THE ROLE OF ALKALI CONTENT OF PORTLAND CEMENT ON THE EXPANSION OF CONCRETE CONTAINING REACTIVE AGGREGATES AND SUPPLEMENTARY CEMENTING MATERIALS

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Abstract

This paper presents results covering the effects of alkali content of Portland cement (PC) on expansion of concrete containing reactive aggregates and supplementary cementing materials (SCM). The results showed that the alkali content of PC has a significant effect on expansion of concrete containing no SCM. When SCM is used, the expansion was found to be related to both the chemical composition of the SCM and, to a lesser extent, the alkali content of the PC used. The concrete expansions were explained, at least partly, on the basis of the alkalinity of pore solution extracted from hardened cement paste samples containing the same cementing blend. An empirical relation was developed correlating the chemical composition (Ca, Si and Na_2O_e) of the cementing blend (PC + SCM) and the alkalinity of pore solution. Results from accelerated mortar bar test (ASTM C 1260) and a modified version thereof are also presented.

Keywords: alkali content, fly ash, slag, pore solution, modified accelerated mortar bar test

1 INTRODUCTION

The level of expansion and disruption in concrete containing reactive aggregate depends on the alkali content of the concrete and the reactivity of the aggregate. It has been reported that the expansion increases as the alkali content of the concrete increases [1-4]. The levels of alkali that trigger the expansion depend, however, on the reactivity of the aggregates. Alkalis from Portland cement (PC) are one of the major sources of alkalis in concrete. The use of supplementary cementing materials (SCM) such as fly ash and slag has been found to be an effective preventive measure against alkali silica reaction (ASR) in concrete [4-6]. One of the mechanism by which SCM mitigate the expansion is by binding alkalis from concrete pore solution and hence, reduce its alkalinity [6].

Fly ash is a type of SCM which performance in mitigating ASR is dependant on its composition [4]. The Canadian Standard CSA A23.5 classifies fly ash based on its calcium content as follows:

Type	CaO (%)	Loss on Ignition, LOI (%)
Low Calcium (F)	≤ 8 ± 1	≤ 8
Intermediate Calcium (CI)	> 8 and ≤ 20 ± 2	≤ 6
High Calcium (CH)	> 20	≤ 6

When used with low alkali Portland cement, low-calcium fly ashes of alkali content in the range of 3.0 to 3.9% have been reported to increase the expansion in concrete containing opaline silica [7] and cristobalite [8]. Thomas et al [9] reported similar findings for samples containing siltstone and siliceous lime stone, and 25% fly ashes. On the contrary 25% of the same three fly ashes (of CaO content ranged from 1.49% to 3.74% and Na_2O_e from 2.98 to 3.86%) were able to reduce the expansion of concrete containing flint sand at all the tested alkali contents which ranged from 4.0 to 6.3 kg/m^3 Na_2O_e [9].

Nixon et al. [10] studied the chemistry of pore solution of pastes containing five Portland cements of Na_2O_e in the range from 0.22% to 1.06% and a fly ash of CaO = 1.85% and Na_2O_e = 3.4%. They found that for all the tested cement and ash replacement levels, 10% to 40%, fly ash increased the pore solution alkalinity at 28 days compared to that of a control paste sample with a

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theoretical dilution of the cement content equal to the ash replacement level. At later ages, 365 days, fly ash introduced to all types of cement except the low-alkali cement (0.22% Na_2O_e) reduced the pore solution alkalinity beyond that of mere dilution. For the low alkali-cement, the tested fly ash increased the alkalinity. The work of Nixon et al [10] explained, at least partly, the expansion trends reported by other researchers [7-9] where higher expansion was obtained for some reactive aggregates when fly ash is added to low-alkali PC. This can be attributable to the increased pore solution alkalinity when the tested fly ashes were used with low-alkali PC.

This paper presents results from a study that investigated the effects of fly ash of a wide range of chemical composition on the expansion of concretes containing two reactive aggregates from North America, and Portland cements of different alkali contents. The effects of fly ash composition on the alkalinity of pore solution of pastes containing PC of different alkali contents are also reported. In addition, a modified version of the accelerated mortar bar test (AMBT) is presented. This modified version is intended to determine the safe level of PC alkali content that can be used with reactive aggregate without causing deleterious expansion. The method also evaluates the efficacy of SCM in mitigating the expansion due to ASR when used with PCs of a wide range of alkali contents.

2 MATERIALS AND METHODS

2.1 General

The aggregates investigated in this study were selected to be of different mineralogy and degrees of reactivity. Also, the fly ashes were selected to represent the range of chemical composition of ashes commercially available in North America. The effects of alkali content of Portland cement (PC) and the chemical composition of fly ash on concrete expansion and pore solution alkalinity were investigated.

2.2 Materials and mix designs

The materials used in this study were three Portland cements (PCs) with different alkali contents (0.36, 0.54 and 0.94% Na_2O_e), 9 fly ashes of different chemical composition and one slag. The chemical compositions of the PCs and SCMs are listed in Table 1. The three reactive aggregates used in the study were: (1) Spratt coarse aggregates (siliceous limestone) from Ottawa, Ontario, (2) Sudbury coarse aggregate (greywacke-argillite) from Sudbury, Ontario, and (3) Jobe sand (chert) from Texas. The 1-year concrete expansion of the Jobe, Spratt and Sudbury aggregates are 0.56%, 0.24% and 0.17%, respectively. These values represent a range of reactivity of the three investigated aggregate with Jobe as the aggregate with the highest reactivity followed by Spratt and Sudbury.

The mixture proportions for concrete and mortar samples were according to the applicable test methods. The paste samples for the pore solution study were prepared at a water-of-cementing materials (w/cm) ratio of 0.5.

2.3 Methods for assessment and analysis

Concrete Prism Test (ASTM C1293)

The concrete prism test (CPT) according to ASTM C 1293 was used to investigate the expansion of different mixtures with and without SCM. In addition to samples tested following the standard test method with the alkali content of the PC raised to 1.25% Na_2O_e, additional samples were tested at different PCs alkali contents to study the role of PC alkali content on the expansion of concrete with and without SCM. For control samples (without SCM), PC alkali levels ranging from 0.4% to 1.25% Na_2O_e were tested. For samples with SCM, Spratt aggregates was tested at PC alkali levels of 1.25%, 0.94% and 0.80% Na_2O_e while Jobe sand was tested at PC alkali levels of 0.94%, 0.80% and 0.60% Na_2O_e. No SCM was used with Sudbury aggregate. These alkali levels were selected to capture, for each aggregate type, the range of alkalis within which a change in the alkali content of the PC is likely to have significant effect on the produced expansion.

For standard concrete prism test, the PC with Na_2O_e = 0.94% was used and the alkali content was boosted to 1.25% Na_2O_e by adding NaOH to the mixing water. For concrete prisms prepared with alkali contents below 0.94% Na_2O_e, the two PCs with Na_2O_e of 0.94% and 0.54% were blended with the right proportion to achieve the required alkali levels.

The different fly ashes were used at a replacement level of 25% with Spratt aggregate. For Jobe sand, low and intermediate-calcium fly ashes (F and CI) were used at 20% while high-calcium fly ashes (CH) were used at 30%. Slag 100 was used with Jobe sand at 35%.

Accelerated Mortar Bar Test (ASTM C1260 and C1567)

The same cementing blends tested in the standard CPT with PC alkali content raised to 1.25% Na_2O_e were also tested using the standard accelerated mortar bar test (AMBT) according to ASTM C 1260 for samples with no SCM and ASTM C 1567 for samples with SCM

Modified Accelerated Mortar Bar Test (Modified ASTM C1260 and C1567)

A modified version of ASTM C 1260 and ASTM C1567 was used to test mixtures with low-alkali PC. The purpose of using this modified method is to take into consideration the effect of alkali content of PC when evaluatig the efficacy of a cementing system in mitigating the expansion. In this modified version, the alkalinity of soaking solution was adjusted to reflect the alkali content of the PC used in the mix, following the relation:

$$OH^- \text{ concentration of the soaking solution (mol/L)} = 0.70 * Na_2O_e \text{ of PC (\%)} \quad (1)$$

The above relation is based on earlier research work [10] that showed the hydroxyl ion concentration in pore solution of cement paste with no SCM to be 0.7 * % Na_2O_e of the PC. The results from the CPT were used as a benchmark to evaluate the efficacy of the modified ASTM C 1260 in predicting the reactivity of the aggregates when used with low-alkali PC.

Pore Solution Study

Pastes samples were prepared at a w/cm ratio of 0.50. The samples were cured at room temperature until the age of testing (i.e. 90 days). Four different alkali levels of PC were used, namely 0.36%, 0.6%, 0.8% and 0.94% Na_2O_e. These levels of alkali content were chosen to represent low, moderate and high-alkali PC with the intension of investigating the role of PC alkali content on pore solution alkalinity at the presence of fly ash. The 0.60 and 0.80% Na_2O_e were achieved by mixing the two PCs of Na_2O_e of 0.94% and 0.54%. In addition to control samples with no SCM, selected fly ashes were tested at a replacement level of 25% with PC at the four levels of alkali contents.

The samples were mixed using a high-speed, high-shear food blender with 3.8-L stainless steel mixing bowl. After casting, the samples were sealed in 50 x 100 mm polyethylene cylinders and rotated at a speed of 12 rpm for the first 24 hours after casting to minimize segregation. The sealed samples were then stored over water at laboratory temperature (23 ± 2°C) until testing. Upon testing, samples were demoulded, broken into fragments (5-20 mm) and squeezed to extract the pore solution following the method described by Barneyback and Diamond [11]. The fragments were exposed to an increasing pressure at a rate of about 2.0 MPa/second until a pressure of 160 MPa was reached. The pressure was then allowed to decrease gradually while the pore solution was collected under vacuum. When the pressure reached zero, the samples were loaded again until a pressure of 320 MPa was reached where the extraction procedure was repeated. The operation of loading and retracting was repeated for a third time with the pressure reaching 480 MPa.

Hydroxyl ion concentrations of the extracted solutions were determined within 20 minutes of extraction by potentiometric titration with 0.05 N H_2SO_4 solution, while sodium and potassium ion concentrations were determined by flame photometry.

3 RESULTS

The effects of PC alkali content on the 1-year expansions of the concrete prisms containing the three reactive aggregates are shown in Figure 1. The PC alkali content has significant effect on the expansion, the higher the alkali content the higher the expansion. The level of PC alkali content required to maintain the expansion below the 0.04% expansion limit at 1 year was 0.70% Na_2O_e, for Spratt and Jobe, and 0.80% for Sudbury aggregate.

The effect of PC alkali content on samples with SCM was not as noticeable as that with the control samples (containing no SCM). This can be seen in Figure 2 which illustrates the expansion of concrete samples with Spratt and 25% fly ash. It is clear from the graph that the role of PC alkalis is secondary compared to that of the fly ash's composition. In other words, the difference in expansions between samples containing the same ash but PC of different alkali contents is not as significant as that between samples containing different fly ashes but PC of the same alkali level. In fact, the only fly ash that showed noticeable difference in expansion when used with PC of different alkali content was FA # 18. This fly ash lies at the lower end of CaO content of high calcium fly ash category (CH), and was not effective in suppressing the expansion when used at 1.25% Na_2O_e. When used at alkali content of 0.94% and 0.8%, it did marginally meet the 2-year expansion limit. However, there was still no difference between the expansion of concrete prepared with PC of 0.8% and 0.94% Na_2O_e.

It is also noticeable that the use of any of the fly ashes with Spratt aggregates at any of the tested alkali level (0.80% to 1.25% Na_2O_e) reduces the expansion compared to control samples at the same Na_2O_e. This was not the case with Jobe sand as can be seen in Figure 3. The high-calcium FA # 71 produces higher expansion than that of the control when used at alkali levels lower than 0.94% Na_2O_e. The high alkali FA # 82 produces expansion values higher that those of the control samples at all alkali levels. On the other hand, low and moderate calcium fly ashes of Na_2O_e content < 4.0% were effective in suppressing the expansion regardless of the alkali content of the PC. This can be explained, at least partly, by examining the pore solution alkalinity of the different samples as illustrated in Figure 4. Pore solution alkalinity of FA # 71 was higher than that of the control sample (with no SCM) at all alkali levels. The same can be said for FA # 82 although its pore solution alkalinity was much higher than that of the sample with FA # 71. It should be mentioned that the alkalinity is represented in Figure 4 by the hydroxyl ion concentration (OH^-) which, as expected, equaled the sum of Na^+ $+K^+$ ions for all the tested samples. In terms of the efficacy of the slag, 35% was sufficient to suppress the expansion of concrete containing Jobe sand at the three tested alkali levels as shown in Figure 3.

Figure 4 also shows that unlike fly ashes of high alkali (> 4.0% Na_2O_e) or high calcium contents, low and moderate-calcium fly ashes of Na_2O_e < 4.0% reduce the alkalinity of pore solution compared to that of samples with no FA. It is also clear from Figure 4 that the effect of PC alkali content on pore solution alkalinity is secondary compared to that of the fly ash composition.

The empirical relation shown in Figure 5 illustrates the relation between the main oxide composition of the cementing bend (PC + FA) and the alkalinity of pore solution extracted from mature paste samples at the age of 90 days. The oxide composition of a blend is calculated as the sum of the products of the oxide content of each cementing component (PC or SCM) multiplied by the proportion of this particular component in the total cementing materials. For example, the CaO content of the paste containing a cementing blend of 25% FA # 1 and 75% HAPC = 0.75 * 61.25% (CaO of the PC) + 0.25 * 1.14% (CaO content of the FA) = 46.04%. It is clear from the graph that the pore solution alkalinity increases as the Na_2O_e and CaO contents of the cementing blend increase and its SiO_2 decreases.

Figures 6 through 8 illustrate the efficacy of the modified accelerated mortar bar test (modified AMBT) in evaluating the expansion of reactive aggregates when used with PC of different alkali contents. Using expansion limits of 0.10% at 14 days for the modified mortar bar test and 0.04% at 1 year for the concrete prism test, the following "safe" levels of alkali can be determined from the data in Figures 6, 7 and 8:

Aggregate	Modified Mortar Bar Test	Concrete Prism Test
Spratt	0.65%	0.70%
Sudbury	0.75%	0.80%
Jobe	0.35%	0.70%

These results suggest that ASTM C 1260 may be used to determine the effect of alkalis by appropriate modification of the host solution composition for Sudbury and Spratt aggregate. However, for aggregates like Jobe sand, the safe alkali level determined by the modified C 1260 is much lower than that determined by the concrete prism test.

The efficacy of the modified AMBT in predicting the two-year expansion of concrete prisms containing SCM is shown in Figure 9. The results from the standard tests (CPT and AMBT) for Spratt aggregate and SCM are also shown on the graph. No standard CPT (at 1.25% Na_2O_e) was available for Jobe sand with SCM. Out of the 30 test points conducted at alkali contents lower than that of the standard tests, 7 test results (3 for Jobe and 4 for Spratt) failed the modified AMBT but passed the CPT. None of the tested samples passed the modified AMBT and failed the CPT. The remaining 20 test points showed agreement between the two tests (either passed-passed or failed-failed). The three test points that lie on the 0.04% expansion limit of the CPT (2 for Jobe and 1 for Spratt at 1.25% Na_2O_e) failed the Modified AMBT (or the standard AMBT in case of the Spratt sample) and had CPT expansion values of 0.039, 0.041 and 0.041%. The three test points can be considered as agreement between the two tests since the CPT expansions are either above or marginally meet the 0.04% expansion limit. Based on the obtained results, one can argue that the modified AMBT is a promising tool that can be utilized as a screening test for evaluating the efficacy of SCM in mitigating ASR expansion in concrete contacting PC of different alkali levels. Samples that pass the modified AMBT at 14 days will most likely pass the CPT at 2 years. However, samples that fail the modified AMBT would require further testing, either using the CPT or testing under site exposure. This is the same criterion governing the current use of the standard AMBT. This criterion is also supported by the results in Figure 9 for the standard AMBT and CPT conducted on Spratt at 1.25% Na_2O_e.

4 DISCUSSION

The results shown in Figures 2 and 3 demonstrate that different reactive aggregates respond differently to the alkali level of PC or the use of SCM. As Figure 2 shows, the use of any type of SCM with PC of any alkali content reduces the expansion of concrete containing Spratt aggregate compared to that of the control samples at the same PC alkali level. On the other hand, only Types F and CI ashes of $Na_2O_e < 4.0\%$, and slag 100 were able to reduce the expansion of concrete sample containing Jobe sand as shown in Figure 3. This can be explained based on the response of each aggregate to alkalis. Shehata and Thomas [4] demonstrated that the expansion of concrete with Spratt increases with the increase in alkali content until reaching a maximum value after which any increase in alkali content does not cause significant expansion. At this high level of alkali, factors other than pore solution alkalinity, such as ion diffusivity or reduction in $Ca(OH)_2$, help reduce the expansion [4]. This seems to be the case with the high-calcium or high-alkali fly ashes investigated with Sprat aggregate in this paper. Although, these ashes increase pore solution alkalinity beyond that of the control samples as shown in Figure 4, the Spratt aggregate was not adversely affected by the increased level of alkalinity, but was positively affected by the reduced $Ca(OH)_2$ or ion diffusivity associated with using SCM. On the contrary, the expansion of the highly reactive Jobe sand increases beyond that of the control samples as the alkalinity of pore solution increases. This is attributable to the nature of this sand which continue to react with increasing alkali levels, even beyond that of the control sample. The response of Jobe and Spartt to increasing alkali levels is illustrated in Figure 1.

The expansion of Jobe sand with FA # 82 decreases with the increase in alkali content of the PC (Figure 3), although the expansion at the three tested alkali levels were way above the 0.04% expansion limit. The reason behind this trend is not clearly understood. The alkalinities of pore solution of mature paste samples at the three tested alkali levels were very close. There is a possibility, however, that at low alkali content of PC, more alkalis were contributed from the fly ash. In the case of concrete, these contributed alkalis were rapidly consumed by the highly reactive sand causing more expansion, instead of being bound by the hydration products of the cementing materials as the case may have been with paste samples. An earlier work by Shehata and Thomas [12] demonstrated that more alkalis are contributed from SCM in solution of lower alkalinity.

In general, the role of alkali content of PC on the expansion of concrete containing reactive aggregate is secondary compared to that of the type of SCM used. In other words, for the reactive aggregates tested in this study, ASR can be mitigated provided that the right Type and level of SCM are used, regardless of the alkali content of the PC. The role of PC alkalis on pore solution alkalinity is similar to that on concrete expansion. This was demonstrated in Figure 4 and can also be concluded from the relation developed in Figure 5 which shows a strong relation between pore solution alkalinity and the parameter $(Na_2O_e * CaO)/(SiO_2)^2$ of the cementing blend. The effect of PC alkali content on this parameter is not as significant as that of the fly ash composition. This can be demonstrated by examining the values of the $(Na_2O_e * CaO)/(SiO_2)^2$ for different fly ashes used at different PC alkali contents:

Fly ash	$(Na_2O_e * CaO)/(SiO_2)^2$	
	PC of 0.94% Na_2O_e	PC of 0.60% Na_2O_e
FA # 1 F-LA	0.07	0.06
FA # 71 CH-LA	0.11	0.09
FA # 82 CI-HA	0.18	0.16

It is clear that changing the alkali content of PC does not result in significant change in the value of the parameter $(Na_2O_e * CaO)/(SiO_2)^2$, contrary to the case when a different fly ash is used.

The results of the modified AMBT showed it to be a promising tool for testing cementing blends with PC of different alkali levels. For determining the safe level of alkali required to mitigate expansion in concrete with no SCM, the test was very effective with Spratt and Sudbury aggregates. However, it overestimated the expansion of concrete with Jobe sand. This is attributable to the sensitivity of this reactive aggregate to alkalis. The large amount of soaking solution provides enough alkalis to promote reaction of this highly reactive aggregate, regardless of the alkali concentration of the solution (Figure 8).

The use of the modified AMBT to evaluate the expansion of cementing blends containing SCM and low-alkali PC showed promising results, although more research work that cover a wider range aggregates is required in this area. It should also be born in mind that testing large blocks in exposure sites provide the most reliable expansion results [12]. Hence, results from test blocks should also be considered as benchmarks for comparing results from accelerated tests.

5 CONCLUSIONS

For the range of aggregate reactivity and chemical composition of the cementing materials used in this study, the following conclusions are drawn:

- The role of alkali content in affecting expansion due to ASR is secondary compared to that of the composition of SCM
- The expansion of concrete containing reactive aggregate and SCM can be explained, to a large extent, based on the effect of SCM on pore solution alkalinity
- The alkalinity of pore solution of mature cement pastes containing SCM and PC of different alkali contents could be predicted based on the silica, calcium oxide and equivalent alkalis of the cementing blend
- Using soaking solutions of various alkalinities, a modified version of the AMBT was introduced and showed promising results in terms of predicting the 2-year expansions of concrete prisms containing SCM and PC of different alkali levels. The test was also successful in determining the safe alkali levels for concrete with no SCM, for two of the three tested aggregates.

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Table 1: Chemical composition of the cementing materials

Sample ID	FA Type	SiO_2	Al_2O_3	TiO_2	P_2O_5	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3 Leco	Na_2O_e	AA[1]
HAPC		20.34	5.06	0.27	0.13	2.55	61.25	2.57	0.26	1.04	3.23	0.94	
LAPC 1		20.36	4.73	0.25	0.09	2.89	63.01	3.26	0.08	0.43	2.87	0.36	
LAPC 2		20.32	4.59	0.30	0.09	3.15	63.16	2.67	0.09	0.69	2.99	0.54	
FA # 1	F-LA[2]	53.56	29.03	1.33	0.14	7.62	1.14	0.90	0.44	3.01	0.53	2.42	0.54
FA # 8	F-LA	57.13	18.71	0.85	0.40	5.31	8.39	3.01	3.02	1.19	0.52	3.81	1.58
FA # 56	CI-LA[3]	49.28	24.59	1.64	0.17	3.74	15.08	2.56	0.34	0.70	0.72	0.80	0.25
FA # 106	CI-HA[4]	34.09	20.50	1.17	0.61	5.07	16.40	3.93	9.18	0.72	4.83	9.66	6.25
FA # 76	CH-LA[5]	40.22	23.28	1.47	0.95	6.01	18.19	4.11	1.41	1.13	1.03	2.16	0.81
FA # 60	CH-LA	35.92	21.18	1.68	1.36	5.85	24.48	4.46	1.70	0.48	1.29	2.01	1.55
FA # 82	CH-HA[6]	32.94	19.14	1.39	0.95	6.24	23.17	5.14	5.52	0.58	3.02	5.91	4.24
FA # 18	CH-LA	37.46	19.39	1.41	1.28	5.94	23.93	4.90	1.84	0.56	1.76	2.21	1.50
FA # 71	CH-LA	31.39	18.54	1.60	1.20	5.22	29.83	5.22	2.10	0.31	2.62	2.30	1.72
Slag 100		39.49	9.57	0.52	0.00	0.39	34.25	12.58	0.46	0.56	2.65	0.83	0.30

[1]AA: Available alkali (ASTM C 311)
[2] F-LA: Type F fly ash with low alkali content (< 4.0% Na_2O_e)
[3] CI-LA: Type CI fly ash with low alkali content (< 4.0% Na_2O_e)
[4] CI-HA: Type CI fly ash with high alkali content (> 4.0% Na_2O_e)
[5] CH-LA: Type CH fly ash with low alkali content (< 4.0% Na_2O_e)
[6] CH-HA: Type F fly ash with high alkali content (> 4.0% Na_2O_e)

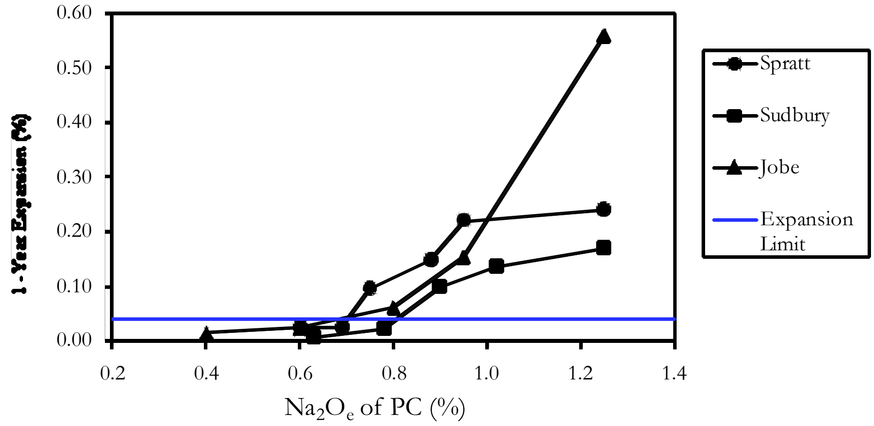

Figure 1: Effects of PC alkali content on concrete expansion.

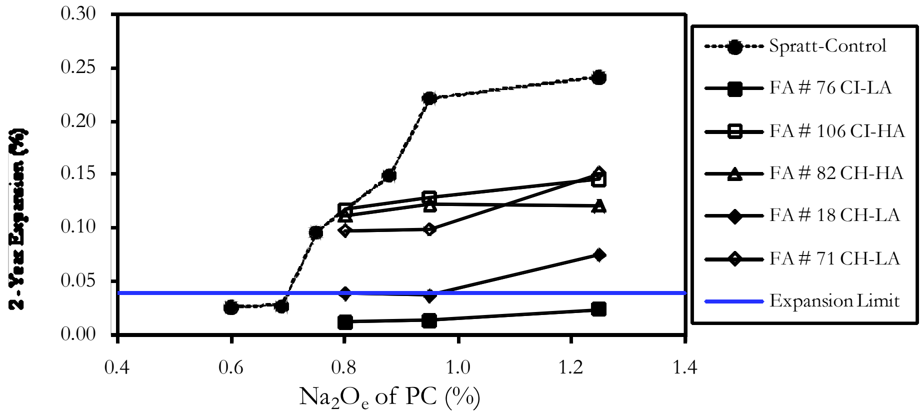

Figure 2: Expansion of concrete containing Spratt aggregate and 25% fly ash.

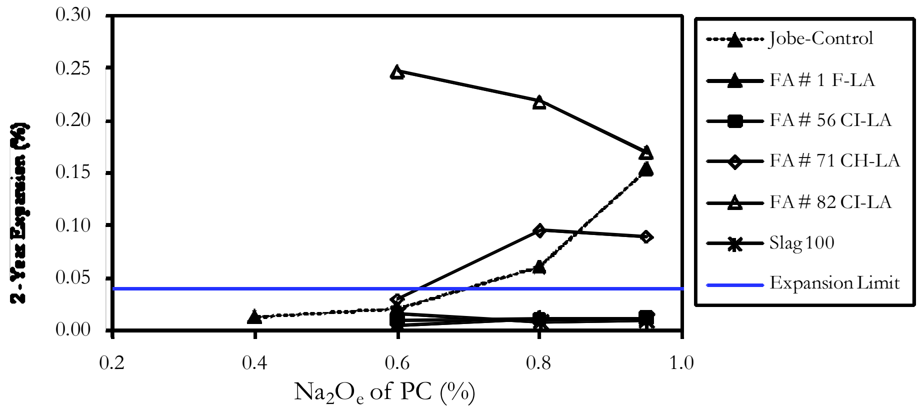

Figure 3: Expansion of concrete containing Jobe sand and SCM.

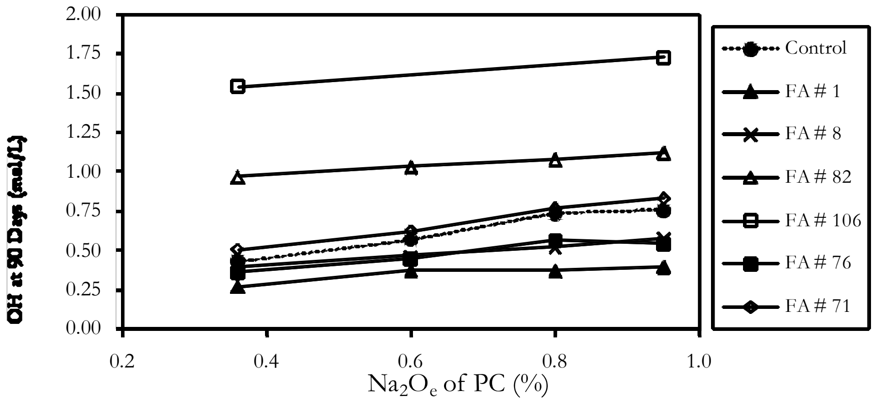

Figure 4: Pore solution alkalinity of mature paste samples.

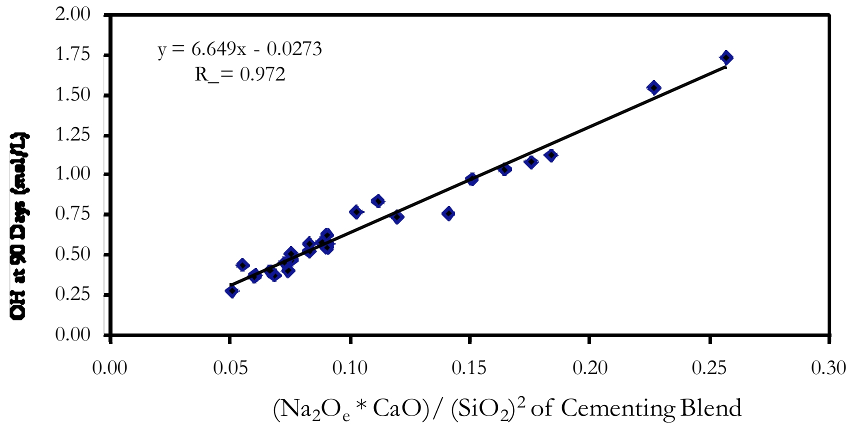

Figure 5: Relation between main oxide compositions of cement blends and pore solution alkalinity of mature paste samples.

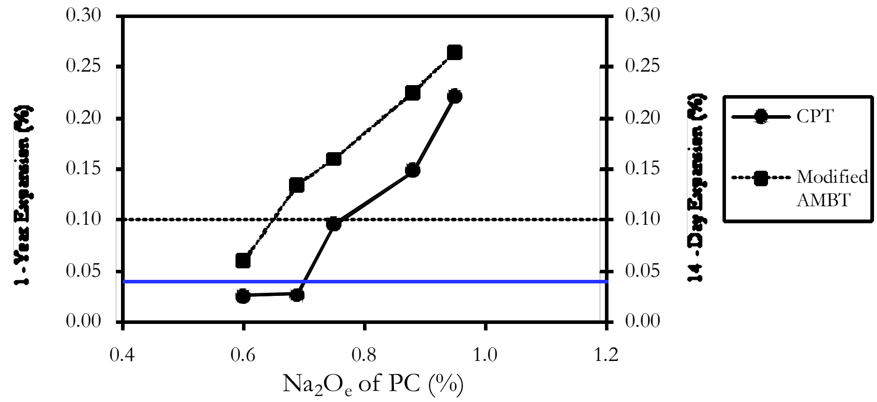

Figure 6: Results of modified AMBT and CPT for control samples containing Spratt.

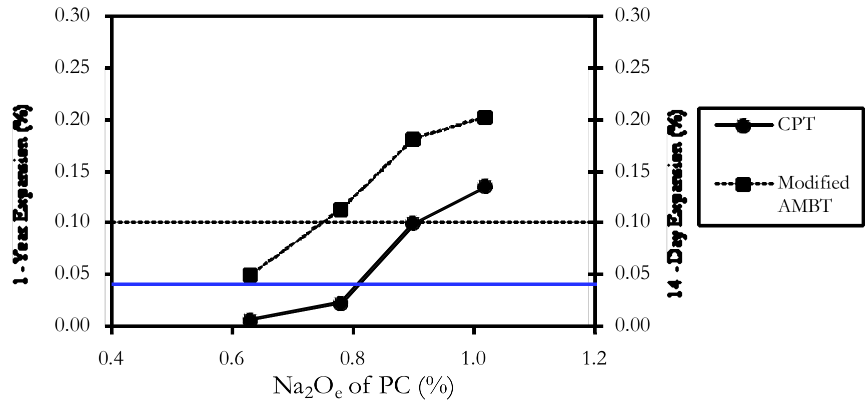

Figure 7: Results of modified AMBT and CPT for control samples with Sudbury.

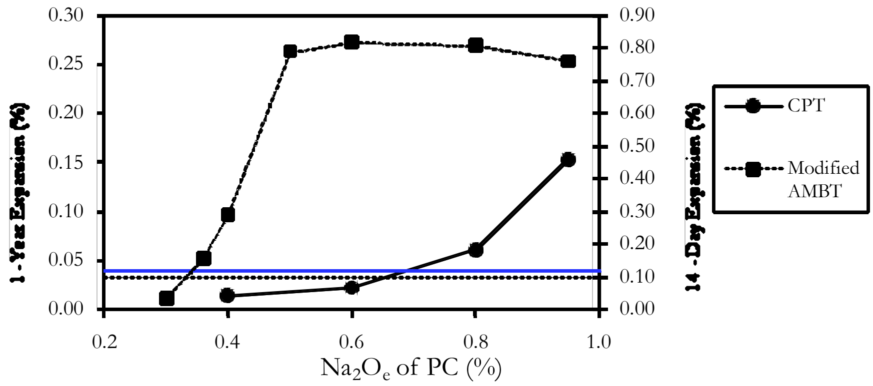

Figure 8: Results of modified AMBT and CPT for control samples with Jobe sand.

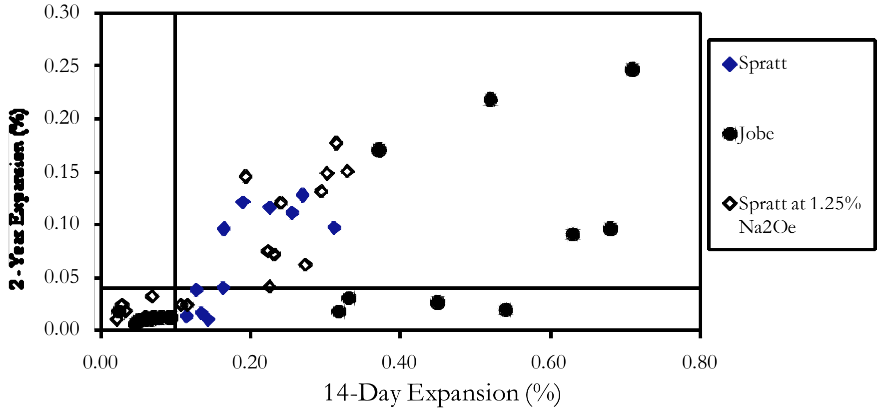

Figure 9: Two-year expansion of concrete containing SCM versus 14-day expansion from the modified AMBT.