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Page | 33

Smart Materials: Self-Healing Polymers for Structural Applications

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ABSTRACT

Smart materials, particularly self-healing polymers, represent a revolutionary advancement in materials science, offering promising solutions to extend the service life and resilience of structural systems. These materials possess the intrinsic ability to sense damage and autonomously initiate repair, inspired by biological healing processes. Self-healing polymers can operate through intrinsic mechanisms, based on dynamic molecular interactions, or extrinsic systems involving encapsulated healing agents activated by external stimuli. Their applications range across infrastructure, electronics, aerospace, and biomedicine. This paper examines the fundamental mechanisms of self-healing, types of healing systems, and the integration of smart polymers in structural engineering. Recent advances such as nanocomposite reinforcement, shape-memory polymer integration, and thermal-induced recovery methods are highlighted. Despite remarkable progress, challenges such as healing efficiency under extreme conditions, scalability for industrial applications aim to develop faster, more efficient, and more sustainable self-healing materials that can autonomously repair major structural defects without external intervention.

Keywords: Smart Materials, Self-Healing Polymers, Intrinsic Healing, Extrinsic Healing, Structural Applications, Responsive Polymers.

INTRODUCTION

Smart materials, or "intelligent" materials, respond to their environment in a controllable manner. Such materials change properties and functions as a response to changes in temperature, humidity, pressure, pH, time, etc., and exhibit sensitivity to the environment. Smart polymers are promising materials for various applications. Their unique properties are derived from special molecular interaction mechanisms of polymers, which include molecular mobility and chain motion, and long-range polymer network structure. Responsive polymers and composites with structural, shape recovery, color-changing, and healing functions are reviewed. They include shape memory polymer systems for structural applications, self-healing polymers and composites, and mechanical self-healing carbon nanotube (CNT)-polymer composites. Responsive polymers with structural applications respond to an environmental stimulus by changing their structure, shape, or conformation. When an equal amount of heat is applied to both types, the one with crystalline phases will respond first and show the most pronounced deformation, undergoing a glassy to molten transition [1, 2].

Overview of Self-Healing Polymers

Polymers are long-chain molecules that have become the backbone of today's high-tech electronics. Not just as insulating materials, they are also used to build circuits, sensors, membranes, motor parts, and more. However, polymeric materials can be damaged by dynamic loads, UV exposure, heat, or chemical corrosion. Such damage can lead to short circuits, leaks, malfunctions, or breakages. Conductors, especially in 3D-printed polymer-based electronics, are structurally more vulnerable and must be designed to heal under ambient conditions. The polymer material comprises healing agents that react with the polymers to repair damage. Healing agents can be microcapsules, vascular channels, or soluble small molecules. An external stimulus must either activate the healing agents or build up the polymer bonds. Alternatively, the healing agents can be intrinsically reactive and self-consume without an outside stimulus. Materials that mimic biology's self-healing ability and intrinsically recover their functionality

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are called "self-healing materials." Self-healing is an emergent, built-in behavior of a material that encapsulates autonomous, energy-efficient, and immediate repair. As a result, a material with memory, sensing, activation, and healing agent is classified as "informed". The self-healing mechanisms utilize energy input to reverse damage accumulation so that a healed material regains its parent functionality. Designed over the years to extend shelf life, self-healing now offers embedded, instantaneous repair. The self-healing materials heal the damage only after the material is loaded. In addition, the healing process requires external activation by high temperatures or light, followed by a complicated phase-in, and the healing is less than complete. Instead, "born self-healing" materials passively recover from a breach across timescales $\lceil 3, 4 \rceil$.

Mechanisms of Self-Healing

The self-healing process in smart materials is an adaptive response occurring under internal or external disturbances to recover the original properties of materials like shape, strength, stiffness, and functionality. In structural applications, self-healing behavior is triggered and regulated by temperature, light, moisture, and a magnetic field. The materials and mechanisms of smart self-healing polymer systems applicable to civil infrastructure systems with promising perspectives are discussed through external stimuli, with insights into the applications in structures, geotechnics, and construction materials in the following sections. Self-healing is described as a method that can perceive any dysfunctional device or system without human intervention and make the necessary adjustments to restore itself to its normal condition. The self-healing method is expected to detect and repair the microcracking in products or systems that help to recover the mechanical or functional performance of the material. This approach is very significant in situations where repairing or inspecting is difficult, hazardous, and costly. Self-healing can be categorized as passive self-healing and active self-healing. Passive self-healing should be more in the material design stage by incorporating stimuli-sensitive healing agents in the structures. By selecting appropriate self-healing agents and stimulation conditions, cracks can be healed within seconds or even microseconds. During active self-healing, the material must sense the damage and trigger the healing process. Hence, sensing materials providing feedback on the internal state of smart structures are required. Over the last decade, self-healing methods have attracted a lot of attention in the research community. Numerous studies have explored self-healing to develop various self-healing capabilities, particularly using polymeric materials [5, 6].

Intrinsic Healing Mechanisms

Combining polymers with intrinsic healing functions creates autonomous self-healing materials that operate beyond traditional timeframes and environmental limits. This innovative concept in polymer chemistry enables materials to autonomously repair damage, differing significantly from extrinsically healing materials that need additives for repair. By eliminating the need for dispersed healing agents, intrinsically healing materials maintain their living function despite impacts. This ability to self-repair can extend the lifecycle of materials while enhancing sustainability. The self-healing process relies on local dynamic interactions and applies to various polymer types, including polyethylene and polystyrene. The growing interest in these materials focuses on preserving structural resilience, especially for safety-critical applications. Intrinsic self-healing occurs through reorganizing polymer chains at crack surfaces without external agents, enabling repeated self-repair after multiple damages. This mechanism has several advantages: rapid self-healing due to the lack of diffusion and control steps, and the capability to adapt and restore minor defects or cracks, thanks to the sensitive, reversible, dynamic bonds operating at the molecular level that quickly initiate multiple healing processes [7, 8].

Extrinsic Healing Mechanisms

Self-healing polymers utilize two mechanisms for repair: extrinsic, which requires external stimuli, and intrinsic, which relies on specific materials. In extrinsic self-healing, the healing agents join at a slower rate than the crack opening, with processes involving diffusion, merging of polymer filaments, or slow fusion of melted regions. The kinetic and thermodynamic challenges of the healing agents make its effectiveness dependent on bonding energy, molecular weight, and compatibility with the matrix. Despite requiring external healing materials, extrinsic systems face limitations compared to intrinsic methods, which offer promising future applications. Functionalities of extrinsic self-healing include anti-corrosion coatings, electrically conductive coatings, and thermo-sensitive composites. This method often involves capsules containing antimicrobial agents in functional coatings. The construction industry could benefit from self-healing asphalt and concrete using embedded capsules with binders. Combining self-healing techniques with shape-memory and self-sensing materials allows for autonomous repair in harsh

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environments. Existing self-healing polymers can restore properties without external sources. They function through encapsulated healing agents released upon fracturing or via intrinsic dynamic bonds that form autonomously. Applications for these polymers span water membranes, lithium batteries, energy transducers, biomedical devices, flexible electronics, and soft robotics [9, 10].

Types of Self-Healing Polymers

Self-healing is an emerging research area in materials science that has gained more attention due to its potential applicability in various fields. Self-healing is described as a method that can perceive any dysfunctional device or system without human intervention and make the necessary adjustments to restore itself to its normal condition. The self-healing method is expected to detect and repair the microcracking in products or systems that help to recover the mechanical or functional performance of the material. It represents the next generation of technology, which helps to improve the important performance of products, as it has been built into various applications for structural, electronic, medical, and aerospace products. This approach is significant in situations where repairing or inspection is difficult, hazardous, and costly. Over the last decade, self-healing methods have attracted a lot of attention in the research community, and numerous studies have explored them to develop various self-healing capabilities, particularly using polymeric materials. Polymeric materials are formed by the bonding of monomers and polymer chains, and they are extensively researched in self-healing because of their versatility. For instance, a self-healing process that requires relatively low temperature can be achieved by the modification and functionalization of polymer systems and can effectively fill the cracks because of the higher viscosity of the polymer. Besides that, the self-healing polymers can transform physical energy into a chemical and/or physical response to heal damage. Polymers are in demand as they offer a lightweight material with good processability and chemical stability. In particular, efficient self-healing polymers and polymer-based fiber-reinforced composites are significant because these materials are utilized in numerous engineering fields, such as for cars, ships, spacecraft, electronics, and biomedicine. There are two types of self-healing mechanisms, which are the extrinsic and intrinsic self-healing mechanisms. Normally, nearly all self-healing mechanisms take place at room temperature [11, 12].

Applications In Structural Engineering

Despite ongoing research and development of various self-healing structural materials, challenges remain in applying self-healing technology successfully in structural engineering applications. Understanding the needs of structural engineers and developing materials that yield satisfactory performance across scales, costs, and user-friendliness are of paramount importance. From aesthetic concerns of the architectural applications to safety considerations of the structural engineering applications, some components and materials are more prone to accidents that induce damage. As the first step towards infrastructure selfhealing, the waterproof material on the concrete bridge decks provides a more realistic demand. Due to the aqueous leaking flow into prestressed concrete girders, the corrosion of the steel strand happens and is leading to a severe maintenance concern. Therefore, a self-healing waterproof material that enables concrete to remain dry even if an accidental impact happens is desired by the precast concrete bridge industry. To be widely used in the construction of infrastructure, sustainable resources should be selected as much as possible to reduce the carbon footprint. This is why, in the proposal of protective waterproof materials on concrete gages, sustainable resources are selected. The pine resin and wax with unique thermal properties are utilized. Emulsifying agents and surfactants are used to form a stable water-based ink for a spray application. The performance of the waterproof material is comprehensively characterized by water-barrier tests. As one of the most promising materials, the formation of water barriers on the surface of porous concrete can be achieved by spraying the ink under warm air to evaporate water. In this case, the liquid water would be stuck on the surface in the area that is still liquefied, while it penetrated the porous concrete in the area that is already dry. As the temperature decreases, the water-barrier performance recovers due to the further solidifying of nanoparticles and the formation of adhesive interactions. Therefore, the waterproofing function is self-dependent, based on the thermal behavior of the material, and works autonomously with no additional mechanical or electronic intervention, which largely contrasts with the active monitoring-based self-healing systems [13, 14].

Advantages of Self-Healing Polymers

One of the important characteristics of these materials is that healing can occur at different temperatures. If the material is damaged by the intrusion of a foreign object, healing can be activated at a temperature well below Tg. On the other hand, if the material is damaged by impacts or violent vibrations, the recombination of the chains occurs within the glassy state, and healing happens. Thus, it is possible to

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adjust the healing process by selecting one of several different temperatures. Also, healing does not require one specialized product. Using outright unmodified binder resin in current applications may already exhibit self-healing properties. These self-healing promotional effects are fully exploited only by adding additional modifiers that restrict the glycidyl functionality of the resin by hindered grouping to minimize initial activity. By varying the amounts of these modifiers, the self-healing temperature can be adjusted to suit demands in applications. Self-healing ability is an attractive potential of polymeric materials, very much in demand for practical applications, such as structural applications. Healing solidstate polymeric materials could be achieved by utilizing reversible weak chemical reactions, thermally induced polymerization, and cross-linking after damage events. Attaching a polymeric soft segment to a hardening poly(urea-urethane) elastomer imparts an intrinsic self-healing behavior to the polymeric system and allows repetitive healing, with retained mechanical strength and elasticity. Nanoparticle dispersion provides a recent self-healing strategy in polymeric composites/mixtures. Catalyst-containing polymeric nanoparticles dispersed in epoxy composites enable a healing ability through dynamic polymerization of healing agents upon crack occurrence. Peptide amphiphiles spontaneously self-assemble into nanofibrous hydrogels, which display autonomous relaxation and high water retention. Active peptide motility is enabled by peptide assembly associated with self-construction and self-destruction. This cooperative mechanism provides reversibility that attracts attention for cell culture and proliferation applications $\lceil 15, 16 \rceil$.

Challenges and Limitations

Despite the potential of self-healing polymers, challenges must be addressed for their use in structural applications. These living materials can maintain structural integrity indefinitely, unlike traditional synthetic materials, which degrade over time. Synthetic materials can gain self-healing properties, but current options have limitations. Self-healing polymers fall into two categories: embeddable and intrinsic. The embeddable type uses healing agents that diffuse to repair damage, while intrinsic polymers create new bonds to restore properties quickly after harm. There is a notable absence of self-healing polymers designed for 3D printing, highlighting a gap that must be filled. Recent efforts have focused on composite fibers from polymer sheets, featuring a self-healing epoxy matrix and boron nitride nanotube fillers for improved durability at low frequencies. Although significant strides have been made in self-healing mechanics, material design must align with production techniques for rigorous applications. Techniques like selective laser sintering, especially through 3D printing, are crucial. Exploring polyurethane with varied induction heating setups and using thermoelectric additives can enhance the design of adaptive circuitry. This approach aims to combine dynamic bond adjustments with effective energy delivery solutions [17, 18].

Recent Advances in Research

Recent research into self-healing materials (SHMs) has emphasized biomedical applications, particularly microcapsule-based and HP-TEA/polyaniline materials in polymers and composites. Innovations include SHMs with dynamic covalent and non-covalent healing bonds. The study of materials capable of intrinsic healing has emerged as a pivotal area of interest. Efforts are underway to develop healing systems capable of addressing substantial structural defects, achieving breakthroughs through fusible bonding methods. However, there are no reports of structural self-healing materials needing no external stimulus. Emerging are lotus-type self-healing materials that recover surface defects via droplet or film-forming methods. Composite SHMs integrating elastomers or plastics with shape memory polymers (SMPs) have shown the potential to self-heal and regain mechanical properties upon simple heating. Thermal-induced transitions between liquid, gel, and solid states represent significant research trends. Recent studies have focused on polymer materials like polycaprolactone (PCL), poly(lactic acid), and bio-based polyurethanes, which heal through swelling, diffusion, and melting. These materials exhibit biocompatibility, biodegradability, and cost-effectiveness. Notable advancements have been achieved in damage repair and function restoration without external aid. This paper summarizes recent developments in various SHMs-covering polymers, metals, ceramics, and composites-highlighting their self-healing mechanisms and performance. Examples of self-healing materials and their industrial applications in aerospace, civil, and marine engineering, batteries, electronics, energy generation, and biomedical sectors are also discussed. A deeper grasp of self-healing mechanisms is anticipated to improve the design of new self-healing materials, enhancing healing efficiency and recovery, thus promoting their use in engineering and beyond $\lceil 19, 20 \rceil$.

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Future Directions

Numerous experts in various fields are exploring self-healing materials, leading to remarkable discoveries. The concept of using artificial materials that replicate biological regeneration to prevent aging or malfunction is intriguing. Self-healing is a cooperative process involving chemistry, morphology, and devices, yet our understanding of materials lags behind that of biological organisms. Furthermore, biological systems lack a consolidated repository of ideas. The demand for polymer products has driven the creation of biomimicry self-healing smart polymers, with many advantages possible through fiberreinforced forms. Out-of-autoclave manufacturing of FRP composites eliminates the need for additional curing, bolstering production speed and reducing costs. This self-healing approach is essential for effective healing efficiency, considering the large volume of FRP composites. Proper crack formation should allow efficient healing, although it's challenging for healing agents to penetrate microcracks. Newly developed 3D printing techniques for self-healing materials could decelerate crack propagation, enhancing longevity, but progress appears slow. Future applications of self-healing polymers will vary spatially, primarily benefiting structural components like buildings and marine artefacts. These materials may support reconstruction adaptations for evolving lifestyles. However, they could be viewed as damage mitigation technologies, limiting operational design. Repairing artefacts at high costs, while risking frustration and inefficiency, would be unwise. Integrating the self-healing method with other techniques could enable structural composites to autonomously recover in hostile environments. Exploring complex biomimetic micro-structures could facilitate effective, real-time repairs without requiring external interventions [21, 22].

CASE STUDIES

Photo-Finishing of Self-Healing Automotive Plastics. Over the last decade, self-healing polymers have gained considerable attention. This is mainly due to their potential for relieving some of the downsides of certain technology trends, such as mobile touch-screens being scratched after daily use, and cameras' surfaces being eroded by environmental stresses, especially in urban environments. Depending on the composition of the poly-material (partly composites), self-healing may occur in various ways. The main processes are: (1) restoring the original chemistry of a damaged polymer via an on-parabole reaction, (2) flow of the material's chain, restoring its dimensions, (3) fusion of the entire surfaces using heat or laser. The recovery time ranges from less than one second (laser/microwave), to days (chemistry flow). Selfhealing technology offers solutions by conforming poly-materials to the properties of healing and recovery. For example, high-glass transition temperature poly-multiparous acrylates (PMMA) can be used for restoring (attaining glass chemical and optical properties) car scratches after minor damage using yellow rust. Tough, flexible poly-isobutylene (PIB) can be used to swiftly restore shapes and detect small scratches in clear or tinted lacquer or plastic-coated surfaces. An experimental coating was developed for protecting plastic car parts. The coating includes self-healing UV absorbers that darken after long UV exposure. The healing components are nanometer-sized soft materials, capable of swelling and allowing slower release of healing agents from their pores. Self-healing clearcoats for protecting plastic automotive parts were developed. The coats rely on Michael's addition condensation/cleavage. The technology is based on pre-polymers for curing with a co-monomer, maintaining high gloss and transparency even when curing. The resulting departing polyurethane can achieve a high 12H hardness value. One of the major challenges is reabsorbing water and oils, due to the hydrophilicity of putative healing network formation (creeping polysaccharides). Coating loss via ablation is an issue for automotive clearcoats, accelerated in a 2500-cycle dirt/dry cleaning experiment using car washing solutions every few thousand miles [23, 24].

CONCLUSION

Self-healing polymers have emerged as a transformative innovation in the field of smart materials, offering new paradigms for durability, resilience, and sustainability in structural engineering. The ability to autonomously detect and repair damage reduces maintenance needs, extends service life, and enhances safety across diverse sectors, from infrastructure to aerospace. Intrinsic healing mechanisms, reliant on reversible molecular interactions, and extrinsic strategies, involving embedded healing agents, both offer unique pathways for material self-repair. However, challenges such as improving healing under extreme environmental conditions, integrating with modern manufacturing processes, and ensuring cost-effective scalability must still be overcome. Continued interdisciplinary research and technological development are essential to unlock the full potential of self-healing materials, steering the future towards smarter, longer-lasting, and more sustainable built environments.

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Page | 39

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