957; Annibale *et al.*, 1983; Cripriani, 1986).

are known to be the part of the cause of ASR in several localities in the USA. These are actually devitrified glassy volcanics and the actual mineralogy of such "fine-grained groundmass" would apparently be a mixture of probably quartz/tridymite and feldspars rather than quartz alone. Pure or nearly pure cryptocrystalline or microcrystalline quartz can actually be classed under the chalcedony group.

Opal

This is regarded as the most reactive of all the silica minerals. Opal, $\text{SiO}_2 \cdot n\text{H}_2\text{O}$, is amorphous and has a varying water content of 6% to 8% (Deer *et al.*, 1967; Dana, 1965). The colour of opal ranges from white, colourless, milky, yellow, red, green, blue and black. In addition some opals show a pearly reflection or opalescence and these are referred to as precious opals.

Under the electron microscope, ordinary opal was found to consist of spheres of amorphous silica of various sizes (from 40 to 4,000 Å) which are randomly arranged. However, in precious opal, the amorphous silica spheres are regular in size and are arranged in a regular manner. It is this regular arrangement which allows diffraction of light to give rise to opalescence in precious opal (Daragh and Sanders, 1965; Cipriani and Borelli, 1986). The sizes of the spheres in the precious opal structures obviously control the colour of the opalescence. Faint cristobalite X-ray patterns in some opal have been interpreted as due to the presence of this mineral interstratified with opal.

Under the petrological microscope, opal is isotropic under crossed polars. Under plane polarized light, the diffraction of light may give rise to various shades of brown or that of the opalescent colour (see plate 3). Its low density (D ranging from 2.008 to 2.160) and low refractive index ($n = 1.441$ to 1.450) are also helpful in its identification. Its hardness on the Moh's scale is 5.5 to 6.5.

Opal is formed through several processes. Firstly opal can be deposited by volcanic hot springs or other hot aqueous solutions. Through this process, opal is found in geyserite or as infillings of vugs, veins, faults, joints etc. largely in volcanic rocks. Other rock types cut by deep seated faults are also prone to opal mineralization. Secondly, opal can be formed by supergene solution (see plate 1). These are percolating meteoric water which remobilize SiO_2 from one area and deposited it as opal in joints, cracks, voids etc. in rocks of all types. Under this condition opal can also be deposited as replacement of other minerals and wood (opalized wood). Thirdly opaline silica is deposited as the tests (protective shell or skeleton) of radiolaria, diatoms and other organisms. When these organisms die, the opaline tests can accumulate to form deposits of chert (see plate 4) and diatomaceous earth (Annabale *et al.*, 1983).

Chalcedonic quartz

Chalcedony is regarded as showing intermediate reactivity. Chalcedony is a compact variety of silica which is composed of minute crystals of quartz forming

a complex network which encloses a large number of micropores (Midgley, 1951 quoted in Deer *et al.*, 1967). Some varieties of chalcedony such as flint contain up to 1% of water. The density of chalcedony ($D = 2.60$) is slightly lower than that of pure quartz.

Several varieties of chalcedony can be recognised. Agate is the banded variety which displays different colours (plate 2 and 5) and is important as a semi-precious stones. Jasper consists of chalcedony admixed with iron oxides. Chert is composed of crypto to microcrystalline quartz derived from the lithification of beds composed of opaline tests of radiolaria, diatoms and other organisms. On lithification the opaline tests may be transformed into fibrous quartz (plate 4). Flint is composed of compact microcrystalline quartz often enclosing carbonaceous material and fine pyrites.

Chalcedonic quartz apparent occurs in a number of geologic environments. Firstly it is a low temperature hydrothermal (aqueous hot solutions) mineral which is deposited in veins, geodes and cavities of mainly volcanic rocks. Geysirites generally contain chalcedonic quartz admixed with opal. Tuffs of acid to intermediate volcanic rocks can contain fragments of chalcedonic quartz derived from geysirites etc. (plate 6). In rock types other than volcanics, chalcedonic quartz is present in veins, cracks, cavities, joints and fault planes when these rocks are permeated by silica rich hydrothermal solutions.

Jasper and flint originated as chemical sediments precipitated out from ancient seas. These two rock types can form beds of great thicknesses or are sometimes interbedded with other sedimentary rocks. Chert and flint are known to segregate out as nodules in limestone, the best example being the chalk of Dover, England (Selley, 1976).

In thin sections, chalcedony can be easily recognized by its fibrous habits. It often shows radiating textures (plates 5, and 6), fine bands (plate 5) and spherules (plate 8). Chert and flint show textures ranging from fibrous to cryptocrystalline to microcrystalline.

Tridymite

Tridymite is regarded as of intermediate reactivity. It occurs as small white blades, isolated crystals, spherules and rosettes largely in the cavities of volcanic rocks. Tridymite is often unstable and at lower temperatures, changes into β quartz.

Tridymite, from the type locality in New Zealand, is found to contain appreciable amounts of alkalis (Deer *et al.*, 1967).

In thin sections, this mineral is granular and often show twins of three wedges (trillings) from which the name of this mineral was derived (Mackenzie, 1980).

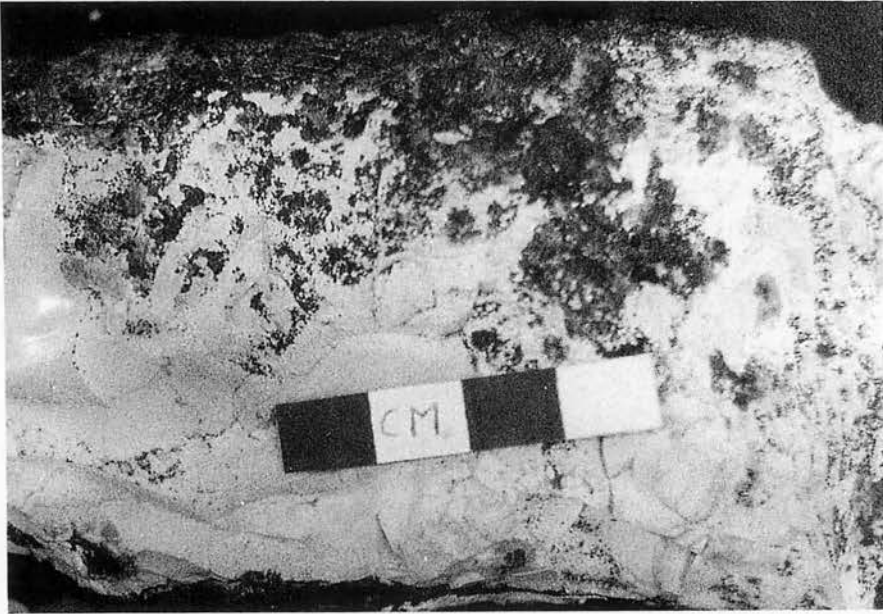


Plate 1: Specimen of milky opal from veins cutting granite. From Selangor, Malaysia.

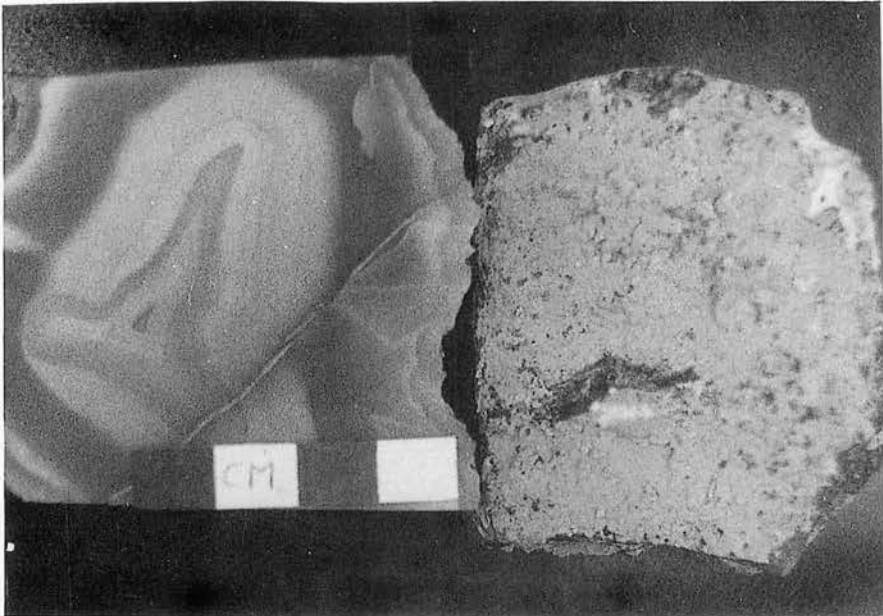


Plate 2: Handspecimens of (left) bluish banded agate in Andesite from Sabah, (right) chalcedony from veins in granite, Selangor.

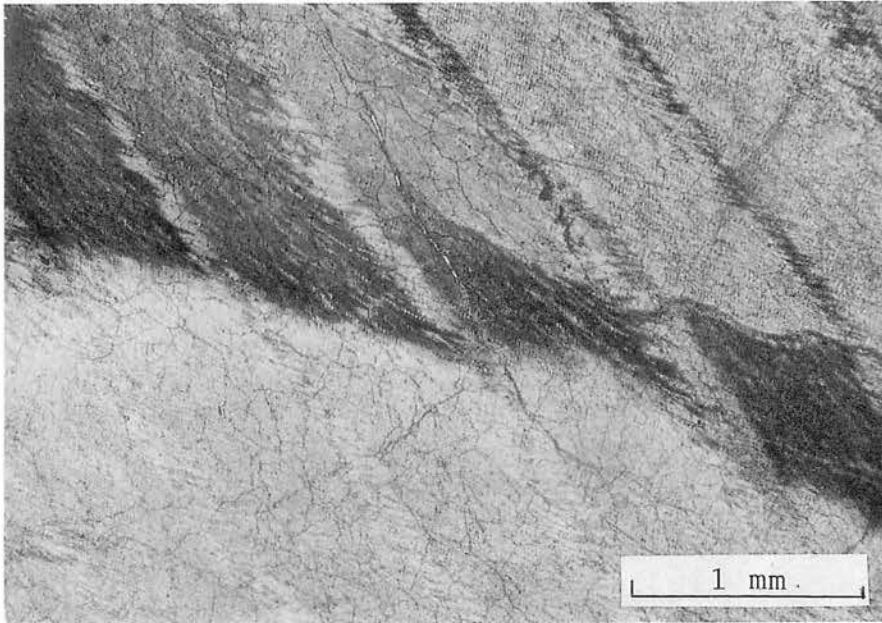


Plate 3: Photomicrograph of thin section of opal under plane polarized light. Dark tones due to light dispersion.

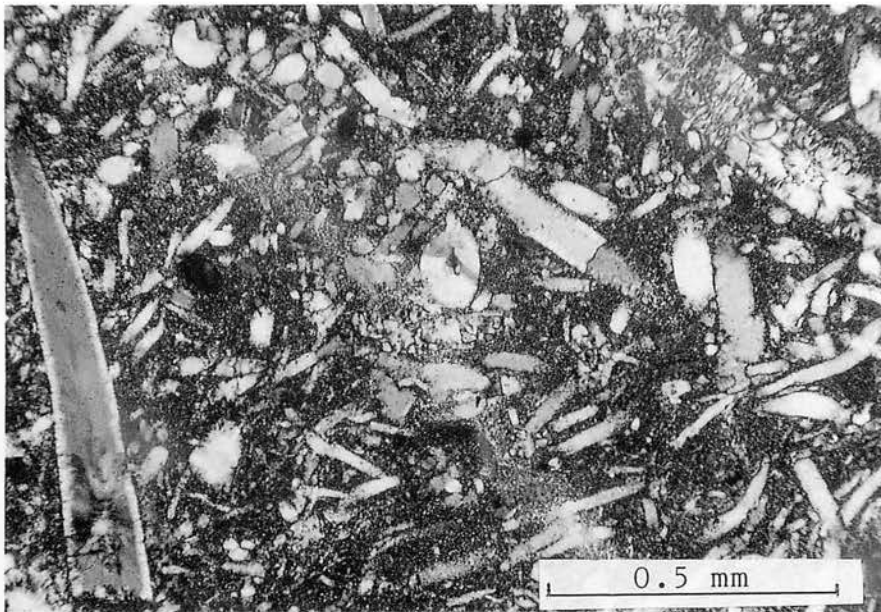


Plate 4: Photomicrograph of thin section of radiolarian chert from Japan. Fibrous quartz (tests) and micro to cryptocrystalline quartz groundmass are observed.

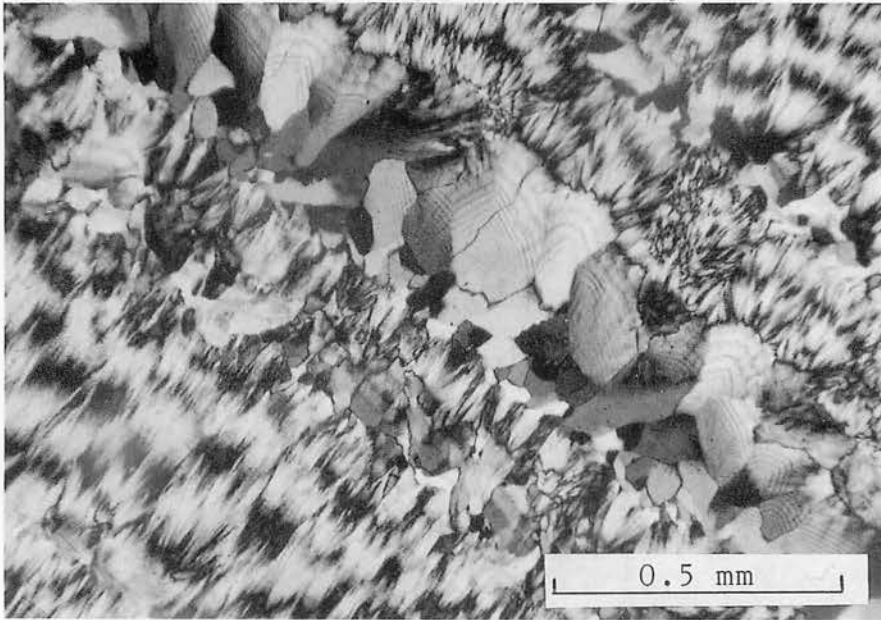


Plate 5: Photomicrograph of thin section of agate from Sabah. Fine and coarse bandings are clear.

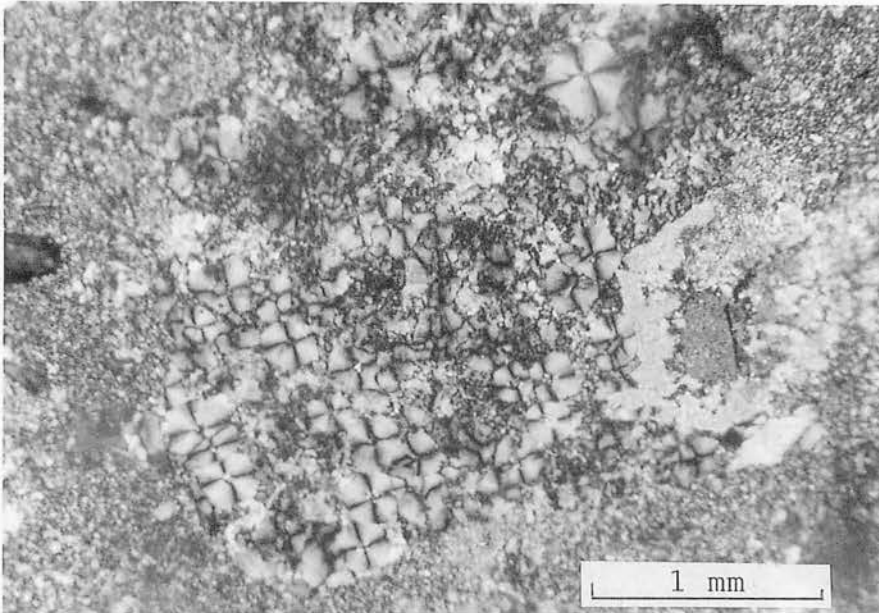


Plate 6: Photomicrograph of thin section of tuff from Johore. Spherulitic (cross patterns) fibrous quartz clasts (centre of photo) are sometimes found.

Tridymite is found in two geologic environments. Firstly it is found as a pneumatolytic (or sublimation) products in cavities in felsic volcanics. Secondly, it is known to occur as a contact metamorphic mineral in sandstone fragments embedded in basaltic volcanic rocks (Annibale *et al.*, 1983).

Strained quartz

Normal quartz is not known to cause ASR. However, BRED (1982) and Diamond (1978) pointed out that strained coarsely crystalline quartz in quartzite has been known to cause ASR.

Physically and chemically, strained quartz and normal quartz do not show any difference. In thin sections strained quartz can be distinguished from normal quartz by the presence of broken, uneven and wavy extinctions in the former. These discontinuities observed in strained quartz grains indicate that crystal structures of such grains have been ruptured. The rupturing is most likely caused by external stresses which had acted upon the rocks, often in a higher temperature environment. Strained quartz is of common occurrence in regionally metamorphosed rocks such as meta-quartzite, schists etc. Strained quartz also occur in granites and other rock types which have been subjected to certain compressive stress which, however, is not strong enough to cause metamorphism.

Cristobalite

Though this mineral is tested to be reactive (Rhoades and Mielenz, 1948), its rarity (in occurrences and amounts) in nature precludes it to be of significance in the cause of ASR.

Acidic volcanic glass

This is a rock type rather than a mineral. Acidic volcanic glass and its devitrified equivalents are known to cause ASR (Stanton, 1940(a), 1940(b); Rhoades and Mielenz, 1948).

Acidic volcanic glass is associated with young volcanics. In older volcanics, generally, the volcanic glasses are devitrified that is converted to quartz, feldspars, chlorite, sericite etc. In most cases, devitrification is aided by the fluids and gases emanating from volcanic centres. Such devitrification process turned the glassy volcanics to a fine to medium-grained rocks. However, in the absence of volcanic fluids or gases, devitrification will be quite slow and the resultant rocks will be crypto- to microcrystalline in texture.

PESSIMUM PROPORTIONS

Reactivity of the silica minerals to alkalis is cement varies. Opal and "silica glass" are regarded as most reactive. Chalcedony, tridymite and cristobalite are

regarded as of intermediate reactivity. Strained quartz is regarded as of low reactivity though sometimes it is not reactive (Diamond, 1978). Non-strained macro-crystalline quartz is not reactive.

Reactivity accompanied by expansion is dependent on the pessimum proportion. Palmer (1981) presented evidence that expansion in concrete is dependent on the proportion of the reactive aggregate in the mix. Below a certain proportion, expansion is negligible and above certain proportion, expansion falls below the acceptable limits.

For opal, Ochard (1973) gave the pessimum proportion as 3 to 5%, while BRED (1982) gave 2-4%. However, Diamond (1978) states that the reactive component (beekite, and opal-chalcedony complex) which caused cracking and distress in the Vale de la Mar Dam on the Island of Jersey is ascertained to be only 0.02% of the total aggregate. For the silica mineral of intermediate reactivity no pessimum has been given, though Mindess *et al.* (1981) quoted ASTM C33 as limiting chert in aggregates to a limit of 5%. This will probably represent the lower limit of the pessimum.

In the case of low reactive strained quartz, evidence available indicate a 100% pessimum (BRED, 1982).

POTENTIAL REACTIVE ROCKS

Based on the review of the mineralogical factor which causes ASR, it is now possible to categorize potentially reactive rocks into several types. There are certain rocks because of its inherent mineralogical composition, must always be regarded as potentially reactive. Some examples are chert, flint and glassy volcanics. Some artificial aggregates like slags (not air-cooled) and fired bricks also come under this category.

Another group of rocks would be potentially reactive because they often can contain pessimum proportions of the reactive silica minerals. However, they may not contain any reactive minerals and therefore would not be reactive. Some examples of these are acid to intermediate volcanics.

The third group of rocks generally would be considered innocuous because the primary minerals of the rock are generally unreactive. However, these rocks can be impregnated with reactive secondary silica minerals (for example opal and chalcedony) which infill joints, fractures, faults, cavities etc. or which sometimes replace the primary constituents of the rocks. Examples of these rocks are granite, gabbro, diorite etc.

Table 2 groups the common occurring rocks into the classification as proposed.

Table 2: Summary of Potentially Reactive Rocks**GROUP I:****Inherently Reactive Rocks****Rock Types****Igneous Rocks**

	Reactive Silica Minerals	Occurrence
(a) Glassy Acidic to Intermediate volcanics	Silica rich glass	Common in young volcanic terrains
(b) "Cryptocrystalline" devitrified Acidic to Intermediate Volcanics	Silica rich glass, Tridymite	Common in young volcanic terrains
(c) Pumice or float stone	"Cryptocrystalline" quartz? Silica rich glass	Common in young volcanic terrains
(d) Geysersites	Chalcedony and opal	Common in young volcanic terrains

Sedimentary Rocks

(a) Chert as beds and nodules	Chalcedony	Very common
(b) Flint as beds and nodules	Chalcedony	Common in some regions
(c) Jasper as beds	Chalcedony	Common in some regions
(d) Diatomite	Chalcedony and opal	Rare
(e) Porcellanite or Porcellanous limestone	Chalcedony and opal	Rare
(f) Siliceous shale	Opal! and chalcedony?	Common

Metamorphic Rocks

(a) Metaquartzite	Strained quartz	Common
(b) Impact rocks	Silica glass, coesite?	Very rare
(c) Mylonized rocks	Amorphous silica?	Rare

GROUP II:**Potentially reactive if pessimum amounts of reactive mineral is present****Rock Types****Igneous Rocks**

	Reactive Silica Minerals	Occurrence
(a) Some rhyolite, dacite, trachyte and andesite flows and tuffs	Crystobalite? Tridymite, chalcedony, opal and silica glass?	Common in young volcanic terrains
(b) Ignimbrites	Tridymite, cristobalite	Common in subaerial volcanic regions

Sedimentary Rocks

Nodular Limestone	Chalcedony and opal?	Common in some regions
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Table 2: Summary of Potentially Reactive Rocks (Cont'd)

Metamorphic Rocks		
Some quartzose hornfels	Tridymite	Common
GROUP III:		
Innocuous rocks unless		
(a) Impregnated by secondary silica minerals (mainly opal and chalcedony)		
(b) Mylonized or vitrified (impact)		
(c) Contact metamorphosed (for quartzose rocks only)		
Rock types		
Igneous Rocks	Reactive Silica Minerals	Occurrence
(a) All coarse to medium-grained plutonic igneous rocks such as granite, adamellite, granodiorite, gabbro, diorite syenite, microgranite, aplite etc.	None in the primary constituents	Very common
(b) Basalt and some acidic to intermediate volcanics such as rhyolite, dacite, trachyte.	None in the primary mineral constituents	Very common
Sedimentary Rocks		
(a) Limestone	None in the primary constituents	Very common
(b) Siltstone and sandstone	None	Very common
Metamorphic Rocks		
(a) Schists, marble and metaquartzites	None in the primary constituents	Very common
(b) Amphibolite, granulite and migmatite	None in the primary constituents	Very common
(c) Hornfels	None in the primary constituents	Very common

(Data and reference to rock type taken from Williams *et al.*, 1965; Annibale *et al.*, 1983; Pettijohn, 1957; Read, 1970; Selley, 1976, 1985; Reading, 1986).

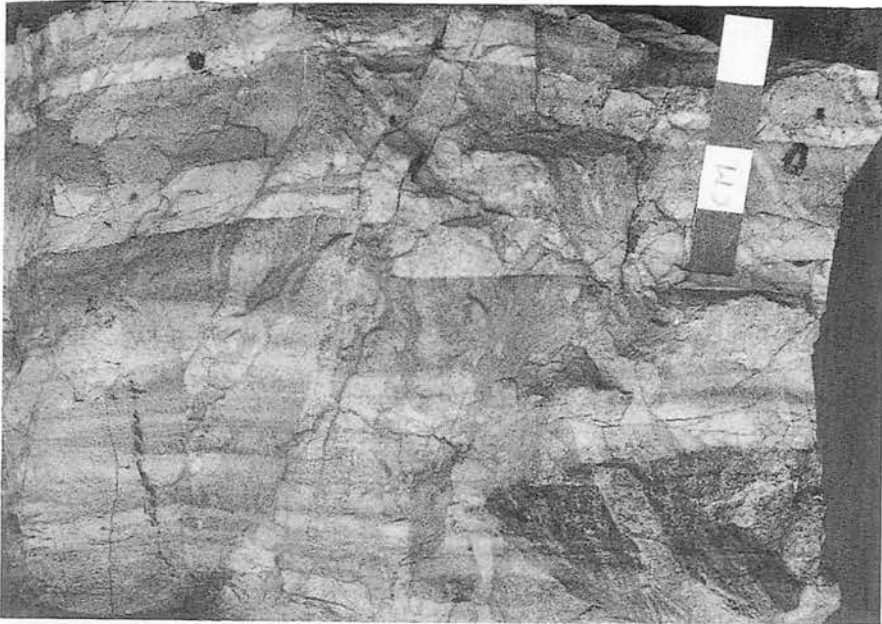


Plate 7: Specimen of flow banded rhyolite from Pahang. Texture of rock is "microcrystalline."



Plate 8: Specimen of pisolitic tuff from Johore. The rounded pisolites represent globules of volcanic dust collected by rain drops during eruption (during geologic past).



Plate 9: Specimen of agglomeratic tuff from Johore. Trace amounts of fibrous silica detected.

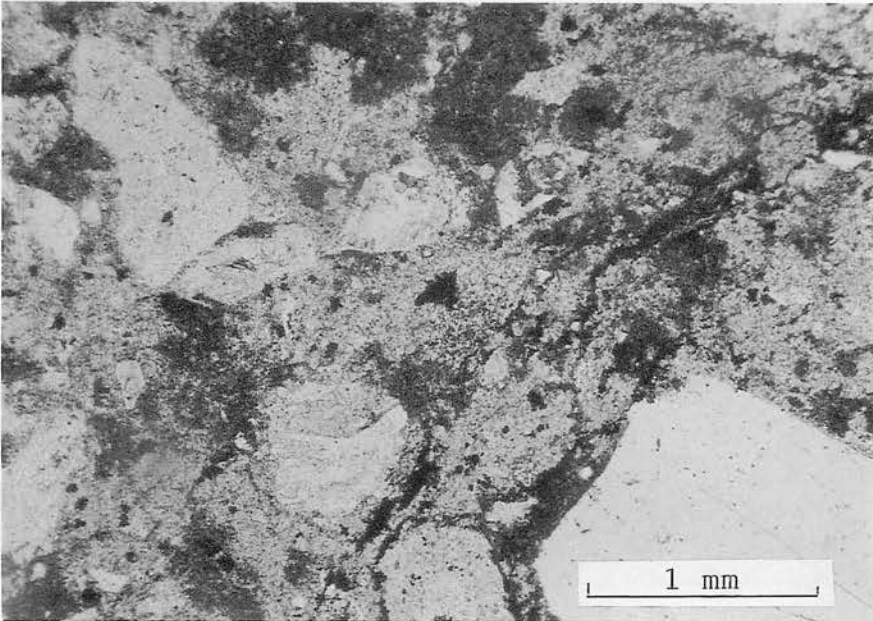


Plate 10: Photomicrograph of thin section of agglomeratic tuff (as in plate 9). Quartz fragment is at right bottom while the rest are lithic fragments.



Plate 11: Field photo showing fine-ash tuff (hammer head) and well sorted crystal ash tuff (hammer handle) from Johore. Fibrous silica absent in both.

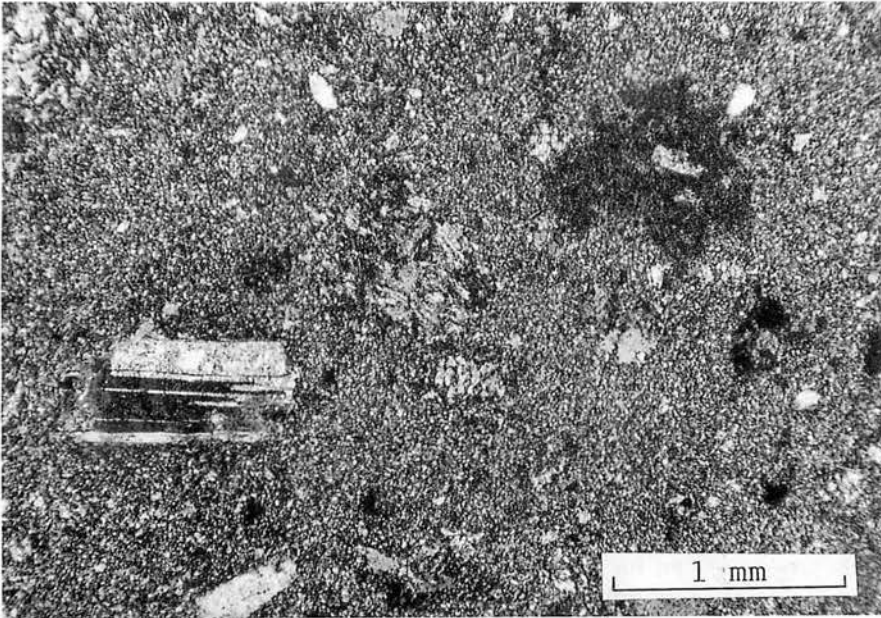


Plate 12: Photomicrograph of thin section of the fine-ash tuff under crossed polarizers.

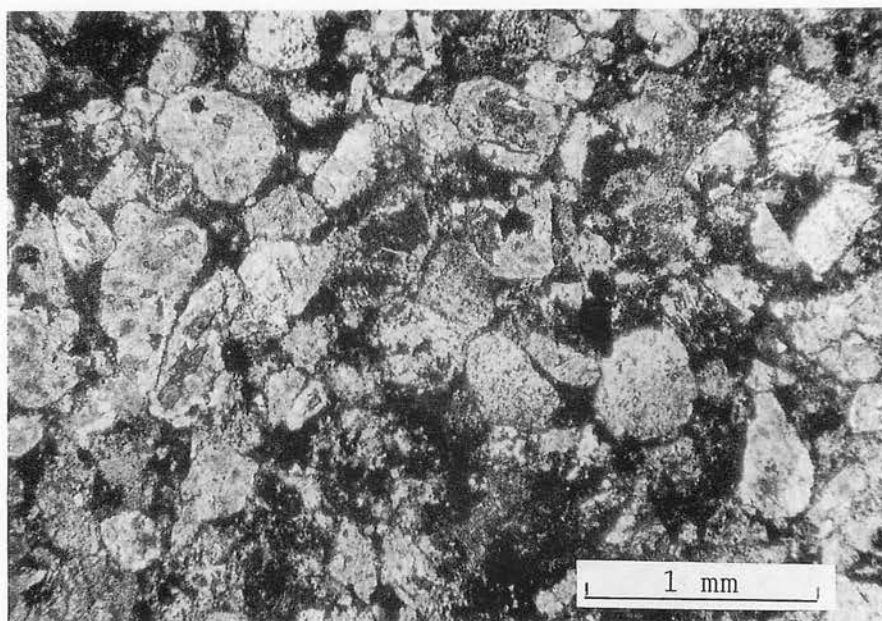


Plate 13: Photomicrograph of thin section of the well-sorted crystal ash tuff under plane polarized light

REACTIVITY OF FINE AGGREGATES

Presence of deleterious minerals in the fine aggregates has been known to give rise to ASR which ended in distress of concretes. Rhoades and Mielenz (1948) pointed out that ASR, in several localities in the USA, is due to the presence of devitrified volcanics, rhyolite tuff, opaline chert and opaline and tuffaceous limestone in the sand fractions of the aggregates. Gutt and Nixon (1981) mentioned that several ASR cases in U.K. could be traced to the presence of chert in the dredged sand aggregate. The pessimum proportions of these deleterious materials in the sand fraction capable of causing distress in concrete are not known.

STATUS OF LOCAL AGGREGATE SUPPLY

Peninsular Malaysia and Singapore are fortunate in that the much preferred crushed rock aggregates are easily available for construction purposes. Several rock types are quarried for the production of aggregates. Granite aggregates are easily available throughout, while limestone (marble) aggregates are popular in Kedah, Perlis, Perak, Kelantan, Pahang and Selangor.

However, limestone/marble aggregates are not suitable for high strength concrete. Volcanic rocks are quarried in Pahang and Johore (see plates 7 to 13). Other rock types such as hornfels, quartzite, sandstone, syenite gabbro etc. are also quarried from scattered localities.

Locally, fine aggregates for concrete consist mainly of quartzose resistate sand obtained from river (and streams), mine tailings and raised beaches or washed from clay/sand deposits and eluvial soil. Deleterious silica minerals are very rarely present in these sand aggregates though micas and clay which can also be reactive with the alkalis are often present, especially, where the sand originated from mica schist or granitic terrains.

While Peninsular Malaysia is self-sufficient in its aggregate requirements, Singapore has to source a large proportion of its aggregate requirements elsewhere.

In order to prevent the possible occurrence of failure of concrete due to ASR, Singapore has banned the import of volcanic aggregates for use in concrete.

ADVOIDANCE OF ASR

Many authors' (Diamond, 1978; BRED, 1982; Palmer, 1981; Gutt and Nixon, 1979; Verbeck and Gramlich, 1986; Mindess and Young, 1981) suggestions for avoiding ASR in concrete are:

1. Use low alkali cement (limit of 3.0 kg equivalent of Na_2O per cubic meter of concrete).
2. Use non reactive aggregates (several tests and monitoring procedures available).
3. Prevent or control external sources of moisture.
4. If use of high alkali cement and reactive aggregates cannot be avoided, addition of pozzolans can prevent or reduce ASR expansion to an acceptable level. Larson (1963) recommended the use of 20 g of pozzolans (finely divided reactive opaline silica) for every 1 g excess of alkalis in one cubic meter of concrete).

MONITORING OF REACTIVE AGGREGATES

Several suggestions (Diamond, 1978; BRED, 1982; Neville, 1978) for the selection and monitoring of non-reactive aggregates are:

1. A good service record of at least 10 years. This is regarded as the most reliable.

2. Motor-Bar Test (ASTM C227-71 reapproved 1976). This test required at least 3 to 6 months. Most reliable of the tests method.
3. Petrological Examination following guidelines of ASTM C295-73 and BS 812 Part I. This method lacks concrete criteria (e.g. pessimum proportions not clearly established) for clear cut conclusions to be made.
4. Chemical Method (ASTM C289-71). This, by its own is not fully reliable. It affords a quick means of assessment which should be confirmed by Mortar Bar Method or supplemented by petrographic examination.

CONCLUSION

A review of the mineralogical and petrological factors indicates that a multitude of rocks can cause or potentially can cause ASR. A sweeping ban on the imports of volcanic aggregates (for use in concrete) may give rise to a false sense of confidence for the prevention of ASR.

High contents of alkalis in cement to be used in concrete not only can cause ASR but also alkali-carbonate and alkali-phylosilicate reactions. A ban on the import of high alkali cement/clinker appears to be a safer and, easier and a more effective way of preventing ASR and other alkali aggregate reaction in concretes.

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