Vietnam Journal of Science and Technology ***64***(3) (2026) 412–427
DOI: https://doi.org/10.15625/2525-2518/23015

REVIEW PAPER

Additive manufacturing of self-healing polymers: A comprehensive review of thermal and photo-activated systems

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Received: 17 February 2025; Accepted for publication: 25 September 2025

Abstract. Self-healing polymer is a polymeric material which, after the occurrence of any form of damage, such as cracks, scratches, or fractures, can independently or externally restore its original integrity and functionality without necessarily replacing the material itself. These polymers represent a transformative approach to enhance the service life and ensure the dependency of the systems, particularly in harsh environments where manual repairs are often impractical. These advanced materials are manufactured using additive manufacturing (AM) techniques that offer precise control over both structural properties and healing functionalities. This review highlights a thorough analysis of AM strategies applied to thermal-activated and photo-activated self-healing polymers and composites, with particular significance on their basic healing mechanisms. It highlights the distinct performance characteristics of these two systems, including activation conditions, healing efficiency, and their suitability for various aerospace applications. By focusing on thermal and photo-activated self-healing mechanisms, the review underscores their potential to reduce maintenance demands, enhance operational safety, and ensure long-term structural integrity. Furthermore, it identifies key challenges and future research opportunities, offering valuable insights for researchers and engineers working to integrate these innovative materials into next-generation aerospace systems.

Keywords: 3D printing, vat photo polymerization, extrusion.

Classification numbers: 2.9.3.

1. INTRODUCTION

Materials science is an extremely intriguing and demanding field, which has advanced from the discovery and utilization of natural materials to the present era of designing and using advanced artificial materials that are more beneficial for production and life, including self-healing materials [1–3]. These materials exhibit excellent mechanical properties, thermal stability, and have high molecular weight, which perform biological self-healing ability to repair micro-cracks through changes in chemical bond or polymerization of stored monomers and maintain the structural durability [4, 5]. In comparison to conventional materials that are mechanically eroded, deteriorated, or degraded and often need to be replaced and repaired, the self-healing materials improve efficiency, durability, sustainability, and lower overall expenditure in various climatic conditions and working settings [6].

Self-healing materials cover micro damage, including cracks, which change the electrical, thermal, and acoustic characteristics that render the material useless [7, 8]. These materials can either work independently or as non-independent structures under certain circumstances. They are deployed in different applications, viz. coatings, ceramics, and concrete, each of which possesses its own self-repairing methods and processes which use healing agents at the time of production [9, 10]. Additive manufacturing (AM) has greatly revolutionized the area by allowing the deposition of healing materials, viz., microcapsules or photo-sensitive resins, using extrusion techniques and vat photo polymerization. These methods increase efficiency, but problems such as volumetric shrinkage and reduction in mechanical properties still occur. However, measures such as employing dynamic bonds can alleviate polymerization stresses and enhance auto-healing characteristics [11, 12].

This dynamic reversible process of bond dissociation and re-formation allows for the release of shrinkage stress that is generated during the polymerization process, hence volume shrinkage is reduced [13, 14]. Self-healing is a revolutionary concept in materials science, where, apart from providing functionality to the materials used, they are also made to self-heal or have an indefinite life span. These materials inherently self-heal after being damaged, which can be a problem in aerospace, automotive, and consumer electronics industries [15]. The steady-state healing procedures, specifically the ones that rely upon thermally reversible Diels Alder, are efficient in healing repeatedly, which makes these self-healing polymers perfect for industries which demand improved performance, viz., aerospace and automobile [16–19]. The Diels-Alder reaction is a type of [4+2] cycloaddition in organic chemistry, where a conjugated diene reacts with a dienophile, which is usually an alkene or an alkyne, to produce a six-membered ring. It can also be mended by UV-induced self-healing, where the cracked areas are repaired by incorporating photo-sensitive polymers, which are suitable for self-healing [20–22].

Self-healing hydrogels are suitable for biomedical uses due to its biocompatibility and biologically-inspired functionality relevant to issues in tissue regeneration and soft robotics. These materials also have great prospects in the aerospace industry, in which such conditions require materials with the ability of instantaneous self-healing [23–25]. Nevertheless, various issues are still present with regard to increasing rates of healing, mechanical stability, as well as the overall feasibility of the project. To increase the durability of self-healing systems, new chemistries and AM driven approaches are being studied by researchers for use in the industrial environments [26]. This review briefs about additive manufacturing of self-healing polymers as well as their self-healing mechanisms. Further, the paper consolidates limitations, challenges and future research opportunities of additive manufacturing of self-healing thermal and photo-activated systems.

2. MATERIALS AND METHODS

The two major techniques followed in AM for 3D printing of self-healing materials are extrusion-based technique and vat photo polymerization. The main extrusion-based method, used for processing self-healing materials, includes thermosetting polymers that offer self-healing, as reported by Roppolo *et al.* [27] and Yuan *et al.* [28]. This method can be used for building the sustainable and reprocessable structures as recommended by Garnero [29] in 2020. Similarly, vat photo polymerization outlined by Caprioli *et al.* [30] demands light-cured resins to create self-healing hydrogels using precise prints. Stereolithography (SLA), a development in vat photo polymerization, is used for the manufacture of intricate self-healing composites, as reported by Sanders *et al*. [31]. More recently, Andru *et al*. [32] have shown progress in incorporating self-healing capabilities into 3D-printed materials, and Liu *et al*. [33] reported that UV-curing was effective for using stimuli-responsive self-healing polymers. In the conclusion of their review, Shafranek *et al.* [34] went further in identifying related future research directions for engaging the stimuli-responsive materials in AM at a greater level so as to expand the scope of self-healing concepts.

2.1. Extrusion based technique

Material extrusion based on 3D printing technology involves the selective deposition of one or more materials on a build platform in which the materials are selectively deposited through a nozzle or orifice attached to a movable print head that travels a predetermined path in the fabrication of each layer [35]. Accordingly, in this 3D printing technology, the materials are deposited in the form of filaments in a line-by-line and layer-by-layer fashion, which eventually leads to the fabrication of a 3D object at specific locations. This 3D printing technology is the most popular and accepted due to its versatility and cost-effectiveness. This technology offers the possibility of printing a wide variety of 3D polymer materials, including thermoplastics, pastes, and polymer solutions, and also offers the possibility of printing multiple materials in a single step [36, 37]. The self-healing polymers are fabricated using fused filament fabrication and direct ink writing (DIW) technologies, as shown in Figure 1. Fused filament fabrication (FFF), also known as fused deposition modelling, involves the melting and subsequent extrusion of a thermoplastic filament through a heated nozzle attached to a print head. In contrast, DIW utilizes viscous or viscoelastic non-Newtonian fluids, which are extruded under room temperature so as to prevent deformation which is caused by melting of the polymers [38]. Nevertheless, as distinguished from FFF, which uses cooling for solidification, DIW involves such methods as pH changes, solution immersion, thermal curing, or photo polymerization, allowing for better flexibility in material processing [39–41].

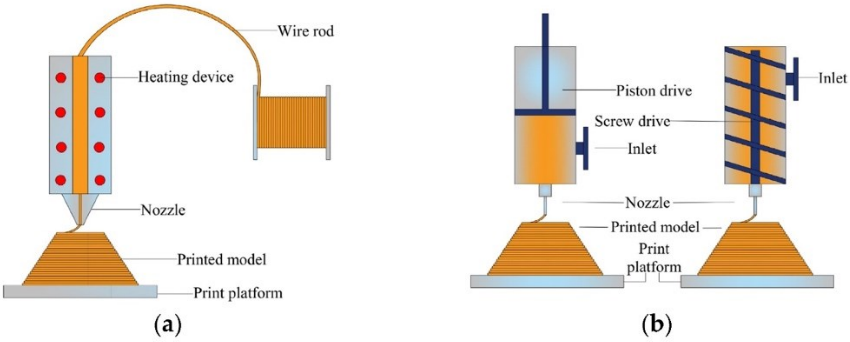

Figure 1. Schematic illustration of extrusion-based additive manufacturing process. a) Fused filament fabrication (FFF) and b) direct-ink writing (DIW). (Extracted and reproduced from [42] under the Terms of the CC-BY 4.0 License, https://creativecommons.org/licenses/by/4.0/).

2.2. Vat photo polymerization technique

Light-triggered additive manufacturing techniques involve spatially controlled light irradiation to solidify a liquid resin contained within a reservoir at specific locations corresponding to a cross-section of the final three-dimensional object [43]. Patterned exposure of ultraviolet or visible light initiates a light-sensitive compound, also known as a photoinitiator, that must be incorporated into the resin formulation [44, 45]. Upon absorption of light, the photoinitiator initiates polymerization by forming reactive species or free radicals that react to form a solid, covalently cross-linked material [46]. A typical resin formulation includes one or more photoactive functional groups, often located along the polymer backbone or at chain ends, to enable cross-linking reactions during curing [47, 48]. Vat photo polymerization techniques can involve linear, planar, or volumetric build techniques depending on the spatial orientation of resin solidification [49]. Stereolithography involves the use of lines laser, while the projection-based approaches, such as DLP, use bidimensional patterns to irradiate an entire layer cross-section at once [50]. Next, continuous liquid interface production (CLIP) cuts build time in half with layerless manufacturing in a bottom-up fashion [51]. Although volumetric 3D printing, which utilizes holographic light patterns directed into a resin vat that rotates to induce rapid polymerization, has not been utilized with self-healing polymers, as discussed in the review [52]. The various methods of vat photo polymerization techniques are highlighted in Table 1.

Table 1. Comparison of common UV-curing 3D printing techniques [18, 19, 33].

Process	Working mechanism	Technique of UV curing	Merits	Demerits	Suitability	Refs
SLA	Laser-beam single-point printing	Free radical and hybrid curing	Suitable for bigger devices	Reduced speed of curing	Automobile mold, dentistry	[53]
LCD	Liquid crystal imaging printing	Free radical curing	Increased curing speed, reduced cost	Reduced operational life span	Jewellery, manufacturing of mold	[53]
TPP	Dual laser beam printing	Free radical curing	Superior precision	High-cost and intricate process	Scientific research, microelectronic art,	[54]
DLP	Projection printing	Free radical curing	Quick curing, superior precision	Produces devices of smaller size	Medical care, jewellery	[54]
CLIP	Projective continuous printing	Free radical and thermocuring	Quick curing	High cost equipment and resin	Cars, sports	[54]
MJP	Multi-nozzle printing	Free radical and hybrid curing	Superior precision with colours	High cost equipment	Medical care, jewellery, consumer goods	[54]

SLA - Stereolithography, LCD - Liquid crystal display, TPP - Two photon polymerization, DLP - digital light projection, CLIP - Continuous liquid interface production, MJP - Multi jet printing.

2.3. Self-healing mechanism

The two widely accepted mechanisms for the repair of damage are thermally activated and photo-activated healing. In the case of thermally activated healing, healing agents are released due to heat, which flow into cracks in the material to heal it. The photo-activated healing follows activation through a chemical reaction caused by exposure to light - mostly UV light - and may activate healing agents or polymerization, bringing the material back to its healthy state. Both mechanisms increase material longevity by giving the ability to repair without human involvement.

2.3.1. Thermal activation

The thermal based mending mechanism is triggered when the self-healing material is put to heating which initiates healing. In this mechanism, the method usually adopts reversible chemical reactions, such as a Diels-Alder (DA) reaction, with which everybody familiar knows that it can interconvert reversibly under heat control. Liu and Chuo [55] stated that various polymers based on DA chemistry may be able to sustain multiple healing through heating, so that such polymers are amenable to the condition found in aerospace components, which are exposed to thermal cycling as shown in Table 2.

Table 2. Self-healing materials and their aspects in thermal-based mending.

Material	Mechanism	Healing temperature (°C)	Healing efficiency (%)	Refs
Epoxy resin	Thermally reversible Diels-Alder reaction	120	90	[18, 19]
Polyurethane	Thermally triggered shape memory effect	80	95	[21, 22]
Polydimethylsiloxane (PDMS)	Thermal remending via chain diffusion	60	85	[24, 26]
Diels-Alder-based polymers	Reversible covalent bonding	110	88	[27, 29]
Polyimide	Thermal re-crosslinking	150	92	[56, 57]

Engel and Kickelbick [56] highlighted that thermally mendable materials can self-heal micro cracks above a certain temperature, restoring strength and structure, especially in hard-to-recycle thermosetting polymers. Zhang *et al.* [57] also focused on the in-situ introduction of thermal self-healing mechanisms into thermosetting matrices as a way towards recyclability of composites. Paladugu *et al*. [58] described that thermal healing bestows enhancement of the mechanical performance of metal matrix composites that are highly beneficial in aerospace structures where high strength is needed. However, repeated thermal cycles may cause degradation, limiting long-term performance, as observed by Ekeocha *et al*. [59]. Figures 2 and 3 show the characterization of self healing materials.

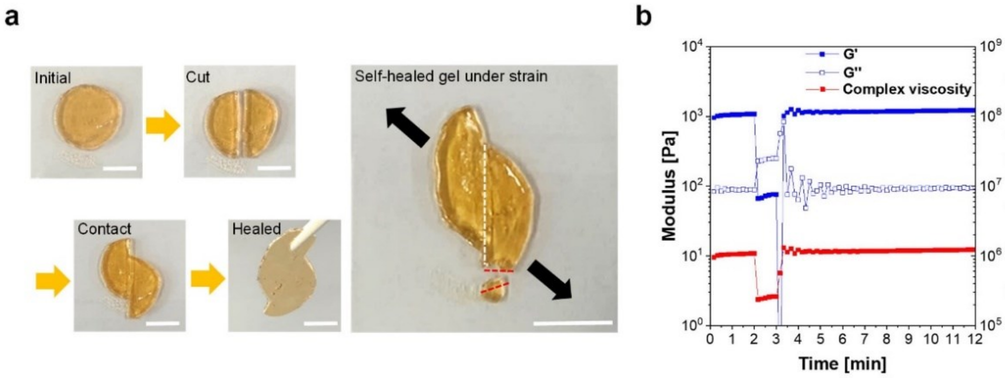

Figure 2. Characterization of the self-healing behaviour of the new PAA-based self-healing hydrogel. (a) Cut-and-contact self-healing experiment (scale bar: 10 mm). (b) Time-dependent recovery experiment of the hydrogel under different conditions of shear strain; 5 % strain for the first 2 min, followed by 400 % strain for 1 min, and 5 % strain for 10 min (at 1 rad/s). (Reproduced from [60] under the Terms of the CC-BY 4.0 License, https://creativecommons.org/licenses/by/4.0/).

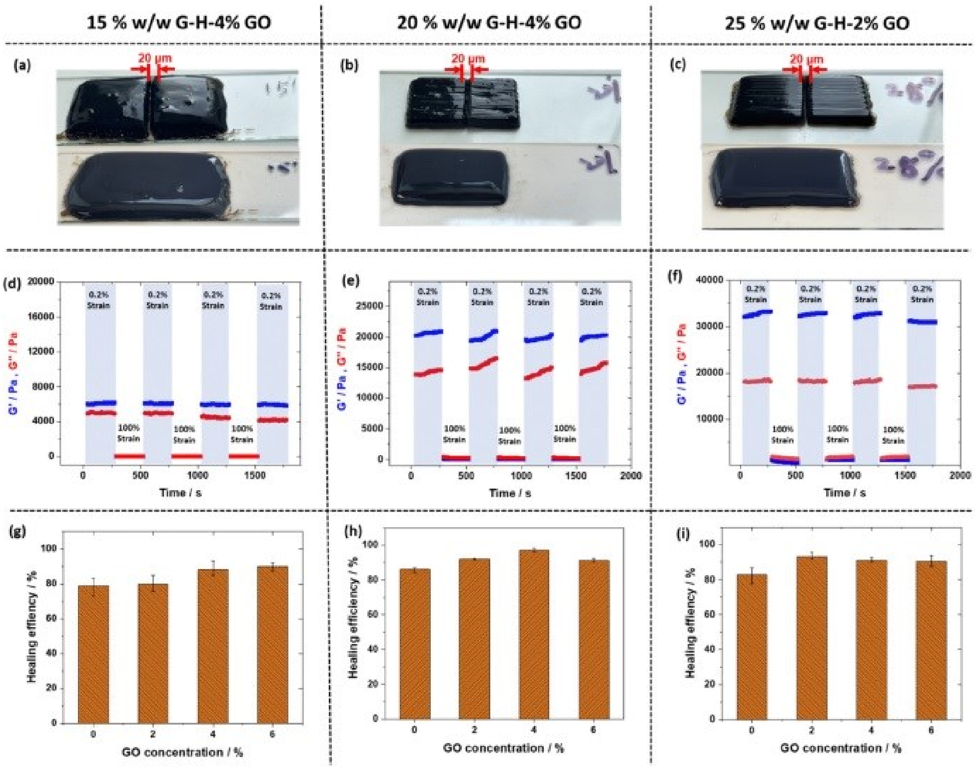

Figure 3. (a)–(c) Photographic images of GO nanocomposite worm gels after 3D printing. Panels (d), (e), and (f) in the above figures represent the oscillatory rheological recovery tests of the GO nanocomposite worm gels utilising continuous strain sweep tests with the application of alternating strain amplitudes (γ = 0.2 % and 100 %) at 25 °C and an angular frequency of 10 rad/s^{-1}. Panels (g), (h), and (i) in the above figures represent the healing efficiencies of the GO nanocomposite worm gels utilising the oscillatory rheology recovery tests. (Reproduced from [61] under the Terms of the CC-BY 3.0 unported License, https://creativecommons.org/licenses/by/3.0/).

2.3.2. Photo activation

Photo-based mending mechanisms make use of light, often within the UV range, to initiate healing. In this context, photo-based mechanisms show excellent utility where localized healing is sought after, particularly without direct heating. Ling *et al*. [62] isolated photochemically remendable polymers that utilized photo-induced reactions for the healing of cracking as well as surface damage for them to be useful in coatings and surface applications. It is possible, as highlighted by Guimard *et al*. [63], that the photoresponsive systems may be included, which could be controlled or activated from a distance to heal precisely in sensitive devices, such as electronic or medical implants, as shown in Table 3. Since the soft electronics need to retain flexibility besides conductivity, photo-activated healing is highly important. Kang *et al.* [64] demonstrated that conductive pathways in a soft electronic material can be healed with UV light - a broad-spectrum application for wearable technologies. However, photo-based self-healing has some limitations, especially concerning penetration depth by light, which limits the healing to the surface. Jiang *et al*. [65] also mentioned that photo-based systems are well suitable with thin films and coatings, but not necessarily suitable for bulk materials or structural applications, whereby the damage is internal. Long exposure to photo-degradation can reduce the extended duration usage of materials, according to Idumah [66].

Table 3. Self-healing materials with their aspects in photo-based mending.

Material	Mechanism	Wavelength (nm)	Healing time (min)	Healing efficiency (%)	Refs
Polyethylene glycol (PEG)	Light-induced disulfide exchange	450	15	87	[62]
Polysiloxane-based elastomers	Photo-reversible [2+2] cycloaddition	320	10	85	[62, 63]
Hydrogel composites	Photo-induced crosslinking	405	8	90	[63, 64]
Chitosan-based hydrogels	Light-activated Schiff base formation	380	6	84	[65, 66]
Acrylate elastomers	Photo-triggered thiolene reactions	450	7	93	[67, 68]

2.3.3. Comparison between thermal and photo-based mending

Both mending techniques have unique advantages and disadvantages when it comes for comparing their self-healing mechanisms. The thermal-based mending is highly effective in terms of bulk materials and structural composite materials; however, it has been greatly successful for applications that can withstand or require elevated temperatures. Such applications include aerospace structures and automotive components. For instance, Balazs [67] proposed the effectiveness of thermally responsive polymers of high stress environments of importance, wherein the structural integrity needs to be held. This is somewhat less practical, however, in temperature-sensitive applications, since it requires external sources of heat and possible thermal degradations [68, 69].

Photogenerated-based mending is robust in localized, non-invasive self-healing with excellent applications onto surfaces and even delicate electronics. According to Cho *et al.* [70], photo-healing polymers can rapidly mend minor scratches and damage in coatings that can, in turn, improve protection on surfaces. This mechanism is of special interest for those applications, where fast and demandable repair is needed, such as for protective coatings on optical devices and sensors [71]. However, low penetration depth of light and sensitivity to environmental factors, such as UV exposure, limit the larger application of photo-activated healing in structural applications. As Wool [72] pointed out, such healing mechanisms, although proved effective, depend on the requirements for the application: repairing bulk versus surface, specific conditions of environmental exposure, and compatibility of materials. However, while giving deeper, more all-round structural repair, thermal-based mending is less precise and less easy to apply than photo-based mending.

According to Hager *et al.* [73], future work on this research will be on hybrid systems for the development, thus giving both mechanisms more versatile self-healing capabilities. Wu *et al.* [74] and Blaiszik *et al.* [75] mentioned that the self-healing polymers had been developed for various types of damage and either thermal or light activation of a healing response. Thermally activated systems are useful for structural composites as they induce reversible reactions by thermal action, thus restoring the mechanical integrity, although such systems suffer from difficulties in terms of thermal degradation [76–78]. Similarly, the photo-activated self-healing was recommended by Yang and Urban [79] and has localized and on-demand UV-induced repairs. This is well tailor-made for surface coatings and electronic components. Dynamic polymer systems can respond to stimuli of light and therefore cause fast and targeted healing [79–82]. These approaches are specifically advantageous in applications where precision is coupled with no thermal exposure. In the case of thermal methods, deep and bulk repairs are favoured; however, the photo-based techniques are appropriate for non-invasive, surface-level fixes [83, 84]. Recent study by Thakur and Kessler [85] suggests that hybrid systems combining both mechanisms can improve material toughness and flexibility.

2.4. Aerospace applications

The Self-healing polymers are generally used in protective coatings, electronics and flexible devices, aerospace and automotive composites, biomedical and healthcare materials, energy storage devices, structural materials and construction, soft robotics and smart materials. In the aerospace industry, the features of self-healing materials are steadily finding their way as adjustments to the durable and safe at construction facets. These progressive composites have features that overcome the obstacle characterizing normal avionics, such as matrix cracking and fibre-matrix debonding [86]. It involves the application of structural health monitoring (SHM) with fibrous composite materials that come integrated with healing agents. The self-repair materials with the help of biological analogy demonstrate the capacity to mend themselves, restoring the strength of aerospace structures, thereby greatly enhancing the life span [87].

Such systems, especially the extrinsic ones such as the microcapsules and hollow fibres, may self-heal impact loads that generate micro-cracks in aerospace parts, increasing reliability of the elements [88, 89]. For instance, carbon-based nanofiller reinforced nanocomposites exhibited an ideal damage recovery mechanism involving matrix-nanofiller interface interactions and thus, appropriate for aerospace applications [88, 89]. Also, the self-healing materials are incorporated into aerospace fuel tanks to reduce the effects of impacts and also for safety reasons. The studies of poly-ethylene-methacrylic acid copolymer ionomer-based system have proved that this material is capable of healing projectile punctures with pressures [90]. These advancements not only contribute to leakage prevention but also reduce the maintenance frequency, thereby lowering operating costs [91].

Diels-Alder (DA) chemistry has been integrated with epoxy resins used in aerospace composite materials to make the material self-healing under thermal action. This makes it possible to have multiple self-repair cycles necessary, especially for the aerospace applications where service reliability is often required during the long-lasting unlettered missions [92, 93]. Additionally, research is being carried out on UV sensitive self-healing coatings to shield aircraft structural surfaces against UV and other conditions that cause deterioration to enhance the life of exterior parts [94, 95]. In general, the deployment of self-healing systems for use in aerospace applications will lead to more durability, less maintenance, and higher safety, concurring with the aerospace industry's current trend toward sustainability and cost reduction [96].

3. LIMITATIONS AND CHALLENGES

There are many challenges in developing self-healing polymers by additive manufacturing, particularly in the aerospace industry, a high-risk field. Healing functionalities are relatively complex when introduced to polymers, as they could potentially interfere with both the mechanical strength of the material and its manufacturability.

1. According to Yang *et al.* [97] in 2015, the efficiency of self-healing significantly depends on the ability to regulate physical and chemical properties of polymers, which complicates large-

scale production processes. Meanwhile, healing efficiency against mechanical performance is still an issue because the incorporation of such healing agents is generally at the cost of compromising in structural integrity, and their use is typically limited to non-load-bearing applications, according to Wen *et al.* [98].

2. Healing agents are normally microencapsulated, though they may have shortcomings coming from long-term performance. The healing agents are mostly monolithic, which thus hinders their durability [99]. Polymers in contrast to metals have modular tail and head; that is why the perception of self-healing is problematic for the metallic elements [100]. Supramolecular forces allow for self-healing, but it comes with penalties: [101] decreased mechanical stability that is unfavourable for structural aerospace components. For instance, temperature and humidity also affect the healing performance of the materials and consequently reduces realism when tested under real aerospace climates [102].

3. The use of such structures in industries still poses a challenge because large-scale synthesis of self-healing materials is challenging [103]. Furthermore, self-healing soft electronics normally suffer from adhesion and flexibility problems that hinder their accommodation into adaptive aerospace platforms [104, 105]. Brochu *et al.* [106] reported that such a challenge has limitations in ensuring biocompatibility and healing efficiency under physiological conditions, so important that they denote a broader challenge related to the utilization of self-healing materials for life-critical aerospace systems.

4. The research by Lin *et al.* [107] on the near-infrared induced self-healing of polyurethane/graphene nanocomposites mentioned that although such conditions are often sensitive, light-induced healing has a precision advantage over other methods of healing, and the activation might fail to be met considering the vast variety of situations encountered in aerospace scenarios. Such application restrictions due to this sensitivity to external stimulation seriously limit photo-activated self-healing systems to less versatile application in a typical aerospace environment where all the components are not always capable of accessing the prescribed light exposure.

5. Overall, self-healing materials have bright prospects in enhancing durability and performance of aerospace parts, even as everyday technological, environmental, and economic issues persist. Specific opportunities include the improvement of mechanical strength of the self-healing system, the development of affordable production processes, and establishment of specified operational conditions for the system. They will establish whether self-healing technologies can provide a solution to these challenges and, therefore, can be part of mainstream aerospace manufacturing.

4. FUTURE DIRECTIONS

The following are important future aspects of self-healing materials:

1. Improvement of hybrid self-healing systems combining thermal and light activation for enhanced durability.

2. Use of nanomaterials to improve healing rates and strength, enabling autonomous repair in extreme conditions.

3. Focus on scalable, cost-effective synthesis methods to support large-scale manufacturing.

4. Integration of self-healing functions in 3D printing to extend part lifespan and reduce waste through material reuse.

5. Advancements in self-healing soft electronics for adaptive, self-repairing technologies, including wearable systems.

6. Design of multi-functional polymers where self-healing capability and mechanical strength are in balance.

7. Development of novel healing agents with enhanced durability, responsiveness, and environmental robustness (e.g., UV-resistant, thermally stable).

8. Optimization of AM process parameters (i.e., temperature, speed, laser power) to maintain healing agent functionality throughout printing.

9. Design of AM-friendly self-healing composites with integrity in structure without the expense of healing efficiency.

10. Real-time monitoring and regulation of healing agent distribution throughout printing through AI and sensor integration.

11. Improving self-healing performance under aerospace-specific conditions like extreme temperature, humidity, and UV illumination.

12. Systems designed for response to multiple stimuli (e.g., light, heat, pressure) to guarantee functionality in a range of environments.

13. Self-healing property tailoring to address particular requirements of aerospace parts (e.g., fuselage, fuel pipes, and electronic enclosures).

14. Soft robotics and adaptive structure integration of self-healing polymers in aerospace systems.

15. Development of standard test protocols for self-healing under representative aerospace loading and environmental conditions.

16. Long-term lifecycle and durability analysis to ensure reliability in missions-critical aerospace applications.

5. CONCLUSIONS

Additive manufacturing of self-healing polymers faces several limitations. Integrating healing agents often compromises mechanical strength and limits use to non-load-bearing parts. Large-scale production is difficult due to the complexity of controlling polymer properties. Encapsulated healing agents may lack long-term durability, and environmental factors like temperature and humidity affect healing performance. Photo-activated systems are limited by sensitivity to light exposure. Additionally, self-healing soft electronics struggle with adhesion and flexibility, posing integration challenges in adaptive aerospace platforms.

Future directions for self-healing polymers made by additive manufacturing include the design of hybrid thermal and photo-activated systems, large-scale synthesis routes, and multifunctional polymers balancing strength and healing. Integration of nanomaterials, smart healing agents, and adaptive soft electronics will improve performance. Tuning AM parameters, allowing for AI-based process monitoring, and designing composites with structural integrity are important. Special attention is also given to aerospace-specific qualification, multi-stimuli responsiveness, and lifecycle assessment to ensure durability and functionality under extreme use conditions.

Acknowledgements. This work was partially funded by the Centre for Research at Chennai Institute of Technology, India, under funding number CIT/CFR/2024/RP/001.

CRediT authorship contribution statement. A.V. Santhana Babu: Conceptualization. H.S. Philip: Writing - Original Draft. D. Jayabalakrishnan: Writing - Review & Editing.

Declaration of competing interest. The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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