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EFFECT OF ACTIVATOR COMPOSITION ON THE PERFORMANCE OF ALKALI-ACTIVATED SCC

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Abstract: This study investigates the influence of varying the concentrations and nature of various dry-powder alkali activator types on the workability and mechanical properties of alkali-activated self-consolidating concrete (AASCC) mixtures. Five dry-powder activator types; calcium oxide (CaO), sodium sulfate (Na₂SO₄), sodium silicate (Na₂SiO₃), sodium carbonate (Na₂CO₃) and silica fume (SF) were incorporated to activate different source materials i.e. ground granulated blast furnace slag (GGBFS) and fly ash (FA). A varying content of GGBFS between 40 to 100% was considered in the study. Fresh and mechanical properties including slump test, visual stability index and compressive strength were evaluated for all mixtures. Results indicated significant variations in the achieved compressive strength and workability as the type of activator was changing.

1 INTRODUCTION

The development of self-consolidating concrete (SCC) that can readily flows and consolidates under its own weight is considered one of the most efficient solutions in the world of construction nowadays. The successful development of SCC is largely depending on its high flowability without the tendency for segregation. A common practice promoted the development of SCC includes the incorporation of high powder content as a filler (i.e. size < 0.125 mm), less water to powder ratio together with new generations of efficient admixtures i.e. superplasticizers and viscosity modifiers as well as controlling the proportions and gradation of coarse aggregates with adequate sizes (typically less than 20mm) in concrete mixtures.

The SCC distinct features of excellent deformability and high resistance to segregation necessitates the use of high volume of cement and chemical admixtures that tend to increase the cost of concrete. It is estimated that the production of 1.0 ton of Portland cement requires about 2.8 ton of raw materials, fuel and other additives along with releasing around 1.3 billion tons of greenhouse gas emissions to the atmosphere due to the carbonation of lime process (Guo et al. 2010 and Malhotra et al. 2002). Therefore, the utilization of composite cements with a considerably larger volume fraction of industrial waste materials, as less expensive cement substitutes, were proven to enhance the SCC fresh and hardened properties (Uysal 2018; Santamaría et al. 2017 and Jerônimo et al. 2018). The two-fold advantages of such a practice are the enhancement of SCC rheological properties without the necessity to increase cement content; at the same time, decreasing the reliance on natural and non-renewable resources to encounter humans' developments. The use of by-product materials as supplementary cementitious materials (SCMs) was manifesting mainly within the partial replacement of cement binder. Whereas, the production of green structures cannot be completed until all the materials utilized are also green. By all means, cement is continuing to be an essential ingredient to make concrete despite the fact that too many environmental problems are associated with the process of its production (Mehta, 1999). Nowadays, the utilization of

entirely greener material that completely replace cement in concrete i.e. alkali activated materials (AAM) will directly contribute to positive environmental impacts.

AAMs, including those specified as geopolymers; derive their binding systems from the reactions between aluminosilicate source materials with alkaline solutions (activators). The resulted polycondensation network binds the aggregates and any inert material. The most commonly used aluminosilicate sources are similar to the used SCMs in the production of conventional concretes. A combination of liquid alkali activators (i.e. sodium silicate and sodium hydroxide) is usually the preference for alkali activation (Yang et al. 2008; Turner and Collins 2013). Although the area of alkali activation was known for about 80 years, the concerns associated with the rapid setting, efflorescence formation and accordingly high shrinkage (Atiş et al. 2009; Zivica V. 2007; Bakharev et al. 1999), carbonation and alkali silica reaction (Puertas et al. 2006; Bakharev et al. 2001a; Bakharev et al. 2001b; Byfors et al. 1989) are still questioned. In addition, the use of highly alkaline corrosive mediums (activators) have limited the scalability of this technique in the industrial-scale.

Up to present time, Alkali Activated SCC (AASCC) was not widely investigated as ordinary SCC as a result of the limited data on its rheological, mechanical and durability performance under high alkaline environments. Most of the previous research in AASCC was carried out using corrosive liquid activators of sodium hydroxide and sodium silicate under accelerated curing conditions (elevated temperature) to overcome the low early age strength of mixtures. Memon et al. (2011) showed that increasing the water content beyond 12% (i.e. 15% and 20%) over the designed mixing water had resulted in bleeding and segregation of mixtures in addition to a significant reduction in the strength. Also, curing samples at 70 °C for 48-hour was considered the optimum condition to enhance the compressive strength of the produced fly ash based AASCCs. Ushaa et al. (2015) showed that replacing fly ash with GGBFS by up to 30% had resulted in later age strength enhancement, while replacing it by silica fume up to 15% had resulted in more cohesive and sticky mixtures with early age compressive strength development. Shafiq et al. (2017) reported a reduction in the flowability of slag/ceramic waste AASCC mixtures as NaOH molarity and slag content were increased. A similar finding was reported by (Patel and Shah, 2018 and Sashidhar et al. 2015) that 12M NaOH represents a threshold, where exceeding this value results in a reduction in the strength. Manjunath and Narasimhan (2018) studied the production of AASCC mixtures using 100% acidic slag and reported that increasing the slag content up to 900 kg/m³ had resulted in a satisfactory mechanical and workability performance satisfying the EFNARC guidelines. However, it was claimed that the viscosity of sodium silicate as part of alkaline solution prevented the occurrence of segregation and bleeding.

Although the development of AASCC from an increasingly diverse range of waste-based precursors, activators, and admixtures has limited its acceptance, standardization, and production. The present study aimed at explore the potential for producing dry-powder AASCCs under ambient curing conditions with adequate flowability and strength. Different parameters were considered by researchers that well-thought directly affecting the fresh and hardened properties of AASCC i.e. type and fineness of mineral admixture(s), nature and dosage of alkali activator(s), chemical admixture(s), curing regime and mixing procedure.

2 Experimental Program

2.1 Materials

Reference concrete mixtures were prepared using ordinary Portland cement (OPC) and binary blended hydraulic cement Type HSb as the main binders for comparison. The OPC cement conforms to the CSA Type 10 (GU) and ASTM C150 (Type I) while the HSb cement conforms to the CSA Type 50 and ASTM Type V sulfate resistant Portland cement (SRPC). The AASCC mixtures were prepared using two types of source materials; ground granulated blast furnace slag (GGBFS) and fly ash (FA Class-F). The chemical analysis of all binders is illustrated in Table 1. For all AASCC mixtures, various dry-powder alkali activator materials were used for the activation of GGBFS and FA namely; calcium oxide (CaO), sodium sulfate (Na₂SO₄), sodium silicate (Na₂SiO₃), sodium carbonate (Na₂CO₃) and silica fume (SF). In addition, two types of chemical admixtures were selected for the study to improve the flowability and stability of the SCC mixtures. A polycarboxylate-based high-range water reducing admixture (HRWRA) conforming to the ASTM C-494 Types A and F and AASHTO M-194 Types A and F. In addition, a viscosity-modifying

admixture (VMA) based on a solution of modified polysaccharide was used conforming the ASTM C-494 (Type S) admixture.

Table 1: Chemical and physical properties of cement and source materials

Oxide (%)	SiO ₂	CaO	Al ₂ O ₃	Fe ₂ O ₃	MgO	K ₂ O	SO ₃	Na ₂ O	TiO ₂
OPC-GU	19.8	63.2	5.0	2.4	3.3	1.2	3.0	0.1	0.3
HSb	23.2	55.7	4.5	3.5	2.4	-	3.9	0.93	-
GGBFS	36.5	37.6	10.2	0.5	11.8	0.4	3.1	0.3	1.0
FA	48.9	3.8	23.3	14.9	0.7	1.7	0.2	0.6	-

To enhance the mixtures' packing density and to guarantee a well grain-size distribution. Natural siliceous sand with a fineness modulus of 2.5, specific gravity value of 2.68 and water absorption of 1.5% was used as fine aggregate. Furthermore, coarse aggregates with a maximum nominal size of 19mm, specific gravity value of 2.71 and water absorption of 0.4% were also used. Their volumetric contents were set to 60% and 40%, respectively. Figure 1 presents the particle size distribution of the used aggregates.

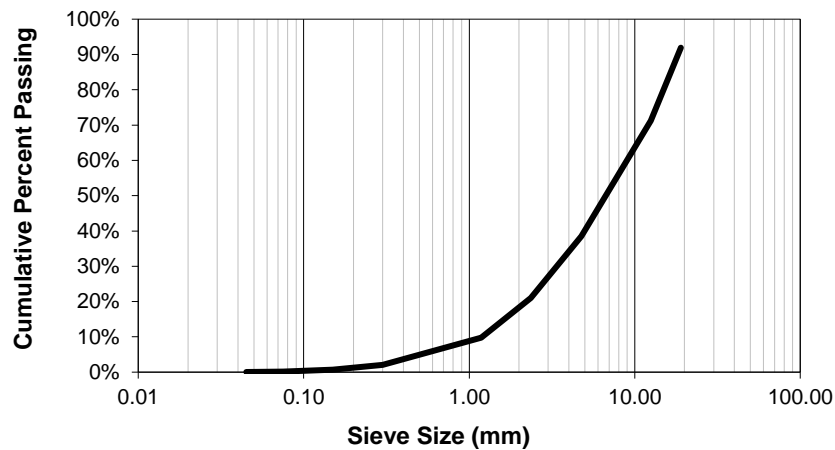


Figure 1: Particle size distribution of “all in” aggregates

2.2 Mixtures proportions

To produce eco-SCC intended for in-situ applications, cement was completely replaced by industrial waste materials activated by various dry-powder activators. The binder content was kept constant at 500 kg/m³ in either single (i.e. cement Type GU, Type HSb and GGBFS) or binary (i.e. GGBFS/FA) combination. Thus, the total AASCC binder content with the added dry-powder activator concentrations of 4.5% (Na₂SO₄ or Na₂CO₃) and 10% (SF or Na₂SO₄) was 522.5 and 550 kg/m³, respectively. The water/binder ratio was 0.38 for all mixtures. The specified binder content was selected to increase the paste volume at constant water/binder ratio for durability purposes (Bernal et al. 2011).

The selection of the various dry-powder activators in the production of AASCC mixtures was based on their contributions to the total greenhouse gas emissions to the atmosphere. To the best of authors' knowledge, the combination between sodium silicate (Na₂SiO₃) and sodium hydroxide (NaOH) solutions is usually the common and preferable choice for the alkali activation process (Yang et al. 2008; Turner and Collins 2013). However, difficulties associated with their corrosiveness, hygroscopicity of NaOH and low solubility of Na₂SiO₃ had limited their effectiveness as dry-powder activators in the alkali activation domain. Therefore, Table 2 presents the composition of 7 AASCC mixtures using various activators with less embodied energy and carbon dioxide emissions.

2.3 Mixtures preparation

The AASCC mixtures were prepared by blending the as-received dry powder source materials and activators for about 2 minute in a pan mixer. This was followed by the addition of sand and blending for 2 minutes. After that, water and chemical admixtures were added to mixtures and mixing continued for 2 minutes. At last, aggregates with a saturated surface dry condition were included and mixing continued for 2 minutes. The total duration of the mixing procedure was 8 minutes.

Several trial batch mixtures were conducted to evaluate the degree of mixtures' flowability using different admixture dosage rates of HRWR and VMA. All mixtures were evaluated based on their fresh behavior relative to (EFNARC, 2005) by maintaining i.e. flowability of 650 ± 30 mm and T_{500} between 2 - 5 seconds. In addition, visual inspection (no segregation and no bleeding) was used as a secondary quality control measure for the fresh AASCC mixtures. After workability testing, both fresh reference mixtures (C-1 and C-2) as well as AASCC mixtures were cast in cylinders with dimensions of 100mm diameter x 200mm height and left in a standard laboratory temperature for 24 h. AASCC cylinders were sealed using plastic sheets to prevent the moisture loss and to eliminate the efflorescence evolution. The mechanical characteristics in terms of compressive strength of the produced mixtures were evaluated after 7 days according to the ASTM C 39 (2005).

Table 2: AASCC cement compositions

AASCC components		Cement ID								
		C-1	C-2	A-1	A-2	A-3	A-4	A-5	B-1	B-2
Source Material (wt.%)	OPC-GU	100								
	HSb		100							
	GGBFS			94	94	90	80	80	44	40
	FA								50	60
Dry-powder activator (wt.%)	CaO			3	3	5	20	20	3	
	SF			3	3	5	10*		3	
	Na ₂ SO ₄			4.5*		4.5*		10*		
	Na ₂ CO ₃				4.5*				4.5*	8
	Na ₂ SO ₃									8

*Added activator by wt. %

3 RESULTS AND DISCUSSIONS

3.1 Workability of AASCC mixtures

In this study, the fresh state properties in terms of the slump flow of AASCC mixtures containing different GGBFS (%) were studied and are shown in Figure 2. According to EFNARC guidelines, the basic workability requirements for acceptable SCC is the excellent deformability, minor blockage rate and adequate stability. A standard slump cone was used where both reference and alkali-activated SCCs mixtures were poured in the cone without compaction. The obtained results from the slump cone represents the value of the mean diameter (measured in two perpendicular directions) after lifting the cone. According to Nagataki and Fujiwara (1995), slump flow values <500 mm might indicate a poor flow of concrete to pass through congested reinforcement while flow >700 mm might cause segregation in the mixtures. The ideal condition according to authors is to have a slump flow diameter ranging between 500 to 700 mm for concrete to be self-consolidating. Results showed that the reference concrete mixtures (i.e. C-1 and C-2) have met the required slump flow values with 733 and 750 mm, respectively. However, AASCC mixtures using 100% GGBFS activated by various activators showed slump flow values ranging between 430 and 470 mm (flow <500 mm). Surprisingly, replacing GGBFS by FA did not enhance the flow but resulted in a very low slump flow values <300 mm.

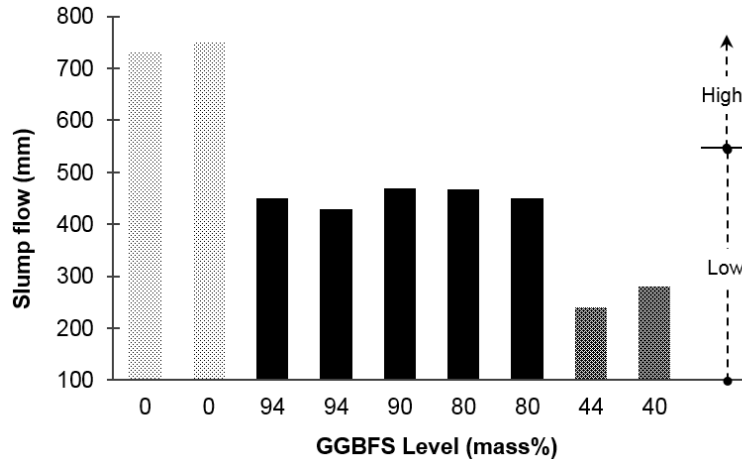


Figure 2: Slump flow test results for AASCC mixtures

3.1.1 Effect of precursor nature and content

Workability is one of the key performance indicators that describes the fresh behavior of mixtures starting from blending the ingredients till placement. Several factors were well thought affecting the workability of AASCC mixtures such as content and fineness of source materials, nature and dosage of the dry-powder activators and chemical admixtures. In 100% ordinary (Portland and HSb) cements, the workability increased due to the formation of cementitious products that led to a decrease in the mixtures' viscosity. On the other hand, increasing GGBFS content in AASCC mixtures tend to increase the ultimate resistance to shear which lead to a more viscous blend (Glukhovskiy et al. 1981). In general, using lower GGBFS-to-FA ratio would offer a better workability (in terms of flowability) performance in their blends due to their different morphology and structural properties. In this study, it was observed that replacing GGBFS with FA by up to 50% produced the lowest flow values. This might be due to the effect of dry-powder activators used in the production of AASCCs, efficiency of the used chemical admixtures and source material fineness. As an attempt to understand the workability behavior of dry-powder mixtures, around 25% extra water was added to most of the blends. The addition of extra water to alkali-activated concretes was observed in mixtures activated by liquid activators to enhance the workability. It should be noted that the addition of extra water over the designed water content showed some bleeding in mixtures (i.e. A-2) activated by dry-powder activators. The addition of extra water had decreased the pH of the blend that led to a decrease in the strength.



Figure 3: Slump flow test for (A-2) AASCC mixture

3.1.2 Effect of admixture type

With respect to the influence of admixtures (HRWR and VMA) on the plastic viscosity of GGBFS-based and control mixtures, Table 3 shows that the presence of only 2% HRWR admixture in the control mixtures have lowered the plastic viscosity of the two blends leading to a satisfactory workability performance meeting the EFNARC guidelines. On the other hand, the presence of the same admixture types in the AASCC blends (at a constant water to binder ratio) was not efficient. Admixtures were added with dosages up to 10% to increase the workability of the blends. A numerous number of studies were oriented to study the effectiveness of water-reducing admixtures, originally designed for OPC concretes, and on alkali-activated concretes showed inconsistent results. This might be due to the use of various source material and activators in addition to the admixture nature. Bakharev et al. (2000) concluded that the efficiency of regular admixtures in alkali-activated concrete depends on the activator type and can be opposite to the effect of OPC-based concretes. Also, admixtures are often have to be used in large dosages for workability enhancement, and may have significant side effects i.e. strength loss and increased shrinkage. Overall, it can be concluded that the high alkalinity of the blend media have affected the stability of the used admixture causing a reduction in its efficiency.

3.1.3 Effect of dry-powder activator

In this study, it was found that the dry-powder activators have a great effect on the rheological properties of AASCCs. The nature, type and concentration of activators have significant effects on the workability of tested mixtures. Generally, replacing GGBFS by FA would increase the workability of mixtures due to its ball bearing effect as a result of its special spherical shape. However, activating mixtures with SF tend to decrease the rheological properties of mixtures due to its high specific surface area and water demand. The substitution of GGBFS by either FA or SF with up to 20% was reported in several studies (Gifford and Gillott 1997; Collins and Sanjayan 1999). These papers reported an enhancement in the workability of concrete mixtures, yet this behavior may vary depending on the nature of activator accompanying. The main reason of workability loss in AASCC mixtures activated by Na_2CO_3 might be the initial precipitation of calcium carbonate that led to a dramatically decrease of the spread flow. In addition, Na_2CO_3 increases the water demand of mixtures.

The improvement of pastes fresh behaviour was observed in OPC-based mixtures while incorporating lime as a filler (Zhu et al. 2005; Svermova et al. 2003). This might be due to the better packing of mixtures as lime particles are usually finer than slag particles leading to a better workability behaviour. Therefore, the incorporation of lime as a key activator in some of the AASCC mixtures was investigated and showed an increase in the flowability of the mixtures to a limit. However, the combination of various activators with lime (i.e. Na_2CO_3) and GGBFS showed a decrease in the spread value due to the reduced volume of water in the mixtures. This might be attributed to the dissolution process of GGBFS particles by Na_2CO_3 that requires water to be completed. A study conducted by (Chen and Liao 1992) concluded that the addition of lime to a Na_2SO_3 -activated slag cement when the silicate modulus is <1.5 caused a quicker loss of workability due to the consumption of water.

3.2 Compressive strength

The early age strength development of the control mixtures in addition to the AASCCs incorporating dry-powder activators are shown in Table 3. It was concluded that the strength development was very sensitive to the activator type and content. Increasing the activator content from 6% to 10 % (lime + SF) in mixtures A-1 and A-3 resulted in about 53% reduction in the compressive strength at age of 7 days. All of the AASCC mixtures were cured under laboratory ambient conditions. It is estimated that heat curing would result in a higher strength elaboration rate. Results presented in Table 3 also showed that the strength development of AASCC was depending on the activator type and nature. Mixtures prepared by Na_2CO_3 dry-powder activator exhibited a very low strength development due to the low solubility nature of the used Na_2CO_3 in pure water under low temperature. The solubility of Na_2CO_3 increases with temperature until reaching 35.4 °C, after which the solubility rate gradually drops. Furthermore, the used Na_2CO_3 also known as “washing soda” occurs in a crystalline decahydrate structure that when dissolved in water absorbs the heat from the blend and elongates the setting time. The crystals of Na_2CO_3 in AASCC mixtures showed a potential to

efflorescence in dry air forming mainly a residue of low hydrates known as monohydrate. Similarly the utilization of Na_2SO_4 in AASCC mixtures exhibited efflorescence (Fig. 4).



Figure 4: Efflorescence problem appeared in AASCC mixtures

Table 3: AASCC fresh and mechanical results

Cement I.D.	GGBFS (%)	Activator (%)	Slump flow (mm)	Admixture (%)	Visual Stability		Compressive Strength (MPa)
					Efflorescence	Bleeding	
C-1	0	-	733	2.0	-		56.6
C-2	0	-	750	2.0	-		63.5
A-1	94	10.5	450	6.5	Moderate	✓	9.1
A-2	94	10.5	430	9.0	Mild		10.8
A-3	90	15.0	470	9.0	Severe	✓	4.8
A-4	80	30.0	467	5.5	-		14.4
A-5	80	30.0	450	5.0	Moderate	✓	12.7
B-1	44	10.5	240	8.0	-		N.A
B-2	40	16.0	280	9.0	-		N.A

N.A: Not Available as the sample did not harden at age 7 days.

4 Conclusions

This study aimed to investigate the potential production of dry-powder Alkali-activated SCC using GGBFS as a main source material and multi-dry powder activators i.e. Na_2SO_3 , Na_2CO_3 , SF, Na_2SO_4 and lime as an alternative to cement in conventional concrete mixtures. The study focused on identifying the main parameters affecting the performance of different AASCC mixtures including the type and fineness of source materials, nature and concentration of activators as well as the efficiency of OPC-based chemical admixtures. The following conclusions can be drawn from the results of this study:

- The workability results (in terms of flowability values) of the AASCC mixtures including different activators nature and Type II /G admixture (HRWR) is not clear and needs further investigation.
- Combinations of lime and SF had accelerated the hardening and strength development of dry-powder AASCC concretes containing GGBFS and Na_2CO_3 to the extent of their promising behaviour for in situ applications and without elevated heat curing.

- To overcome the problems associated with the low early age strength, source materials and dry-activators should go under a milling process to increase the amorphosity of the particles, enhance the dissolution and reactivity of AASCC mixtures (Tang et al., 2012; Paya´ et al., 1997).
- Efflorescence was apparently obvious in dry-powder AASCC mixtures due to the leaching out of alkalis which in turn react with CO₂ in the air to form alkali carbonates. The efflorescence appeared on the surfaces of some mixtures and was controlled by reducing the alkalis concentration (dry-powder activator) in addition to the inclusion of SF.
- Further research is needed to estimate different engineering properties of AASCC concretes containing dry-powder activators.

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4.2 References

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