Fundamentals of Self-healing Construction Materials

Antonios Kanellopoulos and Jose Norambuena-Contreras

1 Introduction

The importance of materials science and engineering is paramount for developing new materials and components that will support our continuously growing needs as a society. At the same time, preserving our natural habitat by minimising our carbon emissions and waste of natural resources has also gained, quite rightly, significant attention over the last two decades. The concepts of structural integrity, durability and performance-based design have been the centre of attention of civil engineers over the previous two decades. In this challenging environment, self-healing materials have a significant role to play. Inspired by nature and the intrinsic healing process of living organisms, self-healing materials aim to tackle localised damage and restore the structural integrity of components.

Construction materials science is a fast-emerging field and a scientific topic that can lay the foundations of a sustainable future built environment. For many decades, the way we manufacture and use construction materials has not changed much, especially compared to other sectors. Construction materials have followed a very conservative evolution path. Nonetheless, in the last ten years, engineers have realised that the "future cities" can be realised by adopting the same standards, materials' properties, processing techniques and eventually carrying over the same problems and issues that materials used in construction have. Construction and building materials

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are used to perform specific functions and satisfy a range of requirements stipulated by design codes and standards. While there are some advancements in aspects of construction materials used, for example, the complex geometries that we can construct today with concrete because of the rheological advances of the last twenty years; there is still a lot of work to be done to transform the functionalities and alleviate the limitations of traditional construction materials.

Ashby introduced the performance index concept where the performance of any material is a direct function of two components: its material properties and a shape efficiency factor which depends on loading conditions and the geometry of the individual components [1]. Construction materials face a continuous and somewhat unfair challenge. Everyone involved in the construction process (engineers, contractors, and clients) expect them to perform. The level of tolerance for lack of performance is limited. Simultaneously, design codes are based on mechanical functionality overlooking the fact that to ensure appropriate levels of mechanical performance the materials must maintain their overall integrity which in many cases is compromised by a combination of actions rather than mechanical fatigue. This need for change is crucial because it will address challenging technical issues such as early degradation. It will considerably improve the life cycle of infrastructure assets yielding a significant reduction in maintenance and replacement regimes. Eventually, this will lead to useless materials in the long run, and it will improve the users' confidence in the reliability and safety of structures. The focus of this book will be on the primary construction materials, providing an overview of strategies, assessment techniques, challenges and the outlook of self-healing applications. This book summarises the main studies recently developed on self-healing construction materials, emphasising in the fundamental concepts, monitoring and large-scale applications of these advanced materials. Due to its applied context, the contents of this book have been focused on the construction materials mostly used in the engineering field, such as concrete, asphalt, metals and alloys. The book should be attractive to the entire community of the science and engineering of materials, working on advanced construction materials. Finally, the technical level of the chapters allows it to be a guide textbook in undergraduate and graduate courses that have a focus on construction materials.

2 Historic Perspective

Natural resources have been the basis of growth in civil infrastructure for at least 100 years. At the same time, the exploitation of natural resources and the pollution associated with their extraction and processing have been already stretching the limits of our natural habitat, putting significant pressure on the environment and human well-being. According to the United Nations, the world's urban population has quadrupled since the 1950s, reaching 4 billion today, and with an estimated 7 billion people living in cities by 2050 [2]. To sustain this rapid urbanisation rate, billions of tonnes of construction materials are required annually. All evidence and projections suggest that civil infrastructure growth will keep expanding to meet the needs of modern urban societies.

The manufacture of construction materials is a process that utilises enormous amounts of raw materials and energy due to the sheer scale of the industrial processes required. Concrete is the dominant construction material and the critical element in most infrastructure assets. However, concrete manufacture on a gigatonne scale per annum imposes extremely high energy and resource demands: with >4 billion tonnes of cement being produced annually, accounting for ~8% of global anthropogenic CO_2 emissions, and yielding an annual production of ~2 tonnes of concrete for every person on the planet [3]. In Europe, the construction sector alone is responsible for 36% of CO_2 emissions and 40% of all energy consumption [4]. Cement manufacture is a significant contributor to the whole clinkerisation process consuming ~3.5 GJ/ton [5]. Of course, environmental burden and maintaining the structural integrity and serviceability are not unique to cement-based materials. The other two dominant players in construction, asphalt and steel, face similar issues. We need all these materials to support our growth. Still, at the same time, we cannot afford to waste natural resources, and most importantly, we cannot afford to produce infrastructure assets with long-term integrity problems.

As mentioned earlier, it is the coupled phenomena that lead to degradation, and this is accelerated with the formation of cracks either on the surface or the bulk volume of construction materials. Although ways have been developed to protect the materials (e.g. porosity reducing admixtures; coatings), the problem of cracking remains. The main issue with the crack repair is that it is not straightforward for several reasons. First, formed cracks may not be visible or may not have reached the surface. Secondly, there are quite a few instances where the cracks are not accessible. Besides the technical difficulties, there is also the cost and nuisance associated with the repair and maintenance of infrastructure assets.

Historically, the concept of self-healing in construction materials is not new. It has its roots back in the Roman era, where the lime-rich mortars used then showed remarkable durability over the millennia, predominantly due to the chemical interaction between the mortar constituents and the surrounding environment. This was not an intentional engineering intervention but rather a coincidental one. It was not until recently that scientists found this feature of the Roman mortars [6, 7]. There were some references about self-healing materials over the past century, but nothing was done systematically or materialised [8]. The first systematic attempt to develop self-healing processes in materials, quite coincidentally, started from concrete by Carolyn Dry in mid-1990s [9–12].

The breakthrough and step change in self-healing materials happened in 2001 with the landmark publication in Nature Journal by the White group [13]. In this work, the researchers have embedded microcapsules of lower stiffness to the host polymeric matrix. Once a crack propagated through the material and reached a microcapsule, this ruptured, releasing an adhesive that repaired the crack. This work caught the attention of materials scientists and engineers worldwide. It inspired several studies on this topic, not only in polymers/composites but also in construction materials. This

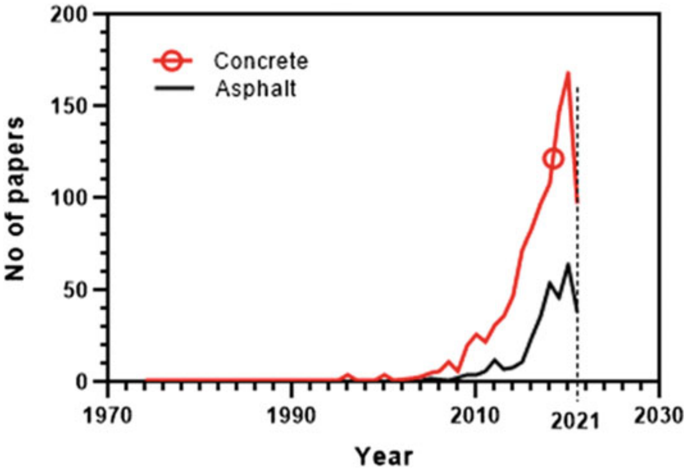

Fig. 1 Number of papers published on self-healing concrete and asphalt (*Source* Scopus; The data for 2021 concerns papers published the first quarter of 2021 only)

translated to many scientific papers published discussing self-healing principles and mechanisms in two highly used construction materials: concrete and asphalt (Fig. 1).

In asphalt, the earliest reported work on self-healing action dates back to 2003 [14], but it was not until the end of that decade when the topic lift-off [15–17]. In concrete, one can find scattered references on self-healing actions throughout the decades, as it is discussed in Chap. 2. Carolyn Dry's work was pioneering in the mid-1990s, but similar to asphalt, it was not until the end of the 2000s that systematic work on self-healing concrete was observed [18–21]. There is a strong incentive to develop a step change in the way materials are being designed and used in the construction sector. Materials' efficiency must be improved as it strongly associated with environmental impact through extensive maintenance and/or replacement regimes. Reinforced concrete structures are well known for their versatility, but they are equally known about their associated degradation problems. Likewise, highway infrastructure in the entire operation supports socio-economic growth, but disruptions, delays and public nuisance begin when issues arise. This is the reality with the two primary construction materials (concrete and asphalt) for many decades, but of course, steel structures and steel and alloys elements cannot be excluded from this discussion. Highlighting these issues does not serve the purpose of downgrading the significant progress over the last decades in manufacturing better construction materials. However, as engineers, we have to be pragmatic and recognise that construction materials are being improved. In the longer run, they still suffer the loss of structural reliability and deterioration and, as a consequence, reduce their durability.

3 Intrinsic and Engineered Healing

As material healing, we can define the recovery of some or all the properties of that material following damage initiation. The intensive research efforts of the last two decades led to the distillation of terms used to describe the mechanisms of healing in construction materials. Thus, self-healing can happen as a result of either of the following broad mechanisms (or a combination of both): *autogenous healing*, which relies on the intrinsic properties of the material and *engineered healing,* which a stimulated process by compounds and components that would not otherwise exist in a typical construction material matrix.

In cementitious systems, the former mechanism could be, for example, the continuous hydration of unhydrated cement particles. In contrast, the latter could refer to the release of an adhesive or mineral in the damaged area to promote repair. De Rooij et al. have in 2013 summarised all the autogenous healing mechanisms in cementitious matrices in three broad categories (see Fig. 2): physical, chemical and mechanical [22].

Of course, the exact manifestation of these mechanisms and how healing is promoted depends on the nature of each material. Continuous hydration works well for cementitious matrices, while elevated temperatures work well in metallic systems. In the subsequent chapters of this book, different materials and mechanisms will be considered and reviewed.

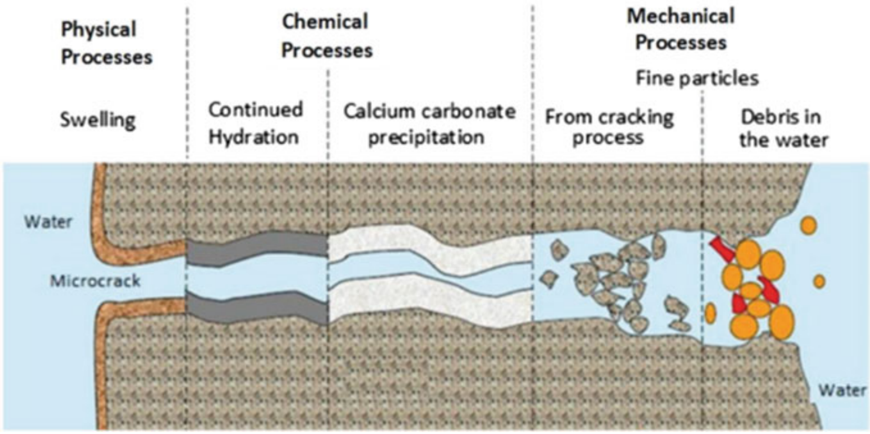

Fig. 2 Main mechanisms contributing to autogenous healing in cement-based materials (Reproduced from Rooij et al. [22])

4 Damage Prevention and Control

Dealing with damage effectively and for a considerable amount of time is the holy grail of materials science as it will enable increased functionality of components and higher levels of structural reliability. As mentioned earlier in this chapter, damage under service conditions is inevitable for any material. However, it should not be the damage leading actions upon the materials that define the performance of components. Instead, it should be our engineering/design interventions that allow materials and components to handle these actions. Therefore, there is a need to transform the way we design materials and elements.

With the progress of materials science and understanding how matter behaves, it became evident that crystal structures and molecular arrangements are essential elements that broadly define the response of materials to stimuli. With the development of advanced instrumentation, innovative manufacturing techniques and further understanding of micromechanics, scientists created a specific set of properties for the material. Advances in the field of fracture mechanics accurately predicted the formation of microdefects and their evolution, leading to the complete rupture. Information about defect creation and propagation was useful to redesign materials and components in such a way that damage is prevented. An early application of this principle was the use of steel reinforcement in concrete elements to enable the material to withstand flexural forces successfully. Nonetheless, the prime example was the rise of fibre-reinforced composites. Fibres incorporated in brittle and quasi-brittle matrices to prevent catastrophic failures. Although fibre-reinforced composites work perfectly without suffering abrupt damage, they still suffer from it. In a way, fibres avoid the occurrence of a sudden event, but they cannot control it in terms of stopping its manifestation or even reverse it. In 2007, Li and Yang [23] identified six attributes to create a concrete with self-healing functionality which at the same time will be attractive for practical large-scale applications. These attributes are summarised in Fig. 3.

Of course, these attributes can be extended to any construction material and any material in general. A self-healing material has to be able to respond immediately when damage occurs ("pervasive") while at the same time the self-healing functionality must remain active throughout the service life of a structural component ("stable"). Damage may occur at any stage of the lifecycle of an infrastructure asset. The construction sector is a bulk materials sector. The volumes used in civil infrastructure are staggering. Being financially viable for large-scale applications is extremely important for self-healing construction material ("economic").

In engineering terms, construction favours materials that have a predicted behaviour and perform reliably in various conditions. The expectation from a self-healing construction material is the same and is/will be expected to cover multiple performance and damage scenarios ("reliable"). Attaining good levels of healing fully recovering as many properties as possible is also very important ("high quality"). And last but not least, a self-healing material ideally must be able to counteract multiple damage events ("repeatable").

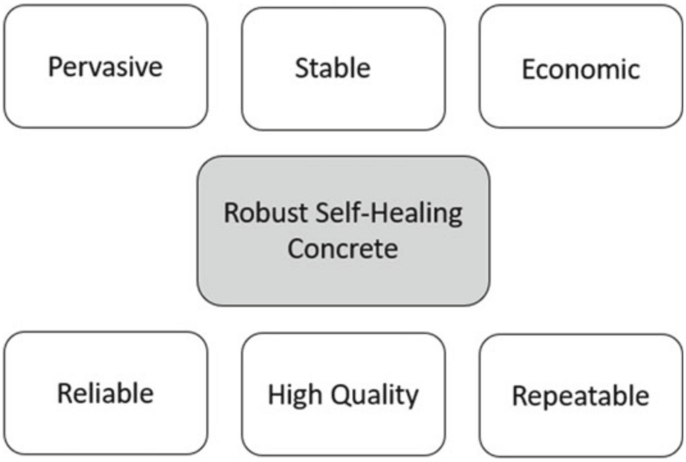

Fig. 3 Six attributes that need to be satisfied towards developing a robust self-healing concrete (according to Li and Yang [23]).

Self-healing materials aim to enhance materials' performance by controlling the effects of the damage on the performance of a component. Let us see a theoretical model of the reliability of a component concerning the service time and the occurrence of damage events. A non-self-healing material (see Fig. 4) exposed to a

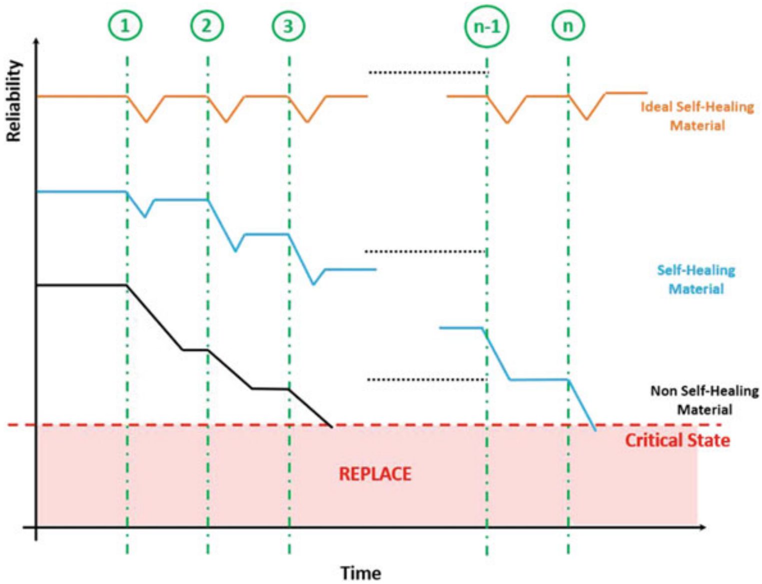

Fig. 4 Theoretical representation of the reliability of different types of materials concerning time and the occurrence of damage events.

"damage" event loses some of its reliability. Following maintenance, which sometimes occurs a long time after the damage event, the material continues to operate safely. A second event reduces the reliability further until at some point the critical state is reached, and replacement is needed. On the other side of the spectrum, an ideal self-healing material would be able to "recognise" quickly the damage event counteracting it and restoring the component's reliability to its original level after every damage event. The existing self-healing material technology provides a good recovery after the occurrence of damage, but repeated recovery is not achieved, especially in self-healing construction materials. Inevitably, the materials reach their critical state and become unreliable.

In reality, the situation is far more complex. The rate of defect formation, the speed of defect "recognition" from the material and the healing rate are all essential factors. Critical is also the loading conditions. Is the loading released after damage to allow for healing to occur, or does the load apply during the recognition and healing processes? As one can understand, these various factors can lead to large number of combinations and scenarios with varying complexity. Also, the healing processes may require minimum distances between crack-planes and even effective "communication" between "damage sensing" and "healing trigger" mechanisms. It is apparent that self-healing materials are not just "materials" but rather they are complex systems of individually engineered components.

Regarding construction self-healing systems, there is still a lot of work to achieve repeated performance and long-term reliability. Self-healing functionality in construction materials, and especially in asphalt and concrete, has been well documented over the last 15 years. Nonetheless, this functionality was demonstrated under very specific and controlled conditions. The need to move towards more complex approaches will take into account the dynamic nature of the built environment and the structural components that make it.

5 Design Considerations and Limitations

From a practical aspect, the implementation of self-healing construction materials in civil infrastructure is very attractive. Theoretically, a well-designed and manufactured self-healing component can offer solutions to the sector's long-standing problems. These include, but not limited to extensive maintenance regimes; congestions/delays; premature replacement of assets and improper or impossible repair/maintenance of difficult to reach elements. All these are also strongly related to sustainability. When it comes to designing structural elements using materials that have "self-healing" functionalities, it is important to identify the acceptable levels of damage in each case. Keeping components within their serviceability limit state is very important for having an effective self-healing framework. Of course, the proper levels of damage vary from component to component and material to material. This can be in the form of maximum acceptable crack as it is the case in concrete elements and asphalt materials, or it can also be in the form of maximum acceptable loss of strain recovery as it is the case in steel elements.

Another important factor to take under consideration is the source of damage. As mentioned earlier in this chapter, it is extremely rare to have a single source of damage on construction materials. It is the combination of mechanical and environmental weathering that will lead to the gradual manifestation of damage. With the process of time and repeated cycles of action, it can be detrimental to engineering components. To counteract this damage, it is important to understand the nature of the detrimental actions, which will help design the best course of action in deploying suitable self-healing mechanisms. What is certain is that there is no panacea in self-healing mechanisms, and not one approach can fit all.

Self-healing systems are multicomponent elements that, in most cases, trade-off their properties in exchange for long-lasting performance. Embedding microcapsules, for example, in a matrix to promote self-healing actions leads to a reduction in that matrix's mechanical performance. The medium becomes less homogenous, and preferential crack pathways are created. Similar issues arise with the use of vascular networks as well. It seems that we are downgrading the mechanical performance of our composites to achieve a better long-lasting performance that will depend less on external intervention.

All these create incompatibility with the current design codes, which in the future need to adapt and make provision for the large-scale implementation of self-healing materials. Practitioners and clients need a higher level of confidence from the design point of view regarding how the use of a "self-healing construction system" will affect the operation of the structure. Are there any knock-on effects from using such systems in bulk volumes and which are these? Currently, we do not have clear answers from the structural design point of view. Another practical barrier, at the moment, is the lack of standardisation in self-healing construction materials. The construction sector is very conservative in adopting new materials and technologies, and the lack of standardisation makes the application of self-healing materials difficult.

Although significant progress has been done the last twenty years, there is a lot of work yet to be done to create a solid framework that will yield robust self-healing construction materials. As researchers, we need to provide conclusive data and strong evidence that the materials we develop can perform: (i) outside the laboratory; (ii) at the large scale; (iii) under multiple and varying exposure conditions; (iv) repeatedly and (v) safely. In addition, we need to consider the dynamic nature of the civil infrastructure. We need materials that ideally can heal under service conditions, and they do that quickly and effectively. Shutting down a bridge, a tunnel or a highway to allow for extended healing periods is not much different from the existing situation. As people at the forefront of this scientific topic, we also need to provide enough information and investigate in detail the whole life aspect of our advanced self-healing materials. We aim for the "ideal" self-healing material with perpetual good performance, but the reality is that materials reach a point that needs replacement. How do our self-healing construction materials fit within the circular economy principles? In some cases, we do embed polymers, chemicals and even bacteria in them to promote healing. Will the material be able to be recycled in 150/200 years' time?

Addressing these technical considerations will make the way towards standardisation and implementation in the large much easier.

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