IS THERE A MARKET FOR SELF-HEALING CEMENT-BASED MATERIALS?

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Cement-based building materials are, by definition, made of cement. Cement particles react with water producing hydration products. These products “glue” cement and aggregate particles together to a cement paste, mortar or concrete. The resulting material is a brittle system, susceptible to cracking under external loads and/or imposed deformations. Cracks are supposed, and known, to jeopardize the strength, stiffness, durability and service life of concrete structures. The majority of degradation studies reveal that transport processes in concrete are the main cause of durability problems. The presence of cracks will increase the transport of aggressive substances into the concrete, thus increasing the risk of degradation of the material. Given the relatively high brittleness of cement-based systems the formation of cracks is almost unavoidable. If it were possible to close or fill these cracks through a self-healing mechanism the service life of concrete structures could increase substantially.

In this contribution examples are presented of proven and potential self-healing of cement-based materials. It will be explained how the heterogeneous nature of cement-based systems can be considered as an asset when it comes to realization of built-in self-healing features. In this respect concrete can be considered as a material different from other building materials since heterogeneity-induced cracking can be considered as an inherent feature of concrete. This unavoidable cracking is, as far as its consequences for the performance of the material are concerned, allowed for in all codes implicitly by well-documented material factors. If this cracking could be used as trigger for starting, or intensifying, a self-healing process, the magnitude of material factors could be reduced, thus reducing building costs and saving raw materials. Finally it will be mentioned briefly that self-healing could contribute to better esthetic performance of concrete.

Keywords: Self-healing, self-repair, self-healing concepts, materials design, market, sustainability, benefits

1 Introduction

Today self-healing is “hot”. This seems to be the case in various fields, particularly in those fields that have the name to be high-tech. Self-healing materials are also named smart or high-tech materials. Building materials, however, are generally bulk materials and qualified as low-tech. Concrete, the most widely used man-made building material, is one of these bulk materials. Although low-tech it is, as will be discussed later, an extremely complex material. In spite of a lack of comprehensive knowledge of reaction processes in hydrating cement and complete understanding of the microstructure of the material, concrete structures have been built since more that 150 years. Many old structures have meanwhile disappeared, and this for a variety of reasons. One of the reasons is undoubtedly the degradation of the material with time. But there are also beautiful examples of old concrete structures that are still in perfect condition. Even concrete structures that have been exposed to similar environmental and mechanical loads may perform differently.
It if where possible to know the reason for different behaviour of concrete structures that are exposed to largely similar conditions, we might have the key for designing durable structures with low or negligible maintenance and repair costs. An enhanced service life of concrete structures will reduce the demand for new structures. This, on its turn, results in the use of less raw materials and an associated reduction of pollution, of energy consumption and CO2 production. Statistics convincingly show the enormous amounts of money spent by society due to lack of quality and durability of concrete structures. The cost for reconstruction of bridges in the USA has been estimated between $20 billion and $200 billion [1]. The average annual maintenance cost for bridges in that country is estimated at $5.2 billion. Comprehensive life cycle analyses indicate that the indirect costs due to traffic jams and associated loss of productivity are more than 10 times the direct cost of maintenance and repair [2]. In The Netherlands one third of the annual budget for large civil engineering works is spent on inspection, monitoring, maintenance, upgrading and repair. In the United Kingdom repair and maintenance accounts for almost 45% of the UK’s activity in the construction and building industry [3]. According to the DEFRA [4] the building and construction industry is estimated to be responsible for up to 50% of the CO2 production in the UK. The CO2 production associated with the production of cement is estimated at 5 - 7% of the total CO2 production in the world. Given the rapid growth of China’s and India’s economy, this figure is expected to increase dramatically if the technology to produce cement remains unchanged.

Enhancing the longevity of our built infrastructure will undoubtedly reduce the impact of mankind’s activities on the stability of the biosphere. Attempts to justify fundamental and risky research by promising solutions of the serious environmental problems society is faced with today may sound a bit modish. This, however, should not keep us from starting this research. Not starting this research will worsen the situation. Moreover, a direct benefit for many parties involved in the building industry and for the society as a whole is conceivable if it were possible to improve the quality and service life of concrete structures. The question here is how self-healing concrete could really contribute to solving the aforementioned environmental and societal problems.

2 Basic features of reinforced concrete

Concrete is the most widely used man-made building material. Concrete structures have been built since the discovery of Portland cement in the midst of the nineteenth century. The reaction of Portland cement with water results in hydration products, which glue the reacting cement particles together to hardened cement paste. If cement and water are mixed with sand the resulting product is called mortar. If the mixture also contains coarse aggregates the resulting product is called concrete. It is a quasi-brittle material, strong in compression but relatively weak in tension. The compressive strength of traditional concretes varies between 20 and 60 MPa. By using a low water/powder ratio, improved particle packing and special additives, high strength concrete can be produced with strength values up to 150 to 200 MPa. On lab-scale cement-based composites have been produced with compressive strength up to even 800 MPa.

Concrete elements loaded in bending or in tension easily crack. For that reason reinforcement is installed. Passive reinforcement is activated as soon as the concrete cracks. The formation of cracks is considered an inherent feature of reinforced concrete. It must be emphasized that in reinforced concrete structures cracks as such are not considered as damage or failure and cracking as such does not indicate a safety problem.
The crack width, however, should not exceed a prescribed crack width. Too wide cracks may reduce to capacity of the concrete to protect the reinforcing steel against corrosion. Corrosion of reinforcing steel is the major reason for premature failure of concrete structures. Apart from these macro-cracks very fine cracks, i.e. microcracks, may occur within the matrix due to restraint of shrinkage deformations of the cement paste (Fig. 1). Microcrack are an almost unavoidable feature of ordinary concrete. If microcracks form a continuous network of cracks they may substantially contribute to the permeability of the concrete, thus reducing the concrete’s resistance against ingress of aggressive substances.

![Typical picture of cracks caused by restrained shrinkage of cement paste. Concrete made with w/c = 0.45.](image)

Even though cracks can be judged as an inherent feature of reinforced concrete and the existence of cracks does not necessarily indicate a safety problem, cracks are generally considered undesirable for several reasons. The presence of cracks may reduce the durability of concrete structures. In case structures have to fulfil a retaining function, cracks may jeopardize the tightness of the structure. Completely tight concrete may be required in case the structure has to protect the environment against radiation from radioactive materials or radioactive waste. Cracks may also be undesirable for aesthetic reasons. Not only cracks, but also the inherent porous structure of concrete can be a point of concern. In case pores are connected and form a continuous network, harmful substances may penetrate the concrete and may chemically or physically attack the concrete or the embedded steel.

### 3 Inherent self-healing potential

Traditional concrete mixtures are made with a water/cement ratio (by weight) between 0.40 to 0.55. Theoretically a water/cement ratio of 0.4 is enough for complete reaction of all the cement. In practice, however, in a mixture with a w/c = 0.4 not more that about 70% of the cement will react. The remaining 30% is left unreacted in the cement paste. The amount of unreacted cement is higher the coarser the cement and the lower the water/cement ratio. In case cracking of the concrete occurs, unreacted cement cores may become exposed to moisture that penetrates the crack. In that case the hydration process may start again and hydration products may start to fill up the crack.
This inherent self-healing mechanism is known for a long time already. Autogenous healing of cracks in fractured concrete has been noticed by the French Academy of Science in 1836 already in water retaining structures, culverts and pipes [5]. According to Hearn [6] the self-healing phenomenon was studied already by Hyde [7] at the end of the nineteenth century. A more systematic analysis of healing phenomena dates back to 1926 and was executed by Glanville [8]. Already at that time a distinction was made between self-healing and self-sealing. In the first case the original strength of the concrete is completely recovered, whereas in the second case leaking cracks will close but no strength recovery is obtained. Glanville’s first studies have been followed by studies of cracks in bridges [9,10]. In 1996 Jacobsen et al [11] observed self-healing of concrete specimens that had been exposed to freeze-thaw cycles. Self-healing of leaking cracks were studied extensively by Clear [12], Hearn [5,6,13] and Edvardsen [14]. Otsuki et al [15] suggested that self-healing of microcracks could have been the reason for densification of the concrete cover, thus reducing the rate of migration of chloride ions into the concrete (Fig. 2). Self-healing of microcracks as a cause of reduced chloride ingress has also been suggested by Fidjestol et al. [16], and Bakker [17]. Reinhardt et al [18] found that self-healing of cracks will benefit from higher temperatures.

In all the afore-mentioned studies self-healing is considered an inherent feature of cement-based systems. This feature makes concrete a material with a lot of ‘forgiveness’. Mankind has taken advantage of this peculiar property of concrete even though it was never designed on purpose to be a self-healing material. Neither the self-healing process itself, nor the required preconditions for this process to happen are completely understood today. A consequence of this is that for the time being this self-healing capacity of cement-based systems is considered a positive feature of concrete, but too uncertain yet to be taken into account explicitly in the design of concrete structures. Only a few exceptions are known, for example in the design of watertight cellars or reservoirs made of reinforced concrete, where designers explicitly count on the occurrence of self-healing of cracks [19,20]. To ensure that these structures will behave liquid tight the crack width should not exceed a certain critical value. The acceptable crack width depends on the pressure drop over the length of the crack, the crack width and the stability of the crack. Almost no criteria for the type of concrete itself are given.
This in fact demonstrates that the self-healing capacity of ordinary concretes is considered a by-product of the material rather than a feature that could be manipulated by sophisticated design of the mixture.

4 Designing for self-healing: The principle

The word healing presupposes that an initially sound material has got damaged. Healing follows on damage. The occurrence of a crack, for example, can be the trigger for the healing process. It can trigger the transport of material from any place in the material to the ‘wound’, where it should then react or precipitate, thus healing the crack. Other healing mechanisms are conceivable as well, but the aforementioned one is probably the one most cited in literature. The internal transport of material presupposes the presence of transportable material, suitable transport channels and a driving force. Porous cement paste, with pores ranging from 2 nm (gel pores) to 10 μm, is a perfect matrix through which transport of a healing substance could take place, particularly through the bigger pores. The healing substances can in principle be mixed in the concrete in the same way as aggregate particles. This can be in the form of capsules, which contain the healing substance until the moment a crack intersects the capsule and the substance starts flowing into the crack and penetrates into the porous cement matrix, thus gluing the crack surfaces together. If compact spherical capsules are used the probability that a crack does not intersect with a capsule is conceivable. The probability of intersection with inclusions will increase if the inclusions are added in the form of randomly distributed needle-like hollow particles [21]. The design of inclusions is still a serious research issue. Inclusions must be strong enough to withstand ‘impact’ forces to which they are exposed during mixing and placing of the concrete. They should be weak enough, however, to break open on intersection with a crack.

Many research questions are still ahead. The question is whether these research efforts are justified. Is the industry really interested in the availability of smart concrete with the potential to heal once being cracked or when suffering from degradation processes? The answer is “yes”. In section 1 it has been mentioned that huge amounts of money are involved today in maintenance and repair of infrastructural works. These costs are a burden for society. Reduction of these costs will save large amounts of money and will, moreover, also improve the profile of the building sector.

5 Self repair versus man-made repair

5.1 The principle

The performance of structures with elapse of time is often presented with graphs like that shown in Figure 3a. Curve A describes how after some time gradual degradation occurs until the moment that first repair is urgently needed. The durability of concrete repairs is often a point of concern. Very often a second repair is necessary only ten to fifteen years later. Spending more money initially in order to ensure a higher quality often pays off. The maintenance-free period will be longer and the first big repair work can often be postponed many years (Fig. 3a, curve B and Fig. 3b). Apart from saving direct costs for maintenance and repair the savings due to reduction of the indirect costs are generally most welcomed by the owner.
The experience is that a higher initial quality of the material results in postponement of repair and, in the end, a reduction of the costs for maintenance and repair. A logic next question concerns the optimum between the increasing initial costs and the costs for maintenance and repair. The extreme case would be that no costs for maintenance and repair have to be considered at all because the material is able to repair itself. Figure 4a schematically shows the performance of a structure made with self-repairing material. On the occurrence of a small crack or the start of any physical or chemical degradation process, the material gradually starts to repair itself and the structure will regain its original level of performance or a level close to that. Figure 4b shows the costs. In this figure inflation and interest are not considered yet. The initial costs will be substantially higher than that of a structure made with traditional concrete. The absence of maintenance and repair costs, however, could finally result in a financially positive situation for the owner.

5.2 Cost evaluation – Completeness and comprehensiveness

If it were possible to convince an owner that a higher initial investment by using a more expensive self-repairing building material finally pays off, he will most probably be prepared to use this material. In his judgement he will consider the savings of costs for maintenance and repair. Since a self-repairing structure does not need any repair there is no need to take the structure out of service during repair works. Substantial savings of indirect costs can then be added to the savings of direct repair costs. Furthermore, reduction of the costs for inspection and monitoring will reduce the exploitation costs.
In a comprehensive analysis of costs and savings the extended service life of a structure should be considered as well. Extension of the service life has a big impact on the total materials cycle and on the associated costs. Demolishing buildings and subsequent transport of rubble, for example, have turned out to be unattractive from the sustainability point of view [22]. Addressing the sustainability issue might complicate the judgement process, but is considered a prerequisite for a fair comparison of alternatives. The use of self-healing materials will increase the service life of structures, it will reducing the amount of waste, it saves scarce raw materials and it saves energy by reducing building activities for new-built. How to value sustainability aspects in a comprehensive and convincing way is still quite difficult and requires an update of currently used judgement formats.

5.3 Radical versus incremental approach

In the forgoing self-healing and self-repair have been referred to as ‘absolute’ or ‘perfect’ phenomena.

In reality, however, the self-healing and self-repairing potential of a material will be limited. This means that it is not realistic to expect that the use of self-healing materials will make all inspections, monitoring, maintenance and repair completely superfluous. The sector, however, can already benefit from incremental improvements of the self-healing capacity of a material. If the maintenance-free period can be extended and the moment of repair can be postponed, high savings are conceivable already. In this respect it is important to realise that huge scale at which concrete is being used. Scenarios are conceivable whereby minor improvements of the material can be realised at only small extra costs. Bulk production of such a material is then possible and a reduction of the costs for maintenance and repair of many concrete structures is plausible. But also situations are also conceivable whereby the degradation of a structure should be avoided at all costs because of extreme high consequences in case of failure (for example leakage of radioactive waste, see section 2). In such a case the use of a robust self-healing material could then be the only solution. If the use of a self-healing material is the only realistic solution, the extra costs of the material will be no limiting factor at all.

6 Developing self-healing material

The fact that under certain conditions traditional concretes exhibit an inherent self-healing capacity already is certainly a strong reason to intensify research in this area. In traditional mixtures the self-healing capacity comes from the still unhydrated cement in the concrete. A positive aspect of self-healing through continuous hydration is that no ‘strange’ material is added to the concrete, which could otherwise cause problems in future recycling processes. From this point of view the use of cast-in capsules containing a self-healing substance on basis of cement is preferable. Irrespective of the content of a capsule, in all cases the production of capsules is still a research topic. An alternative research direction could be the use of coarse cement instead of capsules. Under normal hydration conditions big cement particles will only partly hydrate, leaving large unhydrated cores in the cement paste. Cracks may cause the unhydrated cement cores being exposed to water and the hydration process may continue. For many practical reasons the use of coarse cement, however, is not desirable (cement becomes too slow). Moreover, it is known from past experience that only by using coarse cement the occurrence of self-healing is not guaranteed under all circumstances (see section 3).
7 Who is interested in self-healing?

Owners might be interested in a product that does not exhibit any decay. Claiming that the production of a decay-free concrete is immanent is not considered realistic. With the present state of knowledge of self-healing mechanisms in concrete no contractor will take the risk of promising a maintenance-free and decay-free concrete. In a rapidly changing building industry, however, where Design, Built and Maintenance contracts become increasingly popular, contractors may become interested indeed in the development of concrete mixtures with robust self-healing properties. Such a material could substantially reduce the maintenance costs, which enables them to take a competitive position in the market. Moreover, any reduction of the costs for maintenance and repair will enhance the profile of concrete as a building material and of the entire building sector. It is to be expected, therefore, that the building sector as a whole, and contractors in particular, will stimulate in-depth research on self-healing cement-based materials.

Since it is well known that the quality of a concrete structure does not exclusively depend on the materials characteristics [22], this research should not be restricted to the material itself, but should also include to entire design and building process.

8 Concluding remarks

Concrete is a heterogeneous and extremely complex building material. The complexity follows from the fact that the components of concrete – gravel, sand, cement and other powders - vary in size, properties and function. The presence of cracks further contributes to the heterogeneity of the material. The continuous interaction of concrete with the environment, i.e. exchange of moisture and heat, makes that it is often reasonable to be speak about concrete as a system rather than a material. The heterogeneity of this ‘system’, however, can also be considered as an asset when it comes to the development of self-healing cement-based mixtures. Capsules containing a healing agent can be mixed in the concrete without big impact on the overall performance of the system. Nevertheless, much research is still ahead. Experimental work will be indispensible to verify possible and promising self-healing concepts. In parallel with this experimental work numerical simulation can be of great help. Since the eighties of the last century a few numerical simulation programs are available which can be used for optimisation of concrete mixtures on both the micro- and the mesoscale [24]. Prediction of (micro)cracks, i.e. their number and width, provides input for analysis concerning the required healing capacity of cast-in capsules. Knowing the outcome of numerical simulations, the number of expensive and time-consuming experiments can be reduced and the development of self-healing material can be accelerated.

Once a self-healing concrete is available, the implementation in the building industry should follow. Even though the building industry has often been criticised for its reluctance to pick up new ideas, it is believed that the advantages of self-healing concrete might be so convincing and challenging, that the use of this material will certainly be considered by the industry. Much depends on whether additional changes or adaptive actions in the building process will be necessary. It is important, therefore, that studies are started on the implications of the use of self-healing concrete for the entire design and building process, i.e. the detailing, execution and curing of the concrete.
In this contribution the focus has been on self-healing of concrete through cast-in capsules. This, however, is only one possible concept. For example, hybrid concepts are conceivable as well, whereby an external trigger activates cast-in healing components. For all alternatives it holds that the implementation phase requires thorough studies on the building process on penalty of the rejection of promising high-tech innovations in the building sector.

REFERENCES