In situ Thermal Remending of Multimaterial 3D-printed Thermoplastic Composites

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1. Abstract

Extrusion-based additive manufacturing (AM) of reinforced thermoplastic composites (TPCs) offers high-strength, recyclable structures, yet remains limited by interlayer delamination. Integrating biologically inspired functionalities in these composites could repair such damage *in situ* with minimal human intervention. By embedding low-melting-point thermoplastic copolymers alongside conductive TPCs, Joule heating can thermally remend damaged regions while preserving global structural integrity. This research aims to develop a multimaterial engineering-grade structure with *in situ* thermal remending capability to address interlayer delamination. The selected host composite is a short glass fiber-reinforced polyamide (PA-GF), with polyethylene-co-methacrylic acid (EMAA) as the healing agent and carbon black-reinforced polylactic acid (PLA-CB) for the embedded heaters. Double cantilever beam (DCB) composite specimens are manufactured and tested in mode-I fracture. After crack closure, the heaters are activated *in situ* by applying constant electrical power for 20 minutes, followed by cooling to room temperature by natural convection. The temperature profile obtained by a thermal camera shows a maximum surface temperature of 117°C at the delamination zone, exceeding the healing agent's melting temperature, while staying below the glass transition temperature of the structural composite—thereby maintaining structural properties during the healing event. To assess repair, the specimens are then reloaded at the same loading rate to the same crack length as the first virgin cycle. Repair efficiency is defined as the ratio of the mode-I critical strain energy release rate (G_{IC}) in the repaired state compared to that of the virgin state. The representative specimen undergoes 11 successful repair cycles, with an average repair efficiency of 95%, demonstrating the method's repeatability in repairing interlayer delamination. Overall, multimaterial AM can integrate thermal remending functionalities in TPCs with simplified single-step production. This approach eliminates the need for manual repair, preserves the geometrical integrity, and extends the lifecycle of structural composites.

2. Introduction

Material-extrusion additive manufacturing (MEAM), specifically fused filament fabrication (FFF), is the process of depositing thermoplastic filaments layer by layer. FFF offers high speeds and low costs in comparison to other additive manufacturing methods [1] and can be highly customized with minimal waste generation and possible recyclability [2]. Thermoplastic composites (TPCs) exhibit functional properties when reinforced with additives, many of which are commercially available as filaments for FFF. Short fiber reinforcements with an aspect ratio (length/diameter) of ~21:1 [3] increase strength and specific modulus [4]. Conductive fillers such as carbon black (CB) introduce electrical conductivity for 3D-printed electronics applications [5].

Despite its advantages, a common drawback of FFF composites is their limited interlayer adhesion [6] and a lack of fiber spanning across printed layers, which leads to mechanical anisotropy and interlayer delamination in mode-I fracture. While printing parameters, process conditions, and 3D-printing equipment can be improved to enhance mechanical properties, extending the durability of TPCs through repair can reduce waste and avoid end-of-life challenges. Recycling TPCs is a potential solution to these challenges, but it adds multiple sorting and processing steps that consume time and energy, making repair options more valuable from a circular economy perspective [7].

Repair techniques for TPCs often rely on heating the material above its glass transition temperature (T_g) or melting temperature (T_m). Thermal repair facilitates molecular chain mobility and reentanglement in the thermoplastic matrix. Conventional *ex situ* heating methods, such as induction welding or resistance welding, have been achieved with applied pressure to ensure the fractured surfaces are in intimate contact [8]. However, such techniques require specialized external equipment and the disassembly and transport of components to a dedicated repair facility for processing. Furthermore, they are more practical for surface fusion bonding than for repairing interior damage in a structure, as applying external heat to a TPC up to its T_g or T_m risks deforming the geometry and losing structural properties.

Studies have proposed the use of low-melting-point thermoplastics as a healing agent for thermal remending. As an example, poly(ethylene-co-methacrylic acid) (EMAA) has been incorporated in thermoset composites due to its low melt viscosity and flowability in cracks, as well as its chemical functional groups that increase its reactivity [9]. Moreover, integrated resistive heaters have shown effective focused heating at the damage zone [10]. By locally increasing the temperature above the melting point of EMAA but below the T_g of the composite laminate, repeatable thermal remending has been achieved up to 1000 cycles [11]. To the authors' knowledge, such repeatable and localized thermal remending mechanism has not been exploited in 3D-printed TPC structures. Therefore, this study aims to develop a multimaterial system equipped with *in situ* thermal remending capabilities to repair interlayer delamination in 3D-printed engineering-grade TPCs.

3. Methodology

3.1. Materials and Specimen Fabrication

The experimental framework consists of a multimaterial system fabricated in a single-step manufacturing process using a multimaterial 3D printer (Prusa XL 5T). As shown in a conceptual schematic in Figure 1, this method deposits filaments at various temperatures using multiple printing toolheads. Compared with single-nozzle 3D printing, this sequential deposition avoids nozzle blockage and temperature incompatibility. Furthermore, this process enables the deposition of functional materials into custom 3D geometries. Short glass fiber-reinforced polyamide (PA-GF, FibreX Nylon 12+GF30, 3DXTECH) serves as the primary structural host composite.

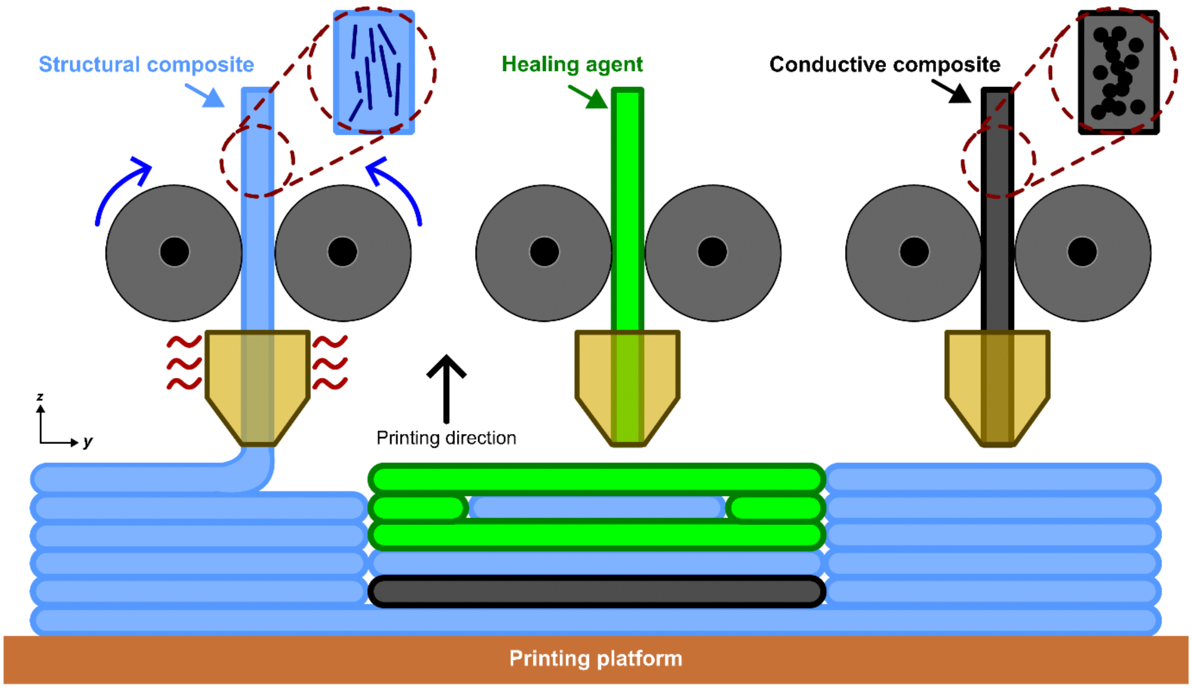

Figure 1. *Schematic demonstrating multimaterial extrusion additive manufacturing using multiple toolheads. The structural composite filament (in blue) is reinforced with short glass fibers, the healing agent filament (in green) is a neat polymer, and the conductive composite filament (in gray) is reinforced with carbon black to pass electrical currents.*

To prepare the healing agent, raw EMAA pellets (Nucrel™ 2940, DuPont, Inc.) are processed into 3D-printing filaments using a single-screw extruder (Filament Maker 2, 3devo B.V.). Carbon black-reinforced polylactic acid (PLA-CB, Protoplant, Inc.) serves as the embedded resistive heating elements. These constituent materials are assembled into double cantilever beam (DCB) composite specimens, specifically designed to evaluate fracture behavior and repair potential at the mid-plane interface. Pinholes are directly incorporated into the 3D-printed structure to ensure precise loading alignment.

The internal architecture is designed to maximize healing efficiency while maintaining structural integrity. Two symmetric PLA-CB heating elements are located on either side of the mid-plane interface. The EMAA healing agent is deposited along the mid-plane across the anticipated crack region in a serpentine pattern, achieving a 36% surface coverage of the fracture area. The healing agent is printed into channels with a width of 1.44 mm embedded within the PA-GF matrix. To ensure controlled crack initiation during mechanical testing, each DCB specimen is printed with an integrated 50 mm pre-crack and side grooves to guide the crack along the midplane.

3.2. Mechanical Characterization and Thermal Repair Procedure

Mechanical characterization is performed through mode-I fracture testing in accordance with the ASTM D5528 standard using an MTS Insight electromechanical testing machine equipped with a 1-kN load cell. During this phase, the DCB specimens are subjected to a constant displacement loading rate of 5 mm/min until a predetermined crack length of 50 mm is achieved. Subsequently, specimens are unloaded at the same rate until they return to a zero-crosshead displacement. The mode-I critical strain energy release rate (G_{IC}) is calculated using the area method and defined as:

$$G_{\mathrm{IC}} = \frac{\Delta U}{b \Delta a}, \tag{1}$$

where b represents the width of the fractured zone and Δa is the measured crack length. ΔU represents the energy required to produce fracture and is calculated as the area under the force-displacement curve [10].

The initial loading cycle establishes the baseline fracture toughness and provides the reference state for subsequent repair assessments. Immediately after the completion of the virgin cycle, the thermal remending process is performed in the load frame by activating the embedded PLA-CB heaters with a constant electrical power of 12 W for 20 minutes. Throughout this phase, the temperature profile is monitored in real time using an infrared thermal camera (FLIR E54) to ensure precise control of the heat distribution. The heating protocol is designed to reach a surface temperature of 110-130°C in the delamination zone. This specific temperature threshold is selected to exceed the melting temperature of the EMAA healing agent while remaining strictly below the glass transition temperature of the structural PA-GF matrix. Following the heating cycle, the specimens are allowed to return to room temperature via natural convection for 45 minutes.

To quantify the effectiveness of the *in situ* repair, the specimens are reloaded under identical experimental conditions, using the same loading rate and target crack length as in the initial virgin cycle. The healing efficiency $\hat{\eta}$ is defined as:

$$\hat{\eta} = \frac{G_{\mathrm{IC},repaired}}{G_{\mathrm{IC},virgin}} \times 100\%, \tag{2}$$

which indicates the ratio of G_{IC} in the repaired state to that of the virgin state [12] as a numerical assessment of restored interlaminar toughness.

4. Results and Discussion

The mechanical performance durability of the system is characterized through a series of consecutive fractures and remending sequences. Figure 2a shows the loading condition and thermal imaging of the DCB specimen. The geometry and placement of the two symmetric PLA-CB heating elements on either side of the mid-plane interface are highlighted in the inset of Figure 2a. The thermal profile shows highly localized Joule heating in the delamination zone while the specimen is held at zero-crosshead displacement.

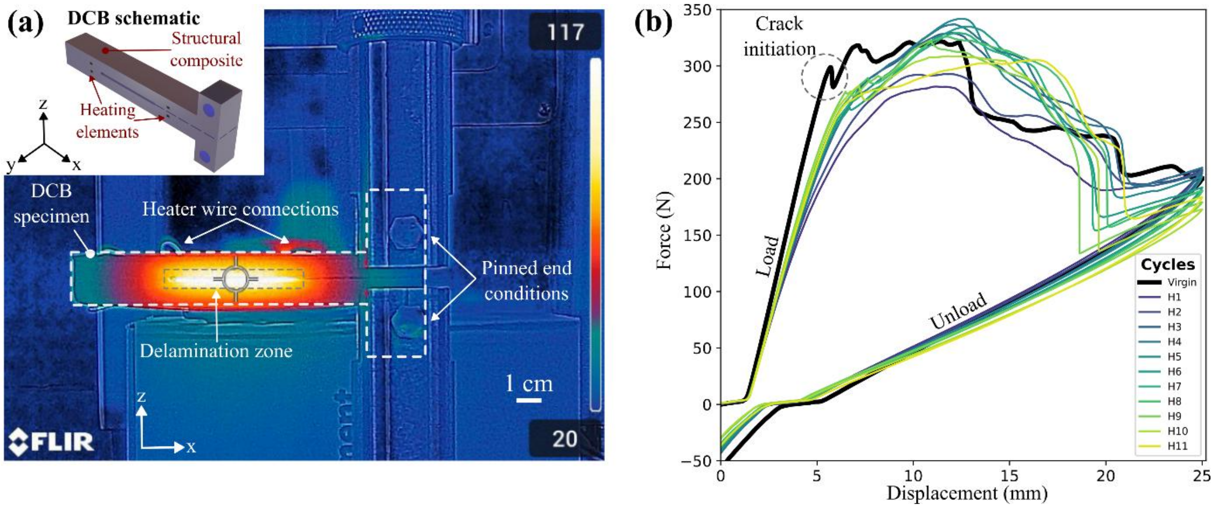

***Figure 2. DCB thermal remending cycles. (a)** Thermal image of the experimental setup during in situ Joule heating at zero crosshead displacement (inset: specimen schematic), **(b)** Force-displacement fracture response of the specimen evaluating the virgin and 11 subsequent thermal remending cycles, showing repeatable fracture response recovery.*

The efficacy of this localized thermally activated healing is reflected in the force-displacement response shown in Figure 2b. Initially, in the virgin cycle, the curve begins with a linear elastic response, leading to crack initiation at the precrack ending tip. As the crack initiates, the load eventually decreases as the crack propagates stably, reflecting the fracture resistance of the interlayer interface. In subsequent healed cycles, a similar fracture pattern appears. However, a notable difference is the slight deviation from linearity observed prior to the peak load. Specifically, in the virgin cycle, the structural composite and EMAA undergo cohesive interlayer fracture. By contrast, during the healed cycles, because EMAA covers the fracture surface, the crack onset becomes blunted. The nonlinearity is thus attributed to the plastic deformation and cohesive fracture of EMAA.

The representative specimen demonstrates significant repeatability, undergoing 11 successful repair cycles with an average healing efficiency of 95% relative to the initial virgin fracture toughness. Notably, such a high repair efficiency is achieved with a heating duration of only 20 minutes, representing a significant reduction in processing time compared to most traditional repair methodologies, which take hours [13] or days [14].

While the fracture response recovery is highly repeatable, a slight reduction in stiffness is observed in the load-displacement curve before fracture in the repaired states compared to the virgin state. This shift is attributed to the inherent material properties of the EMAA healing agent, which occupy the crack interface to facilitate bonding; its lower elastic modulus relative to the structural PA-GF composite results in a slightly more compliant response during reloading. Despite this minor change in stiffness, the overlap of the curves in Figure 2b highlights a consistent recovery of the peak load and G_{IC}, indicating that the EMAA maintains its functional bonding capacity over repeated melt-solidification cycles.

5. Conclusion

The ability to achieve and maintain 95% repair efficiency over 11 consecutive healing cycles suggests that *in situ* thermal repair using a thermoplastic healing agent can substantially extend the operational lifecycle of structural composites. By embedding resistive heating elements and healing agents during the primary fabrication process, the production workflow is simplified compared to traditional post-processing repair methods. The proposed approach offers an alternative to manual repair interventions, which are often labor-intensive and technically challenging. Localized thermal activation ensures that the global geometric and mechanical integrity of the engineering-grade structure is preserved throughout the healing process, enabling the component to remain in the service environment. Ultimately, this research demonstrates that multi-MEAM is a viable pathway to integrate autonomous thermal remending functionality directly into TPC structures.

Despite the repeatable repair efficiency, certain challenges remain for broader industrial adoption. The current selection of EMAA as a healing agent is limited by its relatively low melting point. Consequently, for high-temperature applications or environments with elevated ambient temperatures, the material selection must be reconsidered to ensure the agent does not compromise structural stability prematurely. Additionally, while this study focused on planar delamination, future work should leverage the geometric freedom of 3D printing to distribute the healing agent in three dimensions. Spatial architecture is necessary to address more complex damage modes, such as those resulting from low-velocity impacts, in which damage often propagates through multiple planes and material phases. Expanding the architecture of the embedded thermal remending system will be critical to developing durable, multi-functional aerospace and automotive components.

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