Influence of crystalline admixtures and bacteria on the fresh properties of self-healing concrete

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Abstract. Concrete is undeniably susceptible to cracking due to its low tensile strength in spite of its ability to withstand high compressive stresses. Cracks may jeopardize the concrete core and reinforcements due to the ingress of water and aggressive substances. A promising technique to effectively seal cracks in hardened concrete is using a self-healing technology by introducing healing agents to the fresh concrete mixes. Crystalline admixtures and bacteria emerge as promising self-healing agents to promote stimulated autogenous healing and autonomous healing mechanisms, respectively, improving simultaneously the durability of the concrete. Healing agents are mostly added on top of the normal mix without changing the mix design of the concrete, while in fact, it may induce considerable effects on the fresh and hardened properties of concrete. In this study, two types of cement (CEM I 52.5N and CEM III/A 42.5N) as well as crystalline admixtures and bacterial spores, both in powder form, and PCE-based superplasticizer are utilized to evaluate the changes in consistency and setting time. The dosage of healing agents is taken between 0−5% by weight of powders. To start, tests on paste level were executed. The results show that the addition of healing agents affects the water demand, the consistency and setting time of the pastes. The inclusion of crystalline admixtures increases the water demand, while a declined trend of setting time is observed. The gradual addition of bacteria up to 5% slightly changes the water demand, but a significant delay on the initial and final setting times was detected.

Keywords: self-healing concrete, crystalline admixture, bacteria, fresh properties, workability

1 Introduction

During the past decades, self-healing concrete has been extensively experimented and proposed to upgrade the performance of conventional concrete with an ability to seal the undesirable cracks independently. Crack occurrence in concrete is apparently inevitable and this leads to considerable maintenance cost and labor works if continuous repairs are needed. Furthermore, there is no guarantee that the repair will be efficient and will not induce more defects at other locations. Thus, a stimulated autogenous or autonomous healing of the concrete is proposed by introducing healing agents such as crystalline admixture and bacteria. The use of crystalline admixture is broadened from a porosity reducer to a promotor of self-healing effect [1]. Active chemicals present in this admixture react in the presence of water, forming precipitated crystals that increase the density of calcium-silicate-hydrate gel (CSH), and working in that way as a pore/crack blocker [2]. The implementation of bacteria as a self-healing agent in the concrete matrix emerges as one of the most promising approaches in order to extend the service life of concrete structures by sealing the concrete cracks on a biotechnology basis. Microbial precipitation of calcium carbonate inside the crack occurs due to the metabolic activation of bacteria with suitable nutrients in contact with moisture [3]. The phenomenon of intrinsic crack closure was noticeable on cementitious composite containing healing agents such as bacteria [4] and crystalline admixture [1,5]. Nowadays, the two main goals of adding healing agent into the concrete are to activate the healing mechanism and to improve the healing efficiency, however, the evaluation of fresh properties of concrete mixes with the inclusion of healing agents is often neglected [6]. Workability is an important aspect for fresh concrete to ensure that it can be easily placed and handled in the field. Therefore, understanding the influence of adding healing agent in relation to the fresh concrete properties is of great importance. In this study, two commercial healing agents (i.e. Basilisk bacteria healing agent and Penetron Admix crystalline admixture) are introduced into the fresh paste to investigate the effects on the consistency and setting time of the paste.

2 Materials and Methods

2.1 Materials

Two types of cements (CEM I 52.5N and CEM III/A 42.5N, abbreviated as CEM I and CEM III) and demineralized water were used to make the fresh pastes. The chemical compositions of these cements are presented in Table 1. Penetron Admix crystalline admixture (CA) and Basilisk bacteria spores (BAC), both in powder form, were utilized as healing agents. The particle size distributions (PSDs) of cements, bacteria and crystalline admixtures are presented in Fig. 1. The PSDs were obtained by laser diffraction analysis. Specifically for the bacteria healing agent, laser diffraction analysis was complemented with sieve analysis because particles larger than 300 μm were present, exceeding the upper size limit of the laser diffraction apparatus. The PSD of CA is in the range of the cements, but a bit broader. The median sizes (D_{50}) of CEM I, CEM III, CA and BAC are 17.16, 11.73, 15.20 and 490.24 μm.

According to the manufacturer, the recommended dosage of healing agent is in the range of 0.8–1.0% by weight of cement for Penetron crystalline admixture and 1.0–4.0% by weight of cement for Basilisk bacteria healing agent. However, an attempt was made to use a higher dosage of healing agent to possibly increase the self-healing efficiency without lowering other properties such as workability. Healing agents, both CA and BAC, were finally added in the range of 0.8–5.0% by weight of powder. The incorporation of healing agents is mostly carried out on top of the normal mix, without any adaptation in the mix design. In this study, the healing agents are added into the cement paste by changing the powder proportion of the mixture. For instance, the powder composition of the paste CEM I + 3% BAC is composed of 97% CEM I and 3% Basilisk bacteria. Additionally, the polycarboxylate-ether (PCE) based superplasticizer (SP) of Demula Technofluid P175 supplied by Cugla B.V. was also used as a high-range water reducer (HRWR). The properties of this superplasticizer are given in Table 2.

Table 1. Chemical composition of the cements

Chemical composition (%)	CEM I 52.5N	CEM III/A 42.5N
CaO	64.35	53.58
SiO_2	21.00	27.39
Al_2O_3	4.22	6.83
Fe_2O_3	2.72	1.82
SO_3	3.29	3.02
CO_2	1.00	1.04
Cl	0.07	0.05
MgO	2.04	4.66
K_2O	0.80	0.59
Na_2O	0.24	0.27
P_2O_5	0.49	0.22

Table 2. Properties of superplasticizer (SP)

SP type	Chemical type	Density (kg/dm^3)	Solid content (%)	pH
Demula Technofluid P175	Polycarboxylate ether	1.07	30	2.0 – 8.0

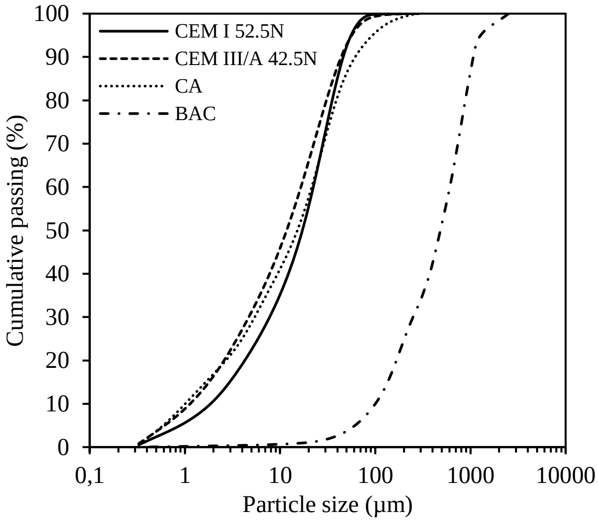

Fig. 1. Particle size distributions of cements and healing materials.

2.2 Testing methods

In this study, initial tests on the fresh cement paste in combination with healing agents were executed. To start, the standard consistence test and setting time test in accordance with EN 196-3:2016 were carried out. The standard consistency test is performed by use of the manual Vicat apparatus (with the use of a plunger) to determine the water demand of each modified powder mixture in comparison to pure cement without a healing agent. On the other hand, the automatic Vicat apparatus is also used to measure the initial and final setting time of the pastes. In practice, superplasticizers are widely used in concrete mixtures to improve the workability. Consequently, an additional experiment was executed to investigate the effect of superplasticizer in relation to the reduction of water demand. The superplasticizer is added in the dosage of 0.1–2.0% by weight of powders (cement + healing agent).

3 Results and discussions

The results on water demands of cement in combination with BAC and CA at standard consistency are presented in Fig. 2. It shows that the gradual addition of BAC slightly reduced the water demand. The combination of CEM III with 5% BAC reduces the water demand from 29.5 (pure CEM III) to 27.7% (CEM III + 5% BAC). In combination with CEM I, the reduction induced by the presence of BAC is apparently minor as the water demand reduces only from 28 to 27%. As shown in Fig. 2b, the behavior of CA is completely different on the use of different cement types which is also in contrast with the result of using BAC. The result shows that the water demand is higher in presence of CA (for all dosages from 0.8 to 5% by the weight of powder). On the use of

CEM III, the amount of water needed to achieve a standard consistency raises from 29.5 to 34.5%. Although it is clear that, in combination with this type of cement, the water demand in case of 4% CA is higher than in case of 5% CA, a linear regression line was plotted (Fig. 2b). Contrarily, the result of combining CA and CEM I shows a different behavior. The tendency seems to be a parabolic regression where the gradual addition of CA up to 3% increases the water demand by 3%, while the higher dosage of CA shows a declined trend as the water demand reduces from 31 to 29.5% when the CA dosage is increased from 3 to 5%. By comparing these two results on water demand, in fact, the addition of CA showed a similar behavior on both types of cements when the dosage was limited to 3%.

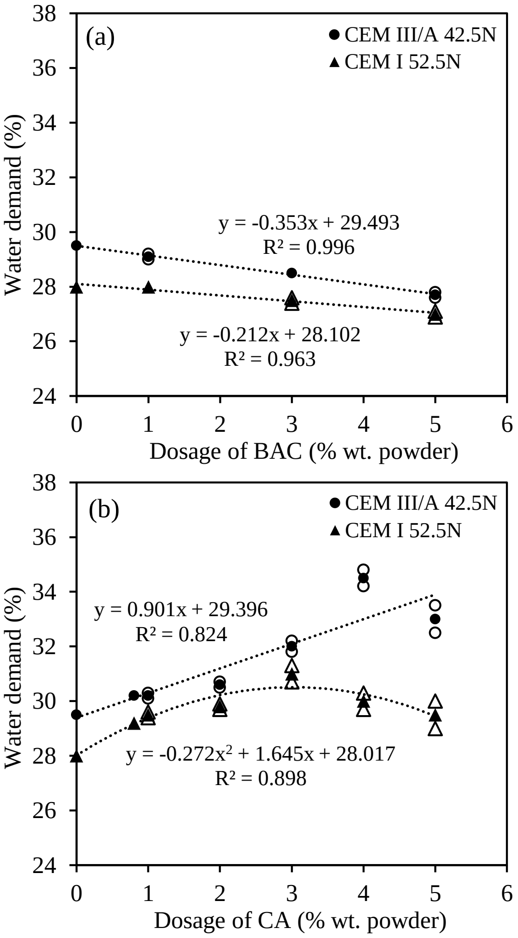

Fig. 2. Water demands of modified cement incorporated with (a) Basilisk bacteria and (b) Penetron crystalline admixture at different dosages (note: the empty markers represent the water demand values, while the filled markers represent the mean values)

As a matter of fact, as reported by [7], in case CA is used, it is important to make a judicious selection regarding the type of cement. Cement with a high content of clinker, such as CEM I, is preferred because CA needs an appreciable amount of calcium hydroxide, which is a typical hydration product of Portland cement, to react. In the case of blended cement like CEM III, less calcium hydroxide is produced and can be even consumed.

Moreover, the setting time test was evaluated and results are shown in Fig. 3. In spite of the rather small effect by BAC on water demand, the setting time of these pastes considerably increases as a consequence of incorporating BAC into the fresh mix. Based on Fig. 3a, both initial and final setting times are delayed regardless the cement type. However, a considerable effect is identified on the mixture containing 5% BAC and cement type I. The final setting times of cement pastes (with CEM I) are 175, 221, 311 and 660 min with the addition of 0, 1, 3 and 5% BAC, respectively. It is clear that the setting time of cement paste with 5% BAC is roughly doubled in comparison with cement paste with 3% BAC. Moreover, this exceptional result is not observed in the paste with CEM III. On the use of CEM III, the addition of BAC from 1 to 5% gradually increases the setting time of the paste. The final setting times of cement pastes (with CEM III) amount to 227, 256, 333 and 430 min with the addition of 0, 1, 3 and 5% BAC, respectively. In general, the mean gap time between initial and final setting time of paste containing BAC is approximately 63 ± 9 min. Rossi et al. [8] investigated the cement hydration with the addition of Basilisk bacteria healing agent by means of an isothermal calorimetry test and they found that the addition of BAC caused a decrease of heat developed by the hydration of cement, resulting in the retardation effect. Due to the reason that BAC is composed of organic compounds, the alkaline degradation of these compounds into molecules containing the α-hydroxy carboxyl group may be responsible for the delay in the setting time of the cement pastes [8,9].

The setting time of paste containing different CA dosage (0.8–5% by weight of powder) is presented in Fig. 3b. Similar with the consistency test results, the inclusion of CA in combination with CEM I and III shows different behavior. On the use of CEM I, the 1% CA addition increases the initial and final setting time of the paste by approximately 63 and 77 min, respectively. The higher the addition of CA the faster the setting time of the paste. Specifically the gradual addition of CA from 1 to 5% shortens the final setting time from 252 to 183 min. A significant effect in using high dosage of CA was the fast setting of the paste. By using the high CA dosage at 4 and 5% by weight of powder, the initial setting times were found to be at 68 and 22 min. From the visual observation by authors, the cement paste incorporated with a high dosage of CA was quickly stiffened after the mixing process. This may be attributed to a chemical reaction occurring when the CA came into contact with water and cement, resulting into a fast setting time. This was supported by the fact that the white powder present in CA particles which presumed to be active chemicals immediately dissolved in water. From the experiment, it was observed that when the CA came into contact with water, the mixing water turned into greenish color, indicating an immediate reaction and a hydrophilic character of CA which can react easily with water [10]. However, further investigations should be conducted to analyze these results in detail. Furthermore, it is clear that the addition of CA in combination with CEM III shows similar setting times regardless the dosage. As a matter of fact, the use of high CA dosage at 4–5% have a comparable result with the low dosage at 1% in the terms of setting time.

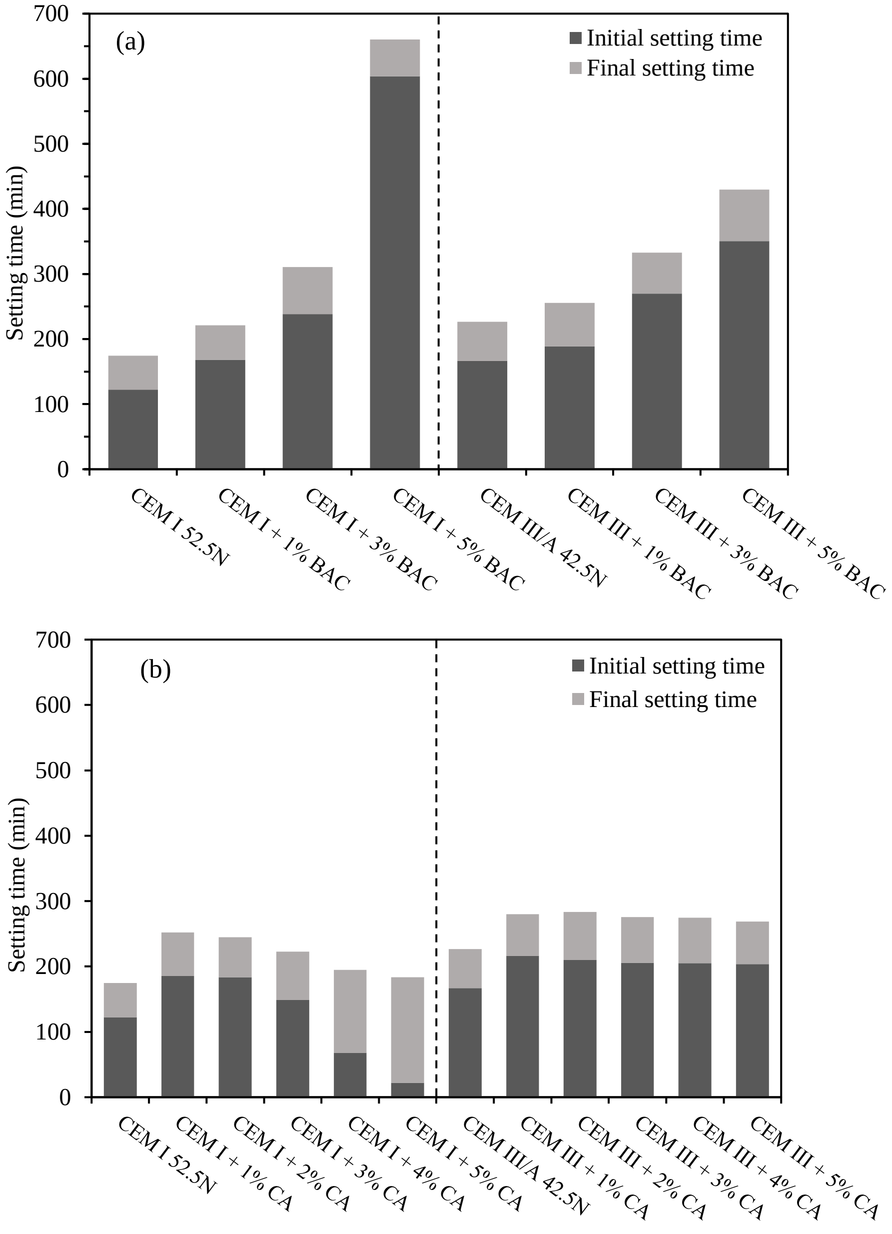

Fig. 3. Setting times of modified cement pastes incorporated with (a) Basilisk bacteria and (b) Penetron crystalline admixture at different dosages

Additionally, the superplasticizer (SP) was added into the modified cement paste with healing agent, either BAC or CA, at the dosage between 0.1 and 2.0% by weight of powders (cement + healing agent) to measure the water reduction of the paste at normal consistency and to observe the compatibility aspect between healing agent and superplasticizer. Fig. 4 shows the relationship between SP dosage and total water demand in combination with different dosage of healing agent. The BAC was fixed at the dosage up to 5%, while the dosage of CA was limited up to 3% to eliminate the unfavorable effect on the use of a higher dosage as explained in the previous results. As a note, the water demand was corrected with the water contained in the SP solution, and is further defined as total water demand.

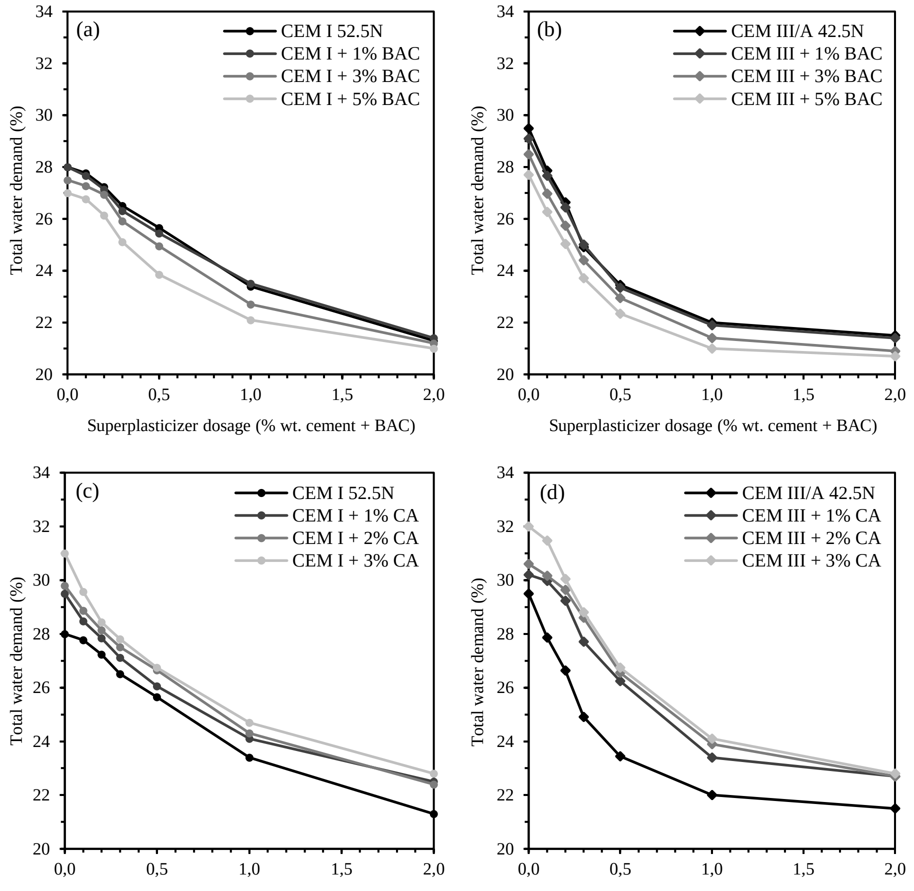

Fig. 4. Changes on total water demand of modified powder mixture due to the incorporation of different healing agents and superplasticizer in combination between (a) BAC + CEM I, (b) BAC + CEM III, (c) CA + CEM I and (d) CA + CEM III

According to Figs. 4a and 4b, the reduction rate of water demand is relatively similar for control paste and paste containing BAC. As shown in Fig. 4c, there is an immediate reduction of water demand on cement pastes with CEM I containing CA at a low SP dosage. However, in general, the reduction rate of water demand with varying SP dosages of all modified pastes was quite similar with the control paste. Contrarily, this was not the case for the modified paste with CEM III and CA. As shown in Fig. 4d, the water demands of CA and CEM III are always higher than the pure cement and in addition, the changes in water demand of CA and CEM III upon increasing SP dosage do not follow the behavior of water demand reduction of the pure CEM III. This behavior can be explained by the fact that the superplasticizer is adsorbed by the CA particles and the incorporation of CA considerably influences the rheological properties of the superplasticized CEM III paste by reducing the workability and increasing the viscosity of the paste [11]. Nevertheless, the incorporation of the superplasticizer decreases the water demand at all dosages of healing agents (CA or BAC), while the variations in the water demand are generally lower at a higher dosage of SP.

4 Conclusions

The effects of incorporating bacteria and crystalline admixtures in relation to the fresh properties of cement pastes were evaluated based on consistency tests and setting time tests. In addition, the reduction of water demand was investigated with the addition of a PCE-based superplasticizer. The major conclusions are presented as follows:

1. The water demand of cement with Basilisk bacteria slightly reduces as the bacteria dosage increases up to 5%, while the water demand of cement with Penetron crystalline admixture increases as the CA dosage increases up to 3%.
2. The addition of Basilisk bacteria prolongs the setting time and the higher the bacteria dosage, the longer the setting time. In contrast, the gradual addition of CA from 1 to 5% by weight of powder into the cement paste with CEM I tends to shorten the setting time. The impact of using a high CA dosage on fresh CEM I paste is rapid initial setting and loss of plasticity. However, the initial and final setting times of paste containing CA and CEM III/A are relatively similar at any CA dosage.
3. As a consequence of incorporating a PCE-based superplasticizer into fresh pastes, the water demand of cement and healing agent reduces with increasing dosage of superplasticizer. In addition, there are no incompatibility issues found between cement, crystalline admixture, bacteria and superplasticizer.

Furthermore, future works will focus on the introduction of bacteria or crystalline admixture into the fresh concrete mixture to identify the influences of healing agent on the workability aspect at concrete level.

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