Mechanical Properties of Bio-Based Epoxy Composites Reinforced with Hybrid-Interlayer Ramie and Recycled Carbon Fibres

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Abstract

The growing environmental concerns have led to attention on bio-based composite materials, such as the natural fibres, recycled carbon fibres and bio-based resins. Herein, the bio-based epoxy composites were reinforced with ramie fibre (RF) and recycled carbon fibre (rCF) via inter-layer hybridisation. The dynamic mechanical analysis, tensile, flexural and impact properties characterisation were conducted to analyse the mechanical behaviour of the specimens. Also, the morphology of fractured surface after mechanical tests was studied under a scanning electron microscope. When the volume ratio between RF and rCF was varied from 100/0 to 0/100, the flexural and tensile strength of composites was significantly increased, while the impact strength was reduced. Thus the maximum values of flexural strength (182 MPa) and tensile strength (165 MPa) were observed for rCF reinforced composite, whilst impact strength of 24 kJ/m² was found for RF reinforced composite. Furthermore, the values of storage and loss modulus were increased with the rCF incorporation due to a greater degree of restriction with the addition of rCF into the matrix. The hybridisation was able to combine the specific properties of RF and rCF and optimise the mechanical performance of composites. Therefore, the alternative low-cost green composites are prepared which can replace synthetic materials for semi-structural applications.

Keywords

Recycled Carbon Fibres, Ramie Fibres, Bio-Based Epoxy, Hybrid Interlayer, Mechanical Properties

1. Introduction

Composite materials are light-weight materials that are employed in transport-
ation (aviation, marine, and railway vehicles) and construction (such as bridges, wind turbine blade) industries due to their excellent mechanical performance, thermal insulation characteristics, fatigue, and corrosion resistance [1] [2]. For the composite matrix, the epoxy resin is widely used due to its good impregnation and adhesion to fibre reinforcement, resulting in excellent chemical and mechanical performance and low shrinkage after curing [3]. Also, the demand for carbon fibres (CF) has increased in the past decade, and with a compounded annual growth rate of 12%, it is expected to escalate from 58,000 tonnes in 2015 to 116,000 tonnes in 2021 [4]. However, even though the use of these materials provides several advantages, they also present some challenges to the environment. The growth of volumes for carbon fibre reinforced composites today would inevitably lead to larger volumes of composite waste in near future [5].

A few studies have demonstrated that recycled carbon fibre (rCF) maintains high strength and modulus when compared with those of fresh carbon fibre [6] [7]. Furthermore, the cost of manufacturing rCF is approximately half of that for fresh CF. Therefore, application of rCF can decrease the price of carbon fibre reinforced composites. Hence, several processes are operating commercially for the recovery of carbon fibres from the end-of-life components, such as aircraft parts and tooling, in-process scrap such as out of shelf-life prepreg, and ply cutter offcuts [8].

Moreover, natural fibre reinforced composites are receiving increasing interest due to light-weight, low-cost, and excellent recyclability, and they provide a solution to the reuse of agricultural wastes [9]. Thus, in recent years, natural fibre (hemp, flax jute, and ramie) composites have been successfully used for light-weight and low-cost applications [10] [11] [12] [13]. Ramie fibre (RF), commonly known as China grass, is one of the oldest vegetable fibres and has been grown in China for several centuries. The fibres possess high strength, good durability, high absorbency with excellent lustre, and better chemical and bacterial resistance than other natural fibres [14]. When compared with synthetic fibres (glass fibre and carbon fibre), ramie fibre is a renewable green material with lower density and production cost [9]. However, significant barriers still exist for the structural applications of natural fibre reinforced composites. These barriers include the lack of confidence in the use and performance of natural plant fibres and their composites, their incompatibility with the hydrophobic polymer matrix, limited understanding of diffusion behaviour, lack of established manufacturing processes, and poor resistance to moisture [15].

Hybridisation refers to the combination of more than one type of fibre in the same matrix, can be an appropriate approach to take the advantages of both natural and synthetic fibres, as incorporation of fibres into a polymer can cause substantial changes in the mechanical properties of composites [15] [16]. This hybrid approach offers an attractive model for the fabrication of products with reduced cost and high specific modulus, strength, and elongation behaviour and impact resistance [17] [18].
Patel et al. [19] investigated the jute/carbon hybrid epoxy composites and found that the mechanical properties were improved with the alkali treatment and acrylation of jute fibres. Noorunnisa Khanam et al. [17] studied the mechanical properties of random sisal/carbon fibre reinforced hybrid composites with different fibre weight ratios and reported that both the tensile and flexural properties of the composites were increased with the carbon fibre loading. On the other hand, Le Guen et al. [20] and Ashworth et al. [21] worked on the damping coefficient of composites reinforced with hybrid flax/carbon and jute/carbon, respectively. The hybrid composites displayed better damping performance than the carbon fibre reinforced composites and higher mechanical performance than the natural fibre reinforced composites.

On the other hand, the researchers have focused on the replacement of petrochemical components in the resin matrix of composites with bio-based renewable equivalents. The renewable resources for resin matrix can provide a sustainable platform to partially or even completely substitute petroleum-based polymer resin via the design of bio-based polymer resin, which can compete with the existing petroleum-based polymers in terms of eco-friendliness values. Several bio-based epoxy resins derived from plant oils [22] [23] [24], lignin [25] [26], and rosin [27] [28] have been reported in literature. Some studies [29] [30] have also addressed the mechanical behaviour of composites made with rCF and epoxy composites.

However, there is no report in the literature on the hybrid, recycled carbon fibre/ramie fibre reinforced bio-based epoxy composites. Herein, the hybrid approach is hybrid-interlayer with a stacking sequence where stiff recycled carbon fibre mats are positioned at the outer-layers and more elastic ramie fabric at the inner-layers. The hybrid composites with “sandwich-structure” were fabricated to explore the effect of volume ratio of RF and rCF on the mechanical properties of the hybrid composites and develop light-weight, low-cost, and green composites with excellent mechanical performance. The nonwoven mat of rCF produced by the papermaking process and compression moulding for composite manufacture were studied. Dynamic mechanical analysis (DMA) was performed for hybrid composites, and tensile, flexural, impact tests were conducted. Scanning electron microscope (SEM) was used for the fracture morphological analysis of prepared specimens.

2. Materials and Methods

2.1. Nonwoven Mat of Recycled Carbon Fibre

A wet papermaking technique was employed to convert the recycled carbon fibres (ReCF™-CP, average length: 3 mm, Nantong Fuyuan Carbon Fibre Recycling, China) into a nonwoven mat with areal density of 100 g/m² (gsm). As shown in Figure 1, the process comprised of three steps. Around 10 g of recycled carbon fibres were weighed, added to a water solution of 10 L, and stirred using a high shear radial impeller at a speed of 800 rpm for 10 min. The water
solution contained 1 g/l of PEO binding agent (Ryoji Organic Chemical, Japan) and 0.3 g/l of anti-foaming agent (KR-XP96, Nanjing Kuorun Chemical, China). The process was undertaken using a tank with two impellers to ensure an efficient dispersion of the short fibres. Then, the fibre suspension was poured into another tank where the mesh was on the bottom. The mesh was slowly moved upwards to collect the dispersed recycled carbon fibres on the mesh to form a nonwoven mat. Finally, the collected mat was dried in an oven at 60°C for 12 hours.

2.2. Composite Manufacture

The composite specimens were manufactured via compression molding technique using a bio-based epoxy resin (AGMEP, ACC (Beijing) Science and Technology, China), and the detailed process is presented in Figure 2. The bio-based epoxy resin was pre-treated by mixing with acetone in 1:1 ratio to reduce the viscosity for easy mat (rCF mat and RF fabric) impregnation. The impregnated mats were left in the fume hood for 8 hours for acetone evaporation. The areal weight of nonwoven rCF mat and plain weave ramie fabric (PR140, ACC (Beijing) Science and Technology, China) were 100 g/m² and 145 g/m², respectively.

![Figure 1](image1.png)

**Figure 1.** Schematic diagram of the recycled carbon fibre papermaking technique.

![Figure 2](image2.png)

**Figure 2.** Schematic diagram of the composite manufacturing process.
The final composites were manufactured via the layer stacking process (see Figure 2). The prepregs were stacked in a sandwich structure where the outer-layer was the rCF prepreg and inner-layer was RF prepreg. The stacked compound was placed in a 2 mm thick mold cavity between two metallic plates, pre-heated in the press at 90˚C for 15 min, and then, pressed at 10 bar for 15 min. This process was followed by curing at the same pressure and curing temperature of 130˚C for 120 min. The mould was then moved to the cooling plates of the press and held under the same pressure while cooling to room temperature.

In all composites, the overall fibre volume fraction was maintained at 30%. The hybrid composites were named based on the ratio between RF and rCF, as shown in Table 1.

### 2.3. Mechanical Properties

The tensile tests of composites were performed according to ISO 527-2 using a Universal Test Machine (E42, MTS, China) at the crosshead speed of 2 mm/min. The three-point bending test was conducted on Universal Test Machine (E42, MTS, China) as per ISO 14125 at the crosshead speed of 1 mm/min. The Izod impact test was performed using the impact machine (TE15, TQ, UK) in accordance with ISO 180 and unnotched specimens were prepared for testing. A minimum of five specimens were prepared for the tensile and flexural test, ten specimens for Izod impact test.

### 2.4. Dynamic Mechanical Analysis

Dynamic mechanical properties of the composites were assessed using a Dynamic Mechanical Analyser (DMA 8000, Perkin Elmer, USA) following ASTM D7028 standard. All the tests (specimen dimensions: 30 × 10 × 2 mm) were performed in dual cantilever mode at 1 Hz frequency from 50˚C to 250˚C at a heating rate of 2˚C/min.

### 2.5. Microscope Observations

The composite samples after flexural test were cooled in liquid nitrogen for 1 min and fractured manually. Care was taken to ensure that the fracture was created outside the zone of damage from flexural testing. Also, putter coating with gold was performed on the samples after tensile tests and the cross-section of the

| Sample code | Layer number ratio (RF/rCF) | Volume fraction ratio (RF/rCF) | Stacking sequence |
|-------------|-----------------------------|---------------------------------|-------------------|
| 8RF         | 8/0                         | 100/0                           | RF₈               |
| 6RF2rCF     | 6/2                         | 75/25                           | rCF₁/RFₑ/rCF₂     |
| 4RF2rCF     | 4/4                         | 50/50                           | rCF₁/RFₑ/rCF₂     |
| 2RF6rCF     | 2/6                         | 25/75                           | rCF₁/RFₑ/rCF₂     |
| 8rCF        | 0/8                         | 0/100                           | rCF₈               |
composite samples was examined under scanning electron microscope (SEM, Sigma VP; Zeiss, Oberkochen, Germany) operated at 10 kV.

2.6. Statistical Analysis

The average values and standard error of all the data presented were calculated and analysed using the Prism software (version 6.0, GraphPad Software, San Diego, CA, USA). A one-way analysis of variance (ANOVA) was calculated with Tukey multiple post-test to compare the significance of the change in one factor with time. The error bars on all the data represent standard error of the mean.

3. Results and Discussion

3.1. Dynamic Mechanical Analysis

As shown in Figure 3, the storage modulus of composites decreased with temperature, being higher for the composite with more recycled carbon fibres (rCF). Even a small content of rCF displayed a pronounced effect on the storage modulus of the hybrid interlayer composite in the glassy and elastomeric regions. The modulus of composites in the glassy state is primarily determined via the strength of intermolecular forces and the packing of polymer chains [31]. An improvement in the modulus can be a result of the incorporation of reinforcement which led to composite stiffness. Besides, an increase in modulus by rCF incorporation should be because of both higher modulus of rCF than ramie fibre and stronger adhesion to the polymer chains at the interface [32].

On the other hand, with an increase in temperature, there was a decrease in the storage modulus (Figure 3), displaying a sharp fall when passing through the glass transition region. This drop was affected by the reinforcing effect of the rCF/ramie fibre in the epoxy matrix. The decrease of modulus with an increase in temperature is based on the Micro-Brownian motion of the polymer chains as the polymer approached the glass transition temperature, $T_g$. The Micro-Brownian movement is related to the cooperative short-range diffusional motion of the main chain segments and to their relaxation stress [32] [33].

![Figure 3](image_url)

**Figure 3.** Storage modulus of hybrid inter-layer bio-based epoxy composite.
decrease in modulus of composites with increase in temperature can also be attributed to the effect of fibre stiffness and fibre matrix adhesion.

**Figure 4** presents the loss modulus as a function of temperature for different relative ratio (RF:rCF). In DMA analysis, the loss modulus is the contribution of the viscous component and is indicative of the energy dissipated by the system [34]. The loss modulus reached the peak values and decreased with the increase in temperature. This behaviour is caused by the free movement of polymer chains at higher temperatures. The rapid rise in loss modulus indicates an increase in the polymer structural mobility due to a relaxation process that allows much more motion along the chain than that is possible when the temperature is below $T_g$ [35].

When rCF was incorporated in the composites, higher loss modulus peak values were obtained. These results can possibly be attributed to the inhibition of the relaxation process in the composites with the increased amount of chain segments and free volume owing to fibre addition [36]. Moreover, an increase in the modulus peak values with rCF addition may be indicative of a structure with higher internal friction that improves the energy dissipation, implying changes in molecular dynamics in this region [36] [37].

The peak value of tan delta is associated with the partial motion of a polymer structure; thus, indicative of the glass transition temperature $T_g$ where a material changes from the rigid to elastic state [38]. The tan delta as a function of temperature for different relative volume ratio (RF:rCF) is shown in **Figure 5** and the glass transition temperature, $T_g$, corresponding to peak value of tan delta are listed in **Table 2**. $T_g$ was found to increase when the recycled carbon fibre replaced the ramie fibre. This temperature shift with the increase in the recycled carbon fibre content is related to the higher energy required to reach maximum dissipation [39].

**3.2. Flexural Properties**

The properties of composites and corresponding failure mode hybrid composites are primarily dependent on the fibre (recycled carbon and ramie fibre) content and layer stacking sequence. According to the literature [34], due to the

![Figure 4. Loss modulus of hybrid interlayer bio-based epoxy composite.](image-url)
difference in surface morphology and functional groups between carbon fibre and ramie fibre, the fibre-matrix adhesion at the carbon fibre-epoxy and ramie fibre-epoxy interface was different. On the other hand, the tensile modulus could be improved by 23% and strength by 10% depending on the stacking sequence, where the carbon fibres at the outer-layer led to higher tensile properties [40]. Therefore, the hybrid interlayer composites were designed as a sandwich structure with recycled carbon fibre in the outer-layer (see Figure 2).

The load-displacement curves obtained from the flexural testing are displayed in Figure 6, and the flexural strength and modulus of composites are presented in Figure 7. Based on the comparison of load-displacement behaviour of composites, composites with pure rCF reinforced exhibited linear curves with a sudden drip of the load after the peak load. However, with the hybridisation of RF with layered structure, the load-displacement curve changed from linear to more prolonged nonlinear. With the substitution of RF by rCF, the peak load for composite failure was increased, and the rCF reinforced composite (8rCF) displayed the highest peak value of 200 N. Meanwhile, the elongation of composites decreased as the addition of rCF could change the composites’ specific properties from “ductile” to “brittle”.

As shown in Figure 7, the rCF reinforced composite (8rCF) displayed the highest flexural strength of 182 MPa and modulus of 19 GPa. In contrast, the flexural strength and modulus for 8RF composite without rCF were 75 MPa and 6 GPa, respectively. When RF was replaced with rCF in hybridisation, an
Figure 6. Load displacement curves of flexural testing for hybrid interlayer bio-based epoxy composite.

Figure 7. Flexural properties of hybrid interlayer bio-based epoxy composite.

increase in flexural strength was observed from 107 MPa (6RF2rCF composite) to 131 MPa (4RF/4rCF composite) and 164 MPa (2RF6rCF composite). Also, the flexural modulus was increased from 7 GPa (6RF2rCF composite) to 11 GPa (4RF/4rCF composite) and 16 GPa (2RF6rCF composite).

Herein, the hybrid composites were designed as a sandwich structure where rCF is in outer-layer and RF in the inner-layer. The flexural strength and modulus were significantly affected by the properties of reinforcement closest to the outer-layer surface of the specimens, of which the top surface experienced the compression force, and the bottom surface tension force [15]. Hence, the total bending deflection behaviour is a combination of the compression and tension deflection. The rCF with better mechanical properties in the outer-layer surface of the composite could provide higher performance in tension and compression during the bending.
3.3. Tensile Properties

The tensile force-extension curves obtained for the composite reinforced by rCF, RF, and hybrid reinforcement are shown in Figure 8, while Figure 9 presents the tensile properties. Similar to the flexural properties behaviour, the tensile strength and modulus increased upon the replacement of ramie fibres with recycled carbon fibres. The hybrid composite with the highest tensile strength (165 MPa) and modulus (17 GPa) was reinforced purely with rCF (8rCF composite). The 8RF composite with fully RF reinforced exhibited the lowest tensile strength of 44 GPa and modulus of 5 GPa. This difference is due to the stronger and stiffer characteristics of rCF in comparison with the RF, even though the reinforcement as nonwoven mat for rCF and plain weave for RF.

Figure 10 shows an SEM image of the fractured surface in the tensile test for different hybridisation structures. The failure mechanism for 8rCF composites fully reinforced with rCF was matrix cracking, fibre fracture, and pull-out. For the 8RF composite with RF reinforcement, there was an indication of matrix cracking and fibre pull-out with fibre extension. However, for carbon fibre hybridised composites, there was a considerable amount of matrix cracking and
pull-out of RF and rCF. In general, the fibre pull-out occurs when the interfacial stress at the fibre matrix interface exceeds the interfacial strength, causing the fibre to debond from the matrix. Therefore, fibre treatment or resin modification should be considered to improve the interface for rCF-matrix and RF-matrix.

3.4. Impact Properties

Figure 11 shows the impact strength results of the composites. When rCF was replaced by RF in the composites, the impact strength was significantly increased and the maximum value of 24 kJ/m² was observed for 8RF composite. As expected, RF reinforced composite showed a better energy absorption capability when compared with other hybrid composites and rCF reinforced composite.

Several studies in literature have reported the improvement on the impact resistance of carbon fibre reinforced composite via hybridisation with natural fibre. Ramana and Ramprasad [41] reported the 46% improvement of impact strength registered for hybrid composites when jute fibres replaced 50% carbon fibres. Sarasini et al. [42] also confirmed the impact absorption energy of hybrid composite could be improved with the addition of natural fibre laminate. They also compared different stacking sequences of hybrid composite, found that
hybrid flax-carbon-flax stacking structure presented better impact absorption performance as flax fibre in the outer-layer could dissipate higher amount of impact energy. As such, the mechanical performance of hybrid interlayer composites with RF-rCF-RF structure where ramie fibre in the outer-layer could be considered in future study, to figure out the effect of stacking sequence on the hybrid composite performance.

As a result, the hybridisation allows the modification of material characteristics and combination of the good properties of rCF (i.e. high stiffness) and RF (i.e. good toughness and elongation behaviour). The volume ratio and fibre distribution of hybrid composites can be designed to follow the requirements of the application. As such, the hybrid composites in this study presented better flexural and tensile performance than 8RF composite and higher impact strength than 8rCF composite. Additionally, such hybrid composites would be entirely “green”. Except for RF and bio-based epoxy matrix, the rCF can be considered as a low-cost renewable material because of direct recycling from the carbon fibre reinforced composites instead of manufacturing from raw material.

On the other hand, the nonwoven mat technique for rCF has the freedom for fibre orientation. The random mats of rCF herein displayed isotropic behaviour, whilst a strong fibre orientation such as UD mat can be obtained to enhance the composite performance in specific direction. Hence, they may obtain multifunctional characteristics to compete with completely synthetic composites. The hybrid RF/rCF composites can be considered as a possible alternative to the current traditional composites, such as glass fibre reinforced composites, for semi-structural applications. However, the compressive and fatigue resistance need to be investigated in future studies for further understanding of the hybridisation of natural fibres and recycled carbon fibres.

4. Conclusions

The growing environmental concerns have led to increased attention towards
the use of bio-based composite materials, such as the natural fibres and bio-based resins. Also, an increase in the waste carbon fibre reinforced composites (CFRP) has resulted in issues about the utilisation of recycled carbon fibre (rCF). As ramie fibre (RF) reinforced composites have been limited to non-load bearing applications due to low mechanical properties, a possible solution can be the hybridisation to combine natural fibre and recycled carbon fibres.

In this study, the fully green hybrid RF/rCF reinforced bio-based epoxy composites were developed. When the hybrid ratio of rCF was increased from 0% to 100%, the flexural and tensile strength of the composites was increased by 141% and 275%, respectively; while a decrease by 50% was observed for impact strength due to RF with better toughness was replaced by rCF. Furthermore, the storage and loss modulus were found to increase with the rCF incorporation. These results can be attributed to the high stiffness of rCF, which resulted in the inhibition of the relaxation process in composites with the increased amount of chain segments and free volume.

The current work elucidates on the preliminary insights into novel green composites via hybridisation, to combine the advantage of rCF with high stiffness and RF with good toughness. The as-prepared hybrid composites with relatively improved mechanical performance can be considered as an alternative low-cost green material to replace the traditional composites for semi-structural applications. For future study, the interface performance study on composite could be taken into account.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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