

Research Progress in Photothermal Self-Healing Coatings

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KEYWORDS

ABSTRACT

Intrinsic self-healing;

*Photothermally
triggered self-healing;*

*Photo-responsive
materials*

Coatings serve as a critical barrier to protect substrates from corrosion, wear, and environmental degradation. Their ability to repair damage directly influences the service life of materials. Traditional coatings lack self-healing capabilities, making them susceptible to rapid performance degradation from micro-damage. Inspired by biological self-healing mechanisms, self-healing coatings have been developed. Among these, photothermal self-healing coatings have gained significant research interest due to their remote, precise, and controllable repair characteristics. This review systematically summarizes the healing mechanisms, material system design, and research progress of photothermal self-healing coatings. It focuses on the roles of carbon-based materials, MXenes, organic materials, and nanoparticles in photothermal conversion and repair behavior. The application prospects in aerospace, marine engineering, electronic devices, and other fields are discussed, along with future challenges. The advancement of photothermal self-healing coatings provides important theoretical support and technical pathways for designing high-performance intelligent protective materials.

INTRODUCTION

Coating technology is the primary defense for protecting materials against corrosion, wear, biofouling, and various environmental factors. The performance of coatings directly determines the service life and reliability of substrates. However, traditional coatings are essentially static, passive protection systems. Micro-cracks, surface scratches, and local delamination inevitably occur during processing, transportation, and long-term service [1]. These micro-damages not only become entry points for corrosive media or stress concentration, leading to exponential decay in protective performance, but their repair is often challenging, sometimes requiring operational shutdowns and resulting in significant economic costs and safety risks.

Inspired by the remarkable self-healing abilities in nature, materials scientists proposed the concept of smart self-healing materials [2]. Self-healing coatings, as pioneers of this concept, aim to mimic biological repair mechanisms, enabling materials to autonomously or externally stimulated recover their structure and function after damage [3].

Intrinsic self-healing relies on reversible chemical bonds

within the polymer network, such as dynamic covalent bonds or non-covalent interactions, enabling repair under external stimuli and offering potential for multiple repair cycles.

Among the various damage-triggering methods, light energy has shown great potential due to its cleanness, easy control, and capability for remote and precise application. Particularly, the combined photothermal repair strategy, which converts light energy into thermal energy to activate dynamic reactions, enables non-contact, spatiotemporally controllable smart repair, opening new pathways for the practical application of self-healing coatings.

Therefore, in-depth research on photothermal self-healing coatings represents not only fundamental scientific exploration inspired by nature but also addresses the urgent need for long-term protection solutions in demanding service environments. This review aims to summarize research progress in photothermal self-healing coatings, systematically explain their core healing mechanisms and key material design strategies, and discuss current

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Received date: November 02, 2025; Revised manuscript received date: November 15, 2025; Accepted date: November, 2025; Online publication date: November 30, 2025.

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challenges and future trends.

1. Intrinsic Self-Healing

Intrinsic self-healing coatings incorporate dynamic reversible bonds into the polymer network, enabling molecular-level damage repair under external stimuli. The healing capability originates from the internal reversible cross-linked network, which can restore integrity and functionality through bond breakage and reformation. Based on the dynamic bond type, healing mechanisms primarily involve dynamic covalent bonds and non-covalent bonds [4,5]. Combining different dynamic bonds through multi-mechanism synergistic strategies is key to achieving high-performance self-healing [6].

1.1. Healing Mechanisms of Dynamic Covalent Bonds

Dynamic covalent bonds combine the stability of covalent bonds with the reversibility of dynamic reactions, providing a robust foundation for coating repair [7].

Disulfide bond systems enable network reconfiguration through reversible disulfide exchange or redox reactions. Studies show that synergizing disulfide bonds with other dynamic bonds significantly enhances performance [8]. Chen et al. constructed a dynamic supramolecular network containing disulfide metathesis and multiple hydrogen bonds, enabling precise tuning of material properties: disulfide bonds drove the self-healing process, while strong and weak hydrogen bonds contributed to mechanical strength and energy dissipation, respectively, markedly improving toughness [9].

Diels-Alder (DA) bond systems are based on the reversible cycloaddition between furan and maleimide, offering excellent thermal reversibility [10]. Truong et al. developed a DA-crosslinked polymer network that also benefited from hydrogen bonds provided by amide groups in the backbone. This DA-hydrogen bond synergy granted good mechanical properties and achieved approximately 85% strength recovery after cutting [11].

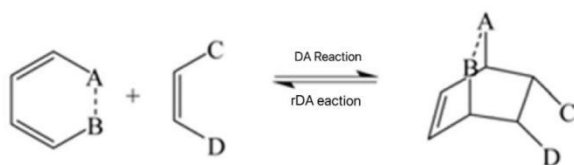


Fig.1. Diels-Alder reversible reaction mechanism

Imine bond systems, formed by condensation of aldehydes and amines, undergo dynamic exchange triggered by heat, pH, etc [12]. Yang et al. synthesized a self-healing Polydimethylsiloxane (PDMS) elastomer containing both strong ureido-based hydrogen bonds and dynamic imine bonds. Their synergy enabled 95% healing efficiency after 24 hours at room temperature, with the conjugated structure enhancing stability in humid environments [13].

1.2. Synergistic Effects of Non-Covalent Bonds

Non-covalent bonds, with their fast dynamic reorganization, are crucial in the initial stages of self-healing.

Hydrogen bonds, as fast, reversible physical cross-links, enable preliminary damage repair at room temperature. Peng et al. built a cross-linked network via multiple hydrogen bonds between tannic acid (TA) and linear waterborne polyurethane (WPU), producing self-curing cationic WPU/TAx composites. These achieved complete interfacial healing at room temperature, but required 7 days. At 50° C, healing time reduced to 3 hours, showing heat accelerates hydrogen bond reorganization [14].

Metal-ligand coordination bonds have moderate bond energy and can act as energy dissipation sites, enhancing toughness. Cui et al. introduced Zn^{2+} -imidazole coordination bonds into a supramolecular network combined with weaker urea hydrogen bonds, producing a self-healing material with excellent mechanical properties and high healing efficiency, where coordination bonds primarily provided tunable mechanical performance [15].

Despite their advantages, intrinsic self-healing materials face challenges for practical application: Firstly, dynamic covalent bond exchange often requires high activation energy, leading to slow healing rates unsuitable for rapid repair. Secondly, most systems need external energy input (e.g., heat) and exhibit limited healing efficiency at room temperature. These limitations restrict their use in applications requiring fast response and high mechanical performance [16].

2. Photothermal Self-Healing Coatings

A key technical bottleneck currently faced by self-healing polymer materials is the trade-off between mechanical properties and healing conditions: systems with excellent mechanical strength often require high healing temperatures, whereas those capable of rapid repair under mild conditions

typically exhibit insufficient mechanical performance. This trade-off severely limits the practical engineering application of self-healing materials. Therefore, developing new polymer materials that combine excellent mechanical properties with rapid repair capability under mild conditions has become an important research direction urgently requiring a breakthrough, with significant practical application value [17].

Based on the external energy source, self-healing polymers can be categorized into photoinitiated, thermally initiated, etc. Photoinitiated systems exploit light's remote, localized control for precise, energy-efficient repair, making them widely studied [18-20]. Different initiation methods offer options for various applications, expanding design possibilities. Optimizing photothermal self-healing coatings hinges on careful selection and integration of photothermal materials. Current research introduces photothermal conversion agents like graphene, carbon nanotubes, MXenes, polydopamine, and Fe_3O_4 nanoparticles into coatings, enabling self-healing under NIR irradiation. The content, dispersion stability, and interfacial interaction with the polymer matrix directly affect photothermal conversion and healing efficiency [10].

2.1. Carbon-Based Materials

Graphene and carbon nanotubes, as typical carbon nanomaterials, demonstrate significant application value in the field of functional coatings due to their excellent light absorption capacity, high thermal conductivity, large specific surface area, and outstanding mechanical properties. Incorporating them into organic coating systems not only enables rapid and precise repair of damaged areas through the photothermal conversion effect but also significantly enhances the comprehensive corrosion resistance of the coating through their physical barrier effect and electrochemical activity [21]. Cai et al. successfully developed nanocomposites with rapid photothermal responsive self-healing capability, excellent mechanical properties, and efficient recyclability by integrating reversible DA covalent bonds with graphene into an epoxy resin system. In this system, graphene not only acts as a photothermal conversion medium significantly accelerating the repair process but also functions to enhance mechanical properties [22]. Li et al. explored the use of carbon nanotubes (MWCNT) for photothermal conversion to trigger

the DA thermal reversible reaction in epoxy resin, analyzing the effects of carbon nanotube content (mass fraction 0.2%~2%), the distance from the near-infrared light source to the sample, and laser power intensity on the repair effectiveness, achieving local high-precision and high-efficiency repair while showing no significant impact on undamaged areas [23].

2.2. MXene Materials

MXenes are a class of two-dimensional inorganic compounds comprising atomically thin layers of transition metal carbides, nitrides, or carbonitrides. They exhibit strong absorption of light and microwaves. Surface hydroxyl or terminal oxygen groups contribute to their metallic conductivity [24]. MXenes' excellent photothermal conversion promotes polymer chain mobility and hydrogen bond reorganization, enhancing self-healing performance. Wang et al. prepared MXene with strong photothermal conversion via etching/exfoliation and fabricated PU and MXene/PU coatings. Scratched MXene/PU coatings healed faster under 808 nm laser irradiation, attributed to MXene's layered structure extending the diffusion path for corrosive media, allowing ongoing crack repair and slowing penetration [25]. Fang et al. found that the corrosion resistance of MXene-composite coatings depends on MXene-polymer interactions and MXene's spatial distribution. Surface functionalization and compositing improve interfacial adhesion, dispersion, and compatibility. Flow-induced or electrophoretic deposition can align MXene, enhancing its barrier effect against corrosive ions [26].

2.3. Organic Materials

Common organic photothermal fillers include Aniline Black (AB), which exhibits good light absorption capacity within both the visible and near-infrared light ranges. Fang et al. adjusted the ratio of monoamine and diamine curing agents, gradually replacing the diamine m-xylylenediamine (MXDA) with the monoamine 4-(heptadecafluorooctyl) aniline (HFOA). This reduction in the epoxy network's glass transition temperature (T_g) and cross-linking density enabled thermal-induced repair above the T_g temperature, based on crack closure and molecular chain diffusion/re-entanglement mechanisms. Subsequently, Aniline Black (AB) was introduced as a photothermal conversion compound. Utilizing the indirectly generated

heat, repair under focused sunlight (with an optical density of $0.7\sim 0.9\text{ W/cm}^2$) was achieved [27].

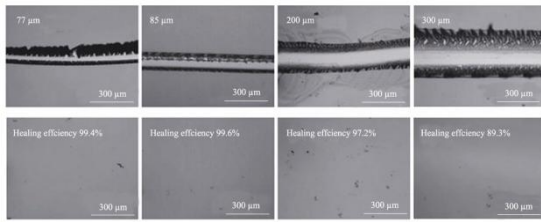


Fig.2.Morphology of Aniline Black coatings with different scratch widths after 10 min sunlight irradiation

2.4.Nanomaterials

Nano- Fe_3O_4 particles offer advantages such as low cost, ease of preparation, and good photothermal conversion performance, making them commonly used nanomaterials for photothermal self-healing [28-30]. Chen et al. prepared Diels-Alder thermally reversible self-healing polyurethane materials modified with nano- Fe_3O_4 . An appropriate amount of Fe_3O_4 nanoparticles (optimal mass fraction 0.3%) can significantly enhance the mechanical properties of the material. The incorporation of Fe_3O_4 not only improved the thermal healing efficiency of the material but also endowed it with near-infrared light-responsive self-healing capability. Compared with traditional thermal healing, near-infrared light-triggered healing demonstrated faster speed and higher efficiency, with the material maintaining excellent repeatable healing performance. After three damage-healing cycles, it still retained a healing efficiency of 55.34% [31].

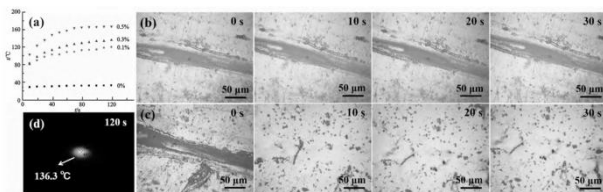


Fig.3.(a) Temperature changes of PU-DA- Fe_3O_4 with different mass fractions of Fe_3O_4 nanoparticles exposed to 808 nm NIR; POM images of cracks in (b) PU-DA and (c) PU-DA- Fe_3O_4 films upon exposed to 808 nm NIR; (d) thermal image of PU-DA- Fe_3O_4 (0.3%) exposed to 808 nm NIR

3.Applications and Future Outlook

3.1.Application Areas

Photothermal self-healing coatings, with their unique remote, localized, controllable repair, are transitioning from lab research to targeted engineering applications. Their core value lies in enabling in-situ repair of hard-to-access or sensitive components, enhancing service reliability and lifespan [32]. Key application prospects include:

Aerospace: Aircraft skins, composites, and engine components are prone to micro-cracks from vibration, fatigue, or impact. Applying these coatings allows rapid crack repair under NIR light during maintenance, preventing damage propagation, extending inspection intervals, and improving safety [33].

Marine Anticorrosion: Ships, offshore platforms, and pipelines face harsh, corrosive seawater environments. Traditional coatings, once scratched, allow rapid corrosion spread. Photothermal self-healing coatings on these structures enable localized NIR irradiation after damage, triggering healing agent release or resin flow/re-fusion to 'heal' scratches, restore barrier function, and provide long-term substrate protection [34].

3.2.Future Prospects & Challenges

Despite their potential, photothermal self-healing coatings face several scientific and technical challenges. Future research should focus on:

Performance Balance & Optimization: Most systems still struggle to combine high strength/toughness with fast, efficient healing. Future designs need deeper understanding of dynamic chemistry and photothermal filler synergies. Multi-network structures and multi-mechanism approaches should optimize the balance between mechanics and healing kinetics at molecular/nano scales.

Intelligent & Precise Photothermal Systems: Next-generation coatings should become smarter, incorporating multi-stimuli responsiveness for complex environments. Self-feedback/regulating systems, e.g., using indicators for visual monitoring or feedback-controlled healing agent release, could enable deeper damage repair.

Long-Term Durability & Environmental Adaptability: Coatings serve for years/decades, requiring evaluation of long-term photothermal stability, aging resistance, and retained healing ability under real conditions.

Long-term reliability data is crucial for engineering adoption.

Scalable Fabrication & Cost-Effectiveness: High-performing fillers and complex syntheses are often expensive, hindering large-scale use. Developing low-cost, green synthesis/compositing methods and scalable application techniques is necessary for widespread use.

Looking forward, interdisciplinary convergence of materials science, chemistry, optics, and AI may help overcome current bottlenecks, evolving photothermal self-healing coatings into truly intelligent material systems capable of sensing damage, deciding on, and executing repair, offering revolutionary solutions for long-life, safe operation of critical engineering assets and sustainable development.

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Acknowledgment : The authors gratefully acknowledge the support from the Youth Project of the Qingdao Natural Science Foundation(24-4-4-zrjj-85-jch) and the Open Project of the Coastal Human Settlement Environment Academic Innovation Center at Qingdao University of Technology(CK-2024-0061).