Article

Industrial application of biological self-healing concrete: Challenges and economical feasibility

Filipe Bravo Silva
is a PhD student at Gent University working in industrial environment. The main goal is to perfom an economic evaluation of the current biotechnology for self-healing concrete application and, if possible and economical feasible, up-scale the existing technology.

Nico Boon
is a Professor in the f aculty of Bioscience Engineering at Ghent University.

Nele De Belie
is a Professor in the faculty of Engineering and Architecture Ghent University.

Willy Verstraete
is a Professor in the faculty of Bioscience Engineering Ghent University.

ABSTRACT

Self-healing concrete has been scrutinized by several researchers and some industrial concrete producers in relation to the remediation of the occurrence of micro-cracks. Such cracks are a quite well known problem that can lead to corrosion of the steel reinforcement and thus to the possible failure of the entire concrete structure. The need to repair these cracks as soon as possible leads to maintenance costs which can be of the order of €130 (direct costs) per m^3 of concrete. Recent scientific studies indicate that a Microbial Induced Carbonate Precipitation (MICP), using microbial spores as active agent, can be an alternative for the actual repair methods. However, the production of bacterial spores is yet imposing considerable costs. According to some concrete producers they would be willing to pay about €15 to €20 per m^3 of concrete for a bio-based self-healing product. However, the actual cost of spores production and encapsulation represent a total cost which is orders of magnitude higher. This article analyzes the costs for the biological self-healing in concrete and evaluates the industrial challenges it faces. There is an urgent need to develop the production of a bio-additive at much lower costs to make the biological self-healing industrial applicable. Axenic production and a possible non-axenic process to obtain ureolytic spores were analyzed and the costs calculations are presented in this paper.

Journal of Commercial Biotechnology (2015) 21(1), 31–38. doi: 10.5912/jcb662
Keywords: Concrete; Self-healing; Spores; Encapsulation; Economics

INTRODUCTION

Concrete is a composite construction material made primarily with aggregates, cement and water. It is the most widely used construction material in the world with a usage, worldwide, twice as much as steel, wood, plastics and aluminum combined.[1]

Due to its wide usage, concrete represents the basis of a large commercial industry. In the United States alone, concrete industry represents a €23.3 billion of sales per year, considering only the value of the ready-mixed concrete sold each year.[2] Despite of its high compressive strength, the tensile strength is low, making it necessary for most applications to add a material (often steel) to allow the structure to maintain its correct form and performance. Reinforced concrete is obtained by adding steel reinforcement bars, steel fibers or glass or plastic fibers to carry the tensile loads. The most widely used reinforcement are the steel bars, forming a net inside the concrete structure.

Correspondence:
Filipe Bravo Silvia, Gent University, Belgium. Email: filipe.silva@aveco.be

Due to the intrinsic heterogeneity in concrete, cracks are almost unavoidable. Cracking in concrete structures is a well-known and studied phenomenon. Cracks can have many causes such as drying shrinkage, thermal stress, weathering, externally applied loads or corrosion of reinforcement.[3] When a crack opens, aggressive compounds such as chloride ions (Cl^-) or carbon dioxide (CO_2) can penetrate the concrete cover, getting to the reinforcement and causing corrosion. The rusting process leads, with time, to the loss of tensile strength, which can cause irreparable damages in the structure. Due to this, it is quite important to fill these cracks avoiding the increase of permeability thus protecting the reinforcement. Moreover, since, on the one hand, cracks in concrete structures can lead to the premature failure of the structure and on the other hand, sustainability is one of the main issues in the modern world,[4] the repair of such cracks is also becoming important from an environmental point of view.

Until now, applying some compounds either to fill the cracks, such as epoxy resins, or to prevent the formation of these cracks, such as plastic polymers applied on the surface of the concrete (repairing and curing compounds, respectively), are the common ways to improve and/or extend the life of concrete structures. However, for both processes, human interventions are required leading to an added cost in labor work. The cost for crack injection in tunnel elements can be estimated to be of the order of €130 per m^3 of concrete (COWI, personal communication). Furthermore, sometimes it is not possible to get to the damaged areas for repairing because of their location and/or environmental conditions. Examples of difficult accessible structures are underground constructions, water tunnels and radioactive waste storage tanks among others.

Due to these facts, self-healing of cracked concrete has been examined for some years. In fact, concrete has always some self-healing ability. The hydration of unhydrated cement particles causes the filling of small cracks. However, this autogenous healing is limited to small cracks (<200 µm) and requires the presence of water.[5] The self-healing concept is quite interesting. It is comparable to the phenomenon occurring when a plant or an animal has a small cut. The latter can be self-healed by the natural biologic repair mechanisms that are pre-existent in these organisms. Hence, the intriguing question for this field of study is "Can we achieve a similar process in concrete?" Several studies have been pointing to an affirmative answer. However, the costs are yet too high to be considered in industrial applications.

It is not our purpose to provide full information about the biological self-healing process in concrete due to its wide spectrum. This paper describes the actual challenges to bring an efficient biological self-healing product to the concrete market with the guaranty that this product can attain legislative requirements and also be cost-effective.

BIOLOGICAL SELF-HEALING CONCRETE

The phenomenon of self-healing in general is already under study since 1970[6] starting with the investigation of this phenomenon in cracks of polymers. However, only after 2001 with the article of White et al.,[7] the topic of self-healing attracted the attention of several investigators. Three main definitions of self-healing and self-repairing have already been provided.[4,8-9] However, the central issue is that for concrete to be considered as self-healing, the concrete should not require any treatment to improve the action of the self-healing agents.

Several authors have dealt with microbial induced carbonate precipitation as being a possible approach for the treatment (self-healing/repairing) of concrete structures.[5,10-16] The microbial hydrolysis of urea ($CO(NH_2)_2$) can be used as a way to place a restoring and protective layer of calcium carbonate ($CaCO_3$) on degraded limestone.[17] The hydrolysis of urea is catalyzed by an urease enzyme and in the process carbonate (CO_3^{2-}) and ammonium (NH_4^+) ions are produced (equations 1 to 4). For each mole of urea two moles of ammonium ions and one mole of carbonate ions are formed (global reaction of hydrolysis of the urea – equation 5).

$$CO(NH_2)_2 + H_2O \rightarrow NH_2COOH + NH_3 \quad (1)$$

$$NH_2COOH + H_2O \rightarrow NH_3 + H_2CO_3 \quad (2)$$

$$2NH_3 + 2H_2O \rightarrow 2NH_4^+ + 2OH^- \quad (3)$$

$$2OH^- + H_2CO_3 \rightarrow CO_3^{2-} + 2H_2O \quad (4)$$

$$CO(NH_2)_2 + 2H_2O \rightarrow 2NH_4^+ + CO_3^{2-} \quad (5)$$

The calcium carbonate precipitation process becomes complete when calcium ions are present and the chemical reaction between carbonate ions and calcium ions results in the deposition of a white precipitate (equation 6).

$$CO_3^{2-} + Ca^{2+w} \rightarrow CaCO_3 \quad (6)$$

It was also described that for a proper deposition of calcium carbonate it is necessary to have what is called sites of crystal nucleation.[18] Due to the negative charge on the bacterial cell wall, calcium ions can be bound to it. This fact, allied to the release of carbonate ions from

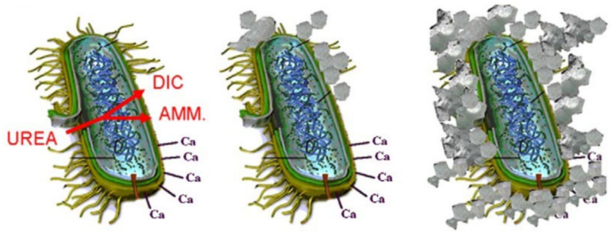

Figure 1: Schematic overview of the ureolytic carbonate precipitation occurring at the microbial cell wall. DIC: Dissolved Inorganic Carbon; AMM: Ammonia[10]

the hydrolysis of the urea, results in the formation of calcium carbonate crystals on the cell wall (Figure 1). Biodeposition of calcium carbonate using bacterial strains can thus be taken as a process to provide a larger and faster calcium carbonate precipitation when compared with the natural precipitation of this compound.

Several micro-organisms have the capacity to rapidly hydrolyze urea with rates of 16,5 grams of urea consumed per gram cell dry weight per hour.[14] Despite the good ureolytic activity of several microorganisms, it was found that the ones closely related to *Bacillus sphaericus* show a greater ability to hydrolyze urea leading to precipitation of a larger amount of calcium carbonate.[19-20] Two of the best performers regarding the ureolytic activity and precipitation of calcium carbonate are *Bacillus sphaericus* LMG22557 and *Sporosarcina pasteurii* DSM33 (previous called *Bacillus pasteurii* DSM33).[10,21] They can produce up to 0,4 grams of $CaCO_3$ per gram cell dry weight per hour.[22]

However, to bring such self-healing agent to the market, some practical aspects should be taken in consideration. The use of pure bacterial cultures with high specific ureolytic activities has considerable importance when related to fundamental research but, generally, these axenic pure cultures, represent a high cost for industrial application. For this specific case, despite of the high ureolytic activity and good calcium carbonate precipitation, production costs are of a very high importance. Thus, there is an urgent need to develop the production of a bio-additive at much lower costs. A possible approach would be the production of a mixed and non-axenic bacterial culture that could perform as well, or even better, than the pure strains regarding urea hydrolysis and calcium carbonate precipitation.

ECONOMIC EVALUATION OF SELF-HEALING BACTERIAL CONCRETE

BACILLUS SPHAERICUS AXENIC PRODUCTION

One of the major problems to apply the bacterial induced calcium carbonate precipitation to achieve self-healing in concrete is the total cost of the product used to incorporate in the concrete. Since concrete itself is relatively inexpensive, costing around €60 to €75 per m^3 of applied concrete at the Belgian market, any product that is added to the concrete with a price above €15 to €20 per m^3 of applied concrete is considered too expensive to be taken in consideration in the normal market (Coeck NV, Belgium, personal communication). Moreover, at industrial scale, besides the price, it is necessary to look also for the warranty provided for such product. According to the European Standard EN206-1:2000,[23] any concrete structure well applied, should fulfill a service life of, at least, 50 years. However, the warranty period in which the contractor is responsible for defects in the concrete structure, is normally 10 years and cracks are not included. Nowadays correct properties of the concrete structure are achieved by means of maintenance using some repairing agents (Coeck NV, Belgium, personal communication). If a bio-based product could give the warranty of a longer life for the concrete, the benefits will overcome the costs and new market can be established. The latter will be enhanced if the bio-based approach is also environmental friendly, thus winning support in the eco-tuned market.

Actually, the bio-based additive for concrete, consisting of encapsulated spores to mix in the concrete before the casting process, results in prices of €5760 per m^3 of applied concrete, making this approach unlikely to be applied (see Box 1). This price is mainly due to the need of aseptic conditions to produce the microbial spores, due to the use of expensive growth media and to the necessary labor. Moreover, the encapsulation process of microbial spores is expensive. Depending on the capsules used, but also on the yield of the encapsulation process and even on the percentage of capsules needed, this step to obtain encapsulated spores can cost between €30 and €50 per kg of spores, contributing significantly to the total price of the final product.

For some applications when there is urgency for the healing/repairing of the structure when a crack appears, the cost of such a product could be acceptable. For example, in an underground museum or library, the quick healing/repairing of cracks is crucial to the maintenance of the right conditions to preserve the highly valuable objects inside. In such instances, the price of such a product is of secondary importance since it provides guaranties that the cracks will be repaired in a matter of days to

Box 1: Estimated total costs to produce a microbial based additive capable to bring about self-healing of micro-cracks in concrete under optimal (submerged) conditions. Estimates based on in house price calculations

1. Costs to produce 1 kg of the effective *Bacillus sphaericus* spores:
   To calculate the cost to produce 1 kg of *Bacillus sphaericus* spores one should take in consideration:

   - MBS sporulation medium used for spores culture[22]
   - Required labor work to assembly and maintain the axenic process
   - Sterilization and energy requirements
   - Production yield of each batch (grams of cell dry weight produced per L)

   Considering the following, one can calculate:

   - Production scale of $1m^3$
   - A labor work cost at 50 €/h
   - 3 hours of required labor work
   - An industrial standard steam sterilization process
   - The electricity cost at 0,09 €/kWh
   - A maximum yield of 3,5 kg CDW/m^3

MBS medium cost	1370	€/m^3
Labor work cost	150	€/m^3
Sterilization cost	5	€/m^3
Total cost	435	€/kg

2. Costs to produce 1 kg of self-healing agent:
   To calculate the cost to produce 1 kg of self-healing agent one should take into consideration:

   - The encapsulation process
   - The addition of the required nutrients
   - The required amount of self-healing agent per m^3 of concrete

   Considering the following, one can calculate::

   - An encapsulation cost of 40 €/kg
   - The capsules do not increase the total weight of the final product
   - The addition of urea (20 g/kg) and $CaCl_2$ (35 g/kg)
   - The addition of 0,5% (w/w) of self-healing agent
   - An average concrete density of 2400 kg/m^3

Encapsulated spores	475	€/kg
Self-healing product (encapsulated spores + nutrients)	480	€/kg
Quantity of self-healing agent	12	kg/m^3
Total cost	5760	€/m^3

3. An important value is the cost per activity unit. Considering the best result of 16,5 grams of urea consumed per gram cell dry weight per hour[14] one can calculate the total cost of spores that can be expressed as about 350 €/g urea hydrolyzed/g CDW.h

crete from the outside will be the treatment of choice. The use of epoxy resins in the case of smaller cracks times less expensive when compared with the application

Table 1. Direct cost in Euro for axenic production of *Bacillus sphaericus* and non-axenic production of an ureolytic bacterial mixed culture

	Axenic pure culture production	Non-axenic mixed culture production	Factor
Spores cost per kg	435	145	30
Self-healing agent cost per kg	480	595	8
Cost per activity unit (i.e. g urea hydrolyzed per g CDW per h)	350	43	8

of bio-based technology (Denys NV, Belgium, personal communication).

To achieve prices of about €15 to €20 per m^3 of applied concrete, one must work with cultures produced under less expensive (non-sterile) environmental conditions. The process must furthermore be optimized to obtain viable spores that maintain ureolytic activity over long storage time to perform the hydrolysis of the urea and provoke a massive calcium carbonate precipitation. It is also necessary to find an inexpensive encapsulation process, providing the necessary protection to the spores, maintaining or slightly altering the concrete proprieties.

Summarizing, from the economical point of view, for a bio-based product for self-healing and/or self-repairing in concrete structures, prices of about €15 to €20 per m^3 of applied concrete are warranted. Even at such levels of costs, for this type of product to be added to concrete, the markets will require that it will be guaranteed to be effective over a certain period, depending on the type of cracks to be healed. This period may range from weeks to months in case of early age cracks due to autogenous or drying shrinkage up to several decades due to the aging of the structure.

NON-AXENIC UREOLYTIC SPORES PRODUCTION

As indicated before, the main problem of using axenic pure cultures is the high production cost of such bio-material. Thus, a possible solution would be the development of a less costly process to obtain ureolytic sporulating bacteria. It must be possible to select an ureolytic sporulating bacterial community starting from soil, wastewaters, activated sludge or any kind of material rich in active microbial activity.

Ureolytic bacteria can be found almost everywhere. Under the right stimulus, one can select the sporulating strains in order to obtain an ureolytic non-axenic mixed culture able to perform as well, or even better, than the pure cultures.

Considering that such non-axenic production is possible and that the main stimulus are the presence of considerable amounts of urea and a thermal shock to induce sporulation one can estimate some costs to produce a non-axenic ureolytic mixed culture. One can consider activated sludge as raw material and a feed containing an easy degradable carbon source (such as sucrose) and urea (in considerable amounts). Considering also a regular activated sludge one can easily get about 12 kg/m^3 of dry organic matter after drying. This value can be assumed as the production yield of such non-axenic process (see Box 2).

Making then a direct comparison between the axenic production of *Bacillus sphaericus* spores and the production of such mixed culture of ureolytic spores (Table 1) one can easily conclude that further studies should be performed using the mixed culture. Furthermore, the development of this new technology might contribute to decrease the production cost.

CONCLUDING REMARKS

In order to use the MICP technology under real applications on concrete structures, the following three points should be taken in consideration:

i. Despite of the lower costs estimated for the non-axenic production process, active ureolytic bacterial spores are still too costly for practical application and prices below €2 per kg of spores dry weight should be strived for.
ii. The encapsulation process of the spores or of vegetative cells should be achieved by means of inexpensive methods so that the overall extra costs per kg of spores decrease from the current €40 to a maximum of €15.
iii. There are at present two markets for the application of the “submersed” MICP technology available i.e. pillar bridges respectively tunnels. Indeed the right conditions to provide the proper microbial activity which depends on ample water supply are, for these two environments, provided.

Box 2: Estimated total costs to produce a non-axenic microbial based additive capable to bring about self-healing of micro-cracks in concrete under optimal (submerged) conditions. Estimates based on in house price calculations

1. Costs to produce 1 kg of the effective ureolytic mixed culture of bacterial spores:
   To calculate the cost to produce 1 kg of such mixed culture one should take in consideration:

   - Culture medium used for spores production
   - Required labor work to assembly and maintain the process
   - Energy requirements
   - Production yield of each batch (grams of cell dry weight produced per L)

   Considering the following, one can calculate:

   - Production scale of $1 m^3$
   - A labor work cost at 50 €/h
   - 3 hours of required labor work
   - Industrial standard equipment
   - The electricity cost at 0,09 €/kWh
   - A maximum yield of 12 kg CDW/m^3

Medium cost	9	$€/m^3$
Labor work cost	150	$€/m^3$
Energy cost	15	$€/m^3$
Total cost	145	€/kg

2. Costs to produce 1 kg of self-healing agent:
   To calculate the cost to produce 1 kg of self-healing agent one should take into consideration:

   - The encapsulation process
   - The addition of the required nutrients
   - The required amount of self-healing agent per m^3 of concrete

   Considering the following, one can calculate:

   - An encapsulation cost of 40 €/kg
   - The capsules do not increase the total weight of the final product
   - The addition of urea (20 g/kg) and CaCl2 (35 g/kg)
   - The addition of 0,5% (w/w) of self-healing agent
   - An average concrete density of 2400 kg/m^3

Encapsulated spores	545	€/kg
Self-healing product (encapsulated spores + nutrients)	595	€/kg
Quantity of self-healing agent	12	kg/m^3
Total cost	714	$€/m^3$

3. An important value is the cost per activity unit. Considering the best result obtained with these non-axenic cultures is the same obtained for the axenic ones (16,5 grams of urea consumed per gram cell dry weight per hour) [14] one can conclude that the total cost of spores can be expressed as about 43 €/g urea hydrolyzed/g CDW.h

ACKNOWLEDGMENTS

This work was supported by the Ghent University Multidisciplinary Research Partnership (MRP) Biotechnology for a sustainable economy and also by the SHeMat project "Training Network for Self-Healing Materials: from Concepts to Market" within the scope of the Seventh Framework Programme by the European Commission's Marie Curie programme. This work was also supported by Avecom NV, Belgium.

BIBLIOGRAPHY

1. Cement Trust. *What is the development impact of concrete?*, Retrieved 10 January 2013 (http://cementtrust.wordpress.com/a-concrete-plan/)
2. NRMCA – National Ready Mixed Concrete Association. *Ready mixed concrete production statistics*. Retrieved 10 January 2013 (http://www.nrmca.org/aboutconcrete/)
3. ACI Committee 224 (2007). *Causes, evaluation and repair of cracks in concrete structures*. ACI 224.1R-07. American Concrete Institute, Michigan (http://mwengineering.net/wp-content/uploads/2241r_07.pdf)
4. Mihashi, H., Nishiwaki, T. (2012). *Development of engineered self-healing and self-repairing concrete-state-of-the-art report*. Journal of Advanced Concrete Technology, 10, 170-184 (https://www.jstage.jst.go.jp/article/jact/10/5/10_170/_article)
5. Van Tittelboom, K., De Belie, N., De Muynck, W., Verstraete, W., (2010). *Use of bacteria to repair cracks in concrete*. Cement and Concrete Research, 40, 157-166 (http://www.sciencedirect.com/science/article/pii/S0008884609002361)
6. Malinskii, Y. M., Prokopenko, V. V., Ivanova, N. A., Kargin, V. A., (1970). *Investigation of self-healing of cracks in polymers*. Polymer Mechanics, 6, 240-244 (http://link.springer.com/article/10.1007%2FBF00859196)
7. White, S. R., Sottos, N., R., Geubelle, P. H., Moore, J. S., Kessler, M. R., Sriram, S. R., Brown, E. N., Viswanathan, S., (2001). *Autonomic healing of polymer composites*. Nature, 409, 794-797 (http://www.nature.com/nature/journal/v409/n6822/full/409794a0.html)
8. Igarashi, S., Kunieda, M., Nishiwaki, T., (2009). *Research activity of JCI technical committee TC-075B: Autogenous healing in cementitious materials*. Proceedings of 4th International Conference on Construction Materials: Performance, Innovations and Structural Implications, 89-96 (http://www.jci-net.or.jp/j/jci/study/tcr/tcr2011/TC091A.pdf)
9. De Rooij, M. R., Schlangen, E. (2011). *Self-healing phenomena in cement-based materials*. Draft of State-of-the-Art Report of RILEM Technical Committee 221-SHC (http://www.springer.com/engineering/civil+engineering/book/978-94-007-6623-5)
10. De Muynck, W., Cox, K., De Belie, N., Verstraete, W., (2008). *Bacterial carbonate precipitation as an alternative surface treatment for concrete*. Construction and Building Materials, 22, 875-885 (http://www.sciencedirect.com/science/article/pii/S0950061806003667)
11. Gollapudi, U. K., Knutson, C. L., Bang, S. S., Islam, M. R., (1995). *A new method for controlling leaching through permeable channels*. Chemosphere, 30, 695-705 (http://www.sciencedirect.com/science/article/pii/004565359400435W)
12. Wang, J., Van Tittelboom, K., De Belie, N., Verstraete, W., (2010). *Potential of applying bacteria to heal cracks in concrete*. Proceedings of the second international conference on sustainable construction materials and technologies. Italy: Ancona, 1807-1818 (http://www.claisse.info/2010%20papers/k48.pdf)
13. Wang, J., De Belie, N., Verstraete, W., (2011). *Self-healing concrete by use of diatomaceous earth immobilized bacteria*. 3rd International Conference on Self-healing materials, Bath, 27-29 June 2011 (https://biblio.ugent.be/publication/2038180)
14. Wang, J., De Belie, N., Verstraete, W., (2011). *A microbial based system developed for self-healing concrete cracks*. 7th International Symposium on Cement Based Materials for a sustainable Agriculture (CSA), Quebec, 18-21 September, (https://biblio.ugent.be/publication/2956354)
15. Wang, J., Van Tittelboom, K., De Belie, N., Verstraete, W., (2012). *Use of silica gel or polyurethane immobilized bacteria for self-healing concrete*. Construction and building materials, 26, 532-540 (http://www.sciencedirect.com/science/article/pii/S0950061811003175)
16. Wang, J., De Belie, N., Verstraete, W., (2012). *Diatomaceous earth as a protective vehicle for bacteria applied for self-healing concrete*. Journal of Industrial Microbiology and Biotechnology, 39, 567-577 (http://www.ncbi.nlm.nih.gov/pubmed/21927907)
17. Dick, J., De Windt, W., De Graef, B., Saveyn, H., Van der Meeren, P., De Belie, N., Verstraete, W., (2006). *Bio-deposition of a calcium carbonate layer on degraded limestone by Bacillus species*. Biodegradation, 17, 357-367 (http://www.ncbi.nlm.nih.gov/pubmed/16491305)

18. Hammes, F., Verstraete, W. (2002). *Key roles of pH and calcium metabolism in microbial carbonate precipitation*. Environmental Science and Biotechnology, 1, 3-7 (http://link.springer.com/article/10.1023%2FA%3A1015135629155)
19. Hammes, F., Seka, A., de Knijf, S., Verstraete, W., (2003). *A novel approach to calcium removal from calcium-rich industrial wastewater*. Water Research, 37, 699-704 (http://www.sciencedirect.com/science/article/pii/S0043135402003081)
20. Hammes, F., Boon, N., de Villiers, J., Verstraete, W., Siciliano, S. D., (2003). *Strain-specific ureolytic microbial calcium carbonate precipitation*. Applied and Environmental Microbiology, 69, 4901-4909 (http://www.ncbi.nlm.nih.gov/pmc/articles/PMC169139/)
21. Qian, C., Wang, J., Wang, R., Cheng, L., (2009). *Corrosion protection of cement-based building materials by surface deposition of CaCO3 by Bacillus pasteurii*. Materials Science and Engineering C, 29, 1273-1280 (http://www.sciencedirect.com/science/article/pii/S0928493108003032)
22. Wang, J., (2012). *Self-healing concrete by means of immobilized carbonate precipitating bacteria*. PhD Thesis. ISBN 978-90-8578-583-5 (https://biblio.ugent.be/publication/3158752)
23. European Committee for Standardization. (2000). European Standard EN206-1. *Concrete – Part 1: Specification, performance, production and conformity (http://beton-profi.ru/standards/EN206-1.pdf)*