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Self-healing concrete using bacillus subtilis natto immobilized in lightweight aggregate

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KEYWORDS

Self-healing Concrete
Bacteria
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ABSTRACT

Concrete is probably the most important and commonly used construction material. However, concrete still faces quality degradation problems caused by cracks. Concrete is probably the most important and commonly used construction material. However, concrete still faces quality degradation problems caused by cracks. In leading to sustainable development, management methods will be better than repairing the cracks in concrete. With high ability to be cultured and $CaCO_3$ precipitation, the potential to use Bacillus subtilis natto in full-scale application is promising. Although the bacteria were transformed into spores before mixing to concrete, their survival is still minimal. In this study, the high survival rate of bacteria immobilized in lightweight aggregate reflected through later-age-crack strength restoration rate contributes essential information for maintaining self-healing for a long time. Experiments also clarifies the bio-mineralization of Bacillus subtilis natto with the effects of nutrient-low medium to find a suitable way to protect and maintain the self-healing abilities in a long time. The effect of cracking age on the self-healing capacity associated with the compressive strength improvement was clarified.

1. Introduction

In the field of construction materials, recent research showed that the crack criteria early age concrete can occur as soon as the cement matrix become hardened and the cracks are usually in micro-size and difficultly controlled. Inspired by natural and biological systems, self-healing concrete was studied and developed. Self-healing in cementitious materials, especially concrete can either be autogenic or autonomic [1, 2]. This phenomenon was demonstrated by fiber reinforcement [3, 4] or using admixture with chemical agents [5], mineral geo-materials [6], and microbial-induced calcium carbonate precipitation (MICP) [7, 8]. Recently, to create a sustainable and cost-effective alternative, microbially calcium carbonate precipitation (MICP) by microorganisms has been studied for application in concrete crack repairing as a self-healing approach. Some bacterial strains can convert carbonate ions (CO_3^{2-}) through urea hydrolysis to bind with calcium ions (Ca^{2+}) to form calcium carbonate ($CaCO_3$). As reported in a study using Diatomaceous Earth to immobilize *Bacillus subtilis* HU58 [9], there was a slight reduction of 2.7 . 10^8 CFU/g from the initial concentration after five months in concrete. This result suggested for a long-life using of *Baciilus* strain in self-healing concrete. *Bacillus subtilis* natto - a native bacterial strain in Japan with spore-forming ability and high resistance to harsh living-conditions can also be a promising choosing. Also, the incorporation of immobilized bacteria in the lightweight aggregate (LWA) in concrete can enhance crack healing by generating $CaCO_3$ due to their metabolic activity and subsequent chemical reactions with other hydrated cement minerals and metabolic by-products. In this study, self-healing repeatability via compressive strength restoration experiments using *Bacillus subtilis* natto immobilized in expanded clay lightweight aggregate (LWA). With high-range cracking age, late hydration as a natural healing phenomenon and self-healing by bacterial bio-mineralization can be clarified. Experimental results add to the understanding of the bio-mineralization of *Bacillus subtilis* natto in self-healing concrete.

2. Materials and Experiments

For the compressive strength restoration test, two groups of concrete specimens (Φ = 50 mm, H = 100 mm), including LWA with and without bacteria, were prepared. Each cycle includes the compression test and 7-day curing in water. First visible cracks appear and developed at around 50 % of compressive strength. For the favorable observation, the load to introduce cracks was defined by 90 % of the compressive strength of the "trial-specimen". All specimens at the same cracking-age were then compressed under the same load. Repeat these processes with the corresponding value of load for the other case of cracking-day (14, 28, and 60 days). Standard procedures are being developed to determine self-healing efficiency. Hence, self-healing capacity was estimated by calculating the strength restoration rate and the crack area

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reduction ratio. Cracking-healing over multi-cycle was performed with the ages of crack, ranging from 7 days to 60 days, to evaluate the self-healing repeatability. Table 1 also shows the mixture followed the study [10] with the w/c = 0.4. The bacterial spores, instead of vegetative cells to increase the survival rate [11, 12] were cultured in a nutrient-low medium with a lack of organic nutrients intentional before penetrating LWA by immersion. The LWA has the component proportion: $m_{bacteria}/m_{lactose}$ = 1.5/1, $m_{bacteria}$ = 0.5 % m_{cement}, m_{urea} = 0.45 % m_{cement}, and m_{CaCl2} = 0.45 % m_{cement}). Visualization of crack closure using optical microscopy was taken with microstructure analysis by Electron Scanning Microscope (SEM) and X-ray Diffraction (XRD) for the phase composition.

Table 1.

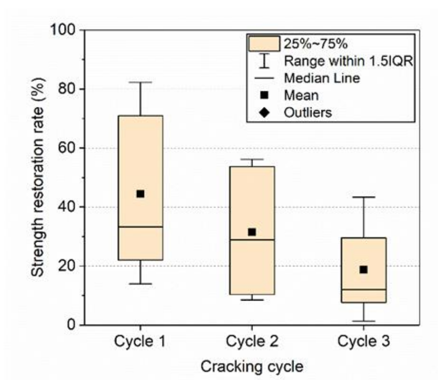
Properties of LWA and mix proportion.

Amount kg/m^3 concrete				
Group	W	C	S	LWA
Bacterial	147	370	953	618*
Reference	147	370	953	618

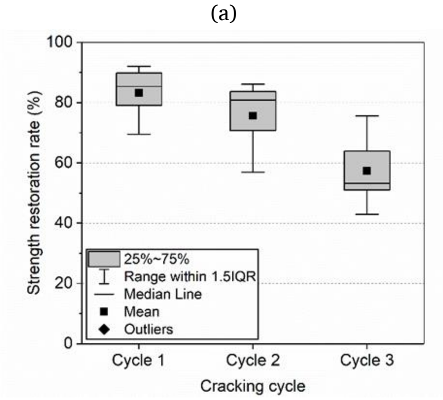
*: LWA with bacteria

3. Results and discussions

3.1. Strength restoration



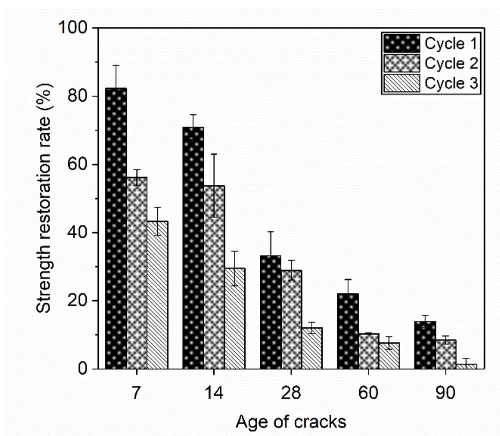
(a)



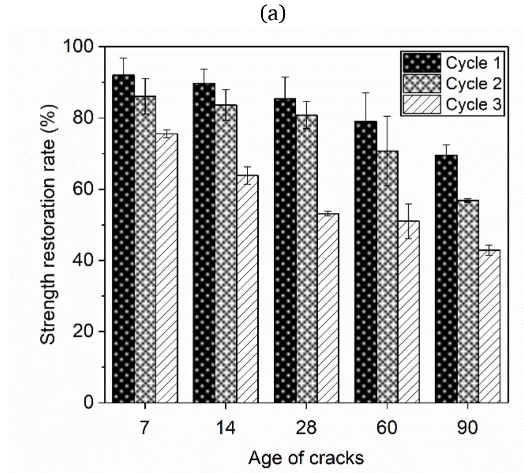
(b)

Figure 1. Compressive strength restoration rate of concrete specimens without bacteria (a) and with bacteria immobilized in LWA (b).

Figure 1 shows the box plots for compressive strength restoration rate of concrete specimens with LWA (Dmax = 20 mm, water absorption = 20 %) immobilized bacteria and the reference specimens range of 7 – 90 days for cracking-age. Data in Figure 1a tend to lean much below the mean, reflecting that regular concrete without bacteria almost cannot restore the compressive strength over multiloading. In contrast, all values greater than 50 % (Figure 1b) and the data tend to lean much above the mean (except cycle 3). The gap between the maximum and minimum values is less than the reference, which indicated the steady bacterial bio-mineralization ability in repairing the damaged structure, compared to the severe damage of the regular concrete after multi cracking-cycle. Figure 2 shows that the late age of cracks could lead to a low strength restoration rate. Late cracks could not be healed by a combination of $CaCO_3$ and other cement minerals as the early-crack with substantial continuous hydration help. A significant difference can be obtained when comparing the reference and specimens with bacteria in LWA. At each cracking-age, especially after a 28-day, the bacterial concrete specimens got a significantly higher restoration rate after the 3-cycle of cracking than the reference specimens.



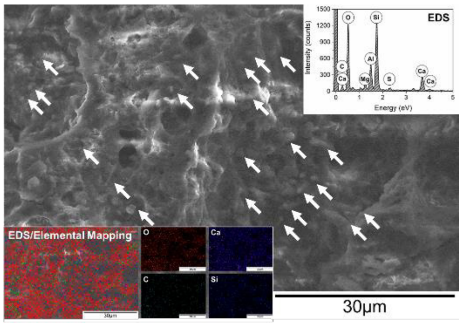
(a)



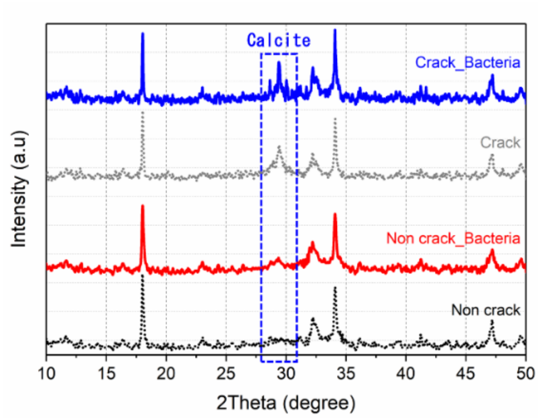
(b)

Figure 2. Relationship between the age of cracks and the strength restoration rate of specimens without bacteria (a) and with bacteria (b).

3.2. Microstructure analysis



(a)



(b)

Figure 3. SEM micrographs and EDS analyses of precipitates at the broken surface of LWA in healed-crack surface (white arrows indicate bacterial imprints covered by healing materials) (a). XRD patterns of precipitated materials inside the crack surface after 7 days (b).

It can be seen from SEM images that the forming of healing products did not occur uniformly in the entire volume of the LWA. The SEM in Figure 3a with EDS and EDS-Mapping shows a continuous and homogenous layer of materials covering the bacterial cells on the broken LWA. This layer filled the porous zones of the LWA and overflowing across the interface of aggregate [13] and the hydrated cement matrix. In this case, the precipitated healing-material could enhance the cement matrix strength as a complex fiber-reinforced compound and fill crack volumes. Also, the mutuality of $CaCO_3$ crystals and hydrated cement minerals as C-S-H and ettringite could make the material robust into cracks with strong bonding with the crack border. This result provides information on the high efficiency of self-healing ability in concrete whose crack age is less than 28 days, in which the late hydration of cement particles can still support healing products. These effects could be explained by the phase composition of the healing products on the XRD analysis of the healed-crack surface.

Figure 3b shows XRD analysis of materials collected from concrete specimens with different situations (un-crack, cracked) to clarify $CaCO_3$ precipitation conditions. The absence of $CaCO_3$ in non-crack reference specimens was already predicted. However, a weak signal of $CaCO_3$ could be obtained in cracked reference specimens, which could be mainly caused by the natural carbonation process when water ingress into the crack. Then, a small peak of $CaCO_3$ can also be seen in the non-crack specimen with bacteria. Although this specimen was not cracked, a small amount of bacterial healing agents could release in the early-state of concrete to heal the micro-cracks, which could be caused by many reasons. It can be seen clearly the high peak of calcite as the result of the bio-mineralization self-healing process.

4. Conclusion

The experimental results of compressive strength restoration and the biological process of *Bacillus subtilis* natto can confirm the possibility of self-healing in concrete. Using bacteria with the nutrient-low medium immobilized in LWA could help repair the crack with considerable damage at high performance at least three times. Besides, bacterial survival in concrete is still inconsistent. However, *Bacillus subtilis* natto with spore-producing ability, particularly under environmental stress or lack of suitable nutrients, could form $CaCO_3$ even at the 90-day age of cracks, suggesting bacterial protection LWA for self-healing repeatability in long-time using. The technique of immobilizing bacterial spores in LWA can also be a reasonable-cost manufacturing process without environmentally unfriendly chemicals for repairing materials production.

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