Self-healing of structural carbon fibres in polymer composites

Yongjing Wang1, Jake Edgell2, Nicholas Graham2, Nathaniel Jackson2, Hengkun Liang2,3 and Duc Truong Pham2

Abstract: Carbon fibre–reinforced composites (CFRCs) are increasingly used in aeroplanes, satellites and offshore wind turbines. Access those systems for repair when the material is damaged may be difficult. Researchers have incorporated vascular systems containing healing agents into CFRCs, enabling them automatically to recover from delamination and debonding. However, self-recovery of the structural fibres that give CFRCs their exceptional mechanical properties is still impossible. This paper describes a method to make CFRCs self-heal following structural fibres’ damage. This involves automatically delivering epoxy-based healing agents containing short carbon fibres (SCFs) to cracks through an embedded vascular system. Cracks are created by disk-cutting through the carbon fibre layer of CFRC specimens. The SCFs in the released healing agents can be aligned in a local electric field produced by applying a potential to the broken structural carbon fibres. The alignment reconnects the structural carbon fibres. Process parameters were investigated to observe their effects on the healing performance and determine the optimum healing agent composition and conditions. In comparison to using conventional healing agents without SCFs and electric alignment which restored 25.2% of a CFRC’s original strength, employing the proposed approach increased the recovery to 47.3%.

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PUBLIC INTEREST STATEMENT
Carbon fibre–reinforced plastics (CFRPs) are strong and light composites containing carbon fibres as reinforcement for the base polymer material. CFRPs are expensive to manufacture but very useful for devices and systems requiring high rigidity and high strength-to-weight ratio. These include structural components for aerospace, automotive and marine applications, medical instruments, sports equipment, and an increasing number of consumer and technical products. We describe a mechanism for damaged carbon fibres in a CFRP to heal themselves, enabling the material to recover some of its original strength. Our experimental results show that the proposed mechanism can help restore as much as 50% of the original tensile strength. This self-healing capability should extend the service life of CFRP parts and structures, reducing their maintenance, repair and replacement costs and the environmental impacts of CFRP wastes.
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1. Introduction
Self-healing composite materials are artificial materials that can automatically heal after damage. A popular method to enable self-healing is to incorporate into the host material microcapsules or microvessels filled with a healing agent capable of repairing cracks, as illustrated in Figure 1. The design was originally inspired by healing processes in biological systems. The healing agent is released to seal the crack when damage to the material causes the capsules or vessels to rupture.

In the past two decades, developments in self-healing materials have enabled some of the issues with early self-healing materials to be overcome. When self-healing materials were first proposed, the reported percentage recovery was only 60% after a 48-h curing period (White et al., 2001). Nowadays, the recovery has been raised to nearly 100% (Cohades et al., 2018; Wang et al., 2015; D. Y. Zhu et al., 2015). The time required for curing has also decreased to a few seconds due to the use of newly synthesised healing agents (Ye et al., 2014). The vessel-based design enables the refilling of healing agents so that recovery from large-scale damage (White et al., 2014) and multi-cycle damage (Patrick et al., 2014) becomes possible. Recovery can also remain strong and active at very low temperatures as the material is able to maintain its core temperatures using internal heating components (Wang et al., 2016).

However, self-healing carbon fibre–reinforced composite (CFRCs) materials are not yet practically applicable. One significant challenge is the inconsistent healing performance. Damage to fibre-reinforced composites is of two general types: matrix damage which can be intralaminar/interlaminar and fracturing of structural fibres. Established self-healing mechanisms can cure debonding (Cohades et al., 2018; Patrick et al., 2014; Blaiszik, Baginska et al., 2010; Neisiany et al., 2017; Trask et al., 2014, 2007) as it is caused by the fracture of the matrix, which could be repaired by polymer healing agents. Reinforcement particles such as multi-wall carbon nanotubes can be added to healing agents to improve the restored interlaminar strength (Kostopoulos et al., 2018), (Bekas et al., 2017). However, when the fracture of structural fibres (i.e. the “bone” of a CFRC) is the primary type of damage, established healing agents are no longer effective.

This paper reports a carbon fibre–reinforced composite that can repair its structural fibres and restore its mechanical properties after damage. The material has an embedded vascular system,
and the healing agents in the vessels contain short carbon fibres (SCFs) which could be aligned in an in situ electric field produced by charging the broken structural carbon fibres. The paper also presents a model that can be used to analyse the alignment of conductive particles in electric fields (Harper et al., 2010; Khan et al., 2013; S. Wu et al., 2015) and evaluate healing performance. Different variables and parameters were experimentally investigated to observe their effects on the healing performance and determine the optimum healing agent composition and conditions.

2. Methodology

2.1. Healing mechanism
The proposed self-healing CFRC material has embedded hollow vessels containing a liquid healing agent mixed with SCFs. The vessels and structural fibres are aligned with one another (Figure 2). When some of the structural fibres rupture, the same happens to nearby vessels, causing them to release the healing agent, which then flows to fill the gaps between the broken fibres. As SCFs are electrically conductive, they can be aligned to an electrical field generated across the gaps by connecting a power supply to the CFRC material.

The direction of the field is the same as that of the structural fibres as shown in Figure 2, and thus the SCFs will become aligned with them and reconnect the broken fibres. Meanwhile, the healing agent in the gap starts to solidify, freezing the location of the SCFs and enabling them to form permanent links between the broken fibres.

2.2. Modelling the effects of additive short fibres and electric alignment on restored strength
The aim of the model is to estimate the strength of the CFRC material after recovery. The effects of aligned SCFs on the strength of the material can be calculated using the Rule of Mixtures, a theory originally developed to model the properties of short fibre–reinforced composites (Bekas et al., 2017; Darlington & Mcginley, 1975; Papathanasiou et al., 1995; Y. T. Zhu et al., 1997; Van Hattum & Bernardo, 1999). According to this rule, orientation and carbon fibre content are key variables...
affecting material strength. For unidirectional continuous fibre composites under isostrain conditions, material strength and the mentioned variables are related as follows (Fukuda & Chou, 1982):

\[\sigma_c = \sigma_f V_f F\left(\frac{l_c}{l}\right) + \sigma_m(1 - V_f)\]  \hspace{1cm} (1)

where \(\sigma_c\), \(\sigma_f\) and \(\sigma_m\) are the ultimate tensile strength of the composite, the structural fibres and the matrix, respectively. \(V_f\) is the volume fraction of structural fibres. \(C_o\) is the fibre orientation factor that can be calculated by using the Krenchel equation (Nairn, 2005) and its value is given in Table 1. The factor \(F\left(\frac{l_c}{l}\right)\) models the effect of the fibre length and its value is defined in Equation (2), where \(l_c\) and \(l\) denote fibre critical length and average length, respectively. The critical fibre length can be calculated using Equation (3).

\[F\left(\frac{l_c}{l}\right) = 1 - \left(\frac{l_c}{2l}\right) \quad \text{when} \quad l > l_c \quad \text{or} \quad \left(\frac{l}{2l_c}\right) \quad \text{when} \quad l < l_c\] \hspace{1cm} (2)

\[l_c = \frac{\sigma_f d}{2\tau_c}\] \hspace{1cm} (3)

where \(d\) is the diameter of the short fibres and \(\tau_c\) is the greater of the matrix tensile strength and the shear strength of the fibre-matrix interface.

Equations (1–3) were used to calculate the strength of the material after recovery. The values obtained were compared against experimental results, as discussed in Section 3.

2.3. Experiment design

Experiments to validate the new healing mechanisms include an investigation of different variables to observe their effects on the healing performance. Table 2 is an overview table with the performed tests.

Experimentation started with investigating the effect of SCFs on the strength of materials after recovery. The effect of the short carbon fibre content of the healing agent was explored to establish trends.

The effect of increasing the strength of the electric field was also studied. This was done by altering the voltage producing the electric field, and comparing the restored strength of specimens healing in electric fields generated by using 9 V, 20 V and 50 V pulse power supplies. Pulses were used to avoid burn damage to specimens through electrical over-heating. A pulse was a spike of voltage of 9 V, 20 V or 50 V that decayed to 0 V over a period of 2 seconds.
Healing agents containing carbon fibres of differing lengths were tested to reveal the effects of fibre length on self-healing performance. The healing performance of an agent containing 150 µm (avg.) length short fibres was compared to that of an agent containing a different batch/brand of 100 µm (avg.) fibres. Finally, the addition of a dispersant was tested to see its effect on the resultant healing performance.

### 2.4. Fabrication of samples

The method to fabricate CFRC samples incorporating 2 mm-internal-diameter hollow vessels is shown in Figure 3. Epoxy resin and hardener (Very High Temperature Epoxy Laminating Resin, EasyComposites Ltd.) were mixed in a vacuum chamber as instructed by the supplier (EL160 High Temperature Epoxy Laminating Resin—170C HDT—Easy Composites, 2019). To make hollow vessels, Nylon fishing lines 2 mm in diameter were fixed on the table using an adhesive tape. The uncured resin was poured to immerse the fishing lines. This was followed by curing at room temperature for 24 h. After curing, the nylon lines were pulled out carefully, leaving hollow vessels inside the material. It has been experimentally found that having such a configuration of hollow vessels only reduced the tensile strength of composites very slightly (Kousourakis & Mouritz, 2010). Prepreg carbon fibre cloth fabrics (CF-PL-200-100, EasyComposites Ltd.) (Carbon Fibre Cloth Fabric Plain Weave, 3k, 200gsm—Easy Composites, 2019) were laid onto the cured resin. After another 24 hours of curing at room temperature, post-cure heating cycles were applied as instructed by the supplier. The fully cured composite which had a thickness of

![Figure 3. Fabrication procedure. Samples were cut into 22 mm × 6 mm specimens. Each specimen contained a vessel.](image-url)
3 mm was cut into identical pieces (22 mm × 6 mm). Each specimen contained hollow vessels and a carbon fibre–reinforced layer on top.

2.5. Healing agent composition and synthesis

In a self-healing material, it is common to use a multi-part curing system (e.g., resin and hardener) with the different parts loaded into separate vessels. Fracturing of the composite causes the vessels to break and release the healing agents the mixing of which causes them to solidify. However, the process is unpredictable due to the local mixing ratio being uncontrolled. To ensure consistency and obtain a true measure of the effect of short fibre alignment, it was decided to employ an identical pre-mixed combination of resin and hardener for all samples in the experiment.

The healing agent was made up from an epoxy resin base (RT151, Resintech Ltd.), short carbon fibres (Carbiso Milled Carbon Fibre, EasyComposites Ltd. and AGM 99–150, Asbury Graphite Mills, Inc.), and dispersant (DISPERBYK-191, BYK-Chemie GmbH). Resin, short fibres and any other additives were mixed in a magnetic stirrer for 1 h using a set speed at 1000rpm, followed by the addition of the hardener and mixing for another 10 min before being injected into the embedded vessels. The concentration of the resin and the hardener followed the instruction by the supplier (resin:hardener = 4:1 by weight) (Resintech Ltd).

2.6. Experimental procedure

For each specimen, the experiment started with cutting through the carbon fibre layer of a specimen using an air grinder to produce a crack as shown in Figure 4 where the depth of the cut was approximately 1 mm. Then, the healing agent was injected into the vessels using a pump, after which the specimen was connected to a pulse power supply for 24 hours at room temperature. The applied potential produced an electric field in the gap to align the SCFs. In the meantime, the healing agent solidified to freeze the position of the aligned SCFs.

Afterwards, the healed specimens were disconnected from the power supply and left for 24 hours in an oven (40°C) to ensure the healing agent was fully cured. The tensile strength of the specimens after recovery was evaluated at room temperature using an MTS Criterion testing machine (MTS Criterion Model 43 Electromechanical Universal Test System), following ASTM D3309 testing standard with a loading rate of 3 mm/min. One-way and two-way analyses-of-variance (ANOVA) adopted from (J. Wu et al., 2016) were performed to detect the significant effects.

3. Results and analysis

3.1. Effects of SCF addition

Equation (1) was used to estimate the strength of the material after healing. Data provided by suppliers indicate that $\sigma_m$ should be in the range 55–68 MPa, $\sigma_f$ = 3150 MPa, $d$ = 7 μm and $\varepsilon_c$ = 75 Mpa. Hence, $l_c$ is calculated to be approximately 147 μm (Equation (3)).

Healing agents containing SCFs ($V_f$ =13.8 Vol%) were compared to a pure epoxy resin to observe the effects of SCFs. Another group of specimens were connected to a 9 V power supply. It was expected that specimens with SCFs aligned by the applied voltage should show better recovery. The test results and calculation results are shown in Figure 5.

As shown in Figure 5, a healing agent containing short fibres increased the restored strength to 37.2%. The recovery in tensile strength incurred by adopting a healing mechanism is significant ($F$ =16.05, $F_{crit}$ =3.24, $p <0.001$; one-way ANOVA). Considering the Non-SCFs group and the two SCF groups only, the effect of adding SCF contents is also significant ($F$ =10.10, $F_{crit}$ =3.89, $p =0.003$). The calculated results based on Equation (1) gave a good prediction of the recovery strength. In terms of the SCFs-Charged group, the minimum value was based on the assumption that $C_o$ did
not increase, or the fibres were still in a random state \( (C_o = 0.2) \), as the orientation of SCFs in the charged specimens was difficult to measure.

### 3.2. Effects of electric field intensity

The inconclusive differentiation between the charged and uncharged specimens suggests that the 9 V voltage was not high enough to allow consistent alignment of the SCFs. Increasing the voltage raises the intensity of the electric field, thus producing stronger forces for fibre alignment which should improve healing performance. Three different pulse voltages, 9 V, 25 V and 50 V were employed. The results obtained are given in Figure 6.

Figure 6 shows that there is a strong correlation between increasing the voltage and achieving a higher healing performance, with the strengths of the 50 V aligned specimens being higher than those of 9 V and 25 V aligned specimens. As discussed previously, specimens that are not aligned and those aligned by the 9 V power supply have similar recovery performance. Hence, \( C_o \) for the 9 V samples should also be near 0.2 (Table 1 and Figure 5). When the voltage was raised to 50 V, the growth in healing strength indicated that \( C_o \) was increased to near 0.31. The reconnection process can be completed in 1 h as shown in Figure 7.

It is worth noting that alignment using a constant power supply was originally attempted. However, specimens were subject to moderate burn damage due to excessive electrical heating.
Figure 6. Effects of voltage on the performance of self-healing.

Figure 7. Reconnection of a bundle of fractured carbon fibres under electric alignment.
which significantly reduced mechanical strength. The use of a pulse power supply allowed the samples to dissipate heat fully in the resting time in each period to avoid thermal damage while also enabling the use of voltages that were high enough to align the SCFs.

### 3.3. Effects of short carbon fibre ratio

Increasing the volume content of SCFs, $V_f$, should allow more fibres to be aligned and the recovery to be stronger. Three different volume contents, 10.1 Vol%, 12.2 Vol%, 13.8% Vol% were tested. The results of the experiment and calculated values are given in Figure 8 which shows that increasing the SCF content in the healing agent increases its healing performance by a significant margin. There is a significant difference in tensile strength with the addition of SCFs from 0 Vol% to 13.8 Vol% ($F =13.65$, $F_{crit} =3.24$, $p <0.001$; one-way ANOVA). Considering the range from 10.1 Vol% to 13.8 Vol% only, the effects of the incremental increase of SCF contents are also significant ($F =6.26$, $F_{crit} =3.88$, $p =0.014$).

However, it is worth noting that the addition of SCFs increases the viscosity of the healing agent, preventing it from flowing to damaged areas to repair them. A volume content of 17.6 Vol% was originally planned to be tested, but it was deemed too viscous to be applied and could not be used. On the other hand, volume content less than 6.6% produced no observable effects.

### 3.4. Effects of short carbon fibre length/quality

As $\sigma_f$ and $F\left(\frac{L}{f}\right)$ in Equation (1) are related to fibre quality and length, respectively, altering their values could lead to different healing performances. The aim of this experiment was to determine their effects.

100 µm and 150 µm SCFs, manufactured by AGM, were compared to 100 µm SCFs, manufactured by EC. For EC SCFs, the $\sigma_f$ is 3150 MPa which is higher than $\sigma_f$ for AGM SCFs (2800 MPa). The calculated and test results are shown in Figure 9.

Figure 9 shows that the strength of the 150 µm length specimens is higher than that of the 100 µm AGM and 100 µm EC specimens. It also appears that the 100 µm EC specimens have more consistent load values. All experimental results are in accordance with the calculated results. They confirm that the adoption of longer carbon fibres led to higher recovery strengths.

### 3.5. Effects of dispersant

This experiment was to examine whether adding a liquid dispersant to the healing agent mixture would improve its healing performance. The motivation of the study came from Equation (1) which shows that increasing the key factor $C_p$ could lead to a better healing performance. This is because the addition of a dispersant reduces viscous resistance during the alignment of SCFs, thus making

![Figure 8. Effects of SCF content ratio on the performance of self-healing.](image-url)
the reconnection easier. The dispersant, BYK-191, was used in an established study regarding the alignment of carbon nanofibres (Ladani et al., 2015). Specimens healed with a healing agent containing 3 wt% BYK-191 were tested.

As can be seen in Figure 10, adding a dispersant to the healing agent does not improve the healing performance. The results were not as expected possibly because, although the viscous resistance was reduced, the modified mixture could also reduce the shear strength of the fibre-matrix interface, $\tau_c$ in Equation (2), which caused the factor $F\left(\frac{1}{2}\right)$ to decrease. As Equation (1) shows, overall healing performance depends on the proportion of $F\left(\frac{1}{2}\right)$ and $C_0$.

3.6. Overall performance
The improvement in the restored strength is shown in Figures 9 and 10, which indicates that using conventional healing agents without SCFs and electric alignment restored 25.2% of a CFRC's original strength, while employing the proposed approach increased the recovery to 47.3%.
3.7. Future work
Based on the model in Section 2.2, future work could further improve the healing performance by increasing volume fraction of structural fibres $V_f$, fibre length factor $F\left(\frac{L}{l}\right)$ and orientation factor $C_o$. In this study, the healing performance reached 47.3% when $\sigma_f = 2800\text{MPa}$, $V_f = 0.138$, $F\left(\frac{L}{l}\right) = 0.5$ and $C_o = 0.31$.

- High-strength carbon fibres with $\sigma_f$ near 6000 Mpa can be adopted.
- $F\left(\frac{L}{l}\right)$ can be increased to 0.6 by employing longer fibres or modifying the surface of the carbon fibre to reduce the critical length $L_c$.
- $C_o$ may be raised to 0.5 by adopting better pulse charging strategies with high voltage spikes and longer resting times.

The above changes may give a healing performance 100% (Equation (1)), suggesting that a CFRC material could fully recover from damage to structural fibres.

4. Conclusion
Significant improvements in the automatic recovery of fibre-reinforced composites after damage to structural fibres have been observed through the introduction of SCFs and the application of an electrical aligning field. A healing agent made with pure epoxy resin was compared to a healing agent containing epoxy resin, 13.8 Vol% short carbon fibres (150 μm in length) aligned in an electric field. Of the original tensile strength, 47.3% could be restored after the structural carbon fibres were completely ruptured. This paper also demonstrated the use of the Rule of Mixture to predict recovery performance. The theory suggests that further improving the alignment process and using stronger short fibres to promote recovery are promising avenues for future work.

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Author contributions
Y.W. contributed to the initial conception, theoretical and experimental data analysis, results presentation and interpretation and drafted the manuscript. J.E. and N.J. contributed to the experimental work and the preparation of the manuscript. N.G. proposed the theoretical analysis method. H.L. was involved in designing the presentation of the manuscript. D.T.P. supervised the research and contributed to the interpretation of experimental results and manuscript writing.

Data accessibility
The methods and materials used in this work are described in the body of the manuscript.

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