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DOI
10.1002/admi.201800736

Publication date
2018

Document Version
Final published version

Published in
Advanced Materials Interfaces

Citation (APA)
Leeuwenburgh, S. C. G., De Belie, N., & van der Zwaag, S. (2018). Self-Healing Materials are Coming of Age. *Advanced Materials Interfaces*, *5*(17), Article 1800736. https://doi.org/10.1002/admi.201800736

Important note
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Self-Healing Materials are Coming of Age

Sander C. G. Leeuwenburgh, Nele De Belie, and Sybrand van der Zwaag

General Introduction

While natural materials such as bone or skin are able to heal themselves in an autonomous manner after mechanical damage, traditional man-made materials generally lack this intrinsic capacity for self-healing. In 1969, self-healing properties were for the first time built-in inside polymeric materials[1] and in the following decades publications about self-healing in thermoplastic and cross-linked systems and concrete appeared.[2] Although, it was only in 2001, through an article about self-healing in polymer based materials published in Nature, that research in the field of self-healing materials research was really triggered.[3] By restoring their functional properties autonomously after damage, self-healing materials would offer tremendous advantages over traditional materials in application areas such as civil, chemical, electrical, aerospace, automotive, and biomedical engineering.

Autonomous self-healing does not require an external trigger (like heat or irradiation), whereas non-autonomous self-healing only occurs in response to such external triggers.[4] Another classification of self-healing materials is based on the incorporation of separate healing agents (extrinsic self-healing) as opposed to intrinsic self-healing which proceeds without the need to include separate healing agents.[4] Irrespective of their classification, self-healing of materials requires the creation of a local mobile phase which can close and repair cracks by physical flow into the crack and subsequent reformation of chemically stable interfaces.

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Current Status and Future Challenges

During the first decade of self-healing materials research, numerous novel concepts for self-healing of man-made polymeric, ceramic, metallic and composite materials have been developed. These concepts were mainly based on trial-and-error and not yet based on thorough understanding of the thermodynamics and kinetics of the process of self-healing. During the past decade, however, the field of self-healing materials research has witnessed a gradual trend towards the development of intrinsic and autonomous vs. extrinsic and non-autonomous self-healing materials. This transition has provided the self-healing materials community with a deeper understanding of the key processes which control self-healing in man-made

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DOI: 10.1002/admi.201800736

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materials. Ideally, this fundamental understanding will pave the way for *a priori* definition of design criteria to render traditional materials self-healing without the need for extensive and iterative experimental optimization cycles.

Individual Contributions to the Special Issue

The current Special Issue consisting of 10 progress reports and/or reviews provides an overview of the state-of-the-art in the field of self-healing materials research (**Figure 1**). The first five manuscripts are dedicated to self-healing of polymer-based materials, whereas the five other manuscripts describe self-healing of hard materials including metals and concrete. Self-healing of polymer-based materials generally relies on dynamic bonds in the polymeric phase which allow for reformation of bonds after cleavage. Two types of dynamic bonds have attracted most interest during the past decade, reversible covalent bonds and supramolecular chemistry. *Article 1800051* defines general design criteria for the synthesis of self-healing polymers based on dynamic covalent bonds, while *article 1800384* shows that supramolecular chemistry offers a versatile toolbox of chemical interactions to render polymeric materials self-healing. Local temporary mobility is the key concept responsible for all classes of self-healing materials, but this mobility is restricted to the outermost layer in case of functional polymeric surfaces. The progress report *1800293* focuses on a specific type of self-healing low-adherence polymeric surfaces. The first decade of self-healing materials research focused mainly on traditional engineering materials, but research interest on self-healing biomaterials has increased considerably during the past decade. Self-healing can be advantageous for polymeric biomaterials in view of their processing, clinical handling and biological performance, as shown in *article 1800118*. Nevertheless, inorganic phases are needed to combine self-healing with mechanical robustness, which is also emphasized in *article 1800177* on self-healing fiber-reinforced polymer composites.

While the combination of a hard civil engineering material and soft biological 'actors' in order to reach self-healing behaviour does not seem intuitively very logical, *article 1800074*, demonstrates a surprisingly effective method of using spore-forming alkaliphilic bacteria in combination with special bacterial food to make microcracks in concrete heal themselves autonomously in the presence of moisture. But this is only one of the discussed methods next to autogenous healing, use of mineral additions, crystalline admixtures or (superabsorbent) polymers, and micro-, macro- or vascular encapsulated polymers and minerals.

The contribution in *article 1701378* discusses models for mechanical self-healing, transport processes in materials with embedded healing systems, fully coupled models and other modelling techniques used to simulate SH behaviour. The mechanics models discussed include those based on continuum-damage-healing mechanics (CDHM), micro-mechanics, as well as models that use discrete elements and particle

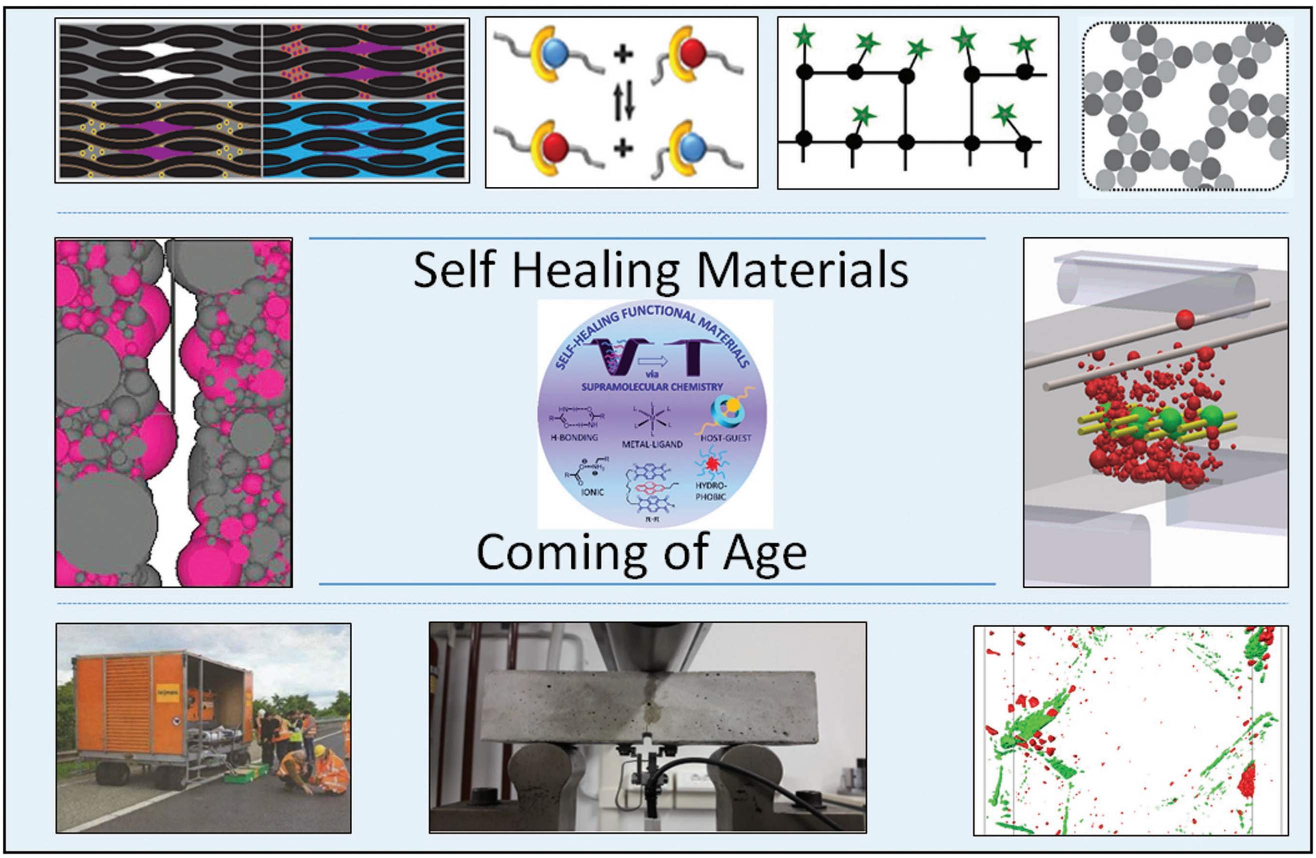

Figure 1. Collage of self-healing materials addressed in the current Special Issue. Reproduced with permission.[5–14]

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methods. While focussed on the self-healing of concrete, the mechanical models presented have a wider application to other materials too. Such quantitative models are of key importance for the introduction of novel materials concepts into a market where guaranteed long term behaviour has to be demonstrated upfront in the design phase of the building or structure.

For predictive modelling of behaviour in conventional materials and their self-healing analogues it is crucial to have reliable data about the damage location and dimensions both prior and after the healing reaction. The detection of such parameters is the topic of contribution *1800179*. The paper makes clear that the determination of the damage characteristics with the required spatial accuracy in multi-phase materials is far from trivial.

While asphalt has a rather weak reputation in the materials science community at large as being too difficult to quantify its structure and therefore to establish general relations between its structure and its performance, this does not do justice to the great steps made to make asphalt and its many variants for road pavements self-healing and ready for a wider market introduction than the current trial motorway track experiments on some European motorways. The publication *1800536* addresses the range of approaches explored successfully.

Finally, the contribution *1800226* focusses on the healing of metals. Given the intrinsically low mobility of the metallic atoms at room temperature, self-healing of metals is restricted to small scale damage formed at relatively high homologous temperatures. However, both characteristics belong to the field of creep damage where the intended long service time yields an attractive time window for healing via atomic displacements from the metallic lattice to the cavities formed at the grain boundaries. While the concept of autonomous self-healing of creep damage has been demonstrated convincingly for Fe-X model alloys, the next step will be to combine self-healing design strategies with conventional design strategies for optimal mechanical properties at high temperatures.

Together the 10 publications in this special edition offer an unprecedented insight into the latest developments in the field and clearly point at the next steps to be taken to bring the field from its current still conceptual explorative stage to the stage of real industrial applications with all the predictive performance models this requires. We are not yet there, but expect to be there in the next five years.

During the final processing of this special issue on Self-Healing Materials we received the terrible news that on May 28th 2018 professor Scott White died of cancer at the young age of only 55 years. Scott White was one of the, if not the, key leaders in the field of self healing materials. His vision and drive not only led to ground-breaking concepts from his own team at Champaign-Urbana, but also inspired many research teams all over the world to enter the field. He coupled a very sharp scientific brain with a fine sense of humour and a generous nature. We extend our sincere condolences to his wife Nancy Sottos, his family and all those who were privileged to work with him closely.

Nele De Belie, Sybrand van der Zwaag and Sander Leeuwenburgh

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