ULTRA-LIGHT WEIGHT THERMOPLASTIC COMPOSITES: TOW-SPREADING TECHNOLOGY

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Keywords: Spread tow; Carbon fibre; Lightweight; Thermoplastic-composites.

Abstract

This paper reports a study of a method for achieving ultra-light weight thermoplastic composites referred to as tow-spreading technology, whereby a conventional 12k carbon fibre (CF) tow is thinned by increasing the tow width from 5mm to ca 25mm, thereby reducing the weight per unit area by ca 500%. Using the tow-spreading technology, sheets of unidirectional and/or woven fabric may be produced. Thermoplastic film of Polyphenylene sulphide (PPS) was used to stabilise and impregnate the spread tow fabric, converting it into a partially consolidated prepreg; woven 12k CF spread tow/PPS (50/50 wt. %). A consolidated laminate was then made from the prepreg, and for comparison, a second laminate was produced from a conventional woven prepreg of 3k CF/PPS (60/40 wt. %). The spread tow laminate exhibited better fibre packing, lower level of crimp, lower void content and improved mechanical properties.

1 Introduction

Continuous fibre reinforced thermoplastic (CFRTP) composites are receiving growing interest owing to many attractive advantages in comparison to the more widely used thermoset composites. The advantages are mainly based on the inherent properties of thermoplastic polymers used, such as fracture toughness and damage tolerance (*higher strain to failure*), ease of shape forming prior to consolidation, significantly faster and lower cost manufacturing, longer shelf life of raw material (i.e. *can be stored in any ambient environment with infinite shelf life as do they not contain solvents which limit shelf life*) and the ability to be reshaped and reused/recycled. However, the use of these materials is mostly limited to secondary and semi-structural aircraft parts. In order to make thermoplastic composites more attractive for primary aircraft structures, the performance/cost ratio has to be improved. Improving the quality of thermoplastic composites may be interpreted as a significant increase in properties with no additional cost.

In addition to the strong interest in exploiting the advantages of thermoplastic composites, there is an increasing demand for lighter weight composite structures through better utilisation of the reinforcing fibre, mainly carbon fibre (CF); this also has beneficial cost implications. The state of the art is to use 3k tows for woven fabrics and conventional 12k tows for unidirectional (UD) tapes. A recent method for achieving ultra-lightweight composite material is referred to as spread-tow technology [1, 2], whereby a conventional 12k CF tow (i.e. comprising 12,000 filaments) is thinned by increasing the tow width from 5mm to 25mm, thereby reducing the weight per unit area by ca 500%.

The use of spread-tows results in very thin plies with optimal in-plane and out-of-plane properties. Ply thickness plays a key role in controlling composite mechanical properties: the thinner the ply the better the properties (First Ply Failure Theory [3]). Typically, the transverse microcracking through the thickness of the ply occurs as the first-ply failure, and the delamination damage follows. The fiber breakage usually happens at the last stage of the failure. Kim and Soni [3] studied the effect of thin and thick plies on composite failure by delamination. Their results show that usually the maximum applied stress prior to failure is greater for composites made of thin plies. Rodini and Eisenmann [4] used a probabilistic method to show that laminates with thick plies are likely to contain, statistically, more defects than laminates made with thin plies. Consequently, the thicker plies are likely to exhibit much lower failure stresses. The thickness effect of plies on laminate failure cannot be adequately explained by stress analysis alone. Wang, et al [5, 6] used an energy release rate technique to describe the facture growth in laminates and found that transverse cracking and delamination damage occurred more readily in laminates made from thick rather than thin plies.

Advanced thermoplastic composites are commonly manufactured from prepreg materials [7-9] where the reinforcing fibres are assembled in unidirectional (UD) form or as a woven fabric and then pre-impregnated with a thermoplastic resin. One of the most commonly used methods of producing prepreg materials is film calendaring [10], whereby a thin thermoplastic film is laid onto an assembly of reinforcing fibre, melted and infused into the assembly under high pressures by heat-calendaring. The principal problem with thermoplastic composites has been that thermoplastic matrices have very high viscosities at processing temperatures, within 500-5000Pa.s [11]. To ensure the void percentage of the ultimate composite part is minimal, it is important that the resin-flow distances are as short as possible, hence the need to totally wet out the fibres by melt pre-impregnation of the prepreg. It is reported that for woven fabric, the film calendaring pre-impregnation is the most effective technique.

The fabric structure used to make a prepreg plays a key role in defining the mechanical properties of the resulting laminate. Non-crimp fabric (NCF) is a promising material to use for better mechanical properties. Textiles manufacturers claimed that for woven structures made with conventional tows the best degree of drape and minimum crimp level/angle that can be achieved is with a satin weave. However, the production for NCF is complex, requires an expensive piece of machinery with a large foot-print. The process essentially involves the action of stitching together an assembly of tows which can cause filament damage. These disadvantages have made the idea of weaving spread tows of much interest, since the thinness of the spread tow should provide a simple woven structure, such as plain weave, with an almost negligible crimp level/angle and the closest proximity of adjacent tows within the woven structure, i.e. tightly woven fabrics. Therefore a woven spread-tow fabric should enable good resin wet out.

In this work the spread tow technology is used to convert conventional 12k CF tow into plain woven spread-tow fabric. Thermoplastic film of Polyphenylene sulphide (PPS) was used to stabilise and impregnate the woven the carbon-fibre spread-tow (CFST) fabric. A consolidated laminate of 50/50 %: CFST/PPS was produced, and for comparison, a laminate was also made from a conventional woven prepreg of 60/40 %: 3k CF/PPS. The physical, structural and flexural characteristics of both composite laminates were determined.

2 Materials

The raw materials used were; carbon fibre (CF) tows (12k-T700-Toray) as the reinforcing fibre and Polyphenylene Sulphide (PPS) film as the thermoplastic matrix, and a conventional CF/PPS 60/40 prepreg. Tables 1, 2 and 3 give the properties of the materials. The weave pattern of CF/PPS 60/40 prepreg is satin 5:1. Among conventionally woven structures the satin weave provides the minimum crimp level/angle, and the closest proximity of adjacent tows within the woven structure, and is therefore a useful comparator for the CFST fabric.

Fibre type	No. of filaments	Tensile strength (MPa)	Tensile modulus (GPa)	Elongation (%)	Diameter (μm)	Density (g/cc)	Mass/length Tex(g/1000m)
T700	12000	4900	230	2.1	7	1.8	800

Table 1. The properties of carbon fibre (CF) tows as received

Resin type	Glass transition (°C)	Melting temperature (°C)	Tensile strength (MPa)	Tensile modulus (GPa)	Elongation (%)	Diameter (μm)	Density (g/cc)
CETEX PPS	90	280	90.3	3.8	3	60	1.35

Table 2.The properties of polyphenylene sulphide (PPS) as received

Prepreg Type	No. of CF filaments	Area Weight (g/m^2)	Resin Content (%)	Fibre Volume Fraction (%)	Thickness (mm)	Density (g/cc)
CFS-F4R1- 127	3000	960	40	60	0.5	0.96

Table 3. The properties of CF/PPS 60/40 prepreg

3 Experiments: Fabrication & Characterisation

3.1 Spread Tow Technology

Figure 1 shows a schematic diagram of the machine used for tow spreading. The mechanism involved is referred to as pneumatic spreading and is well described in the literature [1, 2]. Multiples of CF spread tows creels were produced and subsequently used on a loom specially designed for the weft insertion of spread tows [1].

3.2 Fabrication of Composite Laminate

Consolidated composite-laminate panels of dimensions; 12k-CFST/PPS 200 mm x 200 mm x 1.33 mm and 3k-CF/PPS 200 mm x 200 mm x 2.6 mm were produced by hot compaction using a press of two 300 mm x 300 mm square platens with a pressure capacity of up to 50 ton (600 bars or 60 MPa) and temperature up to 500 °C. A pair of release-coated aluminium plates, 300 mm x 300 mm, were placed between press platens and the plies assembled for consolidation. Figure 2 shows the consolidation profile applied for making 12k-CFST/PPS and 3k-CF/PPS woven laminates.

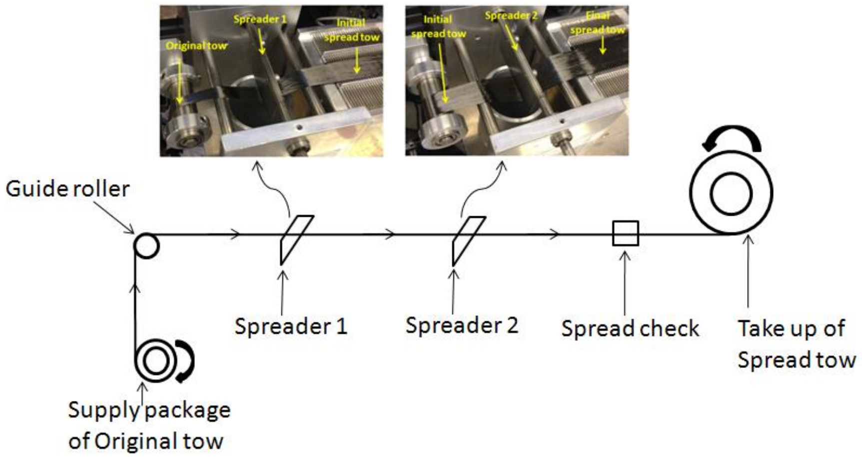


Figure 1. Schematic diagram of CF Tow-spreading by pneumatic method

3.3 Optical Microscopy

Optical microscopic studies were conducted on polished sections of specimens cut from the laminate panels. A Nikon Microscope MM-60 was used to examine the specimens and optical images (micrographs) were captured. Using two-part epoxy compound the samples were cured in 40 mm diameter resin blocks. The polishing procedure is as follows: silicon carbide 800 paper - until flat (30 seconds to a minute), 0.3 micron alumina silica paste for 16 minutes and 0.05 micron alumina silica paste for 16 minutes. Pressure was typically 2.268kg (5lbf) per specimen.

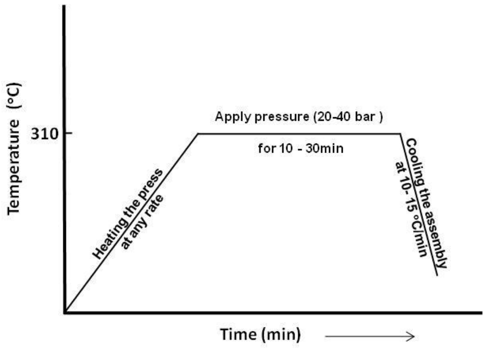


Figure 2. Consolidation profile

3.4 Density, Fibre Volume Fraction and Void Content

Density was measured in accordance with ASTM D792 Method A (zeroed pan immersion) [12]. Specimens, approximately 0.8 g in weight, were cut from the panels using a water lubricated diamond wheel; at least three specimens were tested per material. The specimens were dried for 24 hrs at 100 °C prior to density measurements. Conditioned specimens were weighed in air and then weighed (fully submerged) in a beaker of demineralised water. A Mettler AE240 analytical balance (resolution of 1 mg) was used for weighing specimens.

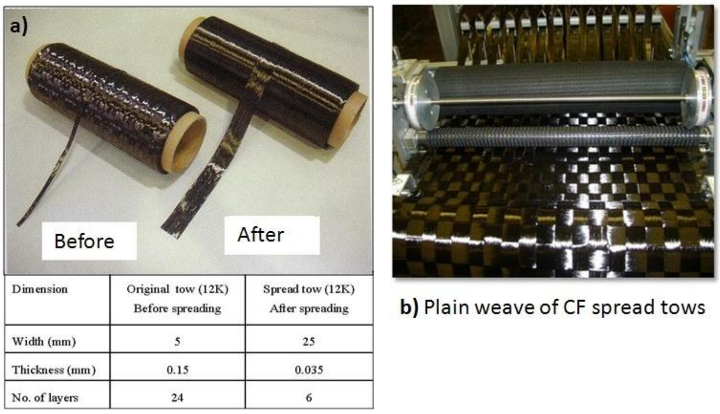
Fibre volume fractions and void content of the composite specimens were determined in accordance with ASTM D 3171-09, Method I, Procedure A acid digestion [13]. The specimens used for the density measurements were re-dried and then digested in 50 ml of nitric acid in a microwave digester, an Anton Paar Multiwave 3000 microwave reaction system. The maximum temperature was set to 105 °C and the exposure time to 90 minutes. It took approximately 15 minutes for maximum temperature to be reached. The PPS was fully digested by the hot concentrated nitric acid. After cooling the liquid and fibres were filtered through a sintered glass filter (Porosity 3) and then washed several times with distilled water and acetone. The filter is then dried at 105 °C.

3.5 Four-Point Flexure Test

The flexural properties (strength, modulus and failure strain) were determined according to ASTM D 7264 Procedure B, four-point loading [14]. The flexure tests were conducted under standard laboratory conditions of 23 °C ± 2 °C and 50% ± 10% RH (relative humidity) at a test speed of 10 mm/min using an Instron 5500 screw-driven test frame with load monitored via a 20kN load cell. Displacement was measured using an ACT 100A linear variable displacement transducer. The load and displacement were monitored throughout the duration of the tests, which were taken to failure. The strain range for the modulus calculation was 0.001 to 0.003. Instron Bluehill® materials testing software was used to control the test machine, and to collect and analyse the test data.

4 Results and Discussion

Figure 3a shows the appearance and dimensions of the CF tow before and after spreading. The spreading of the 12k tow was increased its width by ~ 5 times and reduced its thickness by ~ 4 times. Based on the average fibre diameter (7µm), the number of fibre layers in the 12k tow was reduced from ca. 24 to 6 layers.



Dimension	Original tow (12K) Before spreading	Spread tow (12K) After spreading
Width (mm)	5	25
Thickness (mm)	0.15	0.035
No. of layers	24	6

b) Plain weave of CF spread tows

Figure 3. CF tow before and after spreading

Using the plain weave CFST fabric (Fig. 3b), a 16-ply layered stack was produced consisting of 12k-CFST interlaid with 12 layers of PPS film. The assembly of materials was consolidated by hot compaction using the profile shown in Figure 2 to produce a 50/50: CFST/PPS composite laminate, coded laminate A. A second consolidated laminate was made from 3k-CF/PPS prepreg, coded laminate B. The physical and mechanical properties of the two laminates are given in Table 4. Although laminate A (i.e. CFST) is lighter, the measured densities of both laminates are similar. Theoretically, from their fibre volume fractions of the plies assembled, laminate A (V_F = 50%) should have a density of 1.590g/cc and laminate B (V_F = 60%) 1.632 g/cc. The lower density of the two laminates may be therefore attributable to porosity, which was higher for laminate B. In general the properties of laminate A appear to be better than those of laminate B.

Figure 4 shows optical cross sections of both laminates, and the micrographs would tend to confirm the measured difference in the void contents of the laminates. With laminate B voids appear to be located within the tow, i.e. localised dry regions between and along the fibres (see Fig.4B1 and 4B2). It can be reasoned that in these regions the fibres are more compact together and the air spaces cannot be easily penetrated by the viscose resin flow, an observation also reported by Mitschang et al. [15]. In contrast the laminate A has much wider interfibre spacing (see Fig.4A1and 4A2), which enables easier flow of PPS resin and better fibre wetting, resulting in lower void content.

Property	**Laminate A** **12k-CFST/PPS:50/50**	**Laminate B** **3k-CF/PPS:60/40**
Weight (g/m^2)	2072	4053
Measured density (g/cm^3)	1.558 ± 0.004	1.559 ± 0.007
Fibre volume fraction, V_F (%)	48.1 ± 0.5	51.2 ± 0.1
Void content, V_F (%)	0.9 ± 0.6	1.6 ± 0.5
Flexure modulus (GPa)	54.97 ± 2.1	54.5 ± 1.9
Flexure strength (MPa)	829.7 ± 49.9	748. 7 ± 32.2
Flexural strain to failure (%)	1.98 ± 0.12	1.36 ± 0.20

Table 4. Results of laminates made from CF spread and conventional tows

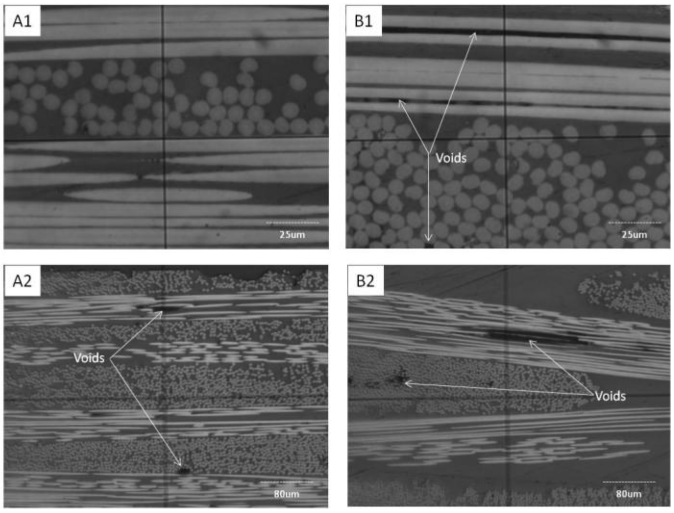


Figure 4. Optical cross sections A) of laminate A, and B) of laminate B. 1 and 2 refer to high (1000x) and low (300x) magnifications.

The tow thickness and weave pattern of the plies used to make a laminate have in combination a significant influence on the mechanical properties of the composite, in that the elastic modulus is inversely proportional to the crimp angle of the weave [16]. Laminate B is made from a 3k satin fabric which provides the lowest crimp for a conventional woven CF material. Laminate A is made from a 12k plain-weave fabric structure, which if it were a conventionally woven fabric would have the highest crimp, but with the reduced tow thickness by spreading exhibits a much lower crimp angle (2^{o}) than laminate B (8.5^{o}), as shown in Figure 5. It can be seen from both Figure 4 and 5 that in contrast to the spread tows of laminate A, the higher crimp of the tow geometry in laminate B result in noticeably resin rich areas (RRA). The factors of more resin rich areas and a high void content for laminate B seem clearly associated with the difference between tow geometries of the two laminates, and is reflected in the better mechanical properties of spread-tow based laminate.

The four-point flexure specimens sectioned from laminate A failed either through interlaminar shear fracture or compressive failure (see Figure 6).The side elevations indicate that compressive failure was initiated in the resin rich areas of warp-weft intersections of the upper surface plies, with interlaminar shear fracture propagating lengthways toward the centre of the span length, before compressive fracture occurred through the specimen’s thickness. The interlaminar shear fracture does not appear to have occurred across the full width of the tested specimens. Inspection of laminate B specimens (Figure 6) revealed that compressive failure and tensile fracture took place; both are acceptable modes of failure [14].

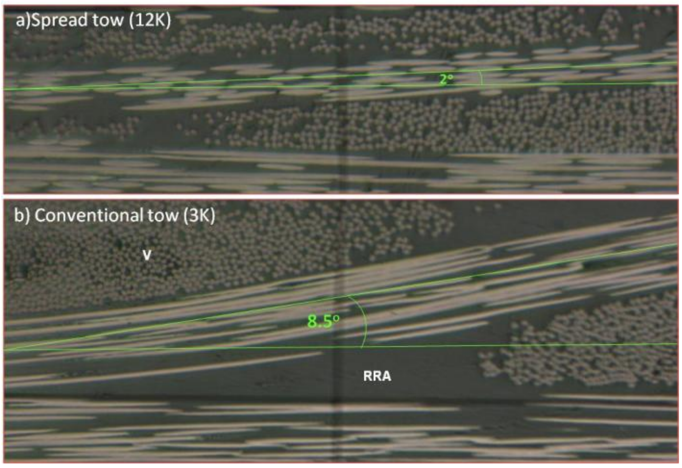


Figure 5. Optical cross sections of a) spread tow and b) conventional tow comparing the crimp level, voids (V) and resin rich area (RRA).

Similar to laminate A, failure was initiated in the resin rich weft-warp intersections of the upper surface ply, but in this case with interlaminar shear facture occurring throughout width and thickness, before tensile facture occurred with each ply of the laminate thickness. Based on the first ply failure theory [3-6], the results in Table 4 and Figure 6, it can be reasoned that the transverse cracking and delamination damage occurred more readily in the laminate made from the conventional 3k-CF/PPS prepreg rather than that from the CFST material.

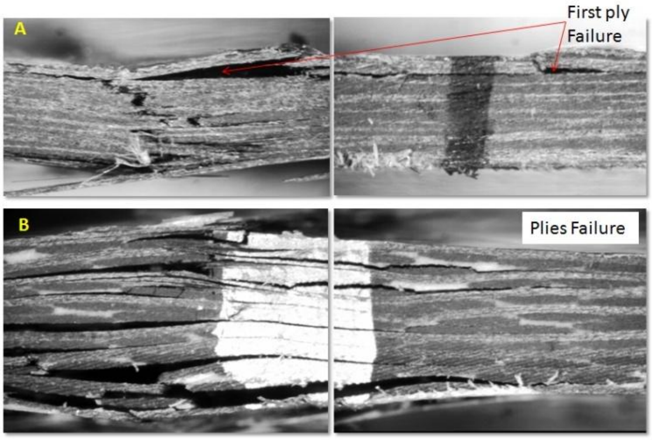


Figure 6. Optical micrographs of flexural tested Laminates A and B

5 Conclusion

Based on the experimental results, the following points can be concluded:

- Tow spreading by pneumatic means is a promising technology for the development of ultra-lightweight composite materials.
- Spread tows enable simple fabrics of plain-weave structure to be produced with lower crimp angles than conventional fabric of satin weave. The spread tows are in closer proximity, but with interfibre spacing that enable adequate flow of a viscous thermoplastic resin to give effective wet-out and impregnation.
- The low crimp angle of a spread-tow fabric reduces the void content and resin rich areas in the composite, resulting in better composite mechanical properties than conventional satin weave fabrics.

Acknowledgement

This work has been co-funded by the UK government-backed Technology Strategy Board under HiPerCom project. The authors would like to thank Technology Strategy Board and HiPerCom partners; University of Leeds, Technical Fibre Products, Tods Aerospace, TilsaTec, Net-Composites and the National Physical Laboratory (UK).

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