PRODUCTION OF CONTINUOUS INTERMINGLED CF/GF HYBRID COMPOSITE VIA FIBRE TOW SPREADING TECHNOLOGY

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Abstract
An air-assisted fibre tow spreader was used to produce continuous commingled glass and carbon fibre tows. Hybrid carbon and glass fibre (CF/GF) reinforced epoxy composites were manufactured from these commingled fibre tows by resin film infusion. The degree of hybridisation of the hybrid CF/GF tow was defined and characterised. Compared with the corresponding continuous CF/epoxy composite, the hybrid composite with a degree of hybridisation of 32.45% exhibits a more gradual tensile failure. The final tensile failure strain increased by 14% as compared to that of the carbon fibre epoxy composite.

1. Introduction

Conventional unidirectional carbon fibre reinforced polymers (CFRPs) have high specific strength and stiffness and a long fatigue life [1]. However, a major disadvantage of these materials is that they fail in a brittle manner with little warning, for example, when subjected to uniaxial tension loading. Due to their brittle failure behaviour, such materials cannot be used in unpredictable load conditions without significant safety factors to ensure the risk of catastrophic failure is acceptably low.

In order to create high-performance unidirectional composites exhibiting a more graceful failure behaviour, a novel method was used to manufacture a continuous intermingled CF/GF hybrid tow. Unlike conventional hybrid composites, which are hybridized at the ply level [2-6], intermingled composites are hybridized at a filament level. Ideally, the nearest neighbouring fibres of one fibre type should be of the other fibre type. When such intermingled unidirectional hybrid composites are subjected to longitudinal tension, the initial failure is most likely to occur in the fibre type with the smaller failure strain while the nearest neighbouring fibres of the other fibre type with a larger strain to failure should still carry load and stop the failure propagating. Such a hybrid composite, therefore, could exhibit a non-linear tensile stress-strain curve with a stepped or gradual tensile failure behaviour. Previously, some investigators have tried to hybridise two fibre types within one layer to create intralayer hybrid composites via simultaneous filament winding [3, 7], co-weaving [8,
and air-texturing commingling processes [10, 11], but we could not find any attempt in the literature to provide a definition of the degree of hybridisation at the filament level. And there were few studies in the literature which established a relationship between the degree of hybridisation at the filament level and corresponding tensile behaviour.

In this research, an air-assisted fibre tow spreading technology was used to increase the spacing between the fibre filaments within carbon fibre and glass fibre tows separately. Then, these two spread fibre tows were intermingled by vacuum airflow to produce a hybrid fibre tow. In order to evaluate the degree of hybridisation at the filament level, we developed a model to describe randomly-distributed, commingled two-fibre-type tows to determine the maximum possible degree of hybridisation. In order to characterise the actual commingled hybridisation, an image recognition programme was developed to analyse cross-sectional micrographs of the hybrid tow. By comparing modelling results with experimental results, we were able to quantify the degree of hybridisation of an intermingled two fibre tow. CF/GF/epoxy hybrid composites were manufactured from the hybrid fibre tow and resin film via resin film infusion. Finally, their tensile behaviour was characterised.

2. Experimental

2.1. Materials

The continuous carbon fibre tow used (TORAYCA®, T700SC-12K) was kindly provided by Toray International UK Ltd. and the continuous E-glass fibre tow (5744-735 Tex) by AGY World Headquarters. An epoxy resin film (HexPly®M21, 37g/m2) from Hexcel Co. was used as matrix. The diameters of CF and GF are 6.90 ± 0.15 μm and 14.92 ± 0.53 μm (measured by modified -Wilhelmy method), respectively.

2.2. Manufacturing route

2.2.1. Manufacturing continuous hybrid CF/GF fibre tow

An air-assisted fibre tow spreading and commingling technology was used to manufacture the continuous hybrid CF/GF tow. The principle of this technology is to let air pass through the fibre tow in a low-tension or tension-free state [12, 13]. Under these conditions the space between the filaments is increased. The overall width of the fibre tow is thereby increased. In order to commingle the glass and carbon fibres, the spread fibre tows are then placed on top of one another, and an air flow is again passed through this combined fibre tow to further spread and commingle the two fibre tows. A fibre tow spreader (Izumi International Inc, US) was used to spread carbon and glass fibre tows individually. Then the spread glass fibre tow was placed on top of the spread carbon fibre tow. The combined GF/CF tow was then passed again through the air-assisted commingling unit, which causes the GF filaments to insert into the space between CF filaments. The volume ratio between the CF and GF in the hybrid tow was 1.55:1. Control CF and GF tows were also manufactured using the same conditions without adding the other type of fibre tow.

2.2.2. Manufacturing hybrid CF/GF reinforced epoxy composites

The CF/GF reinforced epoxy composites were manufactured by resin film infusion. The dry CF/GF hybrid tows with a length of 20 cm and width of 2 cm were carefully laid up on the epoxy films under tension to maintain the fibre alignment. The stacking sequence of the lay-
up process was $E/(H/E)_5$, where $E$ is epoxy film and $H$ is the dry CF/GF hybrid tow. The prepreg was placed into a 20 x 2 cm mould and then heated up from room temperature to 180 °C at a rate of 3 °C/min under a pressure of 0.8 MPa. The sample was kept at 180 °C and 0.8 MPa for 2.5h and then cooled down to room temperature. Then the hybrid CF/GF reinforced epoxy composite panel with dimension of 20 x 2 x 0.5 cm was removed from the mould. The control CF/epoxy and GF/epoxy composites were manufactured using the same method as stated above. The fibre volume fractions in the control CF/epoxy, GF/epoxy and hybrid composite are all 30% - 40%.

2.3. Microscopy analysis of CF/GF hybrid tows

In order to evaluate the distribution of GF and CF within the hybrid fibre tow, the dry hybrid CF/GF tow was carefully fixed using adhesive tape between two microscope slides to minimise accidental changes to the fibre arrangement within the tow. Then it was embedded in a transparent epoxy (EpoxyCure, Buehler Ltd.) and cured at room temperature overnight. The samples were polished and observed using a reflective microscope (AX10, Zeiss Ltd., UK) at a magnification of 20x. A typical microscopy image is shown in Figure 1. The micrographs were analysed using the an image recognition program which we developed to evaluate the fibre distribution within CF/GF hybrid tows.

![Figure 1. Typical micrograph of the hybrid CF/GF tow](image)

2.4. Characterization of the tensile properties of hybrid CF/GF reinforced epoxy composites

The CF/GF hybrid reinforced epoxy, CF/epoxy and GF/epoxy were end tabbed with woven GF reinforced polyester with the thickness of 1.5 mm and then cut to 100 x 5 mm, shown in Figure 2. Five specimens of each group were tested under uniaxial tensile load (Instron 5969, Instron Ltd, Bucks, UK) at a crosshead rate of 0.5 mm/min. A video extensometer (Imetrum, Imetrum Ltd., Bristol, UK) was used to record the strain over the whole gauge length.

![Figure 2. Dimensions of the test specimen used for the tensile testing](image)
3. Results and discussion

3.1. Spreading and commingling CF and GF tows

<table>
<thead>
<tr>
<th></th>
<th>As-received (mm)</th>
<th>Spread (mm)</th>
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</thead>
<tbody>
<tr>
<td>CF tow</td>
<td>5-7</td>
<td>18-22</td>
</tr>
<tr>
<td>GF tow</td>
<td>3</td>
<td>8-12</td>
</tr>
</tbody>
</table>

Table 1. The widths of as-received and spread CF tow and GF tow

After spreading CF tow and GF tow individually, the widths of the fibre tows were increased compared with those of as-received fibre tows, shown in Table 1. As seen in Figure 3, the fibre alignments within spread CF tow and GF tow were good. Figure 4 shows the appearance of the combined CF/GF tow before and during the commingling process.

Figure 3. Photos of spread (a) CF and (b) GF tows

Figure 4. Typical photos of combined CF/GF tow (a) before entering (b) during commingling unit

3.2. Characterisation of the degree of hybridisation of CF/GF tows

3.2.1. Definition of the degree of hybridisation: A model to describe randomly-distributed, commingled two-fibre-type tows

The ideal intermingled hybrid CF/GF tow should be one in which both carbon and glass fibres are randomly distributed without any organised fibre arrangement. A computer model was built to describe the ideally intermingled hybrid CF/GF tow. In this model, elements A and B were randomly distributed in an m-by-m matrix. The dimension of the matrix, m, was calculated using Equation (1),
\[ m = \text{int} \left( \sqrt{N_{GF} + N_{CF}} \right) \] (1)

where \( N_{CF} \) and \( N_{GF} \) are the numbers of filaments of CF and GF in the hybrid tow.

The number ratio between A and B in the model is the same as the one in the hybrid tow to be produced. This matrix was subdivided into a certain number of square analysis windows, shown in Figure 5. Then the proportion of the cross sectional area of CF in the total cross sectional area of GF and CF in each analysis window in the model, \( AR_{CF,Model} \), was calculated using Equation (2):

\[ AR_{CF,Model} = \frac{N_A D_F^2}{N_A D_F^2 + N_B D_F^2} \] (2)

where \( N_A \) and \( N_B \) are the numbers of elements A and B in each analysis window, respectively. D is the fibre diameter and the subscripts CF and GF represent carbon fibre and glass fibre, respectively.

### 3.2.2. Microscopy analysis of hybrid CF/GF tows using our image recognition program

A MATLAB program was coded to evaluate the distribution of CF and GF in the micrographs of hybrid CF/GF tows. The CFs and GFs were distinguished based on their image colour intensity, as seen in Figure 6. The reliability of the image recognition program, R, is characterised via Equation (3),

\[ R = \frac{A'_{CF} \times (A'_{CF} + A'_{GF})}{V_{CF} \times (V_{CF} + V_{GF})} \times 100\% \] (3)

where \( A' \) is the total cross sectional area of each fibre type which is determined by this image recognition program and \( V \) is the volume of each fibre type used in the hybridisation process. It was found that R is 95.37%, which means that this image recognition program is satisfactory.

Then this image was also subdivided into a certain number of square analysis windows. The CF and GF areas in each analysis window were also determined by this image recognition program. The proportion of cross sectional area of CF in the total cross sectional area of GF...
and CF in each analysis window in the micrographs, $AR_{CF,Experiment}$, was calculated using Equation (4),

$$AR_{CF,Experiment} = \frac{A_{CF}}{A_{CF} + A_{GF}}$$

where $A$ is the cross sectional area of CF or GF in each analysis window and the subscripts CF and GF represent carbon fibre and glass fibre, respectively. In order to achieve a representative result, six images (560 x 200 μm) taken from different locations in the hybrid tow were analysed using this program.

3.2.3. Definition and quantification of the degree of hybridisation

Because we could not find any attempt in the literature to provide a definition of the degree of hybridisation at the filament level. We define the degree of hybridisation by comparing the differences between the probability distribution of $AR_{CF,Model}$ and $AR_{CF,Experiment}$.

![Figure 7](image_url)

**Figure 7.** The probability distributions of $AR_{CF,Model}$ and $AR_{CF,Experiment}$ in the analysis windows with different lengths (a) $3D_{nominal}$, (b) $6D_{nominal}$ and (c) $14D_{nominal}$ ($D_{nominal} = \sqrt{(N_{CF}D_{CF}^2 + N_{GF}D_{GF}^2)/N_{CF} + N_{GF}}$, where $D$ is fibre diameter)

When the distribution of CFs and GFs in the hybrid tow is the same as the distribution of elements A and B in the model, the maximum intermingled hybridization of CF/GF hybrid tow was achieved. The distributions of $AR_{CF,Experiment}$ and $AR_{CF,Model}$ in the analysis windows with different sizes are shown in the Figure 7. With increasing size of the analysis windows, the distribution of $AR_{CF,Experiment}$ became closer to the distribution of $AR_{CF,Model}$. However, it is clear that CFs and GFs were not completely randomly distributed in the hybrid tow.

![Figure 8](image_url)

**Figure 8.** Coefficients of variation in $AR_{CF,Model}$ and $AR_{CF,Experiment}$ as the function of length of analysis window

In order to quantify the degree of hybridisation, the coefficient of variation (CV) in $AR_{CF,Experiment}$ and $AR_{CF,Model}$ was plotted as a function of the length of analysis window in Figure 8. The CV$_{model}$ decreased sharply from 0.45 to 0.2 with increasing length of the
analysis window when it is less than $5D_{nominal}$. Hence, when the length of the analysis window is too small (less than $5D_{nominal}$), the distribution of $AR_{CF}$ even in the model has a large amount of scatter. On the other hand, a larger analysis window means that analysis window contains many more filaments, which tends to equalise the distribution of CFs and GFs within one analysis window. Therefore, we selected an analysis window with the length of $6D_{nominal}$ as the optimal analysis window. The degree of hybridisation $H$ was calculated using Equation (5).

$$H = \frac{CV_{Model}}{CV_{Experiment}} \times 100\% = 32.45\%$$

where $CV_{Model}$ and $CV_{Experiment}$ is the coefficient of variation in $AR_{CF,Model}$ and $AR_{CF,Experiment}$ when the length of analysis window is $6D_{nominal}$, respectively.

3.3. Tensile behaviour of CF/GF hybrid composite

Figure 9 shows the tensile stress-strain curves of the control CF/epoxy and GF/epoxy composites, and hybrid CF/GF/epoxy composites. It is interesting to note that hybrid CF/GF/epoxy and control GF/epoxy composites failed more gradually than the control CF/epoxy. There were multiple drops in the tensile stress-strain curves of the hybrid composites, which means that epoxy resins reinforced using intimately commingled carbon and glass fibres do affect the tensile failure mode. As shown in Table 2, the final failure strain of the hybrid composite was 14% higher than that of the control CF/epoxy. This improvement in final failure strain and the difference in failure mode between hybrid composites and control CF/epoxy composites could be associated with the fact that GF and CF were partially intermingled in the hybrid composites with a degree of hybridisation of 32.45%. When some of the CFs in the hybrid composites failed, the nearby GFs and remaining CFs changed the load distribution and delayed crack propagation, which resulted in the stepped and more gradual failure of the hybrid composites.

![Figure 9](image-url)

*Figure 9. The tensile stress-strain curves of (a) control CF/epoxy, (b) CF/GF/epoxy and (c) control GF/epoxy*

<table>
<thead>
<tr>
<th></th>
<th>Max stress (MPa)</th>
<th>Modulus (GPa)</th>
<th>Initial failure strain (%)</th>
<th>Final failure strain (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CF/Epoxy</td>
<td>1055 ± 167</td>
<td>73.0 ± 8.8</td>
<td>1.34 ± 0.06</td>
<td>1.34 ± 0.06</td>
</tr>
<tr>
<td>CF/GF/Epoxy</td>
<td>719 ± 103</td>
<td>51.7 ± 5.0</td>
<td>1.33 ± 0.08</td>
<td>1.52 ± 0.05</td>
</tr>
<tr>
<td>GF/Epoxy</td>
<td>458 ± 14</td>
<td>22.9 ± 0.9</td>
<td>2.04 ± 0.11</td>
<td>2.34 ± 0.14</td>
</tr>
</tbody>
</table>

*Table 2. The tensile strength, modulus, the strain to initial and final failure of referenced CF/epoxy, CF/GF/epoxy and referenced GF/epoxy (Note: $V_i=$30%-40%, $V_{CF}:V_{GF} = 1.55 :1$)*

4. Conclusions

A continuous unidirectional hybrid CF/GF tow containing 12K carbon and 1.63K glass fibres was successfully manufactured via air-assisted fibre tow spreading and commingling. The
degree of hybridisation in the CF/GF tow was defined and characterized by comparing the fibre-to-fibre distribution obtained from a model of a composite containing randomly distributed two-fibre-types and the fibre-to-fibre distribution determined experimentally from micrographs of the hybrid fibre tow using an image recognition program. The degree of hybridization was defined as 32.45%, which means that CFs and GFs are partially hybridised at the filament level within the hybrid tow. Hybrid composites were manufactured from this hybrid tow using resin film infusion. Compared with the control CF/epoxy, the hybrid composite exhibited a more gradual tensile failure and 14% improvement in its final failure strain.

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