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Hybrid composites for the Space industry

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Abstract

Space applications of carbon fibre reinforced polymer (CFRP) composites include several structural components of launchers and satellites, due to their high strength and stiffness properties, provided by carbon fibres. A major weakness of Europe is recognised as its dependency of non-European sources to cover all needs for Space-compliant materials. SpaceCarbon project aims to develop European-based carbon fibres and pre-impregnated materials, enabling a European supply chain capable to reduce the dependency on this critical technology. We present the preliminary mechanical results of laminates containing European fibres, aiming to be used for launcher composite applications.

1. Introduction

The possibility to provide a structural performance at very low weight, leading to significant economical savings, has been the main driver for the use of carbon fibre reinforced polymer (CFRP) composites in Spacecraft applications and the reason with high performance carbon fibres are considered a critical material for such applications.

In these applications, CFRP composites are usually obtained after consolidation of pre-impregnated sheets (prepreg) that consist of semi products in which the reinforcing fibre is pre-impregnated in a thermoplastic or thermoset matrix in a certain proportion. The combined use of heat, pressure and vacuum transforms prepreg into high performance composite materials.

Carbon fibres are typically classified according to two mechanical properties – tensile strength and tensile modulus (Figure 1). The tensile modulus is a measure of the fibre stiffness, and is defined as a ratio between axial stress and axial strain. The maximum stress a material can withstand in tension, above which failure will occur, is the tensile strength. The simplest classification is:

- standard modulus (SM): tensile modulus (TM) < 280 GPa and tensile strength (TS) 3200 – 5700 MPa
- intermediate modulus (IM): tensile modulus (TM) 280 – 350 GPa and tensile strength (TS) 4500 – 7000 MPa
- high modulus (HM): tensile modulus (TM) > 350 GPa and tensile strength (TS) 2800 – 5000 MPa

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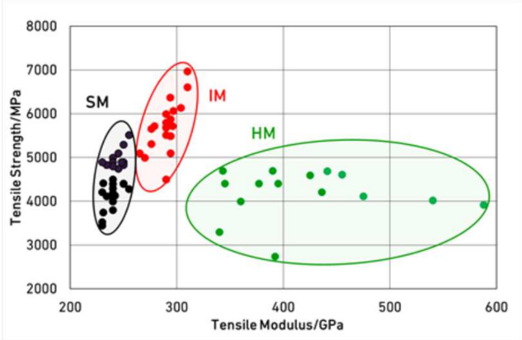

Figure 1: CF distribution according to their tensile strength and modulus

The top five PAN-based CF manufacturers are Toray Industries, Hexcel, Toho Tenax, Mitsubishi Rayon and SGL Carbon. The current global carbon fibre manufacturing industry is predominately located in the United States, Japan, Europe, and China (Figure 2).

Figure 2: Location of carbon fibre manufacturing capacity worldwide [1]

Carbon fibres used in Space applications are supplied by a very limited number of suppliers: particularly for high modulus, two major references M40J and M55J (from Japanese Toray) have been used, while for intermediate modulus, the IM7 from American Hexcel is one of the main choice (see the comparison of properties in Figure 1). Since 2013, Hexcel is producing HMCF in USA facilities (HM63 reference), due to US Government pressure to reduce Japanese dependency. In addition, one should note that these materials are not typically used in the form of fibres by the Space end-users for the manufacturing of composite Spacecraft structures, but in the form of prepregs. In the range of Space relevant characteristics, prepregs are also provided by a reduced number of non-European companies, such as Hexcel or Cytec.

Europe is, therefore, strongly dependent on non-European suppliers to cover all needs for Space-compliant materials. SpaceCarbon project, funded by H2020, aims to develop European-based carbon fibres (CF) and pre-impregnated materials for launchers and satellite applications, enabling a European supply chain capable to reduce the dependency of the European Space

In a previous project (EUCARBON), a European Carbon Fibre just entering the High Modulus range (348 GPa in tensile modulus and 4200 MPa in tensile strength) was obtained at semi-industrial scale for the first time and it was implemented the capacity to develop prepreg materials for Space applications.

In SpaceCarbon, the main objectives are:

- to develop semi-industrial manufacturing process for Intermediate Modulus (IM) CF, targeting mainly launcher applications (Figure 3)

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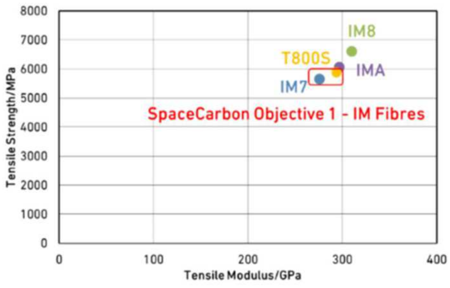

Figure 3: Commercial fibres versus SpaceCarbon objectives for launcher demonstrators

- to improve the properties of the previously obtained High Modulus (HM) CF aiming at reaching a modulus in the range of 440 to 540 GPa, entering the range of mechanical properties of fibres used in satellite sub-system applications (Figure 4)

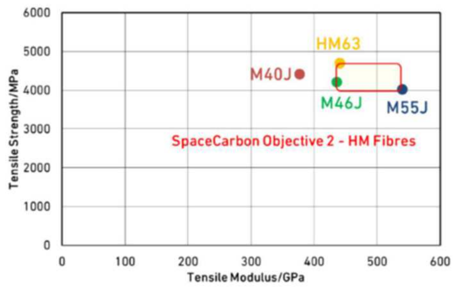

Figure 4: Commercial fibres versus SpaceCarbon objectives for satellite demonstrators

- to scale-up the prepregs manufacturing process to provide such materials at short-term to European Space End-users

- to develop new prepreg formulations at lab scale, in view of enhancing composites performance for future Space missions

- to design and test launcher and satellite demonstrators manufactured with European fibres

The project started in January 2018, and in the first 18 months, European IM fibres with mechanical properties in the range of the requirements for the launcher demonstrators were successfully manufactured. The use of these fibres at both laboratorial and semi-industrial scale allowed to obtain laminates also acquiescent with the requirements.

Further work has been focused on attaining the defined mechanical targets, however, combining two fibres in the same laminate, in order to obtain hybrid materials. The term 'hybrid composite' is generally used to describe a matrix containing at least two types of reinforcements. By combining two or more fibre types, hybrid composites offer a better balance in mechanical properties than non-hybrid composites, which can be a solution, for instance, to toughen composite materials. The purpose of bringing two fibre types in a single composite is to maintain the advantages of both fibres and alleviate some disadvantages. For instance, replacing carbon fibres in the middle of a laminate by cheaper glass fibres can significantly reduce the cost, while the flexural properties remain almost unaffected. Usually one of the fibers in hybrid composites is high cost, with high modulus and high specific strength, hence relatively brittle. The second fibre is frequently cheaper, with low modulus and lower specific strength, therefore more ductile.

In this case, we combined higher modulus 50k fibres (IM) manufactured by SGL Composites with commercial lower modulus 24k fibres (SM), creating interlayer hybrid composites. Typical hybrid configurations include interlayer, intralayer and intrayarn (Figure 5).

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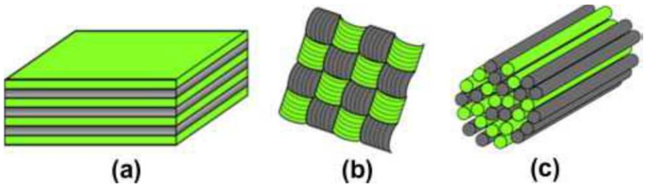

Figure 5: The three hybrid configurations: (a) interlayer, (b) intralayer, and (c) intrayarn

The ratio of higher to lower modulus fibre prepreg sheets, as well as the total amount of prepreg layers, was maintained the same in all materials. However, the location of lower modulus fibres prepreg within the laminate structure was varied, although lower material grades were always located in the inner layers of the laminates.

2. Experimental

2.1 Raw Materials

The resin system used SR121 from Sicomin. European carbon fibres with the properties presented in table 1 were manufactured in the first months of the SpaceCarbon project. For comparison, the requirements defined for launcher applications, as well as the commercial reference from Toray (T800S), are also presented. From these, prepregs and laminates with E2021 and E2022 fibres were developed and characterized. Work is ongoing with12k fibres, and results will be soon available. Reference fibres were tested in previous project EUCARBON, therefore not yet tested during SpaceCarbon project. For hybrid laminates, European fibres E2021 were combined with commercial fibres CT24.

Table 1: Main properties of the fibres used to manufacture prepregs and laminates

	E2021	**E2022**	**E2045**	**CT24-5.o (SGL)**	**Requirements**	**T800S (Toray)**
TM (GPa)	319	315	285	270	275 - 300	294
TS (MPa)	5900	5450	4700	5000	5500 - 6000	5880
Number of filaments	50k	24k	12k	24k	24k	24k

2.2 Prepreg manufacturing

The prepreg sheets were manufactured using the resin dip process. In the dipping process, the fibre tows are directly dipped into a resin bath and impregnated by direct contact with the resin or by the aid of impregnating rolls. The viscosity of resin solution can be controlled by the bath temperature and/or by addition of a solvent content in the bath. Excess resin on the surface is removed by calender rolls. A flowchart for prepregs and laminate production is represented in Figure 6. In the first step, the focus is to obtain prepregs with the required characteristics, such as fibre content, fibre areal weight, among others. If the parameters are within the desirable, laminates are manufactured and mechanically characterized by AAC.

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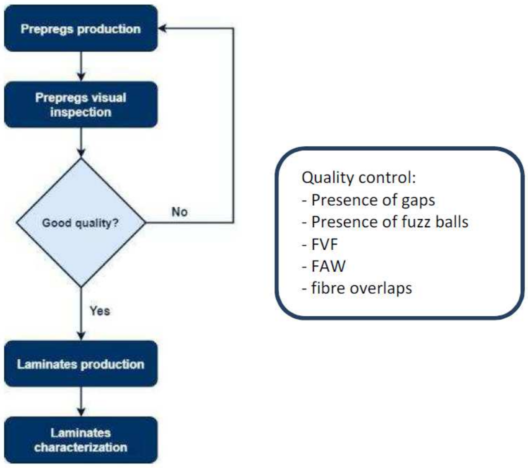

Figure 6: Flowchart for pre-prepregs and laminate production

Some representative pictures of prepreg manufacturing at laboratorial scale using the drum winder are presented in figure 7.

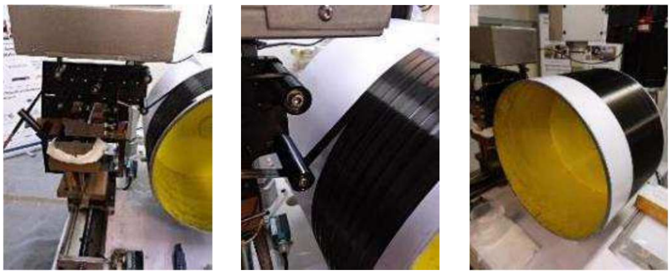

Figure 7: Lab scale prepregs manufacturing

2.3 Laminate manufacturing

There are four basic steps involved in composites part production: impregnation, lay-up, consolidation, and solidification. After prepreg manufacturing, prepreg lamination involves cutting the plies into the required shape, removing the backing paper and placing into a mould. After applying all the prepreg sheets, vacuum bagging arrangements are prepared and the entire assembly is then placed into the autoclave for the resin cure; the curing conditions used to consolidate the materials are shown in figure 8.

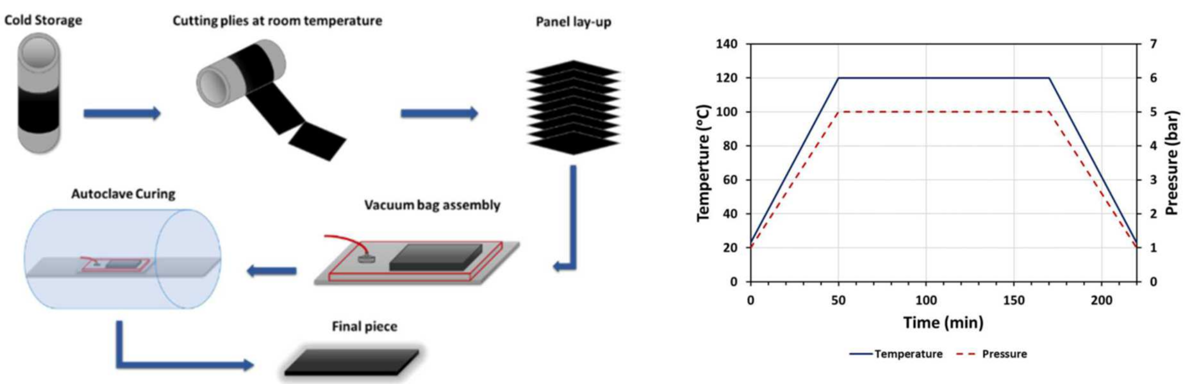

Figure 8: General prepreg processing flow (left) and conditions used in prepreg consolidation (right)

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Figure 9 presents some details of the laminates production, including prepreg layup, vacuum bag preparation and samples after curing cycle.

Figure 9: Details of the laminate production steps

The behaviour of hybrid composites is a weighed sum of the individual components in which there is a more favourable balance between the inherent advantages and disadvantages. As consequence, a balance in cost and performance can be achieved through proper material design. Ideally, the mechanical properties of the hybrid configurations would be closer to the higher modulus laminate properties, leading to important cost savings. Therefore, two types of hybrid composites were manufactured, by changing the stacking sequence of the prepreg sheets (Table 2). The purpose of these preliminary evaluations were to determine the effect of using lower modulus carbon fibre prepregs in the inner layer of the laminate. Both hybrid laminates were manufactured with the same number of layers, but changing the prepreg arrangements in the laminate.

Table 2: Resume of manufactured laminates

	Short description	**Prepreg layers**	**Stacking sequence**
E2021-50k-1	IM European fibres, 50k	8	---
E2022-24k-1	IM European fibres, 24k	10	---
CT24-24k	Reference for hybrids: SM commercial fibres, 24k	10	---
E2021-50k	Reference for hybrids: IM European fibres, 50k	10	---
HYB-1	Hybrid: IM European fibres, 50k + SM commercial fibres, 24k	10 (6 IM + 4 SM)	50k (3) – 24k (4) – 50k (3)
HYB-2	Hybrid: IM European fibres, 50k + SM commercial fibres, 24k	10 (6 IM + 4 SM)	50k (2) – 24k (2) – 50k (2) – 24k (2) – 50k (2)

3. Results

3.1 Fibres

The structure of European IM modulus was compared with commercially available ones, by performing scanning electronic microscopy analyses. Some of the obtained pictures are presented in figure 10.

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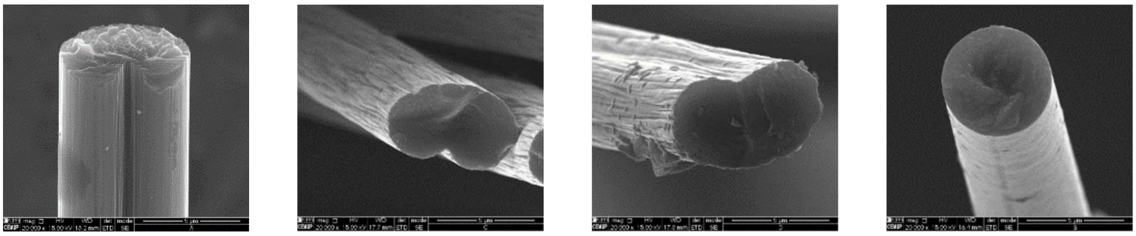

(a) E2021, 50k (b) E2022, 24k (c) E2045, 12k (c) T800S, 12k

Figure 10: Cross-sectional geometry structure observations of carbon fibres for launcher applications

The cross-sectional geometry of the fibres are clearly different: fibres manufactured by FISIPE show a kidney-shaped format, while Toray fibres are circular. W. Steinmann et al. [2] reported that round cross-section fibres provide a uniform mechanical loading of the material, leading to higher specific strength compared to other fibre shapes. The authors explain that non-round filaments (such as kidney shaped) can be formed when inhomogeneous conditions are used in the spinning process, specifically the coagulation conditions. By changing the shape of the spinneret during fibre production or specific post-treatment of the fibres, different fibre cross-sections can be achieved. In a paper from 2008 [3], however, authors claim that the kidney-type fibres with larger specific surface area had a better adsorption characteristic and higher surface free energy compared with the circular fibres. Interlaminar shear loads showed that the kidney-type fibre/epoxy composites outperformed the circular fibre/epoxy composites by more than 12%. The matrix was engulfed by the hollowed-out area of the kidney-type fibre, which allowed the matrix to secure more bonds, resulting in the effective transfer of the load applied to the fibre-reinforced composite system. Many kidney-type fibres overlapped and embraced with each other. Thus the large contact areas between fibres became strong bonding places and interface between fibres and matrix was not damaged easily.

Work is ongoing to study the influence of the fibres geometry in the prepreg and composite properties. The fibre cross section can influence on the processing by changing characteristics such as tribology or wetting behaviour.

3.1 Prepregs

i. Fibre Volume Fraction and Fibre Areal Weight

One of the most important features of a prepreg is its resin content. As expected, the higher the fibre content of the cured composite, the better its weight specific strength and stiffness. On the other hand, a higher resin content prepreg offers more robust and easy to handle manufacturing processes. The fibre volume fraction was determined following standard ASTM D3529/D3529M, which is based on removing the matrix content using a solvent (in this case, acetone was used). In this way, it is possible to calculate the matrix content, expressed as mass loss, and consequently the fibre content fraction, as well as the fibre areal weight.

A simple definition of areal weight (AW) is the weight (in grams) of 1 m^2 of material. Naturally, FAW depends on fibre density, fibre tow size and fibre spread. FAW values can vary from 50 g/m^2 for lightweight applications to 2000 g/m^2 for heavyweight non-crimp fabrics. The prepreg areal weight is gravimetrically determined by weighing a prepreg sample of defined size.

The results obtained for fibre volume fraction (FVF), surface mass (SM) and fibre areal weight (FAW) of the manufactured prepregs are summarized in table 3.

Table 3: Prepreg characterization results

	E2021	**E2022**	**CT24-24k**	**E2021-50k**
SM (g/m^2)	219 - 247	130 - 173	157 - 189	160 - 230
FAW (g/m^2)	132 - 137	81 - 99	100 - 120	111 - 135
FVF (%)	49 - 51	47 - 57	54 - 61	47 - 60

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3.1 Laminates

i. Fibre Volume Fraction

The fibre volume fraction was determined according to standard ASTM D3171 – 15, using the matrix burnoff in a muffle furnace method. This method is more secure and environmental friendly and secure than the chemical digestion method. Several comparison tests were performed using both methods in the same samples, and similar results were obtained. In this way, it was decided to use the matrix burnoff in a muffle furnace method. The main results for each laminate are presented in Table 4.

Table 4: Fibre volume fraction of manufactured laminates

	FVF (%)
E2021-50k-1	49
E2022-24k-1	48
CT24-24k	45
E2021-50k	52
HYB-1	53
HYB-2	50

ii. Tensile Tests

Tensile tests in fibre direction were performed by AAC, according with the standard ASTM D3039. According with this standard, a thin flat strip of material having a constant rectangular cross section (see Table 18) is mounted in the grips of a mechanical testing machine and monotonically loaded in tension while recording load. The ultimate strength of the material can be determined from the maximum load carried before failure. The tensile tests results of laminates with European IM fibres, obtained in fibre direction and normalized to a FVF of 60%, are presented in Figure 11.

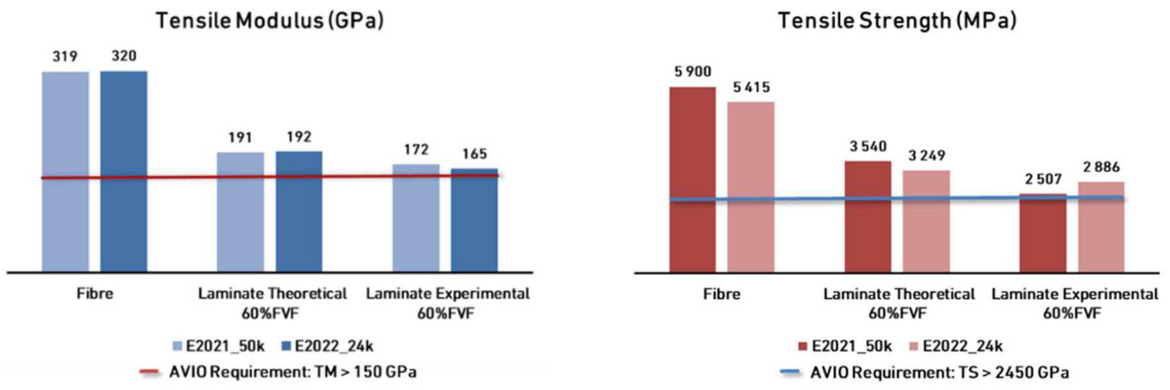

Figure 11: Tensile tests results in fibre direction for laminates produced with European IM carbon fibres

The experimental values obtained for TM and TS in fibre direction are above the requirements for the launcher demonstrator. Based on the carbon fibres values for the TM and TS, theoretical values of TM and TS were calculated for the laminates produced and compared to the ones obtained experimentally. As expected, the theoretical values are higher than the ones obtained experimentally, since the manufacturing process naturally imposes some defects (such as voids, fibre misalignments, etc). This means that the SpaceCarbon European developed fibres are able to fulfil the needs for launcher composite parts.

For the hybrid laminates, the tensile results, in fibre direction, are summarised in figure 12.

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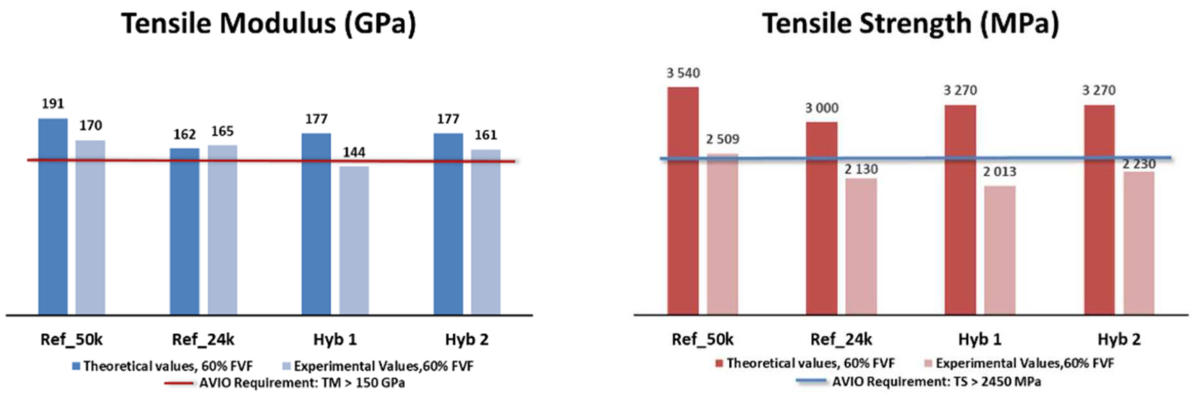

Figure 12: Tensile tests results in fibre direction for hybrid laminates produced with European IM carbon fibres and commercial fibres

Hybrid 1 is a core-shell hybrid composite, composed by four prepregs layers of CT24 carbon fibre sandwiched between two layers of three prepregs sheets of E2021 carbon fibre. Otherwise, Hybrid 2 is an interplay laminate where two prepregs sheets of CT24 carbon fibre is stacked alternately with two prepregs sheets of E2021, in an ordered manner. Both laminates present the same content of each carbon fibre type: 60% of E2021 and 40% of CT24. However, hybrid 2 laminate presents higher TM (161 GPa) and TS (2230 MPa) values when compared with Hybrid 1 (TM: 144 GPa; TS: 2013 MPa). This fact is explained by the difference in the lay-up design of each composite, since in hybrid 1 the lower performance fibres are all concentrated in the centre of the laminate, minimizing the impact on the tensile properties of the laminate. One can conclude that, while maintaining the production costs, it is possible to obtain performance increases in the range of to increase in more than 10% the TM and TS values, just by refining the lay-up design.

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4. Conclusions

This paper intended to present the main drivers and objectives of H2020-funded SpaceCarbon project and highlight some of the main achievements during the first 18 months. Intermediate modulus carbon fibres produced entirely in Europe, and compliant with launcher applications, were successfully manufactured and transformed into prepregs and laminates. These laminates were mechanically characterized in terms of their tensile properties on fibre direction, showing properties within the range of compliance for the defined demonstrators.

Aiming at reducing the overall cost of the final composite materials, a lower modulus and lower cost carbon fibre was combined with intermediate modulus fibre, to obtain hybrid laminates. The results showed that the prepreg layup influences the tensile properties (modulus and strength), since when lower modulus fibre prepregs are located together in the inner layers of the laminate, the properties are higher than when lower and higher modulus prepregs are intercalated. Although in the lower limit of the defined requirement, especially regarding the tensile strength of the laminates, these materials are prone to be successfully used in launcher composite parts, with lower cost.

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