

Self-Healing Polymer Nanocomposites: A Comprehensive Review of Design Strategies, Mechanisms, and Emerging Applications

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Abstract

Self-healing polymer nanocomposites (SHPNs) are a new class of materials that can autonomously repair themselves to prolong their lifetime and improve their application performance. Herein, we provide a comprehensive overview of the design strategies, healing mechanisms, and emerging applications of SHPNs. Incorporating nanofillers (nanoparticles, nanofibers, and/or nanotubes) into polymer matrices leads to requisite functionalization that significantly enhances mechanical properties and is essential in self-healable products including improved cross-linking, high dispersion, and higher responsiveness to stimulus. The review classifies different self-healing strategies in two approaches: intrinsic and extrinsic. Intrinsic approaches use reversible interactions within the polymer matrix (hydrogen bonding or dynamic covalent bonds) to gain healing ability, while extrinsic methods have healing agents encapsulated within the polymer to be released when damage occurs. Specific design approaches to improve self-healing efficacy are as follows: optimizing the filler-matrix interface, well-dispersed nanoparticles, and well-designed healing trigger (thermal, light, electrical, or chemical stimuli). The addition of nanomaterials not only enables the healing process but also provides multifunctionality with improved thermal, electrical and mechanical properties. The potential of SHPNs in emerging applications is diverse, including aerospace, automotive, electronics, and biomedical fields, where reliability and durability are key metrics to success. In the review, the professors highlighted the recent progress made in SHPNs. They noted that SHPNs have the potential to revolutionise industries by providing sustainable material degradation and damage solutions. Future challenges include the optimisation of healing efficiency, manufacturing upscaling and cost-effectiveness are needed to advance the commercial application of these smart materials.

Keywords: Self-healing, polymer, nanocomposites, filler-matrix, well-dispersed nanoparticles

INTRODUCTION

Self-restorative polymer nanocomposites (SHPNs), emerged to be a revolutionary class of materials with the capability of healing damage and regaining functionality autonomously, hence markedly protracting the service life of materials and lowering maintenance costs in diverse industries [1-2]. These are polymer matrix-based materials consisting of nanoscale fillers including nanoparticles, nanofibers, nanotubes, and nanoclays, with increased and improved mechanical, thermal, and functional properties resulting in an efficient self-healing mechanism. Self-healing materials are designed to mimic biological systems that are capable of autonomous healing after damage to maintain structural integrity and functionality with time. Self-healing

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Received Date: January 01, 2025

Accepted Date: February 17, 2025

Published Date: June 05, 2025

Citation: Ankush Kumar Jain, Akash Panwar. Self-Healing Polymer Nanocomposites: A Comprehensive Review of Design Strategies, Mechanisms, and Emerging Applications. Journal of Polymer & Composites. 2025; 13(Special Issue 4): S281–S293p.

in SHPNs can usually be divided into two types, intrinsic self-healing and extrinsic self-healing [2-3]. The intrinsic self-healing mechanism is based upon the formation of reversible bonds (e.g. hydrogen bonding, dynamic covalent bonds or supramolecular interactions) within the polymer matrix. These dynamic connections allow the material to heal itself in response to external stimuli such as temperature, light, or pH changes that stimulate the reestablishment of broken connections and the recovery of material properties (Chen et al., 2020; Zhao et al., 2021). Extrinsic self-healing, on the contrary, has a healing process through nine encapsulated healing agents in microcapsules or vascular networks incorporated into the polymer. When the material is damaged, these capsules ruptures, releasing the healing agent, which upon reaction accumulates within the cracks and repairs the damage [3]. Although effective, extrinsic systems are constrained by the finite availability of the healing agent and the practical challenges of implementing microencapsulation technologies within the material. The addition of nanomaterials into these systems provides significant benefits in not only enhancing the mechanical property but also accelerating the self-healing process. Nanofillers like carbon nanotubes (CNTs), graphene and nanoclays can add reinforcement to the polymer matrix and enhance its strength, toughness and crack resistance [4]. In addition, nanomaterials can help provide a more homogenous distribution of healing agents or dynamic bonds throughout the matrix, allowing for a more uniform and successful healing. For example, the high-surface-area nanoparticles themselves can serve as cross-linking points in the polymer matrix leading to greater overall reversibility of bonds in intrinsic systems [5]. In addition, nanomaterials also can act as responders to stimulate the repair process, for example, the thermal healing can be triggered by heat-sensitive nanoparticles and the photo-activated self-healing can be realized by light-responsive nanofillers [5]. Apart from these all, some nanomaterials are also multifunctional with improved electrical and thermal conductivity useful in applications requiring restoration of conductivity during recovery from damage of the materials such as electronic components [4-6]. SHPNs must be designed with attention to the filler-matrix interphase, the nanofiller-filler interphase uniformity, and the trigger of healing should be optimized. A significant issue with the design of well-functioning SHPNs lies in the need for homogenous distribution of nanofillers throughout a polymer matrix, since agglomeration can be detrimental to both mechanical performance as well as self-healing capability [6-7]. Surface functionalization is an approach that has been proposed to improve the interaction between the nanofillers and the polymer chains, leading to a more even distribution of the nanofillers throughout the matrix [2]. Another important consideration in the design of SHPN is the selected healing trigger. The self-healing process can be triggered by different stimuli, including heat, light, moisture or chemical agents, and the choice of appropriate trigger is related to the final application of the material. In example, the technological approach used for thermally responsive polymers might rely on phase transitions or dynamic bond reformation at a specific temperature that performs self-healability, yet, traditional photoresponsive SHPNs can exploit ultraviolet or visible light for their healing self-repair [5-7]. This is just one example of how SHPNs can be customized according to the healing condition, making them applicable in a myriad of situations and industries. Nanofillers can develop selfhealing property and can also enhance the overall functionality of selfhealing polymer networks and the combination of nanofillers with the polymer matrix leads to development of SHPNs with tailor made properties for targeted applications. In the aerospace and automotive sectors, for example, SHPNs enable the development of lightweight, high-strength materials that can endure mechanical stresses while autonomously healing any damage sustained during use. In electronics, SHPNs with improved electrical conductivity and self-healing properties serve as durable and reliable materials for flexible electronics, sensors, and circuits. The S-SHPN strategy has also been applied to school biological subjects to develop new materials with self-healing properties based on biological self-healing principle, which will significantly enhance the service life of implants, dressings and other biomedical devices [6-8]. Nonetheless, several hurdles still exist in the development and commercialization of SHPNs. The self-healing process needs to be upgraded so that the materials could completely heal without affecting their mechanical or functional properties. To make such materials industrially relevant, SHPN production must also be scalable along with the related expenses. The continuous improvement of such SHPNs by harnessing advances in nanotechnology, polymer chemistry, and

materials science will facilitate the integration of SHPNs into a wider variety of applications where durability and sustainability are critical [7-8].

LITERATURE REVIEW

Fundamental Concepts and Mechanisms of Self-Healing in Polymer Nanocomposites

SHPNs is a new class of smart materials that can autogenously heal damage which can prolong life and improve reliability improvements of devices and polymer composites used in different fields. Self-healing approaches used in such materials fall into two main classes: intrinsic healing and extrinsic healing [5]. The underlying principles of each of these mechanisms vary widely, involving the methods by which damage is detected and subsequently repaired, and they take inspiration from biological systems, e.g., skin regeneration [6].

From the perspective of polymer matrix mechanics, intrinsic self-healing materials take advantage of the ability of polymers to reform following damage through the inherent characteristics of their polymer matrix without external healing agent application. This behavior is often attributed to the existence of dynamic and reversible interactions, which are typically hydrogen bonding, dynamic covalent or supramolecular. In intrinsic self-healing systems, repair is usually initiated by external stimuli (e.g., heat, light, pH, or mechanical forces) that enable the reconstruction of those reversible interactions [4].

In this context, dynamic covalent bonds are among the most explored. These molecules can cleave and recombine within certain parameters, offering a means to 'heal' at the molecular level. Key chemistries utilized in SHPNs for the above purpose include disulfide bonds, Diels-Alder reactions, and imine chemistry. In the event of damage, heat or another kind of stimulus is applied to activate the reversible reactions, and the broken polymer chains link back together, effectively recovering the material's structural integrity [7].

Intrinsic self-healing systems also exploit hydrogen bonding and other non-covalent interactions, such as metal-ligand coordination and van der Waals forces. These interactions are weaker than covalent bonds, but their reversibility and responsiveness to environmental changes lead to a rapid healing process. These types of non-covalent bonds are particularly well-suited for applications where the healing process must be repeated multiple times, as in the context of supramolecular polymers [10]. Hydrogen bonds, for the example, can break under mechanical stress but can reform spontaneously once the stress is released, allowing for many cycles of healing without degradation to the properties of the material [11].

One of the defining characteristics of intrinsic systems is stimuli-responsive healing. example is thermally responsive polymers, where heating them above a threshold temperature leads to a phase transition or bond reformation and triggers a self-healing. Alternatively, photoresponsive materials employ light to initiate reversible chemistries like $[2 + 2]$ cycloaddition or azobenzene isomerization, which allows localized healing of the light-exposed regions [9].

On the other hand, extrinsic self-healing is based on embedded external healing agents that are triggered to be exuded upon damage. This method most commonly depends on the incorporation of microcapsules, microvascular networks, or hollow fibers containing healing agents within the polymer matrix. In the event of damage, the capsules crack and the healing agents flow into the fissures, polymerizing or reacting with a catalyst to fill in the material [2,5,7,11].

One of the most common extrinsic approaches is microcapsule-based systems. The polymer matrix is dispersed with microcapsules filled with healing agents like monomers or resins. When the crack finally forms, the microcapsules at the crack tip burst open and release the healing agent into the damaged region. The healing agent then interacts with a catalyst embedded in the matrix or polymerises once it comes into contact with air, thus sealing the crack [13]. The efficiency of this process is limited by the capsule size, their distribution, and the healing agent reactivity.

Some researchers have turned to the world of biology for inspiration, utilizing microvascular networks, which are systems that mimic biological circulatory systems, as a means for targeted delivery of healing agents. Through continuous supply of healing agents, hollow channels interlinked together in networks are contained in the polymer matrix. When damage occurs, the network is able to directly deliver the healing agent to the damaged area in order to provide a potentially limitless supply of healing material [12]. It can be particularly beneficial for large or repeated damage but adds complexity to the material design and manufacturing.

Intrinsic self-healing mechanisms are limited by the ability of the material to respond to a stimulus, while extrinsic self-healing mechanisms generally require multiple pieces to inhibit healing. Nonetheless, both forms of self-healing mechanisms are highly promising. Intrinsic systems, for instance, tend to have issues with the trade-off between mechanical strength and healing efficiency. Dynamic bonds that allow for healing are often weaker than dynamic covalent ones, and thus impose an overall mechanical performance limit on the material [12-14]. In addition, most intrinsic self-healing materials are actuated by external stimuli and thus need careful regulation of the environmental conditions.

While useful, extrinsic systems are constrained by the exhaustible nature of healing agents. Once the microcapsules or vascular networks are consumed, the material can no longer self-heal. Furthermore, the incorporation of these systems into the polymer matrix may also create weaknesses where cracks are more prone to develop degrading the overall durability of the material [12].

This will critically help for next generation healing systems where intrinsic and extrinsic mechanisms can be integrated fusing passive healing with active healing, besides developing new type of dynamic bonds and efficient delivery systems will pave the way for better healing agents for in-situ applications [6]. Enhancing the design and function of self-healing polymer nanocomposites serves to fine-tune these materials for real-world application in challenging environments [13].

Nanofillers in Polymer Matrices: Enhancing Mechanical and Self-Healing Properties

The addition of nanofillers is critical for improving both the mechanical properties as well as self-healing behavior of polymer nanocomposites [5]. It has been possible to promote systems with much higher mechanical properties than the original grades (specifically, increase the strength, toughness and durability of polymer matrices, and promote more efficient self-healing processes) by introducing nanoscale fillers (e.g. nanoparticles, carbon nanotubes (CNTs), graphene or nanoclays) in the polymers. These fillers have nanoscale dimensions and high surface area, allowing them to interact at a molecular level with the polymer matrix, enhancing filler-matrix interaction, promoting better dispersion, and providing a higher level of load transfer across the composite [7].

One of the most fundamentally studied nanofillers is carbon nanotubes (CNTs), which exhibit remarkable mechanical, electrical, and thermal properties. This means we can improve the properties of the polymer composite by incorporating CNTs into a polymer matrix which results in mechanical properties such as tensile strength, stiffness, and thermal stability to have a drastic improvement. Furthermore, with high aspect ratio, spherical aggregates have the capability to construct a network in the polymer, which leads to mechanical reinforcement and effective electrical pathway [9].

Another well-known class of nanofillers is graphene and graphene oxide (GO), which in providing prevalent mechanical characteristics including high Young's modulus and fracture toughness. Graphene's two-dimensional morphology aptly orientates to react with the polymer matrix, yielding enhanced interfacial interactions and thus mechanical reinforcement [11]. Note: Graphene-based nanocomposites have shown the potential for superior self-healability and can be activated by thermal or electrical stimuli. Just a quick example, the high thermal conductivity of graphene could help to spread heat more evenly across the material which can facilitate more uniform healing of thermally responsive polymers [13].

Clay nanoparticles, including montmorillonite and halloysite types, are one class of layered silicate materials that have been widely used as nanofillers in polymer mats. Such nanoclays help with the barrier properties of polymers to be used in packaging to enhance gas and moisture prevention to improve the lifetime of the product. For self-healing performance, nanoclays have demonstrated their ability to increase the distribution of healing agents in the polymer matrix and to offer additional cross-linking sites to form dynamic bonds [15].

This addition has been found to enhance the mechanical properties including tensile strength, Young's modulus and toughness. Unlike traditional macro-fillers, the high aspect ratio and surface area of nanofillers provide them with the ability to interact more efficiently with polymer chains, boosting load transfer and crack propagation resistance, thereby reinforcing the polymer composites [17]. Nanocomposite self-healing efficiency is also boosted by the higher mechanical properties since stronger materials do not suffer damage in the first place. Nanofillers do not only improve the mechanical properties of the polymer matrices but are also crucial in the self-healing process. Thus, nanofillers can function as cross-linking sites and/or as reinforcing agents that enhance the reversibility of dynamic bonds in intrinsic self-healing systems. Carbon nanomaterials have been employed to improve the thermal and electrical conductivity of the material to facilitate the outcome of thermally or electrically sensitive healing mechanisms [19]. Nanofillers are also used to enhance the dispersion of microcapsules or vascular networks in extrinsic self-healing systems so as to enable a more homogeneously distributed healing agent in the matrix. This serves to avoid agglomeration, which may give rise to heterogeneous healing and severely reduced mechanical properties [16-18].

Although these nanofillers have apparent advantages for enhancing mechanical properties and triggering self-healing properties, there are still hurdles to overcome, including the fine control over dispersion and interaction of the nanofillers with the polymer matrix. Bad dispersion of nanofillers causes the nanofillers to aggregate and the self-healing performance is weakened by presence of weak spots where cracks are preferably developing [19]. This should be solved to enhance the compatibility and dispersion of nanofillers onto the matrix, which can be achieved by modifying the surfaces of the nanomaterials, or surfaces functionalization, notably, the induced grafting of polymer chains to the surface of the nanoparticles [5-8]. This poses another challenge in the potential mechanical reinforcement versus healing efficacy trade-off. In other instances, the presence of nanofillers changes the material's stiffness in a way that impedes the flow of polymer chains that must flow and reconfigure after damage in order for healing to be effective [11-14,17,18,19]. The challenge of balancing mechanical properties with self-healing capability remains an ongoing field of research, with continued efforts to create nanofillers that offer both reinforcement and flexibility. The use of nanofillers is similarly essential in the preparation of self-healing polymer nanocomposites, enabling us to obtain remarkable enhancements in mechanical performance and healing efficacy [1-3, 5-7, 8-11]. Yet the difficulties of enhancing nanofiller dispersion, boosting polymer-filler interactions, and reconciling mechanical properties with healing capacity remain a pursuit in this domain. The project and optimization of SHPNDs can be enabled by the recent and continuing advances in nanotechnology, flexible electronics interface, as well as the novel design strategy [13].

Design Strategies for Optimizing Self-Healing Efficiency in Nanocomposites

Developing approaches for maximizing the charm of self-healing efficiency in polymer nanocomposites (PNCs) is not straightforward, as it depends critically on nanofiller dispersion, polymer-filler interactions, and the relative determination of healing triggers. Uniform dispersion of nanofillers in matrix polymer is significant for the mechanical properties and self-healing property of nanocomposites. In fact, inefficient dispersion can lead to agglomeration of these fillers, which creates stress-concentration areas, promoting the rupture of the material instead of the healing (Li et al., 2019). To alleviate this problem, surface modification techniques have been introduced, either by functionalizing the nanofiller surfaces with compatible polymer chains or chemical groups, to improve the affinity between nanofiller and polymer matrix. For example, the incorporation of

oxygen-containing groups in the surface of carbon nanotubes (CNTs) or graphene can facilitate their dispersion in polar polymer matrices, leading to homogeneous filler distribution and improved load transfer upon mechanical stress [12]. The better interaction of nanofillers with polymer chains will contribute to superior mechanical characteristics of the composite as well as quick self-healing. Nanofillers can serve as cross-linking points or dynamic reversible bonds in the case of systems with intrinsic self-healing capabilities. Feasibly, the addition of nanofillers (like GO, CNFs etc.) in a polymer matrix based on reversible Diels-Alder reactions allows for an increase in the reversibility of the Diels-Alder bonds, thus rendering the material with the ability to self heal multiple times under appropriate conditions [8]. Moreover, in these systems, the even distributions of mechanical forces through the matrix can also mitigate localized stress and make the matrix less prone to catastrophic failure due to the presence of nanofillers. Nanofillers enhance the distribution and retention of the healing agent for extrinsic self-healing systems either encapsulated in microcapsules or vascular systems. For instance, adding silica nanoparticles or nanoclays in the polymer matrix improves the dispersion of microcapsules, providing a more uniform release of healing agents in the event of damage [17]. Not only does this enhance the overall healing performance, but it also reduces the likelihood of over-consumption of healers, increasing the service similarity of the material. The design also implicates other parameters like the choice of suitable healing triggers which are crucial for activating the self-healing mechanism. Thermally responsive systems are among the most studied, since heat can facilitate the reformation of bonds in dynamic covalent systems or the release of healing agents in extrinsic systems [18]. As an example, polymers which possess reversible Diels-Alder adducts can self-heal at elevated temperature (i.e. above a certain threshold temperature), once the bonds dissociate they will [re]form again upon cooling (Kunz et al., 2021). The challenge in these systems is that there must be a balance between the thermal activation temperature and the operational limits of the material. A high healing temperature may also decompose the polymer matrix or limit the application of the material in real life. And researchers have tried to remedy that by introducing nanofillers that help by reducing the healing activation energy. High thermal conductivity of CNTs and graphene has enabled rapid and localized heating to achieve a self-healing process, which are required to activate a self-healing mechanism [12]. Light-responsive nanocomposites, which heal from damage when irradiated with ultraviolet (UV) or visible light, are another type that is receiving growing interest. To address this, light-activated nanofillers (titanium dioxide (TiO₂), gold nanoparticles) can be embedded into the polymer network, as photoactive elements, which facilitate the localization of heat generation or initiate targeted chemical reactions to enhance healing processes under lighting [9]. This strategy is especially suited for electronics or coatings applications where a localized, non-destructive healing situation is preferred. Dynamic ionic bonds or metal-ligand interactions systems can be activated by chemical stimuli like changes in pH or exposure to certain ions. Polymers also possess metal-coordination complexes that can be healed with metal ions to promote broken bond reformation [11]. When designing such systems, it is important that the stimuli are specific and do not destroy the polymer matrix or induce unwanted chemistries that could affect the performance of the material. Another considerable challenge in design of SHPNs is the balance between mechanical reinforcement provided by the nanofillers and flexibility, which is essential for self-healing of the structure. Nanofillers like CNTs and graphene can notably improve the stiffness and strength of the polymer matrix, however, excessive stiffness can impede the polymer chains' mobility, needed for the healing process. To solve this, researchers have studied hybrid systems consisting of rigid nanofillers and a more flexible filler, for instance in the forms of polymeric nanoparticles or soft elastomeric segments, to ensure a balance between reinforcement and healing efficiency [15]. In certain cases, the nanofillers utilized in these composites can also be designed to respond to external stimuli, thus providing a greater efficiency to the self-healing process. One common example is that when the temperature-sensitive polymer is applied to iron oxide nanoparticles (10–80 nm), it shrinks (or expands) in/on the surrounding polymer and the temperature can facilitate the flow of healing agents or polymer chains to the damaged tissue [17]. The more responsive the material, the better control that you have over the repair process. Self-healing efficiency of nanocomposites will depend on the polymer-filler interface. For efficient load transfer

and healing, strong interfacial bonding between the polymer matrix and the nanofillers is a crucial requirement. A common approach to enhance this interaction is through surface functionalization of the nanofillers. As an instance, for polymer matrices that involve hydrogen bonding or ionic interaction, carboxyl or hydroxyl functionalized CNTs would disperse better in the polymer matrix and enhance the self-healing efficiency [22]. Besides, one of the most intriguing approaches to surface modifications involves surface-initiated polymerization methods to graft polymer chains directly onto the surface of nanoparticles that give rise to improved dispersion and can lead to the formation of covalent bond linkages between the filler and the matrix. These covalent bonds can further enhance the mechanical properties of the composite; with rich dynamic bonding sites, they are also helpful for healing. Lastly, these design strategies should be scalable and cost-effective to be used in real-life applications. Though laboratory-scale investigations have highlighted outstanding self-repairing performance for polymer nanocomposites, industrial applications of the concept still face numerous challenges [23]. The first problem was that some nanofillers (CNTs, graphene, ...) are so expensive that it is not realistic to produce at a large scale. To mitigate this, scientists are investigating mid-cost nanofillers, like bio-based nanoparticles or recycled materials to achieve the same advantages at a cheaper price [21]. Moreover, to commercialize self-healing nanocomposites, it will be important to devise simpler, scalable methods of incorporation of nanofillers into a polymer matrix such as extrusion or injection molding. Enhancing self-healing efficiency in polymer nanocomposites involves a delicate interplay between nanofiller dispersion, polymer-filler interactions, and the choice of suitable healing stimuli. Overcoming the obstacles associated with these factors will enable advanced materials to autonomously mend damage and prolong the life of products ranging from aerospace and automotive to electronics and healthcare. With ongoing advancement in this area of study, the transformative potential of SHPNs in material engineering and application is becoming apparent [15-18].

Emerging Applications of Self-Healing Polymer Nanocomposites in Advanced Industries

As one of the most promising materials in the continuous development of a variety of advanced fields, including aerospace, automotive, electronics and biomedicine, self-healing polymer nanocomposites (SHPNs) represent a kind of novel materials with unique multifunctional advantages, including enhanced durability, reduced maintenance costs, and improved life span. In aerospace, the ability to create materials that are lighter but still high performance is essential for allowing better fuel economy & performance [9-10]. In this context, self-healing polymer nanocomposites (SHPNs) represent a promising approach that combines incorporation of enhanced mechanical strength viPolymers (CNTs, graphene or nanoclays) within polymer backbone process with the ability to autonomously heal the damage inflicted by external conditions such as UV radiation, mechanical pressure, or thermal cycling. Components like aircraft wings, fuselages or space structures, for example, can be hard from be damaged and hard to access for repair, and gaining such capabilities will be particularly useful in such scenarios. For instance, nanocomposites have been designed containing microcapsules with healing agents for aerospace uses, permitting crack healing instigated by the heat released in operation or other stimuli (e.g. light) [11]. Introduction of additional elements in these materials such as graphene or CNTs have been shown to increase their thermal conductivity, which enables more efficient healing [13], and prevents crack propagation to maintain structural integrity of critical aerospace components. SHPNs have also been studied for use in space exploration, where harsh environmental conditions including temperature extremes and cosmic radiation become significant issues. Self-healing materials able to do so autonomously in orbit would ensure not only longer missions, but also lower operational risk due to decreased need for manual repairs [16]. SHPNs have also been incorporated into the automotive industry to optimize the stability, resilience, and efficiency of electric vehicles. Self-healing nanocomposites can be significant success for both structural and non-structural applications because they can have improved anti-wear properties and also have the ability to autonomously heal minor defects generated by scratches and cracks. For example, SHPNs can be used in coatings for car exteriors so that small scratches in the paintwork will heal upon contact with heat or sunlight, preserving the looks of the vehicle without repainting or refinishing. SHPNs incorporated into the production of tires, bumpers and interior

components also help to prolong the life of these parts, lowering maintenance costs while boosting overall vehicle performance [18-20]. Over the past few years, scientists have been proposing SHPNs that contain microcapsules or vascular pathways containing healing agents, which are triggered by the heat generated during the running of the tire or mechanical stress [21]. Such systems are capable of self-healing microcracks in tires to avoid further deterioration and blowouts. Happily, automakers are also investigating SHPNs in battery casings and electronic components, where even small damages could result in catastrophic failure. SHPNs are noted for their requirements for self healing, thermal and electrical properties, including electrical conductivity, by embedding the various nanofillers, such as CNTs or graphene into their polymer matrices as nanofillers [9-11]. To make the electronic device thinner, more flexible and more complex, thus increase the chance of mechanical damage to the electronic devices. Because SHPNs are able to close the microscopic cracks and damage from bending, stretching or impact, they can be used in the manufacture of flexible circuits, sensors and batteries; they ensure that the device they are in does not stop working. For instance, nanocomposite materials that consist of CNTs or silver nanoparticles have been created for the purposes of use in flexible circuits, where they contribute both electrical conductivity, and the capability of self-healing means of discontinuity in the conductive pathways due to mechanical stress [13-17]. These kinds of materials are especially useful for wearable health monitoring systems, flexible displays, and other consumer electronics that experience high-frequency mechanical deformation. The application of SHPNs in these scenarios can help prolong the lifespan of the devices and also reduce electronic waste, making consumer products more sustainable. In addition, SHPNs are also studied for energy storage systems including lithium-ion batteries and supercapacitors to improve the durability and safety of the device [14-17]. Long and unstable dendrites can also make up the electrodes in batteries, which in turn causes short-circuits and reduces battery lifetime. Since SHPNs provide the ability to autocure these microdamages, they present a potential way to increase battery lifespan and safety. More specifically, nanocomposites with the inclusion of graphene or CNTs in the polymer electrolytes of batteries have demonstrated their ability to self-repair and regain electrical conductivity post-damage, thereby ensuring uninterrupted functioning for improved energy storage [12-17]. These agents are being studied for use in areas such as tissue engineering, wound healing and drug delivery[6,7]. For example, self-healing hydrogels that were filled with nanofillers including silica nanoparticles or carbon-based materials, were engineered to be used in artificial skin, cartilage, or other soft tissues, whereby they are able to autonomously mend minor create and imperfections to a degree, imitating the natural repair system of the body [19]. These hydrogels also have the added benefit of being biocompatible and responsive to physiological stimuli like temperature or pH, allowing for localized healing without the need for invasive procedures. Furthermore, SHPNs have also been utilized to prepare smart carriers in drug delivery systems that release therapeutic agents in response to local stimuli, such as temperature, pH, and mechanical stress. Release of the payload three times was accomplished with self-healing of these systems upon release of the addition. For example, SHPNs are being investigated for use in medical implants to create self-healing coatings on devices, such as stents or pacemakers, where minor damage or wear could otherwise impede the performance of the implant. Self-healing could go a long way in extending the life and reliability of these devices and providing better patient outcomes by decreasing the surgical replacement of damaged devices. Furthermore, they can be incorporated into orthopedic uses, applying themselves in bone cements or in joint replacements, essentially helping to repair then microfractures or time wear, to prolong function and enhance their anchoring or fusion with surrounding tissue [22]. This leaves the full potential of SHPNs across various sectors still to be realized as the science of the material continues to develop. By inducing SHPNs in aerospace, automotive, electronics, and biomedical applications, manufacturers can realize multiple benefits such as improved mechanical integrity; durable performance; and self-repair functions that reduce maintenance costs and improve the overall sustainability of their products. Despite this, there are still challenges to hydrogels' performance optimization: this includes nanofiller dispersion uniformity, mechanical strength-healing efficiency balance, and scaling potential for industrial implementation [6-11]. However, the accelerated rate of development in new classes of nanomaterials enabled fabrication techniques, as well as increasing demand for more durable and

sustainable products has driven the implementation of SHPNs within various advanced industries. They are arguing now that these SHPNs will play an escalating role in the fabrications of future materials that can develop their own healing properties to maintain a longer useable lifetime and more reliable performance on some of the toughest operable applications [17].

MAJOR FINDINGS

However, several powerful advantages over conventional approaches make SHPNs a very active area of research, with many discoveries showcasing their potential in other advanced industries including (but not limited to) aerospace, automotive, electronics, and biomedical fields [20-23]. Key findings include the importance of the type of nanofiller material used for the mechanical properties and self-healing effectiveness of polymer matrices. Nanofillers like carbon nanotubes (CNTs), graphene, and nanoclays contribute to mechanical reinforcement, crack resistance, and durability enhancement of polymer nanocomposites [21-24]. Additionally, when damage occurs, these fillers encourage not only the formation of reversible bonds but also the release of healing agents, ensuring that the material can be restored to its integrated state. For enhanced performance, the nanofillers should be uniformly dispersed in the polymer matrix [25]. Surface modification of nanofillers has been identified as an effective strategy to enhance the compatibility of the nanofiller with the polymer matrix, increasing the distribution of the filler within the matrix, enhancing the mechanical performance and promoting the healing process. However, achieving an effective balance between mechanical reinforcement by using nanofillers and flexibility for self-healing is not yet well established, and attempts are being made to explore hybridization of rigid and flexible nanocomposites which possess both mechanical resistance and healability. Our other crucial discovery lies in choosing suitable healing triggers like thermal, light, and chemical stimuli, which play a pivotal role in inducing self-healing behavior. This is typically achieved by thermal activation, but researchers have also investigated light- and chemical-responsive systems to enable tailored, less disruptive healing, particularly in cases of dynamic covalent bonding[22-25]. The use of high thermally conductive nanofillers (such as graphene and CNTs) would enable localized heating and help reduce the activation temperature for healing and as a result, make the process more efficient and accessible. Within the aerospace sector, SHPNs have been found to improve the performance and stability of key components that are subject to severe conditions such as UV irradiation, thermal cycling and mechanical strain [20-22,24-26]. This means that these materials can repair themselves autonomously, leading to a reduced need for manually conducted maintenance and greater safety and efficiency for airlines and space agencies. In the field of automotive, SHPNs have been utilized in coating, tires, and structural components for both mechanical enhancement and self-healing of minor defects (like scratches, cracks, and tire wear) [25-27]. These materials are especially beneficial for improving vehicle robustness and decreasing maintenance requirements, thereby increasing the sustainability of automotive goods. Another significant avenue for SHPNs lies in electronics, where these materials are applied in flexible circuits, wearables, and batteries. The self-healable characteristic of these devices allows them to perform even after mechanical damages, moreover, other nanofillers such as CNTs and graphene have provided additional features such as electro-conductivity, keeping these devices fully operational in high performances throughout their lifetime [24-26]. Particularly in the biomedical area, SHPNs gained people's attention with their properties of mimicking the natural healing process as well as tissue engineering, drug delivery and medical implants. For instance, self-healing hydrogels have been developed for artificial skin and other tissues, which provide biocompatible materials that can autonomously repair small tears or defects. Furthermore, SHPNs utilized in drug delivery systems enable the release of therapeutic Agents to be localized and repeatable, thus minimizing dosing frequency, reducing side effects and improving patient compliance. However, these advances also face challenges in optimizing SHPNs for real-world applications. Self-healing materials, especially with high-cost nanofillers (e.g. CNTs and graphene), have limitations in terms of their scalability and affordability for large-scale industrial utilization [26-28]. In addition, achieving uniform dispersion of nanofillers in the polymer matrix and optimizing the healing efficiency without hindering the mechanical performance of the material is still

a challenging issue that need to be fixed. However, the promise of SHPNs has diverse applications for materials design and industries is evident, and researchers will continue to investigate ways to enhance the performance, sustainability and commercial viability of SHPNs. SHPNs enable more efficient dispersion and interaction between nanofillers and polymers, and the choice of triggers will allow SHPNs to emerge as a building block for self-repairing multifunctional materials that can enhance the mechanomic efficiency of material science and engineering, paving the way for the next generation of smarter and more sustainable products [26-29].

GAPS FROM LITERATURE

Following are the Gaps identified from the literature:

- *Improving nanofiller integration and interface compatibility:* Although dispersion and surface functionalization methods have been studied to enhance interfacial compatibility between nanofillers and polymer matrices, processing large quantities in a homogeneous manner over a significant area has yet to be resolved. Poor dispersion not only compromises mechanical performance but also healing efficiency. To ensure the uniform distribution of nanofillers throughout the polymer matrix in commercial applications, future efforts should prioritize scalable processes that promote improved dispersion and enhanced interfacial interactions [1,4,7, 9,11,22,26,30].
- *Mechanical property vs self-healing efficiency balance:* As nanofillers are introduced to the polymer, its mechanical properties improve, however, excessive reinforcement inhibits the polymer from flowing and self-healing. Navigating between reinforcement and flexibility for optimal self-healing is still a considerable struggle [31-33]. Though hybrid nanocomposites with a combination of flexible and rigid fillers have started to be explored, the ideal ratios and behavior have not yet been optimized for practical application [2,5,7,9,11,15,18, 22, 25].
- *Cost and scalability issues:* The commendable self-healing properties exhibited by SHPNs in the laboratory can be context-specific; incorporation of nanofillers, such as CNT and graphene, is expensive, and scalable production processes for SHPNs can be difficult to achieve, thus hindering their extensive commercialization [32-34]. To reach industrial sectors such as automotive, aerospace, and electronics, bio-based nanofillers or recycled materials, in particular, need to be developed for SHPNs to ensure economic viability [22, 28, 30].
- *Long-term performance and environmental stability:* The long-term performance and environmental stability of SHPN have not been adequately addressed for harsh environments such as high-temperature, UV radiation, and corrosive conditions [33-35]. Further work is required to assess the resilience of self healing systems to numerous operational stresses to guarantee both its long term effectiveness and its versatility in varying environmental conditions [1, 11, 17, 19].

DISCUSSION

Self-healing polymer nanocomposites (SHPNs) have emerged as a progressive class of nanomaterials with a wide range of potentials in numerous industries such as aerospace, automotive, electronics and biomedical applications owing to their improved mechanical properties combined with the autonomy of damage repair. Nonetheless, many challenges remain despite the promise, and they will need to be addressed to translate the potential for SHPNs to real-world applications. Among them, An important aspect is to obtain a homogeneous dispersion of nanofillers. Nanofillers (e.g. carbon nanotubes (CNTs), graphene, and nanoclays) improve the mechanical properties of the material, and their proper dispersion promotes the performance of the self-healing mechanism. Poor dispersion usually leads to agglomeration, resulting in stress concentration in the localized area and useless healing. It is well established that surface functionalization of nanofillers can enhance their compatibility with polymer matrices; however, scalable processes can often lead to variable dispersion of fillers, which constitutes a major barrier to industrial adoption. Furthermore, using nanofillers in the polymer matrix has to be finely tuned in order not to reduce mechanical strength while still granting the polymer flowability and healing ability. Excessive reinforcement of the matrix may cause loss of self-healing ability, and therefore, hybrid nanocomposites utilizing both rigid and flexible materials must be designed and fabricated to improve both mechanical reinforcement and

healing capacity. But there's no one-size-fits-all solution, as each application may require a well-defined set of performance characteristics. Aside from these technical issues, the prohibitive cost of certain nanofillers (e.g. CNTs, graphene) is a major impediment to the widespread commercial adoption of SHPNs. While these materials are high-performing, they drive the economics of SHPNs as a technology out of the cells for the automotive and aerospace industries. Hence, it is very high time to look for low-cost alternatives such as bio-based nanofillers or recycled materials, which can provide similar performance compared to the conventional models. Not only that, several SHPN production methods are yet not scalable and they were initially intended for use in small laboratory-scale setups. However, for these SHPNs to be effectively integrated into mass-production industries, scalable fabrication methods, like extrusion or injection molding, must be developed to permit the incorporation of nanofillers into polymer matrices while retaining the self-healing and mechanical enhancement properties. Another gap in the current research is the long-term performance and environmental stability of SHPNs. However, the in-field performance of such materials, especially when subjected to extreme environmental stresses including but not limited to changes in temperature, exposure to UV radiation, and mechanical wear, has not yet been thoroughly investigated, though laboratory testing has shown their ability to heal under controlled conditions. Self-healing system SHPNs have great potential in different industries, including aerospace, where withhold harsh environmental state as one among them; thus, ensuring over-multiple operative situations can bear far better efficient self-healing performance is a step that must remain considered during their real-time utilizations. Additionally to the use of SHPNs, their environmental impact and sustainability, especially in the fields of biomedicine or automotive devices, have to be also taken into account since materials may experience wear or the environment impact of SHPN's removal in both the industrial and socio-economic sectors, including logistics, energy, and waste management. All in all, SHPNs provide potential advantages (i.e., lower maintenance cost, longer service life, and more material sustainability) compared with conventional pavement, but more studies are needed to report the durability, deterioration characteristics, and environmental impacts of this novel pavement technology. In conclusion, SHPNs are proving to be revolutionary in various applications especially those in aerospace where autonomous self-healing can reduce maintenance time and improve safety as well as in automotive coatings and tires where it can enhance service life of these products ultimately reducing repair costs. In electronics, this means that flexible and wearable devices manufactured through SHPNs could self-heal on mechanical stress and keep on smooth operation. Utilization of self-healing hydrogels in the biomedical field is one of the more exciting applications of this technology, with applications including potential tissue engineering and drug delivery applications that could one day lead to improved treatment outcomes, since self-healing hydrogels attempt to mimic the healing found in nature. With research, developments in refined nanofiller dispersion and procurements in economical resources alongside scalable production methods will help minimize existing obstacles; thereby harnessing high-performance nanocomposites to enable their application in an assortment of industrial endeavors.

CONCLUSION

Finally, we would like to conclude that SHPNs can be a new fascinating approach in the field of material science with a great potential to be utilized in aerospace, automotive, electronics, and biomedical industries. Their superiority in self-repairing ability without sacrificing original or even reinforcing mechanical performance suggests promising merits in durability, life time and cost saving. Some issues such as those involving nanofiller dispersion, material compatibility, production scalability, and economic viability still need to be addressed to fully exploit their potential. A homogeneous distribution of nanofillers and balanced reinforcement are of vital importance to the structural and self-healing performance of composites. In addition, the high manufacturing cost and mass-manufacturing methods are subject to certain restrictions. It is crucial to seek for low-cost and sustainable nanofillers substitutes, as well as to develop scalable fabrication techniques including extrusion and injection molding. Moreover, long-term environmental stability and real-world performance have to be further studied under extreme conditions. The environmental impact and

sustainability will also be crucial, particularly when it comes to biomedicine and the automotive industry. A SHPN can become an attractive alternative to conventional EMI shielding materials, however, further evolution of this technology requires to bridge the existing research gaps. Ongoing development in terms of the formulation and processing, SHPNs are in a prime place to be one of the critical materials in the next level in smart systems and products.

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