DEVELOPMENTS IN BACTERIA-BASED SELF-HEALING OF CEMENTITIOUS COMPOSITES

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Introduction

Concrete remains the principal construction material used in buildings and infrastructure worldwide. However, it is susceptible to degradation which, given current design guidance, is nearly always inevitable over its lifetime. This means that intervention and mitigation, in the form of maintenance and repair, is necessary to retain and recover high levels of performance and durability. However, in recent years a range of 'smart' concretes has been developed, including those with autogenous and autonomic self-healing and self-repairing capabilities. Consequently, it is now possible to embed self-immunity and resilience in our concrete buildings and structures such that they evolve over their lifespan. This has significant implications for enhancing durability and serviceability, improving safety and reducing maintenance costs.

Resilient Materials for Life

Research to develop a biomimetic approach to the repair of concrete is being undertaken in the UK as part of the £5M multi-disciplinary research project, Resilient Materials for Life (RM4L), a collaboration between Cardiff University, and the Universities of Bath, Bradford and Cambridge [1]. RM4L has a broad remit to focus on tailoring self-healing cementitious systems for use in specific applications and to address different damage scenarios and conditions. Furthermore, important damage scenarios have been identified, including time-related and cyclic damage as well as chemical damage, including corrosion. In addition to self-healing attributes, RM4L is initiating novel research aimed at embedding self-sensing, self-diagnosing, self-immunisation and self-reporting capabilities into cementitious systems in order to develop truly biomimetic responses in our infrastructure materials and structures.

Within RM4L, research on bacteria-based self-healing of cementitious composites is being undertaken at the University of Bath. In particular, research has been undertaken to overcome a number of application issues with use of bacteria-based self-healing in practice. This paper reports on work that has: (i) determined that different microbial metabolisms result in distinctive mechanisms of precipitation, impacting on performance in application, and (ii) investigated the potential for healing to occur in aged concretes. The extent to which wet/dry conditions are necessary for healing and the possibility for genetically engineering bacteria to obtain more effective healing is also commented on.

Mechanism of Calcium Carbonate Precipitation

Bacteria-based self-healing is achieved by embedding bacterial spores within the concrete and providing them with the nutrients that they need to grow. Whilst the bacteria are in their spore form, they are inactive. However, when conditions become favourable, they germinate into active cells and these multiply. These more favourable conditions occur when a crack forms in the concrete and water and oxygen ingress.

The presence of bacteria near a crack will aid the formation of calcium carbonate should the local environment be rich in dissolved inorganic carbon (DIC) and Ca^{2+} ions. For this reason, growth media containing both carbon and calcium sources are normally used simultaneously with the spores as healing agents. In general, bacteria-based self-healing of concrete can occur because of either ureolytic or non-ureolytic metabolism.

Research at Bath has performed a comparison between how ureolytic and non-ureolytic environmental bacteria precipitate calcium carbonate in order to understand how these mechanisms differ and how we can apply this knowledge to the improvement of self-healing concrete [2]. In this study, environmental bacteria were surveyed for their ability to precipitate calcium carbonate and both the mechanism and resulting precipitates were examined. It was shown that while ureolytic bacteria cause the most rapid precipitation, non-ureolytic bacteria could produce similar amounts of calcium carbonate. In addition, the crystals resulting from non-ureolytic carbonate precipitation appeared to have more organic components and subsequently occupy larger volumes (Figure 1, left), this is likely beneficial for crack sealing application as for the same amount of calcium a larger precipitate can be formed. Indeed, non-ureolytic bacteria caused a more consistent crack healing and subsequent recovery of water tightness (Figure 1, right).

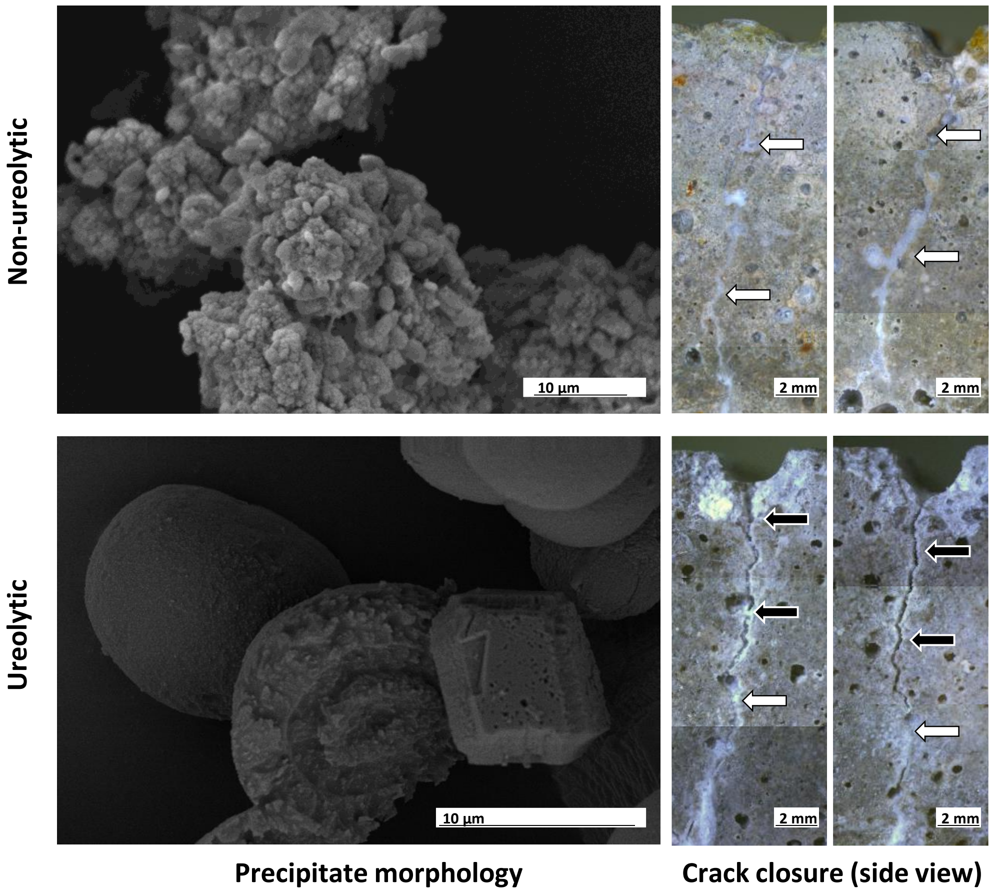

Figure 1: Crystal morphology and crack sealing in mortars containing either non-ureolytic or ureolytic bacteria. Left, electron micrographs of representative precipitate produced by non-ureolytic (top) and ureolytic (bottom) bacteria. Right, crack healing down the side of cement mortars at 8 weeks healing. Non-ureolytic bacteria (top) seal side cracks in a more consistent manner than ureolytic bacteria (bottom). White arrows, complete crack closure. Black arrows, incomplete crack closure.

Engineered Bacteria

It has been considered unlikely that a singular bacterium will be able to meet all requirements for self-healing in concrete and that a mixture of bacteria will be needed in practice. However, speculatively, it has been investigated whether bacteria can be engineered through the introduction of novel "calcite precipitation modules" into weak calcite precipitators to obtain better bacteria for self-healing of concrete. Initial research has shown that this is the case and that *Bacillus subtilis* W168, a bacterium that does not precipitate calcite, can be made to do so by introduction of the correct modules. These results are proving positive in terms of understanding how and why calcite precipitation occurs.

Healing in Aged Concrete

The ability of bacteria to autonomously heal cracks in cementitious composites by precipitating calcium carbonate has been verified in studies using several technologies. Since both Ca^{2+} and CO_3^{-2} ions are required, technologies have been developed that provide these as separate additions (e.g. urea plus calcium nitrate) [3], or by the use of organic calcium salts (e.g. calcium acetate) which supplies both as a single addition [4]. Although the precise source of Ca^{2+} ions used by the bacteria during self-healing is unclear, it has been proposed that calcium hydroxide is likely to be an important source. Therefore, it is interesting to note that in nearly all previous studies self-healing of the cracks has been tested on water-cured mortars or concretes at a relatively young age (~28 days) where calcium hydroxide is present as a hydration product and provides a plentiful supply of soluble Ca^{2+} ions. However, in practice concrete is often subject to alternate wetting and drying, and a process of carbonation can occur before the concrete cracks. The outcome is that Ca^{2+} ions may become trapped in an unusable form. For this reason, research was carried out at Bath to compare mortars that were cast with calcium sources added directly to the mortar and with the calcium source encapsulated in lightweight aggregates.

It was verified that calcium hydroxide is an important source of Ca^{2+} ions for bacteria-based self-healing of mortars. For mortars that do not carbonate prior to cracking this calcium hydroxide is sufficient to provide an efficient level of Ca^{2+} ions for healing. However, carbonation of this calcium hydroxide eliminates this source of Ca^{2+} ions. Consequently, bacteria-based self-healing in mortars that have carbonated prior to cracking is almost totally dependent on the availability of Ca^{2+} ions released from an encapsulated source. Therefore, whilst the direct addition and encapsulation of calcium nitrate are both suitable for providing self-healing of concrete, the conditions of the concrete during service life need to be considered when choosing the most appropriate option.

Wet/Dry Conditions

Water has an important role to play in bacteria-based self-healing. It is essential for germination of bacterial spores and furthermore the mineral precursors need to be dissolved in water in order to be available to the bacteria [5] . Consequently, bacterial precipitation of calcium carbonate cannot occur if no water is present in the crack zone. A number of researchers have looked to determine to what extent self-healing can occur in different environments, and work has been carried out at Bath to compare self-healing in different conditions (Figure 2).

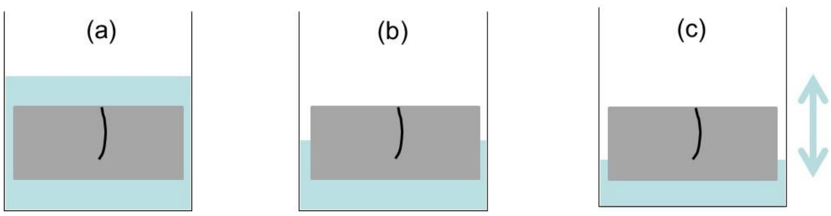

Figure 2: Healing conditions for bacteria-based self-healing concrete: (a) permanently wet, (b) semi-submerged and (c) cyclic wet-drying; 16 hours wet, 8 hours dry

The results show that self-healing is possible in all conditions. However, healing in semi-submerged conditions appears to occur more rapidly and may be related to the continual supply of oxygen. Healing in cyclic wet-dry conditions was better than that in permanently wet conditions.

Conclusions

Bacteria-based self-healing of concrete is a reliable and useful technology for maintaining the durability and resilience of our infrastructure. However, several fundamental and application-related challenges remain to be solved before it will become a readily available and widely used construction material. This paper has reported on work that has further developed bacteria-based self-healing concrete in relation to these challenges.

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