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ARTICLE

Recent Advances in Self-Healing Green Composites for Structural Applications

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ABSTRACT

The development of self-healing green composites offers a sustainable pathway to extend the lifespan of structural materials while reducing environmental impact. This review takes a look at developments in the three principal techniques of self-healing considered namely use of bio-based healing agents, use of microcapsule systems, and also use of vascular networks with particular reference to application in structural engineering set ups. Renewable-based repair materials in the form of bio-based healing agents offer a green-friendly repair agent but limit rebound in the speed of reaction and in stability. Microcapsule systems provide self-contained, readily incorporated repair abilities, but are usually restricted in being used once only. Biologically inspired, vascular networks allow repeatable and even sustained repair, but their manufacturing is more complex requiring careful optimization of the overall network. Each strategy is discussed in terms of material composition, healing mechanisms, advantages, limitations, and recent innovations. The review concludes with a comparative analysis and highlights future directions, including hybrid healing systems, nanomaterial enhancements, and integration with structural health monitoring. These advances underscore the potential of self-healing green composites to meet the dual demands of high performance and environmental responsibility in next-generation structural applications.

Keywords: Self-Healing Composites; Green Materials; Bio-Based Healing Agents; Microcapsule Systems; Vascular Networks

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1. Introduction

The global demand for materials that combine exceptional structural performance with environmental sustainability has intensified over the past two decades. This shift is fueled not only by stricter environmental regulations and the increasing cost of raw materials, but also by a growing awareness of the need to design engineering systems that minimize waste and environmental impact throughout their life cycles. Structural composites—materials that combine two or more distinct constituents to achieve superior mechanical properties—are now indispensable in sectors such as aerospace, automotive manufacturing, civil infrastructure, marine engineering, and renewable energy generation [1]. These are well suited to use in situations that require superior performance, durability and ability to corrode. Nonetheless, even though structural composites offer a number of benefits, they are not resistant to harm. A large number of causes of microcracks, delamination and fiber-matrix debonding include the manufacturing defects, fatigue caused by cyclic loading, impact damage and environmental degradation. Although such defects may start small enough to be imperceptible, they may grow with time to decrease the load-bearing performance, eventually causing a disastrous collapse unless fixed [2].

Composites that traditionally have been repaired by patching, adhesively bonding, or replacing components may be labor-intensive, time-consuming, and expensive. Location and operating conditions of the structure of the structure also adds to the complications of repairs in many cases. Consider the cost to access and maintain wind turbine damage that may occur at great height; or underwater structures that may be of great depth or under sea that require specialized gear in order to obtain information and rectify the source of failure. In aerospace, maintenance windows are firmly booked and unscheduled repairs can put entire fleets of operations into jeopardy. These vulnerabilities and other operational difficulties have spurred great interest in materials that sense and heal themselves, automatically, and so-called self-healing materials [3–5].

The idea of materials that can autonomously repair damage is inspired by biological systems, which have evolved efficient self-repair mechanisms to maintain function despite environmental stress. Human skin, for instance, rapidly heals cuts and abrasions through a cascade of biochemical processes, while plant stems can seal wounds to prevent infection and water loss. Translating these concepts to engineering materials involves embedding within the material an active mechanism—chemical, physical, or biological—that can respond to damage stimuli, deliver a healing agent to the damaged area, and restore lost mechanical properties [6].

Self-healing composites have been investigated extensively since the early 2000s, with significant advances in both healing mechanisms and material integration. More recently, research has shifted toward green self-healing composites—systems designed not only for damage tolerance and extended service life but also for environmental compatibility. These materials aim to reduce or eliminate the use of petroleum-derived polymers and synthetic healing agents, instead favoring renewable, biodegradable, or recyclable alternatives. This approach aligns with the principles of the circular economy, in which materials are designed to retain value across multiple lifecycles, minimizing waste and energy consumption from production to disposal [2,7,8].

Among the various approaches developed to achieve self-healing in composites, three strategies have emerged as particularly prominent for structural applications: bio-based healing agents, microcapsule-based systems, and vascular network systems. Each of these strategies employs a distinct mechanism of healing and presents its own set of benefits, trade-offs, and technical challenges.Bio-based healing agents use renewable material, often of plant, animal or microbial origin as an active medium by which to repair. Examples are plant oils like linseed or soybean oil that could form a polymer upon exposure to the air, chitosan-based hydrogel biomaterials that could crosslink in a humid environment, and enzyme-responsive biopolymers forming covalent networks at the sites of damage. Such materials are necessarily more sustainable than their competitors made of petroleum, and they may be used in conjunction with natural fiber reinforcements. They, however, tend to have a slower curing temperature and can be temperature or humidity sensitive and thus should be formulated carefully to give consistent performance [9,10].

Microcapsule systems are instead a physically dispersible system composed of swellable spherical shells that encapsulate a healing material, and are mixed into the composite matrix during manufacture. In the process, a crack growing through the material breaks the capsules that are immediately surrounding it, passing the healing agent out into the crack plane where it polymerizes and glues the fracture surfaces together. The simplicity of the strategy, its autonomous activation and compatibility with the existing composite manufacturing processes are appreciated. However, even with such systems, the limited supply of healing agent per capsule restricts them to a strictly single healing event per location and it is possible that incorporation of capsules will reduce mechanical strength of the composite [11–13].

Vascular network systems, inspired by the circulatory networks of living organisms, consist of interconnected hollow channels embedded in the composite. These channels serve as conduits for healing agents stored in external or internal reservoirs, enabling delivery of fresh material to damage sites on demand. This design allows for repeated healing cycles and, in some cases, integration with structural health monitoring systems for early damage detection. While vascular networks can provide superior durability, they require more complex manufacturing techniques and may introduce trade-offs in weight or structural performance. Selecting the appropriate healing strategy for a given structural application depends on multiple factors: the mechanical demands of the structure, environmental exposure conditions, desired service life, and the balance between sustainability, cost, and performance. For example, the repair needs of a solar panel support structure in a desert climate differ significantly from those of a marine composite hull subjected to saltwater and constant wave action. Similarly, aerospace applications require ultralight materials with minimal compromise in mechanical performance, while wind energy systems prioritize multi-decade durability with minimal maintenance [14,15].

This review aims to provide a detailed and comparative examination of recent advances in self-healing green composites for structural applications, focusing on bio-based healing agents, microcapsule-based systems, and vascular networks. By consolidating findings from the last decade of research, it seeks to clarify the relative advantages and limitations of each approach, examine their compatibility with sustainable material design, and evaluate their readiness for industrial deployment. The discussion will address healing efficiency, repeatability, mechanical property retention, processing considerations, and environmental impact. Ultimately, the goal is to guide material scientists, engineers, and industry stakeholders toward self-healing solutions that not only extend the lifespan of structural composites but also align with the urgent need for environmentally responsible engineering. The surge in research activity focused on self-healing green composites is evident from bibliometric data. As illustrated in **Figure 1**, the annual number of publications on this topic has increased markedly over the past decade, reflecting its expanding scientific and industrial relevance. Beyond structural applications, self-healing materials show significant promise in biomedical fields, including drug delivery, wound healing, and tissue regeneration, as summarized in **Figure 2**.

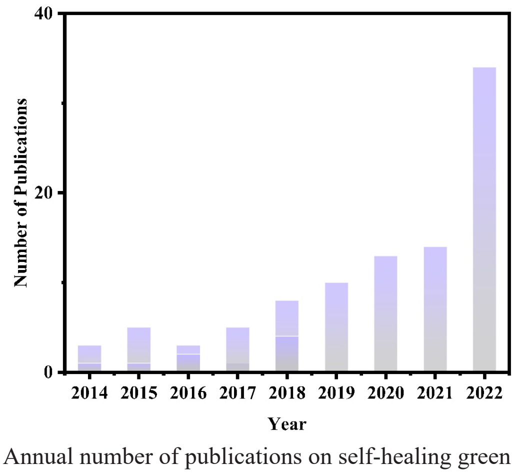

Figure 1. Annual number of publications on self-healing green composites.

Source: Scopus, 2016–2022.

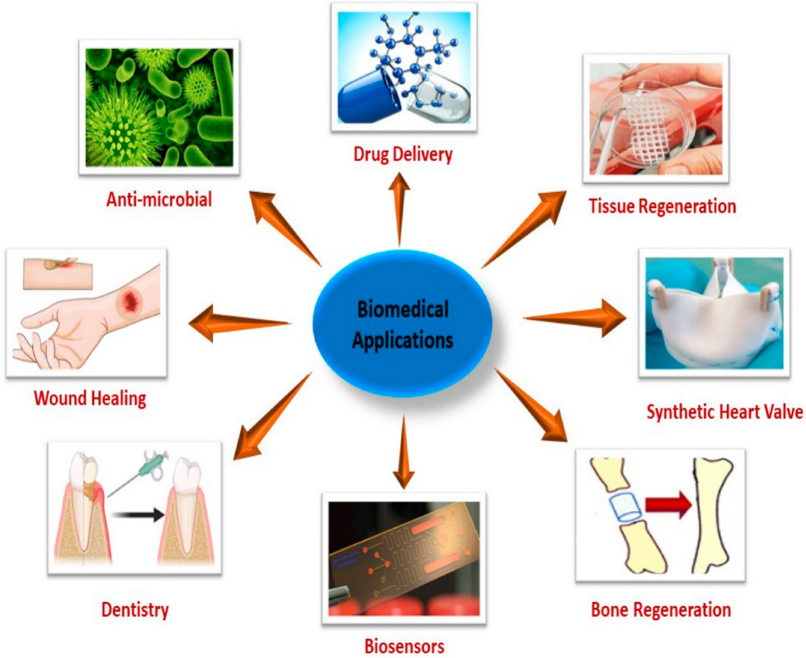

Figure 2. Biomedical applications of SHPs and SMPs.

2. Fundamentals of Self-Healing Green Composites

2.1. Concept and Sustainability Goals

Self-healing green composites represent a convergence of two important directions in materials science: the development of composites that can repair themselves after damage, and the movement toward environmentally sustainable materials. These composites are engineered to provide the strength, stiffness, and durability required for demanding structural applications while also offering the ability to autonomously restore mechanical integrity after damage has occurred. At the same time, their formulation and production are guided by principles of environmental responsibility—using renewable raw materials, minimizing energy consumption during manufacturing, and reducing ecological impact throughout the material's entire life cycle. The dual objective of performance and sustainability makes this class of materials uniquely challenging to design but also highly promising for industries seeking to extend product lifespans while reducing waste and reliance on petroleum-derived resources [2,16].

2.2. Self-Healing Mechanisms and Activation

The principle behind self-healing in such composites can be understood by considering how damage typically develops. In structural composites, damage often begins at the microscopic level—small cracks or voids forming in the polymer matrix or at the fiber–matrix interface. Over time, and under repeated loading, these microcracks can grow, coalesce, and eventually lead to a catastrophic loss of structural performance. In conventional composites, repairing such damage requires human intervention, which may involve detecting the flaw, accessing the damaged area, and applying external repair materials or adhesives. Self-healing systems circumvent these steps by embedding within the composite itself the means to respond to damage as it occurs. When a crack forms, it can trigger the release or activation of a healing agent, which flows into the damaged region, reacts or solidifies, and effectively seals the crack, restoring the material's structural integrity [2,17-19]. **Figure 3** illustrates the key chemical and physical cross-linking mechanisms in self-healing polymers, including dynamic covalent bonds (e.g., Diels-Alder reactions, imine bonds) and non-covalent interactions (e.g., hydrogen bonding, hydrophobic interactions), which enable reversible healing at the molecular level.

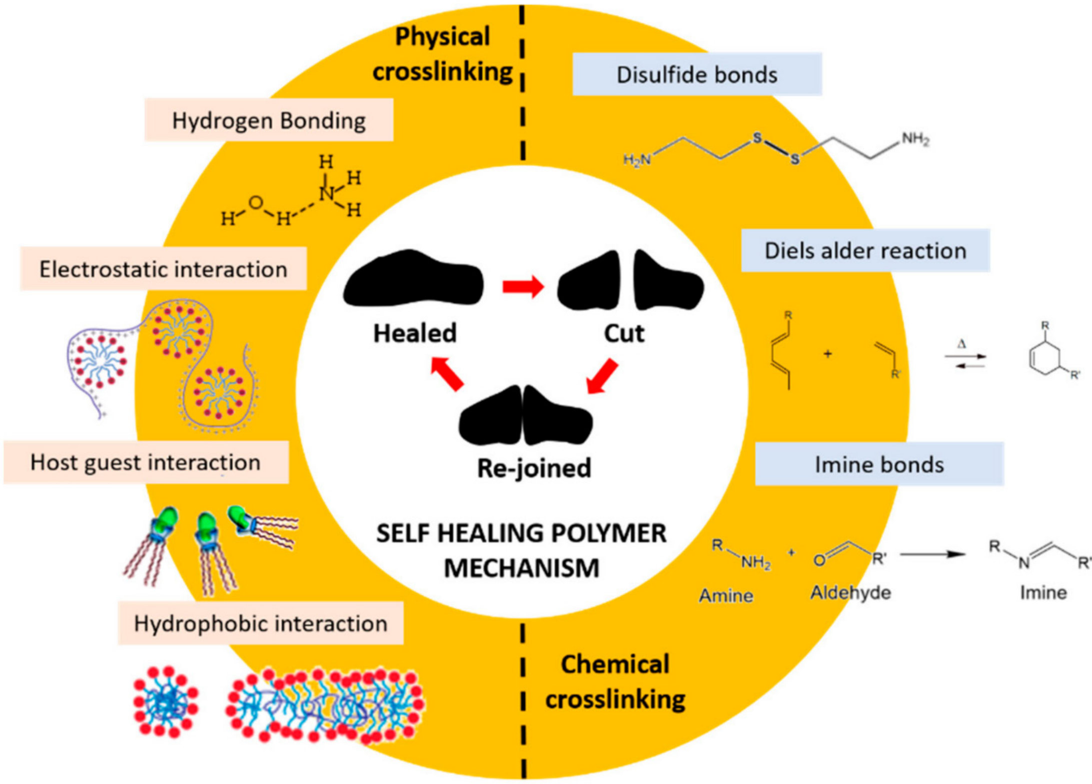

Figure 3. A summary of physical cross-linking and chemical cross-linking use in the self-healing polymer mechanism [19].

Two broad strategies are used to achieve this effect. In intrinsic self-healing systems, the polymer matrix itself is designed with reversible bonding or mobility at the molecular level. For example, some biopolymer networks can contain dynamic covalent bonds, hydrogen bonds, or other reversible linkages that can break and reform under mild conditions such as moderate heating or exposure to moisture. When damage occurs, these reversible bonds at the crack surface can reconnect, effectively "re-knitting" the polymer chains and closing the crack. Because the healing capability is inherent to the polymer, intrinsic systems can often be reactivated multiple times, making them particularly attractive for long-term use. In extrinsic systems, on the other hand, the healing function is provided by a separate component embedded in the composite. This might take the form of microcapsules filled with a liquid healing agent or a network of hollow channels that store and deliver a repair medium. In green composites, these embedded agents are frequently chosen from bio-based or biodegradable chemistries to maintain environmental compatibility [20–22].

The activation of the healing process in these composites can occur through different triggering mechanisms. The mechanical damage per se can act as the initiator, like when a microcapsule crack opens, or a vascular channel is ruptured. In more applications, an external trigger is required: thermostatic polymer networks require heat to move polymer chains in thermoplastic resins, photo-polymerising resins use light to set some bio-derived polymers, whereas hygroscopic biopolymers like chitosan or alginate are crosslinked and swollen in water. Coming up with green self-healing systems, one of the considerations addressed is to ensure that the triggers used are suitable to the expected service environment and do not demand very harsh or energy-intensive conditions so as to make the repair process both feasible and sustainable [23]. The key mechanisms, activation strategies, and outcomes of self-healing in green composites are summarized in **Table 1**.

Table 1. Self-healing mechanisms and activation strategies in green composites.

Category	Mechanism/Feature	Activation Trigger	Examples/Notes
Intrinsic Systems	Reversible bonds (dynamic covalent, H-bonds) in polymer matrix; molecular mobility.	Heat, moisture, or mechanical stress.	Biopolymer networks (e.g., chitosan, alginate); reusable; long-term sustainability.
Extrinsic Systems	Embedded healing agents (microcapsules, vascular networks).	Mechanical damage (crack ruptures capsules/channels).	Bio-based resins or biodegradable liquids (e.g., plant-derived monomers).
Triggers	- Mechanical damage - Heat - Light - Moisture	Crack propagation, external stimuli (mild heating, UV light, humidity).	Prioritize low-energy, eco-compatible triggers (e.g., ambient moisture curing).
Healing Outcome	Crack sealing via polymerization, bond reformation, or agent solidification.	Autonomous or stimulus-assisted repair.	Restores ≥80% mechanical properties in ideal cases; repeatable (intrinsic systems).

2.3. Material Selection in Green Composites

Performance of the green self-healing composites relies on the selection of materials. Flax, hemp, jute, and bamboo are the most common natural fibers used as reinforcement because they are low density, renewable and biodegradable; recycled carbon or glass fibers provide a means to achieve high performance with a lower environmental cost. The resin backbone may be bio-based polylactic acid (PLA), bio-based epoxy compositions, thermoplastic starch or lignin-based thermoset. Healing materials, too, are chosen on their environmental pedigree-plant oils to cure when exposed to air, water-based adhesives that harden without toxic solvents or biopolymer gels that gel in a wet climate. The real task of making such a composite is to harmonize these parts in such a way that the self-healing mechanism is incorporated into the microstructure in a way that does not compromise the minimum mechanical properties necessary to effectively perform the structural function that the composite is assigned. Different chemical and physical mechanisms are at play in the healing process. In some green thermoplastic matrices, heating above the glass transition temperature allows polymer chains to move and re-entangle across the fracture plane, closing cracks in a process known as thermal reminding. Chemical reactions are performed in other systems to make new crosslinked networks at the crack interface. An example is the oxidative polymerization of plant oils, including linseed oil, the oxidation of which by atmospheric air provides the driving force behind a curing reaction that solidifies the oil into a polymeric film that seals the damage. An alternative pathway is enzymatic crosslinking, where monomers of a bio-origin can be homo and hetero crosslinked under mild conditions with the help of natural catalysts, e.g. laccase or peroxidase. Certain curing mechanisms react to moisture, especially when in a marine environment or in a humid climate, the water may make hydrophilic agents swell then fill the crack and create binding adhesions between the injured surfaces [2,24–26].

2.4. Performance Assessment and Industrial Relevance

The efficacy of a self-healing green composite needs to be determined both mechanically and environmentally. Mechanical performance is usually evaluated by determining healing efficiency, the ratio between a mechanical property of the healing, such as fracture toughness or tensile strength, to the non-damaged material. Significant healing efficiency shows that the composite is able to regain a lot of its original strength following damage. The repeatability of healing through a number of healing cycles, the rate of healing response, and long term durability of the remodelled area under periodic loading and environmental exposures are the other major concerns. On environmental front, the renewable content, biodegradability, and life cycle impact of the composite is analyzed together with toxicity profile of chemicals taken in healing respectively [2,27].

The requirements on such composites are intense in the event of structural use. They are also required to resist cyclics, thermal, UV and environmental water during service lives lasting decades. The healing mechanism has to work dependably under these conditions in a way that does not degrade the stiffness, strength or fatigue of the base material. It should also be capable of being used with industrial scale manufacturing processes, such as; vacuum assisted resin transfer molding through to filament winding, without involving prohibitive complexity or costly preparation. When such an extended list of criteria is achieved, we can surely say that self-healing green composites are viable in challenging applications like aerospace panels, wind turbine blades, marine hulls and elements of civil infrastructures. The principles touched upon here are used as the basis of the three core self-healing methods discussed here namely the bio based healing agents, microcapsule systems, and vascular networks that are to be discussed in great depth in subsequent sections [28].

3. Bio-Based Healing Agents

3.1. Introduction to Bio-Based Healing Systems

Bio-based healing agents form one of the most promising directions in the development of self-healing green composites because they allow damage repair to be combined with environmental sustainability. These healing agents are derived from renewable resources such as plants, animals, or microorganisms, and are chosen for their ability to initiate or participate in a healing reaction under service conditions. Unlike many conventional synthetic healing agents—which are often petroleum-derived, non-biodegradable, and sometimes toxic—bio-based systems are typically biodegradable, have lower environmental impact, and can be sourced from agricultural by-products or other renewable feedstocks. In structural applications, their use aims to extend the service life of composites while reducing the ecological burden associated with both the production and disposal of the material.

Bio-based heal promoting agents too do not pass without their hiccups. These types of agents are inherently sustainable but still require high performance criteria, such as reactivity (adequate reactivity to seal cracks in a timely manner), compatibility with the host composite matrix, stability in service, and low negative impact on the baseline mechanical properties of the composite. Not only cost but also processing conditions and long-term availability of the feedstocks used to produce them must be considered when incorporating them into a structural composite according to the price criteria [29].

3.2. Sources and Types of Bio-Based Healing Agents

Depending on its source and chemistry, bio-based healing agents may belong to quite a few categories. Among the most commonly researched there is plant derived drying oils, which include linseed oil, soybean oil and tung oil. The oils have high content of unsaturated fatty acids which are subject to oxidative polymerization upon exposure to air, to form a solid network polymer that block cracks. Examples of self-healing microcapsules include the use of linseed oil to apply in systems where the oil has the capability of being released into the cracks and curing at room temperature to permanently seal the crack.

Healing agents that belong to a different important category are biopolymers. They are natural polymers, i.e. chitosan (obtained by deproteinizing crustacean shells), alginate (extracted of brown seaweed), and starch derivatives (obtained by plants). Such materials gel or form films in presence of moisture or some ions, and thus are also ideally suited to healing conditions in humid or salty surroundings. They tend to be water-soluble or water-swellable so that they can make their way into micro-cracks and form bonds on the fracture surfaces before they set.

A third type of bio-based agent is the enzyme-catalyzed healing systems. This use naturally occurring catalysts (i.e. laccase, horseradish peroxidase, or tyrosinase) to induce polymerization or crosslinking of a bio-derived monomer when damaged. Enzymatic systems can be highly selective, operating under mild temperatures and pH conditions, and they can be tailored to cure in the presence of specific environmental cues, such as oxygen or moisture [30,31]. **Table 2** summarizes the principal categories of bio-based healing agents, their natural sources, activation mechanisms, and key advantages for self-healing applications

Table 2. Classification of bio-based healing agents for self-healing composites.

Category	Source/Examples	Healing Mechanism	Key Advantages
Plant-Derived Drying Oils	Linseed oil, soybean oil, tung oil.	Oxidative polymerization (cures in air to form solid networks).	Ambient-temperature curing; renewable feedstocks.
Biopolymers	Chitosan (crustacean shells), alginate (seaweed), starch.	Gelation/film formation via moisture/ion exposure.	Water-responsive; ideal for humid/marine environments.
Enzyme-Catalyzed Systems	Laccase, horseradish peroxidase, tyrosinase.	Enzymatic crosslinking of bio-monomers triggered by damage.	Mild conditions (pH/temperature); high selectivity; eco-friendly.

3.3. Mechanisms of Healing with Bio-Based Agents

The healing process in bio-based systems typically follows one of several reaction pathways, depending on the chemistry of the agent. Oxidative polymerization is common in drying oils, where exposure to air initiates a free-radical chain reaction that crosslinks unsaturated fatty acid chains, turning the liquid oil into a solid polymeric film. This process can occur at ambient temperatures but may take several hours or days to reach full cure, depending on the oil's composition and environmental conditions. In moisture-activated systems, hydrophilic biopolymers such as chitosan or alginate absorb water, swell, and infiltrate cracks. The absorbed moisture can also trigger ionic crosslinking (as with alginate in the presence of calcium ions) or chemical gelation (as in chitosan crosslinking with glutaraldehyde or genipin). This swelling not only physically closes the crack but also promotes adhesion between the damaged surfaces [30].

Enzymatic systems operate through a catalyst embedded within the composite, which remains inactive until the healing agent comes into contact during a damage event. For example, laccase can oxidize phenolic compounds into reactive radicals that then polymerize into a crosslinked network. Because enzymes are highly specific, these reactions can proceed efficiently under mild, environmentally friendly conditions without the need for toxic catalysts or high heat.

3.4. Advantages of Bio-Based Healing Agents

The primary advantage of bio-based healing agents is their sustainability. They are sourced from renewable feedstocks, which reduces reliance on finite petroleum resources and can lower the carbon footprint of the material. Many bio-based agents are biodegradable or compostable, ensuring that end-of-life disposal has less environmental impact. They often exhibit low toxicity, making them safer for both manufacturing workers and the surrounding environment.

Bio-fertilizing healing agents also favourably match natural fiber reinforcements that are gaining milieu in green composites. An example is that it is easy to make plant oil wetted on lignocellulosic fibers, the adhesion at the interface would be strengthened in the healed area. The chemical compatibility between the bio-based healing agent (in some cases) and the matrix resin (can) lead to superior integration as compared to synthetic agents [32].

3.5. Challenges and Limitations

Though they are advantageous, bio based healing agents are quite challenging. Their reaction rates are slower than those of synthetic counterparts and this may slow restoration of mechanical performance when damaged. In high stress structural loads in particular the speed that load can be recovered is vital. They are also sensitive to the environment (humidity, temperature, oxygen availability) and sometimes display great variability in the results of healing.

The other limitation is the storage stability. Certain bio-based agents, especially highly unsaturated oils are subject to premature volatile polymerization or rancidification during storage which decreases their efficacy. Additives that stabilize them or encapsulate them are commonly required to add shelf life, which may complicate and increase cost. Moreover, specific bio-based polymers might not attain the same final mechanical properties in the repaired area as synthetic thermosetting resins and this might restrict their application in areas where hyperbolic performance is required.

3.6. Recent Developments and Structural Applications

The latest studies concerned the way to break such limitations by modifying materials and by hybridifying. As an example, the plant oils can be modified using nanoparticles including nano clay, graphene oxide, or cellulose nanofiber that will help speed up the curing process and enhance the mechanical properties of the resulting healed material. In enzyme systems, catalyst stability and reusability into several healing cycles has been demonstrated by immobilization of the catalyst in the matrix. There have also been examinations of hybrid systems that incorporate bio-based agents in conjunction with synthetic co-monomers to provide balance between sustainability, fast curing and strong performance. Bio-based healing agents have been demonstrated in a range of structural applications. In wind turbine blades made from natural fiber composites, linseed oil-based microcapsules have been used to repair microcracks from fatigue loading. In marine structures, chitosan gels have been applied for sealing water ingress points, benefiting from their natural resistance to microbial degradation. In civil infrastructure, bio-based epoxies derived from vegetable oils have been used as both matrix and healing agents, enabling cohesive repair of load-bearing components [33–35].

Bio-based healing agents offer a path toward self-healing composites that are both high-performing and environmentally responsible. While their slower reaction times and environmental sensitivities present challenges, advances in material formulation, encapsulation techniques, and hybrid systems are steadily improving their viability for demanding structural applications. In the broader context of self-healing technologies, they represent a crucial bridge between engineering performance and ecological responsibility, aligning with the growing demand for sustainable, long-lived materials in modern industry.

4. Microcapsule-Based Self-Healing Systems

4.1. Introduction to Microcapsule Systems

Self-healing systems based on microcapsules The approach of adding self-healing ability to composite materials by the use of microcapsules is one of the oldest and best-studied methods. Small capsules, which have a diameter of a few, or a few hundred, micrometers are incorporated into the polymer matrix during their manufacture in this strategy. The liquid or semi-liquid healing agent, and in some cases a catalyst, is held in each capsule, isolated and inert, under normal operating conditions. As a crack advances into the material, the crack front breaks all the microcapsules lying along the crack path and the healing agent is released directly into the damaged site. The healing agent solidifies (via polymerization) or comes to solidity when exposed to air or moisture, or a catalyst present as an embedded material, and binds the crack-faces back together and mechanically repairs that part.

So simple and convenient are the microcapsule-based systems that they are rather popular. They can be processed into a wide variety of composites of both the synthetic and the green variety without having to invest in great alterations to the established manufacturing processes. In addition, they have the independent activation, i.e. do not require any external stimulus or manual trigger when they have been damaged. Nevertheless, along with the benefits, these systems possess significant drawbacks, especially in the context of single-use their character, because every capsule is able to treat only a single type of damage [36,37].

4.2. Fabrication and Design Considerations

Microcapsule fabrication in self-healing applications, the manufacture of a microcapsule is usually achieved through in-situ polymerization, interfacial polymerization or solvent evaporation. One of the most popular methods is in-situ polymerization, in which a polymeric shell can be formed around the droplets of the curing agent suspended in a liquid aqueous medium, commonly urea-formaldehyde, polyurethane, or poly(urea-formaldehyde). Within the green composite domain, bio-based shell material is increasingly considered, where instead of non-degradable materials (e.g., fibreglass), newer materials (e.g., polylactic acid, cellulose derivatives, or chitosan) can be used and degrade together at end-of-life with the matrix.

The development of really efficient microcapsules demands a strict control of a number of parameters. The size of capsules is critical: larger capsules have a greater capacity to hold more healing agent and may seal larger cracks but may weaken the matrix of the composite or serve as stress concentrations. In contrast, extremely small capsules do not tend to rupture dependably when the crack goes by, and this lowers the efficiency of healing. Thickness of the shell also plays a part, whereby it would affect the strength of the capsule during processing as well as the ease in rupture when damaged. The content used in capsules, its stability and compatibility with surrounding matrix of the matrix is equally important- at least in structural applications where healing agent should not leak out or be undergone to premature reaction [38].

4.3. Healing Mechanism

The self-healing process in microcapsule systems begins the moment a crack forms in the composite under mechanical loading or environmental stress. As illustrated in **Figure 4**, when a crack propagates through the composite, it ruptures embedded microcapsules, releasing a liquid healing agent that polymerizes upon contact with a dispersed catalyst, thereby autonomously repairing the damage. As the crack propagates, it encounters embedded microcapsules along its path. The mechanical stress at the crack tip ruptures the thin capsule shells, releasing the healing agent into the newly formed crack plane. The healing agent then flows, driven by capillary action, into the damaged region. If the system is designed with a separate dispersed catalyst—such as Grubbs' catalyst in the matrix for ring-opening metathesis polymerization—the agent polymerizes rapidly upon contact. In other systems, the healing agent may cure upon exposure to oxygen, moisture, or heat. In other systems, the healing agent may cure upon exposure to oxygen, moisture, or heat. **Figure 5** provides a complementary schematic of this process, highlighting the roles of the catalyst, healing agent, and polymerized sealant in restoring structural integrity. Unlike **Figure 4**, which emphasizes capillary action and catalyst interaction, **Figure 5** focuses on the sequential stages of crack healing, from rupture to final repair.

Figure 4 shows that the matrix exhibits damage and cracks. When cracks reach the microcapsules, capillary action releases the healing agents into the cracks. Upon contact with the catalyst, the healing agents undergo polymerization, sealing the crack.

The polymerization or solidification process creates a cohesive material bridge between the crack faces, effectively sealing the damage. In ideal cases, this restoration recovers a significant proportion of the composite's original mechanical properties. The healing efficiency depends on several factors, including the volume of agent delivered, the speed of curing, the adhesion between the healed material and the matrix, and the match in mechanical properties between the healed region and the surrounding composite [39].

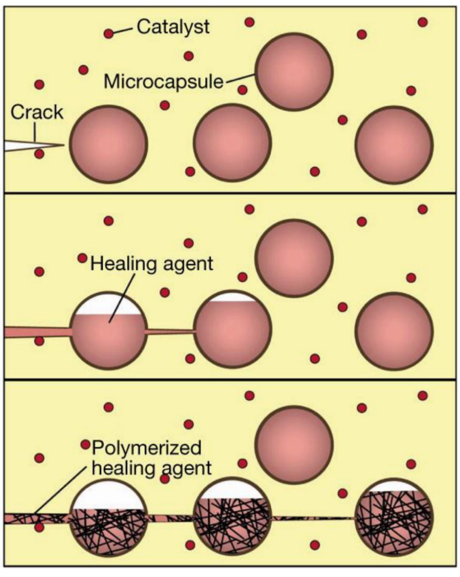

Figure 4. Schematic illustration of microencapsulated healing agents within a composite matrix integrated with catalysts for polymerizing the healing agents.

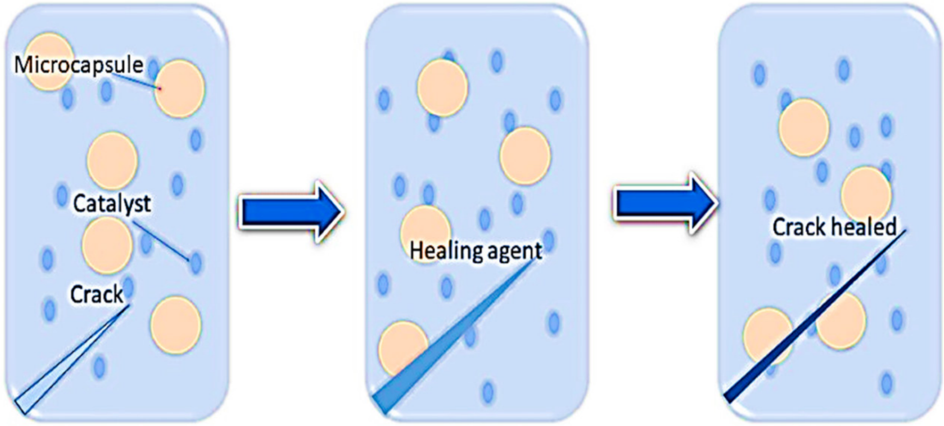

Figure 5. Autonomous self-healing mediated with embedded microcapsules.

4.4. Advantages of Microcapsule Systems

Healing systems with microcapsules have several positive attributes. One is that they can activate in an autonomous way, and after damage is caused healing can start without sensors or other interventions. Such a rapid self-contained response can be important in blunting crack advancement under service loads. Second, its technology is not hard to convert into current composites production methods, including resin transfer molding, vacuum-assisted resin infusion, or even big scale printing, which is attractive to large-scale production. Third, the flexibility of the concept makes it possible to use a broad spectrum of healing agents which makes it tailorable to various environmental and structural needs such as bio-based oils, epoxies and monomers.

The perspective of microcapsules in green composites is that shells could be made biodegradable and renewable healing agents can be loaded into then making the technology in line with the goals of sustainability. This versatility is very helpful in repair in outdoor or marine locations where use of harmful chemicals are required to be avoided [12].

4.5. Challenges and Limitations

Nonetheless, microcapsule-based systems are built on certain disadvantages even though they have strengths that inhibit their use in certain structural cases. Among the most singular features, their single-use character should be listed. That localized repair power is taken away after a capsule has burst and died off. In case the same area is damaged twice, there is no way to heal it any further except when there are other intact capsules around. With cyclic loading structures, this constraint poses special issues as the damage may recur in the same site. A second difficulty is related to trade-offs of the mechanical nature added when the microcapsules are embedded within a composite matrix. Capsules may be stress concentrators, which reduces strength, stiffness or impact resistance-particularly when they are large or weakly bonded to the matrix. Manufacturing may also become complicated and create a possibility of creating voids due to the specific addition of capsules in increasing resin viscosity during fabrication. Another factor is long-term stability. The healing agents cannot be inert until the end of the life of the composite, decades in structural applications. In time, exposure to moisture, oxygen, or high temperatures may destroy the shell of the capsule, or the healing substance within, it; thus diminishing the force. Moreover, the ensuring uniform distribution of capsules within the matrix is necessary to maintain consistency of healing performance, which is difficult to accomplish in large-scale or thick composite structures [11].

4.6. Recent Advances and Innovations

The most recent studies attempted to solve these drawbacks with the better capsule design, coupled with hybrid healing methods, as well as the environmentally friendly materials. A new development is the higher-order microcapsules with dual-core where two diverse healing substances are individually encapsulated and subsequently discharged into the crack where they react with one another to create a stronger, more lasting repair. Self-healing microvascular composites is another strategy that uses capsules as healing-reservoirs attached to recharging channels, which integrate both idea of capsules and vascular in multi-cycle healing [40].

New breakthroughs have also happened in shell chemistry. To limit environmental impact, researchers are shifting towards bio-based capsules, including those composed of polylactic acid, chitosan, or cellulose acetate, to minimize avoiding environmental claims. This can be achieved by the surface treatment of capsule shells to enhance adhesion to the matrix thus increasing the load transmission and decreasing the loss of mechanical properties. Mechanical enhancement after curing and environmental stability The healing agent has had nanomaterial additives, graphene oxide or nanoclay, added to strengthen it and increase its stability to environmental degradation.In structural applications, microcapsule-based healing systems have been demonstrated in natural fiber-reinforced composites for automotive body panels, in green epoxy matrices for wind turbine blade sections, and in lightweight sandwich panels for marine craft. A notable innovation is the adaptation of this technology for nanocapsule-based protective coatings (refer **Figure 6**). Here, nanocapsules (1) are embedded in polymer coatings applied to metal substrates. When damage occurs (2), the ruptured nanocapsules release healing agents (3) that polymerize to seal microcracks (4), simultaneously restoring the coating's barrier properties and preventing substrate corrosion. This approach is particularly promising for bridges, offshore platforms, and other infrastructure exposed to harsh environments. These examples highlight the versatility of the approach, particularly when tailored to specific environmental and mechanical demands.

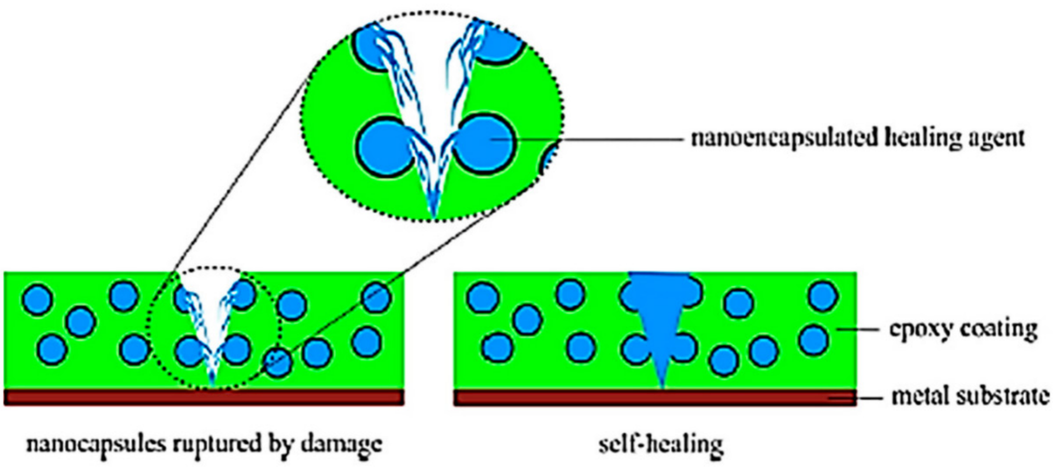

Figure 6. Self-healing mechanism in polymer nanocomposite using nanocapsules.

Microcapsule-based self-healing systems offer a proven, versatile route to autonomous repair in green composites, with the added advantage of straightforward integration into existing manufacturing processes. While their single-use limitation and potential mechanical trade-offs remain significant challenges, ongoing research into biodegradable shells, renewable healing agents, and hybrid capsule–vascular designs is steadily expanding their applicability to demanding structural environments. When combined with sustainable materials and thoughtful engineering, microcapsule systems can play a vital role in extending the service life of structural composites while reducing environmental impact.

5. Vascular Network Systems

5.1. Introduction to Vascular Networks

Such vascular network self-healing systems are literally inspired by nature. Much as the circulatory systems in living creatures and plants pump nutrients and repair molecules to locations that require these molecules, vascular self-healing imparts a network of interconnected channels within the material to deliver healing agents to the area of damage. In contrast to microcapsule systems where a one-time release of limited amounts of healing agent is possible, vascular systems can theoretically be refilled many times and many repair cycles can be undergone at the same site within the service life of the structure. This particularly makes them desirable to use in applications with repeated occurrence of damage or fatigue cracking, e.g., in wind turbine blades, bridge decks, or aerospace parts, etc.

The ability of systems of vascular self-healing is ongoing provision. The reservoir of healing agents lies either outside or within the composite and is linked to the network of channels within the composite. In the case where damage is done, and the crack runs into one of these channels the healing agent spreads either by capillary action or by pressure driven delivery into the damaged area and a repair reaction occurs. As many times as required, this procedure may be repeated provided that the reservoir can be recharged or the healing agent regenerated on-site [41].

5.2. Design and Fabrication of Vascular Networks

Designing a vascular structure in a composite to achieve a balance of mechanical performance, healing coverage, and manufacturability must be considered. This second step requires a network to be sufficiently dense that the path of possible cracks crosses the network many times, but sparse enough not to impose too much stress concentration or cause much weakening of the material. Typical network architectures are parallel networks, intersecting meshes, and branching tree like patterns which resemble the hierarchical structure of veins in the physical world.

There are a number of methods to fabricate vascular networks. In one technique, sacrificial flexible fiber or rod, sometimes in a thermoplastic or water-soluble polymer, is encapsulated in the composite during lay-up. These elements are then melted, dissolved or chemically degraded out of the composite after it has been cured, leaving hollow channels where they were. The other process involves the production of pre-formed channel networks by 3D printing using biodegradable polymers, which are incorporated within the composite during the process of molding. More elaborate techniques are direct ink writing or machining channels with lasers into hardened laminate.

The channels themselves could use bio-based or biodegradable materials in green composite systems which allows the entire system not to be environmentally incompatible. Furthermore, the curative factors found into these networks can be based on renewable sources, e.g., Botanicals, e.g., bio-based epoxies or plant oil-based formulations, to make the technology in line with sustainable objectives [42].

5.3. Healing Mechanism and Operation

The remodelling of the vascular techniques is simple but very efficient. At times when a crack in the composite crosses a channel, it bursts at the contact point with the channel wall. This break permits the restoration material of the linked reservoir to fill the break. Often the liquid is carried through capillary action, along the crack, to fill the gap. Some of the designs have the reservoir pressurized, which guarantees the delivery of the agent at a high rate with complete delivery.

Once in place, the healing agent cures through a chemical or physical process—often polymerization, oxidative crosslinking, or gelation—bonding the crack faces and restoring mechanical continuity. The reaction mechanism depends on the chemistry of the healing agent. In bio-based systems, oxidative curing of plant oils or enzymatic polymerization of bio-derived monomers are common, while in hybrid systems, fast-curing epoxies or polyurethanes may be used for rapid strength recovery.

Because the network is connected to a reservoir, the process can be repeated as often as necessary. If the same region is damaged again, fresh healing agent can be delivered through the same channel or a neighbouring one. This multi-cycle healing capability gives vascular networks a significant advantage over microcapsule systems, particularly in high-cycle fatigue environments [43].

5.4. Advantages of Vascular Network Systems

One of the most important advantages of vascular networks is their ability to support repeated healing events over the lifespan of the composite. This multi-use capability dramatically improves the long-term durability of structural components, especially in scenarios where damage is likely to occur in the same area multiple times. The continuous or on-demand replenishment of healing agent also means that the volume of repair material available is not strictly limited by the size of an embedded container, as in microcapsule systems.

Advantage of another is an opportunity of integration with the structural health monitoring. Since these channels are linked to a reservoir, one can check how much fluid of the healing agent is passed or even observe the variation of pressure along the network in real time to identify damage. Such twofold functionality (repair and monitoring) has the potential to improve security and cut down the expenses of inspection of critical infrastructure.

Vascular networks in green composites provide one more benefit of sustainability. Biodegradable materials used to formulate the channels and incomplete renewing healing agents are also possible. Besides, since the same structure could be repaired repeatedly without replacing the part, there is a much smaller environmental impact throughout a product lifespan [44].

5.5. Challenges and Limitations

Engaging in the development of vascular systems introduces a number of engineering issues. Complex well-distributed networks can technically difficult to fabricate in thick or intricately shaped composite parts. Inserted channels can cause weak points or decrease the entire stiffness and toughness of the composite, especially when the channels are extensive or broad. Care has to be taken by designers not to sacrifice structural performance by under-optimizing the network geometry.

The other difficulty is the provision of uniform coverage of possible crack locations. There is also the possibility in large structures that not all the damage sites are able to overlap with a channel hence causing partial healing coverage. Moreover, it is not always easy to maintain sealed and operating network many years/ decades or longer. The curing resin, debris used or contamination may block the channels and the healing agent may not reach the crack.

There are restraints on the choice of the healing agent itself. The agent needs to be of low enough viscosity to pass through small channels readily, but which will cure quickly and bond well when in the crack. On bio-based systems, this usually implies to achieving an optimum between environmental compatibility and reaction speed and the mechanical strength. Long-term storage stability is of also major concern as the healing agent in the reservoir can stay unused over a substantial length of time before it is alerted to act.

5.6. Recent Developments and Structural Applications

Recent developments in the technology of a vascular system used biofabrication technologies to enhance manufacturability, channel designs, and the performance of healing agents. 3D printing has appeared to be a very powerful technology with the advantage of offering precise control over channel geometry, as well as the capability to reproduce complex and branched geometry to emulate biological vascular patterns. Hierarchical network structures (in which large initial channels feed small secondary and tertiary channels) have been demonstrated to increase both healing coverage and efficiencies with reduced effects on mechanical properties.

Researchers have also developed self-refilling vascular systems, in which capillary action automatically draws fresh healing agent into the channels after each repair cycle, without the need for external pumping. In some designs, two-part healing agents are stored in separate networks and only mix when delivered to the crack, ensuring long-term storage stability.

Practical demonstrations of vascular healing have been carried out in large-scale structural components. In the wind energy sector, vascular systems have been embedded into prototype wind turbine blade sections to repair fatigue-induced microcracks. In aerospace, vascularized sandwich panels have been tested for their ability to seal impact damage, preventing moisture ingress and maintaining structural integrity. In civil engineering, vascular concrete composites have been developed to autonomously repair surface cracks, reducing maintenance costs and extending service life.

Vascular network self-healing systems offer a powerful and versatile approach to creating composites with multi-cycle repair capability. By delivering healing agents directly to the site of damage through an embedded channel system, they overcome the single-use limitation of microcapsules and provide the potential for continuous operation over decades of service. Despite outstanding issues in fabrication, structural optimization, and the development of healing agents, recent progress, especially in bio-based materials, additive manufacturing, and potential hierarchical network formation is fast approaching the attainment of this technology to widespread use. Vascular networks, in the context of green composites, are not only mantled to durability but also is offered a route to a greatly diminished environmental impact in the form of greatly increased service life and even renewable repair chemistry.

6. Conclusions

Green composites structural applications, in search of self-healing green war materials, is a combination of environmental responsibility and performance-based engineering. In the article, three main approaches, namely use of bio-based healing agents, vascular networks and microcapsule systems, have been discussed in details and have unique advantages, disadvantages, and mechanisms.

Bio-based healing agents prove non-autonomous repair can be implemented by making use of bio-sourced and biodegradable raw material, which are renewable and can be derived either out of plants or animals or even tiny organisms. They are made of low-toxicity ingredients which do not negatively interact with natural fibers and follow the premises of circular economy. Nevertheless, the low reaction rate, environmental sensitivity, and storage stability are critical issues of concern, especially to the field of high-stress structural applications where quick regain of mechanical properties is important.

Microcapsule systems are characterised by their ease of use, flexibility, and the ease of integration into the production of existing composites with minor disruption. They are self-activatable on crack formation providing instant repair and, with bio-based healing chemistries and biodegradable shells, are readily tailored to sustainable material systems. However, the disposable nature of capsules and the possibility of losing mechanical properties on inclusion of capsules still pose a challenge to long life durability.

The biological circulatory systems serve as inspiration to offering a radically different solution, the vascular network systems, in which repeated and even continuous healing throughout the service life of any component is possible. This causes them to be especially expensive in use where loads or repeated damage are subjected. When combined with structural health monitoring, this will add value to safety critical systems. However, manufacturing complexity issues, network optimization, as well as years of maintenance have to be resolved and therefore before major industry implementation.

With the comparison of these three approaches, one can notice that none of the systems excel over the other unconditionally. Rather, the decision to use a particular healing strategy should be based on the target use, performance specifications, environment and the sustainability goals. Healing agents based on biology are the best in environmental friendliness; microcapsules are practical and scalable in manufacturing and readily responsive; vascular networks are the best in repeatability and monitoring.

Predictively, going forward, hybrid systems combining the better attributes of multi-strategies, e.g., microcapsules embedded in vascularized matrices or bio-based healing systems tied with inherent self healing chemistry, are likely to be prioritized in future research. Increases in nanomaterial reinforcement, smart sensing ``in-corporation, and additive manufacturing will only further widen design potential. Besides, as a life cycle assessment emerges as a universal material selection tool, the industrial uptake will have an environmental performance-side based on self-healing composites.

As it may happen in the end, self-healing green composites can become a revolution in structural material design a paradigm shift to active lifespan, instead of passive endurance. These materials that allow the structures to mend themselves and have minimal effects with regards to the environment, fit in both the engineering performance requirements and the immediate necessity of sustainable and resource-efficient technologies. The systems are set to take centre stage in next generation structural engineering in the era of active research to translate the innovation that occurs in the laboratory into actual practice.

Author Contributions

Both authors contributed equally to the conception, design, data collection, analysis, and writing of this study. Both authors have read and agreed to the published version of the manuscript.

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The authors declare that there is no conflict of interest.

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