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EFFECTIVENESS OF FLY ASH IN PREVENTING DELETERIOUS EXPANSION DUE TO ALKALI-AGGREGATE REACTION IN NORMAL AND STEAM-CURED CONCRETE

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ABSTRACT

A non-reactive and several reactive aggregates were used in concrete specimens with and without two fly ashes (varying in total alkali content) at binder (cement + fly ash) alkali levels ranging from 0.46 to 2.5% and a binder content of 500 kg/m³ and fly ash/binder ratio of 0.25. The specimens were stored either at 23°C (fog room) or 40°C, 100% RH. Some specimens were steam cured at 75°C for eight hours, and then transferred to 40°C, 100% RH. The expansion behaviour of the specimens was monitored over nearly six years, and showed that the effectiveness of the fly ash in preventing deleterious AAR expansion depended on the alkali content of the concrete. At the highest alkali content of 12.5 kg Na₂O equiv./m³, the fly ashes only had a delaying effect (one to several years), whereas at 6.9 kg Na₂O equiv./m³ they eliminated deleterious AAR expansions. Generally, for more highly reactive aggregates, and at the 2.5% alkali level, fly ash was less effective at 40°C than 23°C because the rate of AAR expansion was much higher at 40°C. At lower alkali levels and for less reactive aggregates, the temperature was not important. Fly ashes were also effective under steam-curing conditions. A measurable amount of chemical shrinkage occurred in the first few months in concretes containing fly ash and with high alkali contents, although some of these concretes later expanded and cracked as a result of aggregate reactivity. Fly ash was used in mortar specimens prepared for the expression and analysis of the pore solution and was found to be very effective in reducing the alkalinity of the pore solution, a factor contributing to its preventive effects on AAR. It is concluded (based on six-year results and the shapes of expansion curves) that the two fly ashes can be used to prevent deleterious AAR expansions in practical situations.

Introduction

The effectiveness of fly ash and other mineral additives in preventing the deleterious effects of alkali-aggregate reaction (AAR) has been known for a long time, and recent reviews (1-5) have

summarised most of the previous work. Due to the use of widely different fly ashes, various authors have correctly expressed opposing views on their effectiveness in suppressing AAR. The controversy is related to the composition of the fly ashes and cements used, as well as to the degree of reactivity of the aggregate employed.

For instance, Pepper and Mather (6) found that a range of fly ashes and other mineral admixtures stopped any deleterious expansion in concrete caused by AAR. Nixon and Gaze (7), Nixon *et al.* (8) and Stark (9) found similar behaviours for slowly reactive aggregates when 20 or 30% of cement (by mass) was replaced by fly ash. However, Hobbs (2–4,10) used highly reactive opal as aggregate, in its most expansive combination with cement, which causes severe AAR with expansions far beyond what happens in practice. In such cases, even very effective fly ashes may not reduce the expansion to safe levels, although the magnitude of the expansion may be reduced significantly. Results of such experiments cannot be applied to practical situations.

Factors other than the composition of the cement and fly ash that may influence the results include the level of replacement and the fineness of the admixture. Dunstan (11) found that 25% fly ash addition suppressed AAR expansion, whereas the addition of 5–10% increased the expansion, depending on the alkali content of the cement used. The amount of fly ash needed to suppress AAR also depended on the Ca content of the fly ash. Lee (12) found similar results for class C fly ash and suggested that for each fly ash a critical $\text{Na}_2\text{O}/\text{SiO}_2$ ratio determined maximum expansion due to AAR. Nagataki *et al.* (13), using eight fly ashes in combination with a high alkali cement and pyrex glass as reactive aggregate, found that AAR expansion depended on the amount of soluble alkali and amorphous SiO_2 content of fly ashes and their fineness, but not on their total alkali content.

Thomas *et al.* (14) found very little evidence of AAR in concrete specimens containing a reactive aggregate and a high alkali fly ash, when the level of replacement was 20% or more. Blackwell *et al.* (15) showed that a fly ash of 4.0% Na_2O equiv. prevented expansion of concrete containing a reactive greywacke and 7.0 kg Na_2O equiv./ m^3 . However, their results are only short term (15 months). Thomas *et al.* (16) provided long-term (30 years) evidence that incorporation of a high alkali fly ash in a dam structure containing a reactive greywacke prevented AAR cracking, whereas the portions without the fly ash were damaged due to AAR cracking.

Hobbs (17), using a highly reactive synthetic cristobalite aggregate, also found that expansion due to AAR was influenced by the interaction between the level of alkali in cement (0.6–1.2% Na_2O equiv.) and fly ash (3.0–3.9% Na_2O equiv.), and the level of replacement, determining the total alkali content of the concrete. Whereas 6% replacement increased the expansion with all the cements, 25% replacement of medium and high alkali cements was able to reduce it.

The influence of mineral admixtures on the alkalinity of the pore solution of concrete and the consequences of this on AAR also depend on the alkali content of the cement (and concrete), as well as the available alkali content of the admixture and its replacement level (8,18,19). Generally, low alkali mineral admixtures are more effective than high alkali ones in reducing pore solution alkalinity and expansion caused by AAR, and their effect is greater than a mere dilution of high alkali cements (18,20,21).

However, fly ashes can be variable in this respect. Whereas two low calcium, high alkali fly ashes did not contribute alkali to the pore solution of concrete made with a cement of 0.7% Na_2O equiv. (22), a high calcium, high alkali fly ash more than doubled the alkalinity of the pore solution of a similar concrete, and a low calcium, high potassium fly ash reduced it considerably (21).

TABLE 1

List of Aggregates and Composition of Cement*

Aggregates		Cement composition		
Name	Alkali reactivity		Cement D	Cement N
Basalt	No	SiO ₂	21.60	22.20
Basalt	Yes	TiO ₂	0.45	0.29
River gravel (1)	Yes	Al ₂ O ₃	4.68	3.70
River gravel (2)	Yes	Fe ₂ O ₃	2.78	2.48
Greywacke	Yes	MnO	0.04	0.04
Ignimbrite	Yes	MgO	1.63	2.16
Quartzite	Yes	CaO	66.30	67.50
		Na ₂ O	0.70	0.24
		K ₂ O	0.13	0.33
		P ₂ O ₅	0.09	0.02
		LOI	1.51	0.94
		SO ₃	2.40	2.50
		Na ₂ O equiv.	0.79	0.46
		Surface area (m ² /kg)	385	372

*Cement D and fly ash (1) were used in combination with all the aggregates except the ignimbrite for which Cement N and fly ash (2) were used.

Duchesne and Berube (23) found that expression and analysis of the pore solution is the best method of estimating the contribution of fly ash alkali to the pore solution of concrete.

Australian fly ashes have been found to be effective in reducing AAR expansion (24,25), although the reported results related to short-term monitoring (one year). This paper provides long-term data for nearly six years, showing that at very high alkali contents (12.5 kg Na₂O equiv./m³) AAR expansion may not be prevented, although the fly ashes employed have an excellent tolerance for all practical levels of alkali, even as high as 7.0 kg Na₂O equiv./m³. Data on the effects of an Australian fly ash on the pore solution composition of concrete is also provided.

Experimental Work

Materials. The list of aggregates (all from Queensland) and their reactivity, determined by field performance and/or laboratory testing, is given in Table 1, together with the composition of the two cements used. A detailed petrographic description of the aggregates has been given elsewhere (25). The chemical, mineralogical and some other properties of the two fly ashes used are given in Table 2. Cement D and fly ash 1 were used in combination with all the aggregates except the ignimbrite for which cement N and fly ash 2 were used. This choice was governed by the geographical location of sources. The sand used in this work has been shown (26; and unpublished work) to be non-reactive in concretes containing less than 7 kg Na₂O equiv./m³, but it caused moderate expansions in concrete containing 10 kg Na₂O equiv./m³ and deleterious expansions in concrete with 12.5 kg Na₂O equiv./m³. This sand is regarded as non-reactive by local practice.

TABLE 2
Chemical Composition and Some Other Properties of Fly Ashes

Chemical composition (%)			Other properties		
	Fly ash (1)	Fly ash (2)		Fly ash (1)	Fly ash (2)
SiO ₂	72.6	57.7			
TiO ₂	1.50	1.0	Glass content	80	93
Al ₂ O ₃	22.6	21.3	Mullite*	12	3.6
Fe ₂ O ₃	0.6	7.35	Quartz*	8	3.4
MnO	0.03	0.08	Relative density	2.20	2.35
MgO	0.16	1.38	Retained on sieve No. 300 (52 µm)	6.8	7.6
			325 (45 µm)	9.5	13.2
CaO	0.07	4.84			
Na ₂ O	0.01	6.30	Pozzolanic activity index with cement (ASTM C311)	96	95
K ₂ O	0.31	1.59			
P ₂ O ₅	0.05	0.69			
LOI	1.10	2.10			
SO ₃	0.10	0.22			
Na ₂ O equiv.	0.21	1.35			

*Determined by quantitative XRD and glass content obtained by difference. Fe₂O₃ in fly ash (2) is distributed in the glassy phase, but a small amount of hematite is also present.

Procedures. Concrete prisms measuring 75 × 75 × 285 mm were used for the evaluation of expansion potential of the various aggregate–cement–fly ash combinations. The concrete mixture contained 500 kg cement per m³ concrete, with a water/cement ratio of 0.40. In mixtures containing fly ash, to evaluate the effectiveness of the fly ashes in controlling AAR, 25% of the cement was replaced by fly ash on a mass basis. The water/binder ratio was kept at 0.40, and this caused a reduction of compressive strength of the concretes containing fly ash.

Three levels of cement alkali were employed in this work. One set of concrete prisms was made with and without fly ash with the cements as received, i.e. with no added alkali. For two other sets of specimens, the level of cement alkali was boosted by adding appropriate amounts of NaOH to the mixing water to achieve cement alkali levels of 1.38 and 2.5% Na₂O equiv. Considering the cement content of the concrete, these cement alkali levels correspond to concrete alkali contents of 6.9 and 12.5 kg Na₂O equiv./m³. The corresponding mixes containing fly ash also had the same alkali contents. The addition of alkali also resulted in a considerable reduction in compressive strength of the concrete, as noted earlier (27) for mortar specimens.

Duplicate specimens were demoulded 24 hours after casting and their initial lengths measured. They were then cured in a fog room (under shelter) at 23°C for one week, after which their lengths were measured again. The specimens were then stored at 40°C, 100% RH and their length change measured periodically for nearly six years. The specimens were transferred into a moist box at 23°C and allowed to recover for 16 hours before each length measurement.

Similar specimens were stored at 23°C in a fog room and measured as above for comparison with the storage conditions of 40°C, 100% RH. Another group of specimens were steam cured at 75°C for eight hours, (three hours precuring, three hours of temperature rise from ambient to 75°C, and five hours of cooling to ambient temperature) and, after initial length measurement on demoulding, were transferred to storage conditions of 40°C, 100% RH for monitoring their expansion behaviour.

To investigate the effects of replacing 25% fly ash by mass of cement in the concrete on the pore solution composition of concrete mortar, cylinders (50 × 100 mm) were made using a cement of 0.84% Na₂O equiv. with and without fly ash and stored at 28°C, 40°C and 80°C under sealed conditions for up to 90 days. At certain ages, the pore solutions of the cylinders were extracted under high pressure and analysed for ionic compositions. The ratios of binder/sand and water/ binder were both 0.5 for the mortar mixes.

Results and Discussion

Expansion curves for concrete prisms containing each of the aggregates tested, and stored under the storage conditions of 40°C, 100% RH, are presented in Fig. 1. The concrete mixes for each aggregate (except the ignimbrite) varied in cement alkali level (% Na₂O equiv.) from 0.79% (no added alkali) to 1.38% and 2.5%, corresponding to concrete alkali contents of 3.95, 6.90 and 12.5 kg Na₂O equiv./m³. For the ignimbrite the lowest cement alkali level was 0.49% (no added alkali), corresponding to a concrete alkali content of 2.25 kg/m³, but the higher alkali contents were the same as above.

Figure 1 shows that concrete made with the non-reactive basalt (without fly ash) expanded and cracked at 2.5% alkali level, whereas at other alkali levels there was no deleterious expansion. However, it was discovered by SEM examination of the concrete specimens that the expansion was caused by the reactivity of the sand component at this high alkali content (12.5 kg/m³), whereas it was very stable at lower alkali contents. All the other aggregates caused relatively rapid deleterious expansion and cracking of concrete at the 1.38 and 2.5% alkali levels, far in excess of the expansion of the non-reactive basalt at the highest alkali level.

Only the quartzite aggregate caused cracking in concrete prisms of the lowest alkali content (3.95 kg/m³, i.e. no added alkali). The expansion curves for this aggregate suggest that it was the most rapidly reactive among those tested. Nevertheless, all of the reactive aggregates may react in field concretes of this alkali content due to probable redistribution of alkali and local high concentrations that would be produced as a result of the moisture movements and drying out of concrete. Alkali ingress from external sources is also likely.

Figure 1 also shows that 25% mass replacement of cement by fly ash has been very effective in eliminating the AAR expansion in concretes that contained the 1.38% alkali level (6.9 kg Na₂O equiv./m³). However, it only delayed the expansion and cracking at the highest alkali level of 2.5% (12.5 kg Na₂O equiv./m³), although this alkali level would be outside the usual range of concrete alkali content.

Results reported for one-year expansion measurement (25) showed that only the ignimbrite aggregate caused cracking in the fly ash concrete at the 2.5% alkali level, whereas Fig. 1 shows that all the reactive aggregates except river gravel (1) caused long-term expansion and cracking in concrete of this mixture. These results emphasise the need for long-term monitoring of specimens to verify the effectiveness of a certain treatment. Field concretes containing fly ash have been reported to undergo AAR expansion and cracking, and Hobbs (3,17) mentions two such cases in the USA and Japan.

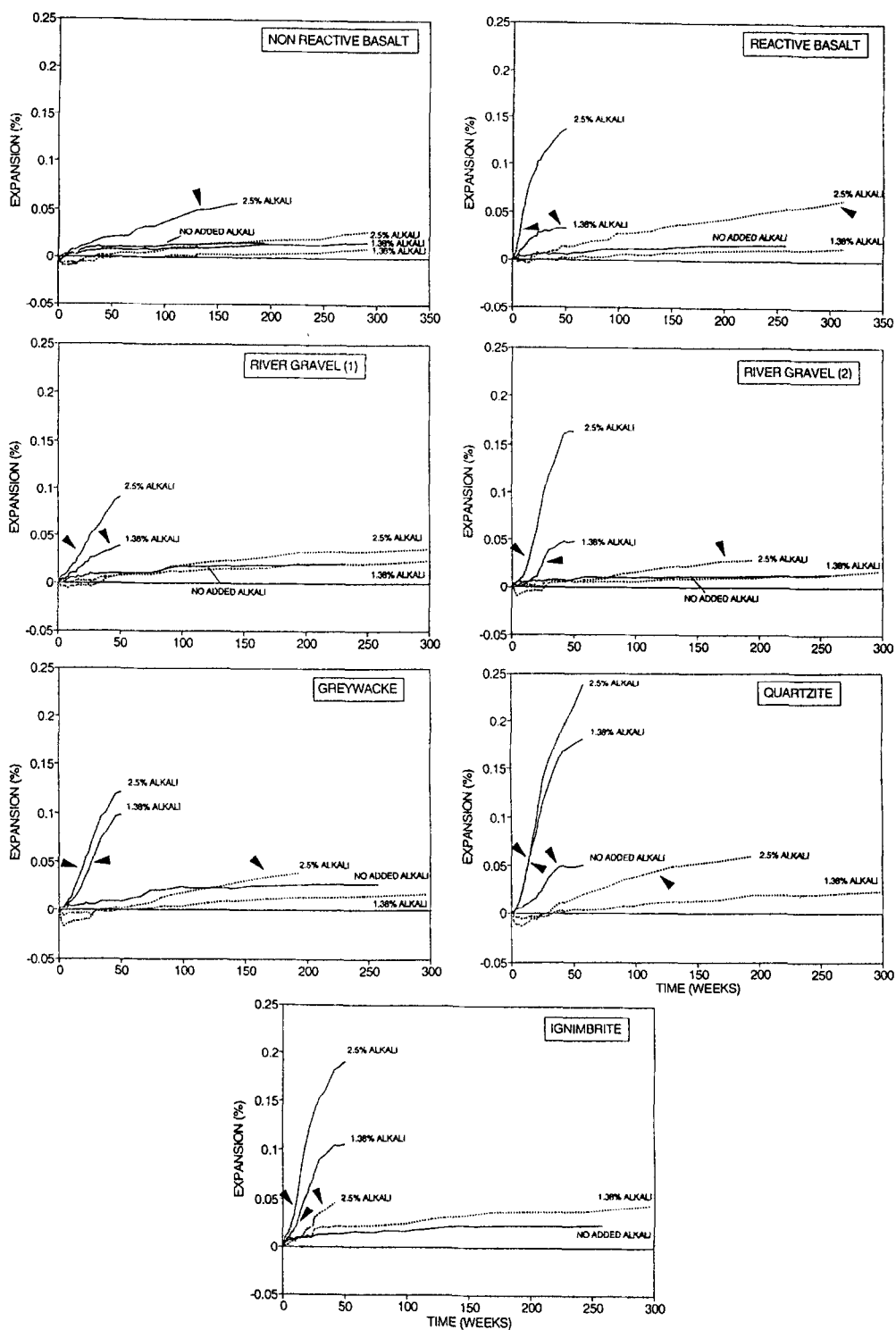


FIG. 1.

Expansion of concrete prisms made with various aggregates and containing various levels of alkali with and without fly ash. Arrows refer to time when cracking was noted. The expansion and cracking of specimens containing the non-reactive basalt at 2.5% alkali level was due to the reactivity of the sand at high alkali. 'No added alkali' refers to 0.79% Na_2O equiv. in the original cement. — no fly ash; fly ash.

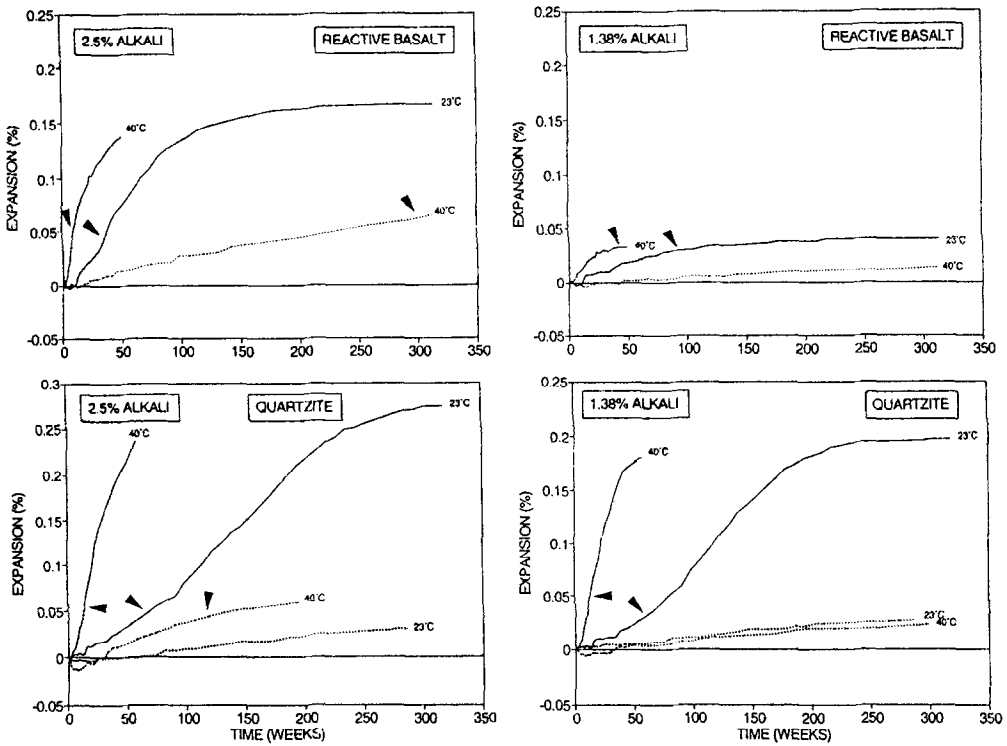


FIG. 2.

Expansion of concrete prisms made with reactive aggregates with and without fly ash at alkali levels of 1.38 and 2.5%, and stored at 40°C, 100 RH, or 23°C (fog room). Arrows refer to time when cracking was noted. — no fly ash; fly ash.

In earlier work (24,25), it was shown that the accelerated mortar bar testing in 1 M NaOH solution at 80°C (28) could predict the effectiveness of fly ash in suppressing AAR expansion in concrete. From the above results, it is clear that the predictions must relate to certain ranges of alkali content in concrete. This is to be expected because mortar bars immersed in 1 M NaOH solution would have pore solutions around 1 M in alkali hydroxide concentration, and the prediction is limited to this upper concentration value. In concrete specimens at the 1.38% alkali level, the pore solution concentration would be equivalent to about 1 M NaOH, and the prediction of the accelerated test agrees with long-term concrete expansion results and effective suppression of AAR.

Figure 2 compares the storage conditions of 23°C (fog room) with 40°C, 100% RH for two aggregates, at the 1.38 and 2.5% cement alkali levels. The other aggregates show similar trends. The expansion curves for the concretes without fly ash show that expansion at 40°C, 100% RH is much more rapid, but that the same magnitude of expansion is reached at 23°C (fog room) after more prolonged storage times. The time lag can vary from about one year to a few years. None of the specimens made with fly ash and stored at 23°C (fog room) showed deleterious expansion and cracking up to the age of six years, regardless of the level of alkali, whereas most of those made at 2.5% alkali level and stored at 40°C, 100% RH have undergone expansion and cracking. This effect was more evident for the more highly reactive aggregates. The difference

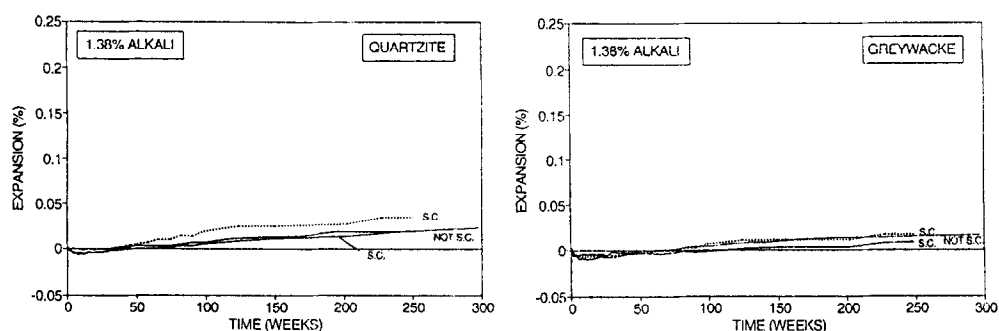


FIG. 3.

Expansion of concrete prisms made with reactive aggregates and the two fly ashes at 1.38% alkali level. Steam curing is designated by 'S.C.' on the curves. The fly ashes behave similarly despite large differences in total alkali contents. — no fly ash; fly ash.

is probably related to the much faster rate of AAR expansion of such aggregates at 40°C, 100% RH, despite the fact that initially fly ash also reacts faster under these conditions (see pore solution concentrations later).

Figure 3 represents the behaviour of steam-cured (75°C) fly ash concrete made at 1.38% binder alkali level and stored at 40°C, 100% RH, and compares the effects of the two fly ashes under these conditions. It is evident that steam curing does not influence the effectiveness of fly ash in suppressing AAR expansion, and that fly ash (2) which contained a much higher total alkali content caused only a slightly higher expansion than fly ash (1). The higher glass content of fly ash (2) (Table 2) probably contributes to its effectiveness.

The effectiveness of various mineral admixtures in reducing deleterious expansions caused by AAR has been attributed to various factors. Bakker (29) proposed that a reduction in permeability for ions and water is responsible for the resistance of blast furnace slag cement to AAR. Tenoutasse and Marion (30) attributed the beneficial role of Belgian fly ashes to the formation of a stable crystalline (hexagonal) hydrated phase which binds large amounts of alkali. Similar products have been reported by Quillin (31) in fly ash/cement mixes cured at temperatures of 50–90°C. Chatterji (32) considered that $\text{Ca}(\text{OH})_2$ formed as a result of cement hydration is essential for AAR to occur. Ming-Shu and SU-Fen (33) and Chatterji and Clausson-Kaas (34) suggested that the beneficial effect of slag cements was to deplete the $\text{Ca}(\text{OH})_2$ in cement paste and therefore inhibit AAR. Hogan (35), in a literature review, refers to the work of Malek and Roy who found that slag cements fixed the sodium ions in the form of $\text{Na}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2 \cdot 5 \cdot 4\text{H}_2\text{O}$. Bhatti and Greening (36) examined 14-year-old high alkali cement pastes made with and without pozzolans. They found that much less alkali could be leached out of the cement/pozzolan paste and this contained much less $\text{Ca}(\text{OH})_2$ than cement paste without pozzolan. They suggested that pozzolans consume the $\text{Ca}(\text{OH})_2$ and form a calcium silicate hydrate (CSH) with a considerably lower Ca/Si ratio than the CSH in the cement paste alone. The low Ca/Si hydrate formed in the presence of Si-rich pozzolans is shown in leaching tests to be capable of retaining much more alkali than higher Ca/Si hydrates that form in plain cement pastes. This is in agreement with findings of Smolczyk (37) that slag limits the mobility of Na ions in concrete and reduces the Na concentration in the pore solution of concrete. Results of Qian *et al.* (38) are also in agreement with this mechanism.

An interesting observation made as a result of this work is that the specimens containing fly

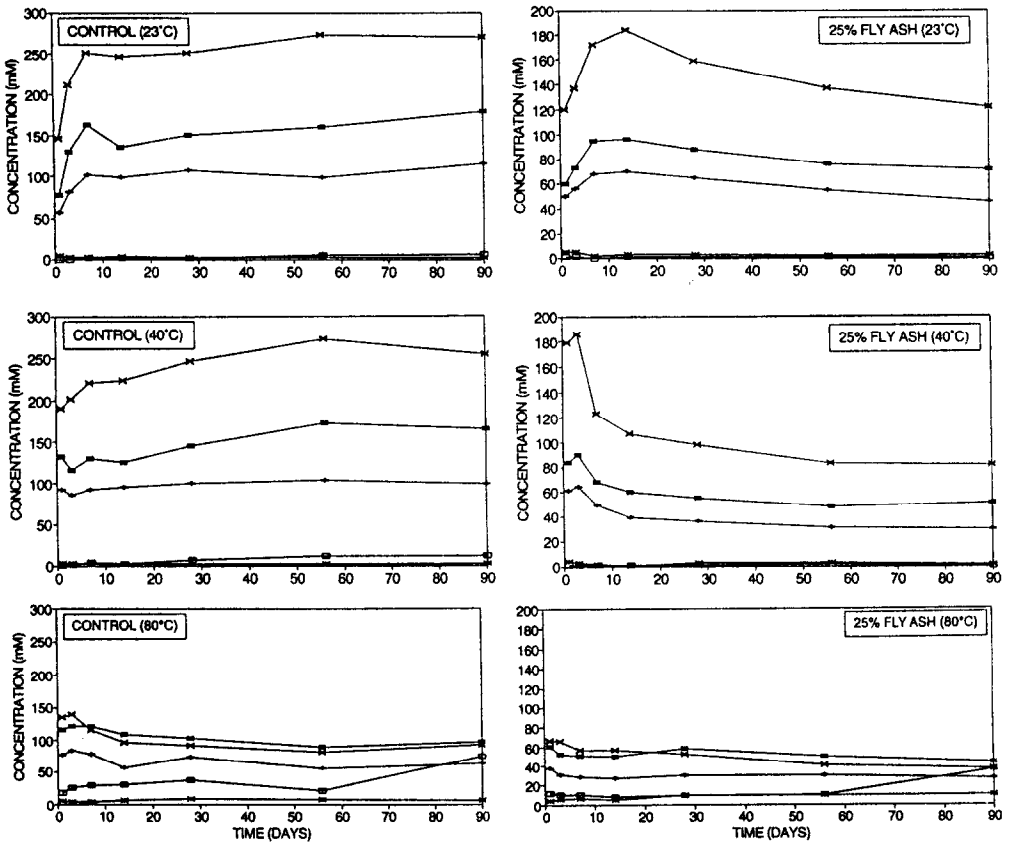


FIG. 4.

Variation with time of pore solution concentration of mortar specimens made with and without fly ash (1), and sealed under storage conditions of 23°C (fog room) or 40°C, 100% RH. ■ Na₂O; + K₂O; * CaO; □ SO₄; × (OH)₂.

ash, particularly those also containing high alkali contents, exhibited an initial shrinkage during the first few months before some expansion took place. This phenomenon is not due to drying shrinkage because it was also observed in large blocks immersed in water. It is suggested that the observed shrinkage in early age is due to the chemical attack of alkali on the glassy phase of the fly ash and its dissolution and formation of a denser CSH-type hydration phase. Therefore, this phenomenon could be regarded as a chemical shrinkage such as that taking place when concentrated NaOH solutions react with unstable silica (39). Further studies are needed to explain the mechanism of this shrinkage and its implications for the use of fly ash as an AAR preventive mineral admixture.

Effects of Fly Ash on Pore Solution Composition. The effects of replacing 25% of a cement of 0.84% Na₂O equiv. by fly ash (1) on the composition of pore solution of the mortar cylinders are shown in Fig. 4. The reaction of fly ash and the consequent reduction in pore solution alkalinity is faster at 80°C than at 40°C and 23°C. At 90 days of sealed storage, the alkali hydroxide concentrations are considerably lower at the higher temperature, but they could probably equalise in the long term. Figure 4 shows that fly ash (1) takes about 10 days at 23°C

and 3 days at 40°C and one day at 80°C to become effective in reducing the pore solution alkalinity of the cement investigated. Similar reductions in the pore solution concentration of concrete mixes containing the fly ash is probably responsible for reduced AAR expansions in these mixes. However, where the ratio of alkali/fly ash is too high it would cease to be effective in reducing the alkalinity of the pore solution below the reactivity threshold. The raised level of SO_4 at the highest temperature is noticeable.

The reduction in alkalinity of the pore solution of concrete made with medium to high alkali cement and containing Si-rich admixtures (8,18,40–42) and the consequent preventive effect on AAR is probably due to the retention of the alkali by the low Ca/Si hydrates mentioned above. However, physical processes such as improved microstructure and reduced permeability of concrete incorporating suitable admixtures, and reduced diffusivity of ions in such concretes also play important roles in improving the general durability of the concrete as well as resisting AAR (29,37). Electrical resistivity of concrete also increases as a result of fly ash reducing the pore solution alkalinity (38). Glasser *et al.* (43) have discussed the hydration processes in cements containing various mineral admixtures, and indicate that both physical and chemical as well as microstructural factors are influential in the processes involved in resisting AAR and improved general durability. It must be noted that achieving the latter requires favourable curing conditions.

Conclusions

Long-term results presented in this work have shown that the two Australian fly ashes studied have been effective in preventing deleterious AAR damage in concretes with alkali contents as high as 7.0 kg $\text{Na}_2\text{O}/\text{m}^3$, but they produced only a delaying effect in concretes containing 12.5 kg $\text{Na}_2\text{O}/\text{m}^3$. The delay was between two and six years, depending on the type of aggregate. A measureable chemical shrinkage occurred in the first few months in the presence of fly ash in the latter concretes, although some of them later expanded and cracked due to AAR.

For highly reactive aggregates and at high alkali contents, the fly ashes were more effective in preventing AAR expansion under the storage conditions of 23°C (fog room) than at 40°C, 100% RH, due to the much faster rate of AAR under the latter conditions.

Fly ash significantly reduced the alkalinity of the pore solution of mortar cylinders, and this occurred more rapidly at 40°C than 23°C, but the difference would be less over longer times.

Although the accelerated mortar bar test (1 M NaOH, 80°C) can be used to predict the long-term effectiveness of fly ash in suppressing deleterious AAR expansions, this prediction only applies to concretes having alkali contents which would produce pore solution concentrations around or below 1 M NaOH.

As field concretes contain usually less than 7.0 kg Na_2O equiv./ m^3 , the two fly ashes should be effective in suppressing AAR in practical applications.

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