New Intelligent Semi-Products based on Recycled Carbon Fibres

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Abstract. The carbon fibre recycling industry is not yet able to operate at full capacity. This lack of potential is a repercussion of a low demand for recycled carbon fibres (rCF) to manufacture new composite materials. As a matter of fact, few semi-products containing recycled carbon fibres are available on the market. Moreover, rCF semi-products available do not allow to manufacture high performances composite parts. The MANIFICA project, based on highly realigned carbon fibres after steam thermolysis, aims at producing new semi-products from recycled carbon fibres for high performance composites. In this article we introduce the I2M/Université de Bordeaux re-alignment process producing continuous tapes made of highly aligned long discontinuous fibres. These tapes are then used to manufacture new intelligent rCF semi-products. In the first part, the mechanical properties of rCF composites based on different semi-products are compared. In the second part, several semi products based on realigned fibres tapes are presented. This work demonstrates that high performance products can be targeted with recycled carbon fibres, thanks to the development of these intelligent semi-products.

Introduction

The aeronautic industry produces an increasing amount of carbon fibre composites parts. As a result, larger volumes of carbon fibre composites waste are generated. Most of them are sent to landfill even though recycling processes have existed for many years. Two steps are required to recycle carbon fibre composites. First the matrix needs to be removed using pyrolysis, solvolysis or steam-thermolysis process. This step is controlled and available at industrial scale. The second step is the reshaping of recycled carbon fibres (rCF), it is more complex and not yet sufficiently studied. It is actually a crucial step to the industry as it provides the final performances of recycled carbon fibres [1]. Several technologies are available to reshape reclaimed carbon fibres. They can be milled, chopped or partially aligned into non-woven fabrics [1]. H2020 Cleansky MANIFICA project (2020-2023) comes up with a new technology to reshape rCF and manufacture highly aligned rCF semi-products. High stiffness and high strength can then be expected from the composites based on these new semi-products.
1. Influence of rCF semi-products shape on the composite performances

1.1. Semi-products currently available on the market

After reclaiming carbon fibres through a pyrolytic or a chemical method, a loss of fibre architecture is observed [1] [2], fibres appear to be entangled, discontinuous and not oriented. These discontinuous fibres can’t be directly implemented to manufacture new composite materials. The remanufacturing challenge is to design interesting semi-products considering discontinuities of rCF.

However, it has been possible for several years to manufacture composites materials based on rCF. Several types of rCF semi-products are currently available on the market:
- Milled fibres (fibre length < 1 mm) [3] [4]
- Chopped fibres (3mm<fibre length<20mm) [4]
- Wetlaid non-woven fabrics (fibre length<20mm) [5]
- Carded non-woven fabrics (20mm<fibre length < 40mm) [5]

These rCF semi-products are industrially produced and available at large scale. Their quality is controlled and thus convenient to produce new “2nd generation” composites. Short fibres are used as fillers in the compounding industry while partially aligned fibres are used as reinforcement in thermoset composites [1].

1.2. Characteristics and performances of current rCF composites

The fibre ratio obtained in composites made of rCF mixed with thermoplastic resin is generally quite low. When Milled fibres are used as a filler, the fibre weight fraction does not exceed 10% [6]. For chopped fibres used in injection moulding process, fibre weight fraction is limited to 40% [7]. When composites are made from rCF non-woven fabrics and thermoplastic matrix through compression moulding process, the fibre volume fraction reaches 25% [8]. However, rCF carded non-woven mixed with epoxy resin yield a fibre volume fraction of 40% [7]. This result can be obtained using carded non-woven fabrics where fibres are partially aligned. Fibre volume fraction is dropping to 20-25% [5] for composites made of non-woven fabrics with randomly orientated fibres. These characteristics are displayed on figure 2 and 3.

![Figure 1: rCF semi-products available on the market](image)

![Figure 2: Fibres length within the different rCF semi-products](image)

![Figure 3: Fibre volume fraction of composites as a function of fibres alignment within rCF semi-products](image)
Tensile strength of composites made of discontinues fibres mainly depends on fibre length and fibre volume ratio. Thus, it spreads from 50 MPa for Thermoplastic composites reinforced with short fibres to 350 MPa for thermoset composite reinforced with carded fabrics. Young modulus of these materials ranges from 10 GPa to 35 GPa (figures 4 and 5) [3], [5], [8], [9], [10].

Therefore, rCF composites offer good mechanical properties, especially when they are made from long and organised fibres. Nevertheless, the specific mechanical properties remain lower than those of metal alloys or glass fibre composites (figure 6).

Considering semi-products currently available on the market, rCF composites do not come up with new mechanical performances to compete with lightweight alloys. It points out in one hand the reason why rCF is struggling to find a position on the market, and in the other hand it explains the reason why recycling is not yet fully integrated in the composites industry.
2. New MANIFICA’s rCF semi-products

2.1. MANIFICA re-alignment technology

After steam thermolysis, solvolysis or pyrolysis, recycled carbon fibres are recovered as superposed layers of fabrics. If the collected composites waste has been cut into pieces (instead of being shredded), it is possible to recover long discontinuous fibres (as long as the original cutting). In order to exploit the high properties of the carbon fibres, the I2M-Université de Bordeaux patented an un-weaving machine for recycled carbon fabrics (Figure 7), able to save fibres integrity while realigning them [11].

Fabric pieces of rCF are transformed into a continuous tape (made of discontinuous carbon fibres). The process was specifically designed to prevent any fibre breakage, in this way recycled fibres from 80 to 250 mm long are processed. The continuous tape is consolidated with a thermoplastic powder (2w%) and thus can be wound on a bobbin. The facility is currently able to produce 7 kg/h (200m/h). MANIFICA project has the ambition to reach industrial scale by building a new facility (30kg/h in 2023).

2.2. Innovative characteristics of rCF semi-products developed by MANIFICA

The benefit of the un-weaving/re-alignment process saving fibres integrity, is to obtain rCF semi-products with a controlled fibre length that can reach up to 250 mm (figure 8).
The second crucial innovation is the high alignment of the fibres. It is obtained in one hand by a vibrating re-alignment technic, and in the other hand thanks to the fibre length. As a matter of fact, long fibres are easier to align than short fibres. The alignment of MANIFICA’s technology reaches 80% of the fibres orientated within 0°± 10°.

2.3. Intelligent semi-products developed by MANIFICA

While highly aligned rCF tapes can be directly used to manufacture new composites, one of the key points of the MANIFICA project is to offer easy-to-process semi-products. Several semi-products will be developed:
- Unidirectional preforms
- Bidirectional preforms 0/90°
- Bidirectional needle punched preforms 0/90°
- Unidirectional tapes impregnated with thermoplastic matrix

3. Properties of composites manufactured from rCF semi-products

3.1. Fibre volume fraction (Vf) in MANIFICA composites

As a result of the high alignment of the fibres, it is possible to significantly improve the compactness of rCF preforms and thus to manufacture high fibre volume fraction composites. As presented on figure 10, Vf obtained lays between 45% and 65%.

Figure 9 MANIFICA rCF semi-products

Figure 10 Fibre volume fraction of composites as a function of fibres alignment within rCF semi-products
3.2. **Strength of MANIFICA composites**

As a result of the fibres length and the high fibre volume fraction obtained, manufacturing unidirectional composites with high strength is now achievable. As displayed on figure 11, a tensile strength of 550 to 850 MPa is attained by MANIFICA composites.

![Figure 11 Tensile properties as a function of fibres alignment within the rCF semi-products](image)

The specific strength of MANIFICA rCF unidirectional composites ranges from 290 to 550 kN.m/kg, acquiring higher performances than high performances metals such as titanium, magnesium or aluminum alloys (100 to 260 kN.m/kg).

3.3. **Stiffness of MANIFICA Composites**

A high stiffness is a direct consequence of the fibre alignment achieved within the UD preforms. It is interesting to note that unidirectional rCF composites stiffness reaching 115 GPa is henceforth getting close to virgin CF unidirectional composites stiffness (figure 12).

![Figure 12 Tensile Modulus as a function of fibres alignment within rCF semi-products](image)
The specific stiffness of rCF UD composites ranges from 58 to 85 MN.m/kg, having consequently the second-best specific stiffness after virgin carbon fibre composites.

3.4. MANIFICA rCF aligned tape position on the market.

The specific mechanical properties of MANIFICA composites places these rCF materials in the category of very high-performance materials, much higher than aeronautical lightweight alloys or glass fibres composites. Moreover, these new composites are positioned where no other materials obtained these performances (figure 13). It demonstrates the great innovation potential of these new rCF composites.

As a consequence, MANIFICA rCF semi-products can be used to manufacture semi-structural parts. Whereas performances of composites from chopped fibres or partially aligned non-woven fabrics limited their use to lightly loaded parts. It might explain the lack of interest toward the use of today’s recycled carbon fibre. MANIFICA materials are dedicated to parts sized for their high stiffness and strength. We assume that the key point to technically and economically boost the carbon fibre recycling industry is to produce high performance materials.

![Figure 13](image)
Conclusion

MANIFICA project is setting up the industrialization of high performances recycled carbon fibres semi-products. As a result of the high alignment of non-damaged recycled fibres, it is now possible to manufacture composites with high strength and particularly high stiffness. With this innovation, recycled carbon fibres can be used to manufacture semi-structural parts. MANIFICA’s new semi-products are easy to process, adapted to manufacture complex shape composite parts. Moreover, tapes are currently improved to be suitable for new processes such as automated tape laying. These intelligent semi-products can be used to manufacture composites with higher performances than most of existing materials. This is the key to develop the carbon fibre recycling industry. Therefore, recycled carbon fibres will not only be preferred for their low environmental impact, but also for their high mechanical performances.

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