

## Performance of Portland-limestone cements with supplementary cementitious materials to mitigate alkali-silica reaction

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### Abstract

Supplementary cementitious materials (SCMs) are frequently used to mitigate or prevent alkali-silica reaction (ASR) in concrete with alkali-silica reactive aggregates. The influence of portland-limestone cements (PLCs) and SCMs together on mitigation of ASR has not been studied extensively. Limited research showed that there was no consistent difference in expansion results for PLCs, up to 15% limestone by mass, with SCMs compared to ordinary portland cements (OPCs) with SCMs in concrete with an alkali-silica reactive aggregate. In addition, previous studies focused only on one aggregate, Spratt – a highly reactive siliceous limestone rock, and mainly slag as an SCM. In this study, ASTM C441 was used as an initial cementitious materials screening test and ASTM C1567 was used to evaluate the performance of interground PLCs with SCMs to mitigate ASR. Various SCM including two class-F fly ashes with different calcium oxide contents, slag, silica fume, and a natural pozzolan were evaluated. Borosilicate glass was used as a fine aggregate for ASTM C441 testing and two very-highly reactive fine aggregates were used for ASTM C1567 testing. The results showed that the interground PLCs with SCMs had similar or better performance when compared to the respective OPCs with SCMs in terms of expansion of the mortar bars. Several additional mixtures are being tested according to AASHTO T380 and ASTM C1293 methods to verify if consistent results are seen in concrete specimens. The results of this study will provide further evidence if the mitigation options that are used with OPCs can be utilized as-is, increased, or decreased when used with PLCs.

**Keywords:** portland limestone cements; supplementary cementitious materials; alkali-silica reaction

## 1. INTRODUCTION

Alkali-silica reaction (ASR) is a well-known durability problem in concrete, which causes extensive cracking and shortens its lifespan. The deleterious chemical reaction occurs between reactive silica that is found in aggregates and alkali hydroxides in the concrete pore solution. Various mitigation methods exist to control the expansion due to ASR in concrete. The most common, effective, and economical ASR mitigation methods involve replacing an adequate amount of cement with supplementary cementitious materials (SCMs) [1]. SCMs suppress ASR expansion mainly by reducing pore solution pH due to alkali dilution and alkali binding, reduced permeability, etc. [1-4]. The effectiveness of SCM to mitigate ASR expansion depends on their chemical composition (mainly calcium oxide, silica, and alkalis) and fineness [1,5]. The replacement level of cement with SCM needs to be increased with increase in SCM's calcium content, decrease in its silica content, and increase in available alkali [1]. SCMs containing alumina are effective in controlling ASR expansion as aluminum in pore solution was found to lower the amorphous silica dissolution rate from aggregates [6]. Fly ash was shown to reduce the amorphous silica dissolution rate from aggregates by providing a large silicate surface area that is accessible to the hydroxide ions in the pore solution thus reducing the attack of hydroxide ions on the reactive aggregates [3].

The expansions of mortars and concrete are monitored in the laboratory accelerated methods (examples: the AMBT - accelerated mortar bar test and the CPT - concrete prism test) as well as in the field exposed blocks. Replacing ordinary portland cements (OPCs) with sufficient amount of SCMs was already extensively studied and proven to be very effective in mitigating ASR. However, the influence of interground PLC on mitigation of ASR has not extensively been studied.

Laker and Smartz used PLC with 10-12% interground limestone to evaluate alkali-silica reactivity. ASTM C 1567 method was used in the study to evaluate the expansions of the mortar bars. They observed that PLC performed comparable to OPC in terms of expansions [7]. Thomas et al. tested PLC with 12%

interground limestone for its resistance to ASR and compared the results to OPC. AMBT (ASTM C1260), CPT, and accelerated concrete prism test (ACPT) were done using alkali-silica reactive Spratt aggregate. The authors observed no consistent difference in expansion results produced with PLC compared with OPC [8].

Hooton et al. [9] compared the performance of three cements with different levels of interground limestone – 3.5%, 10%, and 15% and labeled as GU, PLC10, and PLC15 respectively. Both the AMBT and CPT methods were done for various mixes. When no slag was used, none of the mixes passed the AMBT test, and PLC10 and PLC15 on their own had higher expansions than GU. Even replacing the cements with 30% slag was not effective. When 50% of the cements were replaced with slag, all the cements passed the AMBT test. In combination with SCM it was observed that both the PLC10 and PLC15 cements had less expansion when compared to the GU cement [9]. From the CPT results - GU, PLC10, PLC15, GU Slag 30%, and PLC10 Slag 30% failed the test. Whereas PLC15 Slag 30%, GU Slag 50%, PLC10 Slag 50%, and PLC15 Slag 50% passed the test with less than 0.04% expansion even after 2 years. It was observed that expansions were in an increasing order with the limestone content when no SCMs were used or in the case of the cements replaced with 50% slag. This was not observed in the case of cements replaced with 30% slag [9].

From these previous studies, concrete with PLCs and no SCMs expanded more than their respective 100% OPC. Whereas in the case of mixtures containing SCM, no consistent difference in expansion results was observed for PLC compared with OPC. The previous research was mostly limited to testing one aggregate, Spratt, a highly-reactive siliceous limestone rock, and mainly slag as an SCM. In addition, the majority of literature addresses Type I or GU cement and it does not address Type II or Type V (sulfate resisting) cement. Therefore, there is a need to extensively study the performance of PLCs with various types of SCMs and aggregates especially for regions where Type II and V cements are used to reduce the risk of sulfate attack.

Previous research showed that equivalent strength could be achieved in concrete with PLC having up to 15% limestone by mass by intergrinding the limestone with clinker [10-13]. Portland-limestone cements (PLCs) generally show a synergistic benefit when used with SCMs [14-16]. Limestone particles participate in hydration reactions by acting as nucleation sites and accelerating the reaction and participate in hydration reaction in concrete with alumina containing SCMs through the formation of calcium monocarboaluminate compounds thereby increasing the pore refinement [17-20]. ASTM C595 (AASHTO M240) currently allows up to 15% interground limestone in blended cements.

In this study, the performance of interground portland-limestone cements with up to 13% limestone by mass with SCMs was investigated in order to determine if the same amount of SCM dosage can be used with PLCs as with OPCs to mitigate ASR expansion.

## 2. MATERIALS AND EXPERIMENTAL METHODS

### 2.1 Materials

In this study, three interground PLCs (labelled as A\_L13, B\_L13, and C\_L10) and their respective OPCs (labelled as A\_OII, B\_OIIV, and C\_OV) were studied. In addition, a high alkali (HA) cement was tested as a control mixture. Various SCMs including two low to moderate (class F) fly ashes with different CaO contents (FA1 and FA2), a slag (SL), a silica fume (SF), and a natural pozzolan (NP) were used. Six mixtures that were studied are 25% FA1 (8.61% CaO), 20% FA1 + 5% SF, 50% SL, 25% FA1 + 25% SL, 25% NP, and 25% FA2 (12.54% CaO).

#### 2.1.1 Cements and SCMs

The chemical composition, d50, and d90 values of the cements and SCMs are listed in Table 2.1 and Table 2.2 respectively.

By comparing d50 and d90 values of the cements, OPCs and PLCs from cement B and C have an insignificant difference in their particle size. It was observed that OPC from cement A has a finer particle size distribution than its PLC.

Table 2.1: The chemical composition, d50, and d90 values of the cements used in the study

%	Cement A		Cement B		Cement C		High alkali cement
	A_OII (Type II)	A_L13	B_OIIV (Type II/V)	B_L13	C_OV (Type V)	C_L10	HA
SiO <sub>2</sub>	19.95	18.38	20.54	18.46	19.45	18.77	20.51
Al <sub>2</sub> O <sub>3</sub>	3.95	3.62	4.05	3.71	3.68	3.65	5.26
Fe <sub>2</sub> O <sub>3</sub>	2.28	2.07	3.62	3.46	3.35	3.18	2.11
CaO	63.32	61.69	61.72	60.45	60.32	59.43	64.20
MgO	1.43	1.33	2.52	2.28	4.45	4.33	1.40
SO <sub>3</sub>	2.55	2.48	1.80	1.71	2.73	3.03	4.28
Na <sub>2</sub> O	0.21	0.22	0.17	0.13	0.22	0.22	0.15
K <sub>2</sub> O	0.48	0.44	0.69	0.63	0.36	0.27	1.23
Na <sub>2</sub> O <sub>e</sub>	0.53	0.51	0.62	0.54	0.46	0.40	0.96
LOI	2.71	6.42	1.96	6.75	2.53	4.37	-
Limestone	4.31	13.32	1.79	13.11	4.20	10.00	-
d50 (µm)	10.52	15.70	11.77	12.57	12.53	12.33	12.87
d90 (µm)	27.89	59.18	29.94	37.06	27.90	30.75	37.03

Table 2.2: The chemical composition, d50, and d90 values of the SCMs used in the study

%	Fly Ash 1 (FA1)	Slag (SL)	Natural Pozzolan (NP)	Silica Fume (SF)	Fly Ash 2 (FA2)
SiO <sub>2</sub>	51.86	31.58	66.42	95.88	47.15
Al <sub>2</sub> O <sub>3</sub>	21.70	12.13	11.98	0.69	16.57
Fe <sub>2</sub> O <sub>3</sub>	5.04	0.55	0.86	0.12	5.88
CaO	8.61	41.34	4.06	0.70	12.54
MgO	2.58	6.97	0.18	0.26	4.80
SO <sub>3</sub>	0.78	3.75	0.19	0.15	0.60
Na <sub>2</sub> O	2.58	0.24	3.57	0.16	3.65
K <sub>2</sub> O	1.45	0.28	4.35	0.49	1.72
TiO <sub>2</sub>	1.19	0.47	0.09	0.01	1.17
P <sub>2</sub> O <sub>5</sub>	0.23	0.00	0.00	0.05	0.24
ZnO	0.02	0.00	0.00	0.06	0.01
Mn <sub>2</sub> O <sub>3</sub>	0.03	0.19	0.07	0.04	0.09
Cl	0.01	0.00	0.02	0.01	0.00
LOI	1.42	-	4.09	4.30	-
d50 (µm)	13.01	29.08	13.11	4.96	5.49
d90 (µm)	44.62	87.81	44.28	11.28	16.90

## 2.1.2 Aggregates

Borosilicate glass was used for the Pyrex mortar bar test. Two very-highly reactive fine aggregates (according to ASTM C1778), labelled as F1 and F2, were used for the accelerated mortar bar test. ASTM C1260 14-day expansions of F1 and F2 aggregates were 0.54% and 0.47% respectively.

## 2.2 Test methods

### 2.2.1 Pyrex mortar bar test (ASTM C441)

The Pyrex mortar bar test (ASTM C441) involves casting mortar bars (25 mm x 25 mm x 285 mm) using borosilicate glass with a standard gradation as fine aggregate. This test method is generally used to assess the relative effectiveness of a potential source of SCM to reduce expansion caused by ASR using borosilicate glass as a surrogate for a natural aggregate. For this study, the method was used to compare the performance of PLCs to their respective OPCs as well as with SCMs. As the fine aggregate used in this method is borosilicate glass, which is not used in the field mixtures, the test method only gives an indication of the relative performance of the cementitious materials with respect to alkali-silica

reaction and it does not predict the ability of a combination of cementitious material and a potential reactive aggregate to prevent ASR.

The test method involved casting three mortar bars per mixture. The mortars bars were stored in a vertical position in an air-tight container over water at 38°C. Each container accommodated two mixtures (six bars) per ASTM C441. The total testing period for the method was 14 days. The water to cementitious ratios used for the mixtures were based on the flow test (ASTM C1437) to produce a flow between 100 and 115 as per ASTM C441. The test setup was initially validated using a control set of mortar bars cast with high-alkali cement (0.96% Na<sub>2</sub>O<sub>e</sub>). Twenty-four mixtures were tested and the details are given in Table 2.3.

### 2.2.2 Accelerated mortar bat test (ASTM C1567)

The accelerated mortar bar test method was used to evaluate the effectiveness of SCMs to mitigate or control ASR expansion when very-highly reactive fine aggregates were used. Four mortar bars of dimensions 25 mm x 25 mm x 285 mm per mixture were cast according to ASTM C1567. Following the standard, the bars were immersed in 1 N sodium hydroxide solution at 80°C for 14 days. The length and mass measurements were made periodically. Eighteen mixtures were tested with F1 aggregate and eleven mixtures were tested with F2 aggregate. Most of the mixtures were tested with cement B as it has the highest alkali content among the three cements. The details of the test matrix are given in Table 2.3.

Table 2.3: Test matrix for the study

Cement	SCM proportions	ASTM C441	ASTM C1567	
			F1	F2
A_OII	25FA1	x	x	-
	20FA1-5SF	x	-	-
	50SL	x	-	-
	25FA1-25SL	x	-	-
	25NP	-	x	-
	25FA2	x	-	-
A_L13	25FA1	x	x	-
	20FA1-5SF	x	-	-
	50SL	x	-	-
	25FA1-25SL	x	-	-
	25NP	-	x	-
	25FA2	x	-	-
B_OIIV	25FA1	x	x	x
	20FA1-5SF	x	x	x
	50SL	x	x	x
	25FA1-25SL	x	x	-
	25NP	-	x	-
	25FA2	x	x	x
B_L13	25FA1	x	x	x
	20FA1-5SF	x	x	x
	50SL	x	x	x
	25FA1-25SL	x	x	x
	25NP	-	x	-
	25FA2	x	x	x
C_OV	25FA1	x	-	-
	20FA1-5SF	x	-	-
	50SL	-	x	-
	25FA2	-	-	x
C_L10	25FA1	x	-	-
	20FA1-5SF	x	-	-
	50SL	-	x	-
	25FA2	-	-	x

### 3. RESULTS AND DISCUSSION

#### 3.1 Pyrex mortar bar test results

Figure 3.1 shows the 14-day expansion of the Pyrex mortar bars with only cements tested.

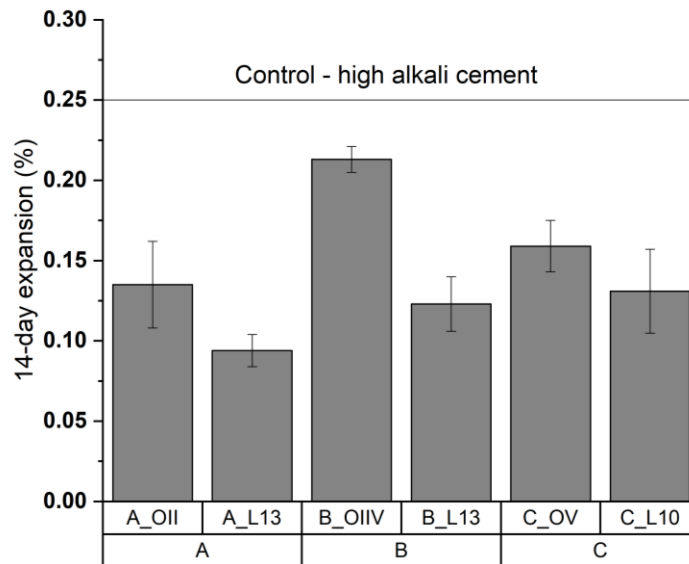


Figure 3.1: Pyrex mortar bar test results for OPCs and PLCs used in the study, the horizontal black line indicates the expansion using a high alkali cement.

From Figure 3.1, it was evident that the PLCs performed better than their respective OPCs. All the cements expanded less than the control-high alkali cement likely due to their lower alkali content compared to the control high alkali cement. B\_OIIV expanded the highest among the cements likely due to its relatively higher alkali content among the cements tested. It was observed that all the mixtures except A\_OII and C\_L10 met the maximum allowed standard deviation limit in ASTM C441. However, it should be noted that the precision and bias statement for ASTM C441 was done with high alkali cement (0.95 to 1.05% Na<sub>2</sub>O<sub>e</sub>) and not for cements with finely ground limestone. Figure 3.2 - Figure 3.6 show the 14-day expansion of the Pyrex mortar bar tests of the mixtures with cements and SCMs.

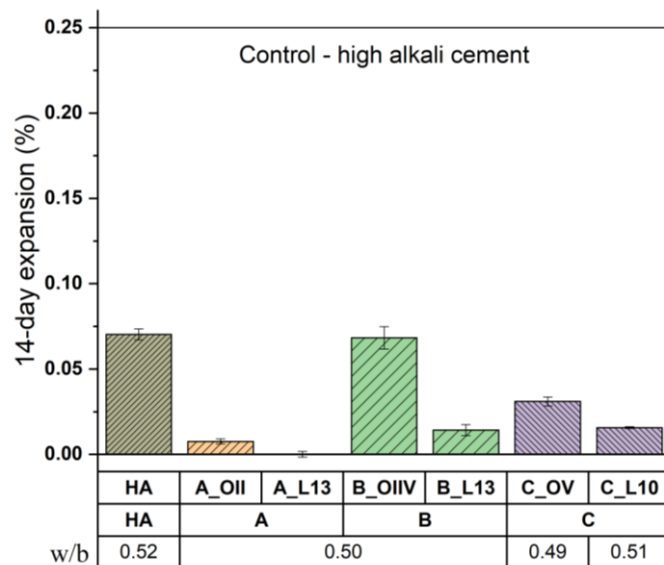


Figure 3.2: Pyrex mortar bar test results for the mixtures with 25% fly ash 1

From Figure 3.2, it was observed that all the PLCs with 25% fly ash 1 performed better than their respective OPCs with 25% fly ash 1.

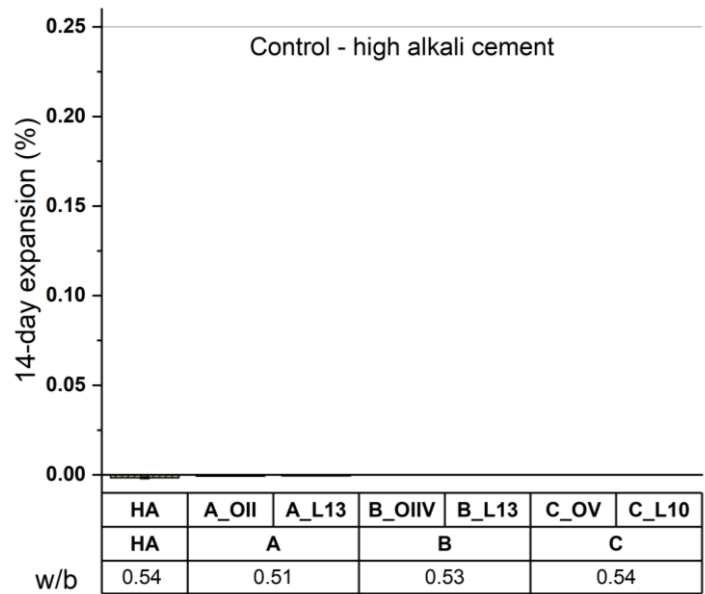


Figure 3.3: Pyrex mortar bar test results for the mixtures with 20% fly ash1 + 5% silica fume

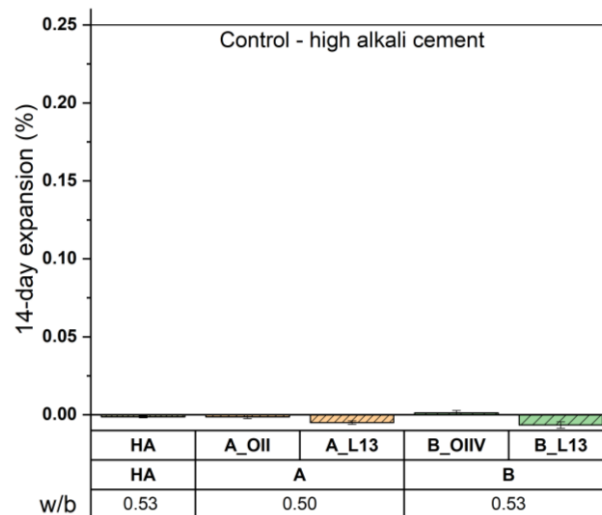


Figure 3.4: Pyrex mortar bar test results for the mixtures with 50% slag

In the mixtures with 20% fly ash 1 + 5% silica fume, 50% slag, and 25% fly ash 1 + 25% slag, the mortar bars did not expand or expanded insignificantly in both cases of PLCs and OPCs as shown in Figure 3.3, Figure 3.4, and Figure 3.5.

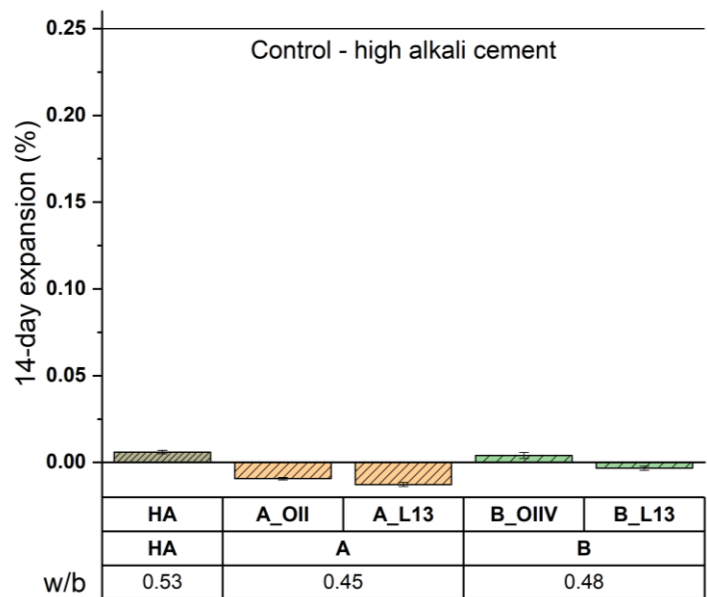


Figure 3.5: Pyrex mortar bar test results for the mixtures with 25% fly ash 1 + 25% slag

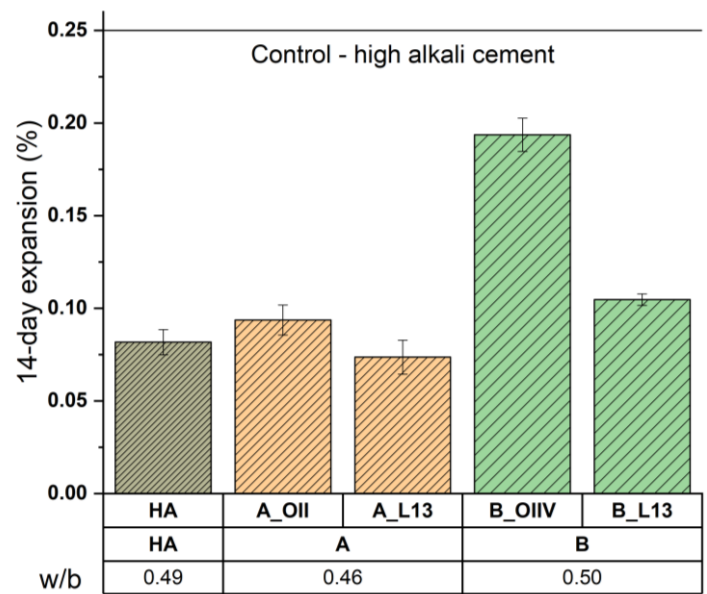


Figure 3.6: Pyrex mortar bar test results for the mixtures with 25% fly ash 2

In the case of mixtures with 25% fly ash 2, the PLCs had lower expansion than their respective OPCs similar to the other mixtures with SCM. Expansions for FA2 were generally higher however than the other SCMs investigated in this study. From the results of the ASTM C441 testing, a subset of PLCs was chosen for further study in ASTM C1567 (AMBT).

### 3.2 Accelerated mortar bar test results

Figure 3.7 shows the 14-day expansion results of the accelerated mortar bar tests with F1 aggregate.

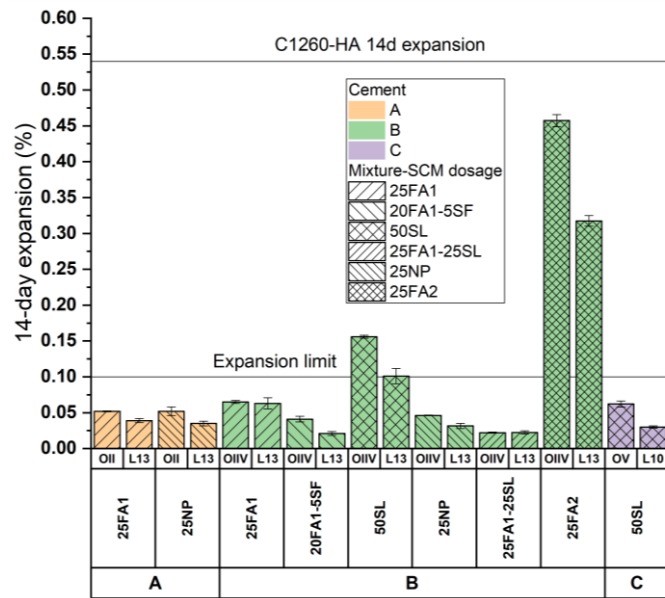


Figure 3.7: ASTM C1567 expansion results of the mixtures tested with F1 aggregate

From Figure 3.7, it was evident that the use of SCMs significantly reduced the expansion of the mortar bars. PLCs performed equal or better than the parent OPCs. All the mixtures expanded less than expansion limit of 0.10% except the 50% slag (B\_OIIV\_50SL, B\_L13\_50SL) and 25% FA2 (B\_OIIV\_25FA2, B\_L13\_25FA2) mixtures that included cement B. One of the reasons that 25% FA2 mixtures had expansion higher than 0.10% is probably due to its higher CaO/SiO<sub>2</sub> compared to FA1 that could have resulted in less alkali binding compared to FA1 and its lower Al<sub>2</sub>O<sub>3</sub> content (~16%) compared to FA 1 (~21%). Figure 3.8 shows the 14-day expansion results of the accelerated mortar bar tests with F2 aggregate.

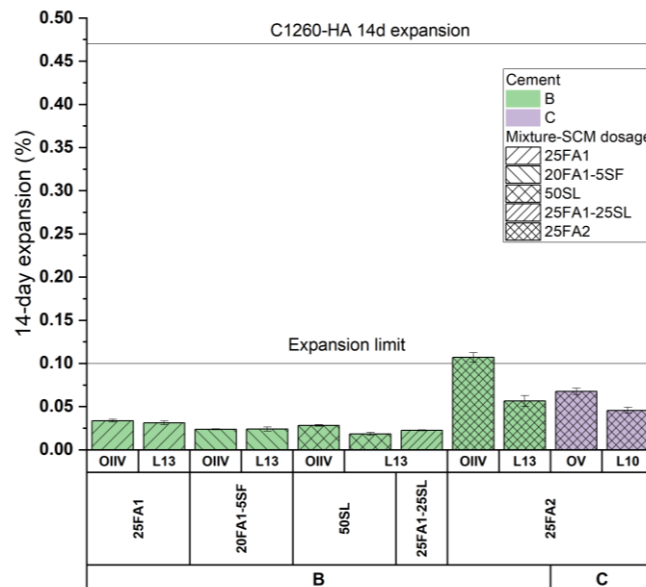


Figure 3.8: ASTM C1567 expansion results of the mixtures tested with F2 aggregate

From Figure 3.8, it was observed that all the PLCs with SCMs expanded equal or less than their respective OPCs with SCMs when they were tested with the accelerated mortar bar method with F2 aggregate. It was also observed that all the mixtures expanded less than 0.10% expansion limit with the exception of B\_OIIV\_25FA2 mixture. Similar to the AMBT with F1 aggregate, 25FA2 mixtures showed the highest expansions among all the mixtures. Though FA2 and slag were much more effective in



controlling ASR with F2 aggregate. It is interesting to note that even though both FA1 and FA2 have  $\text{CaO} < 18\%$  and  $\% \text{Na}_2\text{O}_e < 3$ , 25% FA2 mixtures with B\_OIIV failed the accelerated mortar bar test whereas 25% FA1 performed well. As discussed previously this could be due to higher Ca/Si and lower alumina content of FA2.

When PLCs are compared to their OPCs, PLCs have slightly less  $\% \text{Na}_2\text{O}_e$  but higher  $\text{CaO}/\text{SiO}_2$  when compared to their parent OPCs. Even though there is a slight dilution in total alkali content in PLCs, this alkali dilution would not affect the AMBT expansion due to the high concentration of the soak solution. It was shown that alkali dilution has a negligible impact on lowering the ASR expansion in the mixtures with SCMs in the accelerated mortar bar tests [3].  $\text{CaO}/\text{SiO}_2$  of the binder is directly related to pore solution alkalinity and expansion of mortar bars in the accelerated mortar bar test method [21,22]. Even though PLCs have slightly higher  $\text{CaO}/\text{SiO}_2$  when compared to their parent OPCs; PLCs showed lower AMBT expansions implying this slight increase in  $\text{CaO}/\text{SiO}_2$  of the blend did not influence the AMBT expansion significantly. Therefore, the most dominant mechanism contributing to better performance of PLCs in AMBT could be reduced mass transport as the presence of limestone is known to accelerate the hydration process by acting as nucleation sites for C-S-H formation and increase the pore refinement in cementitious systems. Mass transport properties and pore solutions of these mortars could be studied to better understand the contribution of PLCs with SCMs in mitigating alkali-silica reaction.

## 4. CONCLUSIONS

ASTM C441 (Pyrex mortar bar test) indicated that interground PLCs with up to 13% limestone performed better than their parent OPCs with and without SCMs. The accelerated mortar bar tests with two very-highly reactive fine aggregates showed that interground PLCs with SCMs performed similar or better than their respective OPCs with SCMs in terms of mitigating alkali-silica reaction expansion. The SCM dosages used in the study reduced expansions compared to the control with the majority of the mixtures being below the 0.10% expansion limit. Several additional mixtures are being tested according to AASHTO T380 and ASTM C1293 methods to verify if consistent results are seen in concrete specimens. The results of this study will provide further evidence if the mitigation options that are used with OPCs can be utilized as-is, increased, or decreased when used with PLCs.

## 5. ACKNOWLEDGEMENTS

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