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Smart and repeatable easy-repairing and self-sensing composites with enhanced mechanical performance for extended components life

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A B S T R A C T

Structural composites with smart functionalities of self-healing and self-sensing are of particular interest in the fields of aerospace, automotive, and renewable energy. However, most of the current self-healing methodologies either require a relatively complex design of the healing network, or sacrifice the initial mechanical or thermal performance of the carbon fibre composite system after introducing the healing agents. Herein, an extremely simple methodology based on commonly used thermoplastic interleaves has been demonstrated to achieve repeatable easy-repairing and self-sensing functionalities, alongside enhanced mechanical performance in comparison with unmodified carbon fibre/epoxy system. Moreover, due to the high glass transition temperature of the thermoplastic, the repairable composites are shown to have an unchanged storage modulus up to 80 °C, solving the previous limitation of repairable epoxy matrix systems with thermoplastics. High retention of peak load (99%) and a decent recovery of interlaminar fracture toughness (34%) was achieved. Most importantly, the mechanical properties remained greater than the unmodified system after four consecutive cycles of damage and healing. Repeatable *in-situ* damage sensing was achieved based on the piezoresistive method. This "new" discovery based on an "old" approach, which is fully compatible with current composite manufacturing, may overcome existing conflicts between mechanical performance and healing functions, providing a new solution to extend components' service life towards a more sustainable development of the composite sector.

1. Introduction

Smart materials with integrated self-sensing and self-healing capabilities, which can monitor the internal damage and restore the mechanical performance upon request, are of great interests for the next generation of composite materials in transport and renewable energy sectors. Broadly speaking, two main self-healing strategies can be classified, namely extrinsic healing (e.g. capsule- or vascular-based methods using a healing agent), and intrinsic healing systems (e.g. reversible chemical bonds or supramolecular interactions). Over last few years, great progress has been achieved in many systems such as bioinspired vascular systems which allow healing agents to flow into damaged areas, akin to the human circulatory system, or intrinsic healing systems based on dynamic bond exchanges [1–10]. Nevertheless, adding either liquid or solid healing agents into the load-bearing components often affects their optimised mechanical performance in comparison with neat composite systems, not to mention the relatively complex procedures involved during the manufacturing phase or non-repeatable healing process, resulting in difficulties in applying many self-healable materials to replace existing composite materials in lightweight structural applications. Clearly, smart self-healing composites that can be manufactured in an industrially scalable manner, and that possess similar or even improved mechanical performance in comparison with commercially available composite systems are of particular interests.

Due to the laminated nature of continuous fibre reinforced composites, excellent in-plane properties are often associated with relatively weak out-of-plane properties, the latter often being the limiting factors in advanced composite designs and applications. Numerous research works have been performed with the aim of improving the out-of-plane properties such as interlaminar fracture toughness, ranging from matrix

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modifications and reinforcement to fibre surface modifications and interlaminar reinforcements with nanomaterials [11–18]. Among various routes, the use of thermoplastic interleaves is of particular interest for both academia and industries due to their excellent reinforcement with good preservation of other in-plane properties and temperature resistance, together with ease of fabrication, preserved low resin viscosity, limited overall weight content, and good compatibilities with current industrial manufacturing processes [19–21].

Recently, the concept of thermoplastic interleaving has been advanced in repairing composite laminates, based on both thermoplastic materials and supramolecular polymers [22–27]. Upon heating, the existence of a deformable phase between carbon fibre plies allows the rebonding process to occur, hence restoring the mechanical performance after crack initiation. Kostopoulos *et al.* utilised a supramolecular polymer layer as interleaves in carbon fibre epoxy laminates, with a more than fifteen times increase in interlaminar fracture toughness comparing with reference panels and a healing efficiency of almost 60% [22]. Cescato *et al.* directly electrospun polycaprolactone (PCL) onto carbon fibre plies and manufactured the laminates, with a healing efficiency up to 31% achieved based on 10 wt% of PCL in composites [23]. Poly(ethylene-co-methacrylic acid) (EMAA) has also been used as interleaves for composites to provide healing of interlaminar damages based on a pressure induced condensation reaction between the epoxy resins and EMAA [28,29]. However, some of these pioneering works either reported a decrease in the original mechanical performance similarly to the microcapsule/vascular methods, or sacrificed the performance of composites in high temperature environments due to the low working temperature range of the interleaves (i.e. supramolecular polymers with a glass transition temperature (T_g) below room temperature). The incorporation of thermoplastics in the rubbery state also leads to reduced flexural properties, in-plane stiffness, and creep resistance [30]. This work aims to overcome these drawbacks by achieving similar repairability using a thermoplastic with a higher T_g (92 °C) than previous works, therefore having no obvious effect on the composite's mechanical properties at ambient or even moderately elevated temperatures. Moreover, our previous study revealed that by utilising polyhydroxy ether bisphenol A (phenoxy), which is an epoxy miscible thermoplastic material in composite laminates, the out-of-plane properties can be significantly improved upon phase separation during curing, with good preservation of other properties thanks to the high T_g of the phenoxy [31].

It is also worth noting that the field of composite repair has not been evolved rapidly over the last years, with many conventional routes such as scarf patch repair, bonded repair and injection repair still dominating the sector [32–35]. For most of these repair techniques, dedicated surface and/or scarf preparations are often required, alongside a strong reliance on the experience and skills of the engineers. Clearly, an easy method to repair composite damage such as delamination, without complex procedures or strong reliance on the personnel, while do not affect the original performance of the system, is of great interests and necessity for the field of advanced structural composites.

To realise a truly comprehensive smart repairable composite, self-sensing functionality is highly desired to both monitor real-time structural integrity and assess the quality of the repairing. Several approaches to detect damage in composites have been presented in the literature, including changes in electrical resistivity or through embedded optical fibre sensory networks [36–42]. Some pioneering works have recently utilised these well-established sensing methods alongside self-healing composites to demonstrate comprehensive sensing/healing systems [43–46]. Our work aims to further these advances in multifunctionality with a novel sensing/healing fibre-reinforced composite by real-time monitoring of through-the-thickness electrical resistance during delamination alongside repeatable repairing.

Herein, we explore the potential of the very successful and simple interleaving methodology to easily repair interlaminar damage with added self-sensing capabilities of the damage. In this study, the existing challenges of interleaving self-healing technology have been successfully overcome by utilising a film of phenoxy as interleaf. Although using thermoplastic interleaves to toughen carbon fibre laminates might not appear too exciting, the fact that laminates can repeatedly be healed or repaired with restored properties that exceed those of baseline carbon fibre epoxy laminates is beyond many expectation (Fig. 1a), opening up a new route for taking easy repairing concepts into industrial structural composite components. Both the recovery of peak load and healing efficiency have been measured for four consecutive damage and repair cycles, while the in-situ electrical damage sensing capabilities have also been monitored, with the aim of developing a truly intelligent materials system with repeatable easy repair and self-sensing functionalities. To the best of the authors' knowledge, no literature has previously reported a change in resistance structural health monitoring technique alongside repeatable composite repairing for an epoxy matrix composite system. These new findings will greatly contribute to the sustainable development of composite industries, extending the service life of composites with easy repair functions.

2. Experimental

2.1. Materials

Polyhydroxy ether of bisphenol A (Phenoxy™) in pellet form were procured from InChem, with an average molecular weight (M_w) of 52,000 Da, a glass transition temperature (T_g) of 92 °C and a melt flow index (MFI) of 4 g/10 min at 200 °C. Stitched cross-ply carbon fibre fabric with an areal weight of 303 g/m^2 was purchased from Formax (FCIM151) and used for interlaminar fracture toughness specimens. To demonstrate the healing efficiency after quasi-static indentation tests, phenoxy interleaves were inserted between every carbon fibre ply of a UD fabric (Sigmatex Ltd. PC236, 315 g/m^2) employed instead of stitched cross-plies for indentation and C-scan tests. For both cases, a two-part aerospace grade epoxy (MVR444) from Cytec Ltd. was used, with a T_g of 195 °C according to the datasheet. A polytetrafluoroethylene (PTFE) release film (12 μm, A6000® Aerovac Systems Ltd) was placed at the mid-plane of the laminates to act as the specimen pre-crack for Mode-I tests.

2.2. Interleaf fabrication and laminate manufacturing

Interleaf fabrication. Thin continuous phenoxy films were prepared by melt-compounding. Pellets of pre-dried phenoxy were first heated to 250 °C within a Collin P3100 E Hot Press and then compression moulded at a pressure of 240 bar. The fabricated interleaves had an average thickness between 120 μm and 140 μm.

Laminate manufacturing. The fabrication and repairing process for the interleaved panel is schematically illustrated in Fig. 1b. 24-ply laminates were fabricated in a $[0/90/90/0]_{3S}$ lay-up. For interleaved laminates, phenoxy films were placed between the two mid-plies of the lay-up, covering approximately half of the mid-ply area, with the remaining mid-ply area covered by a PTFE film to generate the pre-crack. Laminates were fabricated using a vacuum-assisted resin infusion (VARI) technique, the details of which can be found in our previous work [31]. The designed curing cycle consisted of a ramp from 30 °C to 120 °C at 3 °C/min and held for 1.5 hrs, followed by a post-cure of 3 hrs at 180 °C. The calculated estimate of fibre volume fraction for both neat carbon fibre/epoxy and phenoxy interleaved laminates for interlaminar fracture toughness specimens was 54%, with a phenoxy loading of 1.0 wt% in the interleaved composite laminates. For indentation test specimens, 8-ply composite laminates with a $[0/90/90/0]_S$ layup were manufactured, with phenoxy interleaves placed in between each interlaminar region. As mentioned earlier, the same repair process and parameters were applied to these specimens. For dynamic mechanical analysis test specimens, neat carbon/epoxy and phenoxy interleaved test coupons were cut from laminates fabricated in the same procedure as for

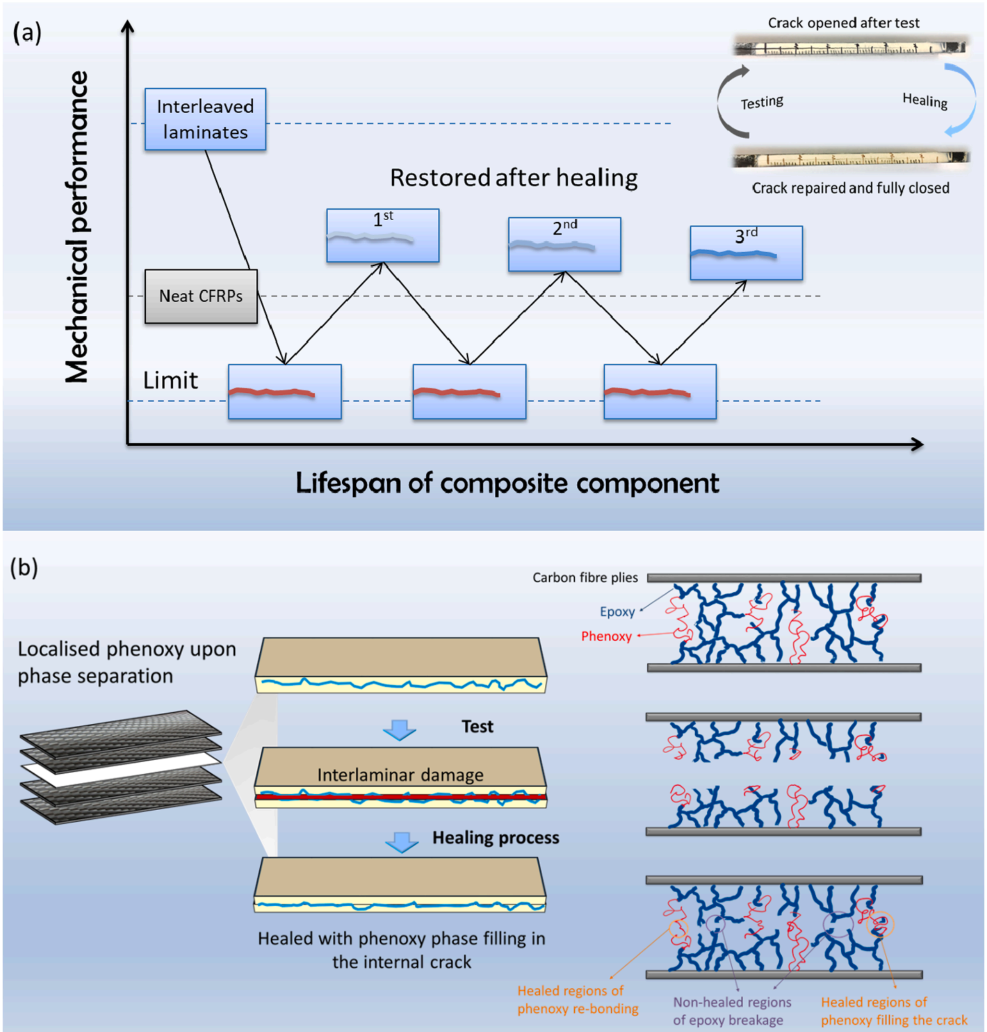

Fig. 1. Schematic illustrations of: (a) enhanced mechanical performance out-performs baseline carbon fibre laminates even after a few repair cycles based on current easy-repair interleaved composites; (b) location, damage and healing process of phenoxy interleaves, showing re-bonding of phenoxy phase and crack filling of phenoxy phase for healing and repair process. Inset in (a) are pictures of specimens after test and repair cycles. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

interlaminar fracture toughness specimens, albeit without the PTFE film pre-crack. I.e., for interleaved specimens, the phenoxy film covered the total midplane surface area. The fibre volume fraction was calculated as 54% and 55% for interleaved and reference specimens respectively.

2.3. Characterisation

Interlaminar fracture toughness and repair. Double cantilever beam (DCB) testing was performed in accordance with ASTM D5528, with specimen dimensions of 130 mm in length and 20 mm in width. Specimens were tested using an Instron 5566 universal testing machine with a 1 kN load cell at a crosshead speed of 1 mm/min. Crack advancement was visually observed using an optical microscope and crack growth was manually recorded, to be subsequently correlated to load–displacement data. As shown in Fig. 1b, to repair the specimens after delamination tests, fractured specimens were placed in the hot press at 180 °C and pre-heated for 10 min, followed by 2 min at 50 bars before cooling to temperature below the T_g of the thermoplastic. Repaired specimens were then retested in the same procedure as previously described. Values of interlaminar fracture toughness were calculated as the average critical strain energy release rate ($G_{IC\text{-}prop}$) in the plateau region of the specimen's R-curve. To compare the repair efficiencies in a like-for-like fashion, a similar region along the R-curve was chosen for each specimen between damage and repair tests in order to calculate the average G_{IC} value. The peak load (P_{max}) sustained by the specimen in each test was directly taken from the load–displacement curves and used to compare the behaviour of the composite between each repair cycle. At least three specimens were tested to obtain average values for each repair cycle.

Fractography. The fractured surfaces of specimens from Mode-I testing were examined through scanning electron microscopy (SEM) using an Inspect™ F from FEI Company (The Netherlands). Specimens were sputter coated with either an Au or Au/Pd target before inspection with a 10 kV accelerating voltage. Specimens were investigated both after the initial test (no repair cycle) and after the 4th repair cycle to compare differences in morphology as a result of the repair regime.

Damage sensing. Electrical resistance method was utilised to perform in-situ damage sensing during Mode-I testing, with the sensing signals to correlate with the internal damage stage of the specimen. Thin copper wires were used as electrodes and attached by a silver loaded epoxy adhesive to the specimen edges. Electrical current would therefore pass through the thickness of the laminate during testing. Resistance values were recorded with an Agilent 34410A digital multimeter connected to a lab-built LabVIEW program.

Ultrasonic scanning of indentation damage. An Instron 5969 equipped with a 50 kN load cell was used for the quasi-static indentation tests, with a 6 mm diameter hemispherical indentor loaded at a speed of 1.25 mm/min. After the introduction of damage from the surface of the laminates, the indentor was unloaded at the same rate, followed by ultrasonic C-scanning using a DolphiCam ultrasonic imaging blackbox with a 5 MHz transducer from Dolphitech AS, Norway, and a scan area of 31 × 31 mm^2. Contact gel was used to improve coupling between the surfaces.

Dynamic mechanical analysis (DMA). A TA Instruments Q800 was used with a three-point fixture and a temperature sweep was chosen to determine the thermal performance of the neat carbon/epoxy and phenoxy interleaved laminates. Specimens were heated from 30 °C to 250 °C at 3 °C/minute, with a frequency of 1 Hz and strain of 0.01%. Average transition temperature values were calculated from the tan delta peaks of three specimens from each laminate.

3. Results and discussion

3.1. Interlaminar fracture toughness and easy repairing

Phase separation of phenoxy/epoxy miscible systems can be expected during the epoxy curing process and resulted in a phenoxy-epoxy co-continuous phase as well as some phenoxy rich regions [20,31,47]. The existence of a phenoxy phase at the interlaminar regions not only enhanced the out-of-plane mechanical performance, but can also act as an efficient healing agent after a crack was generated during the interlaminar failure. As shown from the load–displacement (L-D) curves and summarised G_{IC} propagation values in Fig. 2, the phenoxy interleaves significantly increased the $G_{IC\text{-}prop}$ value, with a more than four times increase from 0.34 kJ/m^2 of the reference laminate to 1.81 kJ/m^2. The peak load values (P_{max}) have also been increased by 110%, from 34.7 N to 72.9 N. These improved out-of-plane properties are consistent with previous studies reported in literature [31]. A stick–slip crack growth behaviour was observed as a result of the thermoplastic toughening from the L-D curves (Fig. 2a), with a decreasing trend of peak load and more stable crack propagation after multiple repairing and testing cycles.

With the phenoxy interleaves in the laminates, interlaminar cracks are closed upon the application of heat and pressure (see Fig. 1) via a rapid repair process to restore their structural integrity. The resistance to crack growth through G_{IC} propagation values for both neat carbon fibre laminates and interleaved laminates were characterised and used as a baseline for comparison on the repairing performance. After the initial tests, both specimens were repaired upon heat and pressure to close the crack, before the specimens were tested again following the same standard testing procedures. The inset images of Fig. 1a show a comparison of the side view of the specimen before and after healing, confirming crack closure after the repairing process.

Considering the nature of this repair process, it is expected that with a higher healing temperature, a better healing efficiency can be obtained due to a higher molecular chain mobility and an easier flow of the thermoplastic rich phase to fill the presented internal cracks. Nevertheless, from a practical viewpoint, repair temperature needs to be set to avoid polymer degradation and to ensure that the localised thermoplastic phase remains within the interfacial regions without "leaching"

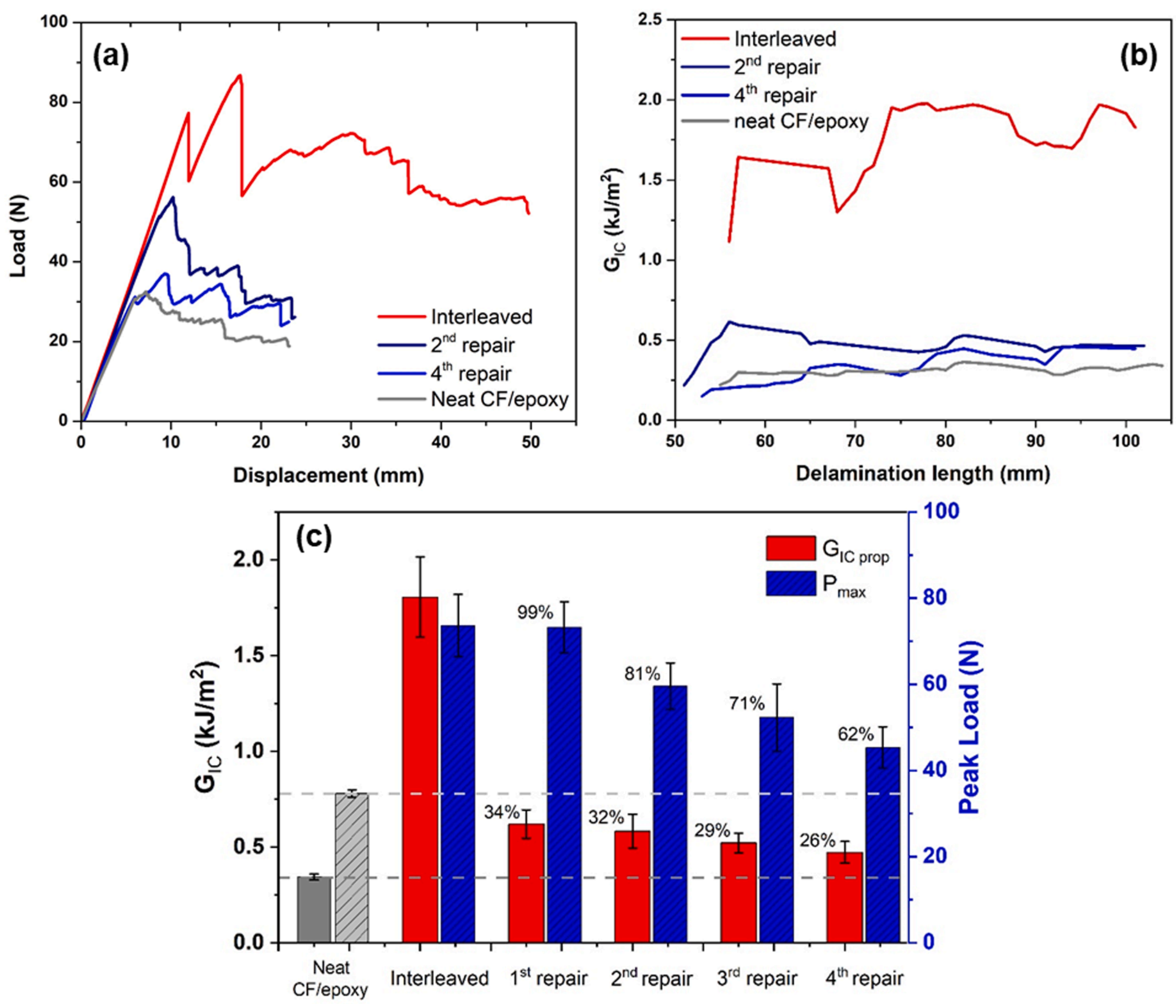

Fig. 2. Interlaminar fracture toughness of composite laminates with significantly enhanced G_{IC} values after introducing phenoxy interleaves: (a) representative L-D curves of interleaved specimens with performance greater than reference laminates after four consecutive test and repair cycles; (b) representative R-curves of specimens after test and repair cycles, with G_{IC} propagation value calculated from the plateau zone; (c) comparison graph of G_{IC} propagation values after each repair cycle, confirming easy repair capabilities with enhanced mechanical properties after 4 destructive test cycles. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

out from the edge of the specimen hence affecting the healing efficiency. As the glass transition temperature (T_g) of phenoxy interleaf is 92 °C, while the T_g of current epoxy resin system is 195 °C, 180 °C was chosen as the healing temperature with the aim of maximizing the healing efficiency.

For the first repair cycle, the G_{IC} value was restored up to 34% of the original values of the phenoxy interleaved laminates, reaching 0.62 kJ/m^2, which is still 80% higher than the interlaminar toughness of the reference carbon/epoxy laminates without modification (0.34 kJ/m^2). The peak load was recovered nearly completely to the original values of interleaved laminates (99%), which is more than double of the neat carbon/epoxy specimens. The repairing capabilities of interleaved specimens are attributed to the thermoplastic nature and reduced viscosity of the phenoxy phase upon heating, leading to better crack healing and crack filling results towards internal damages.

Consecutive testing and healing processes were applied for the interleaved specimens, with the aim to examine the repeatability of healing performance based on the current interleaving concept. With each subsequent repair and delamination, the G_{IC} value continues to decrease, but at a much lower rate, remaining greater than the reference laminates after four complete delamination tests. The healing efficiency values (recovery of G_{IC} value) were reduced to 32% after second repairing, 29% after third time, and reached 26% after the fourth repair cycle. The slight drop in healing efficiency after each cycle can be attributed to two main reasons: (i) for damaged areas where the phenoxy phase cannot reach during the repair cycles, closure of the crack should not be expected; (ii) any fibre breakage during the test cannot be recovered from current method. It is also worth noting that with such a high peak load value, the crack may lead to intralaminar damage or damage in the adjacent interlaminar regions without thermoplastic interleaves, resulting in irreversible damage, especially from the first fracture. Nevertheless, these results demonstrate that phenoxy interleaved composites can be repaired multiple times with a G_{IC}-propagation value greater than the reference carbon/epoxy laminate.

The peak load values decreased linearly after the first repair cycle, reduced to 62% of the initial interleaved laminate value after the fourth repair cycle. It is believed that with each delamination and repair cycle, an increased number of fracture surfaces that cannot be fully re-attached appear, alongside possible reduction of the thermoplastic polymer chain lengths after multiple reheating, which effectively may reduce the molecular weight of the thermoplastic phenoxy along the interface between fracture surfaces. However, it is worth noting that even after four consecutive damage and repair cycles, the mechanical performance of the healed laminates is still higher than that of the neat carbon/epoxy laminates from the initial test. These promising mechanical performances guarantee a second chance for those damaged components to be used as normal after a quick repairing process, greatly reducing both operational and repair costs while extending the components' life with reduced carbon footprint throughout their life cycles. It is worth noting that although the polymer chain interdiffusion and polymer welding at thermoplastic/thermoplastic interface mainly rely on time and heat provided to achieve the healing [48–50], a relatively high pressure was employed in current repair process to promote the crack filling and closure, especially for the aforementioned damaged areas where thermoplastic/thermoset and fibre/matrix interfaces are exposed from the fracture.

3.2. Morphology and fractography

The fracture surfaces of both neat reference and interleaved laminates have been examined using SEM, with images taken from both before and after the repairing process to compare their morphologies. In Fig. 3a, the reference specimen without interleaf highlights a representative fracture surface of a neat carbon/epoxy reference laminate, showing a complete coverage and bonding between epoxy resin and carbon fibres. The smooth surfaces are typical of the brittle Mode-I fracture process associated with highly cross-linked thermoset matrix composites [51].

Fig. 3b shows the fracture surface of phenoxy interleaved specimen after the first Mode-I fracture, revealing a much rougher morphology compared with the reference laminate. Local phase inversion can be seen from the image, with a continuous phenoxy phase covering most of

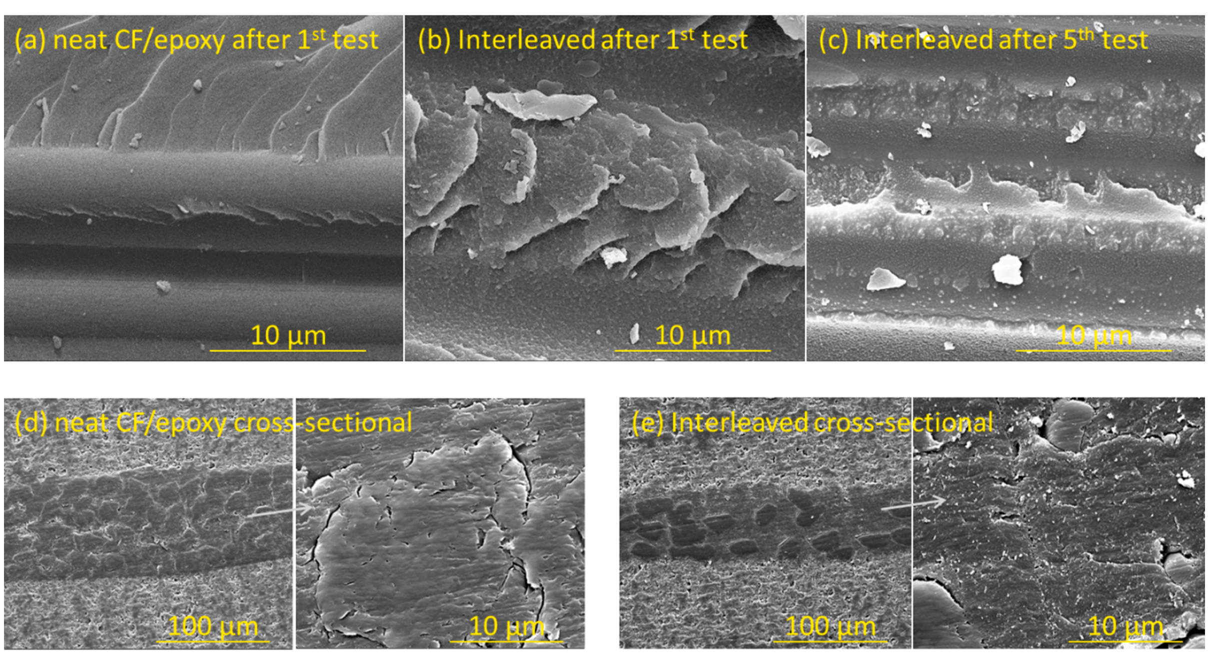

Fig. 3. Mode-I interlaminar fracture surface under SEM: (a) neat carbon/epoxy reference specimen with a smooth fracture surface; (b) interleaved specimens showing a much more ductile fracture surface after phase inversion upon epoxy curing; (c) interleaved specimen after 5th test, showing a similar morphology to 1st fracture with evidence of ductile failure. Polished cross-sectional areas of: (d) reference specimens with smooth epoxy surface; (e) interleaved specimens with phase inversion and plastically deformed morphologies of phenoxy phase (at higher magnification). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the fracture surface. Some evidence of phenoxy debonding from carbon fibre surfaces were also observed (see SI Fig. S1). This is due to the high loading of phenoxy at the interlaminar regions. The continuous nature of the interleaves with limited surface areas has led to a slow dissolution process during the resin infusion and curing process before phase separation, hence a much higher local concentration of the continuous phenoxy phase is formed. The observed morphologies are consistent with literature on phase separation induced toughening in composite laminates [18,20,31].

During the first repairing process (as illustrated in Fig. 1b), the phenoxy phase was able to flow within the damaged areas to fill and close the crack, but with a reduced level of adhesion compared with the phase separation induced interaction between phenoxy and epoxy phases. In addition, unlike the cohesive failure of phenoxy phase which can be repaired upon heating and polymer chain interdiffusion, matrix cracking at difficult to reach regions (e.g. epoxy rich regions with

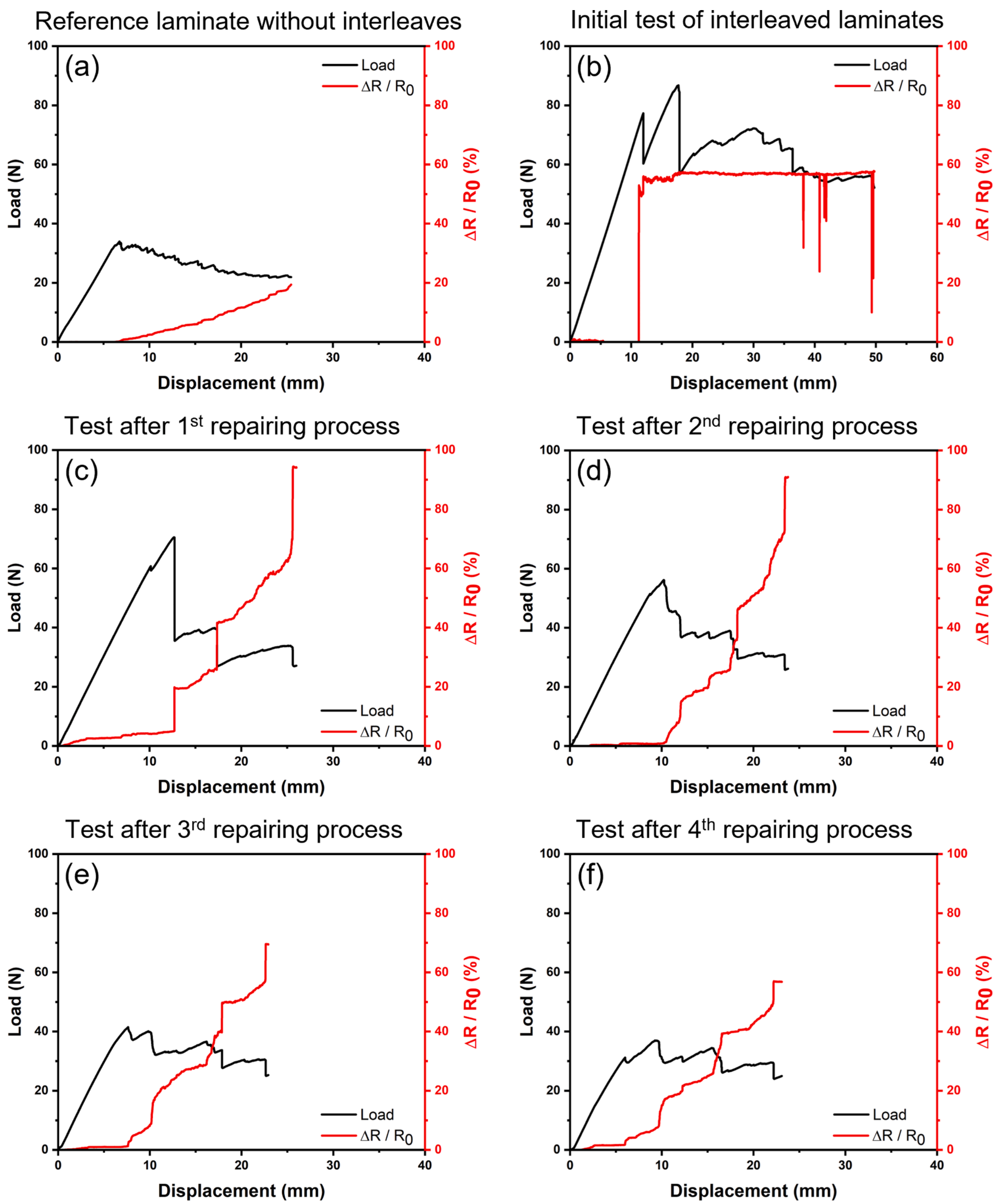

Fig. 4. The in-situ electrical damage sensing of interleaved specimen, showing (a) neat carbon/epoxy laminate without interleaf; (b) initial test of interleaved laminates with no correlation on sensing signals due to the insulating nature of the interleaf; (c) restored electrical sensing signals with good correlation after first repairing process due to improved contact between plies; (d-f) repeatable electrical sensing signals after 4 consecutive damage and repairing cycles, confirming reliable sensing signals to monitor the delamination damages. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

limited amount of phenoxy, or intralaminar regions) cannot be filled easily, hence a reduction in G_{IC} values was observed after the first cycle. This was particularly true for the first test cycle where the peak load and fracture toughness values were significantly increased, resulting in a clear reduction in G_{IC} value after the first repair cycle. From the second fracture onwards, a similar morphology was observed as shown in Fig. 3c and Fig. S2, with a clear trace of phenoxy rich regions from the fracture surface after the 5th fracture cycle. This explains the stabilised trend of G_{IC} values after the first fracture cycle, with similar morphologies and a relatively good coverage of phenoxy phase within the interlaminar regions. The existence of thermoplastic-rich regions is contributing to the significantly enhanced fracture toughness of interleaved laminates with less localized shear yielding around the crack tip, while providing a "meltable" phase to close the crack and repair internal damages upon heating and pressure.

The cross-sectional regions of unfractured specimens were also examined under SEM to reveal the internal microstructure and morphology of the laminates. For the reference laminate (Fig. 3d), a relatively smooth surface after polishing can be observed for the cured thermoset phase, while a co-continuous phase between phenoxy and epoxy can be seen for the interleaved specimen (Fig. 3e), with a much rougher surface from the thermoplastic phase contributing to the improved fracture toughness and embedded repairability.

3.3. In-situ damage sensing

To explore the multifunctionalities of current smart composites apart from easy repairing, the *in-situ* damage sensing functions based on piezoresistive methods was also characterised. Thanks to the electrically conductive nature of the carbon fibres, the electrical sensing signals can be obtained by measuring the resistance of the specimen during the test. Fig. 4 shows the typical load–displacement curves together with the electrical sensing signal, presenting the specimens from initial fracture and after each healing process until the 5th consecutive fracture and repair process (i.e. after the 4th repair). As expected, electrical sensing signals can be obtained from neat reference laminates based on the electrical pathways from conductive carbon fibres bridging the interlaminar regions (Fig. 4a). However, with the introduction of the thermoplastic interleaves between the carbon fibre plies, no sensing signals can be obtained from the interleaved specimen (Fig. 4b) due to the insulating nature of phenoxy film, which inhibited electron travel through the thickness of the laminates. The electrical resistance increased from a few Ohms for the carbon/epoxy reference laminates to a few hundred Ohms of the interleaved laminates, while the measured electrical sensing signals fluctuated from the beginning of the test and maintained at the same level regardless of crack propagation until ultimate specimen failure. This finding is consistent with our previous study on real-time structural health monitoring of interleaved carbon/epoxy laminates [52], but clearly inhibiting the use of in-situ damage monitoring via electrical methods for current composite system. It was shown that the incorporation of conductive nanofillers into the interleaves allowed sensitive signal changes during interlaminar fracture. The incorporation of similar fillers into the interleaves used in this work would allow for damage monitoring during the first test. However, it is worth noting that such addition could decrease the fracture toughness compared to neat interleaving and therefore the repairability of the composite might be sacrificed. A balance between achieving reliable damage sensing signals in the first test with the minimal reduction in mechanical properties and repairability by introducing nanofillers into the interleaves should be carefully considered but is of much interest for further exploration.

Surprisingly, after the first repairing cycle, the electrical sensing signals were successfully restored and can be obtained throughout the subsequent Mode-I test. As mentioned earlier from the morphological observations, some carbon fibres were exposed after the first delamination process (Fig. S1), leading to enhanced electrical conductive pathways through the thickness direction of the laminates especially after the repair process with the applied pressure. This has been confirmed by the measured electrical resistance value of the specimens after first repairing cycle which were reduced to a few Ohms, the same level as reference specimens.

As shown in Fig. 4c, the electrical sensing signals remained at a stable level at the beginning of the test, with a clear step increment when the load suddenly dropped at a crack length of around 12 mm. This jump in sensing signal was due to the internal crack propagation of the specimen. As the test progressed, for each internal crack propagation and load drop in the force curve, a corresponding signal jump is observed for the electrical sensing signal, confirming a good correlation between sensing signals and internal health status. A similar trend was obtained after the second healing process of the composite laminates (Fig. 4d) with clear sensing signals in correlation to load curve and a high sensitivity. Up to the fourth consecutive damage and healing process (Fig. 4e and 4f), the sensing signals remain clear with a good correlation with the load drop, confirming its effectiveness in detecting and monitoring the delamination damage inside the laminates. It is worth noting that the sensitivity of the specimens is slightly reduced with increasing number of damage and repair cycles. For real-world engineering applications, replacement and/or other non-destructive testing should be considered rather than further repairing, should the sensitivity be reduced or unstable sensing signals obtained, although the mechanical performance of the laminates still outperforms the reference carbon/epoxy systems.

3.4. Ultrasonic scanning

To demonstrate the repairing capability for impact induced delamination, laminates with interleaves placed in between each carbon fibre ply were subject to a static indentation test followed by ultrasonic C-scanning to evaluate the damage areas and repairing performance. After inducing a clear indentation damage into the neat reference panel at a load up to around 1.8 kN, the panel was removed from the test frame and scanned, with ultrasonic scanning results shown in Fig. 5a. B-scan images through the thickness of the panel show a clear discontinuity at the front surface, while a delamination area with a diameter of around 6 mm can be seen in the C-scan images (Fig. 5a). After performing the repair process, the same delamination area was observed without any obvious changes (Fig. 5b).

Compared to the reference laminate, a much earlier time-of-flight was obtained from the interleaved laminate (Fig. 5c) due to the presence of continuous phenoxy interleaves between the surface and second carbon fibre plies, reflecting the signals back to the ultrasonic receiver. Clear damage was generated at the surface, with clear fibre breakages observed in the picture in Fig. 5c. A slightly smaller damage area can be found from the C-scan time-of-flight result, with a less pronounced observation in the C-scan amplitude image, although a much higher load (3 kN) was applied for the interleaved laminate to obtain an obvious load drop during the test. After the repairing process, the internal damage area disappeared from the C-scan time-of-flight image in Fig. 5d, with a continuous reflection of signals from the interleaved areas, confirming the feasibility of using the current method to repair composites. It is worth noting that surface damages and fibre breakages (circled area in Fig. 5d) remained unrepaired after the repairing process due to the nature of current interleaving repair concept.

3.5. Dynamic mechanical analysis

To examine the thermomechanical performance of phenoxy interleaved composite laminates, and their capabilities to be used at elevated temperatures, dynamic mechanical analysis was performed to determine the transition temperatures for both neat and phenoxy interleaved laminates. For neat carbon/epoxy laminates, there was no significant change in storage modulus below 200 °C, with a T_g of 227 °C determined

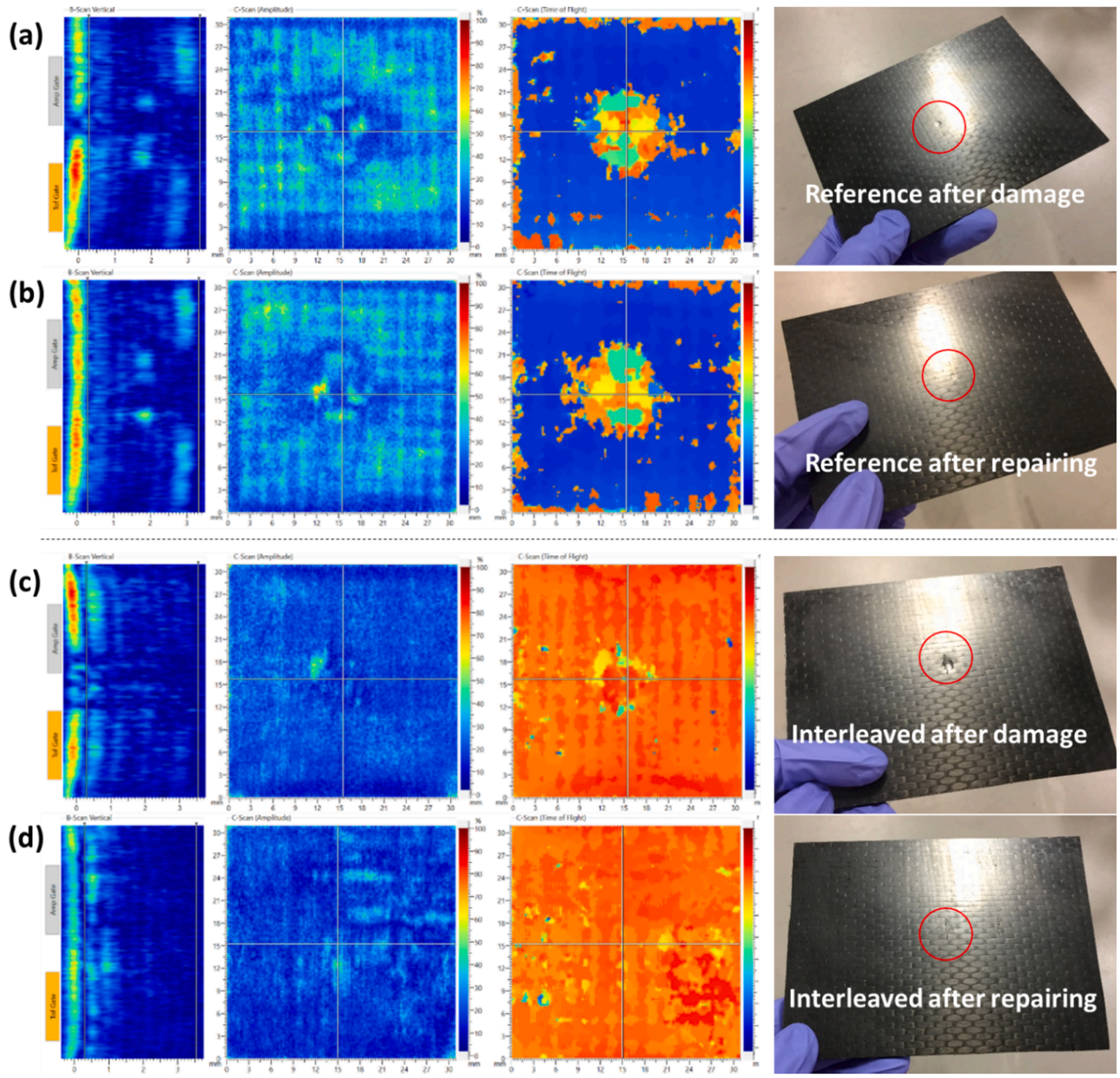

Fig. 5. Demonstration of repairing capabilities by comparing indentation damaged laminates before and after the easy-repair process: (a) and (b) carbon/epoxy reference laminate with the same damage areas without any noticeable changes; (c) and (d) phenoxy interleaved laminate with a clear recovery of damage area after repairing process. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

by the tan δ peak as shown in Figure S3 and Table 1. For phenoxy interleaved three-point bend specimens, the storage modulus remained unchanged compared to the neat laminate (Fig. 6), until near the T_g of the thermoplastic, determined to be 102 °C from the tan δ peak. After this point, the storage modulus is reduced due to the rubbery state of the highly localised and continuous phenoxy phase. The second tan δ peak occurs at the T_g of the epoxy, calculated as 233 °C. Clearly, the phenoxy interleaved laminates demonstrate a similar thermomechanical performance as the neat carbon/epoxy laminate both at room temperature and as high as 80 °C, allowing the interleaved laminates to be used at moderately elevated temperature without reduction in mechanical performance. It is also worth noting that for any specific applications, further analyses should be performed to examine the long-term thermomechanical properties at in-service loading stresses, i.e. creep performance at both ambient and elevated temperatures.

Table 1
Summary of the transition temperature(s) of laminates from DMA tan δ peaks.

Specimen Number	Neat Carbon/Epoxy	Phenoxy Interleaved Carbon/Epoxy	
	T_g (°C)	1st T_g (°C)	2nd T_g (°C)
1	227.5	102.3	232.1
2	226.3	102.8	232.5
3	227.6	102.3	233.5
Average	227.1	102.5	232.7
Standard Deviation	±0.59	±0.22	±0.61

4. Conclusions

A simple process to repair the interlaminar fracture damages in high performance laminated composites has been developed, with restored mechanical properties outperforming those of the neat carbon/epoxy reference laminates, even after four consecutive fracture and repair cycles. Thermoplastic phenoxy interleaves have been utilised as the toughening and healing phase at the mid-plane of the laminates, with a more than four times increase in fracture toughness obtained alongside a good healing efficiency of 34% and high recovery in peak load (99%) upon first repair cycle. Repeatable easy-repairing was successfully achieved by a simple compression moulding process at elevated temperature without any additional healing agents, while both peak load and G_{IC} values remained higher than that of the reference baseline after the fifth test. Integrated *in-situ* damage sensing of current multifunctional composites has also been examined based on the electrical sensing

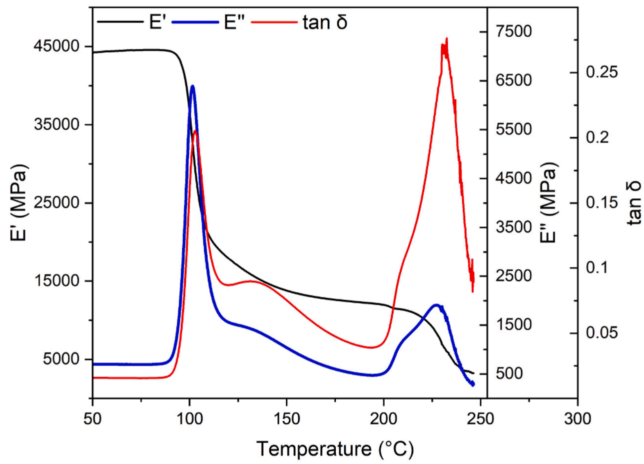

Fig. 6. Representative DMA storage/loss modulus and tan δ plots in three-point bending mode, for a carbon fibre/epoxy laminate containing one phenoxy interleaf film inserted at the midplane of the laminate. Two tan δ peaks are observed, one at 102.8 °C, corresponding to the T_g of the phenoxy, and a further peak at 232.5 °C, corresponding to the T_g of the epoxy phase. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

method, with a clear correlation between sensing signals and internal damage progression to provide useful information for condition-based maintenance.

The incorporation of phenoxy was shown to have no effect on storage and loss moduli up to 80 °C, near the T_g of the thermoplastic. It was demonstrated that the modified laminates have the potential for load-bearing structures even at moderately elevated temperatures and solves a previous limitation of healable composites with reduced thermo-mechanical properties due to the incorporation of thermoplastics.

To explore this simple interleaving concept for future easy-repair composites, a few key points should be borne in mind when designing the system: (i) the T_g of the interleaving materials needs to be within a certain range, e.g. it should not be too low (ideally above room temperature) so that the performance of the composite laminates are not sacrificed at elevated temperatures due to rubbery behaviours and/or low stiffness of the interleaves; (ii) it cannot be higher than the T_g of the cured epoxy resins, so the damaged areas can be filled and repaired by the thermoplastic phase upon heating at the temperature still below the T_g of the crosslinked matrix. A relatively low pressure to close the crack upon heating should be developed, allowing a more practical implementation of current repair system by using vacuum bags and heated blankets for a wider range of applications, especially for those without easy access to both sides of the structures or with curved and complex geometries. The interaction between thermoplastic and thermoset is always an important aspect, especially when adhesive failure at interphase regions, intralaminar cracking or epoxy matrix cracking are expected. Ideally, a cohesive failure within the interleaving thermoplastic phase is favoured for current easy-repair composite systems. It is also worth noting that when introducing a new functionality into a material, it is extremely important to minimise their “side effects” on the original properties. Therefore, when comparing the healing performance, it should not only compare to the system with healing agent before damage, but also the neat system without any modifications. This comparison extends to in-plane and thermomechanical properties when adding thermoplastics to highly crosslinked epoxy matrix composites.

In short, this “new” added function of easy-repairing based on an “old” interleaving concept which is fully compatible with current composite manufacturing could be utilised in various advanced composite systems, providing an industrial feasible and cost-effective solution for many sectors. With the excellent mechanical properties restored after several repairing cycles, alongside the integrated condition-based maintenance capabilities, the components’ service life can be prolonged with reduced environmental impact, contributing to the sustainable development of the composite sector.

CRediT authorship contribution statement

Thomas D.S. Thorn: Conceptualization, Methodology, Investigation, Formal analysis, Data curation, Writing – original draft. **Yi Liu:** Conceptualization, Methodology, Investigation, Writing – review & editing. **Xudan Yao:** Formal analysis, Writing – review & editing. **Dimitrios G. Papageorgiou:** Formal analysis, Writing – review & editing, Supervision. **Paul Robinson:** Methodology, Formal analysis. **Emiliano Bilotti:** Formal analysis, Writing – review & editing, Supervision. **Ton Peijs:** Formal analysis, Writing – review & editing, Supervision. **Han Zhang:** Conceptualization, Methodology, Formal analysis, Investigation, 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

Data will be made available on request.

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Appendix A. Supplementary data

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