INFLUENCE OF AGGREGATE PARTICLE SIZE ON THE ALKALI-SILICA REACTION – A LITERATURE REVIEW

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Abstract

The literature review has shown that the size of a given alkali reactive aggregate particle has a significant effect on the rate on the alkali reaction as well as the potential for expansion and damage in concrete structures. Some very rapid reactive aggregates have a pessimum particle size in the range 0.05/0.2 mm, while coarser aggregate particles give less expansive reaction. This may be due to the high surface area leading to a faster reaction rate. On the other hand, there are a number of reports from the literature showing that very fine alkali reactive particles may have the opposite effect than causing ASR expansion – they rather lead to an inhibiting effect due to pozzolanic behaviour.

Some slowly reactive aggregates may have a pessimum for aggregate particles in the range 1/2 mm, while for Norwegian slowly reactive aggregates there is evidence from field structures that the coarsest particles (> 8 mm) are the most harmful. The latter effect may be explained by fracture mechanics. The effect of the aggregate particle size on the ASR expansion appears to vary greatly depending on the mineral- and rock composition, but is also depending on the exposure conditions. Consequently, the behaviour under accelerated laboratory conditions may differ from the behaviour in a field structure.

Keywords: alkali-aggregate reaction, particle size, reaction rate, lab/field- correlation

1 INTRODUCTION

In the literature, the effect of the aggregate particle size on the expansion due to Alkali Silica Reactions (ASR) appears to vary greatly depending on the mineral- and rock composition and the exposure conditions. As a consequence, the behaviour under different accelerated laboratory conditions may diverge from the behaviour of the same aggregate fraction used in a field structure. To be able to predict the field behaviour of a given alkali reactive aggregate fraction in a reliable way only based on accelerated laboratory testing, it is thus crucial to gain more knowledge about these topics. This is the main motivation for performing this literature review.

Furthermore, the understanding about the link from laboratory testing to field performance must also be increased. Such validation of laboratory results is an important topic in the RILEM TC 258-AAA (2014-2019) and in the Norwegian research project "236661/O30 KPN-ASR" (2014-2018).

The fact that the size of the reactive aggregate particles has a significant effect on the rate of reaction, as well as the ultimate potential to cause expansion and damages to concrete structures has been known since Stanton in 1940 [1] concluded that the aggregate particles (siliceous magnesium limestone containing opal and chalcedony) in the 170–600 μm size range yielded greater expansion than coarser sizes. Contradictory, a comprehensive Norwegian field survey reported by Lindgård & Wigum [2] and Lindgård et al. [3] revealed that mostly the larger particle sizes > 8-10 mm contribute to harmful ASR, while the smaller particle sizes seem to be much less harmful. Obviously, slowly reacting Norwegian aggregates behave completely different from the much faster reactive aggregates tested by Stanton in the 1940-ties in this respect.

Moreover, highly reactive aggregates may even react pozzolanic when ground down to a very fine powder and cause an inhibiting effect rather than a harmful alkali-silica reaction. For example, finely ground Icelandic rhyolite was previously blended with Portland cement to produce a pozzolanic cement. Later on, rhyolite powder was replaced partly by silica fume having much higher pozzolanic activity as reported by Gudmundsson [4].

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FILLER SIZED PARTICLES (< appr. 100 µm)

2.1 ASR versus pozzolanic reaction

The first stage of the alkali-silica reaction is a dissolution reaction. The further stages of reaction involving silica, alkalis as well as calcium may cause either a swelling alkali-silica gel or a non-swelling gel being similar to the C-S-H produced by hydration of Portland cement. The similarities between the alkali-silica reaction and the pozzolanic reaction have been treated by many in the literature, for example Urban [5], Wang & Gillot [6], Xu et al. [7] and Thomas [8], but are still not completely understood. The ratio between available calcium and alkalis near the dissolving silica seems to be of large importance, where a higher calcium to alkali ratio seems to favour the pozzolanic reaction over the alkali-silica reaction according to Hou et al. [9], Dent Glasser & Kataoka [10] and Powers & Steinour [11]. The particle size is of great relevance in this respect. Materials being highly pozzolanic are generally very fine-grained materials, meaning that the alkali-silica gel formed in the initial reaction stage is evenly distributed throughout the cement paste. In this situation, the alkali-silica gel can pick up sufficient amounts of calcium and form C-S-H. Larger sized reactive aggregate particles lead on the other hand to an accumulation on larger deposits of alkali-silica gel, which can become expansive. One example of this is silica fume, which is extremely fine-grained pure amorphous silica. Silica fume is when fully dispersed highly pozzolanic, thus being able to mitigate ASR. However, oversized grains or agglomerates of silica fume in sizes of 100-200 µm or larger may cause harmful ASR rather than mitigate it as described by Diamond [12], Marusin & Shotwell [13] and Gudmundsson & Olafsson [14]. Corresponding examples are given in the next section. It is thus a paradox that the same material may lead to either a harmful alkali-silica gel, or a beneficial C-S-H, only depending on the particle size of the material being applied.

The extra C-S-H being formed by the pozzolanic reaction increases the strength of the concrete. Additionally, it gives a denser matrix phase generally causing a more durable concrete with respect to ingress of aggressive substances. Moreover, effective pozzolans are highly effective in lowering the hydroxyl ion concentration. The main reason for this is that the pozzolanic additions lead to formation of C-S-H with higher binding capacity of alkalis as reviewed by Thomas [8]. Due to the lowered pH in the concrete pore water, less SiO₂ is dissolved from the alkali-reactive aggregate particles. The lowered pH is the main reason for the inhibiting effect of pozzolans on ASR [8].

2.2 Observations from laboratory studies

Highly reactive Icelandic rhyolite has proven to be an efficient pozzolan when ground down to cement-sized particles and mixed with Portland cement according to Gudmundsson [4]. Quingham et al. [15] have demonstrated that andesite with α-cristobalite as the primary reactive component has proven to mitigate expansions caused by ASR when ground to powder with Blaine values in the range from 290 to 1133 m²/kg. The prisms of size 40x40x160 mm were autoclaved before long-term storing at a temperature of 45ºC. The finer the fractions, the higher the resulting mitigating effect on expansion.

Pedersen [16, 17] investigated in his PhD-study the possible mitigation effect of alkali-reactive filler particles from Norwegian cataclastic rocks, along with fillers of Icelandic glassy rhyolite as well as crushed bottle glass. Non-reactive reference fillers were included in the study, as well as silica fume and fly ash known to mitigate ASR. Ground rhyolite and crushed bottle glass significantly reduced the expansions compared to the reference concrete. On the other hand, the finest particles from two different Norwegian alkali-reactive cataclastic rocks had minor effect or even caused an increase in expansion, see Figure 1. These results were based on experiments using the 38ºC Norwegian concrete prism test (CPT) [18], which is believed to provide a realistic picture of the behaviour in real field structures. Pedersen [16] found that the filler particles being able to mitigate ASR had a distinct amorphous silica phase which caused a significant pozzolanic reactivity. On the other hand, the reactive substance in the two Norwegian cataclastic rocks was crystalline quartz, which favoured ASR rather than pozzolanic reaction.

Another important finding from the study of Pedersen [16] was that aggregate fillers leading to increased expansion when tested by the Norwegian 38ºC CPT [18] resulted in reduced expansion when tested by the 80ºC accelerated mortar bar test (AMBT) [18]. Contradictory results were thus obtained depending on the test procedure. The reason for this shift in behaviour when moving from 38ºC to 80ºC is due to the fact that pozzolanic reactivity increases significantly with increased temperature.

Ground glass filler with particle sizes below 38 µm are reported to cause reduced ASR expansion relative to the control mortar containing no glass filler, while coarser particles had less effect when tested by the AMBT according to Shao et al. [19]. Also Hudec & Ghamari [20] reported
similar findings where glass fractions below 75 µm reduced the expansions. On the other hand, glass particles > 75 µm gave the opposite effect and caused increased expansion when tested in the AMBT.

In 1951, Vivian [21] reported that silicious magnesian limestone in particle sizes < 50 µm is innocuous and caused no expansion at all, while particle sizes > 70 µm yielded rapid expansion. For particles between 50 and 70 µm, ASR expansion occurred, but delayed compared to coarser particle sizes. The prisms were stored over water at 20°C. Also Multon et al. [22] found that reactive siliceous limestone ground to particles below 80 µm caused no expansion at all when tested at 60°C at a RH of 95 %, while the coarser fractions expanded, see next section.

Diamond & Thaulow [23] carried out an investigation with opal of α-cristobalite type, with crushed and sieved fractions from 125/150 µm down to 20/30 µm. In this set of experiments, cylindrical mortar specimens with a length of 20 mm and a diameter of 10 mm were stored over water at 20°C. The study did not give evidence to any significant effect of the particle size, as all particle sizes down to 20-30 µm were capable of producing large expansions. The authors concluded that if there is a limiting size below which expansion will not take place, it is less than 20-30 µm.

3 "NORMAL" SIZED AGGREGATE PARTICLES

3.1 Laboratory experiences

Hobbs & Gutteridge [24] examined Beltane opal from California of different fractions in the range between 150 µm to 4.8 mm. They made mortars with different amounts of reactive materials (from 1 to 20 %). Each fraction was tested separately in combination with non-reactive sand. The specimens (25 x 25 x 250 mm) were stored at a temperature of 20°C in water. They found that for a given content of Beltane opal, the expansion increased with decreasing particle size, as can be seen from Figure 2. The mortar bars with reactive grains in the size range 150/300 µm expanded approximately 15 times more than the coarsest particles in the interval 2.4/4.8 mm.

Vivian [21] studied different particle sizes of opaline rock in the range of particle sizes from 50 µm to 2.4 mm. He found that expansions were subsequently high for all particle sizes down to 50 µm, while no expansion was observed for particles below this size (see also previous section). However, the particle sizes in the range 50/150 µm had a delay in the onset of the expansion compared to the coarser particle sizes. The storing temperature was 20°C in this set of experiments.

In a study by Mørtsell and Wigum (reported in Wigum [25]), substitution of innocuous material with reactive materials showed that reactive material in the finer fraction (0.15 to 0.8 mm) gave the highest expansion in the AMBT. The corresponding experiment with reactive material in the larger fractions (0.8 to 4.8 mm) gave significantly lower expansion. A similar test with the same total quantity of reactive material (50%) equally distributed in all fractions gave an even smaller expansion. The described differences were observed for all ages above 28 days, while there was no significant difference at 14 days of exposure.

Zhang et al. [26] performed a study on the influence of the aggregate size. The finest fraction (0.15/0.80 mm) had the greatest influence on the expansion, while coarser sizes caused less expansion. The reactive aggregate tested in this study was a quartz glass. The tests were performed by a method involving steam curing at 100°C followed by autoclaving at 150°C in a 10 % KOH solution.

Wigum & French [27] studied the accelerated expansion of mortar bars cut from real concrete, compared to mortar bars with standard grading according to the AMBT [28]. The mortar bars with the finer grading expanded more rapidly in the beginning, while the identical sized mortar bars with coarser grading yielded an ultimately higher total expansion. By thin section analyses, it was found for most samples that the finest particles below approximately 300 µm did not react.

Nishibayashi & Yamura [29] observed that the ASR expansion of concrete containing reactive fine aggregates increased rapidly to a constant level. Coarser particles on the other hand reacted more slowly during the early period, but continued to react and expand for a much longer period. The experiments were carried out using andesite as the reactive aggregate, while the prisms were stored at 40°C over water. When reactive fine aggregate were mixed with reactive coarse aggregate, the expansions were small and became constant in the early stage due to pessimum behaviour.

In a recent investigation by Binal [30] using the AMBT [28], it was found that the 0.15/0.30 mm fraction yielded the highest expansion, while the finer fraction as well as the coarser fractions yielded less expansion. The difference between the size fractions was much more pronounced for the opal aggregate than for the chert aggregate, as can be seen from Figure 3.

Multon et al. [22] studied different fractions of reactive siliceous limestone in fractions from 0/80 µm to 1.25/2.50 mm. The main results are shown in Figure 4. The particle size interval 0.63/1.25 mm expanded the most, while particle sizes below 0.315 mm yielded very low expansion, or
even no expansion at all for the finest fraction below 80 µm. This set of experiments was performed using an elevated temperature of 60ºC and 95 % RH.

Ramyar et al. [31] studied five different fractions of a natural reactive aggregate combined with non-reactive aggregate. The tests were performed using the AMBT [28]. The intermediate fractions 0.25/0.5 mm, 0.5/1.0 mm and 1/2 mm expanded more than the 0.125/0.25 mm and the 2/4 mm fractions. However, when normalized for different amounts of aggregates in each fraction, which differ from 10-25 % according to ASTM C1260, the differences were relatively small. In addition to testing the natural aggregate, testing was also performed on aggregate sizes crushed from large aggregate particles of the same origin. An interesting observation from these experiments is that the size effect of different aggregates was more pronounced for the crushed materials. For the three intermediate fractions, the crushed material yielded higher expansions than the corresponding fractions with natural uncrushed material. Obviously, not only the particle sizes, but also the angularity of particles may have some influence.

Dunant & Scrivener [32] investigated the effect of single fractions of reactive aggregate combined with non-reactive aggregate in ranges from 0/2 mm to 8/16 mm. The prisms were stored at 38ºC in a simulated pore solution (0.150 mol/l NaOH) in order to minimize leaching of alkalis. The 4/8 mm fraction expanded fastest, followed by the 8/16 mm, while the 0/2 and 2/4 mm fractions expanded identically and slightly slower than the 2/4 mm fraction. However, when the expansions were normalized for weight fractions of reactive material (there were nearly twice as much of the 4/8 fraction as of the 2/4 mm fraction), the ultimate expansions after 2 years were more or less equal for all fractions, with only some differences in the early stage of the expansion curves. The reactive aggregate used in this investigation is described as mixed mineralogy alpine aggregate, the reactive part being mostly chloritic schist. These results were input to numerical simulations. Some findings from these simulations are further discussed in Section 4.

3.2 Field experience and lab-field correlation

For rapid reactive rock types such as chert and volcanic glass, the most damaging size range appears to be from 3/7 mm according to French [33]. He further reported that for aggregates of greywacke and argillite, most damages have been found when they occur as coarse aggregate. In Japan, only a few examples of deterioration due to ASR in structures containing reactive fine aggregates have been reported according to Nishibayashi & Yamura [29].

Lagerblad & Trägårdh [34] reported that for Swedish slowly reacting aggregates the particle size 1/2 mm causes greater expansions than the coarser sizes. The expansion tests were performed using the Danish method (TI-B 51), where the prisms are stored at 50ºC submerged in a NaCl solution. This contradicts observations in structures, where the larger fractions were found to be as deleterious as the sand fractions.

In Denmark, about 90-95 % of all cases with ASR-damaged concrete structures are caused by porous opaline or calcareous opaline flint in the fine (sand) fraction according to Grelk [35]. ASR due to opaline or calcareous opaline flint can occur very quickly (< 5 years) under “severe conditions”, i.e. exposure to water, external alkali supply (e.g. from de-icing salt) and a critical amount of reactive particles in the aggregates. A maximum content of 2 volume-% potential reactive particles in the sand fraction is considered to be a safe upper limit to avoid ASR damage. Before the second half of the 1980’s, most concrete structures in Denmark were built without considering ASR. Furthermore, parts of the concrete business sector probably misunderstood the conclusions from the previous research reports. It seems that they wrongly believed that the ASR problems mainly were connected to the coarse aggregate fraction. By replacing coarse reactive aggregate with non-reactive ones (like crushed granite from Sweden, Norway and Bornholm (Denmark)), and keep on using the reactive sand with porous flint, the ASR problems even increased due to the pessimum effect (Nielsen et al. [36]).

According to Jensen (1993) [37], it has been confirmed by thin section analyses of reacted field concrete that aggregate particles of various slowly reactive Norwegian rock types smaller than 1-2 mm seldom show signs of reactions. Furthermore, he reported that ASR in Norway is mostly caused by the coarse fraction. Both these findings have later been confirmed by SINTEF in many laboratory analyses of concrete samples drilled from field structures with ASR. For example, the comprehensive field survey reported by Lindgård & Wigum [2] and Lindgård et al. [3] confirmed that the most deleterious aggregate particles are larger than 8-10 mm. This was taken into account in the Norwegian regulations revised in 2004 [38, 39], where a “grain size factor” of 2 is applied for coarse aggregates (> 8 mm) when calculating the critical amount of reactive rocks types based on petrographic analysis (Norwegian Concrete Association, 2005). The grain size factor is 1.5 for “fine coarse aggregate” (2/8 mm and 4/8 mm) and 1.0 for sand (< 8 mm). In the AMBT [18], a standard "sand grading" is used...
bar tests (AMBT) of the aggregate show the materials to be highly reactive and expansive, i.e. exhibiting accelerated mortar bar expansions of 0.7% after 14 days of exposure in the NaOH solution. South-East Iceland, containing reactive rock constituents of rhyolite and andesite. Accelerated mortar aggregate tested in this set of experiments was taken from a natural beach sand- and gravel deposit in aggregates (0.28 %) and the mix with 75 % reactive fine and 0 % reactive coarse (0.19 %). The reactive (Lindgård [45]). Other authors have questioned the reliability of test procedures with exposure expansion, which was enormously reduced compared to test procedures with much less alkali leaching 100 % RH or submerged in water, the rate and extent of alkali leaching totally controlled the ASR leaching on the measured ASR expansion for various versions of CPTs. When exposed to 60°C the reviewed test results. For example, Lindgård et al. [44] documented the high influence of alkali leaching in the laboratory followed by the mix with 75 % reactive fine and 0 % reactive coarse (0.55 %), and the mix with 50 % reactive fine and 0% reactive coarse (0.41 %). The concrete mix with 0 % reactive fine and 100 % reactive coarse showed a relatively low expansion (0.22 %). However, contradictory results were observed when examining the expansion results of the same mixes, measured at the outdoor exposure site after close to 8 years of outdoor exposure. It was then clear that higher amounts of reactive coarse aggregate governed the ASR expansion. The mix with 0 % reactive fine and 100 % reactive coarse aggregate exhibited higher outdoor expansion (0.4 %) than the mixes with 100 % reactive fine aggregate and 0 % reactive coarse aggregates (0.28 %) and the mix with 75 % reactive fine and 0% reactive coarse (0.19 %). The reactive aggregate tested in this set of experiments was taken from a natural beach sand- and gravel deposit in South-East Iceland, containing reactive rock constituents of rhyolite and andesite. Accelerated mortar bar tests (AMBT) of the aggregate show the materials to be highly reactive and expansive, i.e. exhibiting accelerated mortar bar expansions of 0.7% after 14 days of exposure in the NaOH solution.

4 DISCUSSIONS
As shown in the previous sections there are conflicting results reported in the literature. Obviously, there are "real differences" in pessimum particle sizes for aggregates with different mineral - and rock composition. However, there is no straightforward correlation between laboratory and field results, as exemplified by the recent report on contradictory Icelandic lab/field results by Wigum & Einarsson [40]. The latter is partly due to the acceleration itself in the laboratory (increased exposure temperature and/or exposure of samples to surplus alkalis) compared to field exposure conditions. These accelerating factors might have different influence on various rock types and various particle sizes. Another example is the huge temperature effect on the pozzolanic behaviour of various fillers, as discussed below (Pedersen [16]).

Furthermore, many known sources of errors in the laboratory might have influenced some of the reviewed test results. For example, Lindgård et al. [44] documented the high influence of alkali leaching on the measured ASR expansion for various versions of CPTs. When exposed to 60°C and 100 % RH or submerged in water, the rate and extent of alkali leaching totally controlled the ASR expansion, which was enormously reduced compared to test procedures with much less alkali leaching (Lindgård [45]). Other authors have questioned the reliability of test procedures with exposure temperature ≥ 60°C (Lindgård et al. [46]). Many of the laboratory tests are performed with the 80°C AMBT that among several researchers is regarded being non-reliable and not suited for testing certain types of aggregates nor be used for performance testing (Thomas et al. [47]).

Several studies have shown that the reaction rate increases with decreasing particle size. For example, Bazant et al. [48] found for waste glass that smaller particle sizes caused higher expansion, which can be intuitively explained by the larger surface area of smaller particles compared to larger particles, causing larger volume fractions of reactive material to undergo ASR. However, they also found a particle size of 1.5 mm, below which the expansions were decreased. This may be explained by using a fracture mechanical approach, which has been done by Bazant et al. [48], Golterman [49] and Bache [50] among others. Given that the expansion caused by the aggregate particle is greater than the strain capacity of the cement paste, a composite with a large particle in a matrix will always lead to local fracture and cracking. On the other hand, for a small particle, cracking may not be initiated even if the strain capacity of the matrix is exceeded. In other words, the strain capacity for a system with small particles is higher than the strain capacity of the matrix. According to fracture mechanics, cracks propagate if the released energy is larger than the fracture energy needed to propagate the crack. Furthermore, the released energy for a swelling particle of radius R is
might be used to mitigate ASR or at least reduce the ASR expansion. However, fillers from other locations are often introduced to concrete and may have different properties.

Alternatively, laboratory investigations using CPTs with concrete having particle size distribution close to the concrete being used in field may give sufficient information. However, as have been shown, not even CPTs with relatively moderate acceleration such as the RILEM AAR-3 [41] are always capable of giving correct information in this respect.

5 CONCLUSIONS AND RECOMMENDATIONS

The literature review has shown that there obviously are some real differences in pessimum particle sizes between aggregates of different origin as has been shown. There is, however, no straightforward correlation between laboratory and field results. For the finest particles, elevated temperatures used in the AMBT (80°C) may cause a false positive effect due to pozzolanic reactions, which can alter the results. However, also documented examples where the 38°C RILEM AAR-3, 2000 [51] gives results that are contradictory to field results. To gain the most realistic information regarding the effect of the potential reactivity of the different aggregate fractions, samples from concrete field structures or concrete blocks stored outdoors should be examined. Alternatively, test methods such as ASTM C-1293 [51] or RILEM AAR-3 [41] with realistic aggregate grading and temperature not too far from real temperatures in field should be used. Test methods like the AMBT (ASTM C1260 [28] or RILEM AAR-2 [52]) using elevated temperature, and only the lower fraction of the aggregate curve, will not give reliable information in this respect. When testing the ASR expansion potential of different size fractions of aggregates with pessimum behaviour, special test procedures have to be developed, involving several tests to identify the combined pessimum amount and pessimum particle size.

Further work on performance testing of concrete and in particular on the link between laboratory results and field performance is in progress in RILEM TC 258-AAA (2014-2019) and in the Norwegian research project "236661/O30 KPN-ASR" (2014-2018).
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**FIGURE 1:** Effect of addition of aggregate fillers on the 2 years expansion using the Norwegian 38°C CPT [18]. From Pedersen [16].
FIGURE 2: Effect of different particle sizes of the reactive aggregate on the ASR expansion for mortar bars contacting 6% of reactive aggregate by weight of total aggregate. The mortar bars were stored in water at 20°C. From Hobbs and Gutteridge [24].

![Figure 2](image)

FIGURE 3: Effect of different particle sizes of the reactive aggregate on the ASR expansion of mortar bars. The test procedure was the AMBT. The aggregates were composed of pessimum ratios of opal (20%) and chert (40%), respectively, combined with non-reactive aggregate. From Binal [30].

![Figure 3](image)

FIGURE 4: Expansions after 500 days of mortar bars containing 30% reactive siliceous limestone of different fractions, combined with 70% non-reactive sand. The mortar bars were exposed to 60°C and 95% RH. From Multon et al. [22].

![Figure 4](image)