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# Impregnation and encapsulation of lightweight aggregates for self-healing concrete

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## HIGHLIGHTS

- Sodium silicate solution was impregnated in lightweight aggregates (LWA).
- Impregnated LWA were coated then embedded in concrete specimens.
- Strength regain was remarkable for specimens with the impregnated LWA.
- Capillary water absorption was significantly improved in the specimens with the impregnated LWA.
- Sodium silicate produced rich silica C–S–H to heal the concrete cracks.

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## ABSTRACT

This study investigated a technique of impregnating potential self-healing agents into lightweight aggregates (LWA) and the self-healing performance of concrete mixed with the impregnated LWA. Lightweight aggregates with a diameter range of 4–8 mm were impregnated with a sodium silicate solution as a potential self-healing agent. Concrete specimens containing the impregnated LWA and control specimens were pre-cracked up to 300 μm crack width at 7 days. Flexural strength recovery and reduction in water sorptivity were examined. After 28 days healing in water, the specimens containing the impregnated LWA showed ~80% recovery of the pre-cracking strength, which accounts more than five times of the control specimens' recovery. The capillary water absorption was also significantly improved; the specimens healed with the impregnated LWA showed a 50% reduction in the sorptivity index compared with the control cracked specimens and a very similar response to the control uncracked specimens. The contribution of sodium silicate in producing more calcium silicate hydrate gel was confirmed by characterisation the healing products using X-ray diffraction, Fourier transform spectroscopy, and scanning electron microscopy.



## 1. Introduction

Surface opening cracks are a common type of defects in concrete structures. They allow penetration of water or other deleterious agents that result in loss of durability earlier than expected. Thus, repairing formed cracks and defects becomes essential and unavoidable. Currently, maintenance and repair of concrete structures generally rely on regular inspection programmes, which are expensive, and they also depend on a combination of non-destructive testing (NDT) and human perception [1]. In case of severe damage, the structural component is replaced entirely while repairs are attempted for less extensive damage. Vast amounts of money are spent each year on inspection and repair as direct and indirect costs, the latter often being much higher than the former. For instance, in the USA, the annual economic impact associated with maintaining, repairing, or replacing deteriorating structures is estimated at \$18–21 billion [2]. The American Society of Civil Engineers estimated that \$2.2 trillion are needed for five years, starting from 2012, for repair and retrofit; a cost of \$2 trillion has been predicted for Asia's infrastructure for the same period [3]. Europe spends more than half of its annual construction budget on repair works [4], while in the UK, repair and maintenance costs account for over 45% of the total expenditure on construction [5].

Moreover, repair works have a significant adverse environmental impact particularly in cases where partial or complete replacement of structures is required. It is known that the production of 1 tonne of Portland cement (PC), as often being the main constituent

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on concrete, releases about 0.85–1.1 tonnes of $CO_2$ [6]. Approximately $3.6 \times 10^9$ tonnes of cement were produced worldwide in 2014 [7]. The $CO_2$ emissions associated with the production of cement are very significant, and are estimated at 7% of the global anthropogenic $CO_2$ emissions [6].

Therefore, developing innovative technologies to overcome these challenges has become an urgent necessity. Over the past few decades, the notion that concrete can be designed with a sufficient healing capability and heal its cracks without any external aid has been inspiring field of work for many research groups around the world. Self-healing as defined by RILEM is "any process by the material itself involving the recovery and hence improvement of a performance after an earlier action that had reduced the performance of the material" [8].

Broadly, self-healing processes within cement based materials can be divided into two categories: autogenic and autonomic. Autogenic self-healing is the phenomenon where the material heals cracks using its own generic components and constituents. Autonomic self-healing however, involves the use of engineered additions that are not conventionally added into cementitious materials. These additions are added specifically to enhance self-healing capability[8,9].

The main mechanisms of the autogenic self-healing are the ongoing hydration of cement grains that have not reacted due to lack of water or the precipitation of the calcium carbonate, which is the result of a reaction between the calcium ions in concrete and carbon dioxide dissolved in water [8,10]. Ongoing hydration is the main healing mechanism in young concrete due to its relatively high content of un-hydrated cement particles, while formation of calcium carbonate is the most likely cause of self-healing at later ages [11]. For attaining effective autogenous self-healing, water is essential and the crack widths are restricted to be less than 100 μm and preferably less than 50 μm [12,13]. Some studies have been carried out to promote autogenous healing by crack width restriction or with continuous supply of water. For instance, fibre reinforced cementitious composites (FRCC) have significantly higher potential of self-healing than ordinary concrete because of their high ductility, the micro-cracking behaviour and tight crack width control [11,14]. Several fibres have been used in FRCC composites such as polyethylene (PE) [15], polyvinyl alcohol (PVA) [16–18], and polypropylene (PP) fibres [18]. Meanwhile some researchers have investigated the possibility to mix super absorbent polymers (SAP) into cementitious materials to provide additional water [19,20]. Others have examined the effect of replacing part of the cement by other pozzolanic and latent hydraulic materials like fly ash, silica fume, or blast furnace slag [21–24]. These materials continue to hydrate for prolonged time enhancing the autogenous healing potential.

In contrast, many systems and techniques have been investigated to heal concrete cracks autonomically such as modifying concrete by embedding microcapsules or hollow fibres with a suitable healing agent. Once the crack occurs the shell of the capsule or the wall of the tube ruptures and the healing agent is released and reacts in the region of damage to produce new compounds which seal the crack and/or bond the crack faces [3]. Zhao et al. [25] have reported that the most utilised shell polymers in development the microcapsules are poly(urea-formaldehyde) (PUF), polyurethane (PU) and poly(melamine-formaldehyde).The healing agents that have been often used to date in the literature include epoxy resins [26,27], methyl methacrylate (MMA) [28], alkali-silica solutions ($Na_2SiO_3$) [29], and cyanoacrylates (CA) [30–32]. Additionally, bacterially induced carbonate precipitation has been proposed as an alternative and environmental friendly self-healing technique [33–35]. Other researchers proposed the use of expansive agents and swelling geo-materials to stimulate the chemical reactions to produce hydration products for filling cracks in concrete [14]. For instance, Kishi and co-workers (2007) have demonstrated the use of a mix of expansive agents ($C_4A_3S$, $CaSO_4$, and CaO), swelling geo-materials such as silicon dioxide and sodium aluminium silicate hydroxide, montmorillonite clay and various types of carbonates as partial cement replacement [36]. Ferrara et al. [37] and Roig-Flores et al. [38] have investigated the self-healing behaviour of ordinary concrete mixtures included crystalline admixture additives, which consist of a mix of cement, sand and active silica. Calcium sulfoaluminate (CSA) has also been utilised as an expansive agent for self-sealing [36,39,40] and recently magnesium oxide has been suggested as a self-healing agent by Alghamri and Al-Tabbaa [41].

Sodium silicate ($Na_2SiO_3$) has been proposed as a potential self-healing agent in different systems. A number of researchers have assessed different aspects of the self-healing capability of sodium silicate. Pelletier et al. [42] enveloped crystalline sodium silicate in polyurethane microcapsules with 40–800 μm size. Thereafter, the synthesised capsules were added to concrete mix of 2% by volume. The concrete samples containing the microcapsules showed 24% flexural strength recovery compared with 12% for the control samples. Huang and Ye [29] embedded 5 mm diameter capsules filled with sodium silicate solution into specimens of engineering cementitious composites (ECC). The results demonstrated that the main mechanisms of self-healing are the reaction between the calcium cations and the dissolved sodium silicate and the crystallisation of the sodium silicate. However, the results showed also a negative effect of the capsules on the mechanical properties of concrete specimens. In another study, Gilford et al. [43] developed sodium silicate and dicyclopentadiene (DCPD) as self-healing agents encapsulated in urea-formaldehyde shell. The two types of microcapsules were examined in concrete cylinder specimens. The results indicated that the addition of 5% sodium silicate microcapsules by weight of cement increased the modulus of elasticity of the concrete specimens by 11% after healing. For the DCPD microcapsules, the healing agent was effective in increasing the modulus of elasticity of concrete after cracking by as much as 30% for the microcapsules at a content of 0.25%. Mostavi et al., [44] also used double-walled polyurethane/urea-formaldehyde (PU/UF) microcapsules to encapsulate sodium silicate. These microcapsules were incorporated into concrete beams with two different proportions (2.5% and 5% by weight of cement) and the healing process was monitored by measuring the crack depth within the healing time using ultrasonic digital indicating tester. It was found that the healing rate with 5% microcapsules was higher in comparison with samples containing 2.5% of microcapsules. In a recent study conducted by Kanellopoulos et al. [45], liquid sodium silicate was stored in a thin walled soda glass capsules. The results indicated that the sodium silicate has a promising capability as a self-healing agents in both regaining strength and improving durability.

Given that the aggregates are the major constituent of any concrete mix, they had been expected to be widely used to host self-healing agents: however, this potential has not been extensively researched. In a study performed by Wiktor and Jonkers [34], porous clay particles with (1–4) mm size were impregnated twice under vacuum by a two-component bio-chemical self-healing agent consisting of bacterial spores and calcium lactate. Upon crack formation the two components were released from the particles by crack ingress water and produced calcium carbonate which led to plug cracks of up to 0.46 mm width. In another study, Sisomphon et al. [46] used expanded clay lightweight aggregates as reservoirs for sodium monofluorophosphate ($Na_2FPO_3$) solution and eventually encapsulated them in a cement paste layer. The developed encapsulated particles were used as a self-healing system in blast furnace slag cement mortars. The characterisation of the healing products indicated that the healing mechanism would