EFFECTS OF ALUMINIUM ADDITION ON INHIBITION OF CONCRETE EXPANSION RESULTED FROM ALKALI SILICA REACTION (ASR)

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ABSTRACT: Alkali-silica reaction (ASR) is one of the major causes of expansion, which ultimately leads to failure in concrete. When reactive concrete is subjected to humidity, alkali silicate hydrate gel (ASR gel) is formed. The swollen ASR gel exerts pressure and creates cracks in the concrete structure. This study, thus, aimed at inhibiting concrete expansion which was caused by the ASR reaction, by creating pores. To achieve pore formation, aluminium powder was used as an air entrainment agent. The mortar bars tested with aluminium content of 0.05, 0.1, 0.15 and 0.2 weight percent of cement were tested according to the ASTM C1260: Standard Test Method for Potential Alkali Reactivity of Aggregates. The results indicated that a larger quantity of entrained pores was achieved at higher aluminium content. The results also showed that when tested according to the ASTM C1260, concrete expansion was reduced as the aluminium content was increased. Microstructural analysis revealed that the entrained pores accommodated spaces for ASR gel, resulting in the decline of concrete expansion.

Keywords: Alkaline-silica reaction, Porous concrete, Aluminum powder, Air entrainment agent

1. INTRODUCTION

Alkali-silica reaction (ASR) is one of the factors causing failure in concrete. ASR involves reactions between alkalies such as Na⁺, K⁺, OH⁻ in cement and silica in aggregates, in the presence of humidity and within temperatures ranging from 30 to 40°C [1]-[2]. The ASR reactions can be expressed according to the following “Eq. (1)-(2)” [3]-[6]:

\[
\begin{align*}
\text{Na}^+ + \text{SiO}_2 + \text{OH}^- & \rightarrow \text{Na}^+ - \text{Si} - \text{H}_2 \quad (1) \\
\text{K}^+ + \text{SiO}_2 + \text{OH}^- & \rightarrow \text{K}^+ - \text{Si} - \text{H}_2 \quad (2)
\end{align*}
\]

The resultant product from ASR is alkali silicate hydrate gel or ASR gel. When subjected to humidity, the ASR gel absorbs water and becomes swollen, causing internal pressure, and ultimately, cracks in concrete [7].

In an attempt to eliminate the failure caused by ASR, effects of adding silica fume, slag, fly ash, meta-kaolin and lithium compounds in concrete and the inhibitive effect on ASR expansion were examined. It has been reported that additives could result in a decline of concrete expansion [8]-[14]. Formation of pores was also reported to inhibit cracks associated with ASR [15]. A number of techniques can be employed to create pores in concrete. One of simplest and the most effective techniques is to add air entrainment agents such as aluminium powder [16]-[19], metallic silicon [20]-[21] and hydrogen peroxide [22]-[23].

This study, thus, aimed at inhibiting concrete expansion caused by the ASR reaction, by creating pores formed by using aluminium powder as an air entrainment agent. Microstructure as well as relationships between aluminium content, apparent porosity and expansion of the mortar bars were also examined.

2. MATERIALS AND METHOD

2.1 Sample preparation

Portland cement Type I was used in casting of mortar specimens, while aluminium powder (Inter-education Supplies co. Ltd.), with amounts of 0.05%, 0.1%, 0.15% and 0.2% by weight of cement, as an air entrainment agent and limestone aggregate from Chaloem Phra Kiat, Saraburi, Thailand. The fine aggregate and cement was proportioned according to ASTM C 1260 standard: Standard Test Method for Potential Alkali Reactivity of Aggregates (Mortar-Bar Method). Water to cement ratio of 0.47 by weight was employed in this study. The mortar specimens were fabricated by mixing the cement, fine aggregates, aluminium powder and water together according to ASTM C305 standard: Standard Practice for Mechanical Mixing of Hydraulic Cement Pastes and Mortars of Plastic
Consistency [24], and then cast into mortar bars with dimensions of 25mm x 25mm x 285mm (1 in x 1 in x 1.25 in).

2.2 Characterization

Chemical composition of limestone and cement were analyzed by x-ray fluorescence (XRF) technique (Bruker, S1 Turbo SD LE) and x-ray diffraction (XRD) technique (Philips, Rigaku TTRAX III), respectively. To identify gel products resulted from alkali silica reaction (ASR), scanning electron microscope (SEM) (FEI Quanta 450) in the backscatter (BSE) mode as well as energy dispersive spectrometry (EDS) (Oxford X-Max) were employed.

The expansion of mortar bars was performed according to ASTM C1260, involving measuring the length of the mortar bars submerged in 1M NaOH solution at 80°C for 28 days. Porosity and pore morphology in the mortar bars were examined using image analysis technique. The samples used in the image analysis were prepared according to ASTM C1260, cut into bars with dimension 25mm x 25mm x 10mm, and polished with SiC paper with with the following grit sizes: 120, 240, 320, 400 and 600. The lapped surface of the specimens then was colored black using permanent black ink. The specimens were heated to 60°C and a paste of alumina powder and petroleum jelly was applied to the surface with a rubber spatula. The paste melted on the surface and flowed into the voids. After cooling to room temperature the excess paste was removed using straight sharp blade, leaving the solid surface black and the voids filled with white paste. The porosity of the specimens was analyzed following procedure c: contrast enhanced method of ASTM C457 standard.

3. RESULTS AND DISCUSSION

3.1 Chemical composition of raw materials

Since the problem associated with alkali-silica reaction (ASR) is mainly attributed to alkali-containing compounds in cement and silica-containing compounds in aggregates. Content of alkali in cement were examined. Results from x-ray fluorescence (XRF) revealed that the cement used in this study had 0.7% alkali-containing compounds by weight, expressed in terms of Na₂O equivalent. The alkali equivalent content was close to the acceptable range for Portland cement type 1, according to ASTM C150 [25].

Phase identification of compounds in limestone was performed using x-ray diffraction (XRD) technique. The XRD results, shown in Figure 1, indicated the presence of phases corresponding to Ca(CO₃)₂ (JCPDF 01-083-0578), magnesium silicate hydrate (M-S-H) (JCPDF 00-052-1558) and quartz (JCPDF 00-033-1161). Attributed to the presence of quartz, the results suggested that the limestone used in the present study was capable of triggering alkali silica reaction (ASR) shown in equations 1 or 2.

![Fig.1 XRD patterns from limestone used as aggregate](image)

3.2 Effects of aluminium concentration on porosity of mortar bars

Image analysis software was used to analyze the cross sections of the mortar bars and area fraction analysis was employed to determine the porosity of the specimens. Total porosity, which took into account pores in all sizes, was shown in Table 1. It was found that total porosity increased with increasing aluminium content. Total porosities of 1.24%, 2.9%, 3.31%, 3.61% and 4.82% were observed in the mortar specimen with aluminium content of 0.0 wt%, 0.05 wt%, 0.1 wt%, 0.15 wt% and 0.2 wt%, respectively.

In general, voids or pores contained in concrete structure were mainly categorized into capillary voids, entrapped air voids, and entrained air voids. Since the present study aimed to examine roles of aluminium as an air entraining agent, the focus was mainly on entrained air voids which were smaller than 200 micrometers in size in concrete [26]. Voids smaller than 5 micrometers were considered capillary voids and voids larger than 200 micrometers were considered entrapped air voids. These two types of voids were ignored when considering the area fraction of entrained air voids. Results from the analysis revealed the entrained porosities of 0.1%, 1.14%, 1.92%, 2.16% and 3.56% in the mortar bars with 0.0 wt%, 0.05 wt%, 0.1 wt%, 0.15 wt% and 0.2 wt% aluminium powder respectively. The results indicated that higher quantity of entrained voids were observed as the amount of aluminium increased.
Table 1 The total porosity of mortar bars with various concentration of aluminium

| Aluminium content (wt%) | Total Porosity (%) |
|-------------------------|--------------------|
| 0                       | 1.24               |
| 0.05                    | 2.90               |
| 0.1                     | 3.31               |
| 0.15                    | 3.61               |
| 0.2                     | 4.82               |

3.3 Effects of aluminium concentration on expansion of mortar bars

In this study, the mortar bars expansion test was conducted according to ASTM C1260 where the mortar bars were exposed to 1M NaOH solution at 80°C and the change in length of the mortar bars were measured, at regular interval up to 28 days, using a length comparator. Average expansion of mortar bars after 14 days exposure were then calculated. According to the ASTM standard, the bars with expansion at 14 days exposure of less than 0.1% are considered non-ASR-reactive, while the ones with expansion of more than 0.2% are considered ASR-reactive. For the mortar bars with expansion in the range between 0.1% and 0.2%, it is useful to look at expansion after 28 days and further investigation, such as petrographic study, is required to make more accurate prediction of the aggregate’s reactivity [27].

In the present study, the expansion at 14 days of exposure to NaOH solution for the mortar bars without aluminium addition was 0.23%, which was higher than the criteria for reactivity according to ASTM C 1260. Therefore, this aggregate could be considered as an ASR-reactive aggregate. As shown in Figure 3, average 14-days expansion of 0.18%, 0.18%, 0.16%, and 0.15% was observed in the mortar bars with 0.05 wt%, 0.1 wt%, 0.15 wt%, and 0.2 wt% aluminium addition, respectively. These results demonstrated that expansion of the mortar bars was reduced with the addition of aluminium powder, as shown in Figure 4. Expansion results of mortar bars at 28 days of exposure were found to be 0.39%, 0.37%, 0.31%, and 0.32% for mortar bars with 0.05 wt%, 0.1 wt%, 0.15 wt%, and 0.2 wt% aluminium addition respectively, as shown in Figure 5. It could be seen that even at longer exposure time to NaOH at 28 days, the aluminium addition was still effective in reducing ASR expansion.

Fig. 2 Relationship between aluminium content and ratio of entrained air void to total air void

Fig. 3 Expansion in mortar bars with 0, 0.05, 0.1, 0.15 and 0.2 wt% Al, tested under the conditions according to ASTM C1260

Fig. 4 Relationship between aluminium content and expansion of mortar bars (tested at 14 days)

Fig. 5 Relationship between aluminium content and expansion of mortar bars (tested at 28 days)
3.4 Microstructural analysis of the mortar bars

The mortar bars that underwent the expansion test were analyzed by scanning electron microscope (SEM). To identify ASR gel, backscatter electron (BSE) and energy dispersive Spectrometry (EDS) detectors were employed. BSE mode generally provides the information related to average atomic number (Z).

It is generally accepted that the specimen with a higher average of atomic number appears brighter. A number of studies, including Rajabipour et al. (2013), Oyan et al. (2013), and Nicholas B. (2012), reported the usage of the BSE mode in identifying the areas with possibilities of finding ASR gel. According to the studies, with a low average atomic number, ASR gel would appear as darker areas. On the other hand, cement paste, with a higher average atomic number, would appear as lighter areas [28]-[30].

Another approach to potentially identifying the ASR gel was to determine ratios of alkali-to-silicon and calcium-to-silicon, using EDS technique. According to Knudsen (1975), Struble (1979), Diamond (2000), [31]-[33] ASR gel tended to have an atomic ratio of Na,K/Si of more than 0.1, while that of Ca/Si less than 1.3. Whereas, in cement paste, the atomic ratio of Ca/Si tends to be larger than 1.3. Aggregates commonly contain Na,K/Si of less than 0.1, as shown in Table 2.

Table 2. Criteria to identify ASR gel and other concrete constituents, according to a compositional analysis obtained from the EDS technique

| Atomic ratio | Materials   | Ref.       |
|--------------|-------------|------------|
| Na,K/Si      | Ca/Si       |            |
| > 0.1        | < 1.3       | ASR gel    | [31-33]     |
| > 1.3        | < 1.3       | Cement paste| [31-33]     |
| < 0.1        | > 1.3       | Aggregate | [31-33]     |

From both approaches, potential locations of ASR gel were identified. As shown in Figure 6, which represented the mortar bar without aluminum addition, ASR gel potentially dispersed throughout cement paste.

The same approaches were also employed to examine mortar bars with aluminum addition. As shown in Figures 7, which represented the mortar bars with 0.1 wt% Al addition, ASR gel tended to form inside and surrounding the pores as well as within cement paste.

![Fig.6 SEM micrograph of mortar bar without aluminum addition](image)

| Area | Atomic ratio | Materials   |
|------|--------------|-------------|
|      | Na,K/Si     | Ca/Si       |
| 1    | 0.19         | 0.2         | ASR Gel     |
| 2    | 0.26         | 1.46        | Cement paste|
| 3    | 0.28         | 0.26        | ASR Gel     |

Table 3. Compositional analysis of the area in Figure 6

![Fig.7 SEM micrograph of mortar bar with aluminum 0.1 wt%](image)
Table 4. Compositional analysis of the area in Figure 7

| Area | Atomic ratio | Materials |
|------|--------------|-----------|
|      | Na.K/Si | Ca/Si |          |
| 1    | 6.10  | 0.17  | ASR Gel |
| 2    | 0.54  | 0.02  | ASR Gel |
| 3    | 0.28  | 0.26  | ASR Gel |

For the mortar bars with 0.2 wt% Al addition, pores within the mortar bar were more noticeable. ASR gel was found in the pore or located around the peripheral area of the pore. From the microstructural and phase analysis, it could be inferred that ASR could occupy and resided within the pores, which potentially led to minimization of cracks within the cement paste.

Fig. 8 SEM micrograph of mortar bar with aluminum 0.2 wt%.

Table 5. Compositional analysis of the area in Figure 8

| Area | Atomic ratio | Materials |
|------|--------------|-----------|
|      | Na.K/Si | Ca/Si |          |
| 1    | 0.80  | 0.73  | ASR gel |
| 2    | 0.19  | 1.39  | Cement paste |
| 3    | 0.17  | 0.10  | ASR gel |

4. CONCLUSION

Aluminum powder was an effective air entrainment agent to create pores within the mortar bars. Higher porosity of 4.8% could be attained in the bars with high aluminum content (0.2 wt% Al addition). An increase in aluminum content resulted in an abundance of pores, which consequently led to minimization of mortar bars expansion when tested according to ASTM C1260. Entrained pores accommodated spaces for ASR gel, resulting in the decline of concrete expansion and potential formation of internal cracks.

5. ACKNOWLEDGMENT

The authors gratefully acknowledge financial support from Kasetsart University Research and Development Institute. Equipment supports from the Department of Materials Engineering, Department of Civil Engineering, Faculty of Engineering, Kasetsart University and the Department of Science Service, Ministry of Science and Technology, Thailand are gratefully acknowledged.

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