

Influence of Silica Fume on the Stresses Generated by Alkali-Silica Reaction

by

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ABSTRACT: Present test methods for determining the suitability of a given combination of cement and aggregates with respect to the potential for alkali-silica reaction (ASR) are based on the measurement of the expansion of the concrete/mortar under prescribed conditions. The scope of this study was to determine the influence of the addition of silica fume as a cement replacement on the stresses generated by the specimens, a parameter that could be useful for the engineer. Cylindrical specimens were placed in a frame equipped with a load cell that fixed the axial strain at approximately 200 and measured the axial stress generated over time. A replacement of cement by silica fume at a dosage of 15% by mass for two different w/c ratios led to a significant decrease of the measured stress. SEM observations helped to provide a tentative explanation for the stress and expansion reduction mechanisms with the addition of silica fume.

KEYWORDS: alkali-silica reaction; mortar; silica fume; stress measurements; stress relaxation

While damage of concrete due to alkali-silica reaction (ASR) may not be the leading cause of the collapse of a constructed facility, it is a major factor in reconstruction. Nevertheless, a small expansion due to ASR can cause overall deterioration or technical problems when ASR is present in massive concrete construction such as dams (Bérubé and Carles-Gibergues 1992). For instance, a minor expansion was reported to jam turbines or overflow valves (Bérubé and Carles-Gibergues 1992). Many studies (Hobbs 1988; Diamond and Penko 1992; Helmuth 1993) have been published on the topic, and the major factors related to the material used contributing to ASR have been identified: water, hydroxide ions, other cations, and reactive silica. Related environmental factors that can play a significant role in ASR are the relative humidity (RH), the porosity of the concrete, and mineral admixtures in the concrete. The cations and other ions are released into the pore solution mainly by the cement, but can in principle be released from other concrete constituents as well (Bérubé et al. 1992).

The scope of this study was to determine the influence of the addition of silica fume as a cement replacement on the stresses generated by the specimen. Comparison was also made between cement replacement with silica fume and cement replacement with an

inert silicon carbide powder. A parameter that could be useful for the engineer is the value of the stresses generated by the ASR reaction. Therefore, a device to measure the axial stresses on a confined mortar cylinder was designed (Ferraris et al. 1995, 1996, 1997).

The stress measured was a combination of any initial applied stress, loads generated by the ASR gel expansion, and loads generated by the expansion associated with water absorption that counterbalances self-desiccation shrinkage. To isolate the stresses due to the ASR expansion, reactive and non-reactive sand mortars were compared.

Using SEM observations, a tentative explanation of the results is given to determine the leading mechanisms for the mitigation effect of silica fume on ASR expansion and stresses. The cement content reduction alone does not reduce the expansion and stresses due to ASR. The addition of silica fume up to 15% replacement of cement reduces, but does not eliminate, the deleterious effect of ASR.

Background

It is apparent that ASR occurs between certain forms of silica present in the aggregates and the hydroxide ions (OH⁻) in the pore water of a concrete (Hobbs 1988; Diamond and Penko 1992). Hydroxide ions in a pore solution result from the hydration of portland cement (Diamond 1983). The pore solution composition depends on the amount of alkalis present in the pore water which, in turn, is related to the amount of soluble alkalis in the concrete. This dissolution leads to a pH in excess of 12.5 (Diamond 1983). The hydroxide ions in solution will attack sites on a silica surface of the aggregates. If the silica is well-crystallized, there are few highly reactive sites, whereas in a poorly crystallized or amorphous silica there are many more such reactive sites. In this case, hydroxide attack may lead to complete conversion of the silica to a calcium and alkali silicate gel (Figg 1983; Helmuth and Stark 1992).

The formation of the gel per se is not deleterious. The deterioration of the concrete structure is usually due to water absorption by the gel and its subsequent expansion. The gel can creep into existing pores or cracks, where it can do relatively little damage. Of course, this scenario is based on the fact that the viscosity of the gel is such that it can flow into the pores and cracks. However, if the void volume is not sufficient to accommodate all the gel, or is not all accessible by the gel, and if the tensile strength of the matrix is locally exceeded, cracks will form and propagate radially from the reactive aggregate. The sites of crack initiation are often randomly distributed in the specimen, with no preferential direction of crack propagation. The points of crack initiation are determined by the location of the reactive silica in the aggregates and the local availability of hydroxide ions. If the relative humidity inside the speci-

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²Deceased.

tion is higher than 80% (Stark 1994), the gel can expand. Therefore, the permeability and the self-desiccation of a concrete specimen play an important role.

Some mineral admixtures have been reported to suppress the deleterious expansion due to ASR (Duchene and Béribé 1994). While the mechanisms are not clearly determined, several possibilities have been proposed:

- reduced permeability that prevents the ingress of water,
- increased strength, probably due to decreased porosity around the aggregates,
- reduced alkali concentrations in the pore solution because of the reduction of cement and/or, the mineral admixture contains less alkali than does the cement it is replacing, and,
- increased pozzolanic reaction, leading to the mineral admixture reacting rapidly with the ions, producing CSH that traps alkali ions and therefore reduces their concentration in the pore solution.

The results of this paper will be used to discuss the validity and relative importance of each of these mechanisms.

Measurements

To measure the axial stress generated by the expansion of a specimen, the following test was developed. Figure 1 shows a sketch of the apparatus. The specimen, a mortar or concrete cylinder, is placed in the stainless steel frame and connected to a load cell. Usu-

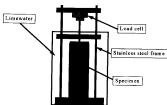


FIG. 1—Schematic of the device used to measure stress due to ASR.

ally some small load is initially placed on the specimen by screwing down bolts set in the frame. The load cell, connected to a computer, monitors the expansive force generated by the specimen in the axial direction only. The specimen is free to expand in the radial direction, subject, of course, to friction at the ends due to the bottom of the frame and the loading disk on the top of the specimen.

If the load cell was infinitely stiff, the sample would be constrained to stay at the prescribed initial strain, which is determined by the load initially placed on the sample. This experiment would then be a pure stress relaxation experiment at a fixed strain level. Because the load cell is not of infinite stiffness, this test is not a pure axial stress relaxation test. It does allow some deformation to take place in the axial direction.

The frame holding the specimen is immersed in a container with linewater. The container is then placed in a water bath with a controlled temperature of $50 \pm 3^\circ\text{C}$. All length and stress measurements reported in this paper were made at this controlled temperature, so that the effects of thermal expansion stresses were eliminated (Ferraris et al. 1995, 1996).

To extract the stresses due to ASR from stresses generated by other phenomena (water absorption, autogenous shrinkage, etc.), the expansions and the stresses generated in comparison samples with non-reactive aggregates were measured. Of course, it is assumed that the stresses due to the ASR are not completely compensated by the stress relaxation of the specimen; otherwise we will observe no stresses at all. All expansions were measured using a digital comparator with a resolution of 0.002 mm (0.0001 in.) with the uncertainty in the measured strain for the size of the specimen used being approximately 0.001%.

Specimen Preparation

Mortars with a low water/cementitious materials (w/c/m) ratio and high cement content were prepared to simulate high-performance concrete. The mixture design is shown in Table 1. A melamine-based high-range-water reducer (HRWR) (4% solid by mass of cement) was used to obtain a usable flow. The sand was prepared by crushing and grading coarse aggregate as described in ASTM Test Method for Potential Alkali Reactivity of Aggregates (Mortar-Bar Method) (C 1260) (non-reactive) or by combining four grades of fused silica sand (reactive) to obtain the specified grading. Consequently, both sets of aggregates had the same size distribution. The only difference between the two sets of samples prepared was in the reactivity: the fused silica aggregates were very reactive (greater than 0.1% expansion at 14 days (ASTM C 1260), and the other ag-

TABLE 1—Mixture design of the mortar specimens tested

Mixture Design	Mix A	Mix B
Water/cement	0.295 by mass	0.338 by mass
Sand/cement	1.411 by mass	1.411 by mass
Type of sand	TECHSIL ² or GENSTAR ³	TECHSIL ² or GENSTAR ³
Sand grading	Grading (Table 2)	Grading (Table 2)
Silica fume	0 or 15% mass replacement	0 or 15% mass replacement
Inert filler	UNASIL 1200 ¹	UNASIL 1200 ¹
	0 or 15% mass replacement	0 or 15% mass replacement
HRWR ⁴	4% by mass of cement	4% by mass of cement

¹ Graded sand provided by C-E Minerals, PA. The composition is fused silica (amorphous).

² Aggregates provided by Genstar, MD. The composition is limestone.

³ High-range water reducer admixture (HRWR), melamine based, was supplied by Master Builders.

TABLE 2.—Gradation of the sand.

Sieve No.	Cumulative Passing, %
8	100
16	63.5
30	43.2
50	22.3
100	2.3

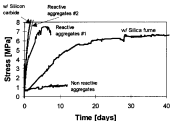


FIG. 2.—Stress versus time measured on the mortar specimens for $w/c = 0.295$. One curve is given for a sample with non-reactive aggregates. All the other samples had highly reactive aggregates. Two plain cement samples are tested since one of them broke in the apparatus (broken specimen is marked #1). Results for silica fume and inert filler cement replacement samples are also given.

gregates were a non-reactive limestone (0.01% expansion at 14 days ASTM C 1260). The fused silica sand was provided in four gradings, namely 4-10, 10-20, 20-50, and 50-100 (sieve size as defined in ASTM E 11, Specification for Wire-Cloth Sieves for Testing Purposes). The silica fume was obtained in a slurry and its characteristics are described in Ferraris et al. (1998). An inert silicon carbide powder was also used as a cement replacement. This powder was UNASIL 1200³, silicon carbide used for polishing. This has been demonstrated to be an inert material in the hydration process.⁴

Cylindrical specimens, 38 mm in diameter and 279 mm long (1.5 by 14 in.), were cast using an ASTM Type I portland cement with a high alkali content (approx. 1.2% Na_2O equivalent by mass). As the high alkali content of the cement was expected to result in a high alkali concentration and pH in the pore solution, this cement was also expected to induce ASR with reactive siliceous aggregates. The high temperature used, $50 \pm 3^\circ\text{C}$, should also have promoted the alkali-silica reaction. After casting, the specimens were cured for 24 h in 100% RH at $20 \pm 5^\circ\text{C}$, and then at $50 \pm 3^\circ\text{C}$ for another 24 h. The stress test was initiated immediately after this second 24-h curing period, which also served to slowly equilibrate specimen temperature, avoiding thermal shock in going from room temperature to the 50°C bath.

³ Brand names and names of manufacturers are identified in this report to adequately describe the experimental procedure. Such an identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the material identified is necessarily the best available for the purpose.

⁴ J. Wernick, private communication.

Results and Discussion

Axial stress and expansion were measured on all specimens. Figures 2 and 3 show the axial stress data obtained at two different w/c ratios, 0.295 and 0.338. Figures 4 and 5 show the results obtained for the expansion of companion specimens that were not confined in the frame.

First consider in Fig. 2 ($w/c = 0.295$) the stress versus time curve for the sample containing nominally non-reactive aggregates. This curve shows only a very small increase in stress level, due to water absorption, as was observed before (Ferraris 1997; Miyazawa et al. 1996). Figure 4 shows the companion expansion versus time curve, showing little expansion. These results confirm that the limestone aggregates were non-reactive under the test conditions.

In Fig. 2, two curves are shown for mortars made with reactive aggregates with no mineral additions. The curve that shows the

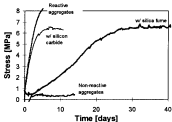


FIG. 3.—Stress measured in the mortar specimens at $w/c = 0.338$. The graph includes mortar with non-reactive aggregates and silica fume, with reactive aggregates, and cement replacement with silica fume or inert powder.

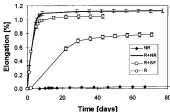


FIG. 4.—Expansion measured on mortar specimens with $w/c = 0.295$. The graph includes mortar with non-reactive aggregates (NR), with reactive aggregates (R), with reactive aggregates and cement replacement with silica fume (R + SF), or inert powder (R + NR). The uncertainty bars represent one standard deviation.

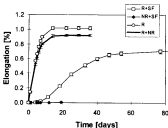


FIG. 5—Expansion measured on mortar specimens with $w/c = 0.50$. The graph includes mortar with non-reactive aggregates and silica fume (NR + SF), with reactive aggregates (R), with reactive aggregates and cement replacement with silica fume (R + SF), or inert powder (R + NR). The uncertainty bars represent one standard deviation.

lowest maximum stress, and even a loss of stress after 5 days, belongs to a specimen that was severely cracked and, therefore, could not bear any further load. The stress measurement was stopped for the other specimen because the load measured exceeded the capacity of the load cell (about 8 MPa). In Fig. 4, the expansion measured for this mixture reached a plateau. Because the expansion is measured on the vertical axis (axis of symmetry of the cylinder), the specimen may continue to expand in the radial direction while the vertical expansion has essentially ceased, thus the plateau in Fig. 4. Therefore, the expansion plateau is considered apparent, rather than an absolute indication that the specimen has ceased expanding.

The stress versus time curves in Fig. 2 for the specimen with silica fume and reactive aggregates shows two phenomena:

- The maximum load and expansion is lower than the mortars without silica fume;
- An equilibrium stress is reached after about 25 days.

This observation signifies that silica fume does mitigate the deleterious response of the concrete to the ASR in two ways: the overall stress level is reduced compared to samples without silica fume, and the stress level equilibrates instead of monotonically increasing. One possible reason for this stress equilibration may be that the amount of dissolved alkalis was depleted. Studies of the pore fluid show that when using silica fume or other pozzolans, alkali concentrations first increase and then decrease, reaching a low equilibrium value after 20 to 30 days. In the absence of silica fume or other pozzolans, the alkali concentration increases continuously over time (Duchesne et al. 1994; Drexel and Feste 1995). In Fig. 2, the stress for the silica fume sample equilibrated after about 25 days. We interpret these results as indicating that the soluble alkali concentrations were reduced to a low value so that the alkali-silica reaction was greatly reduced or stopped. If the reaction cannot continue, the deleterious effects (expansion and stress) would be mitigated. This low value of alkali concentration has been attributed to the formation of hydration products such as C-S-H, which physically or chemically traps the alkali ions (Duchesne et al. 1994).

Mentioned in the "Background" section were three other possible ways silica fume mitigates the deleterious effects of ASR. One was that the lowered permeability induced by the silica fume, which is well-documented in the literature, prevents the ingress of water. Recall that the swelling associated with ASR is thought to be induced by the ASR gel-absorbing water. Of course, a lower permeability will also tend to prevent mix water from escaping by evaporation in air-cured specimens. Since in the present study the specimens were cured and kept in water, it is possible that the low- and permeability of the silica fume specimen did mitigate the ASR expansion by limiting the ingress of water. However, the equilibration of the stress versus time curve for the silica fume specimen might tend to rule against this since the specimen will absorb some water over time if left long enough in the water bath. We plan to let a silica fume sample stay in the load frame in lime water for an extended period of time to study this. It is interesting to note that, as shown in Fig. 4, the expansion of the silica fume specimen also equilibrated and stayed fairly constant for a period of up to 70 days. This finding would tend to rule against the lower permeability hypothesis, as some expansion should have been seen if the ingress of water were only slowed, not stopped. Also, at this w/c ratio, 0.295, even the samples without silica fume have been found to have a low value of permeability, but still showed high values of expansion and stress.

Another possibility for the ASR mitigation effect of silica fume is the strengthening of the material due to the strengthening of the interfacial transition zones. However, the main effect of silica fume is to increase the compressive strength of the material. The tensile strength of the specimen would still have been relatively low, so that the local cracking of the matrix induced by ASR expansion would not have been affected. The increase in elastic modulus reported in the literature for silica fume specimens (Drexel and Feste 1995) would tend to reduce the expansion to some extent by making the specimen stiffer, but not to the extent seen in the data.

The fourth possible explanation for the silica fume mitigation effect on ASR was the reduction of alkali concentrations because of the reduction of cement, as silica fume does not contain any appreciable amount of alkalis. This was tested directly by replacing the cement with an inert filler, silicon carbide, using the same mass percentage replacement as for the silica fume. The induced stress and expansion versus time curves for this scenario are also shown in Figs. 2 and 4. The stress versus time curve was discontinued after a few days, as the load measured exceeded the capacity of the load cell. Up to this point, this curve closely follows that of the sample with no mineral admixtures, which tends to disprove the hypothesis that a reduction in alkalis due to reduced cement content mitigates the ASR stress and expansion.

Figures 3 and 5 contain similar data for a different w/c (water/solid) ratio, 0.338. The same inferences from the data can be drawn as for the $w/c = 0.295$ data, implying that for at least these fairly close values, the w/c ratio used does not appreciably affect the ASR results.

Using SEM imaging, the area fraction of cracks in the aggregates and in the matrix were measured. The specimens were cut perpendicular to the vertical axis and a "disk" was prepared for SEM imaging. Therefore, the cracks that are measured are radial. A point count of SEM backscattered electron images for estimates of area and volume fractions of selected components was made. Fifty-six fields from the approximate center of each specimen were examined at a magnification of $\times 200$, allowing recognition of cracks of less than 1 μm in width. The point count grid of 25 points provided a point spacing of about 100 μm , with a total of about 1400 points

being counted for each specimen. Table 3 shows the results obtained. The error estimates indicate that the specimen with silica fume was different than the other two specimens. The difference appears to lie in the lower volume of cracking within the aggregates of the silica fume specimen. The volume of paste cracking appears to be the same within the experimental uncertainty from that of the other two specimens.

Figures 6 and 7 show the SEM image of the cracks in a specimen with no mineral addition and a specimen with silica fume addition, respectively. The A part of both figures shows the aggregates (light

TABLE 3—Estimated relative crack area in the specimens. The uncertainties are one standard deviation.

Sample	Total	Aggregate	Paste
No additions	14.5 $\pm$ 1.9	10.1 $\pm$ 1.6	4.4 $\pm$ 1.1
Silicon carbide	17.1 $\pm$ 2.0	12.4 $\pm$ 1.8	4.7 $\pm$ 1.1
Silica fume	9.0 $\pm$ 1.5	5.3 $\pm$ 1.2	3.7 $\pm$ 1.0

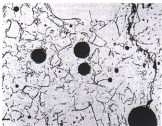
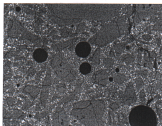


FIG. 6—SEM picture of crack pattern for a specimen with no additions. (A) SEM picture in normal light and showing the aggregates in light gray; (B) the same picture but with the cracks and pores highlighted in black, with everything else, paste and aggregates being white.

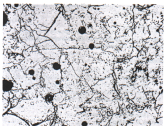
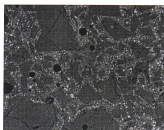


FIG. 7—SEM picture of crack pattern for a specimen with silica fume additions. (A) SEM picture in normal light and showing the aggregates in light gray; (B) the same picture but with the cracks and pores highlighted in black, with everything else, paste and aggregates, being white.

gray) and the paste, while the B part of both figures shows the cracks in black and the solid material in white. As indicated in Table 3, the reduction of cracks in the aggregates for the specimen with the silica fume addition can be seen. Since ASR usually involves gel formation and cracking of the aggregates, leading to cracking in the paste, we tentatively conclude that the silica fume specimen has less reaction in the aggregates, leading to less cracking in the aggregates and less overall expansion.

Conclusion

The work reported in this paper is part of an ongoing program to investigate ASR at a fundamental level by measuring the generated stress rather than just the sample expansion. However, the maximum stress that can be measured with the load cell available to us is about 8 MPa for this size specimen. Therefore, we cannot determine if the stress will reach an equilibrium value for the samples without silica fume. We are planning to install a higher capacity load cell to further investigate this behavior. More SEM observations will be made to study the crack patterns and matrix composition. In addition, we will also study, using a combination of exper-

imental and modeling techniques, the fundamental mechanisms involved in ASR deterioration.

Silica fume, because of its amorphous nature and large surface area, reacts rapidly with the alkalis in the pore solution, forming C-S-H by reacting with the calcium and hydroxide ions. This is essentially ASR. Why does this reaction not cause expansion? With aggregates, the reaction is slower due to the reduced surface area per volume and more concentrated, thus promoting the buildup of higher stresses and consequent cracking. Therefore, the particle size distribution of the aggregates should be investigated.

While the addition of silica fume as a replacement of cement reduces the stresses and the expansion generated by the ASR reaction, expansion and stress are not completely eliminated, at least at the replacement dosage used of 15% by mass (a dosage that is relatively high with respect to common practice). Due to confining reaction of the aggregates in the specimens containing silica fume, the number of cracks is reduced but not eliminated. Future work is planned to determine the fundamental mechanisms of silica fume mitigation of the ASR and the potential effect of the residual expansion and cracking.

Acknowledgments

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