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Influence of mix composition on the extent of autogenous crack healing by continued hydration or calcium carbonate formation

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HIGHLIGHTS

- Crack healing by continued hydration can be improved using alternative binders.
- For $CaCO_3$ precipitation, it is not a benefit to replace cement by fly ash or slag.
- The regain in mechanical properties due to autogenous crack healing is limited.
- Autogenous healing can reduce the water permeability of cracked specimens.
- Autogenous healing seems to be more efficient to close small cracks.

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ABSTRACT

Crack formation is an inherent property for cementitious materials, however, these materials also possess the ability to heal cracks in a completely autogenous way. Upon ingress of water, unhydrated cement particles, available at the crack faces, may further hydrate and cause crack healing. When both water and carbon dioxide become available inside the crack, healing occurs due to carbonation of leached calcium hydroxide.

The influence of using alternative binder materials, such as blast furnace slag and fly ash, on the extent of autogenous crack healing was investigated in this study. Blast furnace slag and fly ash react slower compared to cement and thus more unreacted binder material remains available within the hardened matrix to cause crack healing by further hydration. However, slag and fly ash consume calcium hydroxide during their reaction and thus the potential for calcium carbonate formation may be reduced.

Isothermal calorimetry and water permeability measurements showed that replacement of Portland cement by these supplementary cementitious materials resulted in higher degrees of ongoing hydration upon contact of hardened cement paste with water. The effect of adding blast furnace slag or fly ash on calcium carbonate precipitation was evaluated through microscopic analysis of the change in surface crack width of specimens subjected to wet/dry cycles. Cement replacement by blast furnace slag or fly ash did not seem to improve crystal precipitation. Besides, it was shown from this experiment that smaller cracks closed more completely and faster compared to larger cracks. Also when regain in strength of samples subjected to wet/dry cycles was evaluated, no differences were seen between the different test series.

In general, it can be concluded that cement replacement by blast furnace slag or fly ash improves autogenous healing by enhancing further hydration, while it does not affect the amount of calcium carbonate precipitation. The advantageous effect is most pronounced when blast furnace slag is used as cement substitute.

1. Introduction

The phenomenon of autogenous crack healing in cementitious materials is already known for many years and has already been studied by many researchers [1–15]. Cracks in old structures, like Roman aqueducts or gothic churches, are healed when moisture contacts unhydrated cement grains inside the crack [16]. This causes secondary hydration reactions, leading to crack healing. In addition, crack healing may result from leached calcium hydroxide ($Ca(OH)_2$) which converts into calcium carbonate ($CaCO_3$) crystals when both water and carbon dioxide (CO_2) become available. This is noted as white crystalline deposits at the crack surface.

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However, current concrete structures show a reduced autogenous healing capacity as cement grains are not as coarse as before. Due to the use of finely ground cement particles, less unhydrated material remains available for autogenous crack healing to occur.

Abdel-Jawad and Dehn [12] and Granger et al. [13] demonstrated that using a low water-to-binder (W/B) ratio results in an increased amount of unhydrated cement and thus advanced autogenous healing. Some researchers proposed to make use again of coarse cement grains or to embed encapsulated cement in the matrix for improvement of the autogenous crack healing efficiency [17]. However, increasing the use of cement, to improve autogenous healing, is not beneficial as the production of cement contributes largely to the greenhouse effect. Moreover, using more cement will significantly increase the cost of concrete and may increase the amount of autogenous shrinkage and thus the risk of cracking.

Recently, several researchers [18–28] started to investigate whether the addition of mineral admixtures may promote the autogenous crack healing efficiency of concrete. In this research, it was not our aim to adapt the concrete composition to improve the healing efficiency. However, as it is known that currently used alternative binders such as blast furnace slag and fly ash react slower compared to cement, we wanted to investigate whether the longer availability of unhydrated particles influences the autogenous crack healing efficiency.

While the active component in cement, Portland clinker, is a hydraulic binder, blast furnace slag, a residue from the steel industry, has latent hydraulic properties. This means that the hydration reaction with water needs to be activated by substances such as $Ca(OH)_2$, e.g. from clinker hydration. Fly ash, a waste product from coal combusted power stations, has pozzolanic properties, which means that it binds with $Ca(OH)_2$ that is formed during clinker hydration. So for both binder materials, first cement needs to hydrate partly in order to activate or initiate the reaction.

While blast furnace slag and fly ash concrete seem to be inferior with regard to the early age microstructure and strength development, their self-healing capability can be much higher, precisely because of the slow hydration of slag and fly ash. Upon cracking, the unreacted particles can be activated again by available $Ca(OH)_2$ in order to close the crack and to regain water impermeability and strength. However, as both slag and fly ash consume $Ca(OH)_2$ during hydration, their potential to result in $CaCO_3$ formation, may be reduced.

In their study, Jacobsen et al. [29] noticed that the degree of self-healing varied with the concrete composition. Liu et al. [30] demonstrated that the addition of fly ash is helpful in improving the self-healing performance of concrete. While Şahmaran et al. [31] compared the efficiency of fly ash replacement ratios of 0%, 35% and 55%, Termkhajornkit et al. [32] used percentages of 0%, 15%, 25% and 50%. They noted that the healing ability increased when the fraction of fly ash increased. Other researchers used replacement percentages of 55% and 70% [33] or even 85% [34]. However, they used the binder materials inside strain hardening fibre reinforced cementitious composites to restrict the crack width and they did not study the effect onto the possibility of autogenous crack healing.

Qian et al. [35] replaced part of the cement by blast furnace slag to make their strain hardening fibre reinforced cementitious material more environmentally friendly. Again the effect of this replacement on ongoing hydration was not studied. This was done so by ter Heide [36]. Contrary to her expectations, she noticed that less healing occurred for blast furnace slag mixes compared to mixes made with ordinary Portland cement.

Liu and Zuo [37] were up to now the only researchers who compared the efficiency of both blast furnace slag and fly ash replacement on the improvement of autogenous healing. They concluded that the self-healing ability of concrete increased by increasing the amount of added slag or fly ash.

As blast furnace slag and fly ash react slower compared to Portland clinker, they increase the time span that unreacted binder is available. However, one may speculate how long the slag and fly ash systems can maintain their self-healing ability. This is related to how long blast furnace slag and fly ash can continue to hydrate. As stated by Termkhajornkit et al. [32] there are two factors: first, the available space for the hydration products and second, the essential compounds for the self-healing reaction, unreacted binder and $Ca(OH)_2$.

It seems that hydration products form within pores. When no pores can be filled up anymore, the hydration reaction stops. However, if space is generated by cracks, the reaction may restart again when water becomes present.

Concerning the second factor, Baert [38] found that the hydration degree of fly ash in pastes with a fly ash-to-binder ratio of 0.5 amounted to 28% after 2 years. For pastes (W/B ratio of 0.5) containing blast furnace slag, it was found that the slag hydration degree after 2.5 years decreased from 70% to 40% when the slag-to-binder ratio increased from 50% to 85% [39]. The fact that the hydration degree decreases with increasing proportion of slag in the blend was also confirmed by other researchers [40]. This can most probably be ascribed to the decreasing amount of available produced portlandite. As such a large amount of unreacted binder remains present even after years, slag and fly ash systems exhibit a long-term ability for continued hydration to occur. However, the question is whether enough $Ca(OH)_2$ remains available to activate or initiate the hydration reaction or to result in autogenous crack healing due to $CaCO_3$ formation.

In this study, the influence of different binder types and W/B ratios on the autogenous crack healing efficiency has been studied by using isothermal calorimetry, microscopic analysis, water permeability tests and mechanical tests. A distinction was made between autogenous healing due to continued hydration and $CaCO_3$ precipitation.

Samples used for the isothermal calorimetric measurements and the water permeability test were only exposed to water and thus continued hydration was the only mechanism which could occur. Samples used for the microscopic analysis and the mechanical test were exposed to wet and dry cycles so not only water but also CO_2 became available. Consequently, for the latter experiments crack healing could result from both continued hydration and calcium carbonate formation. However, when cracks were investigated at the surface with the microscope, $CaCO_3$ precipitation seemed to be the main mechanism while strength regain was influenced by both mechanisms.

2. Materials

2.1. Mixture compositions

In order to investigate the influence of the mix composition on the extent of autogenous crack healing by continued hydration or $CaCO_3$ formation, cement paste and mortar samples with varying binder composition and W/B ratio were prepared.

2.1.1. Cement paste

Three different cement types were used to prepare the cement paste mixes: CEM I 52.5 N (CEM I), CEM II/B-M 32.5 N (CEM II) and CEM III/B 32.5 N (CEM III). As represented in Fig. 1, CEM I cement only contains Portland clinker. CEM II cement contains 65–79% Portland clinker while the remaining 21–35% consists of a mixture of blast furnace slag, fly ash and limestone. CEM III cement only holds 20–34% Portland clinker while the remaining 66–80% consists of blast furnace slag. A cement paste mixture was made with each of these cement types. Therefore, 450 g cement was mixed with 180 g tap water, so a W/B ratio of 0.4 was obtained for these mixes (see Fig. 1).

Other mixes were obtained by replacing part of the cement (CEM I) by blast furnace slag or fly ash. For mixtures represented by 50 BFS, 70 BFS and 85 BFS, respectively, 50% (225 g), 70% (315 g) and 85% (382.5 g) of the cement weight was