



Effect of admixture on the pore structure refinement and enhanced performance of alkali-activated fly ash-slag concrete

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HIGHLIGHTS

- Relation between workability and setting adjustment and admixture dosage is derived.
- Compressive strength is significantly enhanced by the plasticizing admixture.
- Microstructure refinement by the plasticizing admixture is observed by SEM analysis.
- Pore structure and porosity are investigated by mercury intrusion porosimetry.
- Chloride migration resistance is affected by the admixture at different ages.

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ABSTRACT

This paper investigates the influence of a plasticizing admixture on the pore structure refinement of alkali-activated concrete and paste mixtures and the consequently enhanced performance. Alkali-activated fly ash-slag concrete and paste are designed using a polycarboxylate-based admixture with different dosages. The pore structure and porosity are analyzed using mercury intrusion porosimetry (MIP). The workability, compressive strength, chloride migration resistance and electrical resistivity of alkali-activated fly ash-slag concrete and paste are determined. The results show that significantly improved workability and strength development are obtained at an increased admixture content. The admixture improves the gel polymerization product layer most likely around the GGBS particles, densifying the matrix. The 28-day Cl-migration coefficient of admixture (1–2 kg/m³) modified concrete is equal to the reference mixture, while at the highest admixture content the Cl-ingress is increased. At the later ages (91-days), the Cl-migration coefficients of all concretes, non- and admixture-containing samples, are comparable and low (about 2.6×10^{-12} m²/s). The MIP analyses show a significant decrease of the total and effective capillary porosity over time at an increased admixture content. The relationships between the porosity and other properties are discussed, at varying admixture contents.

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1. Introduction

Alkali-activated slag-fly ash based binders in comparison with traditional Portland cement possess comparable to moderately modified material properties (i.e. mechanical strength, chloride ingress, acid and carbonation resistance) [1–4]. Designing alkali-activated materials (AAM) with high durability performance lar-

gely depends on the mixture composition (design). This is mainly controlled by the applied precursor minerals such as ground granulated blast furnace slag (GGBS) and pulverized coal fly ash (PCFA), and the concentration, type and combination of alkaline activators (i.e. sodium or potassium silicate or hydroxide). More specifically, a higher GGBS content (0–100 wt%) as a replacement of the PCFA in the binder, favors the matrix densification and strength development [5–8]. By forming mainly calcium dominated gel-structures (C-A-S-H), consequently resulting in a reduced chloride migration rate in concrete [9]. However, to support the practical application and further development of AAM as well as that of Portland cement, both materials are strongly dependent on the availability

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of admixtures [10,11]. Due to the existence of multiple molecular varieties, admixtures (known as superplasticizers (SPs)) can perform very differently in optimizing the fresh concrete mixture state, although this is also dependent on the binder type and composition [12].

For fly ash-dominated AAM systems, the mixture workability, setting time and liquid demand can be relatively easily modified by polycarboxylate and naphthalene type admixtures [13–17]. Although often relatively high admixture dosages (≥ 1 –10 wt% in relation to the binder content) are required in order to gain a high mixture flowability and consistency [15,17–19], compared to that of Portland cement mixtures (mostly ≤ 1 wt%). As a consequence, high dosages lead to unwanted negative side effects, such as increased material porosity and loss of mechanical strength [15,17]. For GGBS-dominated AAMs, almost all related admixtures often do not sufficiently modify the mixture workability [20]. In some cases, mixture rheology improvement and setting time retardation are observed to a certain extent when using only a hydroxide instead of a silicate-based activator [15,20]. Overall, many of these studies indicate that admixtures are able to reduce the liquid to binder ratio or liquid demand of the fresh mixture. Summarizing relevant literature dealing with the effect of mainly Portland cement-based admixtures on AAM systems, the following remarks can be drawn:

- Admixtures shows no improvement to the delay of the mixture setting time and overall mixture workability, which could be associated with their physical and chemical incompatibility or rapid chemical oxidation in the high alkali system [13,15,20].
- Admixtures mainly enhance the AAM mixture workability over a short period of time (≤ 10 –40 min). An increasing GGBS and silicate activator content strongly reduce the workability, therefore AAM is often prone to a non-predictable, very rapid decline of the workability and fast setting [15,16,18,20–22].
- Naphthalene and polycarboxylate admixtures are the most effective SPs, to enhance the mixture workability of mainly alkali-activated PCFA systems [13,15,16] compared to GGBS-based systems [15,18,22,23].
- Admixtures use frequently causes negative effects on the setting time and mechanical property of AAM [13,15–18,20–22,24].
- Admixtures can have either negative or positive effects on the concrete shrinkage [20,24].

Limited experimental studies have been performed on the effect of admixtures on AAM systems as often admixtures are not able to sufficiently modify AAM concrete [20,25]. However, in recent years, more, while still rather limited commercial admixtures, mainly polycarboxylates, are available for AAM. Further knowledge is required in order to improve the physical and chemical understanding of their working mechanisms, as well as that of their predictability with AAM concrete production.

Apart from the rheology-modifying ability [14], another positive effect is that the microstructure development of AAM concrete can be significantly enhanced by a polycarboxylate. Through densification of the interfacial transition zone (ITZ) [26], located between the newly formed AAM gel structure and solid particles (binder and the aggregate minerals), at which the porosity is reduced. This leads to the shift of pore size of hardened AAM towards smaller ranges which improves the material performance by for instance a higher material strength, reduced permeability and enhanced ion diffusion resistance (e.g. chloride). However, the microstructure development (i.e. porosity and permeability) and the chloride migration of AAM concrete under the influence of using admixture have not been studied. Research is needed to understand the potential physical and chemical mechanisms

affected by a working admixture in concrete, contributing to the design of durable AAM concretes for construction structures.

In the present study, a comprehensive approach is applied to investigate the effect of admixture on pore structure refinement and enhanced performance of alkali-activated fly ash-slag concrete. The main objectives of this study are:

- Analyze the influence of the admixture content on the fresh mixture state properties, by measuring the setting time and the workability progression over time;
- Study the influence of the admixture content on the hardened material state properties, by analyzing the AAM compressive strength, chloride migration rate and material electrical resistivity over time;
- Determine the effect of admixture content on the AAM pore structure and progression over time and consequent influence on the material durability (chloride migration);
- Investigate the relation between different system parameters and their significance; such as concrete compressive strength and porosity and their effect on the permeability and chloride migration of concrete over time under the influence of the admixture content.

2. Experimental setup

2.1. Materials

The used mineral binder (MB) is a blend of 73.7 wt% pulverized coal fly ash (PCFA) class F in accordance with NEN-EN 450-1 with 25 wt% granulated ground blast furnace slag (GGBS) and 1.3 wt% technical grade sodium meta-silicate pentahydrate powder (supplied by PQ, The Netherlands). The elemental composition of the MB is determined by X-ray fluorescence (XRF), as shown in Table 1.

River aggregates (sand 0–4 mm and gravel 4–16 mm) were used to produce the mixtures. A commercial 33% liquid sodium hydroxide (NaOH) with a molarity (M) of 11.2 was diluted by tap water to obtain the desired (3M NaOH) system alkalinity. A polycarboxylate plasticizing admixture (supplied by SQAPE Technology), hereafter identified as “admixture”, was used to enhance the fresh concrete workability. The polycarboxylate is highly soluble in water and the backbone contains poly-functional reactive side chains, e.g. carboxyl, which initiate the metal (mainly calcium) adsorption reactions. Preliminary research shows that a chemical oxidation effect is observed when mixing this admixture with NaOH solution that helps to improve the workability. The added additional water, NaOH and the admixture were summed as the total liquid volume (although solids are present). The relevant

Table 1
Elemental composition (%) of the mineral binder (MB), determined with XRF.

Oxides	PCFA	GGBS	MB
SiO ₂	59.7	34.3	51.4
Al ₂ O ₃	24.6	9.8	17.9
CaO	1.5	41.8	13.9
Fe ₂ O ₃	6.8	0.5	6.3
MgO	1.3	7.7	3.8
K ₂ O	3.0	0.6	2.2
Na ₂ O	0.6	<0.1	1.1
TiO ₂	1.2	1.2	1.1
Mn ₃ O ₄	0.0	0.3	0.2
BaO	0.1	0.1	0.1
P ₂ O ₅	0.1	<0.1	0.4
SO ₃	1.0	3.6	1.7
Cl	<0.1	<0.1	<0.1
LOI (950 °C)	0.9		1.6

LOI: loss of ignition.

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