SELF REINFORCED POLYAMIDE 12/CARBON FIBRE HYBRID COMPOSITES

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Abstract

This paper describes the manufacture and properties of self reinforced polymer hybrid composites based on a combination of carbon fibres and oriented polyamide 12 fibres and tapes. Three different hybridisation techniques (intra-yarn, intra-layer and inter-layer) were investigated. In intra-yarn hybridisation, tows were comprised of a combination of carbon fibres and oriented PA12 multifilaments. In intra-layer hybridisation, carbon fibre/PA12 prepreg tapes were co-woven with oriented PA12 tapes. Finally, in interlayer hybridisation, carbon fibre spread tow cloth was impregnated with PA12 matrix polymer and then laminated with layers of a self reinforced PA12 sheet. In all cases, the hybrid composites were manufactured using the Leeds hot compaction technique. Tension and bending results suggested that the intra-layer hybrids had the best combination of properties, although all three hybridisation routes produced interesting materials.

1. Introduction

There is growing interest in hybrid carbon fibre composites with the aim of increasing the ductility of these materials (~2%) by combining with other fibres. Pegoretti et al. [1] described the extensive fibre combinations that had been reported in the literature at that time (2004), including carbon/glass, carbon/Kevlar, carbon/ultra-high-modulus-polyethylene (UHMPE), aramid/UHMPE and UHMPE/glass. In all of these ‘hybrid’ composites the main purpose is to add a proportion of ductile fibres to a carbon fibre composite to increase the failure strain and energy absorbing capabilities of the usually brittle material. In particular, Pegoretti et al. found a significant increase in the ductility index, which is a ratio of the propagation energy to the initiation energy during impact, for hybrid intraply composites of glass and PVA fibres compared to using only glass fibres.

The work presented in this paper (carried out under the European FP7 funded HIVOCOMP project) differs from previous studies into hybrid composites in three crucial aspects. First, although the hybrid composite materials reported here have the same three components as those described in the literature, that is a brittle fibre, a ductile fibre and a matrix, the method of generating the matrix phase is completely different to most of the previously published work. The hybrid composites in this work are produced from the ductile fibres using a process developed at the University of Leeds termed hot compaction. These materials are often termed single polymer, or self reinforced polymer composites (SRPC) and are known for a combination of lightweight and very high toughness. Second, the goal of the research is different to the majority of the previously published studies where the aim is to increase the energy absorbing capacity of a carbon fibre reinforced polymer matrix composite by adding in a fraction of ductile fibres. In the current work the goal was opposite to this, that is to add a small fraction of carbon fibres to an already ductile composite material to increase stiffness without, if possible, compromising ductility. Finally, the fibres we are using have a significantly higher failure strain to the normal 'ductile' fibres often employed.

Self reinforced, or single, polymer composites are a new developing material, where both the reinforcing fibre and the matrix are the same polymer. These materials (mostly based around highly drawn polypropylene tapes - Curv®) are finding increased commercial usage in areas where lightweight and outstanding toughness are key drivers (for instance Samsonite luggage and Bauer ice hockey boots). However, semi-structural applications, such as automotive, remain out of reach due to the moderate stiffness and strength of these materials. While there are a number of published methods for producing self reinforced polymer composites (for example film stacking, powder impregnation and bi-component tapes), the one used for this study was developed and patented at the University of Leeds [2-4] and is termed hot compaction. The production of hybridised self reinforced polymer composites is very much a new research area. Of the few studies carried out, Taketa, Fabich and co-workers [5, 6], have looked at combining discrete layers of a carbon fibre (prepreg) composite and a self reinforced polymer (SRP) sheet, termed inter-layer hybridisaton. Taketa et al. found that the failure strain of the hybrid laminate was improved over the pure carbon fibre laminate and this was associated with a compressive pre-strain associated with the shrinkage of the SRP fraction during consolidation and cooling, while Fabich et al. found some beneficial effects on the impact performance.

In this paper we will present some latest results in the HIVOCOMP project, concentrating on self reinforced polyamide hybrids (using Nylon 12). Three different strategies have been trialled for combining these fibres, which offer hybridisation at different length scales. In the simplest, termed inter-layer, layers of self reinforced polyamide sheet (produced using the Leeds hot compaction process) are combined with layers of a polyamide/carbon fibre composite. In the second, termed intra-layer, highly stretched polyamide tapes are co-woven with carbon fibre/isotropic polyamide tapes, to produce a cloth which is then processed using the hot compaction technique. In the third, termed intra-yarn, tows were sourced which contained comingled carbon and polyamide fibres. These were again processed using the hot compaction technique to produce a hybrid composite

2. Experimental details

2.1 Intra-yarn hybridisation

The co-mingled yarns (carbon fibres and oriented PA12 filaments) were produced by Schappe Techniques, France using their patented stretch breaking technology. These were supplied in a carbon fibre fraction of 13%.

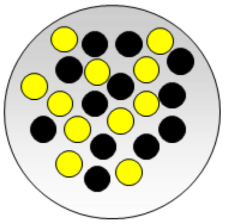

Figure 1: Intra-yarn hybridisation schematic

Microscopy of the combined tows showed that the mingling of the two components was excellent. The carbon fibres used had a modulus of 240GPa and a strength of 4GPa. In order to produce composite sheets with balanced properties, the co-mingled yarns were woven into cloth using a twill 4 x 4 weave style.

Composite samples were produced using the Leeds hot compaction process. The aim of this procedure is to establish a critical temperature at which a fraction of the oriented PA12 fibres are selectively melted, which then forms the matrix of the resulting self reinforced composite. A typical process was as follows. The oriented assembly was placed between aluminium sheets (thickness 0.1mm), two layers of silicon rubber to even out the pressure distribution, and then outer sheets of 2mm thick brass sheets. A thermocouple was placed in the centre of the assembly, and this was used for monitoring the temperature throughout the process. The assembly was then placed into a compression press set at the required temperature. A pressure of 5MPa was immediately applied and temperature monitoring was started. Once the temperature reached the required temperature, it was left for 2 minutes and then rapidly cooled to 50°C (using circulated water cooling) at which point the sample was removed from the press.

2.2 Intra-layer hybridisation

For intra-layer hybridisation carbon fibre/PA12 tapes (~40% carbon fibre fraction manufactured by Jonam Composites) were co-woven with oriented PA12 tapes manufactured at Leeds using a drawing frame (draw ratio 4:1). The PA12 polymer was the identical grade used for the multifilaments in the intra-yarn hybridisation strategy described above. The co-weaving was accomplished on a hand loom, using a warp comprised of only the PA12 tapes and a weft which could be alternated between the carbon prepreg tapes and the oriented PA12 tapes as shown in Figure 3 (the arrow shows the warp direction). Layers of the woven cloth were laid in a 0/90 configuration and processed in using the same procedure as described above

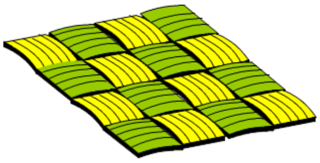

Figure 2: Intra-layer hybridisation schematic

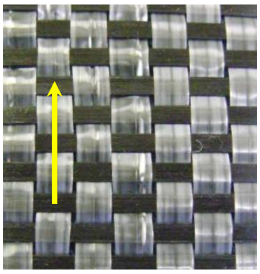

Figure 3: Intra-layer woven cloth

2.3 Interlayer Hybridisation

For the final hybridisation strategy, layers of pure SRPA12 were combined with layers of a carbon fibre/PA12 composite. The composite layers were manufactured using the TeXtreme® spread tow carbon fibre fabric (http://www.oxeon.se/). A PA12 film (once again with the same grade of PA12 as above) was manufactured and this was then used to impregnate the TeXtreme® fabric. PA12 tape was drawn to a 4:1 ratio and woven to produce a pure PA12 woven cloth. Hybrid samples were made by interleaving the impregnated carbon fibre fabric and the woven PA12 tapes using the same procedure as described in section 2.1. Samples were made with the TeXtreme® layers on either the surface or in the interior.

Figure 4: Inter-layer hybridisation schematic

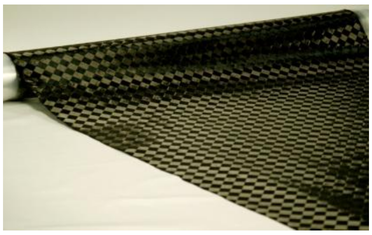

Figure 5: Tow spread carbon fibre fabric

3. Results

3.1 Intra-yarn hybridisation

The relative mechanical properties of the three hybrids were assessed by carrying out tensile and bending tests (ASTM D3039 and ASTM D6272 respectively). The volume fraction of the samples was determined using the burn off method (ASTM D2184).

Figure 6 shows typical results for tensile and bending tests on the intra-yarn hybrid. This had a measured carbon fibre fraction of 13 ± 1% and was manufactured at a hot compaction temperature of 176°C.

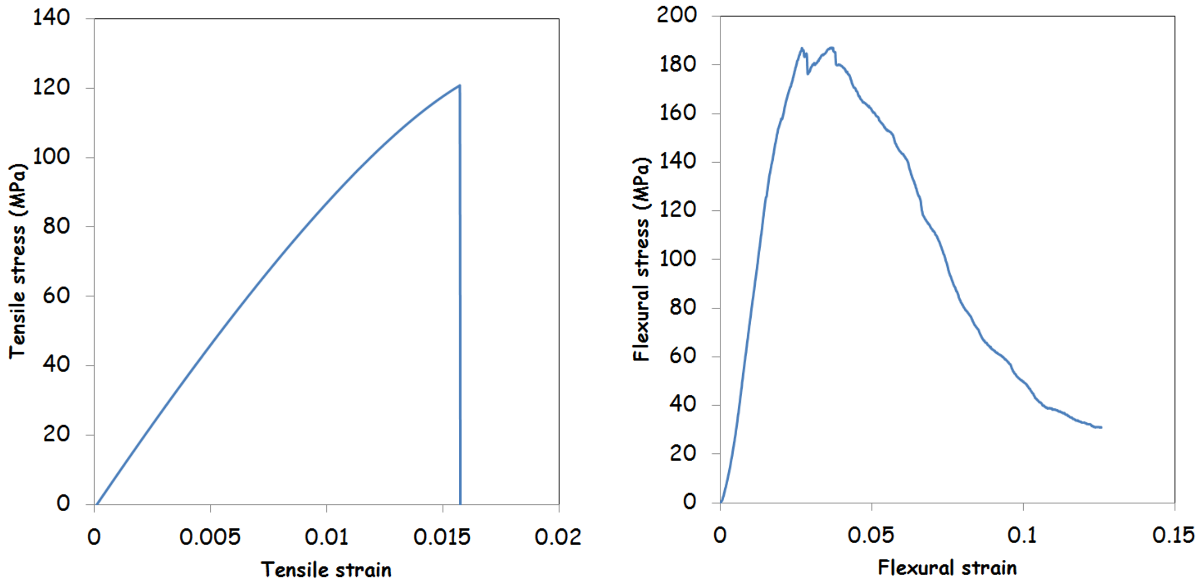

Figure 6: Typical tensile and bending results for intra-yarn PA12 hybrid

The results show that in tension, the samples failed in a brittle manner at a strain of around 1.5%. It was clear that once the carbon fibres broke, the remainder of the sample also broke. In bending the behaviour was different, showing more ductile behaviour and load bearing capacity (on the compression side of the sample), once the carbon fibre fraction had broken. Typical mechanical properties are shown on Table 1 to the right. For a volume fraction of 13%, the expected tensile modus would be 15.6GPa (the carbon fibres have a modulus of 240GPa). The lower measured value is associated with the significant fibre crimp seen in the woven cloth due the thickness of each individual yarn.

	Intra-yarn
Carbon fibre fraction	13 ± 1%
Tensile Modulus (GPa)	9.6 ± 0.8
Tensile Strength (MPa)	111 ± 7
Tensile train to failure (%)	1.9 ± 0.2
Bending Modulus (GPa)	9.5 ± 0.3
Bending Strength (MPa)	185 ± 4
Bending strain to peak stress (%)	3.6 ± 0.1

Table 1: Typical properties for intra-layer hybridisation (13% carbon fibre fraction).

3.2 Intra-layer hybridisation

A similar set of tests were carried out on the manufactured intra-layer hybrid sample. The samples were manufactured at the same temperature as for the intra-yarn samples (176°C). The samples were made from four layers of the woven cloth (with the tapes in the weft direction only), in a 0/90 configuration. The burn off test showed that the carbon fibre fraction for this sample was 8 ± 1%, so lower than the intra-yarn sample. In bending this sample showed different properties depending on whether the carbon fibre prepreg tapes in the outer woven layers were in the direction of bending or perpendicular to the bending direction. The tensile properties were the same in both directions.

Figure 7 shows typical stress-strain curves for these samples in comparison with the intra-yarn samples of section 3.1. In bending the carbon fibre tapes in the outer layer are in the direction of bending.

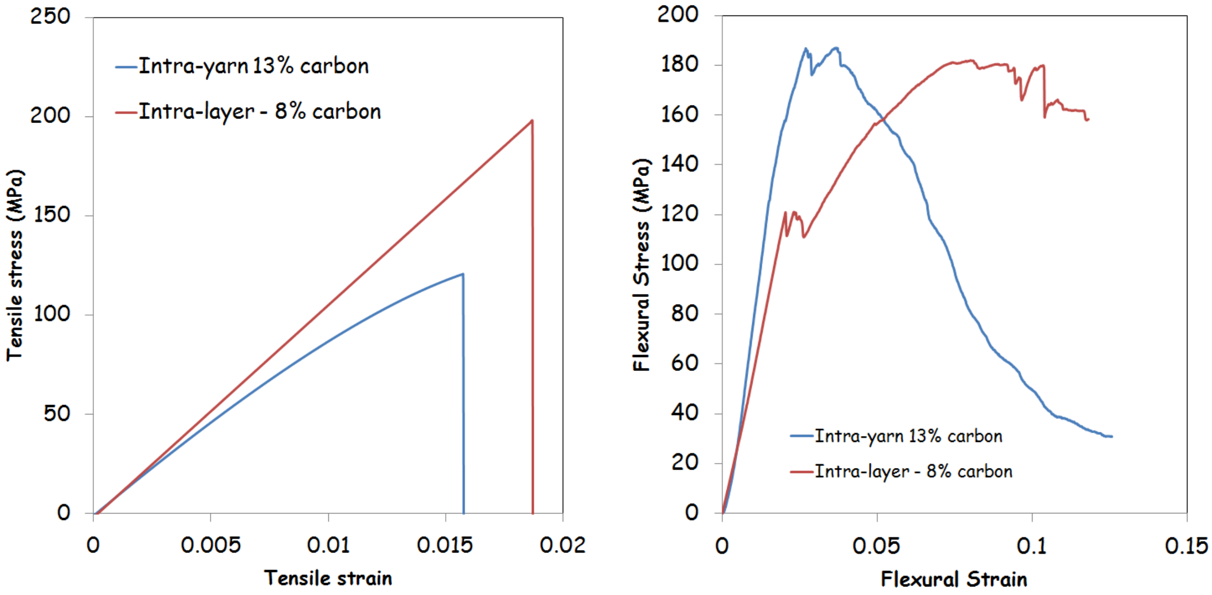

Figure 7: Typical tensile and bending results for intra-layer PA12 hybrids in comparison with intra-yarn hybridisation.

In tension, although the carbon fibre fraction of the intra-layer sample is lower, the stress-strain is much more linear and the failure stress and strain are significantly higher. The measured stiffness (9.2GPa) is also much closer to the predicted one of 9.6GPa. We associate both these aspects of behaviour to the much lower crimp involved in the intra-layer woven cloth. In bending the properties of the intra-layer are again improved over the intra-yarn, showing a much higher strain (ductility) for the peak stress. This could be due to the decreased carbon fibre volume fraction in the intra-layer sample, but might also be due to the way the carbon fibres are distributed. In the intra-yarn sample the fibres are evenly distributed throughout the structure, whereas in the intra-layer sample, there are regions of high carbon fibre fraction (~50%) separated by regions of pure oriented PA12 tapes. Perhaps the combination of these two factors could lead to an increase in ductility.

3.3 Inter-layer hybridisation

The final set of tests were carried out on the interlayer sample. The sample comprised four inner layers of woven pure PA12 tapes plus two outer layers of the impregnated carbon fibre cloth.

Figure 8 show a comparison of the tensile and bending stress-strain of the three hybridisation methods, while Table 2 compares average values for the important measures.

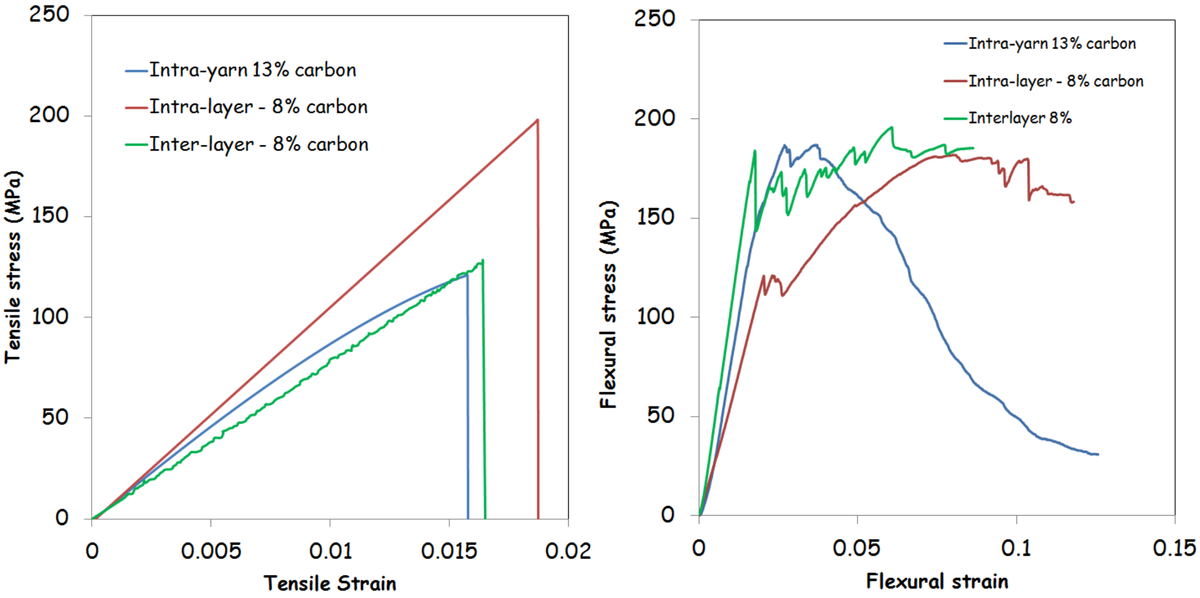

Figure 8: Typical tensile and bending results for the three hybridisation strategies.

	Intra-yarn	Intra-layer	Inter-layer Tex outside
Carbon fibre fraction (± 1%)	13%	8%	8%
Tensile Modulus (GPa)	9.6 ± 0.8	9.2 ± 1.0	8.3± 0.4
Tensile Strength (MPa)	111 ± 7	175 ± 8	126 ± 3
Strain to failure (%)	1.9 ± 0.2	2.0 ± 0.1	1.6 ± 0.1
Bending Modulus (GPa)	9.5 ± 0.3	5.7± 0.4	10.6 ± 1.5
Bending Strength (MPa)	185 ± 4	186 ± 4	186 ± 10
Strain to peak stress (%)	3.6 ± 0.1	9 ± 1	6 ± 1

Table 2: A comparison of the average properties from the three hybridisation methods.

The results show that in tension, the interlayer sample has a comparable stiffness and strength to the lower carbon fibre fraction intra-yarn sample. In bending the stiffness is higher, due to the carbon fibre prepreg layers being on the outer surfaces. The inter-layer sample showed ductile behaviour in bending, in that the sample survived the breakage of the carbon fibre fraction on the surface.

4. Conclusions

To summarise, in tension the inter-layer behaved in a similar fashion to the intra-yarn sample while in bending it was similar to the intra-later sample. In terms of the overall properties, the intra-yarn sample looks to have the best balance. It would be interesting to evaluate an intra-yarn sample with ~8% fibres to assess if the increased ductility seen in the intra-yarn hybridisation is due to the lower carbon fibre fraction or the distribution of the carbon fibres. It will also be interesting to assess other important properties of the hybrids including puncture impact and thermoformability and these will form the basis of future studies.

5. Acknowledgements

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