

SLOWLY REACTING AGGREGATES IN SWEDEN - MECHANISM AND  
CONDITIONS FOR REACTIVITY IN CONCRETE.

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Most of the Swedish bedrock is made up of the very old crystalline bedrock of the Baltic Shield. Of these rocks, some metavolcanics (rhyolites) and deformed rocks (mylonites) give alkali-silica reactions in concrete. These aggregates react slowly and it takes more than 15 years for a cracking pattern to appear. Examination of cracked concrete and laboratory tests show that with these aggregates there is no pessimum and that the stones are as deleterious as the sand. The reactive grains are dissolved from the inside which may explain this. A mechanism is presented.

INTRODUCTION

In Sweden the alkali-silica reaktion (ASR) is fairly unknown as a deterioration process of concrete. Freezing and reinforcement corrosion are regarded as being the most common causes.

The known cases of ASR are from southern Sweden where the gravel, like in the immediate parts of Denmark and Germany, contain reactive chert/flint from Mesozoic limestone formations. It has also been reported from Norway, see Jensen (1), that some of the rocks from the mountainbelt, the Caledonides, may cause ASR. The other parts of Sweden are covered by very old, precambrian, mainly granitoid rocks of the Baltic Shield. An early investigation by Hagerman & Roosaar (2) with a quick chemical and mortar bar test (ASTM C 289-54 T and C 227-52 T) indicated that the rocks were not reactive. With the rocks from the Caledonides expansion was observed but no gel could be identified.

In the mid-eighties, however, it was found the Stockholm city materials testing laboratory that strongly sheared rocks, mylonites, caused ASR in some bridges from the 1930's, see Lagerblad & Nieman (3). Later assessment of damaged concrete revealed that cracking caused by ASR is fairly common in Stockholm. As a result of this the Swedish Cement and Concrete Research Institute (CBI) started a research project, to find out

if ASR occur in other parts of Sweden. A geological survey of potentially reactive rock types was made and concrete structures in areas where such rock types are common were inspected. In cooperation with the road government drill cores were taken from concrete structures with the characteristic "map-cracking" pattern. The cause of the cracking was investigated using polished surfaces, thin sections and SEM. The composition of the gels were investigated by energy dispersive techniques. The results show that ASR is fairly common locally but that the concrete structures are rarely severely damaged.

The reactive components in the aggregate are mainly deformed (kataclastic) rocks with stressed quartz and fine-grained volcanic rocks (rhyolitic porphyries). The stressed quartz is generally very fine-grained and the proper name of the most reactive type is mylonite.

One of the reasons for the occurrence of ASR is that most bridges from the sixties up to the eighties were built with a cement which contained more than 1.1 weight %  $\text{Na}_2\text{O}$  equivalent. Earlier the content of alkalis was generally lower. The ASR cracks are observed in bridges from the early seventies and before. This shows that it takes more than 15 and up to 20 years for the ASR to develop to the stage where cracking can be noticed. Previously the damage was mistaken for frost deterioration as this is often accompanied by ASR.

To investigate the reactivity of different gravels and rock types, accelerated tests were also performed (see below). They show a typical expansion pattern with a slow initial stage.

#### REACTION PATTERNS WITH SLOWLY REACTING AGGREGATES

Polished cores and thin sections prepared from cracked concrete indicate that the gel tends to be formed in the coarse aggregates. Furthermore, the thin section analysis shows that the slow-reacting aggregates have a different dissolution pattern compared to chert/flint from southern Sweden. The causes and consequences of this have investigated in further detail.

#### Petrographic analysis

The deleterious components in the gravel from southern Sweden are porous flint and opaline limestone. The quartz dissolve more or less completely and form a gel. The fine-grained slow-reacting rocks, however, seem to dissolve from the inside. In thin sections, impregnated with fluorescence epoxy and analysed with fluorescence light in the microscope (Figs. 1 and 2), one can

observe a vein-like system, where thin veins lead to thicker veins that eventually lead to the cement paste. It can also be observed that the gel advances in pulses. In the grains no thin veins or other indications of dissolution can be observed in the outer rims, only in the center (Figs. 1 and 2). To investigate this further we carried out SEM with backscatter electron imaging (SEM-BEI), on polished pieces of concrete.

Energy dispersive analyses showed that the gel changed composition from being an alkali-calcium-silica gel at the openings to a calcium-gel with some alkalis in the cement paste. The change is not continuous but occurs in steps as indicated by SEM (Fig. 3). A consumption of portlandite can be observed around the gel in the cement paste. At the rim of the gel closest to the paste the Ca/Si ratio increases indicating a composition closer to a calcium silicate hydrate (C-S-H) gel. The calcium gel is much harder than the alkali gel, at least during the preparation. This is indicated by the fact that most of the gel in the grains disappear during the preparation (drying and vacuum) allowing the fluorescence-exposi to intrude the aggregate grains. However, at very large magnification on the SEM-BEI (Fig. 4) a gel and dissolution phenomenon can be observed. The needles in Fig. 4 are crystallized pure alkali-silica gel. The darker rim, showing lighter elements, indicate a dissolution zone to the light-coloured mylonitic aggregate grain.

### Expansion tests

As a test method we selected a Danish method (NT-build 295). The reaction is accelerated by warm (50 °C) water saturated with NaCl. Originally the method is based on mortar prisms, but for reasons mentioned above it was modified and concrete prisms were used. In the mixture we used a portland cement with 1.1 wt. % Na<sub>2</sub>O equivalent. The content of cement and aggregate is 440 and 1735 kg/m<sup>3</sup> respectively. The water cement ratio is 0.45. The prisms have a dimension of 80 x 80 x 300 mm. Each test is performed with three prisms and the average value is reported.

With this method we tested a whole range of different aggregates. The results correlate well with the expected expansion. We found the same time difference between the chert and the slow-reacting aggregates (Fig. 5) as we do in real constructions.

To investigate the reactive properties of the slow-reacting aggregates we selected two rock types. One is from a shear zone which goes east-west trough the town of Stockholm. Like most cataclastic rocks it is very polymict. Most of it, however, has a texture with very fine-grained quartz, feldspars and micas. Occasionally larger quartz grains with undulose extinction can be observed. In general, however, the quartz grains can only be observed with the largest magnification of the optical

microscope. Even at this size the grains show deformation features like banding etc. The grain boundaries are serrated.

The other rock comes from a 1850 million year old volcanic sequence in the Bergslagen area of central Sweden. It is formed by the deposition of fine-grained volcanic ashes and chemical precipitations in a submarine environment. The rock is not glassy but it is difficult to observe the individual grain in the microscope. It consists of quartz, feldspar and mica. Chemical analyses of the bulk rocks show around 80 wt.% SiO<sub>2</sub> and 5-7 wt.% alkali oxides, i.e the rock has a rhyolitic composition. Thus it is called rhyolite in the diagram (Fig. 5). The rock called "porphyritic volcanic rock" is a rhyolitic devitrified ignimbrite.

The rocks were crushed and separated in different fractions. As a reference aggregate a gravel from the south of Stockholm known to have a good record, was selected. However, at the end of the test we found that also this gravel had a small expansion caused by a small amount of reactive mylonitic rocks. In this reference gravel we exchanged different fractions.

With both the mylonite and the rhyolite the results show that there is no pessimum. The largest expansion is caused by 100 % crushed rocks (Figs. 5 and 6). With the mylonites the second largest expansion is caused by crushed rocks between 1 and 2 mm in size (13 wt.% of the aggregate). All the prisms with reactive grains in sizes larger than 2 mm show an expansion that is significantly larger than the reference prisms. However, the grains smaller than 1 mm do not cause any significant expansion. The expansion is the same although the aggregate in the concrete contains 24 wt. % mylonite. A test showed that the "rhyolite" rock has a pozzulanic activity which might explain this phenomenon. The results from the rhyolite show that an equal amount of crushed rocks larger or smaller than 4 mm show the same expansion, about the same as 100 % porphyritic volcanic rock in Fig. 3.

Thin sections of the prisms after the expansion test show that both the mylonite and the rhyolite have the same petrographical features as the corresponding rock types in real damaged concrete structures. The reactive grains are dissolved from the inside. With a low alkali cement (<0.6 wt.% Na<sub>2</sub>O-equiv.) neither of the rock types show any significant expansion.

#### DISCUSSION

The expansion tests show that the stones in the slow-reacting aggregates are reactive and cause expansion, but that the smaller sizes (1-2 mm) cause more expansion. This contradicts the observation in real structures. However, this may be due to the

time difference. To understand this we must look into the reaction mechanism.

The observations indicate that the reactive grains are dissolved from the inside. An alkali gel is formed within the grain. In contact with the paste and its poor solution it changes composition and becomes more viscous and richer in calcium. The same chemical change phenomenon has earlier been observed by French (4) with highly reactive chert. Ca-rich gel is generally common in old cracked concrete, see Knudsen & Thaulow (5). We have also observed the same chemical conversion as an aging phenomena in badly dispersed microsilica granulates. In the granules the first product is a liquid alkali-silica gel with some Ca rich which is formed in less than 24 hours. It shrinks considerably by the vacuum treatment in the SEM which indicates a high water content. Later a rim of viscous Ca-alkali-gel is formed. The next stage is a hard shell of pure Ca-gel. The liquid nature and high content of water in the alkali gel would explain why it

The change in composition of the gel is caused by either dilution of the alkalis by a calcium compound from the cementpaste or by an cation exchange reaction where calcium replaces alkalis. The relative decrease in the content of  $\text{SiO}_2$  in the gel indicates a reaction with a calcium component from the cement paste. This is obvious for the outer shell. However, there must also exist an extensive calcium/alkali exchange which releases alkalis, see Wang and Gillot (5). The calcium probably comes from disintegrating portlandite in the marginal zone to the gel-filled cracks. The hydroxide ions will be released to the fluid.

The cation exchange would cause the displaced alkali to return to the crack in the aggregate grain. The returning alkalis will keep the alkali content of the fluid in the crack high and thereby capable of dissolving more quartz. The chemical system will only consume a very small amount of alkalis to form Ca-silica gel in the paste. The final product will be a calcium gel with calcium from the portlandite in the paste and silica from the aggregate. The gel will probably form a membrane at the openings of the cracks. It is possible that the higher concentrations of alkalis in the crack solutions induce osmosis which will suck water and/or hydroxide ions into the cracks in the reactive grains.

It will take some time to initiate this process which presumably explains why the aggregates react slowly. Moreover, some initial cracks are needed in the aggregates. However, once the process has started it will continue and can produce large amounts of gel as no alkalis are consumed. Consequently there is ~~no pessimism. More reactive grains will give rise to greater~~ amount of gel and consequently a larger expansion. As long as the concrete has access to water the process will continue.

When the process has started the poor solution will keep a very high content of alkalis and maintain a high pH. It is also

possible that the aggregates provide a source of alkalis. The suggested mechanism could explain why the chemical tests have poor results with slow reacting aggregates.

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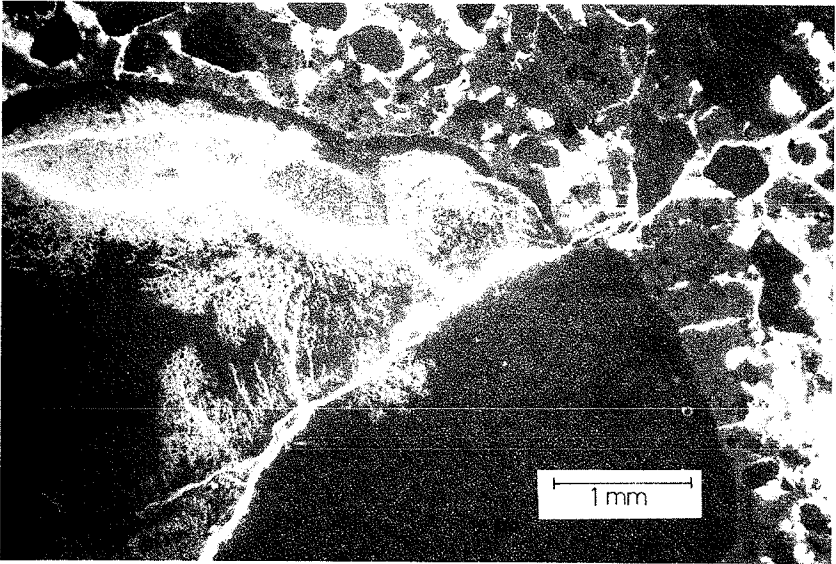


Figure 1 Rhyolitic aggregate being dissolved from the inside. Twenty years old bridge from Dalarna in central Sweden. Thin section with fluorescent light

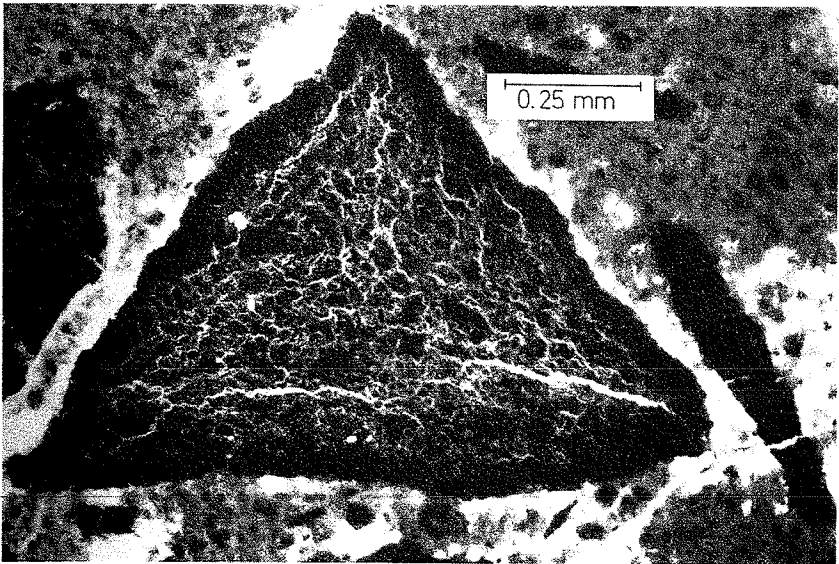


Figure 2 Rhyolitic aggregate from accelerated test being dissolved from the inside. Thin section with fluorescent light.

	①	②	③	④
SiO <sub>2</sub>	67	65	58	50
CaO	13	26	37	48
K <sub>2</sub> O	10	5	2	1
Na <sub>2</sub> O	10	4	3	-

(Portlandite)

Ca<sup>2+</sup> OH<sup>-</sup>

Na<sup>+</sup> K<sup>+</sup>

New alkali  
silica gel  
formed

H<sub>2</sub>O

Na<sup>+</sup> K<sup>+</sup>

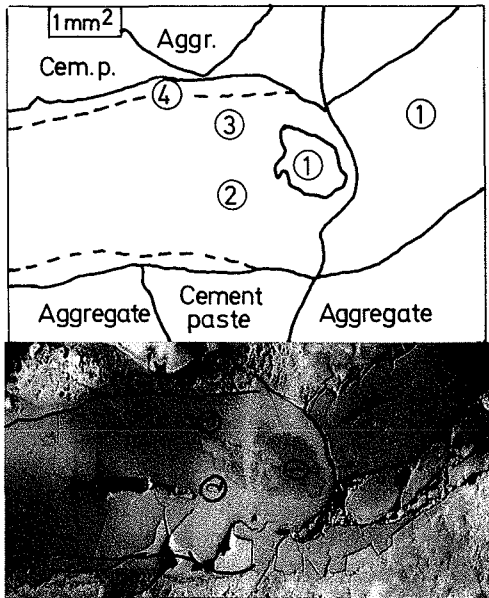


Figure 3 SEM-BEI picture and chemical composition of gel in damaged concrete structure. Analysis normalised to 100%.

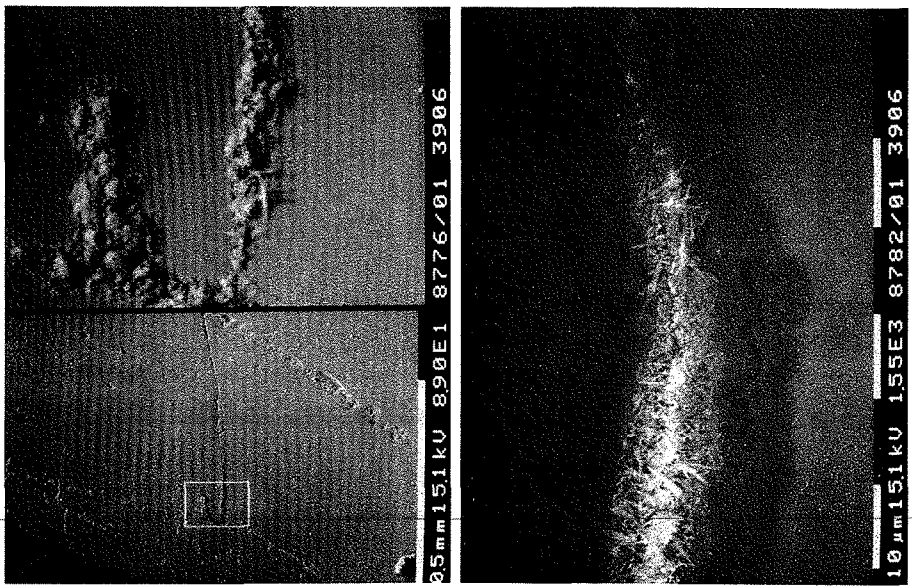


Figure 4 The formation of crystalline alkali-silica gel in microcracks in a mylonite aggregate. Sample taken from a bridge in Stockholm.



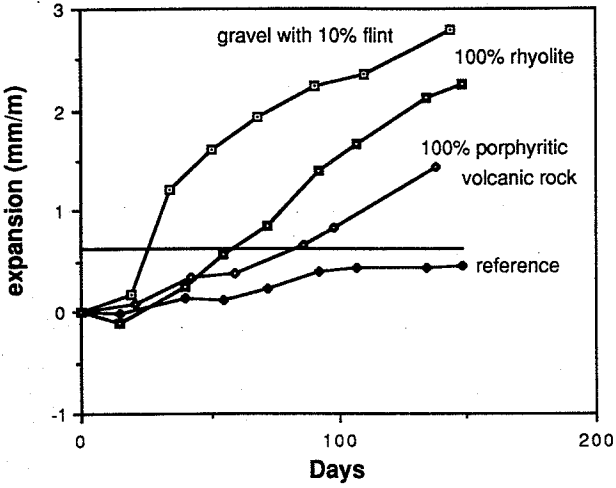


Figure 5 The relative expansion between different rocktypes.

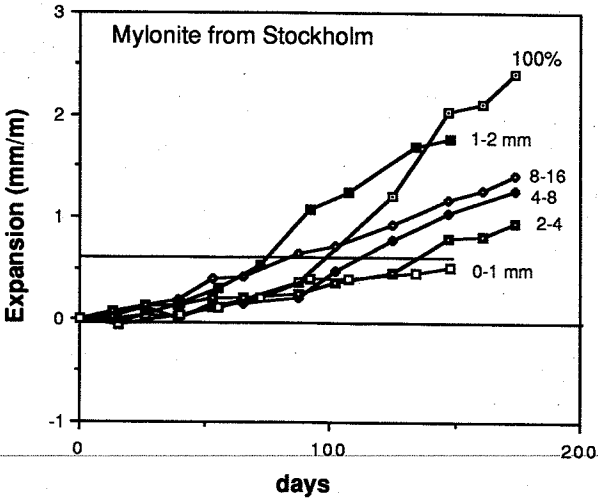


Figure 6 Expansivity of different fractions of mylonite from Stockholm.