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A review on self-reporting mechanochromic composites: An emerging technology for structural health monitoring

Ali Tabatabaeian [a,b,*], Sixin Liu [a], Philip Harrison [a], Erik Schlangen [b], Mohammad Fotouhi [b,*]

[a] School of Engineering, University of Glasgow, Glasgow, UK
[b] Faculty of Civil Engineering and Geosciences, Delft University of Technology, Delft, the Netherlands

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ABSTRACT

Recently emerging mechanochromic systems are becoming highly attractive for structural health monitoring (SHM) purposes in various industries, such as civil, wind, and aerospace, to improve the safety and performance of structures. These are based on self-reporting polymer composites which provide a light-weight sensor with an easy-to-read visual cue for SHM purposes. The present paper reports a critical overview of mechanochromic self-reporting approaches and discusses the outlook for future development in the field. Design principles and cutting-edge applications of the main physical- and chemical-based self-reporting mechanisms, i.e., mechanochromism based on dye-filled materials, modified polymers, structural color materials, and smart hybrid composite sensors, are presented with special attention to SHM. These emerging sensors create a new generation of user-friendly, cheap, and power-free SHM systems, guaranteeing economic and technological advantages that will open up new horizons for innovative, safer, and lighter composite products with significantly lower maintenance costs.

1. Introduction

Safety critical fiber reinforced polymeric (FRP) composite materials and structures are widely used in various industries such as civil, aerospace, biomedical, wind turbine, automotive, medical etc., where they are exposed to harsh environmental conditions, such as temperature fluctuations, humidity, strong winds, UV irradiation, corrosion etc. [1–3]. Moreover, static or dynamic overloading conditions such as impact strikes and fatigue are reported as primary causes of damage in glass and carbon fibre reinforced polymer (GFRP) and (CFRP) composites [4]. For instance, low-velocity impact can happen due to bird strikes or dropping a tool during composite structure repair or assembly. This may not induce any visual damage on the surface while causing multiple internal delamination in ply level. Such internal impact-induced damage, often referred as barely visible impact damage (BVID), might lead to serious structural damage and unexpected failure. In this case, different non-destructive evaluation (NDE) techniques have been developed to facilitate quality control, particularly in large safety critical structures, so that the regions that require repair or replacement could be detected [5]. NDE methods are time-consuming and often employed for post-damage analysis. An alternative to NDE is structural health monitoring (SHM), which is defined as the process of observing a system over time using periodically sampled response measurements from an array of sensors to determine the current state of system health. These sensors can be either attached on the surface or incorporated into the composites in manufacturing, providing information on material degradation and structural integrity over the lifetime of the structure [5,6]. The SHM literature suggests that combinations of different SHM methods must be used to ensure reliable monitoring of complex practical structures. Ultrasound scanning is the most common SHM method that provides reliable information in laboratory scale, but has a lot of complexity and drawbacks for reliable monitoring of real-life scenarios [7]. Acoustic emission is mostly used as an early warning for damage detection and propagation before it reaches a critical status, but it has significant drawbacks due to noise and can be difficult to interpret signals [8]. Fibre optic sensors are also a popular choice and widely used in SHM. Nevertheless, they require lots of cabling, are fragile and tend to break fairly easily, and also cause stress concentrations when incorporated in composite laminates [9]. Accordingly, development of new SHM techniques is essential for reliable and cost-effective monitoring of engineering structure to assure safety and to extend their service life.

Self-reporting mechanochromic composites provide feedback

* Corresponding authors at: School of Engineering, University of Glasgow, Glasgow, UK (A. Tabatabaeian) and Faculty of Civil Engineering and Geosciences, Delft University of Technology, Delft, the Netherlands (M. Fotouhi).
E-mail addresses: 2611578T@student.gla.ac.uk (A. Tabatabaeian), m.fotouhi-1@tudelft.nl (M. Fotouhi).

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mechanisms whereby the structural integrity is reported through visual signals, allowing a timely response to damage, by either further protecting the components or taking actions to repair them. A self-reporting composite can sense its own damage, which makes it an excellent material for SHM of different engineering structures such as concrete, steel, asphalt, and composite structures. Two key advantages of self-reporting mechanochromic composites over other SHM techniques are a) they can monitor the system during operation (not post-operation) and, b) they work completely wirelessly, without the need for data-acquisition systems. Taking advantage of self-reporting materials may foster novel concepts and designs for traditional sensors [10]. For example, FRP composites that are already being used as structural or reinforcing materials can be designed to act both for strengthening and SHM functions, eliminating the need for incorporating extra sensors for SHM purposes [11], or glass fibers functionalized with fluorescent proteins can be embedded in epoxy to be used both as load carrying element and impact-induced delamination indicator [12].

Self-reporting is the characteristic by which a structure senses its own state such as damage, deformation or temperature, etc. due to various stimuli such as thermal, piezoelectrical, mechanical or chemical solicitations [13]., while self-healing is the ability for automatic recovery of the structural integrity when damaged. Self-reporting and self-healing mechanisms have the potential to provide smart materials and structures that can repair themselves [5,13]. A smart composite can be designed to be both self-reporting and self-healing, referred as dual functional [14], however the main focus of this review paper is on self-reporting characteristic.

Engineering systems, in particular aerospace structures, rely heavily on visual inspection as the latter provides the fastest and cheapest inspection option. Self-reporting mechanochromic composites, represent a step forward in visual inspection approaches, and are now an interdisciplinary research theme, recent reviews have provided different insights on this topic [15–18]. However, to the best of our knowledge, there is currently no critical review that discusses and compares the most practical mechanochromic design and manufacturing approaches, considering their challenges and future possibilities, and focusing on their potential in SHM of different engineering structures. In order to limit the scope of this review, the focus here is on self-reporting mechanochromic polymeric composites that visualise damage by changing their fluorescence or other optical features (i.e., absorption, emission, refractive index) when subjected to mechanical excitations [19,20].

2. Self-Reporting approaches

Based on their origins, colors can be categorised according to the following main groups: (1) **chemical-based** and (2) **physical-based** colors. **Chemical-based colors** stem from the selective absorption and reflection of specific wavelengths of electromagnetic radiation. Pigments and dyes are prime examples of chemical-based colors, they are presented in **section 2.1**. The color changing process can occur by incorporating dye-filled materials such as micro-capsules or hollow fibres that are ruptured upon mechanical excitation **(see section 2.1.1)** (Fig. 1(a)) [21]. Alternatively, the polymer or fibre can become 'smart' by adding functional groups that are sensitive to mechanical stimuli (mechanophores), into them. For example, optical changes can be achieved using luminogens that show an aggregation-induced emission (AIE) **(see section 2.1.2)** (Fig. 1(b)) [22].

Physical-based colors are connected to both the shape and refractive index of the material and not to its chemical properties. In fact, physical-based colors originate from the way that light is scattered and diffracted by random or periodic structures **(see section 2.2)**. Fascinating biological-world examples of this group are the color of peacock feathers and of butterfly wings [10]. A prime example of physical-based colors are structural color materials which vary in line with structural dimensions and refractive indices, so that a single set of materials can produce various colors [23] **(see section 2.2.1)** (Fig. 1(c)). Another interesting idea for SHM of engineering components is thin-ply composite sensors, where hybrids comprised of fibres with different strain-to-failure ratios are utilised, for example a carbon layer and a translucent glass layer (see **section 2.2.2)**. The changes in light absorption at the interfacial glass/carbon damaged area can generate a clear visual cue by which damage, such as BVID, can be detected as an early warning to avoid catastrophic structural failure due to hidden damage (Fig. 1(d)).

2.1. Chemical-Based colors

2.1.1. Mechanochromism based on Dye-Filled materials

It is possible to self-report damage in FRP composite structures by adding dye-filled materials such as microcapsules into coatings, films,

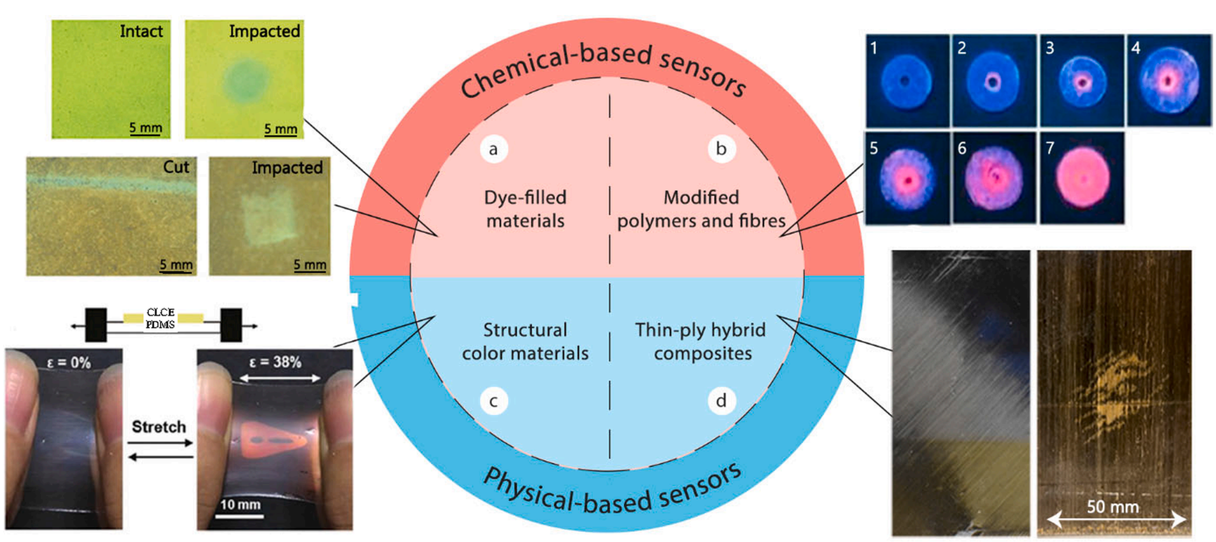

Fig. 1. Examples of different mechanochromic approaches for SHM applications: (a) characterisation of impact damage and cuts in polymer composites prepared with microcapsules. Reproduced with permission [24] Copyright 2020, Wiley-VCH., (b) color changing process in composites under compression, increasing from left to right, indicating the fluorescent activation. Reprinted with permission from Ref [25] Copyright 2017 Elsevier., (c) a mechanochromic film which reveals and conceals a pattern by stretching and releasing, working based on the structural color principle. Reproduced with permission [26] Copyright 2020, Wiley-VCH., (d) comparison of the visual damage detection in a structure with (right) and without (left) hybrid composite sensors under the same impact loading condition [27].

polymer resins, or the interface between polymer and fibre [5]. When applied as a coating layer, dye-filled materials can also be used for SHM of other substrates such as metals and concrete. The rupture of such dye-containing materials at a pre-determined force threshold leads to release of their content into damaged regions, resulting in localisation and quantification of the damage. Fig. 2 demonstrates various approaches to the design of FRP mechanochromic composites using dye-filled materials.

Dye-filled materials are primarily used in the form of dispersed microcapsules in a coating or in the bulk of materials. They provide a visually detectable response when subjected to a mechanical stimulus. The principle is the same as carbonless copy paper and pressure-sensitive recording sheets, where dye precursor solutions are released from microcapsules and instantly react with a developer to create bright color. Findings in the literature [28,29] show that taking advantage of different shells and preparation methods, one can design micro-encapsulated-based composite sensors with the same host polymer but different activation load levels. It is therefore possible to assess SHM quantitatively and visually with multi-capsule systems composed of different materials or thicknesses [30]. Thanks to several advantages such as simplicity, low cost and scalability, microencapsulation by interfacial polymerization is identified as one of the most popular methods to manufacture micro-capsules [31].

Conventional operating modes for capsule-based self-reporting composites are shown in Fig. 3. The first approach, 'simple release', is perhaps the most straightforward method to make smart capsule-based polymers in which the capsule content simply releases its content upon rupture. In this method, the dye in the capsule has the same optical features before and after the capsule rupture, making it difficult to distinguish between the damaged and undamaged regions, particularly for composites with high microcapsule content (Fig. 3(a)). To address this, Postiglione et al. [32] proposed a mechanochromic system based on micro-capsules that had a UV-light screening polyurea shell preventing the dye inside the capsule from fluorescing before its release upon rupture (but being greatly excited and fluorescant after release), creating a clear contrast between damaged and undamaged areas. In another demonstration of the capsule-based system, Di Credico and teammates [33] equipped microcapsules with the UV-sensitive photochromic dye and a UV-light screening shell. Here the yellow colour microcapsules turned red, after undergoing damage and exposure to UV illumination (Fig. 4). These examples underscore that to achieve a high colour contrast, the releasing process must be accompanied by a mechanism that alters the optical behaviour. Therefore, researchers have suggested other operating methods such as a 'turn-on' mechanism and an aggregation-induced optical change (see Fig. 3(b)). In the turn-on mechanism, the switching between the intact (off) and damaged (on)

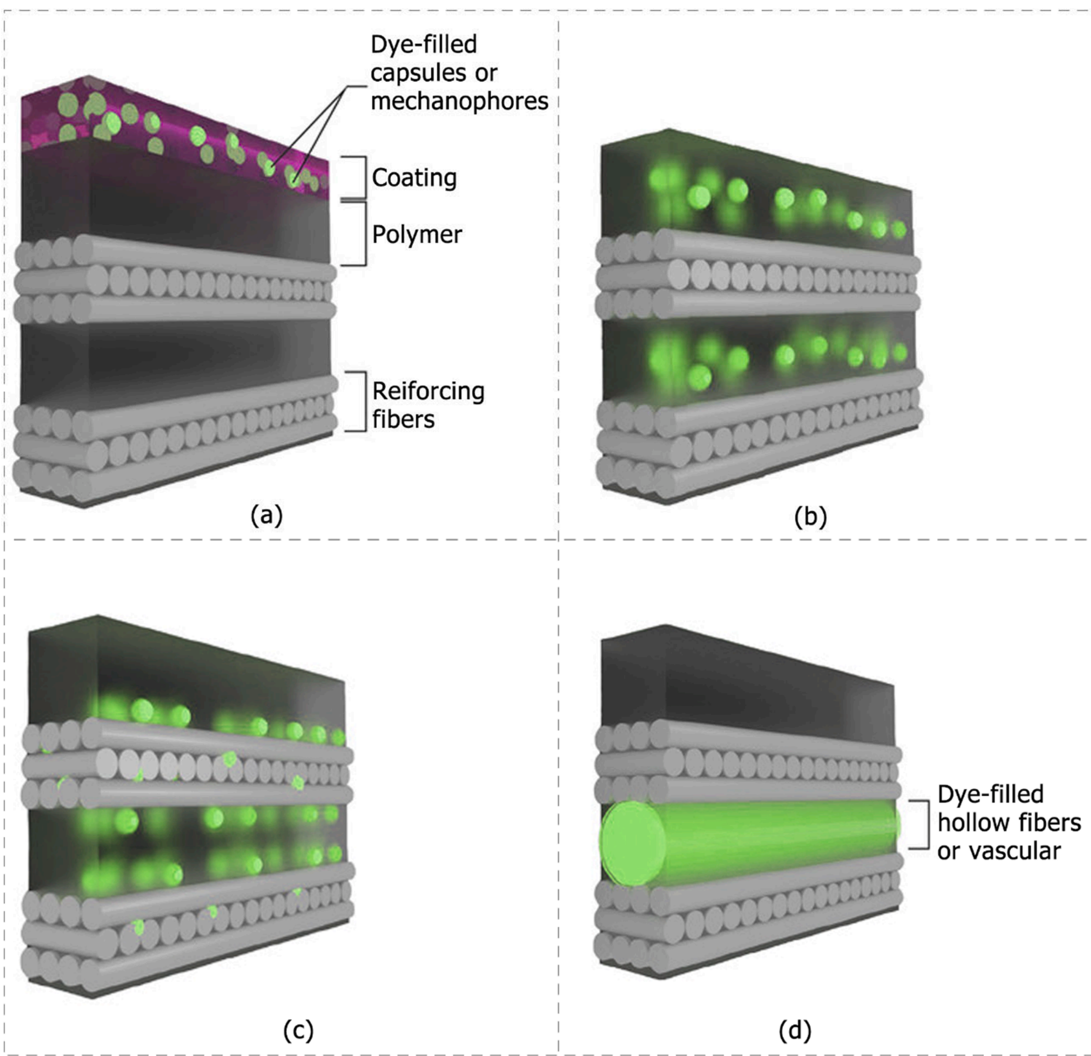

Fig. 2. Different approaches to design FRP mechanochromic composites based on dye-filled materials: (a) by dispersing them into coatings and films, (b) by dispersing them into the polymer matrix, (c) by bounding them onto the surface of the reinforcing fibers, (d) Fibers or microchannels filled with dye can be incorporated into composites. Reproduced with permission [5] Copyright 2018, Wiley-VCH.

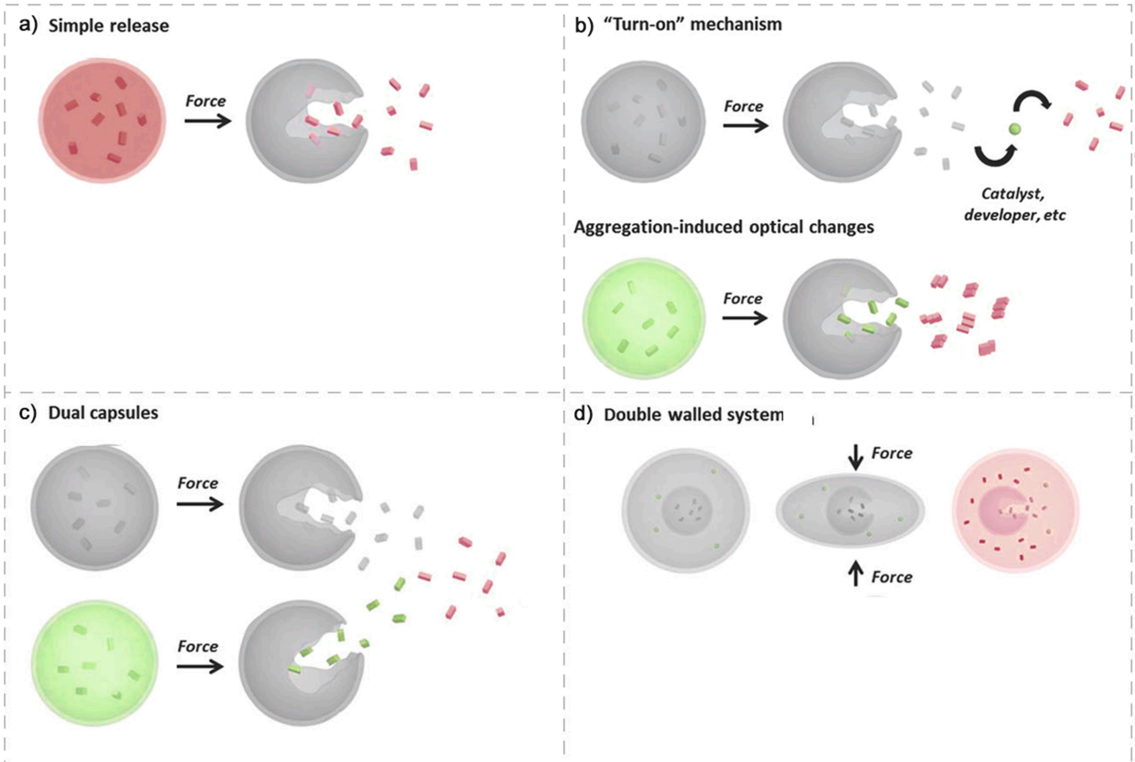

Fig. 3. Self-reporting polymers based on different encapsulation strategies. Reproduced with permission [30] Copyright 2018, Wiley-VCH.

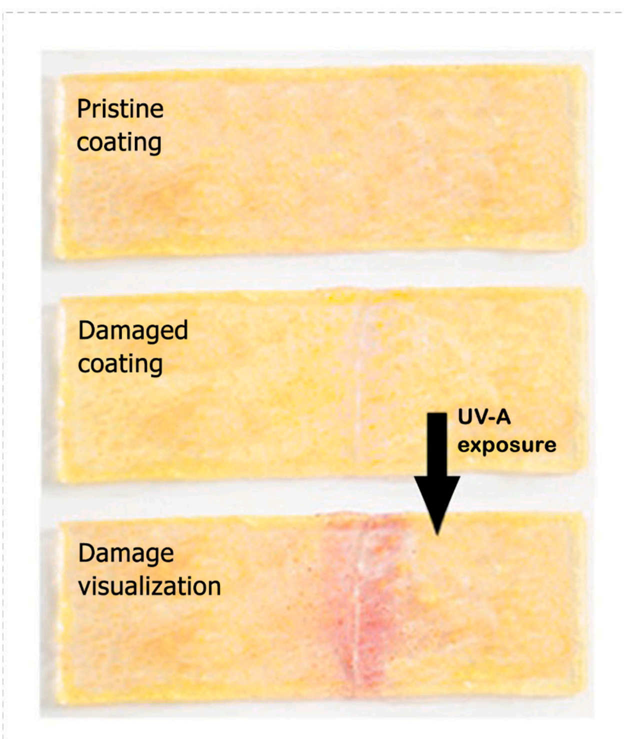

Fig. 4. The visual inspection of damage in microcapsule-based self-reporting films after exposure to UV-A light. Reprinted with permission from Ref [33] Copyright 2013 American Chemical Society.

capsules is provided either using an optical filter or a color developer or catalyst. There are, nevertheless, challenges in using color developers or catalysts [30]; for example, questions remain as to what extent these materials should be incorporated and how they might react with the polymer matrix. Also, the developer's (catalyst's) lifetime might not be well known, and inactivation or leakage are other possible risks. Chemical indicators that can interact with polymers and display a color change without the need for other reagents (e.g., color developers or catalysts) are also a viable solution. The process can be facilitated by aggregation-induced emission microcapsule systems that trigger optical changes when physicochemical changes take place, due to the release of the microcapsules. [30]. AIE-based materials will be discussed in Section 2.1.2.

A third strategy is to combine two different capsules containing the two-component part of a chromic system (see Fig. 3(c)). An interaction between the capsules and their cargo occurs, causing a self-reporting mechanism, reducing the risk of false positives signals [34]. On the other hand, it is well established that more capsule types loaded in the matrix, results in a greater possibility of an unbalanced spread of the capsules in the polymeric matrix, which, in turn, might cause discrepancies in color change or self-reporting function. To avoid such issues, Weder et al. [30] proposed double-walled capsules, a new and optimised encapsulation strategy, in which one capsule is contained within another (Fig. 3(d)). This involves a fragile inner capsule and a more compliant elastic outer one; therefore, the outer elastic container can be deformed without rupture, while the inner one releases its cargo after rupture. A strong visual signal is thus provided by allowing the developer and dye agent to flow into a single location (i.e., at the damaged position) simultaneously, demonstrating a remarkable self-reporting performance (Fig. 5(d)). Schematics of these two encapsulation strategies (dual capsules and double-walled capsules) are illustrated in Fig. 5 (a) and (b).

Chen and colleagues recently developed a dual-compartment microcapsule by Pickering emulsion polymerisation in which two interacting species were encapsulated in a single microcapsule and yellow specimens turned orange after being loaded [35]. Their outcomes suggested that microcapsule concentrations of 1.5 wt% had to be achieved for significant color intensity (Fig. 6). Adding microcapsules also

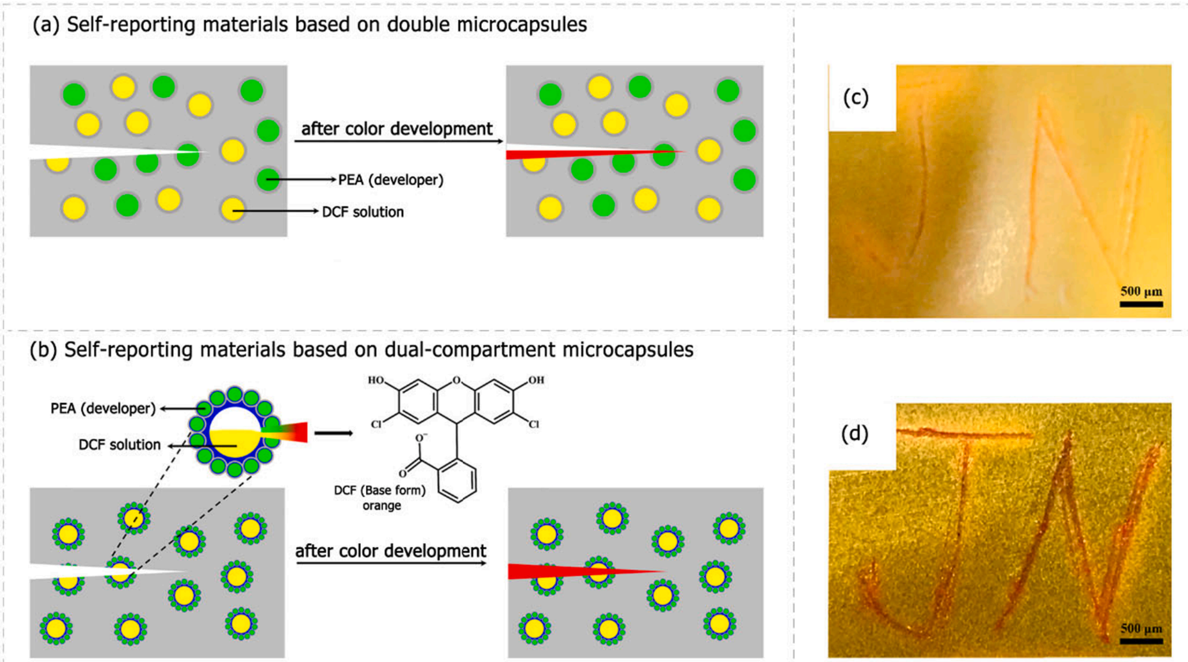

Fig. 5. Schematic of (a) double microcapsule self-reporting, and (b) dual-compartment microcapsule self-reporting systems, (c) photographs of damage to the self-reporting coating made by double microcapsules, and (d) dual-compartment microcapsules. Reprinted with permission from Ref [35] Copyright 2021 American Chemical Society.

increased the elastic modulus of the matrix resin, reflecting its multi-functionality. However, the proposed self-reporting system did not provide a quantitative relationship between deformation and color change. Recent research by Weder's group reported two different methods for preparing micro-capsule-based self-reporting composites that relied on solvatochromic activation schemes. One mechanism was to embed fluorescent poly (urea–formaldehyde) microcapsules containing solutions of a solvatochromic cyanostilbene dye into polymer matrix. When a composite is damaged, the dye solution is released from the microcapsules, diffuses into the matrix, and the solvent evaporates. Accordingly, the polarity around the dye molecules changes, resulting in a change in the fluorescence color. Another method was to blend the dye into a polymer matrix and load microcapsules with a solvent, where the solvent release could trigger the color change. The applicability of encapsulated solvatochromic dyes was demonstrated through self-reporting of the damage under impact loading. Both strategies could provide radiometric signals because the capsules that remain intact or dye molecules that are not exposed to the solvent could be seen as a built-in reference; therefore, a quantitative assessment of the damage inflicted on the material is a priori possible [24]. For example, as shown in Fig. 13(b) in the next section, the emission intensity can be correlated to the impact distance (impact energy).

Smart micro-capsule-based composites can be designed such that the color of the capsulated material not only reports the damage but also quantifies the healing state, referred as dual-function [36] For example, by changing from red, for severe damage, to colourless for complete restoration (Fig. 7) [5]. While researchers have been increasingly investigating smart self-healing polymers over the last decade [37–39], due to the challenges in their manufacturing and implementation, dual function polymers are still limited [30]. White et al. [40] presented the preparation of encapsulated self-healing epoxy composites, where a red dye was used to visualise the releasing process of the healing agent. Accordingly, the composites were self-healing and self-reporting. Nevertheless, the dye had the same color before and after flowing out of the capsules. To highlight the self-reporting feature, i.e. the contrast between intact and damaged sites, the dye must fluoresce or change colour when released into the damaged area. This was demonstrated by Chen et al [41], where a dual-function micro-capsule-based composite was designed in which aggregation-induced emission luminogens (AIEgens) were incorporated. Their smart coating could autonomously report and repair the cracks. As demonstrated in Fig. 8, the damaged areas of a steel plate coated with pure epoxy coating were extremely corroded (Fig. 8(a)). However, the steel plates coated with AIEgens-based microcapsule-embedded epoxy coating remained almost intact (Fig. 8(b)), indicating the great corrosion protection of the proposed coating for the steel panels. Moreover, photos taken both with and without UV illumination (Fig. 8(c) and (d)) exhibited the brilliant blue fluorescence results from the broken AIEgens-based microcapsules in the epoxy coating, demonstrating the excellent self-reporting performance of the proposed coating. Song et al. developed a dual-function micro-capsule-based coating system using a single AIE fluorophore [32]. Their coatings could exhibit weak-intensity red fluorescence after being scratched and the fluorescence color turned to orange after healing. In the healing area, fluorescence intensity increased significantly following the scratch. In addition to encapsulation, dual-function (self-reporting and self-healing) materials can also be created using other methods such as by adding carbon fibre, carbon nanotubes [42–45], graphene [46], incorporating cross-linking hydrogels [47–50] or by taking advantage of magnetic characteristics [51,52], though these mechanisms are outside the scope of this review.

Despite the popular application of micro-capsules as primary dye-filled materials, for some applications, such as FRP composites, when incorporating the dye into a bulk structure, hollow fibres can be advantageous. Fundamental and interesting work has been performed by Pang and Bond [54–58]. Developing the "bleeding composites" idea, they introduced a class of dual-function composites reporting damage by visual cues and subsequently heal it. Smart FRP composites equipped with hollow fibres were examined under flexural bending and low-velocity impact tests. Upon the fracture of the functionalised fibres, they released healing agent into the damaged area. Also, given the fluorescent characteristics of the agent, visual SHM of the BVID could be carried out. Kling and Czigány [59] reported a more efficient dual function system based on the application of very thin hollow fibres. The proposed SHM system could successfully visualise and heal the impact-

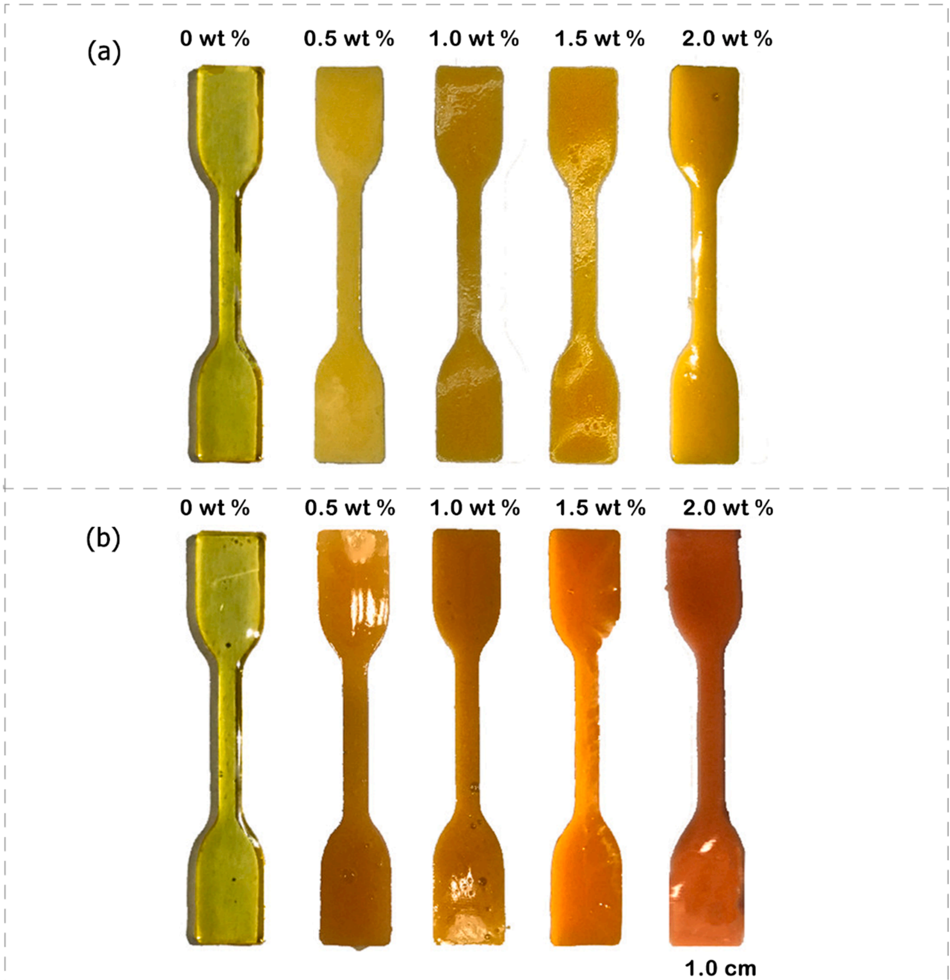

Fig. 6. Specimens with different dual-compartment microcapsule contents show different colors (a) before and (b) after stretching. Reprinted with permission from Ref [35] Copyright 2021 American Chemical Society.

induced damage with the help of a UV lamp.

A challenge in using microcapsules and hollow fibers is that the healing/reporting liquid inside them can only be used for a limited time and will ultimately be used up. Therefore, the SHM via these materials is often a one-time process. Inspired by the transport in vascular biological systems, scientists have developed vascularised microchannel structured materials. These channels can be used for pumping healing/reporting agents into damaged region. As opposed to micro-capsules and hollow fibres, this design strategy can lead to continual healing of a damaged area. In pioneering works by White et al [60,61], microchannels in a smart coating were filled with a solution of the monomer dicyclopentadiene to repeatedly heal the formation of the cracks. Nevertheless, the system was not self-reporting. To make the vascularised channels self-reporting, Trask et al. [47,48] placed solder wire with a diameter of 0.25 or 0.5 mm between the composite plies. The solder wire was removed by heating the material after curing, leaving behind vascular channels. The resulting system could both monitor and heal the impact-induced damage thanks to a fluorescent dye incorporated into the healing solution.

2.1.2. Mechanochromism based on modified polymeric and fibrous materials

Another viable approach to design mechanochromic composites is to functionalise the polymer or fibre with mechanically sensitive functional groups, mechanophores, that change their optical properties such as fluorescence or emit luminescence when exposed to a mechanical excitation such as stretching [35]. Research has shown that if the mechanophores are applied in the interfacial area between the thermoset resin and reinforcing fibres, due to the mismatch in mechanical properties of these two phases and activation of different damage modes in this. Brun's group [62] successfully detected the BVID in CFRPs by applying a yellow fluorescent protein at the resin/fibre interface (see Fig. 9). A significant advantage of self-reporting by this technique over micro-capsules or channels is its ability to monitor damage at the molecular level, making it a reliable tool to analyse damage progression in FRP composites at the nanoscale [5].

The self-reporting of BVID in GFRPs has recently been studied by Shree et al. [12], where they used Spiropyran as a self-reporting functional additive. Spiropyran mechanophores act through a reversible, mechanically-activated ring-opening reaction which converts the colourless and nonfluorescent Spiropyran into the highly coloured and fluorescent merocyanine. It was observed that the GFRPs modified with Spiropyran could change their color from yellow to purple as a result of periodic impact strikes. The number of impact strikes could also be related to the color gradient (see Fig. 10). More recently Magrini et al reported the application of Spiropyran mechanophores to design and manufacture tough bioinspired composites that can report damage. They established a correlation between the tensile deformation and emission intensity of the Spiropyran-modified polymer, leading to visualisation, quantification, and prediction of damage in the composite material before the onset of fracture [63].

Such Spiropyran-containing materials offer good potential for

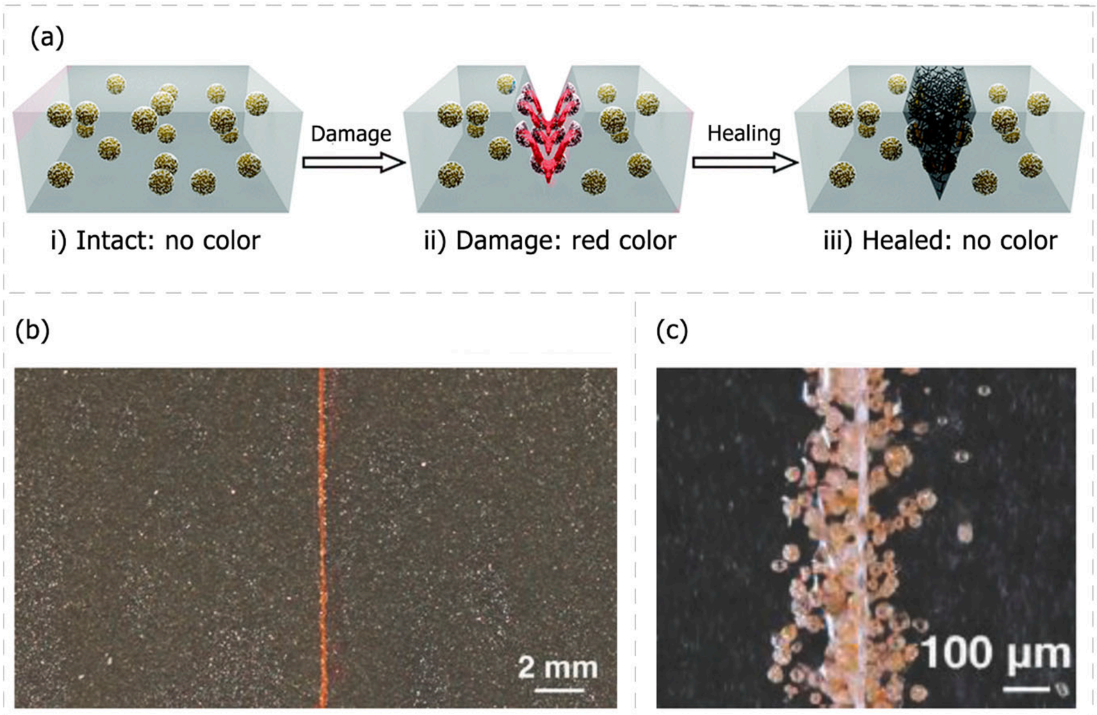

Fig. 7. a) Schematic of a dual-function micro-capsule-based polymer coating to report and heal the crack. Reproduced from Ref [53] with permission from the Royal Society of Chemistry, b) a red color is developed in scratched region of the coating, c) Microscopic demonstration of a scratch, showing mechanochromism in the damaged region. Reproduced with permission [13] Copyright 2016, Wiley-VCH.

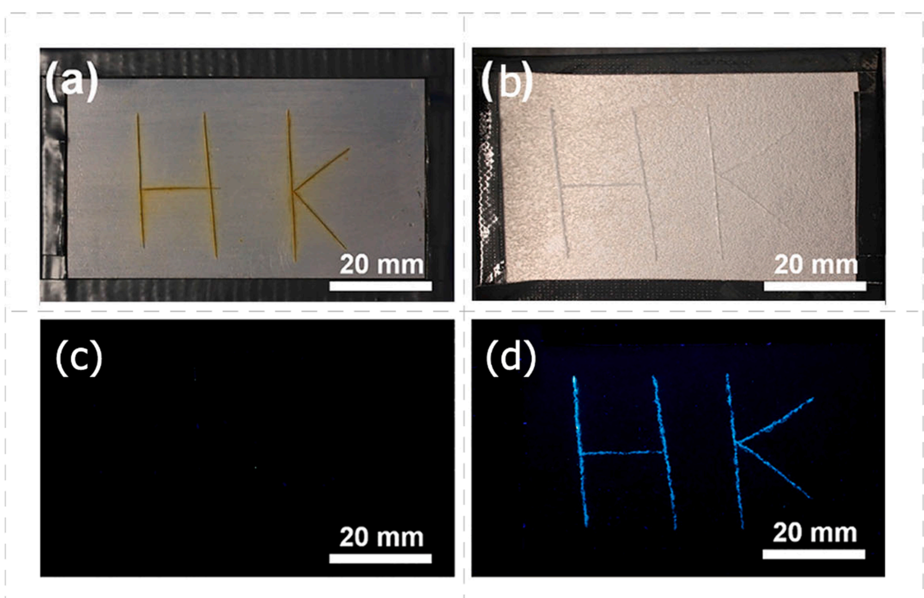

Fig. 8. Demonstration of the simultaneous self-reporting and self-healing functions in smart polymer coatings. (a) Pure epoxy coating, (b) AIEgens-based encapsulated epoxy coating (under the illumination of white light) and (c) Pure epoxy coating, (d) AIEgens-based encapsulated epoxy coating (under the illumination of UV light). Reprinted with permission from Ref [41] Copyright 2020 American Chemical Society.

microscale SHM applications. Nevertheless, it should be noted that initial high-performance or load-bearing properties of the desired materials are not influenced during the synthesis process. Additionally, some mechanophores can react to different stimulus such as mechanical, temperature- or light-related excitations [14]. A viable approach to exploit such multi-responsive mechanophores is the formation of supramolecular complexes. Using rotaxanes as molecular shuttles, Weder et al [64] encapsulated mechanophores non-covalently in polyurethane elastomers. The conceptual framework of such a rotaxane-based molecular shuttle and the respective molecular structure are shown in Fig. 11.

Another example of mechanochromism, based on the concept of supramolecular complexes can be found in [65], where they demonstrated the early-stage damage detection in CFRPs under tensile, compressive and fatigue loading by adding supramolecular cross-links within the matrix (see Fig. 12). Microscale damage in the composite's

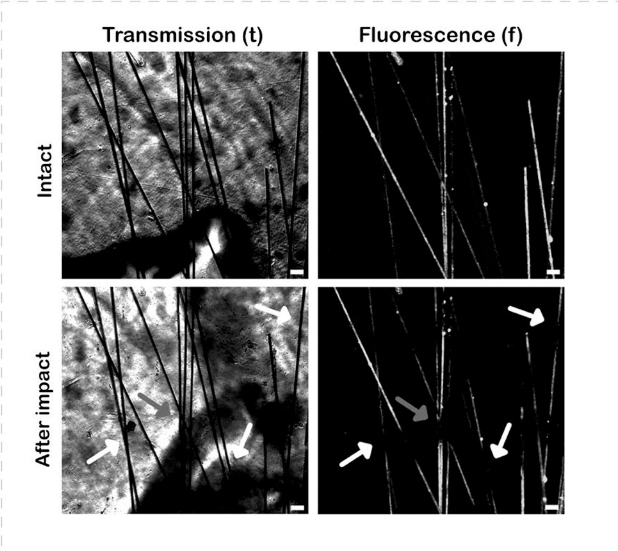

Fig. 9. Detection of BVID in CFRP composites by fluorescent proteins. The yellow fluorescent protein stops fluoresing after the occurrence of low velocity impact damage. Reproduced from Ref [62] with permission from the Royal Society of Chemistry.

first layer was visualised by breaking apart the weak supramolecular links, effectively turning on the fluorescence of the probe by the application of stress.

A mechanochromic response can also be achieved using luminogens with aggregation-induced emission (AIE) features. The working principle in SHM by AIE-based materials is such that they exhibit no significant fluorescence when dissolved in a solvent, given the vibrational and/or rotational modes that promote non-radiative relaxation pathways for electronically excited states. Due to aggregation, intramolecular motion is restricted, thus triggering emission. Since the first report of the AIE concept in 2001 [66], AIEgens have been widely applied in various fields, such as optoelectronic devices, chemosensing, and bioimaging, and they have proved to be good candidates for SHM of polymeric materials [15]. Self-reporting polymer composites modified by AIEgens can be prepared either by chemically linking AIEgens into polymer chain or physically incorporating them into a polymer matrix. The latter is more practical, simpler to carry out and can be performed through encapsulation or direct dispersion into polymer matrix [67,68].

Pucci et al. [69] dispersed a group of AIEgens called tetraphenylethene into poly (b-styrene-*b*-butadiene-*b*-styrene) elastomer to manufacture a smart film with dynamic mechanofluorochromic fluorescence. There was a noticeable decrease in fluorescence when the film was stretched. Following the release of the force, the fluorescence returned to its original state, demonstrating its reversible nature during that phase of the drawing process. Owing to the AIE effect of tetraphenylethene, the stretching-related fluorescence of this composite film could be controlled via the tetraphenylethene concentration and film thickness. In other research, tetraphenylethylene microcapsules were applied as a functional coating to improve the damage visibility of a CFRP composite. As demonstrated in Fig. 13(a), impact-induced damage was clearly discernible under UV light [70]. Exploiting the concept of aggregation-induced excimer-formation of fluorescent dyes, Weder et al [31] developed new microcapsules embedded in a silicone rubber. The materials were tested under impact, tension and compression loadings. An advantage of excimer-forming chromophores is their mechanical response, as opposed to a turn-on or turn-off mechanism, which provides straightforward quantitative assessment of the damage, color analysis can be also conducted to monitor the system's SHM performance (Fig. 13(b)). More recently, Lu et al. designed a multi-layered coating (see Fig. 14), in which AIEgens with red, green and blue emissive colors were encapsulated and incorporated in different layers of the polymeric coating. By reading these emissive colors, one could assess damage depth visually [71].

Another type of nonconventional luminescent polymer has also been proposed to exhibit mechanochromic fluorescence. This type of AIE polymer does not have any classic conjugated chromophores but can show intrinsic solid-state fluorescence by virtue of the clusterisation-triggered emission of electron-rich subgroups [72,73]. In this case, by monitoring their fluorescence during deformation, the failure of polymeric materials can be analysed in precise detail [74]. More details on AIE-based mechanochromism can be found in published reviews [74,75].

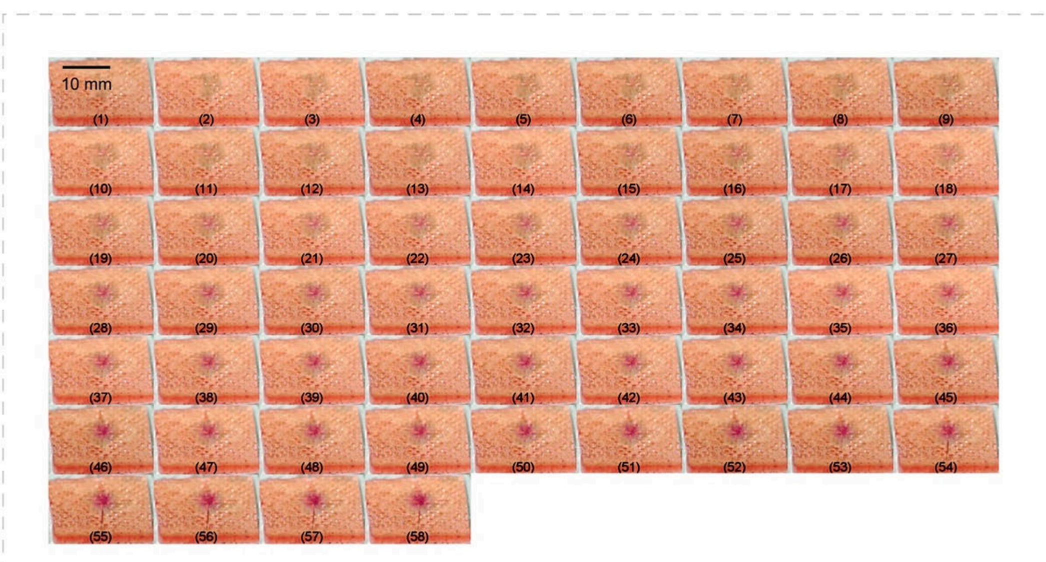

Fig. 10. The self-reporting of BVID in GFRP/ Spiropyran composites through periodic impact strikes. Reproduced from Ref [12] with permission from the Royal Society of Chemistry.

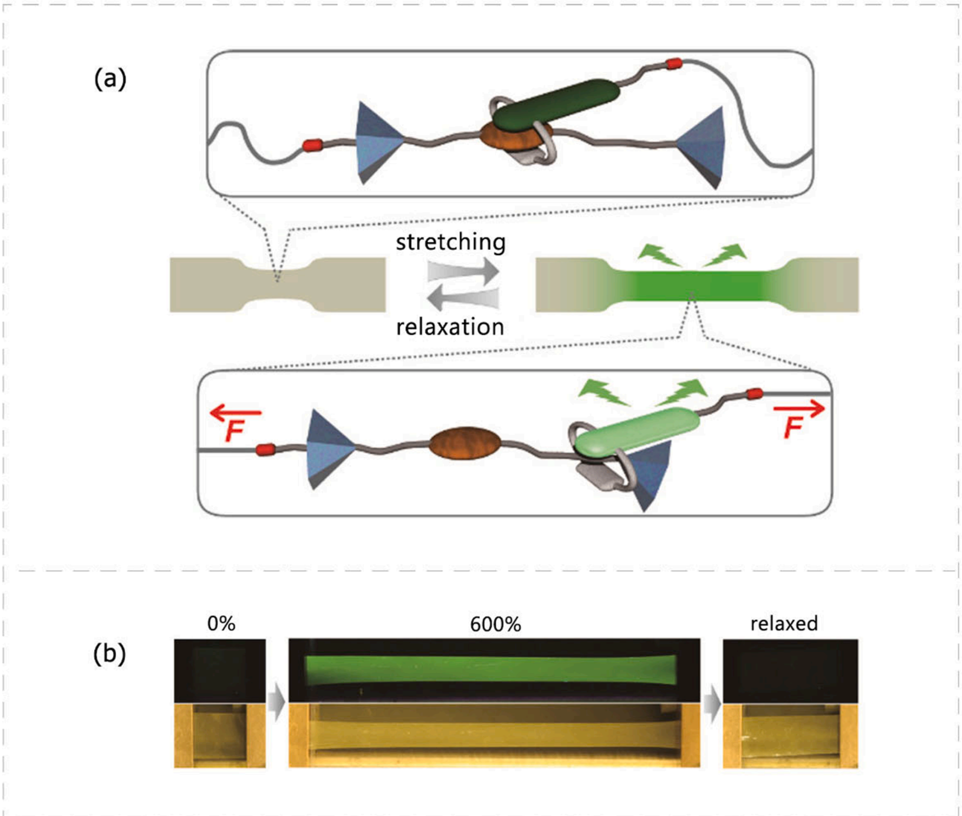

Fig. 11. a) Schematic of self-reporting using rotaxanes being equipped with anchor groups for spreading through polymer chains (red), two stoppers (blue), and a fluorophore-containing cycle (gray/green) located around a suitable quencher (brown). When subject to mechanical load, the fluorophore and the quencher are separated, switching on the fluorescence emission of the fluorophore, b) images of a Polyurethane film, whose fluorescence is turned on and off by stretching and relaxation of the film. Reprinted with permission from Ref [64] Copyright 2018 American Chemical Society.

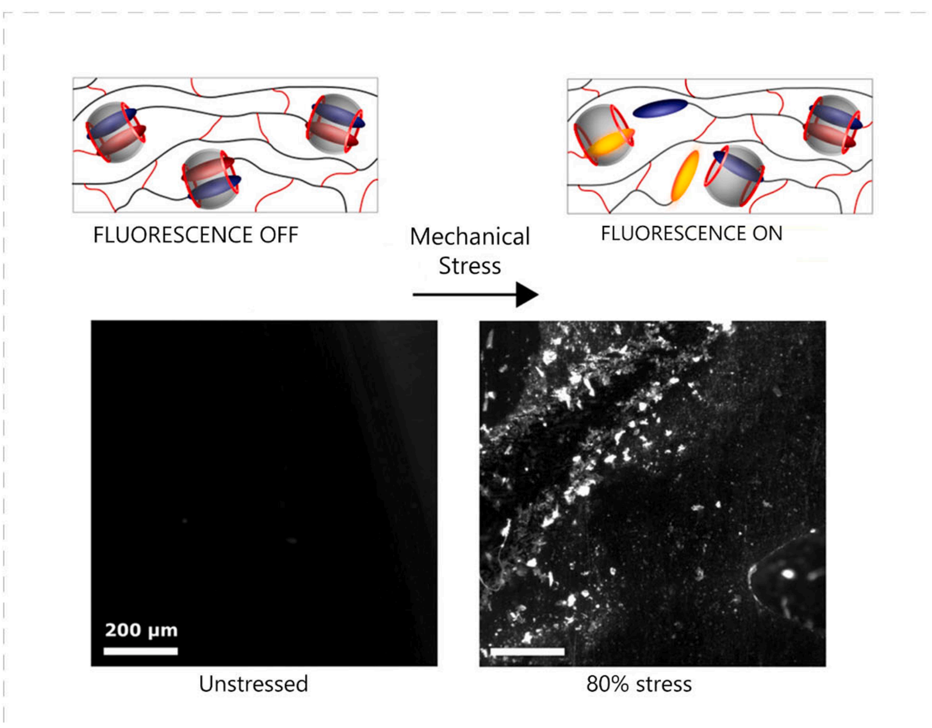

Fig. 12. Schematic (up) and microscopic images (down) of color changing process due to compressive stresses based on the concept of supramolecular complexes. Reprinted with permission from Ref [65] Copyright 2019 American Chemical Society.

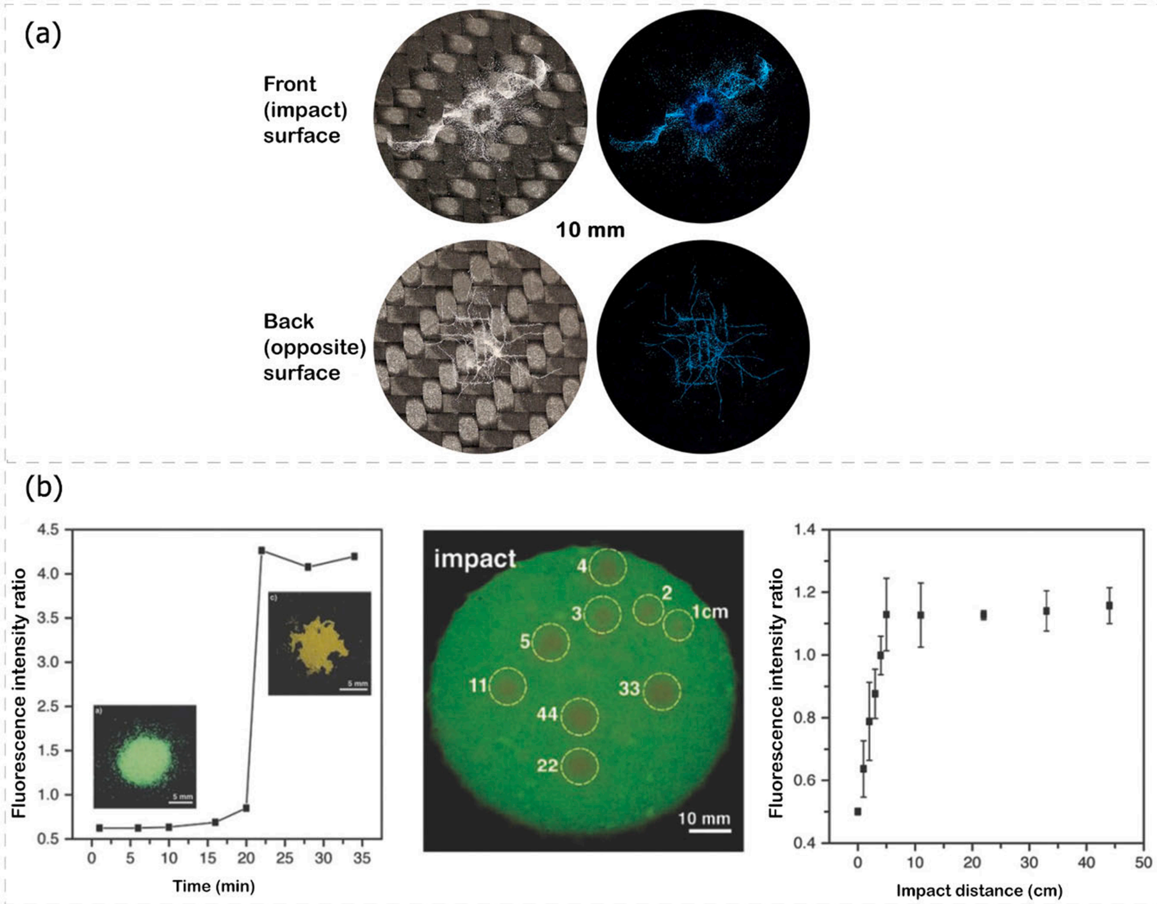

Fig. 13. a) Front face and back face images of micro-capsule-based CFRP panels under white and UV light after impact loading. Reprinted with permission from Ref [70] Copyright 2016 American Chemical Society,b) SHM of polymer composites containing the excimer-forming dye: from left to right: the excimer:monomer ratio as function of solvent evaporation time after breaking the capsules, photographs recorded under UV illumination of microcapsules impacted by a missile from distances between 1 and 44 cm, the excimer:monomer emission intensities in the same sample recorded at 560 and 497 nm as function of the impact distance. Reproduced with permission [30] Copyright 2018, Wiley-VCH.

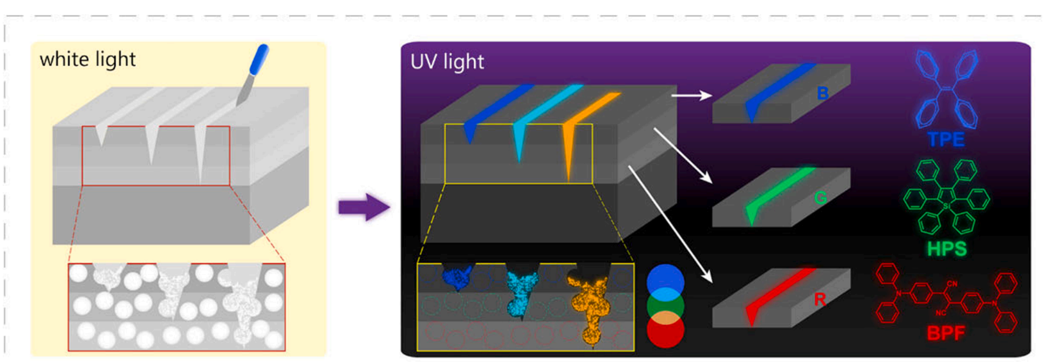

Fig. 14. Schematic of the AIE-based multi-layered coating to self-report damages with varying crack depths. Reprinted with permission from Ref [71] Copyright 2018 American Chemical Society.

2.2. Physical-Based colours

2.2.1. Mechanochromism based on structural color materials

Structural color materials are those with colour based on a physical origin, resulting from visible light interference caused by interaction with random or periodic structural features [76]. For instance, oriented silica spheres in a matrix of silica produce opal's unique color. This is explained by the microstructure, and the fact that the refractive indexes of the matrix and spheres are slightly different. Interaction of different wavelengths of light with this structure produces diffraction in various directions, leading to well-known iridescent colours. Butterfly and chameleon colorations are also examples of natural organisms exhibiting colorful patterning through structural color materials (Fig. 15(a)). The coloration in structural color materials comes from their structural features, so they represent an eco-friendly alternative to pigments and dyes [10]. Moreover, structural color created with inorganic materials can tolerate higher temperatures than pigments. At high temperatures, structured coloration therefore appears to offer greater promise than

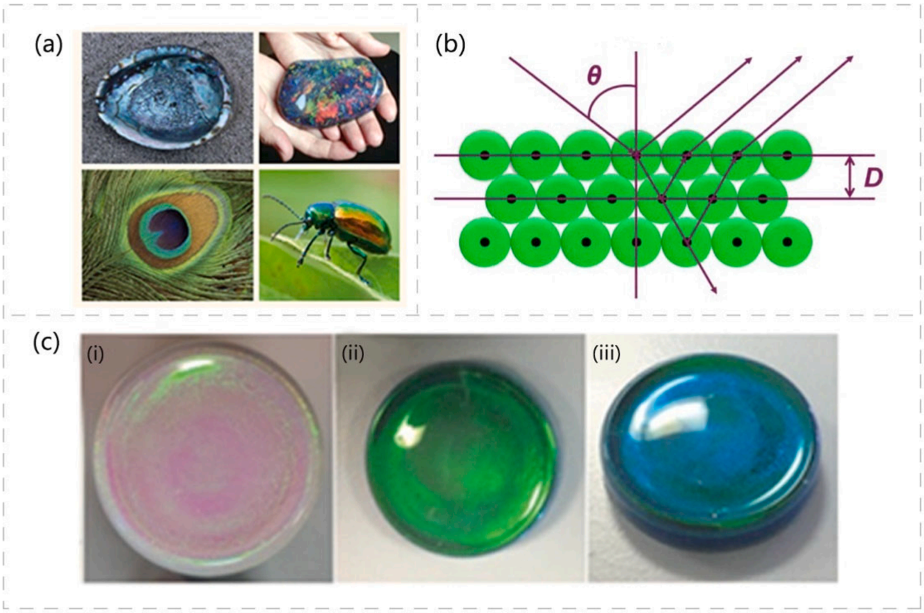

Fig. 15. a) Examples of natural structural colour: (clockwise from top left): mother-of-pearl, an opal, a beetle and a peacock feather. Reprinted with permission from Ref [87] Copyright 2011 Springer Nature, b) incident light with a wavelength predicted by a modified Bragg-Snell equation undergoes diffraction when propagating through a photonic crystal [79], c) colloidal photonic crystals: (i) top-view of a pristine photonic crystal; (ii) top-view of a graphene-based photonic crystal; (iii) the same graphene-based photonic crystal from a different viewing angle (which shows the angle-dependency of opal photonic crystal). Reproduced with permission [90] Copyright 2020, Wiley-VCH.

pigmented coloration.

A photonic material with desired optical characteristics can be manufactured by tailoring the structure at the nanoscale, which would not be possible in bulk materials. Photonic crystals possess vivid structural colours, which arise from the selective Bragg diffraction of visible light by their periodic structures [77,78]. This can be explained by Bragg's and Snell's laws [79]:

$$\lambda = 2D\left(n_{eff}^2 - \cos^2\theta\right)^{1/2}$$

where λ is the wavelength of the reflected light, n_{eff} is the average refractive index of the constituent photonic materials, D is the distance of the diffracting plane spacing, and θ is the Bragg angle of incidence of the light falling on the nanostructures (see Fig. 15(b)). This equation suggests different methods for tuning structural color, for example, by changing D, n_{eff} or θ.

Inspired by the colour tuning behaviour of natural photonic crystals (see Fig. 15(a)), scientists have developed artificial photonic crystals that can change their color under different stimuli such as electricity, heat, light and strain. Mechanical stimulus provides the most efficient color change compared with the other stimuli [80].

Structural color materials can be prepared using bottom-up or top-down techniques. The bottom-up method is based on the self-assembly of various building blocks such as colloids, liquid crystal molecules and block copolymers. An interesting application of colloidal crystals was demonstrated by Escudero et al [81] in which 2D inverse colloidal photonic structures were used in fabricating colour-tunable pressure sensors for optofluidic applications. Here, the characteristic nanostructure dimensions of the 2D photonic crystals could be changed by applying mechanical stress caused by the pneumatic pressure in the microfluidics channels. Therefore, a power-free class of pressure sensor that could change its reflective colour according to the bending pressure was successfully developed.

On the other hand, the top-down approach involves producing increasingly smaller elements by utilising precise macroscopic tools, such as lithography with photons, electrons, atoms and ions [81]. While top-down approaches need expensive lithographic instrumentation, deposition equipment and clean-room facilities, they do however provide more control over the photonic crystalline structure [80]. A combination of top-down and bottom-up fabrication strategies allows for synergy between the two [82].

Stimuli-responsive opaline materials are promising candidates in SHM and sensing devices and actuation systems [83–88]. These well-ordered structures can be generated by various methods; however, the challenge is that the resultant structures are only practical over small volumes and areas and cannot be generated on a macroscopic length scale. A technique to improve opal matrix is to introduce a small fraction of nanomaterial dopant into it. While it does not affect the lattice quality, increasing nanoparticle concentrations causes an increase in colour saturation of the opals. Zhao et al. [89] demonstrated how flexible films of stacked polymer nanoparticles can be directly assembled in a roll-to-roll process using a bending-induced oscillatory shear technique. Nevertheless, a major problem in this method is the lack of true bulk order [76]. Because of its extraordinary optical and electronic characteristics, graphene is an attractive dopant candidate. The wide spectrum of absorbance and high refractive index of this material improve structural color. More notably it works at significantly lower weight fractions compared to other carbon-based fillers, such as carbon [90]. Research suggests that adding pristine graphene into ordered polymer matrices is challenging; accordingly, functionalised graphene oxide is often used instead. In this case, a polymer colloid template is used to achieve a segregated distribution of graphene which has periodicities comparable to the wavelengths of visible light. Wang et al. [91] presented a method to form desired anisotropic structural color particles by phase separation of graphene oxide and silica nanoparticles in droplets. By carefully controlling the drying conditions, the resulting composites exhibited strong angle-dependent structural color and a stop-band which could be repeated in the visible spectrum. It is possible to make these opalescent colloidal crystals in any size and thickness, either crystallised on a substrate or freestanding. (Fig. 15(c)). They are also free of cracks, and the stop-band and mechanical deformability can be controlled throughout the manufacturing process by changing both the lattice parameters and the polymer glass transition temperature. Therefore, they can be seen as potential angle-dependant SHM tools.

As mentioned earlier, photonic crystals have a broad application as mechanochromic sensors in SHM of engineering structures, and for this purpose, they should be attached to the substrate material. However, conventional photonic crystals are made of hard materials which are only elastic up to a few percent strain, and the stop-band shift with mechanical deformation is not large enough to enable optical detection by the naked eye. To address this, polymer-based photonic crystals are swollen with a low-molar solvent to form photonic gels with extreme elasticity [92]. However, gels rely on solvent-induced swelling which can cause challenges [80]. Much effort has been made on improving photonic gels, as the latter show high sensitivity to mechanical stimulation and can therefore act well as mechanochromic sensors. For example, a plasticised photonic gel has proved to be responsive to various mechanical deformations, such as uniaxial tension and shear. Detection and measurement of uniaxial tension is more straightforward than other mechanical loading conditions, as the color change of the gel as a function of deformation can be simply measured by coating it on the elastomeric substrate followed by stretching (see Fig. 16(a)) [92]. This plasticised photonic gel has also been applied in determining the stress distribution during a peel experiment. The latter was similar to a uniaxial tension in the sense that the stress or strain field could be defined along a single principal direction (Fig. 16(b)).

Plasticised photonic gels have also successfully been used in characterising shear loading conditions, showing a strain-rate sensitive mechanochromic response. There was little color change in the green gel at low strain rates. However, intermediate strain rates, changed the color from green to red. At even higher strain rates, the appearance changed from reflective to transparent. Upon unloading of the sample, a slow recovery to the original undeformed state was seen (see Fig. 16(c)) [92]. The mechanochromic response of a photonic gel under uniaxial compression loading was demonstrated in research by Chan et al. [93], where contact mechanical testing was conducted to characterise the applied compressive strain, stress and also the contact area. Given the spherical shape of the indenter, an axisymmetric strain field was developed within the contact area of the gel, which was captured by the local color changes (see Fig. 16(d)). Fig. 16(e) illustrates another SHM application of structural color films. Here, a sensing layer made of a block copolymer and an ionic gel embedded in a polymer medium was sandwiched between ionic hydrogel electrodes, enabling full visual monitoring of strain up to100% [94].

Using strain dependant cracks and folds, Zeng et al. [95] developed various deformation-controlled surface engineering techniques to characterise self-reporting systems with different mechanochromic functions. In all their proposed self-reporting methods, small mechanical stimuli were able to cause a reversible change in the appearance of a composite material. For example, transparency-change mechanochromism shows reversible switching between transparent and opaque states (see Fig. 17(a)). Luminescent mechanochromism emits intensive fluorescence when stretched, with an ultrahigh strain sensitivity when

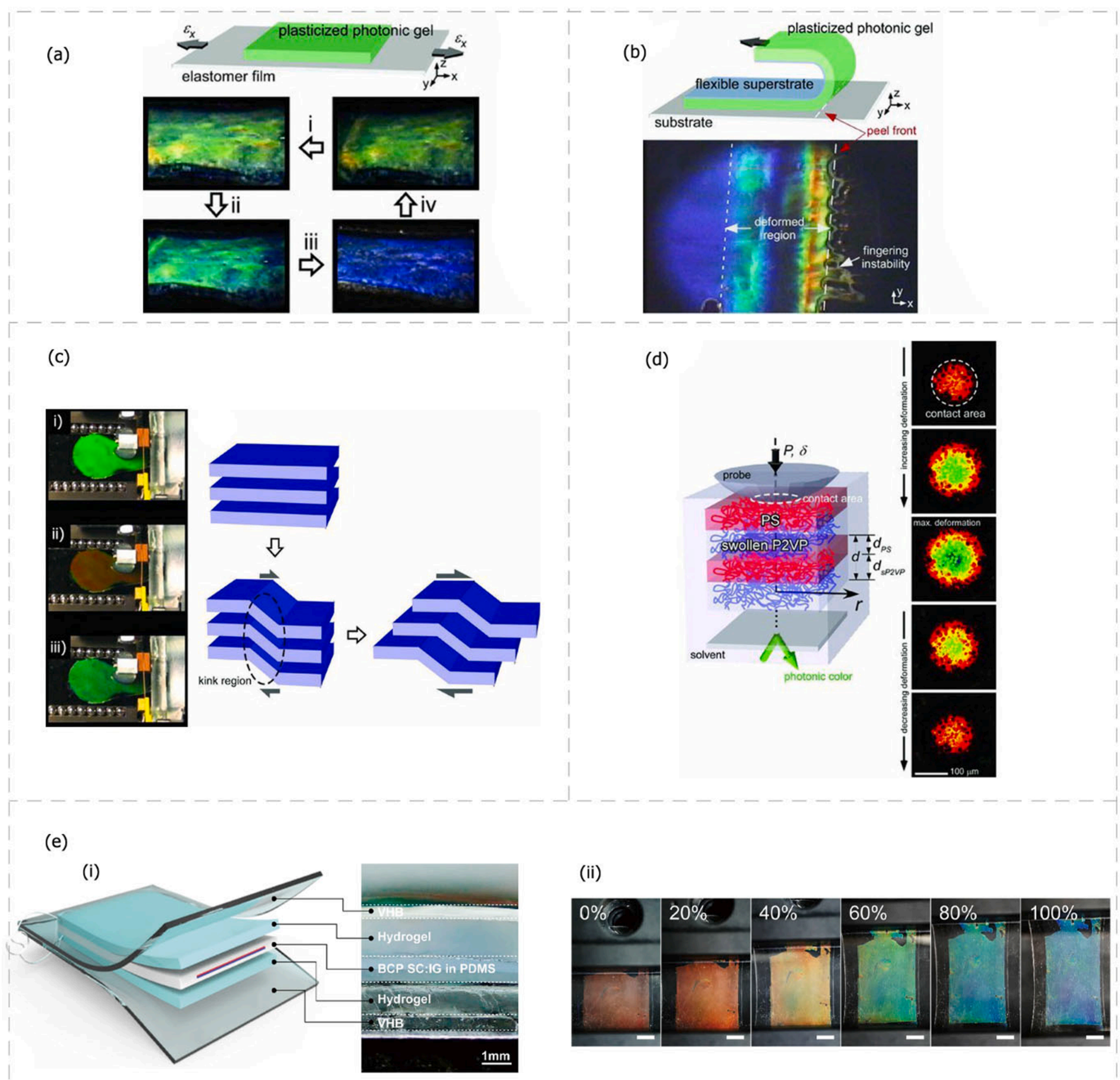

Fig. 16. Mechanochromism of photonic gels under different mechanical stimulus: a) uniaxial tension, b) peeling, c) shear. Reproduced with permission [92] Copyright 2013, Wiley-VCH d) uniaxial compression. Reproduced with permission [93] Copyright 2011, Wiley-VCH, e) a strain sensor based on a block copolymer structural color film. Reprinted with permission from Ref [94] Copyright 2018 Springer Nature.

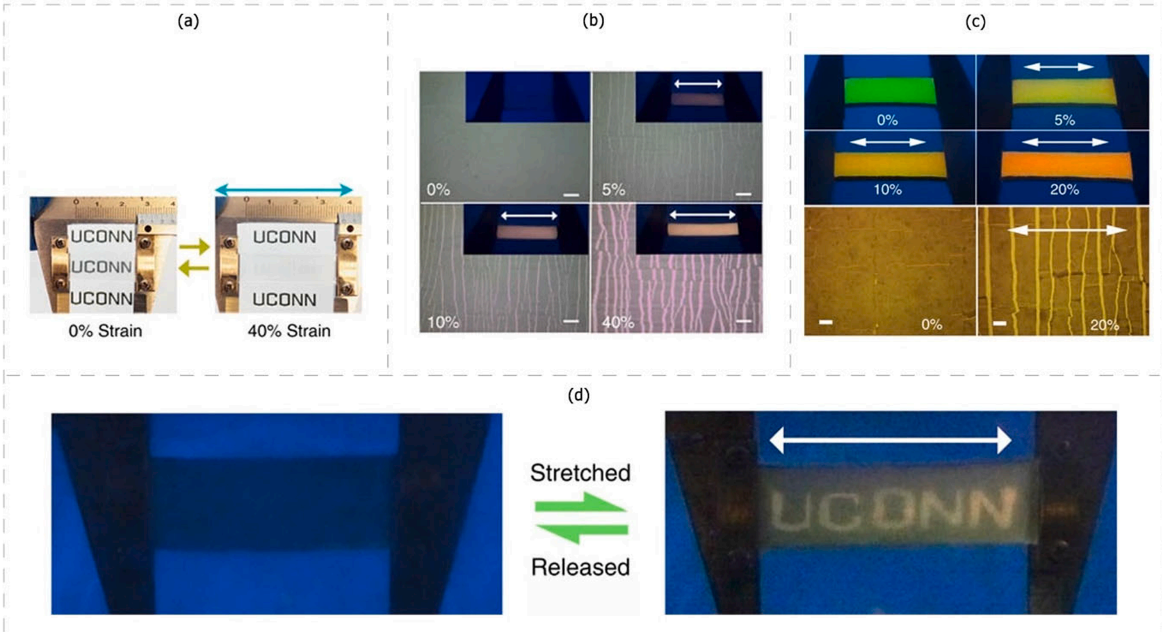

Fig. 17. Various surface-engineering techniques proposed by Zeng et al. [95]: (a) transparency-change mechanochromism, (b) the distribution and size of the longitudinal cracks upon strain in the luminescent mechanochromism, (c) colour alteration mechanochromism at different strains (0–20%) under ultraviolet light, and microscope images of crack size and distribution at 0 and 20% tensile strain (white arrow indicating stretch direction), (d) the hidden 'UCONN' logo concealed at released state and revealed upon being stretched to 17% strain with excellent reversibility. Reprinted with permission from Ref [95] Copyright 2016 Springer Nature.

compared with electrical resistance-based strain sensors (see Fig. 17(b)). Colour alteration mechanochromism turned fluorescent light from green to yellow to orange when stretched by up to 20 % strain (Fig. 17(c)), and the encryption mechanochromism device reversibly revealed and concealed predetermined patterns within a 17 % strain range (see Fig. 17 (d)).

Bae et al. [96] presented an angle-insensitive mechanochromic sensor composed of an elastomeric triblock copolymer as a dielectric spacer sandwiched between two metal layers on a flexible polydimethylsiloxane substrate (see Fig. 18(a)). Stretching the sensor

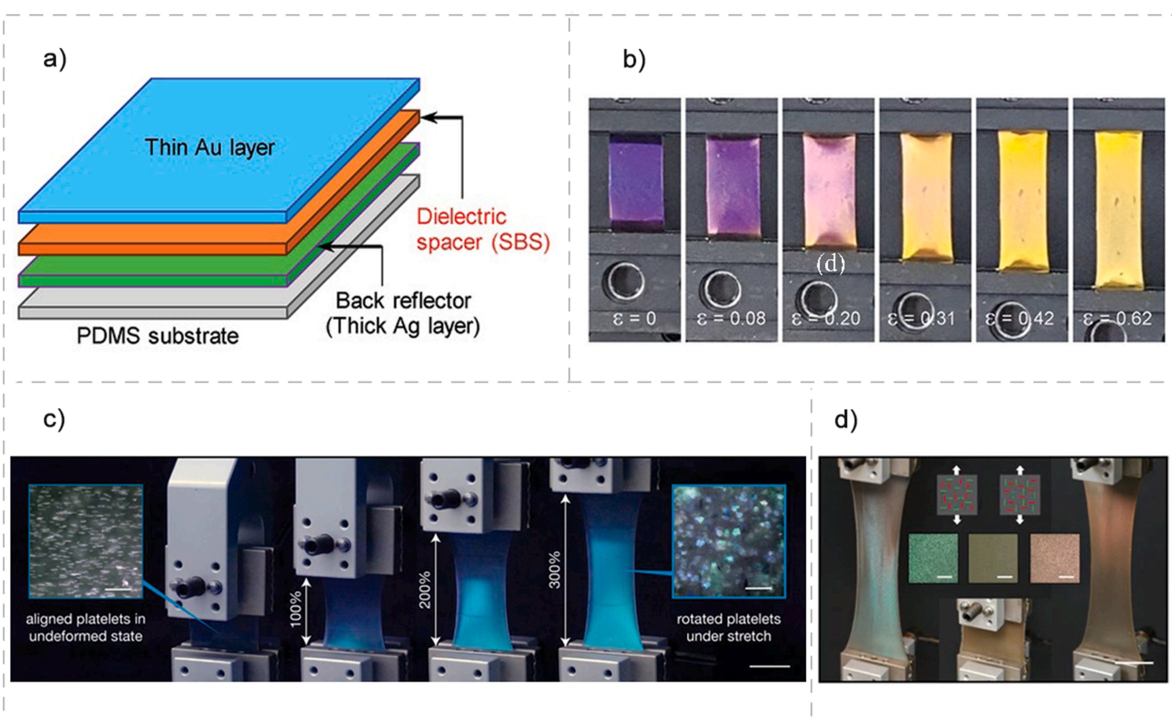

Fig. 18. a) schematic of the proposed sensor in [96], b) color changing process over a range of applied strains. Reproduced with permission [96] Copyright 2021, Wiley-VCH, c) a change in orientation of turquoise-colored platelets embedded in soft composites induces a gradual mechanochromic response over continiual stretching, d) demonstration of anisotrpic coloration in unstreched (middle) and stretched samples in desired directions inducing turquoise and red colors. Reproduced with permission [97] Copyright 2021, Wiley-VCH.

beyond 100 % revealed various colors depending on the strain. Their experiments also demonstrated that mechanical failure in metallic materials can be visually detected using this sensor (Fig. 18(b)). Inspired by the dynamic coloration mechanism of fish, Poloni et al. [97] designed and manufactured soft mechanochromic composites with a programmable platelet architecture. Platelets in a stretched composite reflect a greater percentage of the incoming light, because of an increased reflective surface area (see Fig. 18(c)). Also, an orientation dependent color changing was successfully demonstrated by incorporating turquoise- and red-colored platelets aligned within orthogonal planes in the same composite. Depending on the stretching direction, turquoise or red tilt could be activated selectively (see Fig. 18(d)). Tensile test results confirmed that adding platelets has no influence on the composite stretchability.

Despite great progress in developing structural color materials, they are still remain in the laboratory due to their complex nature, and so their applications in the real world are limited. However, these examples suggest that structural color materials in the form of coated films, are promising as mechanochromic sensors to detect different types of damage in engineering structures.

2.2.2. *Mechanochromism based on Glass/Carbon hybridisation*

Thin-ply hybrid composite sensors have been developed as an easily-implementable approach for visual indication of overload. These sensors are made using commercial prepregs and the sensing mechanism is activated via fracture. Different types of failure in a three-layer uni-directional hybrid laminate made from high-strain-material and low-strain-material are shown in Fig. 19. Damage and failure mechanisms lead to a visual indication of strain overload, where delamination is suppressed and several low strain fractures occur, followed by stable localised pull-out (see Fig. 19(c)). This is achieved by choosing appropriate material properties, relative low-strain-material to high-strain-material thicknesses and absolute thickness of the low-strain-material [98].

Fig. 20 shows two different designs of the uni-directional hybrid composite sensors, where the visual change is achieved by a purpose-designed, thin interlayer of glass/carbon-epoxy hybrid composite loaded by a predefined strain value [99]. Light passes through the translucent glass layer and is absorbed by the intact carbon layer, creating a dark appearance. Fig. 20(a) shows fragmentation of the low strain material followed by gradual, dispersed delamination [100]. The incident light is reflected from the locally damaged glass/carbon interface around the carbon layer fractures, creating light stripes. Fig. 20(b) shows another type of failure in thicker carbon layers: a single fracture of the low-strain-material is followed by sudden delamination [101]. Both can act as good overload indicators since it is easy to visually monitor the delamination through the translucent glass layer.

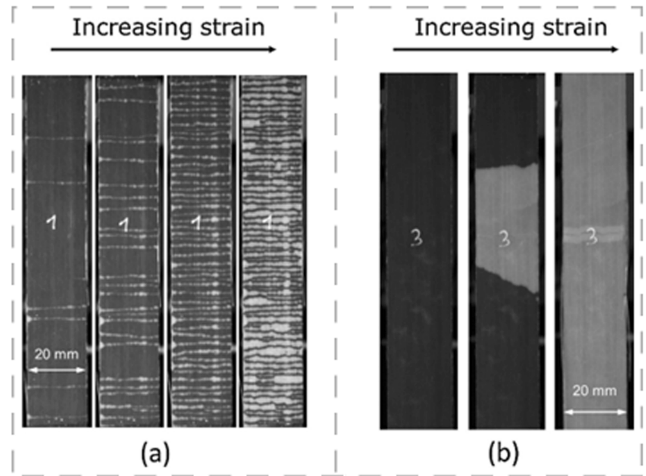

Fig. 20. Visual indication of damage in thin-ply glass/carbon hybrids: (a) carbon layer fragmentation followed by stable, dispersed delamination. Reprinted with permission from Ref [100] Copyright 2016 Elsevier, (b) carbon layer fracture followed by sudden delamination. Reprinted with permission from Ref [103] Copyright 2013 Elsevier.

Hybrid sensors can also be used for fatigue life monitoring, where an increase in the delamination area can be correlated to the number of cycles, as shown in Fig. 21 [102].

Multidirectional pseudo-ductile thin-ply hybrid laminates with improved ductility and notch insensitivity have also been developed [99,104]. These multidirectional laminates showed similar damage mechanisms to the uni-directional laminates but have the advantage of being able to monitor damage evolution, as the damage can be observed around the notches before any catastrophic failure occurs, as shown in Fig. 22.

Typically, hybrid composite sensors are either bonded directly onto the structures' surface or they are incorporated into the structure as a sensing layer (see Fig. 23). By combining different sensing materials activated by different strains, they can provide a more detailed picture of the overload. Also, by designing an array of sensors orientated in different directions, the overload direction can be monitored. A sensor

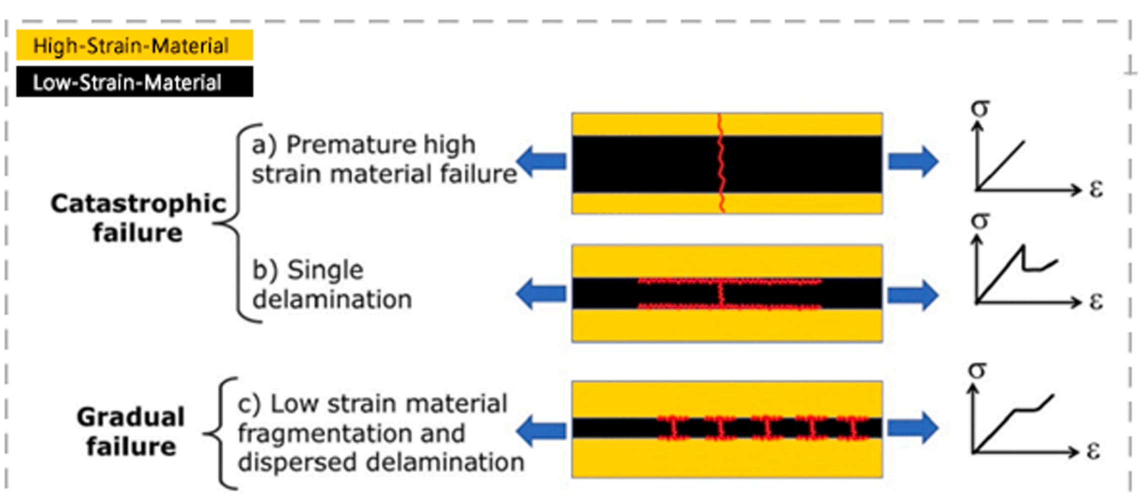

Fig. 19. Different failure mechanisms in a three layer uni-directional hybrid composed of high-strain-material and low-strain-material (red lines indicate fracture) (a) single crack through the whole specimen, (b) single crack in the low-strain-material followed by instantaneous delamination, and (c) multiple fracture and localised stable pull-out of the low-strain-material. Reprinted with permission from Ref [99] Copyright 2017 Elsevier.

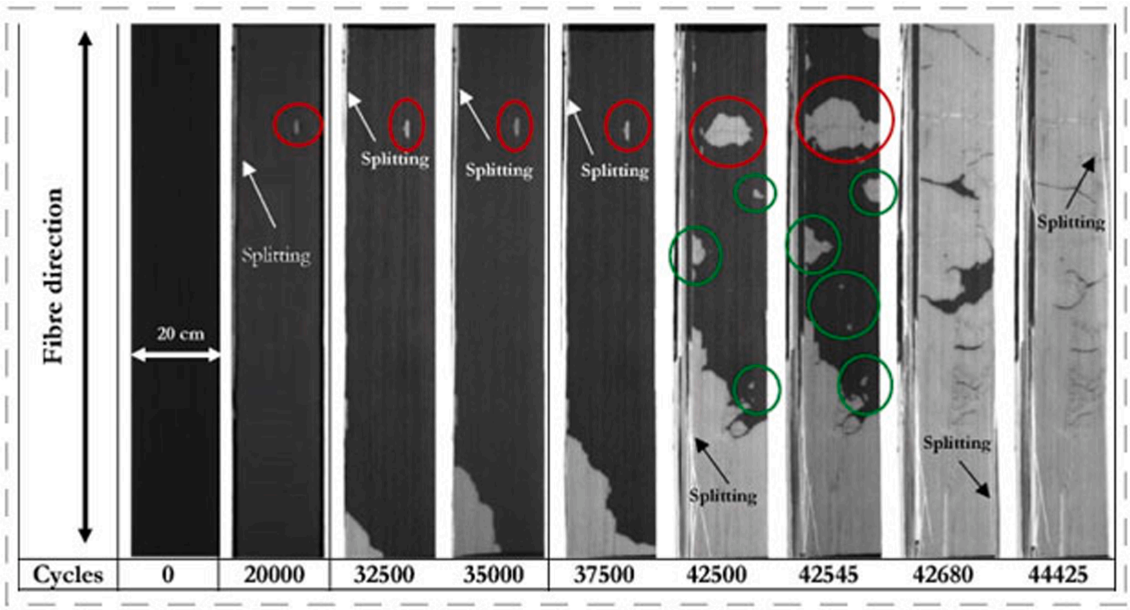

Fig. 21. Fatigue ife monitoring in a uni-directional thin-ply glass/carbon hybrid composite at 90% stress level of the carbon fragmentation initiation. Reprinted with permission from Ref [102] Copyright 2019 Elsevier.

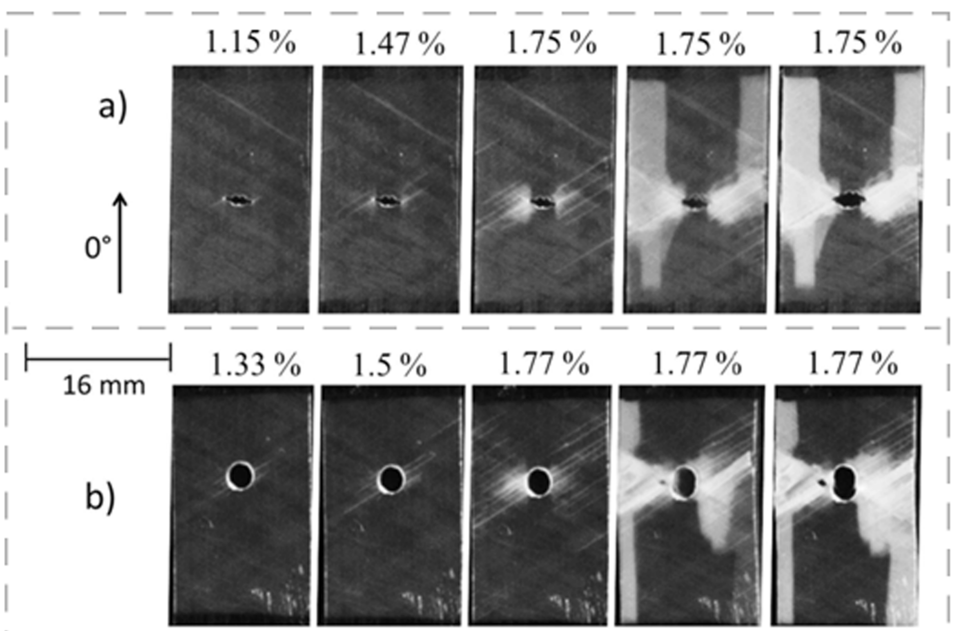

Fig. 22. Visual indication of damage in (a) sharp notched and (b) open-hole ± 60QI/Hexcel laminates, at different strains in tensile test. Reprinted with permission from Ref [104] Copyright 2018 Elsevier.

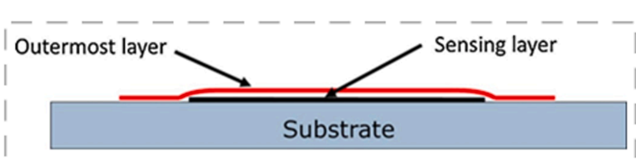

Fig. 23. Schematic of a hybrid composite strain overload sensor attached to a substrate material. Reprinted with permission from Ref [105] Copyright 2019 Elsevier.

mounted on a component is subjected to the same strains as the material below. Carbon and glass layers serve as the 'sensing' and 'outermost' layers in this case.

Unlike other mechanochromic systems reviewed in previous sections the simplicity and practicality of glass/carbon hybrid sensors has led to their relatively rapid use in SHM. For example, a set of short and long sensors made of single ply XN80/EPOXY were applied on some commercially available CFRP bike handlebars, and both handlebar structures with and without sensors were tested under a three-point bending load. Fig. 24 shows the force–displacement response of the tested structures, here the activation point for long sensors is between 1500 N and 2000 N. The graph also shows the load (1000 N) prescribed for testing Racing Bicycle handlebars according to the European

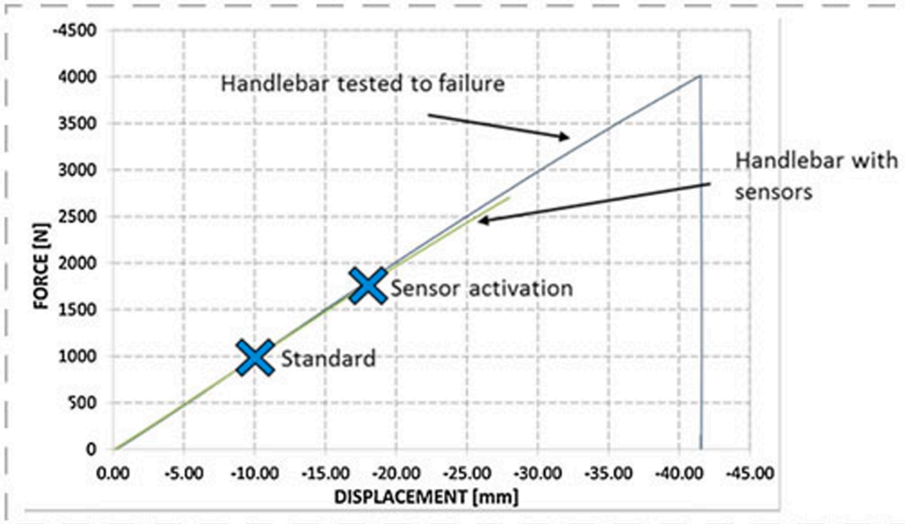

Fig. 24. Force-displacement response of the tested CFRP bike handlebars. Reprinted with permission from Ref [105] Copyright 2019 Elsevier.

Standard (EN14781) [106]. Results of this test suggested that sensors did not induce any noticeable increase in stiffness.

Fig. 25 illustrates a handlebar equipped with both short and long sensors. Here the first fracture of the long sensor was observed at 1750 N. The self-reporting concept could be successfully achieved by using long sensors, providing a warning to the user that a critical loading condition had occurred.

A bonded composite patch is one method used to repair cracks in aluminium panels used in aerospace structures. However, in the case of standard repair techniques, damage beneath the repair patch cannot be detected by simple visual inspection alone. Therefore, hybrid composite sensors can be used as a composite repair patch which can self-report critical situations, e.g. an overload or crack extension in the substrate. As discussed earlier, these sensors are designed such that they do not change the local strain distribution. Nevertheless, in case of a repair patch, it is necessary to have enough stiffness so that the load in the substrate damaged area is reduced and carried instead by the patch. The practicality of the concept was investigated by applying a hybrid composite sensor on an aluminium panel with a 20 mm initial crack length (see Fig. 26).

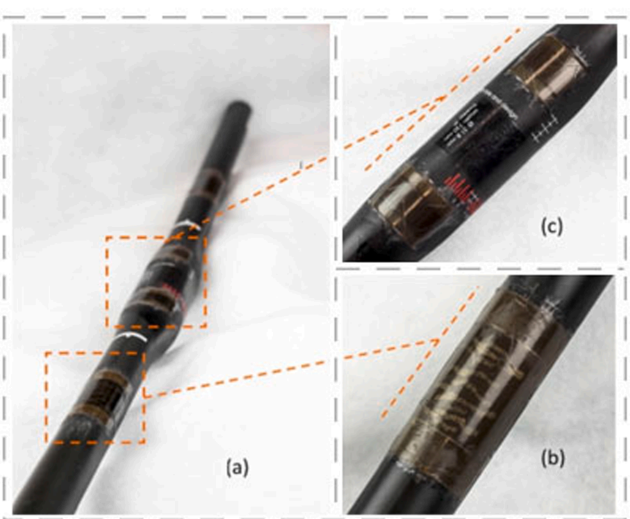

Fig. 25. Bike handlebar eqipped with hybrid composite sensors. (a) MTB Racing flat handlebar, fitted with (b) long (c) short sensors. Reprinted with permission from Ref [105] Copyright 2019 Elsevier.

During the quasi-static tensile test, changes in appearance were evident above the threshold load in the repaired specimen (see Fig. 27). By overloading the specimen, crack propagation began beneath the repair patch. Fig. 27 shows a change of appearance due to delamination, induced by fractured carbon at the centre of the specimen (above the crack), demonstrating the self-warning ability of the repair patch.

A hybrid sensor was also used to detect BVID, which can have a noticeable influence on the mechanical performance of laminates, especially the compressive strength which may decrease by up to 60 % compared with an undamaged laminate [108]. A hybrid composite layer composed of a unidirectional ultra-high modulus carbon (YS-90)/epoxy and a S-glass/epoxy material was used. It was applied to detect BVID in a quasi-isotropic $[45/0/90/-45]_{4S}$ laminate fabricated from unidirectional T800 carbon/MTM49-3 epoxy prepreg. Fig. 28 shows the hybrid composite sensor integrated on both the impacted and back face of a quasi-isotropic composite plate (cured together).

Fig. 29 shows an example of the impacted-face-, back-face and c-scans for samples subjected to a12-Joule drop tower test. The c-scan reveals significant delamination damage for all samples, and the delamination size is slightly higher in the original sample compared to the sensor integrated sample. However, there is no change in the appearance of the original sample on either the front or back faces. In contrast, for the sample with the sensor, a visible colour change is observable on both faces. As discussed, these colour changes are due to damage induced in the hybrid sensors. The size of the visible damage area on the front face corresponds to the level of the impact energy.

3. Summary and future research directions

In recent years, SHM has gradually become a multidisciplinary field of research that seeks novel strategies to improve the lifetime and maintenance of engineering systems. Inspired by nature, scientists have been developing new SHM methods, among which mechanochromic composites are of great interest, in which optical patterns such as different colors can be related to different health levels. This technology has the potential to outperform conventional SHM methods in several ways; firstly, it is wireless and does not need any data acquisition system; secondly, it is light-weight and environmentally friendly; thirdly, it can be applied for online inspection during the system's operation, as opposed to most methods that can only be implemented for post-process inspection, and last but not least, mechanochromic composites can be designed to be both self-reporting and self-healing, which can significantly enhance the durability of the engineering structures. This review

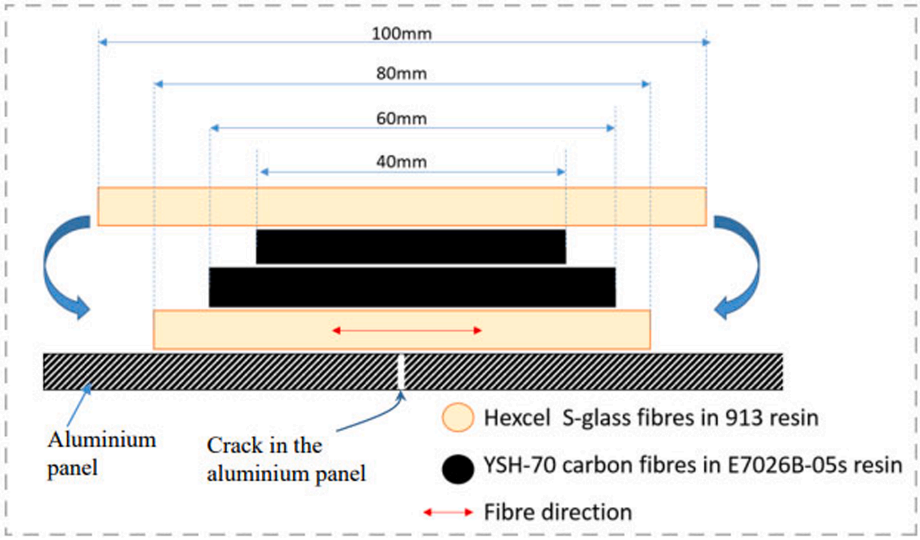

Fig. 26. Schematic of hybrid composite repair patch [107].

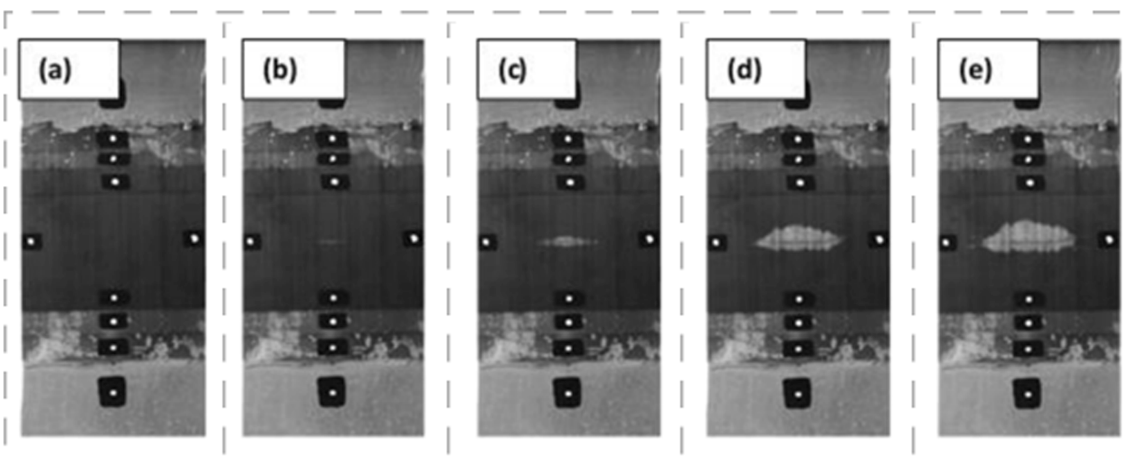

Fig. 27. Self-reporting hybrid composite sensors subjected to increasing loads: (a) 195 MPa; (b) 197 MPa; (c) 304 MPa; (d) 336 MPa; (e) 340 MPa (99 % failure load) [107].

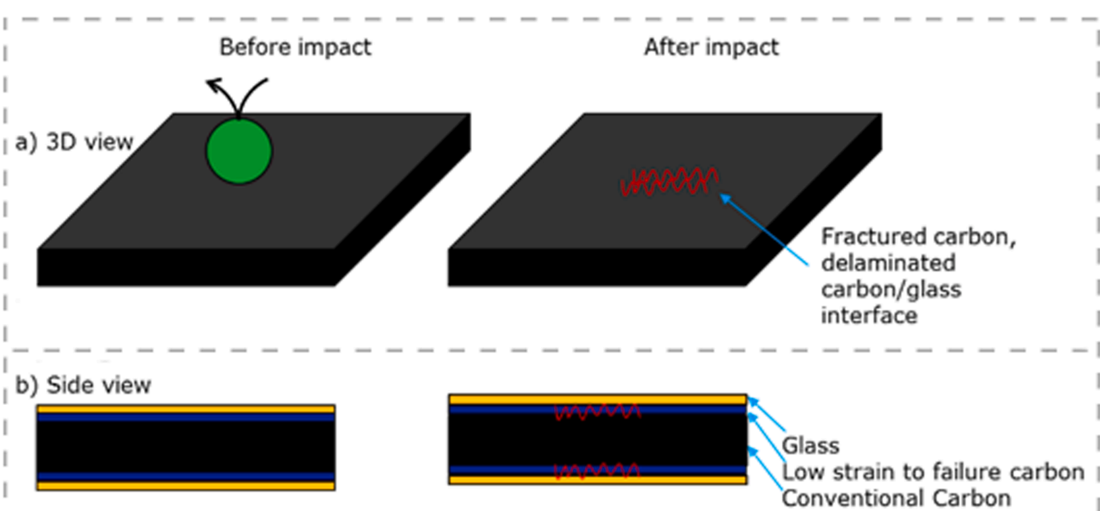

Fig. 28. A carbon/epoxy composite (substrate) with integrated impact detector hybrid composite. a) schematic 3D view, and b) schematic side view of the substrate and the integrated sensor on the sensor [109].

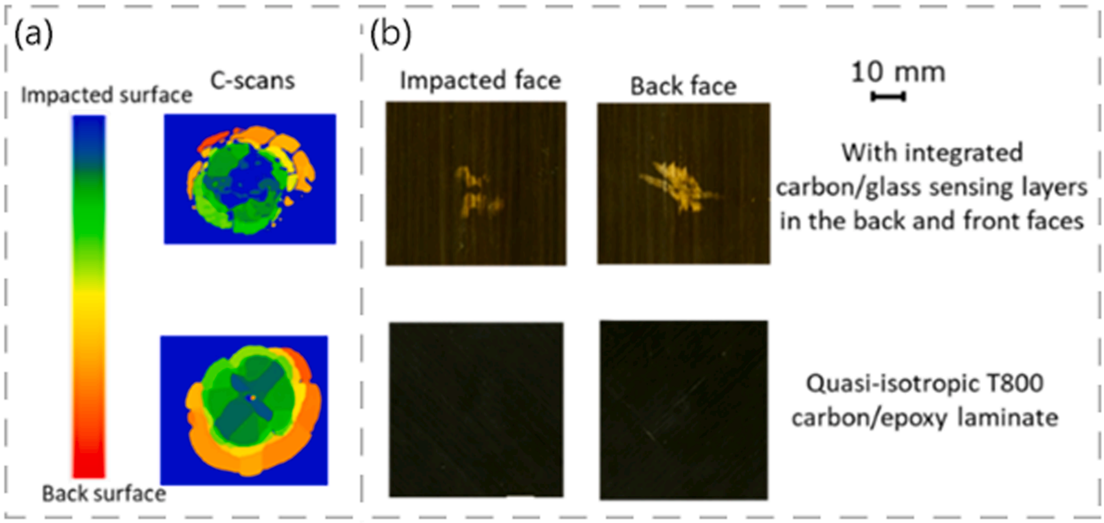

Fig. 29. A carbon/epoxy composite (substrate) with integrated impact detector hybrid composite. a) Schematic 3D view, and b) impacted and back faces of specimens with (up) and without (bottom) sensors [109].

provides background knowledge and highlights the recent progress in self-reporting mechanochromic polymer composites by classifying them into chemical- and physical-based sensors, focusing on their SHM applications.

Mechanochromism based on dye-filled materials is the first approach in chemical sensors in which a dye-containing agent such as microcapsules is embedded into the desired system, and when experiencing strain, it is ruptured and releases its cargo. One strength of this method is its easy-implementation. Double-walled capsules are a new and optimised encapsulation strategy, in which one smaller capsule is contained within another larger capsule and shows greater promise for SHM compared to other encapsulation strategies. These coatings should be as thin as possible, which consequently requires efficient design of self-reporting micro-capsule-based materials. To this end, further research towards the influence of the capsule material, capsule volume fraction, dispersion, and the size as well as the thickness of the shell on the deformation and mechanical behaviour in different polymers is needed. Moreover, research on dual function microcapsules with both self-reporting and self-healing functions is still very limited and there is much room for improvement in this topic.

The second self-reporting approach in chemical sensors is based on modified polymeric and fibrous materials in which desired mechanophores are added to composites to functionalise them. AIE- and Spiropyran-based composites are examples of self-reporting by using functional additives. This method provides a reliable tool to analyse damage progression in FRP composites at the nanoscale. Nevertheless, there are still challenges, especially in processing and applying mechanochromic fluorescent molecules into a polymer matrix, this can be further improved in future studies. Another challenge is that these sensors are not only responsive to mechanical stimuli, but also to temperature and light excitations, which makes strain monitoring difficult as the color change can be due to a combination of mechanical and environmental parameters. Application-wise, this type of sensor has mostly been applied in just polymeric structures, rather than ceramic or metallic ones. Future studies in this field will hopefully involve the design and analysis of mechanochromic functionalised polymers with sensitive and hierarchical responses to different types and levels of mechanical stimuli, to be used in various engineering materials and systems.

The color-changing process in physical sensors, as opposed to the two previous methods, originates from the structural features of the materials. The first approach in physical-based sensors is based on following specific patterns at the nanoscale, to manufacture structural colour materials in which the interaction of different wavelengths of light with the structure can produce diffraction and interference in various directions, leading to well-known iridescent colours. Structural colour materials vary according to structural dimension and refractive index; therefore, a single set of materials can produce various colors which makes these materials more environmentally friendly than chemical-based colour sensors. Despite remarkable achievements, current structural color materials still do not represent a scalable and cost-effective manufacturing strategy that can mimic the complex features of natural creatures. Also, their SHM response is mostly angle-dependent that may hinder the practical application of the sensor. Nevertheless, more advanced mechanochromic photonic crystals for SHM purposes with scalable manufacture, excellent colour perception and angle-independent colour displays are anticipated. This research field would benefit from the development of image analysis software for real-time evaluation of membrane colour changes.

The second physical-based method is related to the composite hybridisation concept, where the reflected light from interfacial damage between two plies can act as a visual sign to detect damage. The manufacturing process of this method is simpler than the previously discussed strategies, and the so-called hybrid composite sensor can act as both a sensing and a repairing or reinforcing system. Also, these sensors can be designed to be direction dependent. These glass/carbon hybrid sensors are simple and easy to implement making them attractive for use in SHM. Practical applications of this method, both in damage monitoring of a bike handlebar and in the repair of a damaged aluminium panel were discussed. So far hybrid composite sensors, have been developed only for static overload sensing, their operation is not reversible, and they are limited to use in specific strain ranges due to the low strain carbon fibre. Currently hybrid sensors cannot be fabricated by conventional hand layup or liquid fusion, due to the thin ply requirement of the sensing layer. Future studies may involve investigating the performance of purposedly pre-damaged sensors in BVID detection, to determine a more precise threshold for the initiation of different damage modes, such as delamination and fibre breakage. Also, hybrid composite sensors should be further tested under various environmental conditions, so that high-performance sensors with a function-oriented design can be manufactured.

A summary of all mechanochromic strategies covered in this review is provided in Table. 1. It should be mentioned that the information provided for each method is relative to the other methods. For example, while there may be reports of reversible visual signals in dye-filled materials, when compared to other methods, it is fair to mention that

Table 1
Summary of mechanochromic approaches presented in this review.

Mechanism	Color origin	Functionalities	Specifications and challenges	Visual signal
Dye-filled materials	Chemical	Self-reporting Self-healing	**Specifications** - easy manufacturing - macro-scale SHM **Challenges** - leakage and diffusion of the encapsulated core - shelf-life and optimal capsule size and volume fraction	- requires UV light - not reversible - angle independent- neither repetable nor continual monitoring (encapsulated coatings)- repeatable and continual monitoring (vascular channels)
Modified polymers and fibres	Chemical	Self-reporting Self-healing	**Specifications** - better SHM by using functional materials such as AIE- or Spiropyran-based additives - implementing AIE method by physical and chemical linking manufacturing strategies - nano, micro and macro-scale SHM **Challenges** - challenging synsthesis process - multi-responsiveness (excited by light, heat, force, etc.)- molecular diffusion and phase separation process (physical-linking strategy)- detailed SHM over desired structures' regions but complex manufacturing (chemical-linking strategy)	- requires UV light - reversible - angle independent - repeatable and continual monitoring
Structural color materials	Physical	Self-reporting Self-healing	**Specifications** - eco-friednly - high temperature tolerance - manufactuing in the form of gels and hydrogels by adding solvents - better SHM by adding graphene or colored platelets - micro and macro-scale SHM **Challenges** - complex manufacturing - scalability for large structures	- requires UV light - reversible - angle dependent - repeatable and continual monitoring
Glass/carbon hybrid composites	Physical	Self-reporting Load carrying	**Specifications** - easy manufacturing - macro-scale SHM **Challenges** - limited strain monitoring range - thermal compatibility of sensing and substrate layers	- does not require UV light - not reversible - angle independent - not repetable but continual monitoring

mechanochromism based on dye-filled materials is not reversible.

4. Conclusions

Based on the comprehensive review presented above, the following conclusions can be made: - It is difficult to make a direct comparison between different mechanochromism strategies and choose one system over the others. The intrinsic features of each approach define potential application scenarios. A more reliable and accurate structural health diagnosis might be expected by combining different SHM techniques. A multi-technique approach could allow the validation or completion of the diagnosis by taking advantage of the redundancy or complementarity of collected data.

- As mechanochromic systems are mostly applied as coatings, some damage types such as impact, indentation, scratches or cuts are best suited to detect. However, all damage mechanisms are apt to occur within the same composite laminate. Therefore, more research is needed to develop mechanochromic-based SHM systems with the capability to differentiate internal damage mechanisms. Moreover, there are still very few studies focusing on the BVID detection of mechanochromic composites, so the topic would benefit from more attention in the future.

- Another topic for future study is the threshold stress. In most cases, current mechanochromic materials do not appear to provide such critical stress levels, though it is a necessary criteria for the design of reliable self-reporting materials. Numerical simulations and experiments should be employed together to systematically analyse the relationship between the visual signals, strain levels and chemical and physical response of smart materials to better design mechanochromic systems with predefined and known threshold stresses.

- The development of scalable and cost-effective production processes for mechanochromic systems is required, before bioinspired self-reporting composites can be applied in SHM practices. To this end, advanced manufacturing technologies such as inkjet printing, roll-to-roll manufacturing and micro melt-processing methods should be developed. In addition, development of new devices for reading, quantifying, and documenting color and fluorescence signals on large surfaces is desirable. A combination of chromogenic material and electronic components makes it possible to continually improve materials and devices that are both smart and efficient. For example, automating the visual cue detection using flying or climbing robots equipped with cameras and fluorescence detection equipment could be used for SHM of large-scale structures such as wind turbine blades, bridges and aircraft.

CRediT authorship contribution statement

Ali Tabatabaeian: Conceptualization, Writing – original draft, Writing – review & editing, Investigation, Data curation. **Sixin Liu:** Writing – review & editing, Visualization. **Philip Harrison:** Writing – review & editing, Supervision. **Erik Schlangen:** Supervision. **Mohammad Fotouhi:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition.

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.

Data availability

No data was used for the research described in the article.

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Appendix

List of abbreviations.
FRP: fibre reinforced polymer.
GFRP: glass fibre reinforced polymer.
CFRP: carbon fibre reinforced polymer.
BVID: barely visible impact damage.
NDE: non-destructive evaluation.
SHM: structural health monitoring.
AIE: aggregation-induced emission.
QI: quasi-isotropic.

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