



The influence of superabsorbent polymers on the autogenous shrinkage properties of cement pastes with supplementary cementitious materials



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ABSTRACT

Fly ash and blast-furnace slag containing binders are frequently used in the construction industry and it is important to know the extent of autogenous shrinkage and its (ideal) mitigation by superabsorbent polymers in these systems as a function of their age. In this paper, the autogenous shrinkage was determined by manual and automated shrinkage measurements. Autogenous shrinkage was reduced in cement pastes with the supplementary cementitious materials versus Portland cement pastes. At later ages, the rate of autogenous shrinkage is higher due to the pozzolanic activity. Internal curing by means of superabsorbent polymers is successful, independent of this long term higher rate of shrinkage in mixtures with supplementary cementitious materials.

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1. Introduction

When fresh mortar is subjected to drying, contraction may take place. This plastic shrinkage may cause cracking during the first few hours after casting [1]. Other forms of shrinkage include autogenous shrinkage as a result of cement hydration, thermally-induced shrinkage and drying shrinkage due to the loss of water to the surroundings and shrinkage due to carbonation. As cement reacts with water hydration products will precipitate in the water-filled spaces between the solid particles in the cementitious material. The water in the remaining small capillaries forms menisci and exerts hydrostatic tension forces. These capillary forces reduce the distance between the solid particles, leading to autogenous shrinkage.

Autogenous shrinkage may result in micro- and macro-cracks which impair strength, durability and aesthetics, especially in mixtures with a low water-to-binder ratio. During hardening, the internal relative humidity (RH) will drop in these mixtures and self-desiccation will occur if no external water source is present. This will lead to self-desiccation shrinkage. The cracks interconnect flow paths for water and gases, possibly containing harmful substances [2]. For example, the ingress of water could induce steel corrosion, frost attack, chemical attack and internal expansive reactions, endangering the durability.

One should mitigate autogenous shrinkage to reduce the amount of shrinkage cracks to increase the service life of a structure. Internal curing is the provision of water within the specimens to maintain the internal RH during self-desiccation and it can mitigate plastic and

autogenous shrinkage. Several materials can be used to promote internal curing in concrete: lightweight aggregates, pumice, expanded clay, and superabsorbent polymers (SAPs) amongst others [3,4]. The application of SAPs for this purpose proved to be promising as autogenous shrinkage was reduced and even counteracted in time [5,6].

SAPs have the feature to absorb up to 500 times their own weight in aqueous solutions due to osmotic pressure, resulting in the formation of a swollen hydrogel. The SAPs are long chains of linear polymers which are interconnected at several points. Nowadays, they are used in the hygiene and medical industry as care articles or smart pills, and they can also be used for firefighting or food packaging [7]. It was only a matter of time until this polymer also found its way as an additive in cementitious materials [8]. SAPs can be used in cementitious materials for reducing the autogenous shrinkage [5,6,9–12], for changing the rheology of the fresh material [5,6,13], for increasing the freeze/thaw resistance [14,15], for self-sealing [16–18] and even to promote autogenous healing [19,20]. Superabsorbent polymers can be used for internal curing, but they can thus also induce other features upon crack formation. The focus in this paper lies in the first original function of SAPs in cementitious materials, namely the mitigation of autogenous shrinkage in cement pastes, and in particular in cementitious materials with supplementary cementitious materials.

During preparation of a concrete mixture, the SAPs will take up mixing water. The SAPs will thus form water-filled inclusions, useful for internal curing [9]. The water released due to self-desiccation during cement hydration can be used for further hydration and reduction of the autogenous shrinkage [10]. The water present in the SAP will hereby be released into the cementitious matrix due to the imminent drop in relative humidity. Due to this water release, the internal relative humidity

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is maintained. The SAP particles shrink and an empty macro pore remains as shown by means of neutron tomography measurements [21].

In systems with a water-to-cement ratio of 0.3 complete hydration is not possible. The hydration stops at a minimum porosity of the cement gel (26–28%). The capillary water reacts until it is consumed and the cement starts to react with the harder bound gel water, causing the relative humidity to decrease and the cement paste to self-desiccate. With an additional (entrained) water-to-cement ratio of 0.054 through pre-described water-filled SAP inclusions, internal curing takes over and the self-desiccation is mitigated [5,22]. There are less gas-filled micro pores and the degree of hydration is higher. Compared to a system with a water-to-cement ratio of 0.35, the maximum degree of hydration is the same and the overall porosity is the same as well. The difference is that in the SAP entrained system, macro pores are more present than the finer capillary pores present in the system without internal curing.

Brüderm and Mechtcherine [23] showed (in uncracked concrete) internal curing due to the presence of SAP and a decrease of the autogenous shrinkage and self-desiccation by the gradual release of water from the SAP particles. Jensen and Hansen [6] measured up to 3700 microstrains ($\mu\text{m}/\text{m}$) of shrinkage in reference cement paste in a period of 3 weeks. When SAPs were used, there was a successful mitigation of shrinkage and even some expansion. Igarashi and Wanatabe [11] showed successful mitigation of shrinkage with an amount of 0.7% SAPs by mass of cement in a paste with a water-to-cement ratio of 0.25. Mönning [15] visualised the densification of the matrix around a SAP particle due to the larger amount of water available in time. The amount of entrained water that is needed to promote the internal curing effectively is 0.18 times the water-to-cement ratio [5]. This entrained water is additionally added to the cementitious mix and held by the SAPs during mixing [5]. The determination of this amount is based on the theory of Powers and Brownyard [22]. The ideal amount of superabsorbent polymers to provide internal curing will be applied in this research. This amount will be based on the theoretical amount of entrained water needed for internal curing [5,6,22].

All abovementioned results were obtained for concrete made with ordinary Portland cement with or without silica fume [6]. As nowadays concrete structures are frequently made with binders containing other supplementary cementitious materials (SCM) like fly ash and blast-furnace slag, the effect of SAPs on the autogenous shrinkage results of those mixtures also needs to be studied in detail. Publications describing the effects of SAPs on internal curing and shrinkage mitigation for concrete with these SCMs, are scarce.

The cement clinker and supplementary cementitious materials hydrate simultaneously and influence one another. The concrete composition and pore fluid affect the overall hydration and pozzolanic reactions (by fly ash) are known to occur later as pozzolans need to react with the calcium hydroxide formed in cement hydration. Yan and Chen [24] found that for mixtures with a water-to-binder ratio of 0.30 a reference with OPC (at 192 days) showed 320 $\mu\text{m}/\text{m}$ shrinkage strain, FA15 (with 85% of cement and 15% of fly ash in the binder) 250 $\mu\text{m}/\text{m}$ shrinkage strain, FA30 220 $\mu\text{m}/\text{m}$ shrinkage strain and FA45 150 $\mu\text{m}/\text{m}$ shrinkage strain. As the degree of hydration of fly ash increases, the autogenous shrinkage also increased in time [25]. De la Varga et al. [26] found that autogenous shrinkage in high volume fly ash systems is lower than in non-fly ash mortar due to the fact that the fly ash systems contained a lower amount of cement, so that the initial reaction rate and water consumption was lower. Fly ash starts to react at later ages and consequently less autogenous shrinkage is monitored at early ages. At later ages, the rate of shrinkage is higher in fly ash mortars compared to pure cement systems. De la Varga et al. found 200 $\mu\text{m}/\text{m}$ shrinkage strain for a mixture with cement and a water-to-binder ratio of 0.30, 100 $\mu\text{m}/\text{m}$ shrinkage strain for FA40 (fly ash), 50 $\mu\text{m}/\text{m}$ shrinkage strain for FA60, 20 $\mu\text{m}/\text{m}$ expansion strain for FA60IC (internal curing) and 60 $\mu\text{m}/\text{m}$ expansion strain for FA40IC

at an age of 14 days. In the latter research, internal curing by means of lightweight aggregates proved to be promising to mitigate autogenous shrinkage in fly ash systems. However, the influence of the internal curing in mixtures containing fly ash should also be investigated at later ages than 14 days. This data is lacking in literature. Data from 14 days onwards would be very useful as the net overall shrinkage for fly ash systems may then exceed the one for the pure cement mixtures.

Blast-furnace slag is another commonly used addition in the building industry. Blast-furnace slag mixtures showed greater autogenous shrinkage than ordinary concrete in the research of Lee et al. [27]. The higher the blast-furnace slag amount, the greater the autogenous shrinkage. They found 310 $\mu\text{m}/\text{m}$ shrinkage strain for a cement mixture with a water-to-binder ratio of 0.32, 380 $\mu\text{m}/\text{m}$ shrinkage strain for 32BFS30 (70% of cement and 30% of blast-furnace slag with a water-to-binder ratio of 0.32) and 420 $\mu\text{m}/\text{m}$ shrinkage strain for 32BFS50 at an age of 200 days. Also, concrete made with BFS showed higher shrinkage in the first days than concrete made with Portland cement [28]. Lura et al. [28] addressed this phenomenon to the supposed denser structure of the BFS cement paste which showed smaller pores. These smaller pores lead to higher capillary forces for self-desiccation, increasing the autogenous shrinkage. One of the few studies found in literature, by Wyrzykowski and Lura [29], focused on the effect of internal curing on ordinary Portland cement and blast-furnace slag blended cement pastes. They found for blast-furnace slag blended cement mixtures, that a part of the autogenous shrinkage was eliminated due to internal curing, but the remaining shrinkage was very high. The shrinkage was approximately 1400 $\mu\text{m}/\text{m}$ shrinkage strain for BFSC-REF (blast-furnace slag blended cement mixture), 600 $\mu\text{m}/\text{m}$ shrinkage strain for BFSC-SAP (mixture with BFS and internal curing), 200 $\mu\text{m}/\text{m}$ shrinkage strain for OPC-REF (ordinary Portland cement mixture) and 50 $\mu\text{m}/\text{m}$ shrinkage strain for OPC-SAP at an age of 7 days. Results from 7 days onwards are again missing in literature.

In this paper, the setting of mixtures with varying amounts of FA or BFS is studied by means of the Vicat needle test and the autogenous strain is monitored in time. Both manual and automated tests are hereby used to study the effects of internal curing by means of superabsorbent polymers on the early age and later age autogenous shrinkage properties. The results will prove to be useful for studying the shrinkage properties of cement pastes with supplementary cementitious materials.

2. Materials and methods

2.1. Materials

A cement paste with a water-to-binder ratio (W/B) of 0.3 was made. The cement used was CEM I 52.5 N (Chemical composition in Table 1). Furthermore, mixtures containing different amounts of supplementary

Table 1

Chemical composition of CEM I 52.5 N, Class F fly ash and blast furnace slag and specific surface (Blaine fineness).

	CEM I 52.5 N mass-%	Class F fly ash mass-%	Blast furnace slag mass-%
CaO	63.12	2.47	40.38
SiO ₂	18.73	49.34	34.35
Al ₂ O ₃	4.94	24.55	11.36
Fe ₂ O ₃	3.99	6.23	0.48
SO ₃	3.07	0.30	1.65
MgO	1.02	1.73	7.57
K ₂ O	0.77	3.84	0.37
Na ₂ O	0.41	0.52	0.29
Cl ⁻	-	-	0.013
S ²⁻	-	-	0.77
Mn	-	-	0.165
Specific surface	390 m ² /kg	330 m ² /kg	400 m ² /kg

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