



The mechanism of limited inhibition by fly ash on expansion due to alkali–silica reaction at the pessimum proportion

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ABSTRACT

The inhibitory role of fly ash (FA) on alkali–silica reaction (ASR) expansion is drastically reduced at the pessimum proportion relative to its role at full proportion. The main contribution of FA to inhibiting ASR expansion is to reduce the alkalinity of the pore solution and it is dependent on the thermodynamic equilibrium between the pore solution and the calcium-silicate-hydrate (C-S-H) gel. C-S-H gel with low Ca/Si ratio can bind a large amount of alkalis. However, when the concentration is higher than the threshold concentration for reaction, C-S-H gel plays a role in buffering the alkali concentration in the pore solution by supplying alkali. Thus, the inhibitory role of FA on ASR expansion decreases at the pessimum proportion because the aggregate can react with alkali even at low alkali concentrations. Therefore, replacing cement with a larger amount of fly ash is essential to reaching the threshold for reaction.

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1. Introduction

Extensive studies have revealed that replacement with a sufficient amount of supplementary cementitious materials (SCM) such as fly ash (FA), blast furnace slag, silica fume, and other natural pozzolans can inhibit the alkali–silica reaction (ASR) in mortar or concrete. The mechanism of the inhibitory role of FA is mainly reduction of alkalinity of the pore solution [1–3], although this remains controversial. The inhibitory role of FA on expansion due to ASR depends not only on its pozzolanic reactivity but also on the reactivity of the aggregate itself. When the concrete mix contains a highly reactive aggregate, the level of FA required for replacement becomes larger [1].

Highly reactive silica minerals such as opal, cristobalite, and tridymite react with alkalis even at low concentrations. Aggregates containing these silica minerals often show the pessimum effect [4]. This effect is generally explained in terms of the balance between the alkali hydroxide concentration of a pore solution and the reactivity of aggregates [5]. Aggregates showing the proportional pessimum effect have high amounts of dissolved silica (S_c) and a high tendency to reduce its alkalinity (R_c) in the chemical test specified by ASTM C 289 [6–8]; they are thus classified as “potentially

deleterious”. A high R_c in the chemical test indicates that such aggregates can reduce the alkali hydroxide concentration of the pore solution. Certain minerals such as clay in the altered aggregate can also bind these alkalis via cation exchange and render them almost unavailable for ASR.

A schematic illustration of the proportional pessimum effect is shown in Fig. 1(a). When the proportion of the reactive aggregate (A_{ag} in Fig. 1) is larger than the pessimum proportion, the net amount of alkali hydroxide per unit volume of reactive aggregate (N_{AOH} in Fig. 1) becomes insufficient for the ASR gel to exert an expansive pressure that exceeds the tensile strength of the concrete or aggregate itself [9]. Conversely, when the proportion of the reactive aggregate is smaller than the pessimum proportion, the expansion decreases with the decreasing amount of reactive aggregate. At the pessimum proportion, the amounts of the available alkali hydroxide and reactive aggregate reach an optimal balance such that the largest expansion occurs.

In this context, the pessimum expansion behavior of mortar and/or concrete is strongly influenced by the alkali hydroxide concentration of the pore solution. This influence implies that the pessimum proportion depends not only on the aggregate characteristics but also on the alkali hydroxide concentration of concrete. During reduction of the alkali content of the cement, the extent to which ASR expansion is reduced varies with the amount of reactive aggregate (Fig. 1(b)). In fact, cement with lower alkali content exhibits a shift to a smaller pessimum proportion of the reactive aggregate (Fig. 1(b)) [10]. When the alkali concentration is sufficiently high

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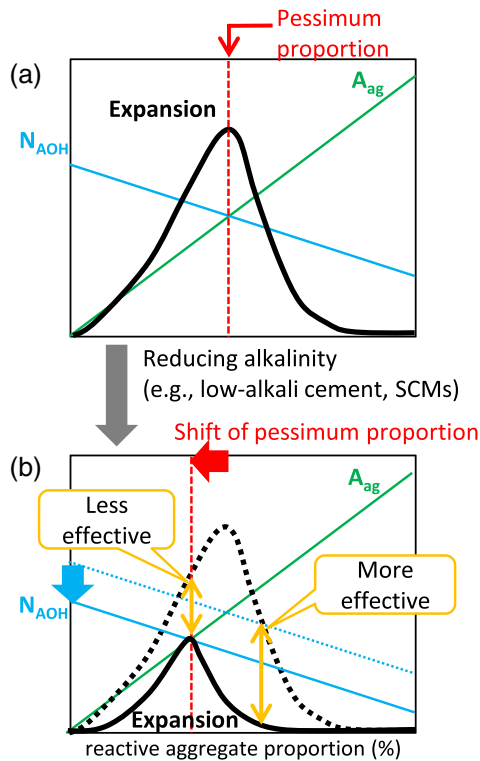


Fig. 1. Schematic illustration of (a) pessimum effect and (b) the influence of reducing alkalinity of the system on ASR expansion (A_{ag} : reactive aggregate proportion (%), N_{AOH} : net amount of alkali hydroxide per unit volume of reactive aggregate).

compared to the binding capacity of the aggregate and/or ASR gel, no pessimum effect is produced.

This discussion can be extended to the inhibitory effect of FA and other SCMs. Since FA reduces the alkali hydroxide ion concentration of the pore solution, replacement of cement with FA has the ability to change the pessimum behavior of the mortar and concrete (Fig. 1(b)). However, little experimental data on this phenomenon has been obtained [11]. The thermodynamic equilibrium of the alkalis between the C-S-H gel and the pore solution is of importance when the cement is replaced with FA; examining such a system may be facilitated by the use of numerical calculations that take into account these interactions. Experiments and numerical simulations focusing on these points are therefore of great importance when designing the ideal mixture for inhibiting ASR expansion by using FA. The goal of this study is to evaluate how the inhibitory effect of FA on ASR expansion decreases at the pessimum proportion and to elucidate the mechanism behind it.

This paper describes the inhibitory role of FA in ASR expansion of mortar, and especially the effect of FA at the pessimum proportion as determined experimentally. Analysis using scanning electron microscopy with energy-dispersive spectrometry (SEM/EDS) of mortar, together with numerical calculations, was also performed to understand the experimental results. From the results, the mechanism behind the reduced effect of FA on the inhibition of ASR expansion at the pessimum proportion is discussed.

Table 1
Chemical composition and physical properties of cement and FA.

Material	Chemical composition (wt%)											Density (g/cm ³)	Blaine SSA (cm ² /g)	
	LOI	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	P ₂ O ₅	Total			
OPC	0.60	21.79	4.98	2.91	65.23	1.21	0.31	0.47	1.72	–	99.22	3.16	3280	
FA	R1	1.50	60.17	22.24	4.29	5.80	1.66	0.48	0.97	0.47	0.81	98.39	2.30	3910
	H	1.93	56.16	26.03	4.82	5.17	1.20	0.55	1.15	0.59	0.43	98.03	2.29	2820

2. Experimental details

2.1. Materials

Ordinary Portland cement with an alkali content of 0.62 wt% was used as the reference material. We used two types of FA (labeled R1 and H), both of which are specified as class II under the Japanese Industrial Standard (JIS) A6201 [12]. Table 1 shows the bulk chemical composition as determined through X-ray fluorescence spectroscopy. Physical properties such as density and Blaine specific surface area are also summarized in Table 1. According to X-ray Diffraction (XRD)/Rietveld analysis [3], the glass contents of FA of R1 and H were 74.1% and 65.7%, respectively.

Four types of reactive aggregates (labeled Ot1, Ot2, Ot3, and Ks) were collected in Hokkaido, northern Japan. Micrographs of these aggregates obtained through polarized microscopy are shown in Fig. 2. Ot1, Ot2, and Ot3 are two-pyroxene andesite containing the reactive silica minerals cristobalite and tridymite that were obtained from different sites in one mine. The major difference between the Ot aggregates is the amount of cristobalite (Ot2 > Ot1 > Ot3). Ks is pit sand consisting of a variety of rock types, including tuff particles containing opal, which is highly reactive.

Results of the chemical test specified by ASTM C 289 are shown in Table 2. The samples were judged as “potentially deleterious” or “deleterious”, meaning that these aggregates have the potential of showing a proportional pessimum effect [4,6]. Pure limestone from Kyushu was found to be a non-reactive aggregate. Andesite coarse aggregate from the Ot mine caused on-site ASR damage in old local structures, but the estimated total alkali contents of these concretes were significantly higher than the limit currently regulated in Japan (3.0 kg/m³). Ks caused ASR damage in several structures with a total alkali content much < 3.0 kg/m³. In the affected structures, Ks was used as a constituent of sand; the non-reactive coarse aggregate was pure limestone. This mix of aggregates is believed to have caused the pessimum effect [13].

2.2. Mixture proportions

Two series of experiments using different aggregates were carried out. In the first series, the andesite aggregate from different locations in one Ot mine was used to study the inhibitory effect of FA at the pessimum proportion. Results were compared against those obtained at full proportion of the reactive aggregate. In the second series, the effects of the amount of reactive aggregate and the level of FA replacement were investigated using Ks.

2.2.1. Series 1

The objective of Series 1 is to evaluate the impact of the proportion of reactive aggregate on the inhibitory effect of FA through the mortar-bar test described in Section 2.3. The water-to-cement ratio and the sand-to-cement ratio were set at 0.50 and 2.125, respectively. The alkali content of cement was boosted to 1.20 wt% of cement by addition of NaOH solution to the mixing water. Non-reactive limestone sand was replaced with reactive aggregate (Ot1, Ot2, and Ot3) at proportions of 30 and 100 wt% of the total sand content. The weight ratio of reactive aggregate content to sand is represented as “ R_w/S_w ”. The final expansions of mortar at 30 and 100 wt% of R_w/S_w are shown in Fig. 3. Pessimum proportions of all aggregates were approximately 30 wt% [11]. At each R_w/S_w

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