PSEUDO-DUCTILE BEHAVIOR OF UNIDIRECTIONAL FIBRE REINFORCED POLYAMIDE-12 COMPOSITE BY INTRA-TOW HYBRIDIZATION

H. Diao¹, A. Bismarck^{1*}, P. Robinson², M. R. Wisnom³

¹Polymer and Composite Engineering (PaCE) Group, Department of Chemical Engineering and Chemical Technology, Imperial College London, London, UK, SW7 2AZ ²The Composite Centre, Department of Aeronautics, Imperial College London, London, UK, SW7 2AZ ³Advanced Composites Centre for Innovation and Science (ACCIS), University of Bristol, Bristol, UK, BS8 1TR *a.bismarck@imperial.ac.uk

Keywords: Hybrid composites, polymer matrix composites, pseudo-ductile behaviour, intratow commingling process.

Abstract

Conventional carbon fibre reinforced plastics (CFRP) possess high specific strength and stiffness, can provide good chemical resistance and achieve long fatigue lives. However these materials are relatively brittle and have a low strain to failure and so structural failure can be catastrophic with little warning. In order to enhance the ductility of CFRP and change its catastrophic failure mode into a progressive one, carbon fibre tows with different failure strains were carefully selected and combined together into intra-tow hybrid reinforcement by using a gas-flow-assisted commingling process. This hybrid reinforcement was used to manufacture a polyamide-12 matrix composite using a polymer powder suspension impregnation method. By controlling the manufacturing parameters (speed and air flow rate in commingling process), a hybrid composite with significantly improved failure characteristics was obtained. Compared with corresponding single-fibre type composites, this hybrid composite has an improved tensile failure strain and still retains good tensile strength and stiffness properties.

1 Introduction

Conventional fibre reinforced polymers (FRPs) have high specific strength and stiffness, chemical resistance, long fatigue life and flexible mechanical properties which can be obtained by careful choice of reinforcement and matrix. However, a major disadvantage of FRPs is their brittle nature [1]. This implies that these materials will fail catastrophically when an extreme load is applied. Therefore, FRPs cannot be used in unpredictable load conditions without significant safety factors to ensure the risk of catastrophic failure is acceptably low. In order to improve the ductility and thereby introduce a more progressive failure mode in FRPs, a reinforcement hybridization method has been investigated.

The concept of reinforcement hybridization is that two or more types of high performance fibres with different failure strains are intimately mixed to reinforce a ductile thermoplastic matrix. Unlike conventional laminated hybrid composite (ie. where hybridization is achieved by using laminae of different materials), this intra-tow hybrid composite has a higher degree of commingling, which consists of different fibre materials randomly dispersed throughout in a single lamina. Due to this structure, the hybrid effect, which is defined as the improvement in failure strain over that of the strain to failure for the single fibre composite [2], is potentially more significant than in conventional laminated hybrid composites. This is due to the fact that the low elongation fibre failure can be bridged by adjacent high elongation fibres. Therefore, this reinforcement hybrid composite can provide a progressive failure instead of catastrophic failure while still retaining high strength and stiffness [3]. Such a hybrid material can potentially be used in civil engineering, aeronautics and sports industries as a structural material to replace conventional brittle FRPs.

A key challenge of this study is to intimately commingle the different fibre tows together at filament level to achieve intra-tow hybrid reinforcement while maintaining their fibre alignment in the laminate. Previous works have used parallel winding and air-texturing technologies to process reinforcement fibres yarn and thermoplastic matrix fibre yarn into one hybrid tow, which is then manufactured into continuous fibre reinforced thermoplastic material by pultrusion [4] [5]. In this study, the aim is to commingle two or more different reinforcement fibre yarns rather than reinforcement and matrix yarns; and so it is important that after hybridization the fibres retain their alignment. Only air texturing technology can meet this requirement. In 2006, Herath et al. used this technology to commingle two reinforcement fibres (carbon fibre and glass fibre) and investigated commingled fibre reinforced thermoplastic matrices (PES and PEEK). However, the distributions of different fibres in the matrix and tensile behaviour of these hybrid composites were not characterized in that paper [6]. In this project study, two different carbon fibre tows were hybridized into one reinforcement fibre tow by a gas-flow-assisted commingling unit developed in-house. Then, hybrid carbon fibre reinforced PA-12 composite was manufactured via an in-house laboratory-scale composite production line. The distribution of different fibre types in the composite was measured by microscopy of polished cross-sections. Finally, the impact of hybridization on the tensile behaviour of the composite was investigated.

2 Experimental

2.1 Materials

The reinforcements used in this study were 12K unsized intermediate modulus carbon fibre (IM7, Hexcel Corporation, Cambridge, UK) and 12K sized high strength carbon fibre (T700SC, Toray International Corporation, London, UK). The matrix was PA-12 powder (VESTOSINT 2159, Evonik Degussa GmbH, Essen, Germany). Surfactant (Cremophor A-25, BASF Company, Manchester, UK) was used to disperse the PA-12 powder in water. N₂ (BOC, UK) was used in the gas-assisted commingling unit. Carbon fibre (T700SC) reinforced PA-12 lamina (T700/PA-12, Sulzer Innotec, Winterthur, Switzerland) was used to manufacture a baseline composite laminate for comparison with the hybrid composite.

2.2 Inline production of unidirectional hybrid composite lamina

Two types of unidirectional composite tapes, IM7/PA-12 (width x thickness=7x0.13mm) and IM7/T700/PA-12 (width x thickness=12x0.13mm), were manufactured using an in-house lab modular composite production line (Figure 1), which is based on the wet powder impregnation thermoplastic-matrix manufacture method [7][8]. Firstly, two fibre tows were passed through the tension controlling unit (tension force=1.4N) to hold the fibre spools and stabilize the manufacture speed (0.75m/min), prior to the in-house designed gas-assisted

commingling unit to provide a higher hybridization degree in the commingled tow. This commingled fibre tow was passed through a 2L polymer suspension bath (5 wt-% PA-12 in water, surfactant/ polymer weight ratio=1/20) which used 13 pins to spread the fibres and help the fibre tow pick up the thermoplastic powder. Drying oven (temperature=120 °C) is used to evaporate the water carried by the fibre tow and then followed by melting oven (temperature=205 °C) to melting the polymer powder. Heated shear pins (temperature=205 °C) were needed to further spread and consolidate the melt-impregnated fibre tow into a composite tape. The two-belt puller was used to control the speed of the manufacturing process. In order to keep the concentration of polymer suspension constant, concentrated polymer suspension (15 wt-% PA-12 in water) was added into the polymer bath at 10minute intervals.



Figure 1. Schematic of UD fibre reinforced thermoplastic polymers production line

The detail design of the commingling equipment is shown in Figure 2 (a) and (b). As N_2 (pressure=2.5 bar) passes through the small holes (diameter 1mm) located on the PTFE roller, the two fibre tows which pass onto the rollers can be spread into filaments or small bundles and then commingled.



Figure 2. Detailed design of commingle unit (a) photo (b) schematic diagram

2.3 Preparation of composite laminate

The produced composite tape was cut into 200mm sections, then cleaned with ethanol and dried in the vacuum oven at 105 °C overnight. 15 composite laminae were stacked up in a stainless steel mould (dimension of cavity: 200x12x5 mm) coated with mould release agent (Loctite 700-NC, Henkei Corporation, USA) and then compression moulded. The manufacturing parameters (temperature, pressure and time) are shown in Table 1. The edge of the composite was smoothed by grinding using P180 sand paper.

	Temperature (°C)	Pressure (ton)	Time (min)	
Pre heat	195	0	10	
Hot pressing	195	1	2	
Cool pressing	80	0.5	10	

Table 1. The detailed manufacturing parameters of compression moulding process

2.4 Microscopy analysis of composite lamina and laminate

The cross section of the composite lamina and laminate were embedded into a transparent epoxy (EpoxyCure, Buehler Ltd, Warwick, UK). The epoxy resin was cured at room temperature overnight before being polished. In order to achieve the mirror-fine and stretch-free surface of the microscopy samples, they were ground by P320, P800 and P2500 sand papers respectively, for 5 mins at a pressure of 0.2MPa and a speed of 150 rpm. Finally, they were polished using 6μ m, 3μ m and 1μ m diamond suspensions (MetaDi, Buehler Ltd, Warwick, UK) respectively for 2 mins at the same pressure and speed as the previous grinding process. The samples were observed by using a reflective microscope (BH2, Olympus, Tokyo, Japan).

2.5 Tensile test of unidirectional fibre composite

The dimensions of the tensile specimen are shown in Figure 2. The 100 mm gauge section in the middle of the composite laminate specimen was first protected by a masking tape. Then the two ends of the specimen were sand blasted and cleaned. The end tabs (woven glass fibre/polyester) were glued to the specimen using cyanoacrylate adhesive (CN, Techni Measure Co, Japan). Finally, all the specimens were placed beneath a 5kg weight overnight to allow the adhesive to cure.



Thickness (t) =0.6-0.7mm. Width (w) = 12mm Free length (L) =100mm Length of end tabs=50mm

Figure 2. The dimension of the modified tensile specimen

The tensile specimens were clamped in mechanical wedge action grips and tested in an Instron machine (model 4566, UK) equipped with a 100 kN load cell, loaded at 2mm/min speed. 2mm-long strain gauges (FLA-2-11, Techni Measure Co, Japan) were bonded at the centre of the specimens to measure the strain ε during mechanical testing. The tensile stress (σ) and modulus E of the specimens were calculated according to ASTM D3039 [9].

3 Results and Discussions

3.1 Integrity of produced composite lamina

Figure 3(a) and (b) are photos of the carbon fibre T700/IM7 hybrid reinforced PA-12 composite tape manufactured with and without the gas-assisted commingling process. Clearly, there was a slit in the middle of the un-commingled hybrid composite tape (Figure 3 (a)). This could possibly mean that the two carbon fibre tows were just aligned parallel to each other instead of being mixed. However, this defect was not apparent in the hybrid composite tapes when manufactured using the air-assisted commingling unit (Figure 3 (b)).

(a)







Figure 3. Carbon fibre T700/IM7 hybrid reinforced PA-12 composite tape manufactured (a) without and (b) with commingling process

3.2 Fibre distribution of intra-tow carbon/carbon fibre hybrid composite

According to Figure 4 (a), some T700 carbon fibre filaments (with a diameter of 7 μ m) were inserted into the IM7 carbon fibre tow (with a diameter of 5 μ m) within a single layer hybrid composite tape as expected. However, sometimes these two types of fibres were simply overlapped to form the hybrid tape, seen in Figure 4 (b). These two microscope pictures were taken from two randomly chosen composite tapes, which show that the distribution of the two types of carbon fibres was not uniform during the manufacturing process. However, there were no obvious matrix-rich regions or voids in the hybrid composite tape. In addition, both of these photos show that there was no big gap at the junction area between these two fibres.

(a)



Figure 4. Microscopy pictures of polished surface of two randomly chosen single tape cross section

The polished surface of a T700/IM7 hybrid laminate sample is shown in Figure 5. The fibre volume fraction was relatively uniform and there was no obvious void or porosity in the consolidated laminate. However, there were some IM7- or T700-fibre-rich regions in this hybrid composite (Figure 5), but the degree of hybridization is higher than that obtained by Lauke [10]. It shows that some level of hybridization was achieved by the in house gas-assisted commingling unit. However, the degree of hybridization could be improved by selecting a more effective commingling unit and using unsized carbon fibre as the reinforcement.



Figure 5. Microscopic image of a polished surface of a T700/IM7 hybrid laminate sample

3.3 Tensile behaviour of intra-tow carbon/carbon fibres hybrid composite

Table 2 demonstrates the tensile strength, modulus and failure strain of the non-hybrid composites (IM7/PA-12 and T700/PA-12) and the hybrid composite T700/IM7/PA-12. Figure 6 shows the load-crosshead displacement curves of five hybrid composite samples. There were two peaks in each of these curves for samples 2-5, which shows that this type hybrid composite could still carry a relatively high residual load after first failure had occurred. It is also important to note that there was no sudden load drop before the final failure in sample 1, which indicates that this specimen failed progressively and smoothly. The typical tensile

		Strength (MPa)		Initial Modulus (GPa)		Failure strain (%)	
IM7/PA-12		2137±164		171.2±15.8		1.37 ± 0.06	
T700/PA-12		2017±186		125.7±3.2		1.81 ± 0.10	
T700/		1 _{st} failure	2 _{nd} failure	1 _{st} failure	2 _{nd} failure	1 _{st} failure	2 _{nd} failure
IM7/	Predicted	1932	1506	141.0	83.2	1.37	1.81
PA-	Measured	1786±230	1633±153	142.5 ± 8.0	94.8 ± 18.1	1.26 ± 0.10	1.47 ± 0.10^2
12 ¹	Comparison ³ (%)	-7.56	8.43	1.04	13.88	-8.03	-18.78

behaviour of the un-hybrid and hybrid composite can be seen in Figure 7, which indicates that the hybrid composite can provide some degree of pseudo-ductility.

Table 2. Tensile properties of IM7/PA-12, T700/PA-12 and IM7/T700/PA-12 composite

Note:

1 T700/IM7 volume ratio=1.96/1

2 2_{nd} failure stain of the hybrid composite is the final strain recorded the failure before strain gauge fails

3 Comparison (%) = Predicted value (based on the rule of mixtures)/measured value x 100% -1



Figure 6. Stress-crosshead displacement curves of hybrid IM7/T700 composite samples



Figure 7. Typical tensile behaviour of the non-hybrid (IM/PA-12 and T700/PA-12) and hybrid (IM7/T700/PA-12) composite

It was promising to see that the failure strain of IM7/PA-12 composite was slightly increased through adding the high elongation fibre T700 to form the hybrid composite (Table 2). However, the tensile strength of the hybrid composite was lower than both of the non-hybrid composites, which may be due to the surface damage of the reinforcement from the blowing during the commingling process, shown in Figure 8.



Figure 8. Average tensile behaviour of non-hybrid (IM7/PA-12 and T700/PA-12) and hybrid (IM7/T700/PA-12) composite

Based on the measured tensile properties of these two non-hybrid composites and using the rule of mixtures (ROM), the tensile behaviour of the hybrid composite was predicted. Figure 9 shows the comparison between predicted behaviour and measured behaviour of the hybrid composite. The measured initial modulus was very similar to the predicted value, but the 1st failure stress and 2nd failure strain from the experiment were lower than the predicted ones. This again may be due to the surface damage on the reinforcement caused by the commingling process. Furthermore, the experimental value of the residual strength of the hybrid composite after 1st failure was slightly higher than the predicted value.



Figure 9. Comparison between predicted and measured behaviour of the hybrid (IM7/T700/PA-12) composite

3 Conclusions

The carbon fibres T700 and IM7 were chosen as the reinforcements based on the ROM to produce an intra-tow hybrid PA-12 matrix composite with pseudo-ductile behaviour. This composite was then manufactured by a gas-flow-assisted commingling process and a continuous composite production line based on the powder suspension impregnation method. The fibre distribution in this composite showed that this commingling process could provide some degree of hybridization. The hybrid composite showed improved failure characteristics with slightly increased tensile failure strain compared to the non-hybrid low elongation fibre composite while retaining good tensile strength and stiffness properties compared to the single fibre type composites. The tensile failure mode of this hybrid composite was initially gradual rather than catastrophic, which showed the potential of intra-tow hybridization as a means of achieving pseudo-ductile behaviour.

Acknowledgement

This work was funded under the EPSRC Programme Grant EP/I02946X/1 on High Performance Ductile Composite Technology.

References

- [1] Matthews FL, Rawlings RD. *Composite materials: Engineering and science*. Woodhead Publishing Limited, Cambridge, UK (1994).
- [2] Hayashi T. On the improvement of mechanical properties of composites by hybrid composition in 8th International Reinforced Plastic Conference. British Plastic Federation; UK, (1972).
- [3] Bunsell AR, Harris B. Hybrid carbon and glass fibre composites. *Composites*, **5**(4): pp 157-164 (1974).
- [4] Svensson N. Manufacturing of thermoplastic composites from commingled yarns a review. *Journal of Thermoplastic Composite Materials*, **11**(1): pp. 22-56 (1998).
- [5] Pradhan AK. Tribological properties of aramid-polypropylene composites from commingled yarns: Influence of aramid fibre weight fraction. *Journal of the Textile Institute*, **100(8)**: pp. 702-708 (2009).
- [6] Herath CN, Hwang BB, Ham BS, Seo JM, Kang BC. An analysis on the tensile strength of hybridized reinforcement filament yarns by commingling process in 5th International Conference on Processing and Manufacturing of Advanced Materials, Vancouver, Canada, (2006).
- [7] Ho KKC, Shamsuddin S, Laffan M, Bismarck A. Unidirectional carbon fibre reinforced poly (vinylidene fluoride): Impact of atmospheric plasma on composite performance. *Composites Part A: Applied Science and Manufacturing*, **42**(5): pp. 453-461 (2011).
- [8] Ho KKC. Wet impregnation as route to unidirectional carbon fibre reinforced thermoplastic composites manufacturing. *Plastics, rubber and composites*, **40(2)**: pp. 100-107 (2011).
- [9] ASTM D3039, Standard test method for tensile properties of polymer matrix composite materials (2007).
- [10] Lauke B. Effect of hybrid yarn structure on the delamination behaviour of thermoplastic composites. *Composites.Part A: Applied science and manufacturing*, **29(11)**: pp. 1397-1409 (1998).