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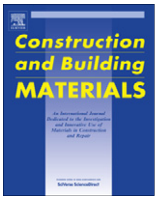

Biotech cementitious materials: Some aspects of an innovative approach for concrete with enhanced durability

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HIGHLIGHTS

- ▶ Bioinspired materials are eco-efficient.
- ▶ Biotech concrete may have enhanced durability.
- ▶ Biomineralization is a low toxic crack repair technique.

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ABSTRACT

The deterioration of reinforced concrete structures is a very common problem due to the fact that this material has a high permeability which allows water and other aggressive media to enter, thus leading to corrosion problems. The use of sealers is a common way of contributing to concrete durability. However, the most common ones are based on organic polymers which have some degree of toxicity. The Regulation (EU) 305/2011 related to the Construction Products Regulation emphasizes the need to reduce hazardous substances. Therefore, new low toxicity forms to increase concrete durability are needed. Recent investigations in the field of biotechnology show the potential of bioinspired materials in the development of low toxic solutions. This paper reviews current knowledge on the use of bacteria for concrete with enhanced durability. It covers the use of bacteria in concrete mix and also biomineralization in concrete surface treatments. Investigation gaps are described. Results from practical applications in which there is exposure to environmental conditions are still needed in order to confirm the importance of this new approach.

1. Introduction

With an annual production of about 10 km^3/year [1], Portland cement concrete is the most used construction material on Earth, the majority of which being used in the execution of reinforced concrete structures. However, many concrete structures face premature degradation problems, in fact, in the USA alone about 27% of all highway bridges are in need of repair or replacement. Also, the corrosion deterioration costs due to deicing and sea salt effects are estimated at over 150 billion dollars [2]. The reasons for such a low durability performance have to do with the fact that some of them were built decades ago, when little attention was given to durability issues [3], but also due to the fact that concrete has a high permeability. This allows water and other aggressive elements to enter, leading to carbonation and chloride ion attack resulting in corrosion problems. The importance of concrete durability in the context of the eco-efficiency of construction materials has been rightly put by Mora [4], when he stated that increasing its durability from 50 to 500 years would mean a reduction of its environmental impact by ten times. The importance of concrete durability for their eco-efficiency has also recently been recognized by other authors [5,6]. Concrete structures with low durability require frequent maintenance and conservation operations or even its integral replacement, being associated with the consumption of raw materials and energy. Concrete durability means, above all, minimizing the possibility of aggressive elements to enter the concrete, under certain environmental conditions, for any of the following transport mechanisms: permeability, diffusion or capillarity. The use of concrete surface treatments with waterproofing materials (also known as sealers) to prevent the access of aggressive substances is a common way of contributing to concrete durability. However, the most common surface treatments use organic polymers (epoxy, siloxane, acrylics and polyurethanes) all of which have some degree of toxicity. Polyurethane is obtained

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from the isocyanates, known worldwide for their tragic association with the Bhopal disaster. The production of polyurethane also involves the production of toxic substances such as phenol and chlorofluorocarbons. Besides, chlorine is associated to the production of dioxins and furans that are extremely toxic, and also, bio-cumulative. Several scientist groups already have suggested that chlorine industrial based products should be prohibited [7]. Recently, the European Union recently have approved the Regulation (EU) 305/2011 related to the Construction Products Regulation that will replace the current Directive 89/106/CEE, already amended by Directive 1993/68/EEC, known as the Construction Products Directive. A crucial aspect of the new regulation relates to the information regarding hazardous substances [8]. New low toxicity materials and techniques that increase concrete durability are therefore needed. An innovative approach to solve this and other current technological problems faced by the human society which can encompass a new way of perceiving the potential of natural systems [9]. The continuous improvement of these systems carried out over millions of years, has been leading to materials and ''technologies'' with exceptional performance and that are fully biodegradable. Recent nanotechnology achievements regarding the replication of natural systems may provide a solution to solve the aforementioned problem [10,11]. Analysis of bioinspired materials requires knowledge of both biological and engineering principles thus constituting a new research area that can be termed as biotechnology. Although this area has rapidly emerged at the forefront of materials research, the fact is that the study of biological systems as structures dates back to the early parts of the twentieth century with the work of D'Arcy W. Thompson, first published in 1917 [12]. In this context biotechnology seems to be able to provide a solution to concrete durability enhancement by means of biomineralization, a phenomenon by which organisms form minerals and were used for crack repair by Gollapuddi et al. [13]. In this paper the most relevant investigations on the field of bacteria modified concrete are reviewed.

2. Bacteria mineralization mechanisms

Bacteria are relatively simple, unicellular organisms. There are typically 40 million bacterial cells in a gram of soil and a million bacterial cells in a milliliter of fresh water; in all, there are approximately five nonillion (5×10^{30}) bacteria on Earth, forming much of the world's biomass. Under optimal conditions, bacteria can grow and divide extremely rapidly and bacterial populations can double as quickly as every 9.8 min [14]. Biomineralization is defined as a biologically induced precipitation in which an organism creates a local micro-environment, with conditions that allow optimal extracellular chemical precipitation of mineral phases, like calcium carbonate ($CaCO_3$) [15]. Decomposition of urea by ureolytic bacteria is one of the most common pathways to precipitate $CaCO_3$. The microbial urease enzyme hydrolyzes urea to produce dissolved ammonium, dissolved inorganic carbon and CO_2. Furthermore, the ammonia released in the surroundings subsequently increases pH, leading to accumulation of insoluble $CaCO_3$ in a calcium rich environment. Fig. 1. shows a simplified representation of the events occurring during the ureolytic induced carbonate precipitation. Ramachandran et al. [17], reported that a high pH hinders the growth of bacteria, these authors also state that the optimum pH for the growth of *Bacillus pasteurii* is around 9. According to Arunachalam et al. [18] *Bacillus sphaericus* bacteria induced calcium carbonate precipitation at pH = 8 yields the highest results. Urease-catalyzed ureolysis is also temperature dependent and the optimum temperature ranges from 20 °C to 37 °C. In fact, increasing the temperature from 15 °C to 20 °C increased the rate of ureolysis, k_{urea} 5 times and 10 times greater than k_{urea} at 10 °C [19]. The same authors reported that k_{urea} is more dependent on the bacterial cell concentration than initial urea concentration so long as there is enough urea to sustain the bacteria. At 25 mM Ca^{2+} concentration, increasing bacterial cell concentration from 10^6 to 10^8 cells mL^{-1} increased the $CaCO_3$ precipitated by over 30%. However, when Ca^{2+}concentration was increased 10-fold, the $CaCO_3$ precipitated increased by over 100% irrespective of initial urea concentration.

3. Bacterium type

Different bacteria lead to very different calcium carbonate precipitation results. Table 1 summarizes the several bacteria used by different authors. Ramachandran et al. [17] found that the contribution of *Pseudomonas aeruginosa* related to calcium carbonate precipitation was insignificant. Ghosh et al. [32] mentioned that the use of *Escherichia coli* showed no evidence of biomineralization. Achal et al. [29] developed a phenotypic mutant of *Sporosarcina pasteurii*. The mutant named Bp M-3 was found to be more efficient in improved urease activity and also showed to be able to survive at very high pH values. In the majority of the studies, ureolytic bacteria of the genus *Bacillus* were used as agent for the biological production of calcium carbonate based minerals. The mechanism of

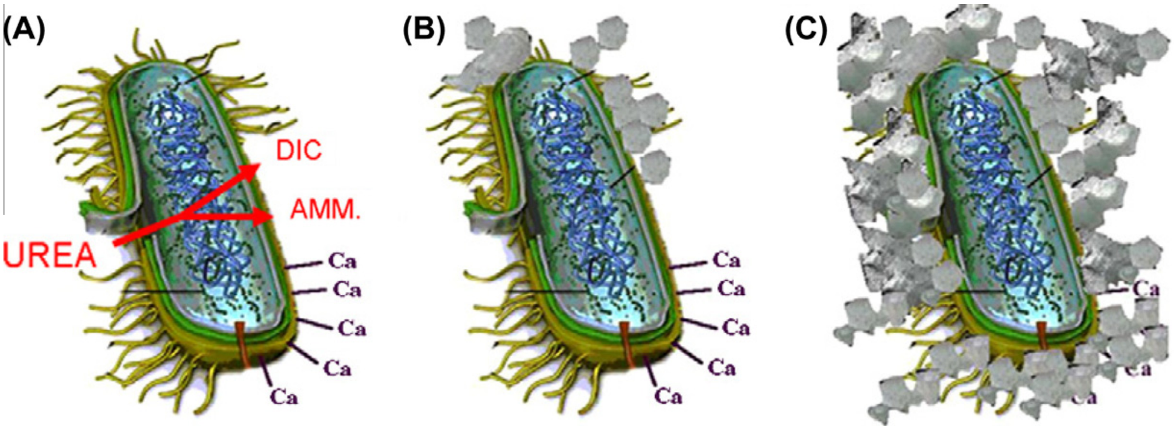

Fig. 1. Simplified representation of the events occurring during the ureolytic induced carbonate precipitation. Calcium ions in the solution are attracted to the bacterial cell wall due to the negative charge of the latter. Upon addition of urea to the bacteria, dissolved inorganic carbon (DIC) and ammonium (AMM) are released in the microenvironment of the bacteria (A). In the presence of calcium ions, this can result in a local supersaturation and hence heterogeneous precipitation of calcium carbonate on the bacterial cell wall (B). After a while, the whole cell becomes encapsulated (C) [16].

Table 1
Bacterium type.

References	Bacterium type
[20,21]	*B. sphaericus* LMG 225 57
[22]	
[18]	
[23]	
[24]	
[17]	*S. pasteurii* ATCC 11859
[19]	
[25]	
[14]	*S. pasteurii* (formerly *Bacillus pasteurii)*
[26]	
[27]	*Bacillus subtilis*
[28]	
[29]	Phenotypic mutant of *S. pasteurii* (Bp M-3)
[30]	
[17]	*Pseudomonas aeruginosa* ATCC 27853
[31]	*Bacillus pseudofirmus* DSM 8715
[32,33]	*Shewanella* sp.
[34]	*B. megaterium* ATCC 14581
[35]	*Bacillus* sp. CT-5,
[36]	*Bacillus alkalinitrilicus*

calcium carbonate formation by these bacteria is based on the enzymatic hydrolysis of urea to ammonia and carbon dioxide. A potential drawback of this reaction mechanism is that for each carbonate ion two ammonium ions are simultaneously produced. This may increases the risk of reinforcement corrosion [31]. Besides atmospheric ammonia is recognized as a pollutant that contributes to several environmental problems [16].

4. Using bacteria in concrete mix

Ramachandran et al. [17] mentioned that using *B. pasteurii* has a positive influence on the performance of cementitious composites. Ghosh et al. [33] reported that anaerobic hot spring bacterium leaches silica and helps in the formation of new silicate phases that fill the micro pores. They also mention that a concentration of 10^5 cells/ml optimizes the microstructure of cementitiuous composites. In order to overcome the problem of excessive ammonia production associated with the use of genus *Bacillus*, Jonkers et al. [31] used bacterial spores (*Bacillus cohnii*). They reported a loss of bacteria linked to the continuing decrease in the matrix pore diameter sizes with the progress of concrete curing age. In order to avoid bacteria loss, these authors suggest their encapsulation prior to addition to the concrete mixture, or else, the addition of air-entraining agents. Reddy et al. [27] reported that the use of *bacillus subtilis* bacteria for a cell concentration of 10^5

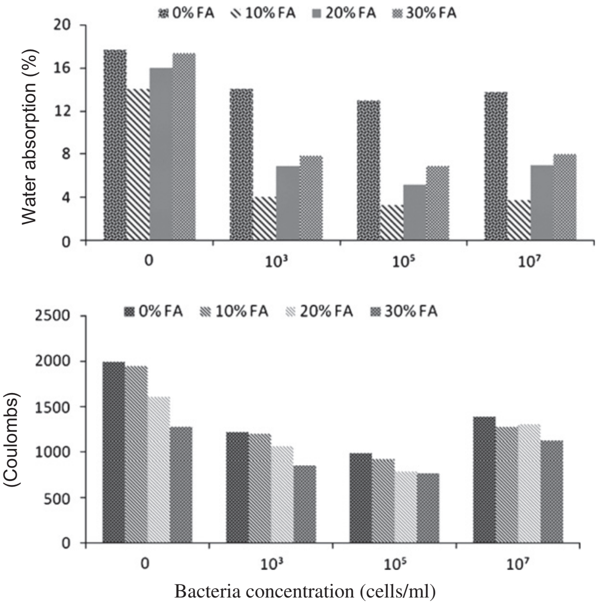

Fig. 3. Effect of bacteria *S. pasteurii* on (a) water absorption of concrete at 7 days and (b) chloride permeability of fly ash (FA) concrete at 28 days [14].

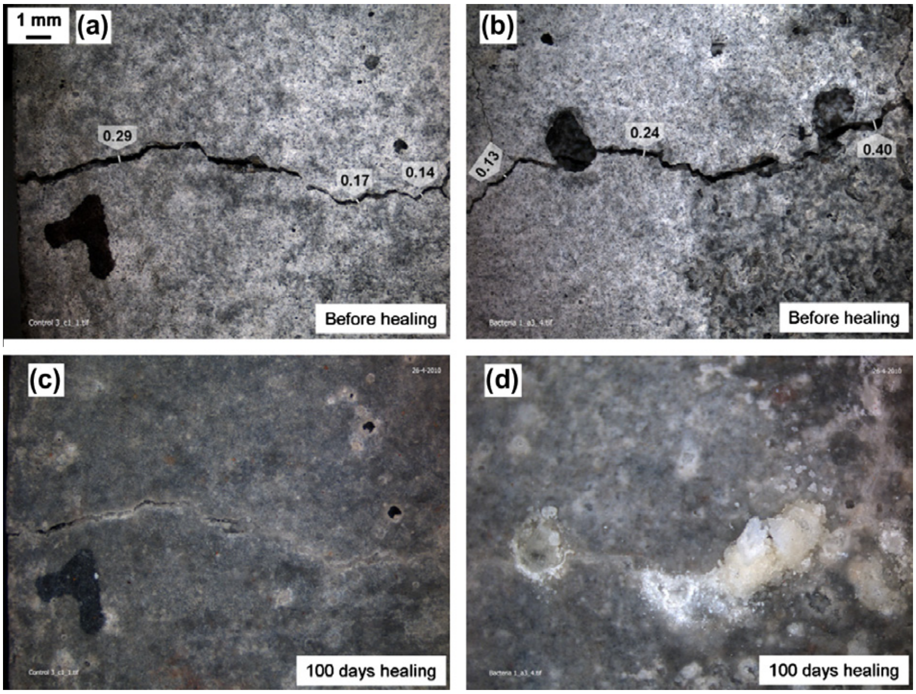

Fig. 2. Stereomicroscopic images of crack-healing process in control mortar specimen before (a) and after 100 days healing (c), in bio-chemical agent-based specimen before (b) and after 100 days healing (d) [36].

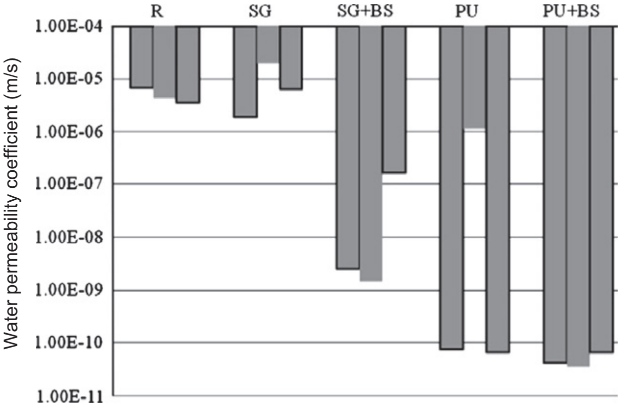

Fig. 4. Water permeability [24]: (R – reference; SG – silica gel; BS – bacterial suspension; PU – polyurethane immobilized bacteria).

cells per ml of mixing water increases the concrete resistance to sulphuric acid attack. For the same bacteria an optimum concentration of 10^6 cells per ml was reported by other authors [28]. Van Tittelboom et al. [22] confirm that the use of bacteria can help to reduce the water permeability of concrete, however, they mention that the highly alkaline pH of concrete hinders the growth of the bacteria. To overcome this problem they immobilized the bacteria in silica gel. Other authors have already suggested the use of polymer encapsulation [37]. According to Achal et al. [34] fly ash concrete containing *Bacillus megaterium* cells absorbed nearly 3.5 times less water than the control concrete. They also found that the permeability of the concrete with bacterial cells was lower than that of the control concrete. Wiktor and Jonkers [36] reported that the combined effect of viable bacterial spores plus calcium lactate embedded in porous clay capsules significantly enhanced mineral precipitation on crack surfaces further resulting in the healing of cracks (Fig. 2) with a maximal width of 0.46 mm. They also mentioned that since bacteria consume oxygen, it may provide an additional benefit associated with the potential to inhibit reinforcement corrosion. Chahal et al. [14] studied the influence of *S. pasteuri* bacteria on fly ash concrete. The optimum performance was achieved for a 10^5 cells/ml of

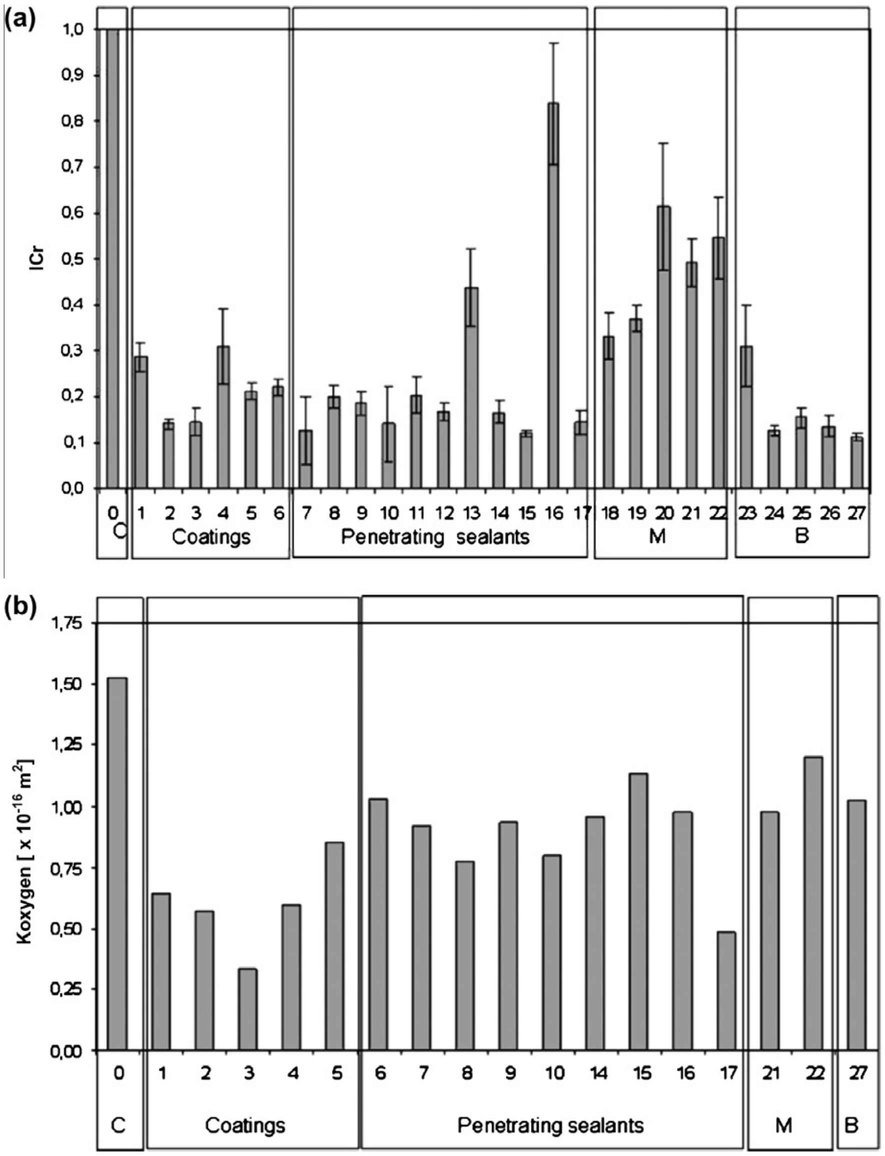

Fig. 5. Concrete durability: (a) Capillary water suction results expressed as the relative capillary index and (b) permeability towards oxygen of treated and untreated concrete specimens expressed as specific permeability coefficient K to oxygen (C = control, M = mixed ureolytic cultures, B = *Bacillus sphaericus*) [20].

bacteria concentration. These authors reported a four-time reduction in water absorption and an eight times reduction in chloride permeability due to calcite deposition (Fig. 3). Wang et al. [23] suggests the use of diatomaceous earth to protect the bacteria *B. sphaericus* from the high-pH of the concrete matrix. These authors report that the bacteria immobilized in diatomaceous earth had much higher ureolytic activity (12–17 g/l urea was decomposed within 3 days) than that of un-immobilized bacteria (less than 1 g/l urea was decomposed within the same time span) in cement slurry. The optimal concentration of diatomaceous earth for immobilization was 60% (w/v, weight of diatomaceous earth/volume of bacterial suspension). Wang et al. [24] compared the performance of two different techniques (silica gel and polyurethane) to protect bacteria when immobilized inside concrete. The silica gel technique uses Levasil®200/30% sol with a specific surface area of 200 m^2/g and a solid content of 30% was used to embed bacterial cells. The immobilization of bacteria into polyurethane uses a two-component polyurethane MEYCO MP 355 1 K (BASF), to encapsulate bacterial cells. The incorporation of the bacteria into mortar specimens is made by glass tubes with a length of 40 mm and an inner diameter of 3 mm. Experimental results show that the silica gel immobilized bacteria exhibited a higher activity than polyurethane immobilized bacteria, and hence, more $CaCO_3$ precipitated in silica gel (25% by mass) than in polyurethane (11% by mass), which was demonstrated by thermogravimetric analysis. However, cracked mortar specimens healed by polyurethane immobilized bacteria had a lower water permeability coefficient (10^{-10}–10^{-11} m/s) compared with specimens healed by silica gel immobilized bacteria, which showed a water permeability coefficient of 10^{-7}–10^{-9} m/s (Fig. 4). The use of bacteria in concrete is associated with mineral precipitation that helps to fill micro pores and cracks thus reducing its permeability. However, the highly alkaline pH of concrete hinders the growth of the bacteria. To overcome this problem different authors have suggested the use of different immobilization solutions (clay capsules, silica gel or polyurethane encapsulation).

5. Concrete surface treatment

De Muynck et al. [20] compared the durability (concerning capillary water uptake and gas permeability) of concrete when its surface was treated with pure and mixed cultures of ureolytic bacteria. They concluded that the type of bacterial culture and the medium composition had a profound impact on $CaCO_3$ crystal morphology, being that the use of pure cultures resulted in a more pronounced decrease in the uptake of water. They also concluded that the durability performance obtained with cultures of the species *B. sphaericus* was comparable to the ones obtained with conventional water repellents (silanes, siloxanes) (Fig. 5). De Muynck et al. [21] studied different durability parameters (carbonation, chloride penetration and freezing and thawing) confirming that the biodeposition treatment showed a similar protection towards degradation processes when compared to some of the conventional surface treatments under investigation. They also mention the need for investigations regarding the durability of the treatment under acidic media. They further mentioned that biological generated calcite is less soluble than the one inorganically precipitated, thus suggesting a higher performance. Okwadha and Li [25] used bacterium *S. pasteurii* strain ATCC 11859 to create a biosealant on a PCB-contaminated concrete surface reporting a reduction on water permeability by 1–5 orders of magnitude. They also state that the treated concrete had a high resistance to carbonation. Achal et al. [35] mention a six times reduction in water absorption due to the microbial calcite deposition. In a different study the same authors [30] used a phenotypic mutant of *S. pasteurii* (Bp M-3) with improved urease activity also reporting a significant reduction in water absorption, permeability and chloride permeability. Li and Qu [38] confirms that bacterially mediated carbonate precipitation on concrete surface reduces capillary water uptake, leading to the carbonation rate constant to be decreased by 25–40%. Nevertheless, the cost of biodeposition treatment still remains a major drawback to be overcome being dependent on the price of the microorganisms and the price of the nutrients (5–10 € per m^2), which is far from being cost-efficient [16]. More recently Achal et al. [30] used corn steep liquor, an hazardous industrial effluent, as a nutrient source reporting a biodeposition cost of just (0.3–0.7 €) per m^2.

6. Conclusions

Bioinspired materials can lead to a more sustainable construction industry, especially when providing new low toxic solutions. Much research has already been carried out on the field of bacteria based concrete; however, it is still far from being a proved and reliable technique capable of replacing current common concrete surface treatments based on organic polymers sealers. The use of bacteria in concrete is associated with mineral precipitation that helps to fill micro pores and cracks thus reducing its permeability. However, the highly alkaline pH of concrete hinders the growth of the bacteria. To overcome this problem different authors have suggested the use of different immobilization solutions (clay capsules, silica gel or polyurethane encapsulation). The use of bacteria in concrete mix also needs further research efforts. Several issues still need to be addressed in this field:

(a) Which calcite producing bacteria are more efficient in highly alkaline environment?
(b) Can air-entraining agents be effective in preventing bacteria loss associated due to reduction in pore size?
(c) Which is the most eco-efficient encapsulation method?
(d) Will biologically deposited calcite endure the test of time?
(e) Can biomineralization be made cost-efficient?
(f) What are the environmental implications related to the use of corn steep liquor as a nutrient source?
(g) Are there any health implications involved in the use of bacteria?
(h) What is the life cycle analysis of biotech concrete?

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