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Effectiveness of mineral additives in mitigating alkali-silica reaction in mortar

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Abstract. The effectiveness of mineral additives in suppressing alkali-silica reactivity has been studied in this work. Experimentation has been performed in accordance with the procedures prescribed in ASTM C 1567. In the scope of the investigation, a quarry aggregate which was reactive according to ASTM C 1260 was tested.

In the experimental program, prismatic mortar specimens measuring 25×25×285 mm were produced. Ten sets of production, three specimens for each set, were made. Length changes were measured at the end of 3, 7, 14 and 28 days and then expansions in percentage have been calculated. Fly ash, silica fume, and metakaolin have been used as cement replacement in different ratios for the testing of the alkali-silicate reactivity of the aggregate. In the mixes performed, the replacement ratios were 20%, 40%, and 60% for the fly ash, and 5%, 10%, and 15% for the silica fume, and 5%, 10%, and 15% for the metakaolin. Mixes without mineral additives were also produced for comparison.

The beneficial effect in suppressing alkali-silica reactivity is highly noticeable as the replacement ratios of the mineral additives increase regardless of the type of the mineral additive used. Being more concise, the optimum concentrations of using silica fume and metakaolin in mortar in suppressing ASR is 10%, respectively, while it is 20% for fly ash.

Keywords: alkali-silica reactivity; mortar; sodium hydroxide; mineral additives; expansion

1. Introduction

Alkali-silica reaction (ASR) is one of the chief concerns associated with the deterioration of concrete structures in many countries, notably North America, Scandinavia, India, Australia and New Zealand and was first recognized by Stanton (Stanton 1940, Lane 1994, State of the art ACI 1998). ASR occurs between the alkalis from mostly cement and reactive constituents from the aggregate like amorphous opal, cherts, chalcedony, kuvars (Ramachandran 1998). Although the mechanism of ASR has not been fully understood, there is a unanimous agreement on the factors affecting ASR such as the quantity of available alkali, the nature and the content of the reactive silica, the porosity of concrete, particle size and the amount of available moisture in the surrounding environment (Mindess *et al.* 1981, Helmuth 1993, Baingam *et al.* 2015, Zahira and Aissa, 2015).

Alkali-silica gel forms in concrete owing to this reaction. The amount of silica gel that accumulates at the aggregate-cement paste interface depends basically on the type and the quantity of reactive silica, and the alkali hydroxyl ion concentration. The volume of the alkali-silica gel increases over time by imbibing moisture in the surrounding environment. Mortar expands depending on the volume increase inducing tensile stresses in concrete that lead to concrete cracking (Neville 1991). The mechanism of ASR takes place in two sequential steps as follows:

Step 1: Reactive silica + Alkalis → Alkali-silica gel (sodium silicate)

$SiO_2 + 2NaOH + H_2O \rightarrow Na_2SiO_3 \times 2H_2O$ (2KOH can replace 2NaOH)

Step 2: Alkali-silica gel + Moisture → Expansion

In the absence of any one of the factors affecting ASR will effectively suppress ASR-induced damage.

The formation of tensile stresses in concrete does not occur simultaneously with gel formation in as indicated in Step 1. That is to say, the presence of silica gel does not actually mean that serious ASR-induced damage will occur. For instance, ASR damage doesn’t occur at all as long as the concrete remains dry. The ingress and the movement of moisture in concrete will be difficult when the permeability of concrete is low. Temperature is the most important accelerating factor of ASR since most of the aggregates become more reactive at higher temperatures. Due to wetting-drying, the migration of the alkalis in concrete speeds up and they accumulate in dry areas of concrete (Neville 1997).

Mitigating or suppressing the ASR-induced damage in concrete can be achieved by: i) using non-reactive aggregates; ii) minimizing alkalis; iii) limiting moisture; iv) using mineral additives; v) using lithium compounds (Mindess *et al.* 1981).

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The aim of this study is to evaluate and compare the effectiveness of using fly ash, silica fume and metakaolin at certain ratios in controlling alkali-silica reactivity in concrete produced with a potentially reactive aggregate.

1.1 Mitigation of ASR with mineral additives

The importance of mineral additives in mitigating ASR in concrete has been a subject for extensive research for a long time. A variety of mineral additives such as fly ash, ground granulated blast furnace slag, silica fume, calcined clay have been incorporated in concrete as an ingredient (Duchesne and Berube 1994a, Duchesne and Berube 1994b, Bagel 1998, Shehata and Thomas 2000, Shehata 2001, Shehata and Thomas 2002, Shehata and Thomas 2006, Thomas *et al.* 2011, Shafaatian *et al.* 2013, Kandasamy and Shehata 2014, Afshinnia and Rangaraju 2015, Vayghan *et al.* 2016). It has been reported in these studies that in addition to their suppressing effect on ASR-induced expansion, mineral additives have beneficial effects on the pore structure and permeability of concrete as well. This is because more hydration products forming due to mineral additives reduce the total volume of pores in the mortar phase of concrete (Dongxue *et al.* 1997). The various proposed mechanisms of control ASR expansion can be summarized as follows. i) The permeability of the cement paste is decreased and consequently the mobility of ions in concrete is reduced as a result of pozzolanic reaction between mineral admixtures and cement hydrates. ii) The improved strength developed by the mineral additives provides higher resistance to the expansive stresses produced by ASR. iii) Mineral additives reduce the alkalinity of the pore solution by consuming the hydration product of calcium hydroxide. iv) Mineral additives deplete $Ca(OH)_2$ in the cement paste. v) The secondary hydrate produced by pozzolanic reaction entraps alkali ions (Neville 1991, Duchesne and Berube 1994b, Xu *et al.* 1995, Detwiler 1997, Shafatian *et al.* 2013, Ansah *et al.* 2014, Latifee 2016, Abbas *et al.* 2017).

Pozzolans reduce and/or eliminate expansion due to ASR by producing additional C-S-H which can retain additional alkali; thus, reducing the amount of alkali available for reaction with reactive aggregate (Bhatty 1985). However, the mechanism by which pozzolans control expansion is not merely the effect of dilution of the alkalis (Gaze and Nixon 1983). Small additions of silica fume can delay the onset of ASR by reducing the rates of ions transports (Chatterji *et al.* 1986). Various researchers have reported opposing results on the effectiveness of mineral additives mainly because of the wide range of available mineral additives with different levels of alkali contents and the different properties and reactivity of aggregates being investigated (Shayan *et al.* 1996).

The most commonly used mitigation measure for ASR is to control the amount of alkali in the concrete for the purpose of reducing the hydroxyl ion concentration and eventually reducing the pH of the concrete. Controlling the amount of alkali of cement has been proven to decrease ASR-induced damage. Over the years, the 0.60% Na_2Oeq limit in the cement has been proven to be very effective in obstructing concrete damage due to ASR (ACI 1998).

Table 1 Chemical composition, physical and mechanical properties of cement

Chemical Composition		Physical and Mechanical Properties		
Components	(%)	Retained on sieve 45 μm (%)		9.8
SiO_2	19.46	Retained on sieve 90 μm (%)		1.0
Al_2O_3	5.11	Specific surface (Blaine) (m^2/kg)		412.6
Fe_2O_3	3.31	Specific gravity (g/cm^3)		3.12
CaO	60.23	Setting Times (Vicat) (min.)	Initial	140
MgO	2.08		Final	200
SO_3	3.05	Water Demand (%)		29.2
Na_2O	0.27	Soundness (mm)		1.0
K_2O	0.69	Compressive Strength (MPa)	2 days	28.0
Cl^-	0.02		7 days	40.4
Loss on ignition	3.00		28 days	51.5

Table 2 The physical properties of the aggregate used

Physical properties	Content
Dry particle density (Mg/m^3)	2.79
SSD particle density (Mg/m^3)	2.91
Apparent particle density (Mg/m^3)	3.17
Water absorption, by mass (%)	4.24

Table 3 Physical and chemical properties of fly ash, silica fume, and metakaolin

	Fly ash	Silica fume	Metakaolin
Density (g/cm^3)	2.34	2.20	2.79
SiO_2 (%)	61.57	93.00	54.93
Al_2O_3 (%)	20.25	0.58	41.00
Fe_2O_3 (%)	7.00	2.79	2.00
CaO (%)	1.54	0.60	0.04
MgO (%)	-	1.00	0.06
Na_2O (%)	-	1.00	0.06
K_2O (%)	-	0.10	0.00
SO_3 (%)	-	0.50	0.21
Moisture content	-	0.10	0.03
Loss on ignition	-	0.50	0.21

Pozzolans also contribute to the mitigation of ASR by decreasing the pH of the lime in cement paste and by decreasing the solubility of silicate in concrete (Hill 1996). Another advantage of using pozzolans in concrete in relation with ASR is that they reduce the permeability of concrete significantly (Afshinnia and Rangaraju 2015).

In a study carried out for determining the effectiveness of using metakaolin on ASR, it has been accentuated that the expansion due to ASR decreases greatly by the increase of metakaolin replacement in concrete mix (Ramlochan *et al.* 2000). It has also been stated in the study that the optimum ratio for the metakaolin in controlling the expansion due to alkali-silica reaction is between 10% and 15%.

In a study carried out for reducing ASR by using a reactive aggregate powder, it has been pointed out that the expansion due to ASR was reduced by 35% to 90% (Cyr *et al.* 2009). This shows how effectively ASR has been taken under control by using siliceous and calcareous powder. In another study, it has been stated that using amorphous silicate as mineral additive reduces the expansion resulting from ASR in concrete by 120% (Davraz and Gündüz 2008).

Among the techniques used for determining the effectiveness of using mineral additives in controlling the ASR in concrete, ASTM C 1567 Accelerated Mortar Bar Method and ASTM C 1293 Concrete Prism Test Method have been reported to be the most reliable methods.

2. Experimental Setup

2.1 Materials

In this work, the effects of fly ash, silica fume, and metakaolin on ASR has been investigated. For this purpose, a quarry aggregate was studied. Concerning the alkali-silica reactivity of the aggregate determined in a preliminary testing, it is considered to be reactive according to the criteria prescribed in ASTM C 1260. The cement used in the experimental program was CEM I 42.5 R type with an equivalent Na_2O content of 0.72%, manufactured by Trabzon Cement Factory which is a local cement factory. The fly ash was provided by Zonguldak-Çatalağzı power generation plant. The silica fume, along with the metakaolin was provided by private companies. The sodium hydroxide (NaOH) used for the alkali-silica reactivity testing was provided by a private laboratory. The properties of the materials used in the experimental program are given in Tables 1 to 3.

2.2 Methodology

ASTM C 1260 was followed for the mixing, preparation, and the expansion calculation procedures of the mortar bars. The expansion or the length change of the mortar bars due to ASR was measured at certain intervals. The method prescribed in ASTM C 1260 was followed as it is a quick way of determining the potential reactivity of the aggregates in the mortar mix.

The gradation of the aggregates, determined in accordance to ASTM C 1260 guidelines, is given in Table 4. By keeping the water to cement ratio constant as 0.47, the mortar mixes were prepared so as to have 990 grams of aggregate, 440 grams of cement, and 206.8 grams of water for each casting. Three mortar bars, measuring 25x25x285 mm, were prepared for each casting. After 24 hours of casting, the initial comparatory readings of the mortar bars were recorded and then the mortar bars were numbered. Following the initial readings, the mortar bars were placed in a container with sufficient tap water at 20ºC. The containers were then placed in a water bath at 80ºC for a period of 24 hours and then the zero readings (L_o) for each mortar bar were taken. After zero readings, the mortar bars were placed in steel containers with sufficient 1N NaOH solution at 80ºC throughout the experimentation. To determine the expansion due to ASR, the length of the mortar bars were measured and recorded at the end of 3, 7, and 14 days of exposure in 1N NaOH solution, which is a mixture of 40 grams of sodium hydroxide and 900 ml of water.

To accomplish the objective of the study, along with the control mix a total of 10 mixes were produced using the aggregate with the gradation given in Table 4. Fly ash, silica fume, and metakaolin were used. For the mixes produced, the adopted replacement ratios were 20%, 40%, and 60% for the fly ash, 5%, 10%, and 15% for the silica fume, and 5%, 10%, and 15% for the metakaolin. Type F fly ash,

Table 4 The gradation of the aggregate used with ASTM C1260

	Sieves				
	2.36 mm (No.8)	1.18 mm (No.16)	600 μm (No.30)	300 μm (No.50)	150 μm (No.100)
	%10	%25	%25	%25	%15
Weight (%)	10	25	25	25	15
Weight (grams)	99	247.5	247.5	247.5	148.5

Table 5 Mix proportions for three mortar bars (in grams)

Mixes	Cement	Aggregate	Mineral additive
FA20	352	990	88
FA40	264	990	176
FA60	176	990	264
SF5	418	990	22
SF10	396	990	44
SF15	374	990	66
MK5	418	990	22
MK10	396	990	44
MK15	374	990	66

Table 6 Interpretation of the alkali-silica reactivity measurements with ASTM C1260

Expansion (%)	Interpretation
< 0.10	It indicates that the aggregate is innocuous.
between 0.10 and 0.20	It indicates that the aggregate is potentially reactive.
> 0.20	It indicates that the aggregate is reactive.

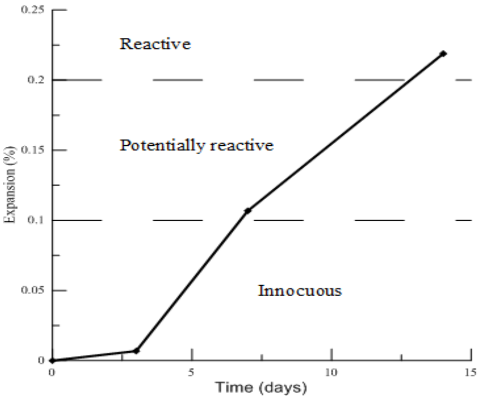

Fig. 1 Expansion-exposure time relationship for the quarry aggregate

Fig. 2 Mortar bars measuring 25×25×285 mm

produced from bituminous coal which includes a total amount of silicon dioxide (SiO_2) plus aluminum oxide (Al_2O_3) plus iron oxide (Fe_2O_3) of 88% is used in the study. The proportions used for the mixes of the mortar bars, measuring 25×25×285 mm, are given in Table 5. Abbreviations were used for the mineral additives as FA for the fly ash, SF for the silica fume and MK for the metakaolin. For example, FA20 indicates that the substitute ratio for the fly ash is 20%.

The interpretation of the alkali-silica measurements of the mortar bars at the end of 14 days of exposure in 1N NaOH solution was based on the criteria provided by ASTM C 1260 as given in Table 6.

3. Results and discussion

3.1 Preliminary work

A preliminary testing was performed for the aggregate used in the study to determine whether it is alkali-reactive or not. The testing has been performed in accordance with the procedures prescribed in ASTM C 1260. The alkali-silica reactivity measurements obtained for the aggregate are illustrated in Fig. 1. Considering Fig. 1, it can be seen that the aggregate is reactive in relation with the criteria given by ASTM C 1260, regarding the measurements obtained at the end of 14 days of exposure in 1N NaOH solution. The molds used in the experimentation along with the prismatic mortar bars are illustrated in Fig. 2.

3.2 Reactivity measurements

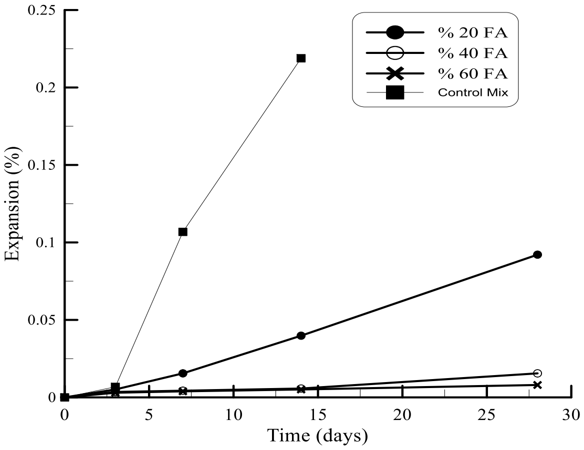

Fig. 3 Alkali-silica expansion of the mortar bars containing fly ash

Fig. 3 illustrates the relationship of the expansion of the mortar bars due to ASR with exposure time produced using fly ash at different ratios. In the mixes produced, the replacement ratios of the fly ash were as 20%, 40% and 60%. As can be seen from the figure, the effect of fly ash on the alkali-silica reactivity of the mortar bars changes depending on the replacement ratio of the fly ash. In other words, it can be seen that as the replacement ratio of the fly ash in the mixes increases the expansion decreases noticeably. At the end of the 14 days of exposure, the expansion of mortar bars with the replacement ratio of 20% fly ash decreased by approximately 82% compared to that of the mortar bars produced without fly ash. Similarly, the expansion measured for the mortar bars produced with a substitution ratio of 40% fly ash decreased by 97% and the decreasing in the expansion was about 98% for those of produced with a replacement ratio of 60% fly ash. This finding is quite compatible with the study carried out for determining the effectiveness of using fly ash in concrete as it has been stated that 20-30% F type fly ash had indicated an adequate mitigating effect on controlling ASR (Thomas 1996). An expansion level of 0.040% was measured for the mortar bars containing 20% fly ash. In a study (Shafatian *et al.* 2013), the expansion level measured for mortar bars containing 20% fly ash is 0.03%, which is quite compatible with the value obtained in present study. The expansions were 0.006% and 0.005% for 40% and %60 fly ash replacement ratios, respectively. It is quite obvious that the expansion due to ASR decreases as the fly ash replacement ratio increases.

Fig. 4 demonstrates the time dependent expansion of the mortar bars due to ASR depending on the replacement ratio of the silica fume. As can be seen from the figure, the silica fume has a beneficial effect in suppressing the adverse effects of the ASR. The expansion measured for the bars with the replacement ratio of 5% silica fume is approximately 0.16% at the end of 14 days of exposure. This indicates that there is a reduction of 25% in the expansion of the mortar bars compared to those without silica fume. However, a replacement ratio of 5% silica fume seems to be not sufficient for bringing the expansion level caused by the alkali-silica reactivity below 0.10% which is the limit value for the aggregate being accepted as innocuous according to ASTM C 1260 (see Table 6). However, there is a noticeable reduction in the alkali-silica reactivity of the mortar bars in the case of 10% and 15% silica fume replacement. This means that the aggregate used becomes innocuous in relation with the alkali-silica reactivity for replacement ratios of 10% and 15% of silica fume. This is quite compatible with the findings obtained by the work (Aquino *et al.* 2001), as indicated that silica fume showed a superior performance in reducing expansion due to ASR. The reduction in expansion of the mortar bars containing 10% silica fume is about 69%, while it is 71% in the present study, both are quite close to each other.

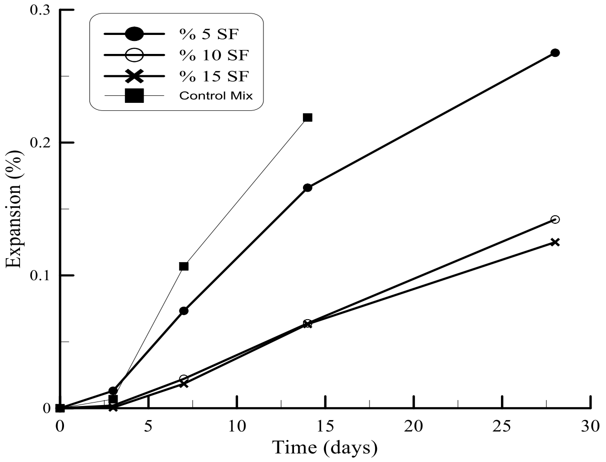

Fig. 4 Alkali-silica expansion of the mortar bars containing silica fume

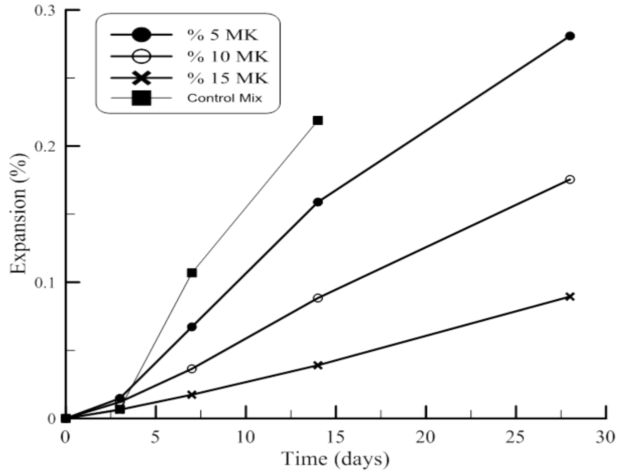

Fig. 5 Alkali-silica expansion of the mortar bars containing metakaolin

Fig. 5 illustrates the correlation between the expansion of the mortar bars produced with metakaolin at different ratios and the exposure time. As seen from the figure, the replacement ratios of 10% and 15% metakaolin seems to be sufficient for bringing the expansion due to ASR below 0.10% which is the limit value for the aggregate being accepted innocuous according to ASTM C 1260 (see Table 6). For an exposure period of 14 days in 1N NaOH solution, a reduction of 28% in the expansion of the mortar bars due to ASR is obtained for a replacement ratio of 5% metakaolin. The reduction in the alkali-silica reactivity is 60% for the mortar bars containing metakaolin with a replacement ratio of 10% and it is 82% for a replacement ratio of 15% metakaolin. In relation with the effectiveness of using metakaolin in suppressing the ASR, it has been reported elsewhere (Ramlochan *et al.* 2000), the optimum ratio for metakaolin in controlling the expansion due to ASR is approximately 10% and 15%. It has been stated elsewhere (Moser *et al.* 2010), that a reduction in the expansion of the mortar bars containing 8% metakaolin is 65%, while the reduction is about 85% for the mortar bars containing 15% metakaolin. These results are quite close to the measurements obtained from the present study.

Inspection of Figs. 4 and 5, it can be observed that silica fume and metakaolin have yielded fairly the same beneficial effects concerning the alkali-silica reactivity when both are used at the same ratios.

5. Conclusions

The alkali-silica reactivity measurements obtained from this investigation are limited to the experimental conditions in which the tests were performed. Due to the results of this work, the main conclusions obtained are given below:

- The expansion obtained for the mortar bars at the end of 14 days of exposure in 1N NaOH solution is approximately 0.22% which indicates that the quarry aggregate used in the study is reactive according to the criteria provided by ASTM C 1260.
- An expansion level of 0.040% was measured for the mortar bars containing fly ash at a substitution ratio of 20%. The expansions were 0.006% and 0.005% for 40% and %60 fly ash replacement ratios, respectively. Therefore, it is quite obvious that a replacement ratio of 20% of fly ash is sufficient in controlling ASR expansion.
- In case cement was substituted by silica fume at 5%, an expansion of 0.166% was measured for the mortar bars. The expansions measured were 0.064% and 0.063% for replacement ratios of 10% and 15% silica fume, respectively. Hence, it can be seen that a replacement ratio of 10% silica fume seems to be enough to control ASR-induced expansion.
- An expansion of 0.159% was measured for the quarry aggregate with metakaolin for a replacement ratio of 5%. The expansions due to ASR measured were 0.088% and 0.039% for the replacement ratios of 10% and 15% metakaolin, respectively. This clearly indicates that a replacement ratio of 10% metakaolin seems to be sufficient in controlling ASR expansion.
- Consequently, silica fume and metakaolin yielded almost equitable beneficial effects on mitigating alkali-silica-reaction with a substation ratio of 10% based on the measurements obtained from this study. As the replacement ratio increases, the beneficial effect of fly ash in controlling ASR has been more pronounced as compared to the beneficial effect of the silica fume and metakaolin provided at used ratios in this study.

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