

# **REPAIR PERFORMANCE OF DISCONTINUOUS FLAX FIBRE REINFORCED VITRIMERS**

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Keywords: Circular Economy, Discontinuous Reinforcement, Mechanical Testing, Repair

#### ABSTRACT

This study examines the utilization of vitrimers in flax fibre reinforced composites, highlighting their potential as sustainable solutions. Vitrimers, possessing characteristics of both thermosets and thermoplastics, offer self-healing and recycling capabilities. By incorporating vitrimers into the composites industry, the principles of a circular economy can be realized. A commercial vitrimer resin, Vitrimax T100<sup>TM</sup>, was reinforced with aligned short flax fibre preforms produced by the HiPerDiF method. The mechanical recovery performance of aligned short flax fibre reinforced vitrimers through two repair strategies, end-to-end and single patch methods, was investigated. The results demonstrate the feasibility of reusing, repairing, and recycling these materials, presenting a significant step towards achieving sustainability and circularity in fibre reinforced composite applications.

#### **1 INTRODUCTION**

The sustainability of fibre reinforced polymer composites has become important for reaching some of the global sustainable development goals. Natural fibres, particularly flax, and bioderived matrices are possible sustainable solutions for the composites industry, due to the reduction in the constituents' embedded environmental impact. According to the circular economy paradigm, sustainability can also be achieved by delaying the disposal of materials. Vitrimers form a comparatively new class of polymer family that combines the beneficial properties of both thermosets and thermoplastics, and are able delay disposal since they show self-healing/repairability features. [1]

For sustainable composites, vitrimers outweigh other potential matrices due to their remarkable potential for use in the circular economy [2]. It is also possible to derive vitrimers entirely from renewable sources [3]. It has been shown that a glass fibre reinforced vitrimer can be repaired by applying heat and pressure due to exchangeable disulfide crosslinks in the vitrimer [2]. It has also been demonstrated that vitrimer-composite specimens can be reshaped in a hot press machine and can be recycled via mechanical or chemical recycling methods. Furthermore, Tanyton et al. [4] have reported an energy-neutral closed-loop recycling process for a malleable polyimine networked vitrimer resin/carbon fibre composite in which it was possible to completely recover and reuse both components. Tanyton et al. [4] have also shown that delamination damage on carbon fibre reinforced vitrimer can be perfectly repaired through simple heat pressing.

The High Performance Discontinuous Fibre (HiPerDiF) method, invented at the University of Bristol, is a new and high throughput water-based manufacturing process to produce high-performance highly aligned discontinuous fibre composites [5]. Even though natural fibres are typically hydrophilic in nature, it has been shown that flax fibres can be processed *via* the HiPerDiF method, also in combination with reclaimed carbon fibres, to manufacture more sustainable composites with good stiffness and high vibration damping properties [6].

In this study, a vitrimer was used for flax fibre reinforced composites since it satisfies the principles of a zero-waste hierarchy. Aligned discontinuous flax fibre reinforced vitrimer (ADFFRV) specimens were produced using the HiPerDiF method and investigated. Two strategies were applied to repair ADFFRV, and the mechanical recovery performances are presented for the ADFFRV.

### 2 MATERIALS AND METHOD

#### 2.1 Materials

The flax fibres were grown and processed by Eco-Technilin- Flaxtape<sup>™</sup> (Normandy, France) using a proprietary approach and were used as received.

Vitrimax T100<sup>TM</sup>, an imine-linked vitrimer procured from Mallinda Inc., was used. To prepare the resin blend, the hardener and resin were blended in the ratio (2.5:1), then carefully mixed manually, and cured in an air circulating oven at 135°C for 60 min.

#### 2.2 Fibre alignment and composite manufacturing process

The HiPerDiF method was used for producing highly aligned discontinuous flax fibre preforms. 6 mm short flax fibres suspended in water were accelerated through a nozzle and directed in a gap between two parallel plates. Due to the sudden change of liquid momentum, the fibres were aligned, and water medium was removed by suction and drying stage in which the aligned fibre preform is dried with infrared radiation to allow the resin impregnation process. A schematic of the HiPerDiF short fibre alignment machine is shown in Figure 1.



Figure 1: The HiPerDiF fibre alignment machine [6].

To manufacture composite specimens, four layers of aligned 6-mm long flax fibre preforms (75–85 gsm) produced via the water based HiPerDiF fibre alignment method were sandwiched between five layers of Vitrimax T100<sup>TM</sup> resin film (150–250 gsm) sourced from Mallinda Inc. The composite stack was then cured by vacuum bag moulding in an autoclave at 135 °C at 1 bar pressure for 30 min followed by another 30 min at 6.5 bar pressure.

#### 2.3 Methods

The ADFFRV samples were tested in tension on an electro-mechanical testing machine at a test speed of 1 mm min<sup>-1</sup> following the ASTM D3039/D3039M-17 [7]. Strain was measured using a video

extensometer and converted to strain. A 10 kN load cell was used to record the load. The nominal sizes for specimens' widths and lengths were 5 and 150 mm, respectively; no end-tabs were used, and the specimens were gripped with hand-screwed clamps leaving a 50 mm gauge length.

A low-temperature and rapid technique was devised to assess the repair performance of ADFFRV. Broken samples were placed in a semi-closed mould that was then located between the heated plates of a servo-electric tensile test machine equipped with a 50 kN load cell. A 0.69 MPa pressure (on each sample) was applied for 5 min at 120  $^{\circ}$ C to repair the samples. The broken ends of the samples were placed in contact with one another, and two repair strategies were tested; (i) unpatched and (ii) single-patched on samples that had already been broken and repaired using method (i). Methods (i), (ii), and the repair process are represented schematically in Fig. 2.



Figure 2: Schematics of two composite repair strategies specimens; (i) end-to-end repair (eoe) and (ii) single-patched, and a schematic representation of the repair process. The patch is identical to the original specimen. [1]

## **3 RESULTS & DISCUSSION**

The mechanical properties of the ADFFRV samples were determined, both on original and repaired specimens. In total, six specimens were tested, of which five (four for single side patched ADFFRV) successfully fractured within the gauge length and were used to calculate the mechanical properties. The mechanical properties of fractured and repaired ADFFRVs are summarised in Table 1 and shown in Figure 3.

Sample	Young's modulus (GPa)	Tensile Strength (MPa)	Failure Strain (%)
Original	$12 \pm 1$	$122 \pm 14$	$1.2 \pm 0.1$
1 <sup>st</sup> eoe repair	$12 \pm 1$	$58\pm 6$	$0.5 \pm 0.1$
2 <sup>nd</sup> eoe repair	$13 \pm 1$	$47 \pm 8$	$0.4 \pm 0.1$
3 <sup>rd</sup> eoe repair	$16 \pm 1$	$44 \pm 8$	$0.3 \pm 0.1$
Patched	$13 \pm 1$	$81 \pm 15$	$0.7\pm0.2$

Table 1: Mechanical properties of as-manufactured (original) and end to end (eoe) repaired aligned flax fibre reinforced vitrimers. Errors represent the standard deviation from the mean.



Figure 3: Tensile strength of original, end-to-end repaired & tested, patched ADFRV specimens.



Figure 4: Images of original, end-to-end repaired & tested, and patched & tested ADFFRV specimens.

As seen in Table 1 and Figure 3, the results demonstrate that the eoe repair strategy was able to recover a portion of the mechanical properties, with the single-patch repair method showing better recovery. Young's modulus of the repaired specimens varied, while the tensile strength and failure strain decreased compared to the original composite. An almost doubling of tensile strength was seen for the patched repaired ADFFRV samples compared to the three times repaired materials; this in turn is nearly two-thirds of the value obtained for original samples. Figure 4 shows scanned images of repaired and tested ADFFRV specimens. It was seen that most of the eoe repaired specimens failed in the same area of the specimen and with a similar shape. However, the patched repaired specimen failed in a slightly

different area and the failed shape was found to be different than other specimens.

Figure 5 shows the scanning electron microscopy images of the ADFFRV specimens. It was observed that there are aligned fibres in the failure region of the original ADFFRV. There are however fewer aligned or distorted fibres, and in some regions no fibres at all, in the end-to-end repaired ADFFRVs. After the patch repair, the number of aligned fibres increased, which explains the increase in tensile strength, and some distorted fibres were observed. It was found that the existence of aligned fibres or protecting alignment of the fibres during the repair process affects the mechanical recovery in ADFFRV.



Figure 5: Electron microscopy images of original, end-to-end repaired & tested, and patched & tested ADFFRV specimens.

## 4 CONCLUSIONS

The findings showed that a rapid and low-temperature end-to-end repair strategy can recover half of the strength of the as-manufactured composites; however, the single patch repair method showed increased recovery (67%) compared to end-to-end repair. It was found that recovery is highly dependent on presence of the aligned fibres in the repaired area. Therefore, better strategies need to be applied to protect aligned fibres during the repair process to increase the recovery performance of the short fibre reinforced vitrimer composites. In conclusion, aligned natural fibre reinforced vitrimers can be reused, repaired, and recycled: this represents a step towards a circular economy and sustainability in fibre reinforced composites applications.

#### ACKNOWLEDGEMENTS

This work was funded under the UK Engineering and Physical Sciences Research Council (EPSRC) Project [grant EP/P027393/1] "High Performance Discontinuous Fibre Composites—a sustainable route to the next generation of composites" and the EPSRC Centre for Doctoral Training at the Advance

Composites Centre for Innovation and Science (ACCIS, Grant number EP/L016028/1). A.K. acknowledges support from Turkish Ministry of National Education YLSY grant. The authors also thank Mallinda Inc. for providing vitrimer matrix material.

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