

Self-Healing Structural Polymer Composites Incorporating Bioinspired Nanofillers

A. N. Shankar¹, Shrujal Barvaliya^{2*}, Prasadaraju Kantheti³, Tasneem K. H. Khan⁴, Santosh Yerasuri⁵, P. K. Hemalatha⁶

Abstract

Self-healing polymer composites become an attractive family of intelligent materials that are capable of autonomously repairing damage, which will enhance their durability, reliability and service life in extreme engineering applications. The materials are based on the principles of nature, using the nanofillers that are derived from biological systems to improve mechanical properties and self-healing capabilities by utilizing hierarchical structures and multifunctional interface interactions. The recent developments on the formulation of self-healing polymer composite with nanocellulose, graphene and graphene oxide, MXenes, engineered nanoclays, chitin nanofibers, catechol-functionalized nanoparticles, and other bioinspired nanofillers obtained from sustainable sources are reviewed. The intrinsic, extrinsic and vascular self-healing systems have been explored with a detailed discussion on the effect of these nanomaterials on crack arrest, stress transfer, interfaces adhesion, reversible covalent and supramolecular bonding, healing kinetics and mechanical reinforcement. In addition, the review compares different manufacturing methods, such as solution casting, melt blending, additive manufacturing, electrospinning, and in situ polymerization, and also discusses the impact of the manufacturing process on the dispersion of nanofiller, composite structure and healing properties. Recent advances are also explored in mechanical characterization, fracture toughness, fatigue resistance and thermal stability, electrical conductivity, and multifunction sensing capabilities to illustrate the wide range of possibilities of these advanced composites. The review also covers major issues associated with large scale commercialization such as nanofiller agglomeration, limited healing cycles, scalability, cost, recyclability, environmental stability for extended period, manufacturing complexity, etc. The next generation of autonomous structural materials is highlighted by emerging research trends of vitrimer chemistry, dynamic covalent networks, bio-derived sustainable polymers, machine learning-assisted materials design, digital manufacturing and artificial intelligence-driven optimization. In conclusion, the bioinspired nanofillers are a highly promising approach for the development of strong, multifunctional and sustainable self-healing polymer composites for application in various sectors including aerospace, automotive, civil engineering, marine, biomedical, and energy industries, and also the future research directions required for successful industrial scale implementation of the materials.

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INTRODUCTION

Structural composites with the ability to heal themselves automatically are a paradigm change in materials science, with the potential to make components more durable and reliable by stopping damage from occurring to the point of catastrophic failure [1]. These materials replicate the natural biological systems in that they repair themselves, without the intervention of humans, and they are damage tolerant and resilient [2]. In this dynamic environment, introducing bioinspired nanofillers into monolithic polymer matrices holds promise as a novel approach to address the mechanical and durability constraints associated with traditional polymer matrices [3]. Synergistic nanofillers serve as both mechanical support and initiation points for the stimuli-responsiveness needed for fast and repeatable healing, filling the gap between extrinsic healing agents and structural performance [4, 5]. This strategy tackles the classical dilemma of mechanical reinforcement versus material mobility and allows for designing systems that are damage-tolerant and have high structural integrity, while also enabling efficient healing kinetics [6].

BACKGROUND AND MOTIVATION

The need for such tough and sustainable materials comes from the fact that the growth of micro-cracks in them is one of the most severe problems in the mechanical integrity of a structure the existence of which may only be guessed when the mechanical integrity of a structure is fundamentally shaken [7]. Various multi-scale damage modes are especially prone to occur in conventional FRP composites, requiring costly manual handling that may not be economically or environmentally sustainable [8]. Moreover, these composites suffer from impact degradation, which can be severe, especially with stiff fibre architectures, and makes them unsuitable for maintenance-free applications such as safety-critical applications [9]. To overcome these shortcomings, researchers have focused more on the self-repairing ability of the material, as well as on the introduction of dynamic cross-linked networks and bioinspired architectures, which enable the material to repair itself with superior mechanical properties needed for structural applications [10, 11].

Scope and Objectives

- Analyses the role and mechanism of bioinspired nanofillers in the autonomous self-healing property of polymer composites with high performance [12].
- Investigates the role of nanoscale reinforcement in dynamic interface bonding, and its ability to enhance crack repair while maintaining structural stiffness and mobility for healing [13, 14].
- Discusses development of bioinspired hybrid architectures from basic design principles to advanced structural composite applications for fracture toughness, mechanical performance and environmental stability [15, 16].
- Categorizes self-healing systems into groups according to the chemistry they employ: supramolecular interactions, reversible covalent chemistry and dynamic covalent chemistry. Assesses the value of bioinspired nanofillers for enhancing the structural integrity, damage tolerance, repeatable healing capability, and multifunctional properties of advanced polymer nanocomposites. Emphasizes the ecological and industrial value of autonomous, adaptive, and sustainable self-healing structural materials and their promise in next-generation engineering applications [17, 18].

Bioinspired Nanofiller Architectures

The classic stiffness-compliance dilemma is often solved in nature using hierarchically graded, functionally adapted composite architecture made of soft, energy-dissipating matrices and stiff structural parts [19]. For example, nacreous structures are optimally designed using brick-and-mortar structures to achieve maximum fracture toughness and, therefore, offer a template for designing synthetic structures using nanofiller-reinforced composites that emulate such load-bearing hierarchies [20]. In the same vein, complex vascular systems that enable the localized delivery of healing agents to damage sites are inspired in the creation of a synthetic vascular system that is made up of nanofiller channels that allow healing agents to be continuously delivered to damage sites [21]. Biological

strategies are mainly based on active metabolic pathways with centralized locations to repair, while synthetic hybridization is based on embedding nanofillers with dynamic interfacial interactions that can be repaired autonomously without external energy [22]. The main bioinspired nanofillers, their properties, healing mechanisms, advantages, and applications are summarized in Table 1.

Table 1. Comparison of bioinspired nanofillers for self-healing structural polymer composites.

Bioinspired nanofiller	Key characteristics	Self-healing mechanism supported	Mechanical advantages
<i>Nanocellulose</i>	Renewable, high aspect ratio	Hydrogen bonding, intrinsic healing	High toughness, crack deflection.
<i>Graphene/Graphene Oxide</i>	Excellent electrical & thermal conductivity	Thermal-triggered healing, sensing	High stiffness, conductivity.
<i>MXenes</i>	2D conductive nanomaterials	Electrothermal healing	Self-sensing, EMI shielding.
<i>Cellulose Nanocrystals</i>	Hierarchical bioinspired structure	Dynamic interfaces	Reinforcement, flexibility.
<i>Engineered Clays</i>	Layered morphology	Barrier effect, reversible bonding	Improved modulus.
<i>Carbon Nanotubes</i>	High aspect ratio	Joule heating-assisted healing	Fatigue resistance.

Source: Compiled from literature [9, 12, 15].

Natural Hierarchical Templates

Precisely ordered architectures at multiple scales are used by organisms – like nacre, bone, and wood – to dissipate energy and prevent crack propagation, enabling them to operate in lightweight frameworks that are robust to defects [23, 24]. The architectures are built around the complementary properties of inorganics with high strength and flexibility, with bio-polymer matrices with high toughness and modularity that are essential for survival [25]. These biological blueprints are now used to synthesize and mimic the staggered, interlocking topologies that are effective at stopping brittle fracture propagation and deflecting stress [26]. Nanocellulose is an example of such resilience because it has a naturally ordered Bou ligand structure that encourages uniaxial orientation, which effectively guides the paths of cracks as well as improves impact resistance [27]. In addition to these static geometries, there is recent work on the dynamic mechanical properties that organisms can change, such as sea cucumbers, which will change the stiffness of their internal structures through manipulation of stress transfer between their collagenous fibre networks [28].

The Biomimetic Role of the Bioinspired Nanofillers

Either bioinspired nanofillers for multifunctional self-healing through structural mimicry, reversible interfaces, and localized repairport, or catalytic in nature, which enable the restoration of mechanical integrity and increase the durability of the synthetic composite [29]. These enhancements bring together advanced reinforcement technologies, with the goal of breaking the inherent trade-off between mechanical performance and self-healing kinetics to enable the repaired structure to retain its performance under service conditions [30]. Future progress in this area needs to focus on optimizing the interphase between the matrix and the nanofiller since the interphase is the most critical weak link at which crack initiation and propagation usually occur [31]. In addition, the long-term reliability of these systems requires moving beyond passive reinforcement strategies and towards dynamic (embodied intelligence) designs that can detect damage and precisely control repairs in an autonomous manner [32]. In this direction, the considerable difficulties in scaling these intelligent architectures are huge, since the attempts to reproduce the synergic, multi-mechanism, repair processes that occur in biological systems in the laboratory have still not been successful [33].

Overcoming these manufacturing constraints is crucial for realizing the potential of composites in real-world applications where they are subjected to challenging environmental conditions, such as UV exposure and thermal cycling [34]. To apply these systems to a practical use, however, advanced

manufacturing process techniques are necessary to achieve complex hierarchical structures that will enable targeted repair chemistries [35]. Furthermore, the use of stimuli-responsive components, such as magnetic or electric field responsive components, opens opportunities to control these repair processes as needed, rather than relying on passive diffusion-based healing, which is constrained by their limited ability to generate new tissue [36, 37]. The principle is illustrated by examples of biological growth, such as biologically-grown composites, in which hierarchical networks of mycelial hyphae form the bonds between the lignocellulosic substrate network [38]. The structural arrangement of hybrid natural-fibre/metal laminates is also shown to be significant, with studies revealing that reinforcement architecture and stacking sequence are significant factors in the tensile, compressive, impact and interlaminar properties, indicating that arrangement is as crucial as component selection for the creation of robust, bioinspired composites [39].

Synthetic Biomimetic Strategies

Here, they have used engineered clays and cellulose nanocrystals in place of fillers to mimic the natural binding interfaces of mineralized tissues and have surface-functionalized them to mimic the binding mechanisms [40, 41]. Such material modifications are often made synthetically; and include supramolecular grafting or covalent grafting, which mimic the sacrificial bonding motifs found in nacre to allow for reversible interfacial slip, leading to energy dissipation [42]. The functionalized nanomaterials form a network of dynamic cross-links, exert structural rigidity while retaining the ability to reconfigure themselves quickly and arrest crack propagation [43, 44]. These bio-mimetic reinforcements, therefore, successfully move synthetic matrices from an “unbroken” to a “self-healing” state, restoring mechanical integrity upon damage, and make them into self-adaptive networks [45]. Surface treatment, Interfacial compatibility. Since this work is of a review nature, there was no single surface treatment/coupling agent used by the authors. Rather, comparatively reported interfacial-modification strategies for the bioinspired nanofillers are considered.

Surface treatment is of special interest because it is possible that the untreated nanofillers do not have good wettability properties, have incompatible surface energies or strong filler–filler interactions that lead to aggregation. Reported methods are: silane coupling, alkali treatment of lignocellulosic reinforcement, oxidation or introduction of oxygen containing functional groups, polymer grafting, surfactant assisted dispersion, and catechol/dopamine inspired functionalization. They can add functional groups, such as hydroxyl, amine, epoxy, and other groups, which can enhance the adhesion between the layers by covalent bonding, hydrogen bonding, mechanical interlocking, or supramolecular interactions. In self-healing systems, however, it is desirable to avoid interfaces that are too strong and/or irreversible that inhibit chain mobility or dynamic bond exchange. Therefore, good surface modification must be adequate to ensure good filler–matrix compatibility and must also ensure reversible interactions at the interfaces for crack closure and multiple healings [46–49].

Self-Healing Mechanisms

Self-healing composites can be developed through three main concepts: intrinsic self-healing, where healing is achieved through molecular interactions that can be reversible within the polymer matrix; extrinsic self-healing, where healing agents are encapsulated and are released only after they are broken; and vascular networks, which allow for the ability of multiple sites to be damaged for healing [50, 51]. Combining these healing mechanisms with bioinspired nanofillers demands a compromise between the limited molecular mobility of hard minerals, and the effective delivery of healing agents, which necessitates transport of mass. The inclusion of these healing strategies into bioinspired nanofillers demands compromise: while the hard mineral assemblies have restricted molecular mobility, they require efficient mass transport of healing agents [52]. Intrinsic and vascular methods take advantage of the dynamic nature of the interfaces between the nanofiller and rubber to allow for localized movement of the rubber molecules; however; some extrinsic methods may also have difficulties integrating because the nanofiller concentration can be high, resulting in tortuous pathways to allow the diffusion of the monomeric precursors [53, 54]. Table 2 compares the major self-healing strategies used

in structural polymer composites, including intrinsic, microcapsule-based, vascular; and hybrid systems. It summarizes their healing mechanisms, key advantages, and limitations. Table 2 provides a quick overview of their suitability for advanced structural applications.

Table 2. Comparison of self-healing strategies in structural polymer composites.

Self-healing strategy	Healing principle	Advantages	Limitations
Intrinsic Healing	Dynamic covalent/supramolecular bonds	Multiple healing cycles	Lower stiffness in some systems.
Microcapsule-Based	Rupture of capsules releases healing agent	Autonomous repair	One-time healing.
Vascular Networks	Continuous delivery through channels	Repeatable healing	Complex fabrication.
Hybrid Systems	Combination of intrinsic and extrinsic	High healing efficiency	Increased processing complexity.

Source: Compiled from literature [55, 56].

Intrinsic Healing Systems

Intrinsic healing systems utilize dynamic chemistries that respond to stimuli, such as reversible covalent chemistry or supramolecular interactions, and will heal without the need for any healing agents that must be stored within the material [57]. The materials are designed to form Diels-Alder linkages and/or supramolecular networks through hydrogen bonding, which can be reversed by applying a certain stimulus, allowing polymer chains to be mobile. These systems use high-aspect-ratio nanofillers to reinforce the structure and assist chain diffusion and reconnection through sacrificial bond breakage and reformation [58, 59]. In particular, hydrogen bonding is often used in such matrices because it has a very strong directional and adaptable strength, and the ability to heal the material quickly under various conditions without requiring complex external stimuli [60]. Self-healing polymer composites can heal damage in either a reversible bond reformation or by releasing an embedded healing agent, depending on the healing strategy. Intrinsic healing allows the cracks to be repaired repeatedly by dynamic bonding; and extrinsic healing restores material properties by sealing the cracks with polymerized healing agents [61] as illustrated in Figure 1.

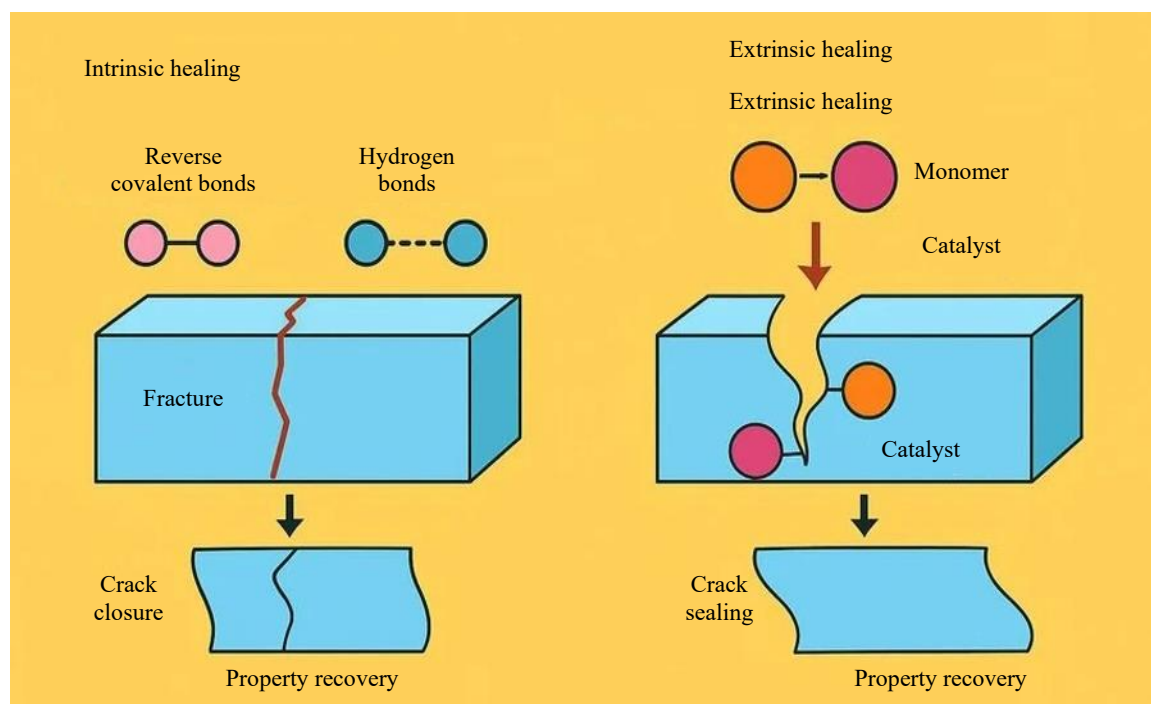


Figure 1. Comparison of intrinsic and extrinsic self-healing mechanisms in polymer composites, illustrating reversible bond-mediated healing and microcapsule-based crack repair for restoring the mechanical integrity of damaged materials [62]. Licensed under CC BY 4.0.

Microcapsule-Based Approaches

In extrinsic systems, monomers or catalysts for polymerization are encapsulated in a matrix and only released during mechanical damage for the autonomic in situ polymerization [63]. The effectiveness of these systems largely relies on the choice of shell chemistry meeting the criteria of long-term environmental stability and a carefully designed threshold for triggering rupture when cracks are propagated [64]. For the microcapsules to interact effectively with propagating cracks, they need to be accurately localized in the fibre/matrix interphase, while their mere presence directly along the crack line would only release the healing agent in that specific location [65, 66]. The bioinspired nanofillers, however, add to the high local stiffness and density that can hinder mechanical rupture of these shells, thus requiring careful engineering of the interfaces to ensure high healing efficiencies [67, 68].

Vascular Network Designs

Unlike microcapsules, which are only applicable for one event repair, these systems have the advantage of using interconnected microchannel architectures for delivering healing agents to the damage sites and are applicable to repeated repair cycles [55]. They can be classified into 1D, 2D or 3D architectures where 3D interconnected channels offer better fluid distribution than 1D bioinspired vascular conduits. To achieve the optimal vascular integration, the diameter of the channels and the volume fraction of nanofillers need to be optimized to prevent degradation of the structure yet provide enough hydraulic conductivity to transport fluids [69]. The use of biomimetic design principles has increased, and more researchers are now using 3D-printed vascular geometries that navigate around filler-rich areas, allowing the self-healing agent to reach regions of high structural stress [70]. Further, the change from 1D to 3D vascular architectures improves the volume coverage of the healing agent and thus provides a way to circumvent the issues of localized repair, which is observed with single-channel pathways [71, 72]. This enhancement of the network dimensionality enables excellent in-situ fluid mixing and allows the entire mode-I fracture resistance to be recovered by a replenishable, cycle-independent, repair mechanism [73].

Composite Fabrication Techniques

The main difficulty to overcome in the manufacturing of such advanced materials is the control of both the concentration and spatial distribution of nanofillers, as their excessive loading can lead to the agglomeration of nanofillers, to increase the viscosity of the matrix, to create local stress concentration, and can decrease the mobility needed for the efficient self-healing [74, 75, 76]. A uniformly dispersed and uniform filler architecture is crucial for minimising localized defects that may be created during the process, which can be obtained by techniques like liquid resin infusion and ultrasonic dispersion [77]. Additive manufacturing enables the creation of complex, bioinspired vascular geometries that are difficult to achieve using sacrificial templating or soft lithography. To satisfy the need for scalability in industry, current research is focusing on modular additive manufacturing processes that can integrate interconnected networks into Fibre-Reinforced polymer laminates without any detrimental impact on the structure's performance [78]. There are two ways to disperse and mix. There are two methods of dispersion and mixing: High shear sonication is often used to disperse clusters of nanofillers and assure that the nanofillers are evenly dispersed in the monomer precursor prior to the start of in-situ polymerization of the monomers. High-shear sonication is often used in combination with surfactants or coupling agents or surface functionalization of nanofillers to prevent re-agglomeration due to van der Waals interactions and to improve the filler-matrix compatibility during curing. The in-situ polymerization approach inside the scaffold structure ensures accurate immobilization of these nanofillers, enabling more precise localization in the desired directions at specific stress points [79]. This homogeneity can also be further enhanced during processing by using centrifugal mixing to ensure that the fillers do not settle in the initial stages of network curing, as the viscosity rises [80].

PROCESSING AND CONSOLIDATION

Two fundamental techniques used to pre-process thermoplastic matrices for controlled incorporation of nanofillers are solvent casting and melt extrusion, enabling the retention of both the rheological properties needed for high-resolution additive manufacturing and the nanofillers. The two basic

approaches that can be used to pre-process thermoplastic matrices for controlled incorporation of nanofillers are solvent casting and melt extrusion, both of which can preserve the needed rheological properties for high-resolution additive manufacturing and nanofillers. Yet, the solvent-free approach is still of paramount importance in the scaling process, as it can affect the final quality of the print and its mechanical properties [81]. Manufacturers can optimize the thermal history of melt extrusion and remove the aggregation that is common with high-loading composites [56, 82] by making the interface between the filler and matrix in the composites smooth. For AM, it is crucial to control the cure schedule precisely, which ensures a rate of polymer matrix vitrification that is favorable for the development of a stable interface between the polymer matrix and the filler, as this phenomenon reduces [83, 84].

Analysis of Mechanical & Functional Performances

Standardized protocols use the tapered double cantilever beam test to measure the recovery of mode-I fracture toughness after thermal/chemical stimuli to validate structural recovery [85]. The digital image correlation method is used to map the strain distribution to investigate the load-bearing capacity restoration efficiency of the damaged interfaces by these mechanical tests [86]. To evaluate the effectiveness of these systems, the balance between the reinforcing benefits of the nanofillers and the decrease in healing agent mobility at high filler loads needs to be assessed, as high concentration of nanofillers can lead to high viscosity of the matrix which can hinder the ability of the matrix to fill the cracks during healing [87].

Healing Efficiency Characterization

Typical metrics used for determining recovery are the ratio of impact energy measured in tension or failure number per cycle before and after successive cycles of damage-healing [88]. These metrics tend to focus on healing efficiency and cyclic durability, indicating that achieving high-aspect-ratio nanofiller can considerably increase impact resistance, yet it's essential to precisely optimize their properties so as not to hinder the movement of healing agents [89]. Specifically, it was shown that while static modulus and toughness both increase with higher concentrations of nanofiller, shearing high concentrations can result in the formation of percolating networks, which can impede the ability of the chains to move in order to undergo dynamic bond exchange and interface re-adhesion [90]. Furthermore, rational design of the filler morphology (from spherical to shapes with high aspect ratio) plays a key role in achieving a good balance between reinforcement of the structural matrix and diffusion of the agent [91, 92]. Advanced material designs that adopt multivalent polymer-grafted nanoparticles can overcome these limitations, achieving much better improvements in bulk modulus without compromising the chain motion at the segmental level required for efficient crack healing [93].

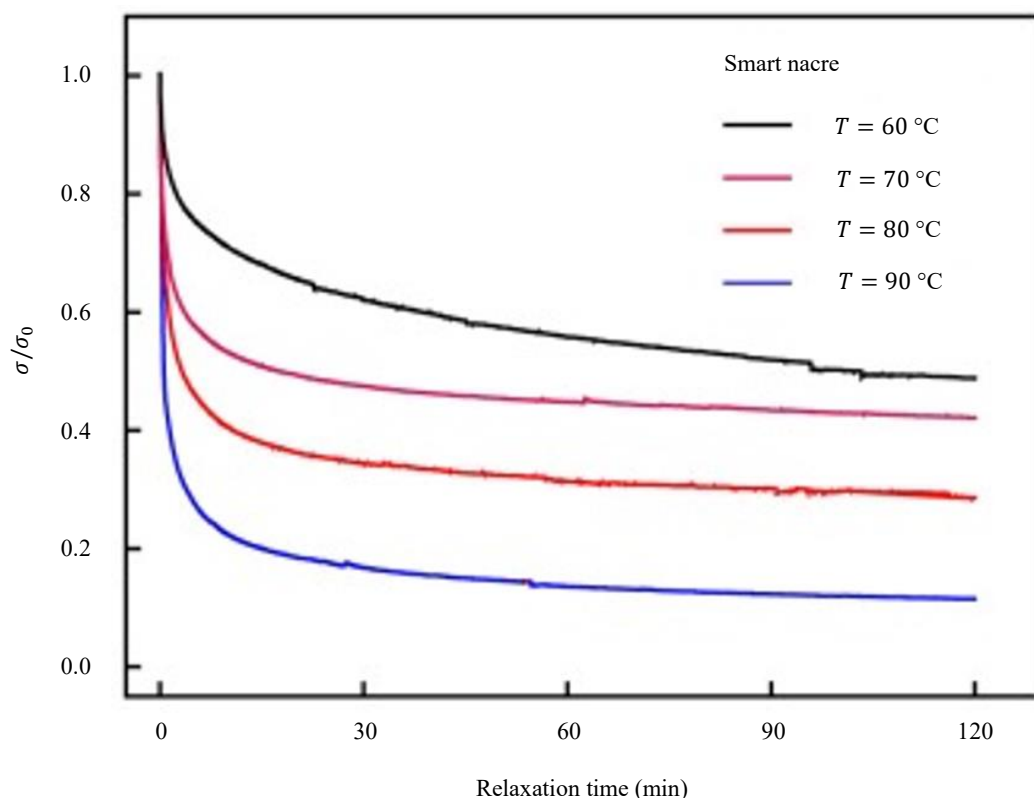
The effect of Nanofiller loading and optimum concentration. One of the most important design parameters is the concentration of the nanofillers because reinforcement and self-healing are not always proportional to the concentration of fillers. Well dispersed nanofillers can enhance stress transfer, crack deflection, interfacial adhesion, thermal/electrical transport, and the number of reversible interactions for healing at relatively low to moderate loading. However, above the optimum concentration, nanoparticle aggregation, increasing resin viscosity, poor matrix wetting, lack of polymer-chain mobility, and diffusion restriction of the healing-agent can lead to a decrease in mechanical properties and healing efficiency. The optimum concentration, therefore, can vary depending on the chemistry of the nanofiller, aspect ratio, surface functionality, polymer matrix, dispersion method and healing mechanism for all self-healing nanocomposites. This concentration dependent behavior has been observed from the literature on polymer composites. For instance, optimum mechanical performance was obtained with the composites made from nano-Aristida hystrix loaded with 5 wt% of the nanofiller, while 7 and 9 wt% of nano-Aristida hystrix resulted in a decrease in the mechanical properties due to the agglomeration of the nanofiller and the limited wetting of the polymer. In the same way, the mechanical properties of Calamus tenuis/epoxy composites were found to be maximum at about 10 wt% fibre loading. Indeed, such results argue for the need to find an optimum value for the application instead of expecting that much higher fill levels will always lead to better performance [94, 95].

Multi-Cycle Damage Recovery

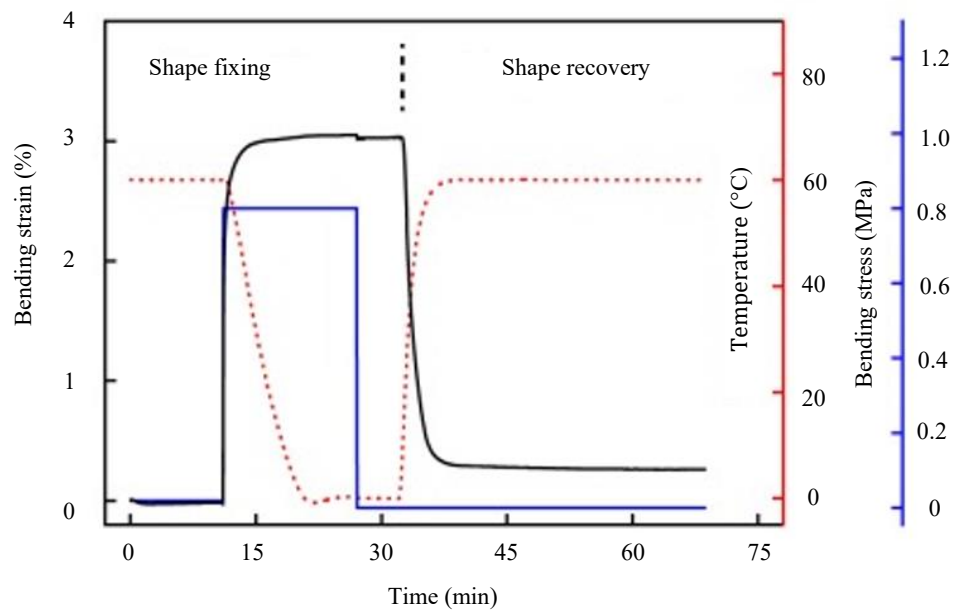
These systems have fatigue limits and can be tested by subjecting composites to alternate loads how long the restorative response lasts depends on the stiffening or failure of the structure and the depletion of the agents. It is important to perform systematic investigations into the coupling of degradation of structural integrity during autonomous cycles' downtime between cycles, in order to evaluate the degradation of structural integrity during the intervals between the cycles, since the assessment of the degradation of structural integrity between autonomous cycles is often neglected [96]. Healing efficiency has been found to vary on repetitive cycles of break/heal in long-term durability testing, and the decrease in healing efficiency is generally attributed to the depletion of the local healing reservoirs and formation of unhealed interfacial micro-voids [97]. In addition, the shift from static fracture mechanics to cyclic fatigue characterization is important since monotonic tests do not reflect the complex relationship between crack closure and loading frequency and stress intensity ratios [98, 99]. However, such restrictions require a more detailed examination of the damage-loading cycles, with the aim of maintaining the cumulative fatigue life without being affected by the intrinsic dualism between the rigidity of the structure and the reversible and dynamic nature of the bonding network [100].

Multifunctional Property Integration

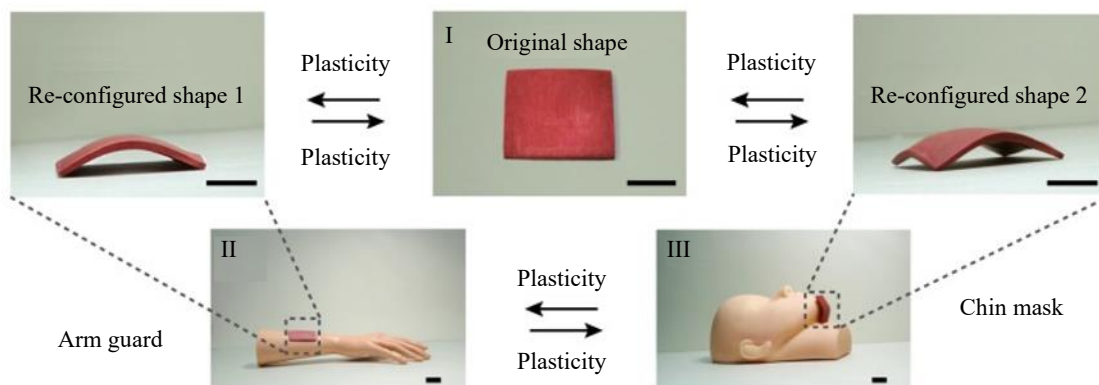
To realize multifunctionality, mechanical self-healing should be synergically coupled with electrical, thermal, or sensing functions, which can be realized by forming percolating networks in the polymer matrix which are dynamic [101]. The conductive pathways enable auto-diagnosis of damage by sensing the mechanical deformations and transducing them into measurable changes in electrical impedance, for real-time monitoring of the health condition of the structure [102]. At the same time, coupling thermally conductive nanofillers allows for the generation of localized, remotely triggered heating to accelerate bond reorganization and recovery of structural properties in environments where the passive autonomic recovery is kinetics-limited [103]. As shown in Figure 2, the nacre-inspired self-healing composite exhibits excellent shape-programming and shape-recovery capabilities, demonstrating its potential for adaptive structural applications [104].



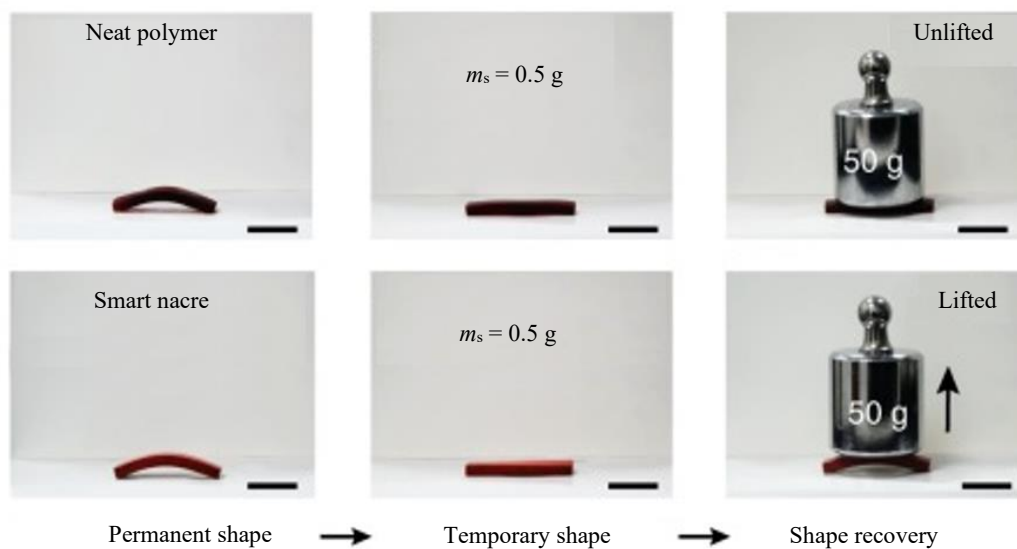
(a)



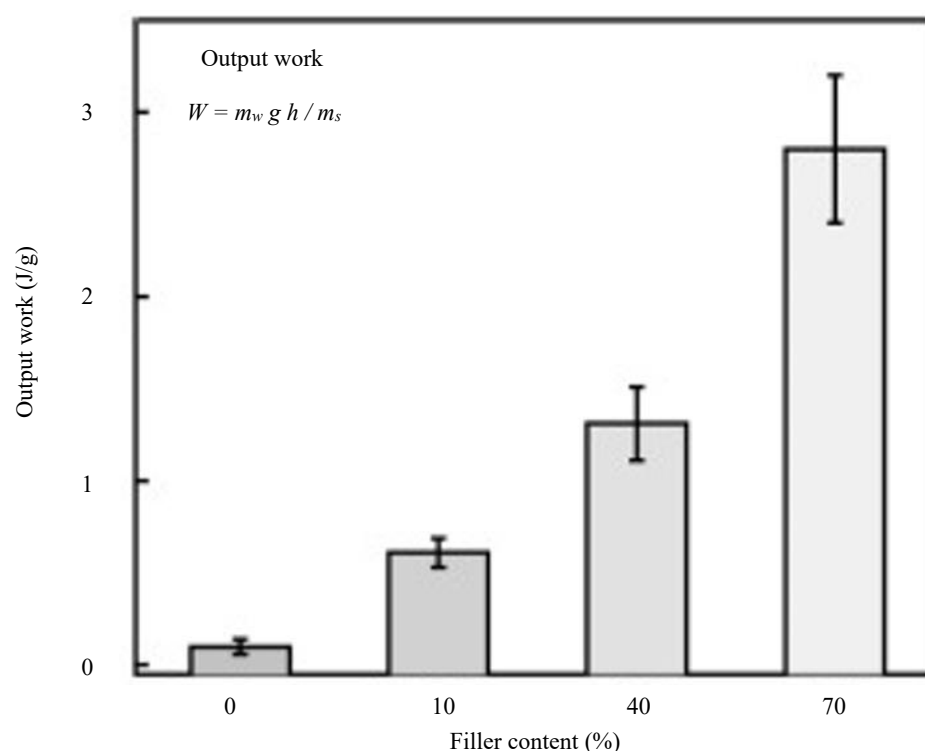
(b)



(c)



(d)



(e)

Figure 2. Multifunctional performance of a nacre-inspired hierarchical composite illustrating stress relaxation, shape-memory behavior, plastic reconfiguration, and enhanced mechanical properties enabled by its bioinspired brick-and-mortar architecture [20]. Licensed under CC BY 4.0.

Electrical and Thermal Properties of Materials

The electrical percolation threshold is reached by the establishment of conductive pathways between nanofillers, with the use of high-aspect-ratio fillers to allow the presence of low filler loading conductive pathways without compromising the structural toughness of the matrix [105, 106]. Likewise, hierarchical networks of fillers substantially enhance thermal anisotropy, facilitating efficient heat dissipation and thermally induced filler healing [62, 107]. The composite matrix can be designed to obtain high-performance mechanical properties and retain mobility for dynamic bond reorganization by using these network architectures [108].

Deliverable's Sensing and Monitoring Capabilities

By incorporating piezoresistive nanofillers, micro-cracks or cracks in the structure can be precisely located using real-time fluctuations in electrical resistance [109]; thereby different through-thickness resistances allow one to determine the degree and direction of damage caused by fatigue, such as delamination growth, in the vicinity of stress concentrations [110]. Also, the seamless integration of these functions of self-reports forms closed-loop system of sensing signals triggering thermal activation of autonomous healing mechanisms [111, 112]. These dual-functional composites can be used to reduce the likelihood of overheating by controlling the energy delivery during the recovery process with the use of magnetic particles exhibiting different Curie temperatures, for example [113].

Applications and Future Perspectives of the New Technologies

Self-healing polymers are useful for high-value structural applications, such as wind energy infrastructure and aerospace, where they can suppress catastrophic fatigue failure by self-healing crack arrest at critical interfaces [114, 115]. In space exploration, where repairs in inaccessible environments are vital for mission critical success, these materials are particularly important and can repair structural damage automatically [116]. Likewise, such composites can be used in this deep-sea infrastructure

applications to minimize the need for complicated and redundant back-up systems in high-risk areas [117]. To move these laboratory findings towards industrial deployment; the focus of the research roadmap needs to shift to scaling up the manufacturing process, for example, by adopting additive manufacturing or resin transfer moulding, that will promote uniform distribution of the filler without creating stress concentrating agglomerates [118].

STRUCTURAL ENGINEERING APPLICATIONS

In civil infrastructure and automotive applications, use of these materials in load-bearing components offers an enabling way to achieve damage tolerance and helps minimize the high maintenance costs of manual inspections and service-life replacement, especially for these applications [119]. With the transition from the traditional maintenance philosophy of maintenance schedules and intervals to the new one of an autonomous, proactive healing system, the stakeholders can obtain a significant reduction in life-cycle cost in fatigue-prone assets [120]. Taking the next step from traditional non-destructive evaluation methods to integrated sensor-based health monitoring that can quantify the true condition of structural restoration is an important economic consideration for these deployments [121–123].

EMERGING RESEARCH DIRECTIONS

The use of bio-derived and sustainable nanofillers and dynamic matrices is becoming a key focus of future developments, while maintaining the intrinsic healing kinetics of the polymer network, which is in line with the objectives of the circular economy [124, 125]. broader roadmap is needed to overcome these challenges, which involves the standardization of long-term reliability tests for these multi-component, complex vitrimeric systems and the development of greener chemical pathways for their recycling at the end-of-life [126, 127]. Current research is investigating computer modelling and computational methods to speed up the discovery and optimization of these vitrimer systems to make them more attractive to industry where the empirical cycle times are currently limiting [128]. Furthermore, the need to develop dynamic covalent chemistry that are compatible with existing thermoset & thermoplastic industrial manufacturing is one of the most important challenges for successful industrial applications [129]. Moreover, there are great challenges in retaining mechanical, stiffness and toughness properties after recycling multiple times, which depends on mechanical depolymerization processes that can degrade the interfacial fibre-matrix bond [130, 131]. Researchers are looking into ways to keep this structural integrity intact through multiple reprocessing cycles by using associative dynamic covalent bonding such as transesterification [132]. Lifecycle assessments and techno-economic analyses need to be incorporated early in the development process to account for the net reductions in energy consumption and waste produced in both production and recycling life cycle stages [133, 134].

CONCLUSION

In this work, the embedding of nanoscale fillers into the polymer matrix to create autonomous repair mechanisms, like those found in natural structural materials, will be reviewed. This study explores the synergistic effect of bioinspired fillers and polymer network toward the mechanical reinforcement and reconstruction of structural integrity by synthesizing the latest advancements in materials science. Moreover, such strategy can overcome the major drawbacks of traditional polymers – low specific surface area/volume ratio of nanomaterials to optimize interfacial load transfer and multifunctional performance. Such nanocomposites can be used to design smart, mechano-responsive systems that sense damage and trigger specific repair cycles. The current studies indicated that future research would focus on manufacturing at a larger scale and on the standardization of performance parameters to ensure the reliability of materials in future applications in the aerospace and automotive industries. Additionally, there is a need for closer integration of the multiscale modelling techniques to predict the behavior of materials subjected to complex service conditions to bridge the gap between fundamental mechanism development and deployment at the industrial scale. In conclusion, the standardization of characterization procedures and the development of quality life cycle assessments will be crucial for transposing the laboratory achievements into possible and sustainable solution for infrastructure and high-performance operating systems.

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